# The saturation of interband Faraday rotation in $Bi_2Se_3$

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**Abstract** – Bismuth selenide – the prominent topological insulator – has been recently reported as a material exhibiting an extraordinary strong Faraday rotation. This effect has been identified as due to interband excitations in bulk promoting electrons from the valence to the conduction band, in which the electron gas is partially spin-polarized due to the Zeeman effect. In this work, we test the Faraday rotation in $Bi_2Se_3$ in high magnetic fields, when the conduction band electrons reach their full spin polarization. We find that the Faraday angle becomes in this regime almost independent of the applied magnetic field. This contrasts with the Faraday effect observed in this system at low magnetic fields, where the Faraday angle scales linearly with $B$ and may thus be described by a conventional Verdet law.

**Introduction.** – Bismuth selenide ($Bi_2Se_3$) is a narrow-gap semiconductor widely explored in the past due to its promising thermoelectric properties [1]. More recently, this compound [2,3] has been identified as a three-dimensional topological insulator, with a characteristic band inversion of its bulk electronic bands and typical surface states hosting massless electrons [4–6]. The electric and optical properties of bulk $Bi_2Se_3$ are mostly determined by the electronic states at the Γ-point of the Brillouin zone, where nearly parabolic and doubly spin-degenerate conduction and valence bands are separated by a direct band gap, see fig. 1(a). This band structure with a fairly high electron-hole symmetry can be reasonably well described by a simple Dirac-type Hamiltonian for massive particles, which does not imply more than two material parameters: the band gap energy $E_g$ and velocity parameter $v_D$ [7].

Recently, an extraordinary strong for non-magnetic materials Faraday rotation has been reported on $Bi_2Se_3$ [8]. The origin of this effect has been traced back to the presence of conduction band electrons – appearing in this material due to native defects (selenium vacancies). Nevertheless, in contrast to the well-known Faraday rotation due to free charge carriers [9,10], the present effect is not related to intraband, but instead, interband excitations. These excitations promote electrons from the full valence to the partially occupied conduction band, which both are spin-split due to fairly large effective $g$ factors of electrons and holes, while Landau levels are still not well resolved (fig. 1(b)).

The observed Faraday angle $\theta_F$ has been found to be proportional to the applied magnetic field $B$ and may thus be described by the conventional Verdet law, $\theta_F = VBd$, where $d$ stands for the sample thickness and $V$ for the Verdet constant, exceeding in this particular case $10^3\,\mathrm{deg}/(\mathrm{T}\cdot\mathrm{cm})$ [8]. Other contributions to the Faraday rotation, such as due to intraband (cyclotron resonance-like) absorption in bulk or at the surface [9–11], remained in the explored middle infrared spectral range negligible.

In this paper, we probe the Faraday rotation in bulk $Bi_2Se_3$ in high magnetic fields, which allow us to achieve the full spin polarization of electrons in the conduction

