Polarization-sensitive infrared magneto-optical studies in two-dimensional materials ranging from graphene to high $T_c$ superconductors

by

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Dedicated to my parents.
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Abstract

This Dissertation presents experimental mid-infrared Faraday and Kerr effect studies of a variety of materials: graphene, Al-doped ferromagnetic SiC, high Tc cuprate superconductors (YBCO, LSCO, LBCO) and iron-based superconductors (BaFe$_2$As$_2$). The Faraday and Kerr effects are optical analogs of the Hall effect and are sensitive to the off-diagonal conductivity $\sigma_{xy}(\omega)$. By measuring the polarization of the transmitted (Faraday) and reflected (Kerr) radiation, these measurements are sensitive to the underlying symmetries of the system. The main goal of this research is to explore the symmetries that can give us new insights into novel materials.

The main challenge in measuring the Faraday and Kerr response of a material lies in the ability to measure tiny changes in the polarization of transmitted/reflected light. Using a unique magneto-polarimetry setup we have measured Faraday/Kerr angles as small as 10 $\mu$rad.

The primary results of this work include the discovery of a colossal enhancement of the magneto-optical Kerr effect in films on SiC. In epitaxial graphene, the Kerr response gets enhanced by a factor of 68 near the reststrahlen band of SiC. The enhancement effect works for any polar substrate and provides a new way of enhancing the infrared Kerr response from complex materials in which the signal usually is very small. Moreover, we predict enhancement in Faraday response in films grown on metamaterials.

By studying the changes in the polarization of reflected light in epitaxial graphene grown on terraced substrates of SiC, we also have discovered Kerr rotation and ellipticity at zero magnetic field. This optical effect originates from the anisotropy in graphene’s
conductance when it grows over a terraced substrate like SiC, acting as a wire-grid linear polarizer and producing a Kerr signal even in the absence of an external magnetic field. Proposed next-generation displays will combine liquid crystals and graphene, so understanding the polarizing properties of graphene is also important for technological applications. Our technique of measuring the polarization properties of graphene at zero magnetic field and at room temperature gives a new contactless- noninvasive way of probing the anisotropy of graphene.

We also explore broken symmetry states in high-temperature cuprate superconductors (HTCS) by measuring the zero-field Faraday response. In HTCS we have found linear dichroism (preferential absorption of light polarized along one axis) in mid-infrared and also at visible wavelengths at room temperature and down to 10 K. Earlier studies in THz and near IR have shown that the signal in HTSC is linked to different broken symmetries in the pseudogap region such as stripes. The Faraday rotation signal is strongest in underdoped films, depending on both temperature and sample orientation, which suggests that there is linear symmetry breaking in the pseudogap region.

With its sensitivity to electronic structure and magnetic ordering, we have studied Faraday response in iron-based pnictide superconductors to understand the interaction of superconductivity (SC) and ferromagnetism (FM). Faraday and Kerr effects studies have been used in the past to explore these two phenomena separately. The pnictide (Co-doped BaFe$_2$As$_2$), films that we have studied show both SC and FM behavior. We use the frequency and temperature dependence of our Faraday response measurements to explore how the SC and FM phases interact with each other.
1 Introduction

Two-dimensional (2D) layered materials exhibit rich and diverse physical phenomena. Their unique properties have grabbed the attention of condensed matter community, making them one of the most extensively studied classes of materials. Layered transition metal chalcogenides, graphene, copper oxides, and iron pnictides are few examples of systems whose properties are dominated by their 2D structural units.

In this Dissertation, we have primarily focused on three different 2D layered materials: (i) graphene (ii) copper-oxide superconductors, and (iii) iron pnictide superconductors. Before we introduce the content of this Dissertation in detail, we will first introduce the materials and highlight the questions this Dissertation focuses on.

**Graphene:** A one atom thick, two-dimensional crystal made up of carbon atoms arranged in a hexagonal honeycomb formation. It was first studied theoretically by Wallace [1] in 1947 to calculate the band structure of graphite. He identified graphene as a zero band gap semiconductor and predicted extraordinarily high mean free path in the graphene sheets. Earlier theoretical work by Peierls [2, 3] and Landau [4, 5] had predicted that a strictly 2D crystal could not exist, as it is thermodynamically unstable. Contrary to this prediction, in 2004 Geim and Novoselov [6-8] used micromechanical cleavage techniques to extract a stable single layer of graphene, and experimentally confirmed the existence of massless Dirac fermions [7]. Graphene’s unique properties include extremely high mobility at room temperature (more than 200,000 cm²V⁻¹s⁻¹ [9]), ballistic transport, and high optical transparency [10]. Owing to its linear dispersion, graphene shows novel phenomena such as the room-temperature integer quantum Hall effect [11]. Although exfoliated flakes are great
for studying the intrinsic properties of graphene, for applications, large area samples, typically on a substrate, are usually required. The properties of graphene strongly depend on the underlying substrate, which can modify the local electronic environment and can also cause mechanical deformations. Understanding this substrate-film interaction is of great interest as it can alter the intrinsic properties of graphene. For optical studies, the complex refractive index and surface morphology of the substrate plays a significant role in the overall optical response of the thin film/substrate system. This Dissertation investigates these substrate induced effects in the magneto-optical properties of multilayer graphene grown on SiC substrates. In our investigations of graphene, we discovered a colossal enhancement of the magneto-optical Kerr effect in multilayer graphene/SiC, occurring around 120 meV (Chapter 3). A series of systematic measurements and analysis confirmed that this enhancement is linked with the refractive index of SiC substrate. We also discovered optical anisotropy induced in graphene by the surface morphology of the substrate, which makes the graphene act like a linear polarizer (Chapter 4).

Copper-oxide superconductors: Also known as cuprates, they were discovered in 1986 by Bednorz and Muller. The material they discovered, lanthanum barium copper oxide (La$_{2-x}$Ba$_x$CuO$_y$) had a critical temperature ($T_c$) of 35 K [12]. Later, by varying the type and proportion of other elements, new cuprates were discovered with $T_c$ reaching up to 133 K [13]. The cuprates are layered materials, in which the copper and oxygen atoms reside in a 2D plane. Atoms of other elements are sandwiched between the Cu-O$_2$ planes. Superconductivity takes place in the CuO$_2$ 2D planes, which is why cuprates are often referred to as 2D superconductors. The parent compounds of the cuprates are insulators; superconductivity appears when the parent compound is doped to become more metallic. By
varying the type and proportion of doping the $T_c$ of the system can be changed. Depending on the temperature and doping level, the system can be in one of the following phases 1) antiferromagnetic insulator 2) superconductor 3) strange metal 4) pseudogap, and 5) normal metal. In this Dissertation, we are primarily focused on the pseudogap phase which is a poorly understood metallic-phase with anomalous properties. It is seen in underdoped cuprates as a weak energy gap, which begins to appear at $T^*$ as the sample is cooled before the superconducting gap fully emerges at the lower temperature $T_c$. In the pseudogap phase, Cooper pairs start forming but without any macroscopic phase coherence. Scientists believe that understanding the enigmatic pseudogap phase is the key to understanding high $T_c$ superconductivity. It is still not clear whether the pseudogap phase is a distinct phase or just a precursor to superconductivity. For the pseudogap to be a distinct phase, it should have its own broken symmetries. In this Dissertation, we optically study this pseudogap phase by examining the presence of broken symmetries in the mid-infrared and visible photon energy ranges (Chapter 5).

**Iron-pnictide superconductors:** Discovered by Hosono and co-workers in 2009 [14], they form another class of high $T_c$ superconductors which have structural similarity with cuprates and superconductivity arising from 2D iron arsenic layers. The highest $T_c$ reported in these materials is about 56 K [15]. The presence of iron in a superconducting material was unexpected because of iron’s large magnetic moment which favors parallel alignment of spin, inducing ferromagnetism. Superconductivity, on the other hand, requires the formation of Cooper pairs which are pairs of electrons with opposite spin. The discovery of iron pnictide superconductors is significant because it has given another platform to understand high $T_c$ superconductivity. In this Dissertation, the primary question which we want to investigate is
the interaction of intrinsic superconductivity of iron-pnictides with extrinsic ferromagnetism from the unintentional presence of iron impurities. We investigate this interaction using Faraday and magnetization measurements in Chapter 6.

In this Dissertation, we use Faraday and Kerr spectroscopy to probe the 2D materials listed above. In these measurements, we observe the changes occurring in the polarization of light after it interacts with a medium. We report the discoveries made during our investigation, which includes novel optical effects such as colossal Kerr enhancement and optical anisotropy in epitaxial graphene. In addition to increasing our understanding of the electronic structure of two-dimensional materials, both of these optical effects have possible applications in future technologies. We also report the discovery of nematicity in high $T_c$ cuprate superconductors, which can help the condensed matter community better understand these materials that have perplexed scientists for over thirty years. We also report our studies on iron-based superconductors and show how Faraday measurements can be used to investigate competing phases and also to obtain information about the electronic structure of the material.

Chapter 2 discusses the theoretical and experimental aspects of Faraday and Kerr spectroscopy. We start by describing the concepts of magneto-polarimetry in the Faraday/Kerr geometries and the definitions of related quantities. We then discuss the origin of Faraday and Kerr signals in a system and link them to the presence of broken symmetries. The most common reason for observing magneto-optical effect is the breaking of time reversal symmetry, which is achieved either by applying an external magnetic field to align the magnetic moments in a sample or by spontaneous magnetization. Another mechanism that produces changes in the polarization of reflected/transmitted light is structural
asymmetry (e.g., helical structure in a DNA molecule). In this Chapter, we first discuss the situation of broken time-reversal symmetry, which induces circular birefringence and circular dichroism in the sample. We perform a model calculation of Faraday response assuming a Lorentzian absorption. Next, we proceed to discuss structural symmetry breaking which includes linear and circular asymmetries. We also discuss optical setups to distinguish between the two asymmetries. We end the theoretical section by discussing the application and significance of Faraday and Kerr effect measurements in condensed matter physics. We then describe in greater detail the experimental setup for performing polarization modulation spectroscopy. We present a brief overview of the optical setup and discuss the relevant equations for extracting Faraday and Kerr angles from raw data. We provide a brief description of the main individual components used in the experimental setup and their roles. We describe an in-house built cryogenic rotator which allows us to rotate a sample inside a cryostat under high vacuum and at cryogenic temperatures. This setup allows us to study linear symmetry breaking in samples. At the end of the Chapter, we discuss the issue of background noise arising from having an imperfect hole on the sample plate and our method of removing it.

Chapter 3 presents our first discovery, the dramatic enhancement of Kerr effect (polarization change in reflected light) in SiC substrates at around 120 meV. We describe the observation in multilayer graphene grown on SiC substrates where the Kerr response is enhanced by a factor of 68 near the reststrahlen band of SiC. We also observe a similar enhancement in bulk Al-doped SiC. By analyzing the thin film equation for the Kerr angle, we predict that the enhancement is not intrinsic to the film but is linked with the refractive index of the substrate passing through unity near 120 meV. We use this effect to engineer Kerr enhancement in films of Fe grown on SiC substrates. We validate the mechanism for
enhancement by measuring the complex refractive index ($\bar{n}$) of the SiC substrate in the mid-infrared. Finally, we make two predictions that should be readily observable: 1) an analogous Faraday enhancement for transmitted light in thin films grown on metamaterials with $\bar{n} = -1$ and 2) the enhancement of Kerr response at all wavelengths for a suspended film.

Chapter 4 presents our second discovery, the observation of optical anisotropy in epitaxial graphene. The Chapter starts with a brief introduction to the process of growing epitaxial graphene on SiC substrates and the formation of steps due to the sublimation of Si atoms from SiC substrate. Yakes et al. [16] have shown a dc conductance anisotropy in epitaxial graphene due to the differences between conductivity along the steps and normal to the steps. We observe the optical analog of this conductivity anisotropy by performing Kerr angle measurements at zero magnetic field at room temperature on epitaxial graphene. We model the effect using a wire-grid polarizer, which creates differences in amplitude and phase of reflected light with polarizations parallel and perpendicular to the wires. We conclude the Chapter by discussing the possible applications of this effect in optoelectronic devices.

Chapter 5 reports the discovery of linear dichroism (LD) and linear birefringence (LB) in cuprate high $T_c$ superconductors at room temperature in the mid-infrared and visible energy ranges. We review previous work done in the THz and near IR and describe why these experiments are essential to understanding the nature of the elusive pseudogap phase in cuprates. We then present our result where we observe Faraday rotation at zero magnetic field, from room temperature down to 10 K for samples over a broad range of doping levels. We show the presence of LD and LB signals for a set of samples grown at two different institutions. We also found that the LD and LB signals persist from the infrared to the visible energy ranges and from 10 K to room temperature. We experimentally rule out substrate
terraces as the mechanism for the LD/LB signals by making Faraday measurements on a metal film grown on a LSAO substrate. We think the results are intrinsic to the electronic states of cuprates and may be linked to the presence of stripes (nematicity) of the samples.

Chapter 6 describes our ongoing work on iron-based superconductors (IBSC). We provide a brief historical introduction to IBSC and describe its importance in understanding high \( T_c \) superconductivity. The samples are characterized using x-ray diffraction (XRD) and SQUID magnetometry. The magnetization measurements reveal the presence of ferromagnetism (possibly from iron impurities) along with superconductivity (intrinsic to the IBSC). We observe a Faraday signal that is hysteretic with respect to \( B \), and resembles the magnetization signal from a soft ferromagnet. Near zero magnetic field, the response is a step function while at higher fields, the response is linear with \( B \). We analyze the Faraday response by fitting a straight line to the linear part to determine the slope \( \theta_{F, \text{slope}} \) and also by measuring the zero-field step height \( \Delta \theta_f \). We then study the temperature and energy dependence of \( \theta_{F, \text{slope}} \) and \( \Delta \theta_f \) separately. We conclude by suggesting that \( \Delta \theta_f \) might be linked with extrinsic ferromagnetism and the linear response \( \theta_{F, \text{slope}} \) with intrinsic superconductivity.

In Chapter 7, we summarize the results of individual projects and discuss their importance and implications. We also discuss future directions of our research.
2  Polarization modulation spectroscopy

Magneto-polarimetry extends dc Hall measurements into the optical regime. These measurements are sensitive to the complex Faraday $\tilde{\theta}_F$ and Kerr $\tilde{\theta}_K$ angles, which are closely related to the complex Hall angle $\tilde{\theta}_H$ [17]. Faraday/Kerr spectroscopy is a sensitive tool to probe the magneto-electronic structure and symmetries in a system. In this Chapter, we describe the theoretical and experimental aspects of magneto-polarimetry. We first explain the Faraday and Kerr effects and then discuss the underlying mechanism producing the Faraday/Kerr response, which is due to symmetry breaking (e.g., time reversal, or structural/electronic symmetry breaking) in the system. Then we proceed to review the experimental setup used to study the changes in the polarization of infrared light after it gets reflected (polar Kerr effect) or transmitted (Faraday effect) from the sample. Many publications are dedicated to the experimental details and measurement process [18-21], so in this Chapter, we only discuss the major details of the magneto-optical setup and provide brief description of the individual components. We describe in greater detail a new in-house built cryogenic rotator developed to perform rotation of sample inside a cryostat at low temperature (10 K), high vacuum and in the presence of a magnetic field. We discuss the issue of background polarization signal in the rotator and our technique of eliminating those signals. In the end, we discuss the technique of extracting complex Faraday and Kerr angle from raw data using three lock-in amplifiers.
2.1 Faraday and Kerr effects

In 1845, Michael Faraday discovered that when linearly polarized light passes through a sample, which is placed inside a magnetic field, the plane of polarization of the transmitted light rotates [22]. A few years later in 1877, a similar experiment was also discovered in reflection by Rev. John Kerr [23] where he observed the polarization of reflected light coming from a polished pole of a permanent magnet. In both the Faraday and Kerr effects, the magnetic field \( \vec{B} \) was parallel to \( \vec{k} \) the propagation direction of light. The later discovery by Voigt in 1899 also found similar effects for in-plane magnetic fields (\( \vec{B} \perp \vec{k} \)). In this Dissertation, we will study a variety of samples using polar geometry (\( \vec{B} \parallel \vec{k} \)).

\[ \text{Rotation} \quad \text{Re}[\theta_{\text{Far/Kerr}}] \]

\[ \text{Ellipticity} \quad \text{Im}[\theta_{\text{Far/Kerr}}] \]

**Figure 2-1 Faraday and Kerr geometry.**
Linearly polarized light is incident on a sample, placed in an out of plane magnetic field. The reflected (Kerr) and transmitted (Faraday) light shows rotation and ellipticity.

Figure 2-1 shows the schematic for Faraday and Kerr effect measurements. The incident polarization is purely linear and is aligned vertically. After interacting with the
sample, which is placed in an out of plane external magnetic field \( H \), the transmitted/reflected beam acquires a new polarization. It is useful to treat the Faraday/Kerr angle as a complex number. The real part ( \( \text{Re}[\theta_r] \), \( \text{Re}[\theta_k] \) ) is related to the rotation of the polarization axis with respect to the incident polarization. The imaginary part ( \( \text{Im}[\theta_r] \), \( \text{Im}[\theta_k] \) ) is referred to as the ellipticity and is defined as the ratio of minor to major axes for the ellipse that is swept out by the oscillating electric field.

