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Beating oscillations of magneto-optical spectra in simple hexagonal graphite

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ABSTRACT

The magneto-optical properties of simple hexagonal graphite exhibit rich beating oscillations, which are dominated by the field strength and photon energy. The former has a strong effect on the intensity, the energy range of the beating and the number of groups, and the latter modulates the total group numbers of the oscillation structures. The single-particle and collective excitations are simultaneously presented in the magnetoreflectance spectra and can be precisely distinguished. For the loss function and reflectance, the beating pattern of the first group displays stronger intensities and broader energy range than other groups. Simple hexagonal graphite possesses unique magneto-optical characteristics that can serve to identify other bulk graphites.

1. Introduction

Graphene-based materials have attracted many researchers to investigate their physical properties due to their potential for novel applications. The material properties demonstrate their strong dependence on the stacking configurations [1–6], layer numbers [7–12], and interlayer atomic interactions [13–15]. Graphite is composed of van der Waals coupled graphene layers [16,17]. Three prototypes of periodical stacking along the z-direction exist: AA-stacked simple hexagonal graphite (SHG), AB-stacked Bernal graphite (BG), and ABC-stacked rhombohedral graphite (RHG). The graphites discovered in nature are mainly composed of BG and RHG. Recently, SHG has been successfully synthesized in the laboratory [18]. The interlayer couplings play an important role in determining the low-energy electronic properties; thus, different periodic stacking configurations exhibit their own unique characteristics. It is known that the neighboring electronic states congregate and form the Landau subbands (LSs) along $\widehat{k}_z$ in a perpendicular uniform magnetic field $\mathbf{B} = B_0\widehat{z}$. The magneto-electronic properties demonstrate very interesting phenomena, e.g., the anisotropy of low-energy electronic structures [16,19–21], the de Haas-van Alphen effect [22,23], quantum Hall effect [24–27]. In this work, we mainly focus on obtaining the magneto-optical properties of SHG by means of evaluating the dielectric function $\varepsilon(\omega, B_0)$. Comparisons with BG and RHG are also made.

The LSs of graphites present many important features. SHG possesses very strong k_z-dependent energy dispersions with a broad band width about 1 eV, and each LS can be described by a simple relationship with k_z [19,25]. Many LSs cross the Fermi level ($E_F = 0$) [19,28]. Moreover, the excitations related to the densely low-lying LSs own wide energy ranges which can overlap for different optical transition channels. On the contrary, RHG exhibits weak k_z-dependent dispersions with a narrow band width ($\sim$10 meV) [21]. Only one LS crosses E_F, and there is no coexisting energies for different optical excitations. The LS can be characterized by the approximate solution [29]. The energy dispersion of BG has a band width of $\sim$0.2 eV, which lies between that of SHG and RHG, and two LSs cross E_F [30,31]. The low-lying LSs are complex and cannot be easily described by k_z. The characteristics of LSs would be reflected in the magneto-optical spectra.

The magneto-optical properties are closely associated with the stacking configurations of graphites [28,31–33]. The low-energy magneto-optical absorption spectrum of SHG is dominated by intraband and interband optical excitations which induce a multi-channel threshold peak, several two-channel peaks, and many

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double-peak structures [28]. In the magneto-optical absorption spectra of BG, the prominent peaks originate from the interband excitations at both the K and H points. The peaks associated with the K point display double-peak structures [20,30]. Moreover, the field evolution of the absorption lines for the K-point type shows an approximately linear dependence, while the dependence of the H point is square-root like [20,30,34]. The magnetoreflectance $R(\omega, B_0)$ spectra of BG displays irregular oscillations [35,36]. Up to now, no theoretical calculations and experimental measurements for the magneto-optical absorption or reflection of RHG have been performed.

The magneto-optical properties are evaluated based on the Peierls tight-binding model, which can be exactly diagonalized even with the inclusions of field-induced Peierls phases and important atomic interactions in the Hamiltonian [28,31,37]. This study shows that the beating patterns of the dielectric function can be formed mainly owing to the strong overlap of different optical transition channels in a wide frequency range. Such beating patterns are also exhibited in the higher-frequency absorption spectrum, loss function, and reflectance. The single-particle and collective excitations can be precisely identified, respectively, based on the shoulders (peaks) and dips of specific structures in the magnetoreflectance spectra. The regular beating magneto-optical spectra can be controlled by the field strength and the photon energy, which provide a theoretical basis for future experiments to clarify the optical responses of the graphite configurations.

The generalized tight-binding model deserves a closer examination in numerical calculations. We developed this model to study the magneto-electronic and magneto-optical properties by the exact diagonalization method. In studying the magneto-electronic properties, the earlier work can only cope with eigenvalues and eigenfunctions at strong magnetic field strength [38] because the Hamiltonian matrix gets too large as the field strength decreases. For example, this matrix is 40 000 × 40 000 for monolayer graphene at 7.9 T. By means of rearranging the tight-binding functions, it is possible to transform the huge matrix into a band-like one. Therefore, the eigenvalues and the wave functions can be efficiently solved at weaker field strength (∼1 T) [37]. In this work, the magneto-optical absorption spectra, which are determined by three large matrices due to the Hamiltonian, the initial state and the final state, can be obtained by using the localized features of the magnetic wave functions. The PC clusters are sufficient in calculating numerical data. The acquired features of LS spectra and the reliable characterization of the LS wave functions provide a guideline for other physical properties, such as Coulomb excitations and transport properties. As for the discussion of the optical properties in our previously published works, the generalized tight-binding method has been successfully applied to investigate the magneto-optical absorption spectra of few-layer graphenes. The optical selection rules are well defined through the detailed analysis on the wave functions. It is also applicable to bulk graphite with layers stacked in any sequence. Furthermore, the generalized tight-binding model can be used in the cases of spatially modulated fields and combined magnetic and electric fields.