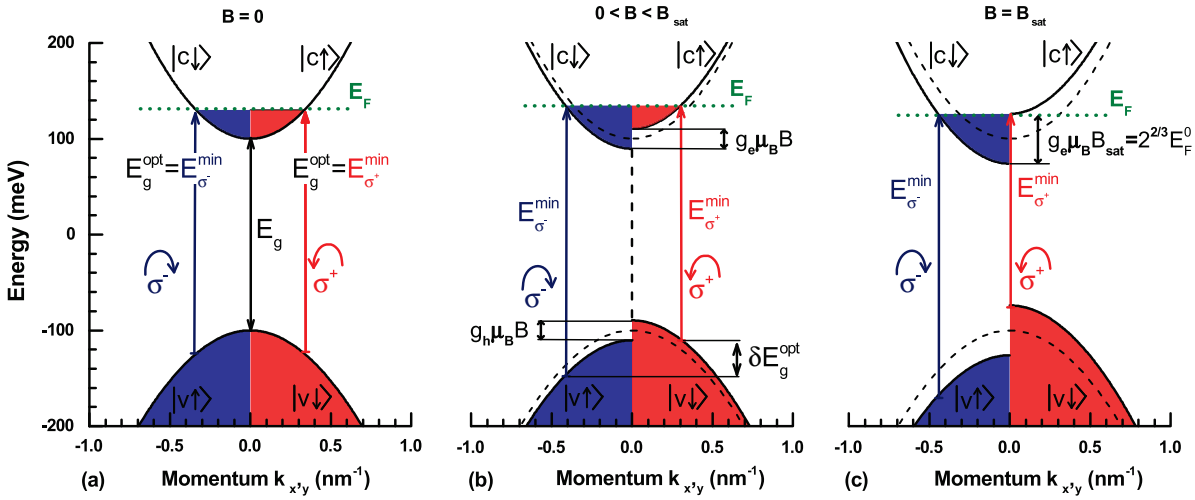


Fig. 1: (Color online) A simplified sketch of electronic bands in $n$-type $Bi_2Se_3$ in the vicinity of the Brillouin zone center. The vertical arrows indicate the lowest in energy interband excitations that are active in a given circular polarization ($E^{\rm min}_{\sigma^+}$ and $E^{\rm min}_{\sigma^-}$) and which are allowed by the occupation effect (Pauli blocking). (a) At $B = 0$, the absorption onset $E^{\rm opt}_g = E^{\rm min}_{\sigma^+} = E^{\rm min}_{\sigma^-}$ follows the standard Burstein-Moss shift of the interband absorption edge [13], $E^{\rm opt}_g = E_g + (1 + m_e/m_h)E^0_F \approx E_g + 2E^0_F$. (b) At $0 < B < B_{\rm sat}$, the splitting of the interband absorption edge $\delta E^{\rm opt}_g = \mu_B B[g_e(m_e/m_h) + g_h]$ appears, thus giving rise to magnetic circular dichroism that scales linearly with $B$. (c) At $B = B_{\rm sat}$, the splitting of the interband absorption edge reaches $\delta E^{\rm opt}_g = \Delta_{\rm sat}$, cf. fig. 3. At $B > B_{\rm sat}$, the splitting $\delta E^{\rm opt}_g$ remains constant (assuming $g_e \approx g_h$), unless sizeable effects of the Landau quantization appear.

band (fig. 1(c)). We show that achieving this full spin polarization is associated with a pronounced change in the Faraday rotation response, giving rise to the Faraday angle nearly independent of $B$ over a fairly broad interval of magnetic fields.

**Experimental details.** – The measurements presented in this manuscript were carried out on (001)-oriented free-standing layers, which were sliced using a microtome machine from a bulk $Bi_2Se_3$ crystal prepared by the standard Bridgman method, see ref. [8] for details. Due to native defects (selenium vacancies), the crystal was $n$-type doped, with the electron density close to $10^{18}\,{\rm cm}^{-3}$.

To measure the Faraday rotation, the studied $Bi_2Se_3$ layer was placed between two collinearly oriented wire-grid polarizers deposited on a silicon substrate. A relatively large thickness of the sample, $d = 225\,\mu$m, was chosen to resolve the Faraday angle with a sufficient precision and resolution in the photon energy. The macroscopic area of this sample ($\approx 2 \times 2\,{\rm mm}^2$) was then exposed to the radiation of a globar, which was analysed by a Fourier transform spectrometer and, using light-pipe optics, delivered to the sample placed in a high-field resistive magnet. The transmitted light was detected by a composite bolometer, mounted directly below the sample and kept at the same temperature as the sample ($T = 2\,{\rm K}$). The obtained data, corrected for magnetic-field–induced variations in the response of the bolometer, are plotted in fig. 2(a)–(c).

The complementary magneto-transmission experiment (fig. 3) was performed, apart from the removed linear polarizers, in an identical experimental configuration. To resolve the field-induced changes in transmission close to the interband absorption edge, a relatively thin layer ($\approx 10\,\mu$m) was prepared for this measurement. The sample with approximately the same position of the interband absorption edge $E^{\rm opt}_g$ (fig. 1(a)) was selected for these measurements to minimize the influence of a certain variation of the electron density present in bulk $Bi_2Se_3$ crystals.