### 2.2 Time reversal symmetry breaking

One of the fundamental reasons for the origin of magneto-optical effect is the breaking of time-reversal symmetry, which can arise due to the application of an external magnetic field to align the magnetic-moments in a sample or due to the presence of spontaneous magnetization.

The polarization of light can be studied in either a circular or linear basis. In a circular basis, the incident linearly polarized light consists of equal parts of right (+) and left (-) circularly polarized light. The complex refractive index for the right and left circular polarization is given by \( \tilde{n}_+ = n_+ + ik_+ \), where \( n_+ \) (\( n_- \)) represents the real refractive index of right (left) circularly polarized light and \( k_+ \) (\( k_- \)) is the extinction coefficient of right (left) circularly polarized light. The refractive index gives information about the speed of light in the medium while the extinction coefficient is related to the absorption of light in the medium.

The presence of a magnetic field breaks the time reversal symmetry and changes the optical properties of the sample. It induces circular birefringence, which means that the refractive index for left and right circularly polarized light are no longer same (\( n_+ \neq n_- \)) inside
the medium. Because of circular birefringence, left and right circularly polarized beams travel at different speeds inside the sample and this leads to a phase shift between the two beams.

Due to this phase shift, the polarization of the transmitted light gets rotated, and we get a Faraday rotation signal. Figure 2-2 (b) shows the Faraday rotation obtained due to circular birefringence. The magnetic field can also induce circular dichroism ($k_+ \neq k_-$), where the extinction coefficients for right and left circularly polarized beams becomes different and the transmitted beam becomes elliptically polarized. Figure 2-2 (c) shows the ellipticity due to circular dichroism.

The expressions for the Faraday and Kerr angles are derived in M.H. Kim’s Dissertation and are as follows: For the Faraday angle (transmission), the expressions for $\text{Re}[\theta_F]$ and $\text{Im}[\theta_F]$ are given by:

\[
\text{Re}[\theta_F] = \frac{\omega d}{2c} (n_+ - n_-)
\]

\[
\text{Im}[\theta_F] = \frac{\omega d}{2c} (k_- - k_+)
\]

Where $\omega$ is the frequency of incident radiation, $d$ is the thickness of the sample and $c$ is the speed of light. From the above equations, it is clear that the Faraday rotation is associated to the difference in $n$ while Faraday ellipticity is associated to difference in $k$ between left and right circularly polarized light.
For the Kerr angle (reflection), the expressions for \( \text{Re}[\theta_k] \) and \( \text{Im}[\theta_k] \) are given by:

\[
\text{Re}[\theta_k] = \frac{1}{n^2 - 1} (k_- - k_+) \\
\text{Im}[\theta_k] = \frac{-1}{n^2 - 1} (n_- - n_+)
\] (3)

Unlike the expressions for Faraday rotation, the expressions for Kerr rotation and ellipticity depends on both \( n \) and \( k \).
Gaj and Kossut [24] have discussed in detail the origin of the Faraday rotation signal in semiconductors. We will follow their analysis here in this section. For a single Lorentzian optical transition line, the complex refractive index $\tilde{n}(\omega)$ is given by

$$\tilde{n}(\omega) = 1 + \frac{A}{\omega_0 - \omega - i\Gamma}$$  \hspace{1cm} (5)

Where $\omega_0$ is the resonance frequency, $\omega$ is the frequency of incident light, $2\Gamma$ is the full width at half maximum (FWHM) and $A$ is the amplitude. The refractive index $n$ and extinction coefficient $k$ are the real and imaginary parts of $\tilde{n}$ in Eqn. (5). As an example, the $n$ and $k$ for a Lorentzian absorption with parameters $\omega_0=1$, $A=1$, $\Gamma=0.01$ are shown in Figure 2-3(a). The line shape of the Faraday angle for such a Lorentzian absorption will depend on how the magnetic field influences the absorption parameters [25]. In general, it can affect (1) the resonance energy $\omega_0$ (2) the amplitude of the transition $A$ and (3) the linewidth of the transition $\Gamma$.

In the first situation when the magnetic field changes the resonance frequency $\omega_0$ (line position) for different circular polarizations, we get the Zeeman-type Faraday response. In the low B-field limit, it is given by

$$\frac{\theta_F(\omega)}{d} = \frac{\omega}{2c} \Delta \tilde{n} = \frac{\omega}{2c} \Delta \frac{d\tilde{n}(\omega)}{d\omega_0} = -\frac{\omega}{2c} \Delta \omega_0 A \frac{(\omega_0 - \omega)^2 - \Gamma^2}{(\omega_0 - \omega)^2 + \Gamma^2}.$$  \hspace{1cm} (6)

Where $\Delta \tilde{n}$ is the change in the complex index of refraction for left and right circular polarizations and $\Delta \omega_0$ is the difference in the resonance frequencies for the two polarizations. Figure 2-3(b) shows the Faraday angle for a Lorentzian line (for
\( \omega_0 = 1, \ A = 1, \ \Gamma = .01, \ c = 1, \ \Delta \omega_b = .001 \). The Faraday rotation is even and symmetrical about the resonance frequency while the ellipticity is odd and crosses zero at \( \omega_0 \).

**Figure 2-3 Faraday response from a single Lorentzian optical transition.**
Shows the complex refractive indices (a), Zeeman splitting (b), amplitude-type (c) and line-width type (d) Faraday response spectra.

The magnetic field can also influence the amplitude and/or the line width of the transitions. The expression for the amplitude-type Faraday response is given by
\[
\frac{\theta_\varepsilon(\omega)}{d} = \frac{\omega}{2c} \frac{\Delta^2}{\tilde{c}A} + \frac{\omega}{2c} \frac{\Delta^2}{(\omega_0 - \omega)^2 + \Gamma^2},
\]

(7)

Where \(\Delta A\) is the difference in the resonance amplitudes for left and right circularly polarized light. Similarly, the Faraday response for linewidth-type effect is

\[
\frac{\theta_\varepsilon(\omega)}{d} = \frac{\omega}{2c} \frac{\Delta^2}{\tilde{c}A} - \frac{\omega}{c} \frac{\Delta^2 \Gamma}{(\omega_0 - \omega)^2 + \Gamma^2},
\]

(8)

Where \(\Delta \Gamma\) is the difference in the resonance amplitudes for left and right circularly polarized light. Figure 2-3 (c) and (d) shows \(\theta_\varepsilon\) for amplitude-type and linewidth-type effects, respectively. The values of the parameters used to generate the plots are \(\omega_0 = 1, A = 1, \Gamma = .01, c = 1, \Delta A = 0.01, \Delta \Gamma = 0.001\). For both these effects, \(\text{Re}[\theta_\varepsilon]\) is odd and \(\text{Im}[\theta_\varepsilon]\) is even and peaks at \(\omega_0\).

An important feature of \(\theta_\varepsilon\) is that that we can measure the effects of symmetry breaking in an optical transition even when the probe energy is far from that transition. This makes probing symmetry breaking using the Faraday effect much easier since one does not need to precisely tune the probe energy to a narrow resonance.

### 2.3 Structural symmetry breaking

Structural asymmetries in a medium can also produce Faraday and Kerr signals. A chiral material such as a metal in an out of the plane magnetic field or a chiral molecule such as DNA will respond differently to the left and right circularly polarized light. The difference in magnitude and phase between the two polarizations will produce rotation and ellipticity as shown in Figure 2-4 (a and b) shows that the circular anisotropy signal is independent of the
sample orientation. Linear anisotropy generated due to uniaxial strain or nematicity can also produce polarization signals. In this situation, it is best to use the linear basis, the horizontal and vertical polarization. Figure 2-4 (c) shows how a material with linear anisotropy interacts differently to horizontal and vertical polarizations, and thereby generates a rotation and ellipticity signal. Unlike circular anisotropy, a linear anisotropic sample will produce polarization signal that depends on the sample orientation as shown in Figure 2-4 (d) . In samples where both linear and circular asymmetries are present, the polarization signal is offset due to the circular anisotropy and is modulated sinusoidally with sample orientation due to the linear anisotropy.

---

**Figure 2-4 Probing structural symmetry using polarized light.**
A sample with circular anisotropy (a) and linear anisotropy (c) and combination of both these symmetries (e).
2.4 Applications of Faraday and Kerr effect

The magneto-optical Faraday and Kerr effects are powerful and sensitive probes of the electronic band structure of materials. They have been applied to reveal fundamental material properties such as magnetic anisotropy [26, 27], electron spin polarization [28] and magnetic excitations [29]. Faraday and Kerr spectroscopy in the visible and infrared (IR) spectral ranges have been widely used to study metals [17], magnetic semiconductors [21, 30-33], superconductors [34-36], and more recently graphene [37, 38] and topological insulators [39]. These effects also play a vital role in modern technology, for example, Faraday isolators are commonly used to prevent unwanted feedback between devices in telecommunications. The magneto-optical Kerr effect (MOKE) also offers exciting possibilities for ultrahigh density data storage in magnetized media [40]. In addition to probing the electronic structure in novel materials, we can use the fundamental sensitivity to symmetry breaking that Faraday and Kerr measurements offers to investigate symmetries in materials.

2.5 Experimental setup

The experimental setup has been discussed extensively in earlier publications [18-21], so in this Section, we will only discuss the essential details of the setup. Figure 2-5 shows a schematic of the optical beam line. The gas lasers emit linearly polarized light which gets focused by Lens 1 on to the blades of an optical chopper. After that, the light reaches a Brewster’s reflector which purifies the vertical linear polarization by removing any undesired horizontal polarization component. The vertically polarized light then passes through the BaF2 window of the magneto-optical cryostat and interacts with the sample. The magneto-optical cryostat can produce an out of plane magnetic field of up to 7 T and can cool the sample down.
to 10 K. The polarization of the transmitted/reflected beam is measured by the optical detection system consisting of a PEM, a wire grid polarizer, an infrared detector (mercury cadmium telluride (MCT) or InSb) and three lock-in amplifiers.

2.6 Description of major optical components

In this Section, we overview the primary optical components used in the magneto-optical polarimetry system as shown in Figure 2-5. The major components of the magneto-optical system are 1) lasers, 2) optical chopper, 3) magneto-optical cryostat, 4) photoelastic modulator (PEM), 5) wire-grid polarizer, 6) infrared detector, and 7) lock-in amplifiers.

I. Lasers: Most of the experiments in this Dissertation uses infrared molecular gas lasers namely CO\textsubscript{2} (9-11 µm, 138-112 meV), CO (5-6 µm, 248-206 meV) and HeNe (3.4 µm, 365 meV). The discrete laser lines originate from transitions between various vibrational and rotational levels.

II. Optical chopper: Light from the laser gets focused on to the blades of the chopper. The blocking and unblocking of the incident beam creates a square wave modulation usually set to a frequency of \( \omega_0 = 2000 \) Hz. The chopper modulated beam yields the intensity of the laser, \( I_0 \), in the lock-in amplifier that is referenced to \( \omega_0 \).
Figure 2-5 Schematic of the magneto-optical polarimetry setup. The setup shows the optical setup to perform Faraday (transmission) measurements. Light from laser passes through optical elements and interacts with the sample, placed inside the cryostat. The transmitted light passes through a PEM and gets focused on the detector. Figure from Ref. [20].

III. **Magneto-optical cryostat:** Allows the sample to be cooled to 10 K using a cold finger contact. A split-coil superconducting magnet produces magnetic fields up to 7T, normal to the sample. The cryostat does not have cold windows which helps reduce additional background polarization signal, i.e., windows located where the magnetic field is large will produce their own Faraday signal. The outer windows are made up of BaF$_2$ and are mounted on 9 cm extension tubes to reduce background polarization signal which arises due to the presence of stray magnetic field. The cryostat can be translated horizontally.
and vertically with an accuracy of 25 µm which allows centering the beam on the sample.

IV. Photoelastic modulator (PEM): Two different PEMs have been used depending on the laser wavelengths. A ZnSe PEM (II/ZS50, Hinds Instruments) works for the CO\textsubscript{2}, CO and IR HeNe lasers while a fused silica (FS) PEM (I/FS50, Hinds Instruments) provides the polarization modulation for shorter wavelengths 0.17-2 µm. The PEM’s were set to a retardation of 0.383 wave for the measurements.

V. Polarizer: A polarizer (P\textsubscript{2}) at 45 degrees after the PEM was used to mix the horizontal and vertical components of the polarization. A BaF\textsubscript{2} holographic wire-grid polarizer was used with the ZnSe PEM while a Glan-Taylor polarizer was used with the FS PEM.

VI. Detector: Two different liquid nitrogen cooled detectors were used to span the measurement energy range. An MCT (HgCdTe) detector was used for the CO\textsubscript{2} laser while an indium antimonide (InSb) was used for the CO and visible wavelengths.

2.7 Cryogenic rotator

Many of the experiments described in this Dissertation require the ability to rotate a sample at low temperature (10 K) and under high vacuum. A special sample holder was constructed, enabling a 360 ° rotation of the sample around an axis parallel to the incident radiation direction [33]. The sample rotation is achieved by two Kevlar threads wrapped around the sample holder and around two brass cylinders that are placed at the top of the sample stick. The Kevlar threads pass through separate vacuum feed-throughs inside the
cryostat. The actual rotation of the sample holder was done by rotating the brass cylinders. A schematic illustration of the sample holder is shown.

![Schematic illustration of cryogenic sample rotator.](image)

**Figure 2-6 Schematic illustration of cryogenic sample rotator.** Cold finger (1), the sample holder (2) with a sample mounted (black square). The arrows indicate how the sample holder is rotated when Kevlar threads (3) which are wrapped around two brass cylinders are pulled/loosened. Figure from Tesarova et al. RSI (2012).

### 2.8 Issue of background noise

It was found that a background polarization signal was measured even when rotating a hole (without any sample) red in Figure 2-7. As seen, the background signal is sinusoidal with a maximum amplitude of 2 mrad and mimics a signal from a real sample hence it must be eliminated. It is to be noted that the FWHM of the laser beam at the center of the hole is
≈ 0.7 mm while the diameter of the hole is 7.1 mm. It was speculated that although the beam is Gaussian and has a diameter of only 1/10 th of the hole, the tails of the beam might still be extending to the inner wall of the sample holder. Boundary conditions will force the electric field vector to become normal to the edge of the circular aperture and hence will disturb the overall polarization of the beam. The is a good indication of the sensitivity of our technique and that a hole drilled in a copper plate may not be as round as one may have expected.

Figure 2-7 Polarization signal from rotating a hole. The signal from rotating an empty hole (red). The signal from the hole with an iris in front (black).

To eliminate this problem an iris (hole in a Delrin sheet) of diameter 4.5 mm was placed in front of the sample holder. The blocks the outer edges of the infrared beam which may have been interacting with the edge of the hole in the copper sample plate. On repeating the polarization test by rotating the empty copper sample plate with the iris in front, no appreciable polarization signal was recorded (black square in Figure 2-7). Figure 2-8 shows
the photographs of the rotating sample holder from front and back (with and without the plastic plate).

Figure 2-8 Photograph of the rotating sample holder. Photograph of the sample holder at different orientations (a) back side (b) front side with a sample mounted at 45° (c) front view of the sample holder with an additional white plastic plate in front (d) side view with the plastic plate in front.
2.9 Extracting complex Faraday/Kerr angles from raw data.

In this Section, we discuss the steps involved in extracting Faraday/Kerr angles from the raw data. Figure 2-9 shows the schematic of the optical setup. The optical chopper modulates the incident light at $\omega_0$ before it reaches the sample. The reflected/transmitted light acquires a small y-component of polarization after interacting with the sample. The PEM oscillates at a frequency $\omega_{PEM}$ along the y-axis at, thereby periodically shifting the phase of this y component with respect to the much stronger x component. This relative phase shift $\delta(t)$ is given by $\delta(t) = R_y \cos(\alpha_{PEM} t)$ where $R_y$ is the retardance of the PEM.

![Figure 2-9 Experimental setup for measuring $\dot{\theta}_F$ and $\dot{\theta}_K$](image)

The polarizer $P_2$ after the PEM set at 45 degrees to the $x$ axis mixes the $x$ and $y$ components of the polarization. The infrared detector records the intensity of the light. Three lock-in amplifiers measure the RMS voltage at $\omega_0$, $2\omega_{PEM}$ and $3\omega_{PEM}$. The voltages recorded
by the lock-in amplifiers are $I_0$, $I_{2\omega}$ and $I_{3\omega}$ respectively. The Faraday/Kerr angle are extracted using the following equations (derived in M. H Kim’s Dissertation):

$$\text{Re}(\theta_p) = \frac{I_{2\omega}}{I_0} \frac{\tan(\alpha_2)}{\tan(\alpha_2)} \left[ \frac{4J_2(R_d)^2}{4J_2(R_d) - 2J_0(R_d) I_{2\omega} I_0} \right]$$

$$\text{Im}(\theta_p) = \frac{\frac{I_{3\omega}}{I_0} \left[ 1 + 2J_0(R_d) \text{Re}(\theta_p) \tan(\alpha_2) \right]}{4J_3(R_d) \tan(\alpha_2)}$$

where $J_n$ are nth order Bessel functions and $\alpha_2$ is the angle, nominally $45^\circ$, at which polarizer $P_2$ is oriented with respect to vertical. The equations for $\text{Re}(\theta_p)$ and $\text{Im}(\theta_p)$ can be simplified by choosing $R_d$ to be 2.406 rad, which makes $J_0(2.406) \approx 0$. Further orienting $P_2$ at $45^\circ$ makes $\tan(\alpha_2) = 1$.