**Results and discussions.** – The relative magneto-transmission spectra $T_B/T_0$ of the $225\,\mu$m thick sample placed between two collinearly oriented wire-grid polarizers are plotted in fig. 2(a) for selected values of the magnetic field. The Faraday effect in $Bi_2Se_3$ is manifested as a pronounced modulation of these spectra. The minima correspond to the Faraday angle $\theta_F = \pi/2$ and its odd-integer multiples, $\theta_F = \pi(k + 1/2)$, $k = 1, 2, 3\ldots$, as shown in the detailed analysis of the spectrum taken at $B = 18\,{\rm T}$ (fig. 2(b)). The amplitude of the modulation, decreasing with $B$, points towards the field-induced ellipticity, which appears due to non-zero absorption in $Bi_2Se_3$ at sub-band-gap photon energies.

The analysis of $T_B/T_0$ spectra, namely the read-out of the minima, maxima and inflection points, allowed us to extract the Faraday angle as a function of the magnetic field. This field dependence has been plotted for three selected photon energies in fig. 2(c). At low magnetic fields, the Faraday angle increases linearly with $B$ and thus may be described by the conventional Verdet law, $\theta = VBd$. This regime has been described in our previous work [8], where the Verdet constant exceeding $10^3\,{\rm deg}\cdot{\rm T}^{-1}\cdot{\rm cm}^{-1}$ in the middle infrared spectral range was reported. The origin of the magnetic circular dichroism, which stands

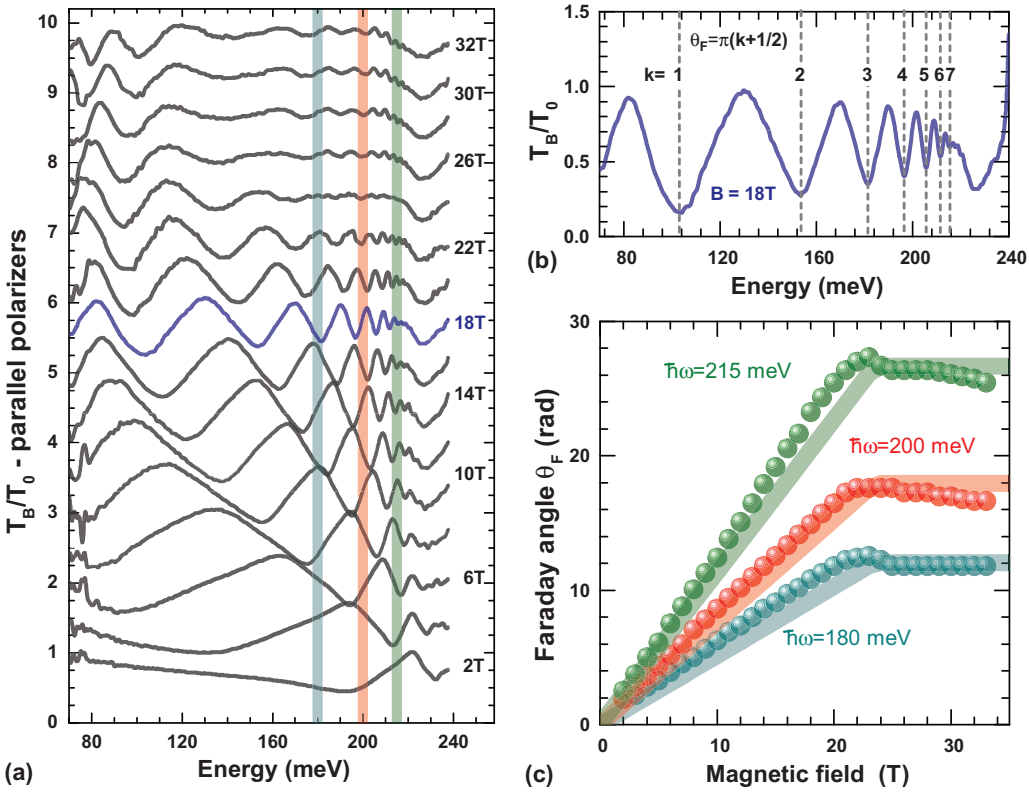


Fig. 2: (Color online) Part (a): relative magneto-transmission spectra $T_B/T_0$ of the 225 $\mu$m thick layer of $Bi_2Se_3$ placed between two collinearly oriented wire-grid polarizers. For clarity, the successive spectra are shifted vertically by 0.6. The minima in $T_B/T_0$ curves correspond to odd-integer multiples of the Faraday rotation angle $\pi/2$ as indicated in detail in the part (b) for the spectrum taken at $B = 18$ T. Part (c): the Faraday angle extracted at three different photon energies indicated in (a) by vertical color lines. The size of the used symbols represents the corresponding error bars. The solid lines correspond to the expectations based on a simple model described in the text.

behind this strong Faraday effect, was identified as due to interband excitations, from the bulk valence to the conduction band with a partly spin-polarized electron gas (fig. 1(b)).