The expressions for $\text{Re}(\theta_p)$ and $\text{Im}(\theta_p)$ now reduces to,

$$\text{Re}(\theta_p) = \frac{1}{4J_2(R_d) I_0} I_{2\omega}$$

$$\text{Im}(\theta_p) = \frac{1}{4J_3(R_d) I_0} I_{3\omega}$$
3 Colossal Kerr enhancement in SiC Substrates

In this Chapter, we present our discovery of the colossal Kerr enhancement effect that we observe in SiC substrates. We observe polarization changes in reflected light that are enhanced by over a factor of 60 by the SiC substrate. The chapter begins with an overview of Kerr enhancement results observed by us and also briefly review the different ways in which other groups have tried enhancing the Kerr response and compare our approach with others. We then discuss the individual systems where we observe the effect, namely: 1) Multilayer graphene epitaxially grown on a c-face 4H-SiC substrate. 2) Al-doped bulk SiC and 3) Iron films deposited on 4H- SiC substrates. For each system, we describe the sample and discuss the experimental results. To understand the Kerr enhancement and for theoretical modelling, we need the optical constants of 4H-SiC which we obtain by reflectance measurements on the SiC substrate. Therefore, we next discuss the experimental setup and then describe the fitting process to extract the optical constants. We then present our theoretical models. Finally, we discuss the mechanism for Kerr enhancement and predict Kerr enhancement in suspended films along with Faraday enhancement in metamaterials. We conclude the chapter discussing the importance of the enhancement effect.

3.1 Overview of Kerr enhancement

In this Chapter we investigate the enhancement of the MOKE signal at infrared photon energies \(E_{ph} \sim 100\) meV, which is achieved in films when the substrate’s index of refraction \(\tilde{n}\) is near unity. While similar MOKE enhancements have been observed at visible photon energies for magneto-optical films that exhibit \(\tilde{n}=1\) [41], in the present work the enhancement relies upon the optical properties of the substrate rather than the film of
interest itself. As such, this greatly improves the flexibility of such an enhancement since it is usually possible to choose a substrate material independent of the film in order to optimize enhancement effects over a desired spectral range. Furthermore, the prior work on MOKE enhancement in the visible regime relies on the plasma edge induced by free-carriers to obtain $\tilde{n}=1$. It is well known that these free-carrier based systems exhibit optical losses that are especially strong at lower, infrared frequencies. However, in the present case, we exploit the optical properties of polar-dielectric substrate materials, such as SiC, that achieve similar results without the presence of free-carriers. In the polar-dielectric case, the $\tilde{n}=1$ enhancement condition is achieved through the collective oscillations of bound lattice charge that is driven by optical phonons. As such, the optical losses in polar-dielectric materials are much lower for these materials compared to free-carrier systems, which stems from the fact that optic phonon lifetimes are over an order of magnitude larger (~ few picoseconds) than the plasmon lifetimes (~ tens of femtoseconds)[42-44].

Large intrinsic Kerr effects have been observed in various systems such as in transition-metal and rare-earth compounds [45]. Mechanisms proposed to explain these large Kerr signals include inter/intraband transitions [46], plasma edge splitting [47] and plasma resonance of charge carriers [41]. While most of the earlier work has focused on enhancing the Kerr signal in visible wavelengths (1.5-3 eV) [41, 47, 48], the present work focuses on enhancement of Kerr signal at infrared energies (~100 meV). As mentioned in Chapter 1, magneto-optical Kerr effect (MOKE) measurements at these infrared energies are critical to understanding the electronic structure of wide variety of materials. However, MOKE signals are often weak. As such, the enhancement of infrared MOKE signals can play a critical role in improving such measurements. Furthermore, it was recently suggested that MOKE in graphene films may be exploited to produce fast, infrared polarization modulators that are
capable of supporting arbitrary waveforms [38]. It is expected that the maximum amplitude of polarization modulation that can be achieved in such a system will rely on the exploitation of substrate-mediated MOKE enhancement that is presented in this Chapter.

3.2 Multilayer graphene on 4H-SiC

In this Section, we will discuss the MOKE measurements on multilayer epitaxial graphene. We will discuss the sample details followed by the magnetic field and photon energy dependence of Kerr response.

3.2.1 Sample details

Multilayer graphene was epitaxially grown on a c-face 4H-SiC substrate. The substrate was etched in hydrogen and then annealed at 1600°C while maintaining a 10⁻⁴ mbar vacuum in a chemical vapor deposition system. The samples were grown at the Naval Research Lab (NRL). The details of the growth was reported in Jernigan et.al (Nano Letters 2009)[49].

3.2.2 Results

In the presence of an out-of-plane magnetic field $B$, the electronic levels in graphene condense into discrete Landau levels (LLs) [6]. For bilayer and multilayer graphene, the LL energies are proportional to $B$, whereas for a monolayer, the energies are proportional to $\sqrt{B}$. It is possible to optically excite electronic transitions between LLs when $E_{ph}$ matches the energy of an allowed transition between LLs (cyclotron resonance). The selection rules for such transitions allow for two energetically degenerate transitions (assuming electron-hole band symmetry) that are excited by opposite handedness of light. Owing to the fact that the MOKE signal is proportional to the difference between the complex, ac conductivity of left
(σ+) and right (σ−) circularly polarized light, these measurements provide a sensitive probe of chiral asymmetries that may exist between these two degenerate transitions. Earlier MOKE measurements on these epitaxially grown multilayer samples have identified over 18 different cyclotron resonances (CR) in a single 0-5 T field sweep [38]. This prior work was able to associate these CR features to distinct monolayers and multilayers with various stacking geometries and attributed the chiral asymmetry to the Pauli blocking of LLs.

Figure 3-1 shows the $B$ dependence of $\text{Re} [\theta_K]$ measured for multilayer graphene grown on a 4H-SiC substrate at $E_{ph}$ of 120.99, 121.27 and 133.60 meV. By determining the scaling behavior of CR features with $B$ for a range of $E_{ph}$, Ellis et al. (2013) has shown that the low-field features $|B| < 2 \, T$ follow $\sqrt{B}$ behavior and thus originate from interband LL transitions in monolayer graphene while the other CRs features at $|B| > 2 \, T$ scale linearly with $B$ and originate from multilayer graphene. As shown in Figure 3-1, the magnitude of $\text{Re} [\theta_K]$ shows a strong dependence on $E_{ph}$. This is evidenced by not only the magnitude of Kerr features that correspond to CR transitions, but also the linear background component of the signal that results from many high-order overlapping transitions that cannot be distinguished. In order to further explore this dependence on $E_{ph}$, we focus on the behavior of low field slopes, by performing a linear fit for the data in the range of $|B| \leq 1 T$. While the peak/dip CR Kerr features may also be used to explore this dependence, doing so is more challenging due to the fact that the lineshape of CR features evolves throughout this range of $E_{ph}$. At 133.60 meV (blue curve), the slope is nearly zero (-0.05 mrad/T) and the CR features are less than 5 mrad. However, for $E_{ph} = 120.99$ meV (black curve), we find an enhancement in the slope (35 mrad/T) and in the magnitude of the CR features. As shown in Figure 3-2 the largest
enhancement of slope occurs at 120.99 meV (black curve), where the slope of $\text{Re}[\theta_K]$ at its peak is 68 times larger than its corresponding values at higher energies. It is interesting to observe such a dramatic change in slope and magnitude of features for energies differing by less than a few tenths on an meV.

Figure 3-1 Mid-infrared MOKE measurements of multilayer graphene
Magnetic field dependence of Kerr rotation at various photon energies. The individual lines shows the various features originating from CR.

In addition to the $E_{\text{ph}}$ dependent enhancement of the Kerr angle, the lineshape of CR Kerr features also shows an interesting dependence on $E_{\text{ph}}$. In Figure 3-1 the high-field features are denoted as $\alpha (\alpha')$, $\beta (\beta')$ and $\epsilon (\epsilon')$ for $+B (-B)$ for $E_{\text{ph}} = 120.99, 121.27$, and
133.60 meV, respectively. Arrows indicate the sign of the features where up arrows represents a dip and down arrows represents a peak in the Re[θk] signal.

Figure 3-2 Colossal Kerr effect in multilayer graphene. Energy dependence of low field slope. The slope shows a maximum around 121 meV.

The high-field features always exhibit odd symmetry θk(−B) = −θk(B), as indicated by the lineshape inversion that occurs between correlated CR features in the −B and +B regimes (also indicated by the orientation of arrows). This is consistent with the expected MOKE signal, which is proportional to the off-diagonal conductivity σxy and therefore should have an odd symmetry with respect to B [50]. Interestingly, this is not the case for the observed CR
features from monolayer graphene that occur at low-$B$. For $E_{ph} = 121.27$ meV the typical odd symmetry is maintained for low-$B$ CR features, as indicated by the arrow orientation for peaks labeled as $\mathcal{D}$ ($\mathcal{D}'$) in the $+B$ ($-B$) regime. However, for $E_{ph} = 120.99$ meV, an even symmetry $\theta_k(-B) = \theta_k(B)$ is observed for low-$B$ CR features, as indicated by the arrow orientation for peaks labeled as $\mathcal{E}$ ($\mathcal{E}'$) in the $+B$ ($-B$) regime. This behavior is completely unexpected for MOKE signals. Interestingly, for all spectra measured below and above $E_{ph} = 120.99$ meV, the low-$B$ Kerr signal exhibits CR features with odd and even symmetries, respectively. Furthermore, the crossover from odd to even symmetry features is bounded by the same photon energy ($E_{ph} = 120.99$ meV) that results in the maximum Kerr signal. While it is tempting to conclude that both the enhancement and symmetry effects are related to the same mechanism (i.e., $\hat{n}$ passing through unity, as discussed later), these symmetry effects are not captured by our modeling. As such, the origins of such a symmetry change is not understood and remains to be investigated in future studies.

3.3 Al-doped bulk SiC

The Kerr enhancement observed in multilayer graphene was also observed in a bulk system, namely Al-doped SiC.

3.3.1 Sample details

Al-doped bulk SiC was prepared in an inductively heated furnace by using a physical vapor transport method. The details of the growth are reported elsewhere [51, 52]. The samples were grown in Institute of physics (IOP), China. The samples show ferromagnetic response from room temperature and down to 4 K. The dc magnetization was measured at UB using Dr. Zeng’s PPMS system. Figure 3-3 shows the measured in-plane magnetization at
15 K and 200 K. At 15 K, nonlinear ferromagnetic magnetization response is seen. At 200 K, the sample shows nonlinear response till 5000 Oe, after that diamagnetic response from the sample holder starts dominating.

Figure 3-3 Magnetization measurements of Al-doped SiC.
(a) At 15 K and (b) at 200 K.

3.3.2 Results

Figure 3-4(a) shows the $B$ dependence of $\text{Im}[\theta_x]$ for bulk Al-doped SiC with $E_{ph} = 117, 121$ and 224 meV. Although this material shows a ferromagnetic response in its dc magnetization [53], both $\text{Re}[\theta_x]$ and $\text{Im}[\theta_x]$ are linear in $B$ which could be due to the fact that magneto-optical measurements, namely Faraday and Kerr, depend not only on magnetization, but also are functions of photon energy and temperature. The dc magnetization, measured using a SQUID, measures the total magnetization of the sample. In the infrared, where the photon energies can be similar to magnetization energies, the MOKE and magnetization signals can look quite different [21, 31, 54]. On the other hand, in the
visible energy range (where the photon energy is much greater than the magnetization energy), the magnetization and MOKE signals typically behave very similarly.

Figure 3-4 Kerr response of Al doped SiC.
(a) Magnetic field dependence of Al-doped SiC. (b) Energy dependence of slope of $\text{Re}[\theta_K]$ and $\text{Im}[\theta_K]$.

In Figure 3-4(a) as we change $E_{ph}$ the slope of $\text{Im}[\theta_K]$ changes dramatically near 120 meV. At 121 meV (green) the slope is negative. When tuned to 117 meV (red), the magnitude of the slope increases and also the slope changes its sign and becomes positive. At 224 meV (blue) the slope approaches zero and the ellipticity signal is weak. Figure 3-4(b) shows the slope of $\theta_K$ as a function of $E_{ph}$ at 10 K for the Al–doped SiC sample. A strong energy dependence can be seen for both $\text{Re}[\theta_K]$ and $\text{Im}[\theta_K]$. The sign of slope for both $\text{Re}[\theta_K]$ and $\text{Im}[\theta_K]$ changes very sharply as the photon energy crosses 120 meV demonstrating Kerr enhancement in this bulk system.
3.4 Fe film on SiC

To test these effects more systematically, we deposited several iron control films on SiC and GaAs. Iron was chosen since its ferromagnetic behavior produces two important advantages: 1) the ferromagnetism enhances the applied magnetic field, producing robust Kerr signals and 2) the hysteretic behavior of the Kerr signal in iron allows the nonlinear contributions from the iron film to be easily separated from linear background signals coming from the substrate and cryostat windows.

3.4.1 Sample details

Iron films of thickness 3nm and 10 nm were sputtered onto a semi-insulating 4H-SiC substrate. The plasma was turned on using 50W and the film was sputtered under 30mTorr vacuum. These test films were grown by Dr. Zeng’s group at UB. Since iron gets easily oxidized, the sample was kept in a dry nitrogen container. However, the possibility of oxidation cannot be ruled out.

3.4.2 Results

Figure 3-5(a) shows the results of Kerr measurements performed on a 3.2 nm thick iron film on 4H-SiC. The ferromagnetic iron shows a hysteretic and non-linear Kerr angle response with respect to $B$, saturating near 2 T. For this sample, the Kerr enhancement can determined by measuring the $E_{ph}$ dependence of $\Delta \text{Im}[\theta_k]$ (vertical arrow in Figure 3-5(a)), where the latter is the difference between the $B = 0$ intercepts of fits to the high-field signal where saturation occurs (indicated by dashed guidelines in Figure 3-5(a)). An enhancement of $\Delta \text{Im}[\theta_k]$ is clearly seen when comparing the response between 114 meV and 120 meV.
Figure 3-5 Kerr angle measurements of iron film.
In a) Magnetic field dependence of $\text{Im}[\theta_K]$ at 120 and 114 meV b) Energy dependence of $\Delta\text{Im}[\theta_K]$ for iron film of 3nm and 10 nm thickness on SiC and GaAs substrate.

Figure 3-5(b) shows the film thickness and substrate dependence of the Kerr enhancement for iron thin films. For a 10 nm Fe film on GaAs (red curve), $\Delta\text{Im}\theta_K$ exhibits little energy dependence. When the same thickness film is deposited on SiC (black curve), $\Delta\text{Im}\theta_K$ is a factor of 5 larger over the entire energy range. This shows the strong role of the substrate in enhancing the Kerr signal. For an even thinner, 3 nm thick Fe film on SiC (blue) $\Delta\text{Im}\theta_K$ is further enhanced by more than a factor of 2, with the peak value near 121 meV.

3.5 Measuring the refractive index of 4H-SiC

In order to confirm that the optical properties of SiC are critical to the Ker enhancement, we need to measure the complex index of refraction $\tilde{n}$ of SiC itself. The near
normal incident reflectance spectrum of 4H-SiC is measured using a Fourier Transform Infrared spectrometer (FTIR) at room temperature. Subsequently, the complex dielectric functions \( \tilde{\varepsilon}(\omega) \) and complex refractive index \( \tilde{n} \) of 4H-SiC are calculated by modeling the infrared reflectance spectrum.

### 3.5.1 Experimental setup: Reflectance measurements using FTIR

The near-normal-incidence, infrared reflectance spectrum of 4H-SiC was measured at room temperature using a Bruker Vertex 70 Fourier transform infrared (FTIR) spectrometer. A KBr beam splitter and a HgCdTe (mercury cadmium telluride, MCT) detector are used to measure the spectrum from 400 - 7000 cm\(^{-1}\).

Figure 3-6 shows the experimental setup. Broadband light from a globar exits the external port of the FTIR spectrometer and gets focussed by the lens, with a focal length of 10 inches. A similar lens \( L_2 \) collimates the beam. Mirror \( M_1 \) directs the collimated beam towards the sample holder. The sample holder has two chamfered holes of the same size, and the sample holder is painted black. The sample holder is mounted on a translation stage to align the sample/reference with the FTIR probe beam. The sample and a gold reference mirror are attached to the back side of the sample holder. Light reflected off the sample is focused by lens \( L_3 \) towards the MCT detector. The angle of incidence was around 5°. A gold mirror is used as a reference to normalize the reflectance spectra. The signal from the detector is fed back into the FTIR processor. The internal software, OPUS determines the reflectance spectra.
3.5.2 Drude-Lorentz oscillator model

To model the infrared properties of polar molecules such as SiC, the Drude-Lorentz (DL) oscillator model works very well. In this model, the lattice ions are approximated as simple harmonic oscillators subject to the driving force of the incident light.

For normal incidence, the reflectance $R$ is given by

$$R = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2}$$  \hspace{1cm} (13)

where $n$ and $k$ are the index of refraction and extinction coefficient, respectively.
The infrared reflectance spectrum of 4H-SiC is fitted with a frequency-dependent DL dielectric function [55]

\[
\tilde{\varepsilon}(\omega) = \varepsilon_\infty + \frac{\omega_p^2}{\omega_{\text{TO}}^2 - \omega^2 - i\gamma\omega} \quad (14)
\]

where \( \tilde{\varepsilon}(\omega) \) is the complex DL dielectric function, \( \omega \) is the frequency of incident light; \( \varepsilon_\infty \) represents the high-frequency dielectric constant; \( \omega_p \) is the plasma frequency of lattice vibration; \( \omega_{\text{TO}} \) is the frequency of the transverse (TO) optical phonon and \( \gamma \) is the scattering rate (damping) of the Lorentz oscillator. For this model, only the TO phonon frequency is required since the frequency of longitudinal optical phonon, \( \omega_{\text{LO}} \) can be determined by the Lyddane–Sachs–Teller relationship [55, 56]

\[
\omega_{\text{LO}}^2 = \omega_{\text{TO}}^2 + \frac{\omega_p^2}{\varepsilon_\infty} \quad (15)
\]

From the best fit of the reflectance data, the real and imaginary parts of dielectric function namely, \( \varepsilon_1 \) and \( \varepsilon_2 \) respectively, are obtained. The coefficients \( n \) and \( k \) are related to the complex dielectric function \( \varepsilon(\omega) \) by,

\[
\tilde{\varepsilon}(\omega) = \varepsilon_1 + i\varepsilon_2 = \bar{n}^2 = (n + ik)^2 \quad (16)
\]

On simplification,

\[
\varepsilon_1 = n^2 - k^2 \\
\varepsilon_2 = 2nk \quad (17)
\]
$n$ and $k$ can be written in terms of $\varepsilon_1$ and $\varepsilon_2$ as,

\begin{align*}
n &= \sqrt{\frac{\varepsilon_1^2 + \varepsilon_2^2 + \varepsilon_1}{2}} \\
k &= \sqrt{\frac{\varepsilon_1^2 + \varepsilon_2^2 - \varepsilon_1}{2}}
\end{align*}

(18)

### 3.5.3 Determination of optical constants

The measured IR reflectance (red squares) of 4H-SiC at room temperature as a function of $E_{ph}$ is shown in Figure 3-7. The incidence angle was close to normal. The reflectance spectrum was obtained by dividing the spectrum of 4H-SiC with the spectrum of a gold mirror under identical conditions. The high reflectance region between 100-120 meV is the reststrahlen band, where reflectance is close to 100% for a polar dielectric such as SiC. At 124 meV, the reflectance drops to its minimum and approaches zero. Later analysis will show that at that minimum reflectance point, the refractive index of 4H-SiC becomes 1 and we get the colossal Kerr enhancement effect. The solid black curve is the calculated fit using the DL dielectric function. The best-fit parameters are $\varepsilon_\infty = 6.74$, $\omega_p = 1441.56$ cm$^{-1}$, $\omega_{TO} = 797.55$ cm$^{-1}$ (98.8 meV) and $\gamma = 5.03$ cm$^{-1}$. Plugging the best-fit parameters into the equation (15), $\omega_{LO}$ comes out to be 972 cm$^{-1}$ (120.5 meV).
Figure 3-7 Reflectance spectra of 4H-SiC.
The reflectance spectra of 4H-SiC was measured using a FTIR at near normal incidence at room temperature.

Figure 3-8 shows the $E_{ph}$ dependence of the dielectric function $\varepsilon(\omega)$ evaluated using the fit-determined parameters. The real ($\varepsilon_1$) and imaginary ($\varepsilon_2$) parts are shown in blue and red curves, respectively. $\varepsilon_2$ is a sharply peaked function of energy with a maximum value at $E_{TO}$ while $\varepsilon_1$ crosses zero at $E_{LO}$. The real part $\varepsilon_1$ is large and positive below TO phonon energy, and it has a negative value at slightly higher energy than TO phonon. Between $E_{TO}$ and $E_{LO}$, $\varepsilon_1$ is negative and behaves like a metal producing high reflectance. At 124 meV $\varepsilon_1$ is unity.
Figure 3-8 Dielectric functions of 4H-SiC.
Dielectric functions obtained using best-fit parameters. The TO and LO phonons are shown at 98.8 meV and 120.5 meV respectively. The inset shows the position where $\varepsilon_1$ crosses zero (the position of LO phonon).
The frequency and energy dependence of $n$ and $k$ are shown in Figure 3-9. Approaching $E_{\text{TO}}$ from the low energy side, both $n$ and $k$ grow and attain peak value at $E_{\text{TO}}$, where both the indices have the same magnitude. Inside the reststrahlen band both $n$ and $k$ fall in magnitude as energy increases. The real part of the index, $n$, however, falls sharply by over two orders of magnitude, passing through a minimum around 115 meV before steeply rising again close to $E_{\text{LO}}$. The imaginary part of the index, $k$, on the other hand, falls more slowly with energy and always maintains a value greater than 1. The decrease in $n$ with increasing energy, inside the reststrahlen band is the well-known anomalous dispersion.
exhibited by the refractive index whenever it crosses the region of strong absorption. At photon energies above the reststrahlen band, in the transparent regime, $n$ flattens out attaining a nearly constant value, while $k$ keeps decreasing.

To explain the Kerr enhancement, we will only focus on the values of $n$ and $k$ over the measurement range. In the 100-180 meV range, $n$ passes through unity at two points, first at 100 meV with $k = 9.3$ and second at 124 meV with $k = 0.0371$. As will be seen later, the observed Kerr enhancement occurs only when the value of $\tilde{n}$ is unity (i.e., $n=1$ and $k \to 0$), which is satisfied only at 124 meV.

### 3.6 Theoretical modeling of complex Kerr angle

In this Section, we will model the Kerr response for all the three systems. We compare the theoretical Kerr angle with the experimental data.

#### 3.6.1 Kerr angle for graphene on SiC

The theoretical modeling of Kerr response for graphene was done using the thin film formalism. Kim et al.[21] have derived simplified analytical expressions for the complex Kerr angle for a thin film on a substrate using the Fresnel’s equations. The model takes into account multiple reflections within the film and the substrate. For a thin film of thickness $d$ deposited on a thick substrate of complex refractive index $\tilde{n}$, the complex Kerr angle is given by:

$$
\tan(\vartheta_k) = \left( \frac{-2}{Z_0 d} \right) \left( \frac{\tilde{\sigma}_{xy}}{\tilde{\sigma}_{xx}} \right) \left( \left( 1 + \frac{1}{Z \tilde{\sigma}_{xx}} \right) \left( 1 + \frac{1}{Z \tilde{\sigma}_{xx}} \right) \right)^{-1}
$$

(19)
where \( Z = \frac{Z_0 d}{\hat{n}} \pm 1 \), \( Z_0 = 376.7 \Omega \) is the vacuum impedance, \( \tilde{\sigma}_{xx} \) is the complex longitudinal conductivity and \( \tilde{\sigma}_{xy} \) is the complex transverse (Hall) conductivity. Gusynin et al. [57] have derived the analytical expressions for the complex conductivities \( \sigma_{xx}(\omega) \) and \( \sigma_{xy}(\omega) \). These expressions are given by

\[
\sigma_{xy}(E_{ph}) = -\frac{1000^2 v_f^2 |eB|}{\pi} \times \sum_{n=0}^{\infty} \left[ \left| \left[ n_f(\Delta E_{N=1}) - n_f(\Delta E_{N=1,J=1}) \right] \right| - \left| \left[ n_f(-\Delta E_{N=1,J=1}) - n_f(-\Delta E_{N=1,J=1}) \right] \right| \right]
\times \left[ \frac{1}{(\Delta E_{N=1,J=1} - \Delta E_{N=1,J=1})^2 - (E_{ph} + 2i\Gamma)^2} \right] \left[ \frac{1}{(\Delta E_{N=1,J=1} - \Delta E_{N=1,J=1})^2 - (E_{ph} + 2i\Gamma)^2} \right]
\]

and

\[
\sigma_{xx}(E_{ph}) = \frac{1000^2 v_f^2 |eB| E_{ph} + 2i\Gamma}{\pi i} \times \sum_{n=1}^{\infty} \left[ \left| \left[ n_f(\Delta E_{N=1,J=1}) - n_f(\Delta E_{N=1,J=1}) \right] \right| + \left| \left[ n_f(-\Delta E_{N=1,J=1}) - n_f(-\Delta E_{N=1,J=1}) \right] \right| \right]
\times \left[ \frac{1}{(\Delta E_{N=1,J=1} - \Delta E_{N=1,J=1})^2 - (E_{ph} + 2i\Gamma)^2} \right] \left[ \frac{1}{(\Delta E_{N=1,J=1} - \Delta E_{N=1,J=1})^2 - (E_{ph} + 2i\Gamma)^2} \right]
\]

Where:

- \( v_f \) is the Fermi velocity in m/s.
- \( B \) is the magnetic field in T.
- \( E_{ph} \) is the photon energy in meV.
- \( \Gamma \) is the disorder parameter in meV which broadens the Landau Levels.
- \( n_f \) is the Fermi occupation function that is given by
\[ n_f(E_{ph}) = \frac{1}{1 + e^{(E_{ph} - \mu)/1000eK_B T}} \]

- \( \mu \) is the chemical potential in meV.
- \( \Delta E_{N\rightarrow 1,j} \) is the LL transition energy, given by \( \Delta E_{N\rightarrow 1,j} = E_{N\rightarrow 1,j} - E_{N\rightarrow 1,j} \)

For modelling the Kerr response of graphene the following values were used,

\( v_f = 1.036 \times 10^6 \) m/s, \( \mu = 50 \) meV, \( T = 10 \) K, \( \gamma = 10 \) meV.

---

**Figure 3-10** Calculated Kerr response of monolayer graphene on 4H-SiC. Enhancement of \( \text{Re}[\theta_K] \) and \( \text{Im}[\theta_K] \) seen at 124 meV.
The complex conductivities used in Eq. (19) are 3D conductivities. The 2D formulation of Eq. (19) can be made by substituting $\sigma_{ij} = \sigma_{ij}^{2D} / d$, thereafter making Eq. (19) independent of $d$.

Figure 3-10 shows the calculated Kerr angle slope for a monolayer graphene on SiC. The sharp enhancement of slope is seen for both Re[$\theta_k$] and Im[$\theta_k$]. The overall lineshapes of the curves are in excellent agreement with the measurements. The calculated peak and the measured peak differ by 2.7 meV and the measured line shapes are broader than in the calculation. This may be attributed to the fact that the calculation is only true for a single monolayer. However, in reality, the epitaxially grown graphene sample consists of dozens of layers on the substrate. Also, the calculation assumes graphene to be directly placed on substrate while the graphene under measurement may be separated from the substrate by many graphene layers. Furthermore, the optical constants ($n$ and $k$) used in the theoretical model for the three samples were obtained at room temperature whereas all the Kerr measurements were performed at 10 K. The temperature dependence of the optical constants has not been taken into account. All these factors complicate the system, and alter the optical properties of the system that may cause a broadening and spectral shift of the Kerr enhancement. Further, the model cannot explain the existence of even symmetry features seen for $|B| < 2T$ for 120.99 meV Figure 3-1 ($\mathcal{D}$ ($\mathcal{D}^\dagger$)). Since the MOKE signal is proportional to $\sigma_{xy}$, which is fundamentally odd in symmetry with respect to magnetic field, these even symmetry features are unexpected and are not understood at this moment.
3.6.2 Kerr angle for iron film on 4H-SiC

A simple Drude model is used to calculate the MIR Kerr angle response of an iron film on SiC. The ac conductivities are given by \( \sigma_{xx}(\omega) = \frac{\sigma_0}{1 - i\omega\tau} \) and \( \sigma_{xy}(\omega) = \frac{\sigma_0\omega\tau}{(1 - i\omega\tau)^2} \), where the dc conductivity \( \sigma_0 = \frac{Ne^2\tau}{m^*} \), cyclotron frequency \( \omega_c = \frac{eB}{mc} \), \( N \) is the electron density, \( e \) is the electronic charge, \( \tau \) is the scattering time, \( m^* \) is the electron effective mass and \( B \) is the magnetic field which is treated as a fitting parameter. The values from Refs.[58, 59] used for the different parameters are \( N = 17 \times 10^{22} \text{ cm}^{-3} \), \( \tau = 1.11 \times 10^{-15} \text{ s} \), \( m^* = 2.3 \times m_e \). For \( B = 400 \text{ T} \), the calculated Kerr angle matches the experimental value. This value is reasonable for a ferromagnetic sample where the effective magnetic field, as a result of exchange interaction between the electrons are of the order of 100 to 1000 T [60, 61].

Figure 3-11 shows the calculated Kerr angle for 3 nm and 10 nm Fe films on SiC as well as for a 10 nm Fe film on GaAs. The optical constants for GaAs were taken from Refs.[62, 63]. The Fe films on SiC shows enhancement at 120 meV while that on GaAs shows enhancement at 40 meV, which is where the reststrahlen band of GaAs is located. With the increase of film thickness, the Kerr angle decreases in magnitude. This behavior is expected since increasing film thicknesses decrease the amount of light that interacts with the substrate, which is responsible for the enhancement effect.
3.6.3 Kerr angle for bulk Al doped SiC.

For a bulk material, the Kerr angle equations are derived by Reim and Schoenes [45]. The real and imaginary parts of the Kerr angle for Al-doped SiC is modeled using the equations for a bulk sample given by

\[
\text{Re}[\theta_k] = \frac{4\pi}{\omega} \left[ \frac{B\sigma_{1xy} + A\sigma_{2xy}}{A^2 + B^2} \right] \tag{22}
\]

\[
\text{Im}[\theta_k] = \frac{4\pi}{\omega} \left[ \frac{A\sigma_{1xy} - B\sigma_{2xy}}{A^2 + B^2} \right] \tag{23}
\]
where \( A = n^3 - 3nk^2 - n \), \( B = -k^3 + 3n^2k - k \) and \( \sigma_{xy} = \sigma_{3xy} + i \sigma_{2xy} \).

Figure 3-12 Modelled Kerr response for bulk Al doped SiC.
Sharp enhancement peak is seen at 120 meV.

To make a quantitative theoretical model of the Kerr angle for bulk Al-doped SiC, the value of \( \sigma_{xy}(\omega) \) is needed over the measured frequency range. For our calculation, we assume \( \sigma_{xy} \) to be constant (\( \sigma_{xy} = 1 + i \)) over our energy range for two reasons. First, we do not have sufficient information to calculate \( \sigma_{xy}(\omega) \) over our energy range, and secondly, we want to show that the enhancement effect seen in this present work can be theoretically predicted to
occur at the correct $E_{ph}$ even if we assume a constant $\sigma_{xy}$ and only takes into account the
dispersion of $n$ and $k$. Figure 3-12 shows the calculated $\text{Re}[\theta_k]$ and $\text{Im}[\theta_k]$ for bulk
ferromagnetic SiC using Eqns. 10 and 11. A sharp peak at 124 meV is seen for both $\text{Re}[\theta_k]$ and
$\text{Im}[\theta_k]$. Away from the resonance peak, both $\text{Re}[\theta_k]$ and $\text{Im}[\theta_k]$ are close to zero.

3.7 Mechanism for colossal Kerr enhancement

The Kerr angle enhancement observed in 4H-SiC arises when passes through unity at
the reststrahlen band edge near the LO phonon. For 4H-SiC, this occurs near $E_{ph} \approx 124$ meV
as shown in Figure 3-9. The expression for the Kerr angle Eq. (1-3) for both thin film and bulk
becomes large when $\bar{n} \rightarrow 1$ ($n = 1$ and $k \rightarrow 0$). Looking in more detail at the thin film equation
(Eq. 1), the term $Z = \frac{Z_0 d}{\bar{n} - 1}$, becomes large when $n = 1$ and $k \rightarrow 0$. This results in the colossal
enhancement of the magnitude of Kerr angle slope (in graphene and Al-doped SiC), zero-field
Kerr intercepts (in Fe-film) and also the magnitude of Kerr features due to CR absorption in
graphene.

The colossal enhancement of Kerr effect observed in this work is the long wavelength
infrared analog of the MOKE enhancement in the visible spectral range, that occurs for metals
near the plasma edge [19]. Although there are no free carriers in the SiC, and therefore, there
is no plasma edge, the presence of the reststrahlen band causes SiC to behave like a metal at
optical frequencies as indicated by the high reflectance and the negative $\varepsilon_1$. This metal-like
behavior is derived from the polar nature of the SiC lattice, where there is a charge imbalance
between the Si and C atoms. When the SiC is illuminated with a frequency between the LO
and TO phonon frequencies, the lattice and bound lattice charges are set into motion. It is
this oscillating bound charge that gives rise to the metallic nature of the reststrahlen band. Similar to the metals in Ref. [19] we have a $\epsilon_1 < 0$, which gives rise to an effective plasma edge (without free carriers) near the LO phonon. However, since Ref. [19] is looking at metals the Kerr enhancement is in the few eV energy range, whereas our phonon derived enhancement is in the hundred meV range due to the phonon frequencies. Furthermore, Ref. [41] focuses on investigating Kerr enhancement phenomena that arise when the magneto-optical material itself satisfies the condition $\epsilon_1 \sim 1$. This is in contrast to our work, where we also observe enhancement in films when the substrate material satisfies the Kerr enhancement condition $\epsilon_1 \sim 1$, as demonstrated in this work for both the graphene and iron films.