The Faraday angle retains the linear-in-$B$ dependence up to $B_{\rm sat} \approx 23$ T, when the character of the Faraday effect abruptly changes. For $B > B_{\rm sat}$, the Faraday angle remains constant or even a weak decrease of $\theta_F$ with $B$ appears. This saturation behavior is clearly associated with the full spin polarization of the conduction band electrons established in high magnetic fields (fig. 1(c)). For a 3D parabolic band, this is achieved when $g_e\mu_B B_{\rm sat} = 2^{2/3}E_F^0$ [12], where $\mu_B$ is the Bohr magneton and $E_F^0$ is the zero-field Fermi energy, which can be roughly read out from the Burstein-Moss shift [13] of the absorption edge in a system with a small electron-hole asymmetry ($m_e \approx m_h$), $E_F^0 = (E_g^{\rm opt} - E_g)/2 \approx 25$ meV. Assuming the electron $g$ factor estimated from our recent magneto-optical data $g_e \approx 27$ [7] or, alternatively, taken from the recent EPR experiment $g_e = 27.5$ [14], we obtain the theoretically expected saturation field $B_{\rm sat}^{\rm theor} \approx 25$ T, which is in very good agreement with the experimental value.

The magnetic circular dichroism due to interband excitations, responsible for the observed Faraday rotation, is also manifested in the complementary magneto-transmission experiment (fig. 3) by pronounced splitting of the interband absorption edge $\delta E_g^{\rm opt}$. This splitting reflects different onsets of interband absorption for left- and right-handed circularly polarized radiation, as graphically explained in fig. 1(b). As discussed in detail in our previous work [8] this splitting increases at low fields linearly with $B$ and is described by a simple formula $\delta E_g^{\rm opt}(B) = \mu_B B[g_e(m_e/m_h) + g_h]$. Taking parameters deduced in ref. [7] ($m_e/m_h \approx 0.8$, $g_e \approx 27$ and $g_h \approx 24$), we obtain $\delta E_g^{\rm opt}/B = 2.6$ meV/T in good agreement with the experimentally determined value 2.3 meV/T.

In high magnetic fields, the splitting of the interband absorption edge saturates at $\delta E_g^{\rm opt}(B \geq B_{\rm sat}) \approx \Delta_{\rm sat}$ (inset of fig. 3). This implies, consistently with conclusions of ref. [7], that $g_e \approx g_h$. This is because at $B > B_s$, the absorption edge splitting should follow the linear-in-$B$ dependence $\delta E_g^{\rm opt}(B > B_{\rm sat}) = \delta E_g^{\rm opt}(B_{\rm sat}) + (g_h - g_e)\mu_B(B - B_{\rm sat})$, as may be inferred from fig. 1(c). Let us also note that the difference in saturations fields found in the transmission and Faraday rotation experiments in figs. 2(c) and 3 ($B_{\rm sat} \approx 20$ *vs.* 23 T) is related to slightly different electron densities in the investigated samples.