The Kerr enhancement in 4H-SiC at 124 meV can also be understood from the definition of the Kerr angle, $\tan \theta_K = r_{xy} / r_{xx}$, where $r_{xy}$ is the transverse reflection coefficient and $r_{xx}$ is the diagonal reflection coefficient. An enhancement in $\theta_K$ is expected whenever $r_{xx}$ has a local minimum, assuming that $r_{xy}$ is unaffected by the substrate optical properties and that the magneto-optical film is adequately transparent. Figure 3-7 shows that at 124 meV, the reflectance of 4H-SiC approaches zero and it is also where we observe the enhancement. This explanation is easier to understand for the case of bulk Al-doped 6H-SiC, where one does not have to account for contributions to the reflectance from both the film and the substrate. For thin films where the reflectance is dominated by the properties of the substrate, this argument should also be valid.

The calculations of complex $\bar{\theta}_K$ for the three different samples show enhancement and produce line shapes that are in good semi-quantitative agreement with the measurements, with enhancements occurring at the expected energies.
3.8 Faraday and Kerr enhancement in metamaterials

While our analysis has focused on the reflectance case, similar effects should be observed for light that is transmitted through the sample. The Faraday angle for a thin film on a thick substrate is given by [21]

$$\tan(\tilde{\theta}_F) = \left( \frac{\hat{\sigma}_{xy}}{\hat{\sigma}_{xx}} \right) \left[ 1 + \frac{1}{Z_x \hat{\sigma}_{xx}} \right]^{-1}$$  \hspace{1cm} (24)

The term $Z_x = \frac{Z_0 d}{\bar{n} + 1}$ in the denominator will produce an enhancement of Faraday angle for a metamaterial whose refractive index $n$ passes through -1.

**Figure 3-13** Faraday enhancement for a iron film on metamaterial substrate.
Figure 3-13 shows the enhanced Faraday response of a layer of iron on a metamaterial whose index passes through \( n = -1, k \to 0 \). Such a metamaterial will also display Kerr enhancement at \( n = -1, k \to 0 \) due to the presence of \( Z_\lambda \) factor in the Kerr angle expression in Eqn. (19). Figure 3-14 shows the enhanced Kerr response for \( n = \pm 1, k \to 0 \).

![Kerr angle enhancement for thin film on metamaterial](image)

**Figure 3-14 Kerr angle enhancement for thin film on metamaterial.**
Calculated Kerr response for a iron film on a metamaterial substrate. The Kerr enhancement occurs at both \( n = \pm 1 \).

Figure 3-15 a) shows the measured refractive index by Doling et.al.[2006][64], with negative refractive index from 1.3 - 1.6 \( \mu m \). The refractive index \( n \) passes through -1 at \( \lambda_1 = 1.403 \mu m \) and \( \lambda_2 = 1.478 \mu m \). The optical constants \( (n,k) \) at \( \lambda_1 \) and \( \lambda_2 \) are (-1, 0.35) and (-1, 2.83) respectively. As seen earlier in the case of Kerr enhancement \( n = 1 \), \( k \to 0 \) is the
condition for observing an enhancement in Kerr angle. Faraday enhancement will be similarly observed when $n = -1, k \to 0$. Figure 3-15 b) shows the calculated $\theta_F$ response for a 3.2 nm thick iron film on a metamaterial described by Doling et al., Im[$\theta_F$] shows a peak at 1430 nm.

---

![Figure 3-15](image)

Figure 3-15 (a) Refractive index of a metamaterial (b) calculated $\theta_F$. Refractive index data from Ref. [64].

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### 3.9 Kerr enhancement in suspended films (air substrate)

Vacuum has a refractive index of $n=1, k=0$ [65], which makes it a perfect substrate for observing Kerr enhancement at all wavelengths. Suspended graphene sheets has shown rich physics including the presence of strain induced pseudo-magnetic field of 300 T [66] and zero-field quantum Hall effect [67], fractional quantum Hall effect [68, 69] and the presence of ultrahigh ballistic electrons [70, 71]. Figure 3-16 a) and b) shows the calculated Kerr response for suspended graphene and graphene on the 4H-SiC substrate for Re[$\theta_K$] and Im[$\theta_K$], respectively. The suspended graphene shows an enhanced Kerr response in Re[$\theta_K$]...
and \( \text{Im}[\theta_k] \) over the entire energy range. The \( \text{Re}[\theta_k] \) changes sign at 120 meV, the significance of which is not clear at this moment.

![Graph showing Kerr angle for suspended graphene.](image)

**Figure 3-16 Kerr angle for suspended graphene.**
Calculated Kerr angle for suspended graphene and graphene on 4H-SiC a) \( \text{Re}[\theta_k] \) and b) \( \text{Im}[\theta_k] \).

### 3.10 Conclusion

Magneto-optical Faraday and Kerr effects probe the off-diagonal (Hall) conductivity \( \sigma_{xy} \), which is very sensitive to the electronic structure of the material. Studying the energy dependence of \( \sigma_{xy} \) through Faraday and Kerr measurements provides new information about the energy scale of a system such as plasma frequency, carrier relaxation rate, cyclotron frequency [36]. In materials showing unusual dc Hall properties, it is especially interesting to explore how the Hall effect behaves at higher frequencies [17, 34, 36]. In spite of the rich physics that Kerr and Faraday measurements can access, it can be very challenging to measure Kerr signals, which can be very small and difficult to separate from the background signals.
and noise. In this scenario, the reststrahlen band assisted Kerr enhancement mechanism provides a new way to enhance this signal by almost two orders of magnitude. The reststrahlen band in ionic crystals is known to vary between 7-200 µm [56, 72]. Therefore, although the enhancement only occurs over a narrow energy band, this narrow band can be varied by choice of substrate. Moreover, our model indicates that the enhancement be achievable at all frequencies for free-standing films in air or vacuum.
4 Optical anisotropy in epitaxial graphene

This Chapter will cover another discovery that we made in exploring infrared polarization effects in graphene grown on SiC. Due to terraces in the SiC substrate, graphene forms a wire-like pattern, and this anisotropic conductivity acts as a linear dichroic polarizer. This effect is not only interesting for the study of graphene and possible applications, it also could be important for understanding similar signals in other materials, such as the high-temperature superconducting cuprates, where substrates terraces also produce steps in these films. We will first discuss dc conductivity anisotropy discovered in epitaxially grown graphene on SiC substrates [16]. We will then discuss the analogous optical anisotropy measurements, which are the main result of this Chapter. We will conclude the Chapter by discussing a theoretical model to explain the observed anisotropy.

4.1 Background

The unique electronic properties of graphene have attracted a lot of attention over the last decade [73-75]. Studies with suspended graphene monolayers have shown electronic mobility exceeding $200,000 \text{ cm}^2\text{V}^{-1}\text{S}^{-1}$ [70, 71, 76, 77]. Furthermore, the mobility shows little temperature dependence between 10 K and 100 K [7, 9, 78]. This temperature independence suggests that defects are the main scattering mechanism.

For practical applications on an industrial scale, large area graphene on suitable substrates are required. Hence, the influence of the substrate on the electrical properties of graphene is highly important. Understanding this interaction will play a key role in designing devices for nano-scale electronics [7, 8, 79, 80].
4.2 Growing epitaxial graphene on SiC

The growth of a crystalline layer on top of a crystalline substrate is known as epitaxy. When SiC is heated, the Si atoms sublimate at a rate faster than C and leave the surface before the C atoms. This is due to the fact that Si has higher vapor pressure than C [81]. The leftover C atoms rearrange to form graphene. Graphene formed in this manner is called epitaxial graphene. Figure 4-1 shows the schematic of the growth process. One of the main advantages of growing epitaxial graphene is that the graphene layer directly forms on the substrate, thus eliminating contamination risk associated with transferring films on to substrates. Epitaxially grown graphene on SiC can be made large area and high quality [82].

Figure 4-1 Epitaxial growth of graphene.
(a) Shows the honeycomb lattice structure of graphene (b) SiC with C and Si atoms (c) SiC is heated/annealed, Si atoms sublimate (d) on Si face monolayer graphene formation (e) on the C face, rotationally decoupled multilayer graphene formation.
Due to lack of inversion symmetry, 4H-SiC and 6H-SiC have polar axes and have two inequivalent faces the (0001) and (000\overline{1}). The former is known as Si face and the latter is known as C-face of SiC \[83, 84\]. Figure 4-2 shows the two faces of SiC crystal. The morphology and transport properties of graphene depend on the face of SiC crystal on which it is grown. Graphene grown on the C-face forms multilayers whereas only a single monolayer grows on the Si-face. The Si-face monolayers have much lower mobilities of about \(1000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}\) \[85, 86\]. The multilayers in C-face graphene are typically rotationally decoupled and exhibit high carrier mobility, with values greater than \(18000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}\) at 300 K \[86\].

Infrared transmission and magneto-optical studies have also shown that the 10-20 layer thick epitaxial graphene on the C face behaves as an isolated monolayer \[87, 88\]. To understand this puzzle, first principle calculations by J. Hass et al. [2008]\[89\] have revealed that rotationally decoupled graphene sheets on the C face of SiC, produces a band structure which is nearly identical to isolated monolayer graphene. When the crystal axes of two neighboring graphene layers are rotated with respect to each other, they are essentially
decoupled and behave as two independent monolayers. In other words, we can imagine that the graphene layers that are buried inside the multilayer film are better protected from the external environment (surface and substrate), thus preserving their pristine intrinsic monolayer band structure.

The C face and Si face also differ morphologically. The C face samples are marked by meandering steps and distinctive “giraffe stripes” lining the sample [16] whereas step bunching and micron size terraces are observed on the Si face samples. Although the SiC substrate before the growth of graphene is atomically flat, when graphene grows at high temperature ~1600 °C, the movement of surface atoms reconstructs the surface and forms wide terraces and high steps. This phenomenon is called step bunching. Ideally, we would like to have the devices on a step-free substrate and several groups have succeeded in this attempt, however, a millimeter scale step free surface is difficult to make [90-92]. The step bunching phenomena and its sequential control has been studied by Bao et al. [90] and according to their study, “large step bunching” (~10 nm) takes place when the SiC is heated slowly whereas “minimum step bunching” (~1 nm) is a result of fast heating of SiC substrate. Figure 4-3 from Bao et al. shows this effect of fast and slow heating resulting in minimum step bunching and large step bunching respectively.

Characterizing the change in the electronic properties of graphene due to its interactions with substrate steps is highly essential for applications. The interaction of epitaxial graphene with atomic terraces and steps has been studied by various groups. Studies find that the resistance increases when current flows over a step [16, 93], compared to staying within a single terrace, and that this increase is greater when the step height is larger [94, 95].
Figure 4-3 Bunching of steps in SiC.
AFM images when (a) the sample was heated fast while in (b) the sample was slowly heated. Slow heating produce large step bunching while fast heating produces minimum step bunching. The AFM images shows the height and width of the steps and terraces. Figure from Bao et al. [2016].
4.3 Conductance anisotropy

Yakes et al. (2010) of NRL have shown conductance anisotropy in epitaxial graphene by measuring the angular dependence of resistance. Studies were done on both Si face and C face. To measure angular dependence of resistance, the authors used rotational square micro four probe method [96, 97] in which the four probes were rotated with respect to the substrate to measure the resistance parallel and perpendicular to the steps Figure 4-4 (a) shows the SEM image of Si face epitaxial graphene. Parallel step bunches are clearly seen. The resistance vs sample orientation is shown in Figure 4-4 b. The resistance parallel to the step edge is 260 Ω while resistance perpendicular to the step edge is 440 Ω. The model fit of resistance is shown by the fitted curve (black).

![SEM image of Si face epitaxial graphene](image1.png)
![Resistance vs sample orientation](image2.png)

**Figure 4-4** (a) SEM image of Si face epitaxial graphene (b) Measured resistance vs sample orientation angle.
Figure 4-5 (a) shows the SEM image of the C face of epitaxial graphene. Because of multilayer growth of graphene, the buried terraces and steps of the SiC substrate cannot be seen. However, the resistance measurements show angular dependence similar to the Si-face samples.

The authors have argued that the primary scattering mechanism responsible for the observed conductance anisotropy originates from the trapped silicon atoms between neighboring graphene layers and in between the lowest graphene layer and the substrate. They discuss that in a terraced substrate, the trapped atoms (Si in this case) tends to move towards the step edge and this build-up of charges enhances the scattering at the edges.

### 4.4 Optical anisotropy

With its high electrical mobility and optical transparency along with mechanical flexibility and strength, graphene provides excellent opportunities for future optoelectronic devices [98-102]. Traditionally, indium tin oxide (ITO) has been used for making transparent
electrodes, however, due to its ceramic nature, making flexible electronics is challenging. Moreover, indium is a strategic element with limited supply. Graphene, on the other hand can be produced cheaply and in bulk quantity once the growth technology matures. In this scenario, studying the optical properties of graphene will be crucial. In this study, we have focused on the polarizing properties of graphene on a substrate.

We have observed the optical analog of the conductance anisotropy in the same samples as used by Yakes et al. [16]. We measured $\theta_\kappa$ at 120 meV without any magnetic field ($B=0$ T ) and at room temperature for different sample orientations. The sample was rotated about an axis going out of the plane. Figure 4-6 shows the measured $\theta_\kappa$ response. Both Re[$\theta_\kappa$] and Im[$\theta_\kappa$] vary sinusoidally with sample orientation. From these data, it is clear that graphene is showing an optical anisotropic response, which is consistent with the difference in conductivity for directions parallel and perpendicular to the steps. A familiar example would be a wire grid polarizer.
Figure 4-6 Angle dependence of infrared Kerr response at zero magnetic field and room temperature measured at 120 meV.

4.5 Theoretical modeling

To understand the origin of optical anisotropy in epitaxial graphene, we will model it as a linearly dichroic (absorption is different for light polarized along and perpendicular to the steps, which changes the reflected amplitudes of the two polarizations) and linearly birefringent (index of refraction is different for light polarized along and perpendicular to the steps, which produces a phase shift between the two polarizations) material.
Figure 4-7 Optical setup for measuring the polarization change due to a rotating sample with linear dichroism (LD) and birefringence (LB). Light from a laser with electric field $E_0$ is incident on a sample exhibiting linear dichroism and birefringence. The sample is mounted on a rotational mount, and is spinning in XY plane. The reflected light passes through a PEM and a second polarizer P2 fixed at 45°. Finally the light is detected by a MCT detector.

Let us consider a sample which is dichroic ($k_x \neq k_y$) as well as birefringent ($n_x \neq n_y$). As a result, the reflection coefficients are $r_s$ and $r_p e^{i\eta}$. Figure 4-7 shows the optical setup in which the sample is spinning and the change in polarization is detected by the combination...
of PEM and a polarizer at 45 degree. The intensity of the reflected light is measured by a MCT detector.

The Jones matrix for the sample can be written as

\[
(M_{\text{Sample, LD}})(M_{\text{Sample, LB}}) = \begin{pmatrix} A & 0 \\ 0 & B \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & e^{i\eta} \end{pmatrix} = \begin{pmatrix} A & 0 \\ 0 & Be^{i\eta} \end{pmatrix}
\]

Where \( A = r_x \) and \( B = r_y \) and \( \eta \) is the phase .

The Jones matrix for the optical setup is given by

\[
E_{\text{det}} = (M_{\text{p-e5}})(M_{\text{PEM}})(M_{\text{Rot(-\theta)}})(M_{\text{Sample, LD}})(M_{\text{Sample, LB}})(M_{\text{Rot(+\theta)}})E_{\text{inc}}.
\]

\[
\begin{pmatrix} Ex \\ Ey \end{pmatrix} = \begin{pmatrix} 1 & 1 \\ 2 & 1 \end{pmatrix} e^{i\delta} \begin{pmatrix} \cos(\theta) & -\sin(\theta) \\ \sin(\theta) & \cos(\theta) \end{pmatrix} \begin{pmatrix} A & 0 \\ 0 & Be^{i\eta} \end{pmatrix} \begin{pmatrix} \cos(\theta) & \sin(\theta) \\ -\sin(\theta) & \cos(\theta) \end{pmatrix} \begin{pmatrix} E_0 \end{pmatrix}
\]

\[ (25) \]

\[
I_{\text{det}} = E_0^2 \left[ A^2 + B^2 + (A^2 - B^2)\cos2\theta - 2AB\sin\theta\sin2\theta\sin\delta + \cos\delta \left[ 4A^2 \cos^3 \theta \sin\theta - 4B^2 \cos\theta\sin^3 \theta - AB\cos\theta\sin4\theta \right] \right]
\]

Using the Bessel function expansion of \( \cos\delta \) in the above equation and collecting the terms of different harmonics we get,

\[
I_0 = E_0^2 \left[ A^2 + B^2 + (A^2 - B^2)\cos2\theta + J_0(z)[4A^2 \cos^3 \theta \sin\theta - 4B^2 \cos\theta\sin^3 \theta - AB\cos\theta\sin4\theta] \right]
\]

\[ (27) \]

\[
I_{2\omega} = -2J_1(z)E_0^2 \left[ 4A^2 \cos^3 \theta \sin\theta - 4B^2 \cos\theta\sin^3 \theta - AB\cos\theta\sin4\theta \right]
\]

\[ (28) \]
\[ I_{3\omega} = 4J_3(z)E_0^2[AB\sin\eta \sin 2\theta] \]  
(29)

\[
\begin{align*}
I_{2\omega} &= \frac{2J_2(z)\left[ 4A^2 \cos^3 \theta \sin \theta - 4B^2 \cos \theta \sin^3 \theta - AB \cos \eta \sin 4\theta \right]}{A^2 + B^2 + (A^2 - B^2) \cos 2\theta + J_0(z)\left[ 4A^2 \cos^3 \theta \sin \theta - 4B^2 \cos \theta \sin^3 \theta - AB \cos \eta \sin 4\theta \right]} \\
I_{3\omega} &= \frac{4J_3(z) [AB\sin \eta \sin 2\theta]}{A^2 + B^2 + (A^2 - B^2) \cos 2\theta + J_0(z)\left[ 4A^2 \cos^3 \theta \sin \theta - 4B^2 \cos \theta \sin^3 \theta - AB \cos \eta \sin 4\theta \right]} 
\end{align*}
\]

Setting \( R_d = 2.406 \text{ rad} \) and \( J_0(2.406) \approx 0 \) simplifies the equation for \( \text{Re}[\theta_e] \) and \( \text{Im}[\theta_e] \). The simplified equations are,

\[
\begin{align*}
\text{Re}[\theta_e] &= \frac{1}{4J_3(R_d)} \times \frac{2J_2(z)\left[ 4A^2 \cos^3 \theta \sin \theta - 4B^2 \cos \theta \sin^3 \theta - AB \cos \eta \sin 4\theta \right]}{A^2 + B^2 + (A^2 - B^2) \cos 2\theta} \\
\text{Im}[\theta_e] &= \frac{1}{4J_3(R_d)} \times \frac{4J_3(z) [AB\sin \eta \sin 2\theta]}{A^2 + B^2 + (A^2 - B^2) \cos 2\theta}
\end{align*}
\]  
(30) (31)

Equations (30) and (31) can be seen as a general equation which reduces to the situation of a rotating polarizer, waveplate and LD with appropriate choice of the variables \( A \), \( B \) and \( \eta \).

1. If \( A = r_x = 1 \), \( B = r_y = 0 \), the situation reduces to the case of rotating a linear polarizer (Appendix 8.1).

2. If \( A = r_x = B = r_y = 1 \) and \( \eta \neq 0 \), the situation reduces to the case of rotating a waveplate (Appendix 8.2).
3. If \( \eta = 0 \), the situation reduces to the case of rotating a pure dichroic sample without any birefringence (Appendix 8.3).

Figure 4-8 shows the rotation and ellipticity signal produced by the sample for \( A/B = 1.04, \eta = 0.06 \). The rotation and ellipticity signal thus obtained matches the experimental data for epitaxial graphene grown on SiC with a phase difference of about 135 degrees.
5 Linear dichroism in cuprate superconductors

Even after 30 years of its discovery and more than 200,000 referred publications [103] later, understanding high-temperature superconductors (HTS) remains one of the most challenging problems in condensed matter physics. Its rich behavior is evident from its complex phase diagram, where different phases compete, and each phase has its own long-range order and associated broken symmetries.

A key problem in the realm of HTS is to understand the pseudogap phase. Whether it is just a precursor to superconductivity or it represents a full-fledged thermodynamic phase with its own broken symmetry is yet to be settled. In this Chapter, we focus on the presence of broken symmetry states in HTS and use Faraday measurements at B=0 to identify the broken symmetry.

We observe a Faraday rotation signal, which shows sample orientation dependence (linear dichroism and linear birefringence). Although we see changes in both the polarization rotation (Re[θr]) and ellipticity (Im[θr]) as the sample is rotated, we will focus on Re[θr] in this Chapter. Since the Re[θr] signal that we measure has a two-fold rotational symmetry, it can be best associated with linear dichroism (LD), which occurs when radiation polarized along one direction is absorbed more strongly than radiation polarized in the perpendicular direction. We measure the amplitude of LD and study its temperature, energy, and doping dependence. We investigate possible artifacts from the substrate. We do not find any appreciable signal coming from the substrate. We conclude that the LD signal may be due to nematic charge ordering in cuprates, as has been observed in dc Hall transport measurements [104].
5.1 Background

Copper oxides, now known as cuprates are insulators which when doped with lanthanum or barium become superconductors. The remarkable discovery in 1986 by Bednorz and Muller of a material (La\textsubscript{2-x}Ba\textsubscript{x}CuO\textsubscript{4}, LBCO) superconducting at 35 K was beyond the maximum limit (25-30 K) set by the BCS theory. Soon, other cuprates with even higher T\textsubscript{c}, for example YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7} (YBCO) with T\textsubscript{c} of 93 K, were discovered. The evolution of an insulator to a superconductor is remarkable. Researchers discovered that by doping the copper oxides, electrons were freed from the copper atoms and could then flow freely in the lattice. When the samples are cooled below T\textsubscript{c}, the electrons move in a highly ordered state exhibiting zero resistance- they become superconducting. The phase diagram of hole-doped cuprates is shown in Figure 5-1. The parent compound is an insulating antiferromagnet (AFM). As the system is doped, the AFM phase gets suppressed and for a critical range of doping a superconducting dome appears. At the highest doping, the system turns into a normal metal. In underdoped HTS cuprates, a weak energy gap begins to appear at T' as the sample is cooled before the superconducting gap fully emerges at the lower temperature T\textsubscript{c}. Above T\textsubscript{c} and below T' is the region of enigmatic pseudogap (PG) phase. In the pseudogap phase, there is a depression of density of states which resembles a gap (the density of states does not go to zero, as in a true gap). This PG phase exhibits anomalous transport [105, 106], thermodynamic [107, 108] and optical [109-112] properties. Whether the PG phase is a new phase [113-115] with its own broken symmetries or is it just a precursor to superconductivity [116-118] is a question that is intensely debated.
Figure 5-1 Phase diagram for hole doped cuprate showing different competing phases[119].

5.2 Optical anisotropy measurements

Recent measurements done in the THz [120] and near infrared [112] energy ranges have discovered the presence of linear and circular asymmetries, respectively. In the NIR (800 meV) measurement done by Prof. Kapitulnik’s group [112], a small polar Kerr rotation ($10^{-6}$ rad) was measured in the absence of any external applied magnetic field in YBCO in the pseudogap regime, shown in Figure 5-2a. The Kerr rotation signal was independent of sample orientation and therefore originates from a circular asymmetry in the system. A circular asymmetry can arise, for example, from the breaking of time reversal symmetry (out-of-plane magnetization) or from presence of optical gyrotropic effect [109, 121]. In gyrotropic effect, charge stripes oriented in different directions for adjacent CuO$_2$ planes, produce a helical structure for light propagating perpendicular to the plane, thus breaking circular symmetry.

In a similar measurement done in the THz (2-6 meV)[120] range by Prof. Armitage’s group, a large Faraday rotation signal ($10^{-3}$ rad) that depends on sample orientation was
observed in YBCO, shown in Figure 5-2(b,c). This orientation dependence suggests the presence of a linear asymmetry in the sample, which can arise from the presence of stripped states in the pseudogap regime.

The frequency dependence of the optical anisotropy is crucial to resolving the microscopic origin of the broken symmetry. For example, since metals typically have a higher...
conductivity at lower frequencies, one would expect that metallic stripes or wires would produce larger optical anisotropy as the probe frequency decreases. Gyrotropic models predict a $\omega \gamma(\omega)$ dependence where $\gamma(\omega)$ is the gyrotropic parameter [120]. In this Dissertation, we have studied the optical anisotropy of YBCO thin films (of various doping levels), in mid-infrared energies (132 meV, 228 meV) and in visible energy (2330 meV). Figure 5-2d shows the energy range of our current and future work.

5.3 Samples

Two groups provided samples for this study. 1) Prof. Koren’s group, Technion, Israel and 2) Prof. Wei’s group, University of Toronto, Canada. The list of samples is given below.

<table>
<thead>
<tr>
<th>No.</th>
<th>Sample name</th>
<th>Doping</th>
<th>$T_c$ (k)</th>
<th>Thickness (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Technion samples</td>
</tr>
<tr>
<td>1</td>
<td>UD 52</td>
<td>Underdoped</td>
<td>52</td>
<td>100</td>
</tr>
<tr>
<td>2</td>
<td>UD 78</td>
<td>Underdoped</td>
<td>78</td>
<td>100</td>
</tr>
<tr>
<td>3</td>
<td>OPD 91</td>
<td>Optimally doped</td>
<td>91</td>
<td>100</td>
</tr>
<tr>
<td>4</td>
<td>OVD 85</td>
<td>Overdoped</td>
<td>85</td>
<td>100</td>
</tr>
<tr>
<td>5</td>
<td>LBCO</td>
<td>$X = 1/8$</td>
<td>20</td>
<td>100</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Toronto samples</td>
</tr>
<tr>
<td>1</td>
<td>UD 70</td>
<td>Underdoped</td>
<td>70</td>
<td>200</td>
</tr>
</tbody>
</table>
Table 5-1 List of cuprate samples and specifications.

<table>
<thead>
<tr>
<th></th>
<th>Sample Type</th>
<th>Doping Level</th>
<th>Experimental E (meV)</th>
<th>Starting Temperature (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>UD 50</td>
<td>Underdoped</td>
<td>50</td>
<td>200</td>
</tr>
<tr>
<td>3</td>
<td>OPD 90</td>
<td>Optimally doped</td>
<td>90</td>
<td>200</td>
</tr>
<tr>
<td>4</td>
<td>LSCO</td>
<td>X=1/8</td>
<td>20</td>
<td>200</td>
</tr>
</tbody>
</table>

Figure 5-3 Angle dependence of $\text{Re}[\theta_F]$ for YBCO/LSAO Technion samples at different energies.

5.4 Zero field Faraday rotation measurements

Faraday angle at zero external magnetic field was measured for YBCO, LBCO and LSCO thin film samples grown on LSAO substrate. Figure 5-3 shows angular dependence of $\text{Re}[\theta_F]$ measured for Technion samples at 290 K at photon energies of 132 meV, 228 meV and 2330 meV. All the samples show clear LD response. The signals are sinusoidal with 180 degree periodicity. The vertical offsets are random and have no physical significance. Comparing the peak-to-peak amplitudes (the difference between maximum and minimum of $\text{Re}[\theta_F]$) of the
sinusoidal signal, UD 78 and UD 52 show a large LD response at all the measured energies. OVD 85 has an amplitude of 4 mrad at 132 meV but the amplitude decreases to 0.3 mrad at 2330 meV. Similarly OPD 91 shows an amplitude of about 1 mrad at 132 meV but decreases to less than 0.5 mrad at 2330 meV.

Figure 5-4 shows the angular dependence of Re[θ_e] for the Toronto samples. UD70 shows the largest amplitude of 18 mrad at 228 meV. UD 50 shows the smallest amplitude at all the three energies. OPD 90 shows a similar amplitude (~4 mrad) at 132 meV and 228 meV while decreasing to < 1 mrad at 2330 meV. The LSAO substrate does not show any appreciable LD signal (see Figure 5-4a).

Figure 5-4 Linear dichroism for Toronto samples at different energies. Angle dependence of Re[θ_e] for YBCO/LSAO samples with different dopings.
The amplitude of the LD signal $\Delta \text{Re} \theta_r$ for all the samples was usually largest at 228 meV, as can be seen by the peak in Figure 5-5. The response of optimally doped (OPD 91) and overdoped (OVD 85) was minimal. We plan to measure these samples using a broadband FTIR to generate a continuous spectra in the MIR.
Figure 5-6 shows the temperature dependence of $\text{Re}[\theta_f]$ when the sample was cooled when oriented at $\pm 45^\circ$ with respect to the polarization of the incident light. $\text{Re}[\theta_f]$ signal changes sign when the sample is rotated by $90^\circ$. No special signature is seen at $T_c$. The difference $\Delta \text{Re}[\theta_f]$ between the cooling curves increases as the sample is cooled, which indicates that the LD grows stronger at lower temperature. The LSAO substrate did not show any appreciable temperature dependence. Figure 5-7 tracks $\Delta \text{Re}[\theta_f]$ as the sample cools from room temperature to 77 K. The underdoped and overdoped samples shows an increase in $\Delta \text{Re}[\theta_f]$ as the sample is cooled while the optimally doped sample shows no temperature dependence.
Figure 5-7 Temperature dependence of $\Delta \text{Re}[	heta_r]$ at 132 meV.

Figure 5-8 shows linear dichroism of LBCO and LSCO samples at 132 meV. Both the samples show strong LD at room temperature. $\Delta \text{Re}[	heta_r]$ for LBCO is about 50 mrad, which is the largest signal among all the cuprates that we measured at room temperature.

Figure 5-8 Linear dichroism of LBCO and LSCO (x=1/8) samples.
5.5 Issue of phase shift and asymmetry

The LD signal shown in Figure 5-3 and Figure 5-4 shows phase shifts and small asymmetry in the lobe amplitudes. If stripes/wires are responsible for the LD signal, one may expect that the phase shift depends on how the stripes/wires are aligned with the edge of the sample, which is used to determine the sample orientation. Figure 5-9 shows the LD signal measured at 132 meV and the orientation of stripes/wires that would be consistent with the measured signal. The lobe asymmetry can occur if the angle of incidence of the probing radiation is large (> 10°). In the measurements, great care was taken to minimize the angle of incidence, which we estimate to be below 5°.

Figure 5-9 Phase shifts and possible orientation of wires.
5.6 Role of terraces and steps in producing the LD signal.

We have seen the effect of terraces on the substrate producing optical anisotropy in graphene films on SiC (Mukherjee et al. [in preparation]). In bare SiC, which is insulating, no LD signal was observed. However, when metallic graphene was placed on top of the SiC, the terraces produced anisotropic conductivity due to graphene forming wires along the terrace edges. This has also been observed in dc transport of graphene on SiC [16]. Therefore, although no LD signal was observed in bare LSAO, which is also an insulator, we wanted to make sure that the unavoidable steps/terraces in LSAO [104] do not produce wire-like conductivity when a metallic film is deposited in the LSAO. To test this, a 100 nm thick metal film (LNO) was deposited on a LSAO substrate. The metal film produced no LD signal, which makes us confident that the LD signals are intrinsic to the cuprate films.

---

**Figure 5-10** LD signal of YBCO compared to a LNO metal film (green and red).
5.7 Discussion

Linear symmetry breaking was observed in cuprate superconductors with a wide range of dopings grown on LSAO substrates. The tetragonal substrate does not contribute to the $\theta_F$ signal. The $\theta_F$ signal from YBCO, LSCO and LBCO are seen at room temperature and down to 10 K without the application of any external magnetic field. We observe strong probe energy dependence of the LD signal. The LD signal persists from MIR to visible energies. The strongest LD tended to be from underdoped samples, where $T^*$ is highest. These results are similar to those found by THz measurements [120]. Although the samples are polycrystalline films, one would expect the contributions from crystal structure and Cu-O chain from different domains to average out, but the strong linear asymmetry signals suggest that the effect is macroscopic and persists across many domains over the large area ($\sim 0.08 \text{ mm}^2$) of the probing beam. The signal is consistent with conductive stripes spread across the entire probe region. The effect strengthens with decreased doping. If the signal has its origin in the greater conductivity of Cu-O chains, the trend should be opposite since underdoped samples are less metallic. Additionally our observations have ruled out the possibility of substrate terraces inducing optical anisotropy. All these observations points towards the existence of conductive stripes which are persisting at the normal state of the cuprates. The recent observation [104] of anisotropy in in-plane dc transport measurements in the cuprates is consistent with our observations. The authors in Ref [104] suggest that the anisotropy is due to electronic nematicity, that may not necessarily be aligned with the crystal axes. By extending the in-plane anisotropic conductivity to the infrared, we hope to provide new tests of this claim.
6 Infrared Hall effect studies in iron based superconductors (IBSC)

The infrared Hall effect probes the off-diagonal conductivity \( \sigma_{xy}(\omega) \) and is sensitive to electronic structure and magnetic ordering. These measurements have played a vital role in understanding strong correlation effects in the superconducting cuprates [34, 35] as well as understanding electronic structure of magnetic materials like GaMnAs [30] and CaSrRuO\(_3\) [31]. Iron pnictide based superconductors are unique systems in that they contain typically ferromagnetic elements such as iron, which one would expect to oppose the formation of Cooper pairs. The iron can also produce extrinsic ferromagnetism in IBSC, leading to a highly unusual and interesting situation where both superconductivity and ferromagnetism can coexist in the same sample. Since infrared Hall measurements are sensitive to magnetic and electronic order and can probe a broad and important energy range (superconducting gap, spin-wave gap, etc. are in the infrared energy range), these measurements are particularly useful for studying these materials. In this Chapter, we study Co-doped BaFe\(_2\)As\(_2\) by exploring its temperature and energy dependence. We use both Faraday measurements, which are sensitive to magneto-electronic structure, and SQUID measurements, which probe magnetic moments in a complementary fashion, to understand the interaction of superconductivity and ferromagnetism.
6.1 Background

Ferromagnetism and superconductivity are opposing effects. Iron due to its large magnetic moment aligns the spin and favors ferromagnetism while superconductivity arises with the formation of Cooper pairs, which require dynamic antiparallel spin orientation. So the discovery of superconducting iron-based compounds in 2009 was a big surprise for the condensed matter community. Fluorine-doped lanthanum oxide iron arsenide (LaO$_{1-x}$F$_x$FeAs) was discovered to be a superconductor with $T_c$ of 26 K [14]. Figure 6-1 shows a timeline of the discovery of major superconductors. With the advent of IBSC, a new route to high $T_c$ superconductivity was discovered which rejuvenated the area and started extensive search for other IBSC families with higher $T_c$. The theory of high $T_c$ superconductivity remains one of the most challenging problems in condensed matter physics although it was discovered more than 30 years ago. The discovery of IBSC demonstrates that unconventional (non-BCS) superconductivity is not just limited to cuprates. With IBSC, a new system with a new order parameter and a possibly different mechanism for achieving high $T_c$ superconductivity has been discovered, which gives a great chance in solving the puzzle of high $T_c$ superconductivity. For an interesting comparison between cuprates and IBSC see “Superconductivity gets an iron boost” by Igor Mazin, Nature 2010 [122].