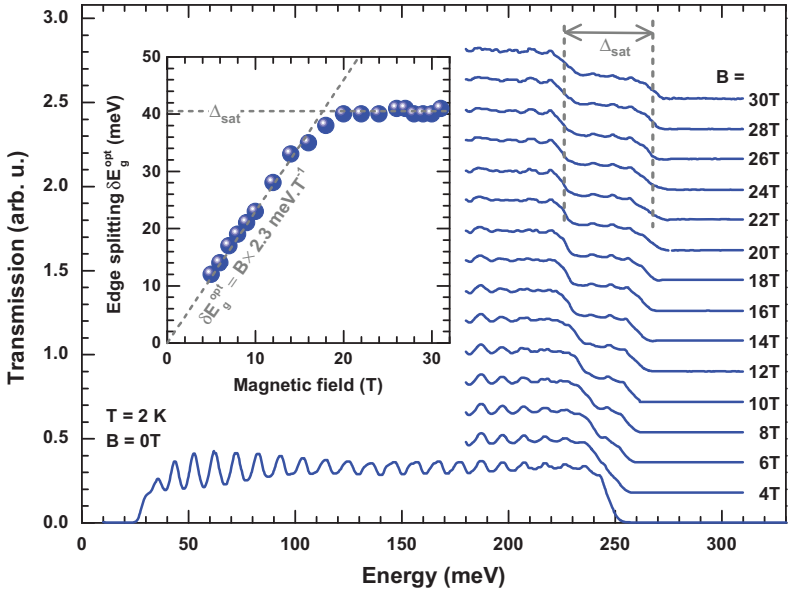


Fig. 3: (Color online) Magneto-transmission of a 10 $\mu$m thick layer of $Bi_2Se_3$ for selected values of $B$. At $B = 0$, the sample is transparent in the spectral window defined at low energies by the plasma frequency and at high energies by the interband absorption, implying the optical band gap $E_g^{\rm opt} = E_g + (1+m_e/m_h)E_F^0 \approx E_g + 2E_F^0$ to the Burstein-Moss shift. With increasing $B$, the interband absorption edge becomes split with respect to the circular polarization of light (see ref. [8] for polarization-resolved spectra). This splitting increases linearly with $B$ and above $B_{\rm sat}$ becomes saturated at $\Delta_{\rm sat}$. The modulation of the curves correspond to the interference fringes appearing due to nearly plan-parallel surfaces of the sample. The separation of fringes allows us to estimate the index of refraction at sub-band-gap photon frequencies: $n \approx 6$.

Let us now describe the observed Faraday rotation quantitatively. At low magnetic fields, when the Faraday angle increases linearly with $B$, the simple model developed in our previous work [8] is applicable and the Verdet constant can be expressed as

$$V(\omega) = \zeta \frac{v_D^2}{n} \frac{\sqrt{B_s}}{E_g^{\rm opt}} \frac{\hbar\omega}{(E_g^{\rm opt})^2 - (\hbar\omega)^2}, \qquad (1)$$

where $\zeta = 8e\alpha\sqrt{2^{1/3}\hbar e}/\pi$, $n$ is the refraction index and $\alpha$ the fine-structure constant. This formula has been derived for a system described by the Dirac-type Hamiltonian for massive particles [7,15,16] which is characterized by a full electron-hole symmetry, and thus implies simplified expressions for effective masses and $g$ factors: $m_e = m_h = E_g/v_D^2$ and $g_e = g_h = 4m_0 v_D^2/E_g$ ($m_0$ is the bare electron mass) [7]. A correction due to the anisotropy of the system, implying the effective mass roughly twice larger along the $c$-axis has been introduced [17,18].

We emphasize that the formula (1) does not include any freely tunable parameters: the velocity parameter is known, *e.g.*, from ARPES experiments, but also from our recent Landau level spectroscopy of thin $Bi_2Se_3$ layers: $v_D = 0.47 \times 10^6$ m/s [7]. The refractive index $n \approx 6$ and the optical band gap $E_g^{\rm opt} \approx 250$ meV are directly read from the zero-field transmission spectra taken on a thin sample (fig. 3), from the interference fringes and the position of the interband absorption edge, respectively. Taking these parameters together with the saturation field $B_{\rm sat}$, the calculated Faraday angle $\theta_F = VBd$ fairly well describes the experimental data in fig. 2(c) at magnetic fields below $B_{\rm sat}$.