Currently $T_c$ values up to 56 K have been reported for IBSC. Superconductivity has been discovered in many families of IBSC such as 122-type (XFe$_2$As$_2$, X=Ba, Sr,Ca), 1111-type (XFeAsO, X=La,Ce,Pr,Nd), 111-type (XFeAs, X=Li, Na), etc.
IBSC shows similarities with their siblings, the copper-oxide-based superconductors. Both pnictides and cuprates are layered materials and superconductivity is supposed occur in the 2D planes of FeAs and CuO respectively [122]. In both systems, electron-electron correlations play an important role in the properties of the normal state as well as in the superconducting state [123].

There are also important differences; the parent compounds are semi-metals in the pnictides while they are Mott insulators in cuprates. Superconductivity in IBSC arises when anti-ferromagnetism (AFM) is completely eliminated (in 1111-type) or diminished (in 122-type) either by applying external pressure [124] or by chemical pressure, which is achieved by partially substituting arsenic with phosphorous [125] or replacing iron with cobalt [126].
In cuprates, strong electron correlation leads to electron localization in the parent compounds, which makes them Mott insulators. Superconductivity appears when extra charge carriers are doped into the system, which screen the electron-electron repulsion and produces superconductivity when the doping is high enough.

IR reflectance measurements [127-129] performed on IBSC have measured the longitudinal conductivity $\sigma_{xx}(\omega)$ and revealed the spectrum of electronic excitations. By studying the temperature and doping dependence of the spectral weight, these measurements have revealed the presence of pseudogap phase both in the parent compound and in optimally doped Ba122. At present, it is not clear how the pseudogap phase is related to superconductivity. Is it a precursor to superconductivity or a distinct phase. IR Hall measurements can provide a new perspective on the nature of pseudogap by measuring $\sigma_{xy}(\omega)$ at different temperatures and for different doping levels.

In this Chapter, we present measurements on optimally doped BaFe$_2$As$_2$ samples. We study the temperature and energy dependence of IR Hall effect. We discover that in addition to the signal coming from Ba122, there exists an extrinsic FM signal induced by iron impurities, which complicates the analysis of the results. We try to separate the two signals by studying their temperature and energy dependence. At the time of writing this Dissertation, the analysis of the results is still ongoing and many questions remain unanswered.

### 6.2 Samples

Epitaxial films of optimally doped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ were grown by Prof. Hosono’s lab, where IBSC was first discovered. Films were deposited using pulsed laser deposition (PLD)
techniques on BaF$_2$ and LaSrGaO$_4$ substrates, which were specially chosen by us since they are transparent in MIR. Three different films were grown, they are:

<table>
<thead>
<tr>
<th>#</th>
<th>Film</th>
<th>Substrate</th>
<th>Thickness (nm)</th>
<th>$T_c$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$</td>
<td>BaF$_2$</td>
<td>230</td>
<td>26</td>
</tr>
<tr>
<td>2</td>
<td>Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$</td>
<td>BaF$_2$</td>
<td>160</td>
<td>26</td>
</tr>
<tr>
<td>3</td>
<td>Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$</td>
<td>LaSrGaO$_4$</td>
<td>125</td>
<td>20</td>
</tr>
</tbody>
</table>

Figure 6-2(a) shows the temperature dependence of resistivity of Sample 1. The onset of superconductivity is seen at $T_c = 26$ K. The XRD data for Sample 1 is shown in Figure 6-2 (b). It shows intense diffraction peaks assigned to BaFe$_2$As$_2$: Co 00l and BaF$_2$ 00l. No peak is seen from Fe crystals. Figure 6-3(a) shows the temperature dependence of resistivity for Sample 3 with $T_c = 26$ K. In the XRD data shown in Figure 6-3(b), intense diffraction peaks assigned to BaFe$_2$As$_2$: Co 00l and BaF$_2$ 00l are seen along with a small peak from Fe at 65°. In similar epitaxial films (BaFe$_2$As$_2$: Co/LSAT), the presence of Fe and FeAs were seen via strong diffraction peaks at 65° and 35°, respectively [14]. The absence of any peaks at these angles in the XRD data of Sample 1 rules out the presence of Fe and FeAs metal within the detection limits of XRD.
Figure 6-2 Characterization of Sample 1.
(a) Temperature dependence of resistivity (b) XRD, peaks are from Ba122 and BaF$_2$. The presence of iron crystals is not seen.
Of the three samples, only sample 1 was chosen for infrared Faraday measurements for two main reasons. Firstly, the Faraday angle is proportional to thickness, so a thicker sample (which is still transparent) will produce greater Faraday rotation. Since sample 1 is the

**Figure 6-3 Characterization of sample 3**
Temperature dependence of resistivity (a) and (b) XRD peaks are from Ba122 and BaF$_2$. Presence of iron is seen at 65°.
thickest it was the natural choice. Secondly, in the x-ray diffraction characterization, sample 1 did not show any impurity phase from Fe or FeAs, whereas sample 3 shows a clear Fe peak.

### 6.3 Infrared Hall effect measurements

The complex Faraday angle \( \tilde{\theta}_F \) in the mid-infrared range (MIR: 100-1200 meV) was measured in Ba122 film as a function of temperature \( (T) \) and magnetic field \( (B) \). In general, all our MIR \( \tilde{\theta}_F \) measurements on the Ba122 films produce a hysteretic response with respect to B, which is consistent with that of a soft ferromagnet.

This is surprising since the level of Fe impurities should be very low in these samples and previous studies in GaMnAs \([18, 30]\) suggests that these ferromagnetic islands contribute very weakly to the Faraday/Kerr signals, which result from the overall optical properties due to the band structure of the bulk material \([30]\). In order to make better sense of the data, we will separate the signal that is linear in B from that which shows hysteretic behavior with B. We may expect that the hysteretic signal is arising from FM domains of Fe impurities, but we think that this analysis is useful regardless of the origins of these two types of magnetic field behaviors. The strong hysteretic signal the saturates above 1.5 T. If the hysteretic signal is from Fe-domains, one could argue that the signal above 1.5 T (where the FM signal has saturated) is the linear response from the Ba122 superconductor. We discuss the results and the approach taken to extract the signal of Ba122 superconductor from the overall Faraday response.
Figure 6-4 Temperature dependence of (a) $\text{Re}[	heta_F]$ and (b) $\text{Im}[	heta_F]$ of Sample 1 at 117 meV photon energy.
Figure 6-4 shows the temperature dependence of the complex Faraday angle $\tilde{\theta}_F$ for the Ba122 film, measured as a function of an out-of-plane magnetic field and at a probe energy of 117 meV. Both the real (Faraday rotation) and imaginary (Faraday ellipticity) components of $\tilde{\theta}_F$, exhibit ferromagnetic hysteresis. Figure 6-4(a) shows $\text{Re}[\theta_F]$ measured as a function of applied magnetic field up to ± 4 T at various temperatures. The measurements were performed from 200 K down to 10 K. At all those temperatures, the magnetic field sweeps yielded hysteresis curves similar to that of a soft ferromagnet, with a step like feature at ~0 T (remnant magnetization) and a linear response at higher magnetic fields above 1.5 T. The slopes of the hysteresis curves at higher fields show clear temperature dependence. Figure 6-4(b) shows the temperature dependence of $\text{Im}[\theta_F]$ as a function of applied B field. $\text{Im}[\theta_F]$ also exhibits ferromagnetic hysteresis similar to the $\text{Re}[\theta_F]$. The linear slope in $\text{Im}[\theta_F]$ above $\approx 1.5$ T shows smaller temperature sensitivity when compared to that of $\text{Re}[\theta_F]$.

In order to trace the origin of the hysteresis signal, we measured the magnetization (M) of the sample in both the normal and superconducting states. Figure 6-5 (a) shows the M-B loop obtained for the Ba122 film using a SQUID magnetometer at 10, 40, 100 and 300 K. At 10 K, in the superconducting state, the M-B curve is a typical diamagnetic loop associated with superconductivity. Figure 6-5(b) shows the ferromagnetic hysteresis curves in the normal state after subtracting the diamagnetic response of the BaF$_2$ substrate. Above $T_c$, the magnetization curves show a ferromagnetic hysteresis response, possibly coming from iron impurities present in the film.
Figure 6-5 Temperature dependence of magnetization of Ba122 sample.
(a) Raw magnetization data for Ba122 (b) Field dependence of magnetization after subtracting the diamagnetic linear response of the BaF$_2$ substrate.

Figure 6-6 Shows the fitted slope at higher B field and the obtained intercept at zero field.
The hysteresis curves of $\tilde{\Theta}_F$ can be analyzed by separating the total response into two parts. The linear slope at high field ($B \geq 2$ T) and the step like feature near $B = 0$ T. The step like feature near $B=0$ T can be attributed to the magnetization-dependent anomalous Hall effect (AHE), which is typically found in metallic ferromagnetic systems and whose origin lies in spin-orbit coupling. In the Ba122 film the step is most likely due to iron particles/islands present in the film as impurities. The linear dependence observed at higher B field, after the magnetization of iron has saturated, is associated to ordinary Hall effect (OHE) and we assume that it is mainly coming from the Ba122 film. Regardless of the detailed sources of these two signals, we think new insights can be obtained by separating these two responses. We explore the temperature and energy dependence of both the slope and step. Figure 6-6 illustrates this analysis technique. A slope ($\Theta_{F \text{ slope}}$) is fitted to the linear part of the hysteresis curve at high $B$ and the slope extrapolated to $B = 0$ T, gives the step height $\Delta \Theta_F$.

To further distinguish the signal from Ba122 and that from Fe particles/islands, a Fe film of 15 nm thickness was grown on a BaF$_2$ substrate and its MIR Faraday response and dc magnetization were studied. We studied the temperature and energy dependence of both the Ba122 and the Fe films. Figure 6-7 shows the temperature dependence of the slope $\Theta_{F \text{ slope}}$ for both the $\text{Re}[\Theta_F]$ and $\text{Im}[\Theta_F]$ for the Ba122 and Fe reference film. For the Fe film, $\Theta_{F \text{ slope}}$ shows very little temperature dependence. On the other hand, Ba122 film shows very strong, non-monotonic temperature dependence. As the sample is cooled from 225 K, the value of $\Theta_{F \text{ slope}}$ goes up by almost a factor of 10 at 35 K where it peaks. $\text{Re}[\Theta_{F \text{ slope}}]$ for the Fe reference film is significantly smaller and stays nearly constant with $T$. 

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Figure 6-7 Temperature dependence of Faraday angle from Ba122 and Fe films at 117 meV.

Figure 6-8 Temperature dependence of $\Delta \text{Re} \theta_F$ from Ba122 and Fe films at 117 meV.
Figure 6-8 shows the temperature dependence of $\Delta \text{Re}[\theta_f]$, which is associated with the abrupt change in magnetization near $B=0$ for a ferromagnet. For Fe, the step height shows very small temperature dependence in the range of 10-200 K, which matches the expectation from Fe since it has a Curie temperature close to 1000 K. $\Delta \text{Re}[\theta_f]$ also doesn’t show much temperature dependence, although it starts rising as $T$ is lowered below 50 K, suggesting that the iron might be playing a dominant role in the zero field AHE signal of Ba122.

Figure 6-9 shows the temperature dependence of $\text{Im}[\theta_f \text{slope}]$. In the case for the Fe film, after the magnetization saturates, a very weak slope remains and that slope does not show any appreciable change over the temperature range. On the other hand, the slope for Ba122 film increases monotonically and is maximum at the lowest temperature. Figure 6-10 shows the temperature dependence of the step size $\Delta \text{Im}[\theta_f]$ at zero field. The response of both Ba122 and Fe film shows very weak temperature dependence, suggesting that the $B=0$ step is dominated by Fe islands that produce an $M$ that is nearly independent of $T$. 

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Figure 6-9 Temperature dependence of $\text{Im} [\theta_F \text{ slope}]$ for Fe and Ba122 films.

Figure 6-10 Temperature dependence of $\Delta \text{Im} [\theta_F]$ for Fe and Ba122 films.
Subfigure 6-11 Energy dependence at 10 K for a) $\Re[\theta_f]$ slope, b) $\Delta \Re[\theta_f]$, c) $\Im[\theta_f]$ slope, d) $\Delta \Im[\theta_f]$.

$\tilde{\theta}_f$ was also measured at different photon energies. Figure 6-11 (a) and (c) show the slopes of $\Re[\theta_f]$ slope and $\Im[\theta_f]$ slope respectively as a function of probe energy at 10 K. At higher energies $E$, the background in $\Re[\theta_f]$ slope becomes large (grows as $E^2$), so the results are less reliable, although we are confident that the sign changes/differences in Figure 6-11(a) are real. $\Delta \Re[\theta_f]$ and $\Delta \Im \theta_f$ for the Ba122 and Fe films in Figure 6-11 (b) and (d) are very similar, suggesting the step in the Ba122 is mainly due to Fe islands. On the other hand, the low energy behavior of $\Im[\theta_f]$ slope is very different for the Ba122 and Fe films. The energy dependence is qualitatively different for both the films, suggesting that $\Im[\theta_f]$ slope from the Ba122 film is dominated by the superconducting Ba122 rather than Fe impurity domains.
In order to better understand the effects of magnetization on $\theta_f$, magnetization measurements were also done on the Fe film. Figure 6-12 shows the magnetization data (total step height at 0 T) for Fe and Ba122 films. Both Fe and Ba122 films show similar temperature dependence. As the sample is cooled, magnetization increases for both the samples.

![Figure 6-12 Temperature dependence of magnetization of Ba122 and Fe films.](image)

### 6.4 Discussion

The coexistence of ferromagnetism and superconductivity in our Ba122-Co doped film makes it a very interesting sample to study proximity based interactions. Even though the ferromagnetism is most likely extrinsic in origin, our results suggest that the SC and FM domains are interacting with each other. Using Faraday measurements, we can probe the interactions occurring below $T_c$ where SQUID only detects the large diamagnetic signal from the superconductor. The temperature dependence of step height is independent of $T$ so its
origin is most likely linked to the FM impurities present in the sample. Looking at the slope, we find there exists a strong $T$ dependence. The slope peaks close to $T_c$ and also its magnitude is similar to other superconductors such as YBCO [36]. Therefore, we can assume that these signals are coming from the superconductor. We cannot be absolutely sure that the linear response in $\tilde{\Theta}_F$ in the Ba122 film is not coming from the Fe domains, since a linear response has been observed in diluted ferromagnets [21, 30, 31, 54]. However, this analysis gives us a good starting point in trying to understand the complicated, multi-phase behavior of Ba122.
7 Conclusions

In this Dissertation, we have explored magneto-optical Faraday and Kerr effects in a variety of 2D materials, which includes graphene and superconductors. Using a unique magneto-polarimetry system, with a sensitivity of 10 µrad, the complex Faraday and Kerr angles were measured in the infrared and visible energy ranges. These magneto-optical effects are very sensitive to the symmetries of the system and provide rich information about the magneto-electronic structure of the material. During this research, we have made many exciting discoveries, some of which could play an important role in studying material properties and the others show promise for future technology. Our main findings are as follows:

1. **Colossal Kerr enhancement effect**- We first discovered this interesting optical effect in multilayer graphene grown on the carbon-terminated face (c-face) SiC. At one particular energy, 121 meV, the Kerr response was enhanced by almost two orders of magnitude. Upon further investigation, we also discovered the same effect in ferromagnetic bulk Al-doped SiC. Our research revealed that the enhancement of the Kerr response is a consequence of the complex refractive index of SiC passing through unity around 121 meV. We tested our model by engineering several iron films on SiC and GaAs substrates. We have modeled the theoretical Kerr response in all the samples and found good agreement with the experiments. Magneto-optical effects are often weak, making them challenging to measure and only practicable in materials with significant optical activity. However, in the present work, we have shown that these effects can be amplified.
through the careful selection of the substrate. This discovery will impact research on novel materials and the development of experimental techniques that exploit the Kerr effect to elucidate the electronic and optical properties of matter. In addition, this enhancement may have technological applications for infrared polarization modulators. Based on this research, we have made two predictions (i) enhancement in the Faraday response (in transmission rather than reflection) for thin films grown on a metamaterial with an index of refraction -1 and (ii) enhancement of Kerr response at all wavelengths for suspended, free-standing films.

2. **Optical anisotropy in graphene**: We have discovered substrate induced optical anisotropy in epitaxial graphene. For this experiment, we have measured Kerr response from epitaxially grown graphene at room temperature without the presence of any external magnetic field. The zero-field Kerr response reflects the presence of optical anisotropy in epitaxial graphene. Transport measurements [16] had earlier revealed dc conductance anisotropy in similar epitaxial graphene samples, grown by the same group that made our samples. The anisotropy in dc transport and optical measurements originates from the interaction between the terraces present on the SiC substrate with the graphene. Trapped Si atoms between the substrate and graphene tend to accumulate near the step edges causing enhanced scattering at the edges. The difference in conductivity for directions parallel and perpendicular to the steps leads to anisotropy in transport and optical properties. We have modeled the Kerr response of the system by treating it as a wire grid polarizer. We believe our technique of detecting optical
anisotropy in graphene at room temperature without the application of a magnetic field can find industrial application in graphene-based flexible displays.

3. **Nematicity in cuprate superconductors**- Recent polarimetry measurements in the THz [120] and near IR [112] energies in cuprate high-temperature superconductors have found the presence of linear and circular asymmetry, respectively. Our aim in this project was to study the broken symmetries in the MIR and visible energy ranges. We performed Faraday spectroscopy at zero-magnetic field for infrared and visible wavelengths and discovered the presence of linear asymmetry. We studied a variety of films with different dopings that were grown on LSAO substrates at two different institutions. For this experiment, a new sample rotator was designed that is capable of rotating the sample inside a cryostat at low temperature and under high vacuum. The measured Faraday response varied sinusoidally as the orientation of the sample is rotated with respect to the polarization of the probing radiation, which indicated the presence of linear asymmetry in the cuprates. We studied the temperature and energy dependence of the Faraday angle for samples with different doping levels. The underdoped samples tended to produce larger signals than the optimally and overdoped films. We checked that the observed linear asymmetry is intrinsic to the cuprate film and not induced by terraces on the substrate (as observed in epitaxially grown graphene on SiC) by observing a null signal from a metal film deposited on a LSAO substrate. All these observations suggested the presence of conductive stripes in the system. Our conclusion is consistent with the THz measurements [120], which also suggest the presence of conductive stripes that persist into the normal state of the superconductors. However, unlike the THz measurements, we observe a
robust Faraday signal even at room temperature. Our discovery is very similar to the dc transport anisotropy in the cuprates that was recently reported in Ref. [104], which concluded that the anisotropy was due to nematic order in the system.

4. **Proximity effects in iron-based superconductors**—Magneto-optical spectroscopy has played an important role in the study of strongly correlated systems such as superconducting cuprates [34, 35] and diluted ferromagnetic systems such as GaMnAs [21, 30]. We have performed Faraday measurements in iron-based superconductors (optimally doped Co-BaFe$_2$As$_2$) in which ferromagnetism (extrinsic) and superconductivity (intrinsic) coexist. The measured complex Faraday angle signal was hysteretic with respect to $B$, which is commonly observed in the magnetization signal from a soft ferromagnet. At higher magnetic fields, the response was linear with $B$ while near zero-field the response was a step function. We studied the temperature and energy dependence of the high field slopes and low field steps separately. The linear slopes showed strong energy dependence with the signal peaking near the critical temperature of the sample (27 K), which suggests that this signal may be affected by the intrinsic superconductivity of the sample. The steps, on the other hand, did not show any significant temperature dependence, suggesting that it may be originating from the ferromagnetic impurities present in the sample. Although one may expect the Faraday signal from ferromagnetic domains to saturate when the magnetization saturates, it is still not clear if the linear response at higher $B$ is only due to the intrinsic response of the superconductor since a linear response at higher magnetic field can also arise from spins located far apart, as in a diluted ferromagnetic phase. However, the analysis technique offers a new way of studying the response of competing
phases in a complex system. Faraday and Kerr spectroscopy can be used along with magnetization measurements to study the nature of electronic structure and magnetic ordering in such systems.

Although this work has deepened our understanding of a wide variety of 2D materials, it has also opened new avenues for future research in all the projects. In the Kerr enhancement project, we hope that future work on thin films grown on metamaterial substrate will detect the proposed Faraday enhancement. Further measurements can be made on an exfoliated graphene flake placed on a micron-sized aperture to verify the presence of Kerr enhancement at all wavelengths. In the cuprate project, we are currently in the process of measuring the broadband Faraday rotation spectrum in the mid-infrared using a FTIR spectrometer. Current measurements in this project were made at only three distinct energies (in the mid-infrared at 132 meV and 228 meV, as well as in the visible 2.3 eV. The broadband measurements will connect these points and provide a continuous spectrum of L.D. With an FTIR, measurements can also be extended down to far-infrared (down to to 4 meV). Continuous measurement from 232 meV upto 3 eV (visible) can also be performed by a custom-built broadband spectrometer which is present in the lab. Measurements of the frequency dependence of the linear asymmetry will provide new insights into the mechanism for broken symmetry. A systematic study of temperature dependence down to 10 K is also planned for all the cuprate samples. Extended studies can also be done on LSCO and LBCO samples with different levels of doping. Measurements are also planned for Kerr measurements on single crystals which will eliminate the substrate interaction. In the pnictide project, the role of extrinsic ferromagnetism and its interaction with intrinsic superconductivity could be further explored by studying samples with high and low level of
ferromagnetic impurities. The proximity effect can be studied systematically for samples with different levels of doping. Future work can also test the presence of nematicity in pnictides as recent measurements have shown an anisotropic optical response in these superconductors [130].

To sum up, our research has shown that Faraday and Kerr spectroscopy is a powerful probe with unique capabilities to reveal the magneto-electronic structure and broken symmetries in complex materials. Several exciting discoveries were made during our research starting from colossal Kerr enhancement in graphene to revealing broken symmetries in superconductors. We believe that our research will serve as a base for future discoveries on 2D materials.
8 Appendix

8.1 Kerr response from a spinning wire grid polarizer.

Figure 8-1 Optical setup to measure Kerr response of a spinning wire grid polarizer.
Light from a laser with electric field $E_0$ is incident on a linear polarizer mounted on a rotational mount, and is spinning in XY plane. The reflected light passes through a PEM and a polarizer P2 fixed at 45°. Finally the light is detected by a MCT detector.

We can model the rotation and ellipticity signal produced due to a spinning wire grid polarizer in Kerr geometry. This method can be used to calibrate the Faraday/Kerr optical setup as shown in Cerne et al. [2003] [36].
The optical setup is shown in Figure 8-1. The probing monochromatic radiation passes through the spinning polarizer and the change in polarization is detected by the combination of PEM and a polarizer at 45 degree. The intensity of the light is finally detected by a MCT detector.

The Jones matrix train for the setup is

$$E_{\text{det}} = (M_{p=45})(M_{\text{PEM}})(M_{\text{Rot}(-\theta)})(M_{p=1})(M_{\text{Rot}(+\theta)})E_{\text{inc.}}$$

The Jones matrix train for the setup is

$$E_{\text{det}} = (M_{p=45})(M_{\text{PEM}})(M_{\text{Rot}(-\theta)})(M_{p=1})(M_{\text{Rot}(+\theta)})E_{\text{inc.}}$$

$$\begin{pmatrix} E_x \\ E_y \end{pmatrix} = \frac{1}{2} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \begin{pmatrix} E_0 \\ e^{i\mu(t)} \end{pmatrix} \begin{pmatrix} \cos(\theta) & -\sin(\theta) \\ \sin(\theta) & \cos(\theta) \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} \cos(\theta) \\ \sin(\theta) \end{pmatrix} \begin{pmatrix} 0 \end{pmatrix}$$

(32)

The initial intensity of light incident is $$I = E_0^2$$ while the intensity detected at the detector is $$I_{\text{det}}$$.

On solving the matrix train we get,

$$E_x = E_y = \frac{E_0}{2} e^{i\mu t} \cos^2 \theta + E_0 \sin \theta \cos \theta$$

(33)

The intensity at the detector $$I_{\text{det}}$$ is given by

$$I_{\text{det}} = E_x^* E_x + E_y^* E_y$$

(34)

After simplification

$$I_{\text{det}} = \frac{1}{2} E_0^2 \cos^2(\theta)[1 + \cos \delta(t) \sin(2\theta)]$$

(35)

The functions $$\cos \delta(t)$$ can be expanded in terms of Bessel function

$$\cos(R_d \cos \omega t) = J_0(R_d) - 2J_1(R_d) \cos 2\omega t + 2J_2(R_d) \cos 4\omega t - 2J_1(R_d) \cos 3\omega t$$

$$\sin(R_d \cos \omega t) = 2J_1(R_d) \cos \omega t - 2J_2(R_d) \cos 3\omega t$$

(36)
Where, $I_0$ is the 0th harmonic dc signal not intensity.

$$\frac{I_{2w}}{I_0} = -\frac{1}{4J_2(R_d)} \times \frac{2J_2(R_d)\sin 2\theta}{1 + J_0(R_d)\sin 2\theta}$$  \hspace{1cm} (37)$$

$$\frac{I_{3w}}{I_0} = 0$$

At $R_d = 2.406 \text{ rad}$, $J_0(2.406) \approx 0$ which simplifies the equation for $\text{Re}[\theta_k]$ and $\text{Im}[\theta_k]$.

The simplified equations are,

---

**Figure 8-2** Kerr angle for a rotating polarizer.

---

$$\frac{I_{2w}}{I_0} = -2J_2(R_d)\sin 2\theta$$  \hspace{1cm} (38)$$

$$\frac{I_{3w}}{I_0} = 0$$

Figure 8-2 shows the calculated $\text{Re}[\theta_k]$ and $\text{Im}[\theta_k]$. The output produces a sinusoidal rotation and no ellipticity.
8.2 Kerr response from a spinning waveplate.

Figure 8-3 Optical setup for measuring the polarization change due to a spinning waveplate.

Light from a laser with electric field $E_0$ is incident on a waveplate mounted on a rotational mount, and is spinning in XY plane. The reflected light passes through a PEM and a polarizer P2 fixed at 45°. Finally the light is detected by a MCT detector.

The optical setup is shown in Figure 8-3. Monochromatic light passes through the spinning waveplate and the change in polarization is detected by the combination of PEM and a polarizer at 45 degree. The intensity of the light is finally detected by a MCT detector.

The Jones matrix train for the setup is

$$E_{det} = (M_{p=45}) (M_{PEM}) (M_{Rot(-\theta)}) (M_{wp}) (M_{Rot(\theta)}) E_{inc}.$$
\[
\begin{pmatrix}
E_x \\
E_y
\end{pmatrix} = \frac{1}{2} \begin{pmatrix}
1 & 1 & e^{i(t)} \\
0 & 0 & 1
\end{pmatrix} \begin{pmatrix}
\cos(\theta) & -\sin(\theta) & 1 & 0 \\
\sin(\theta) & \cos(\theta) & 0 & e^{i\theta}
\end{pmatrix} \begin{pmatrix}
\cos(\theta) & \sin(\theta) \\
-\sin(\theta) & \cos(\theta)
\end{pmatrix} \begin{pmatrix}
E_o \\
E_i
\end{pmatrix}
\] (39)

\[
l_{\text{det}} = \frac{1}{2} E_0^2 \left[ 1 - \sin R \sin \delta(t) \sin 2\theta + \cos \delta(t) \sin^2 \frac{R}{2} \sin 4\theta \right]
\] (40)

For a quarter wave plate \( R = \pi / 2 \),

\[
l_{\text{det}} = \frac{E_0^2}{2} \left[ 1 - \sin \delta(t) \sin 2\theta + \frac{1}{2} \cos \delta(t) \sin 4\theta \right]
\] (41)

Expanding \( \cos \delta(t) \) and \( \sin \delta(t) \) in terms of Bessel function and collecting the harmonics together we get,

\[
\frac{I_{xu}}{I_0} = -\frac{J_1(R\delta) \sin 4\theta}{1 + 0.5 J_0(R\delta) \sin 4\theta}
\] (42)

\[
\frac{I_{yu}}{I_0} = -\frac{2J_3(R\delta) \sin 2\theta}{1 + 0.5 J_2(R\delta) \sin 4\theta}
\] (43)

This system produces a rotation as well as ellipticity with \( 4\theta \) and \( 2\theta \) angle dependence respectively. Figure 8-4 shows the angle dependence of rotation and ellipticity.
Figure 8-4 Kerr angle calculated for a rotating QWP
8.3 Kerr response from a spinning dichroic sample.

A purely dichroic sample has $k_x \neq k_y$ while $n_x = n_y$. The optical setup is shown in Figure 8-5. Light from the laser reflects the rotating linear dichroic sample and the change in polarization is detected by the combination of PEM and a polarizer at 45 degree. The intensity of the light is finally detected by a MCT detector.

Figure 8-5 Kerr setup for measuring the polarization change due to a spinning sample with linear dichroism. Light from a laser with electric field $E_0$ is incident on a dichroic sample mounted on a rotational mount, and is spinning in XY plane. The reflected light passes through a PEM and a polarizer P2 fixed at 45°. Finally the light is detected by a MCT detector.
The Jones matrix for the setup can be written as.

\[ E_{\text{det}} = (M_{p,45})(M_{\text{PEC}})(M_{\text{Rot}(-\vartheta)})(M_{\text{Sample,LD}})(M_{\text{Rot}(\vartheta)})E_{\text{inc}}. \]

\[
\begin{pmatrix}
E_x \\
E_y
\end{pmatrix} = \frac{1}{2} \begin{pmatrix}
1 & 1 & e^{i\vartheta} & 0 & \cos(\vartheta) & -\sin(\vartheta) & A & 0 & \cos(\vartheta) & \sin(\vartheta)
\end{pmatrix}
\begin{pmatrix}
0 & 1 & \sin(\vartheta) & \cos(\vartheta)
\end{pmatrix}
\begin{pmatrix}
0 & B & -\sin(\vartheta) & \cos(\vartheta)
\end{pmatrix}
\begin{pmatrix}
0
\end{pmatrix}
\]

(44)

\[ l_{\text{det}} = \frac{E_0^2}{2} \left[ A^2 + B^2 + (A^2 - B^2) \cos 2\vartheta + (A - B) \cos \delta \sin 2\vartheta (A + B + (A - B) \cos 2\vartheta) \right] \]

(45)

Using the Bessel function expansion of \( \cos \delta \) in the above equation and collecting the terms of different harmonics we get,

\[ l_0 = \frac{E_0^2}{4} (A^2 + B^2 + (A^2 - B^2) \cos 2\vartheta + (A - B) J_0(R_d) [(A + B + (A - B) \cos 2\vartheta) \sin 2\vartheta] \]

(46)

\[ l_{2\omega} = -\frac{E_0^2}{2} [J_0(R_d) [(A + B + (A - B) \cos 2\vartheta) \sin 2\vartheta] \]

(47)

\[ l_{3\omega} = 0 \]

(48)

\[ \frac{l_{2\omega}}{l_0} = \frac{-2 [J_0(R_d) [(A + B + (A - B) \cos 2\vartheta) \sin 2\vartheta]]}{(A^2 + B^2 + (A^2 - B^2) \cos 2\vartheta + (A - B) J_0(R_d) [(A + B + (A - B) \cos 2\vartheta) \sin 2\vartheta]}} \]

(49)

Again we set the \( R_d = 2.406 \text{ rad} \) and \( J_0(2.406) = 0 \) which simplifies the equation for \( \text{Re}[\vartheta] \) and \( \text{Im}[\vartheta] \). The simplified equations are,

\[ \frac{l_{2\omega}}{l_0} = \frac{-2 (A - B) J_1(R_d) \sin 2\vartheta [(A + B + (A - B) \cos 2\vartheta)]}{A^2 + B^2 + (A^2 - B^2) \cos 2\vartheta} \]

(50)
\[ \frac{I_{\infty}}{I_0} = 0 \]  

(51)

This system produces only rotation and no ellipticity signal. Figure 8-6 shows the angle dependence of rotation and ellipticity with A=1.4 and B=1. The rotation signal is similar to the experimental rotation as shown in Figure 4-6, however we are missing the ellipticity.

Figure 8-6 Kerr angle calculated for a rotating sample with LD.
References


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