It is important to note that the simple analytical expression for the Verdet constant (1) was derived only for low magnetic fields when the spin-splitting $E_s = g_e\mu_B B$ is small with respect to the Fermi energy, $E_s \ll E_F$. Nevertheless, it remains approximately valid up to the magnetic field of $B_{\rm sat}$ when $E_s = g_e\mu_B B_{\rm sat} = 2^{2/3}E_F^0$. This is due to the fact that the (joint) density of states $jDOS^{\pm}(\omega) \propto \sqrt{\hbar\omega - E_g}$ is the same for both circular polarizations. Together with the occupation of the spin-split conduction band, this implies that the Verdet constant is proportional to the spin polarization of the electron gas: $V \propto \delta N = N^{\downarrow} - N^{\uparrow}$. This approximation is valid for relatively low-doped systems, $E_F \ll E_g$, and for photon frequencies not too close to the optical band gap, $\hbar\omega < E_g^{\rm opt}$. The Faraday rotation due to interband excitations thus becomes sensitive to the net spin polarization, or in other words, to the magnetization of the electron gas, described in the model of Pauli paramagnetism for non-interacting electrons [19]. Indeed, the spin polarization of a 3D electron gas remains almost linear in $B$ up to the saturation field of $B_{\rm sat}$, which validates the formula (1) up to the magnetic fields as high as $B_{\rm sat}$.

At $B > B_{\rm sat}$, when the full spin polarization is established ($N = \delta N = N^{\downarrow}$), the Faraday angle becomes independent of $B$ and stays at the value of $\theta_F = VB_{\rm sat}d$. The saturation of the Faraday angle above $B_{\rm sat}$ thus may be

viewed as a signature of the full spin polarization (magnetization) of the electron gas. At the same time, however, we should keep in mind that the saturation of $\theta_F$ can only be observed when the condition $g_e = g_h$ is fulfilled. A small deviation from this condition ($g_e \gtrsim g_h$) may imply the weak decrease of the Faraday angle at $B > B_s$, see fig. 2(c). A more significant deviation from this condition would lead to a more complex dependence of the Faraday angle, which can only be described by more sophisticated models for Faraday rotation, see, *e.g.*, refs. [10,20,21].

Let us also note that Faraday rotation proportional to the spin polarization of electrons may be reminiscent of the Faraday effect due to the Raman spin-flip of electrons bound to indium donors in CdS, reported by Romestain *et al.* [22], and later on, of free conduction-band electrons in InSb and HgCdTe [23,24], which also depends linearly on $\delta N$. In our case, however, the excitations responsible for the studied Faraday rotation are dipole-active interband excitations and no spin-flip Raman processes are involved.

Interestingly, the presented quasi-classical description of the observed Faraday effect remains valid up to high magnetic fields, when the system is in the quantum regime, with well-resolved Landau levels ($\mu.B > 1$), and even the quantum limit is approached (with only the lowest Landau level occupied). As a matter of fact, the Zeeman splitting in the conduction band of $Bi_2Se_3$ is nearly twice the cyclotron energy, $g_e\mu_B B \approx 2\hbar\omega_c$ ($\omega_c = eB/m_e$) [7,25]. This allows us to estimate the magnetic field at which the quantum limit should be achieved $B_L = (1+\sqrt{2})^{2/3} B_{\rm sat} \approx 40\,\mathrm{T}$.

Indeed, the Faraday angle calculated in this quantum limit, when the magneto-optical dichroism is induced due to excitations into the partially occupied lowest electronic Landau level, implies a nearly identical result, with the spectral profile $\theta_F \propto x/(1-x^2)$, where $x = \hbar\omega/(E_g+\hbar\omega_c)$. The crossover from the quasi-classical regime, with spin-split but still parabolic bands, to the quantum regime, with fully resolved Landau levels, is thus not manifested by any pronounced change in the Faraday effect at sub-band-gap photon energies. The effects due to the Landau quantization are expected in magneto-optical response at photon energies above the band gap. Such interband inter-Landau level resonances profoundly modulate the transmission spectra (for $Bi_2Se_3$ see ref. [7]) and also give rise to a fairly complex Faraday rotation response (seen in various narrow-gap semiconductors, see, *e.g.*, [26,27]). In our case, however, the investigated samples, with the thickness of ten microns and more, are fully opaque in this spectral region.

**Conclusions.** – We have studied Faraday rotation in $Bi_2Se_3$ in a broad range of magnetic fields at sub-band-gap photon energies, where this effect is governed by interband excitations, from the valence to partly occupied conduction band. We have found that the Faraday effect basically follows the spin polarization of the conduction band electrons. At low fields, the Faraday angle increases linearly with $B$, reflecting thus the partial spin polarization of the electron gas. Once the full spin polarization of electrons is achieved, the character of the Faraday rotation changes and the Faraday angle intriguingly remains nearly constant with $B$.

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