2. Methods

For calculation purposes, the geometric structure of simple hexagonal graphite is regarded as a stacking sequence of infinite layers of graphene with an AA-stacked configuration along $\widehat{z}$. All honeycomb structures in SHG have the same projections on the x–y plane. The interlayer distance is $I_c = 3.50$ Å [18] and the C–C bond length is $b' = 1.42$ Å. A primitive unit cell consists of two atoms. The associated hopping integrals γ_i's taken into account are the one intralayer atomic interaction ($\gamma_0 = 2.519$ eV) and three interlayer atomic interactions ($\gamma_1 = 0.361$ eV; $\gamma_2 = 0.013$ eV; $\gamma_3 = -0.032$ eV) [15].

When SHG is subjected to a $B_0\widehat{z}$, the path integral of the vector potential induces a periodical Peierls phase (details in Ref. [19]). The phase term of the associated period is inversely proportional to the magnetic flux ($\Phi = 3\sqrt{3}b'^2B_0/2$) through a hexagon. To satisfy the integrity of the primitive cell, the ratio $R_B = \Phi_0/\Phi$ ($\Phi_0 = hc/e$ flux quantum) has to be a positive integer. As a result, the extended rectangular unit cell has $4R_B$ carbon atoms. The π-electronic Hamiltonian built from the $4R_B$ tight-binding functions is a $4R_B \times 4R_B$ Hermitian matrix. To solve this huge matrix problem, one can convert the Hamiltonian matrix into a band-like form by rearranging the tight-binding functions [19,29,31]. Both eigenvalue $E^{c,v}$ and eigenfunction $\Psi^{c,v}$ are efficiently obtained, even for a small magnetic field. The superscripts c and v, respectively, represent the conduction and valence bands.

The main features of the electronic properties can be directly manifested by optical excitations. As materials absorb photons, electrons are excited from occupied states to unoccupied states. Within the relaxation-time approximation [39], the transverse dielectric function at zero temperature is expressed as

$$\varepsilon(\omega, B_0) = \epsilon_0 - \frac{e^2}{\pi^2}\sum_{n,n'}\sum_{h,h'=c,v}\int_{1stBZ} d^3\mathbf{k}\frac{\left|\left\langle\Psi_{n'}^{h'}(\mathbf{k})\left|\frac{\widehat{E}\cdot\mathbf{P}}{m_e}\right|\Psi_n^h(\mathbf{k})\right\rangle\right|^2}{\omega_{hh'}^2(n,n';\mathbf{k})} \times\left\{\frac{1}{\omega-\omega_{hh'}(n,n';\mathbf{k})+i\Gamma}-\frac{1}{\omega+\omega_{hh'}(n,n';\mathbf{k})+i\Gamma}\right\}, \quad (1)$$

where $\epsilon_0 = 2.4$ is the background dielectric constant [40]. $\omega_{hh'}(n, n';\mathbf{k}) = E^{h'}(n',\mathbf{k}) - E^h(n,\mathbf{k})$ is the optical excitation energy which comes from the intraband ($c \to c$; $v \to v$) or interband excitations ($v \to c$); $\Gamma(= 3.5$ meV) is the broadening parameter due to the deexcitation mechanisms. In these optical excitations, the momentum of the photons is nearly zero and thus the excitations can be regarded as a vertical transition between two LSs. The initial and final states have the same wavevector, i.e., $\triangle k_x = 0$, $\triangle k_y = 0$, and $\triangle k_z = 0$ [28]. The velocity matrix element $D_m = \left\langle\Psi_{n'}^{h'}(\mathbf{k})\left|\frac{\widehat{E}\cdot\mathbf{P}}{m_e}\right|\Psi_n^h(\mathbf{k})\right\rangle$ is evaluated within the gradient approximation [39,41]. As $|D_m|^2/\omega_{hh'}^2$ is set to be a constant, the imaginary part of $\varepsilon(\omega, B_0)$ is simply the joint density of states $D_J(\omega, B_0)$. The evaluation of $\varepsilon(\omega, B_0)$ can be employed to study the absorption spectrum, loss function, and reflectance.

3. Results and discussion

The perpendicular magnetic field causes the cyclotron motion in the x–y plane; therefore, the Landau levels lie on the k_x–k_y plane and the LSs along $\widehat{k}_z$. The energy dispersions of the LSs along the K–H line ($0 \le k_z(\pi/I_c) \le 1$) exhibit a strong dependence on k_z, as shown in Fig. 1. Based on the node structure of the Landau wave functions, the quantum number $n^c(n^v)$ for each conduction (valence) LS can be identified by the total number of nodes [28]. The LSs with n^c and those with n^v are asymmetric about $E_F = 0$ because of the interlayer atomic interactions. In optical excitations, electrons are excited from occupied LSs into unoccupied LSs. For the sake of convenience, the excitations between two LSs with quantum numbers $n^{c,v}$ and $m^{c,v}$ are represented as $[n^{c(v)}, m^{c(v)}]$ and (n^v, m^c) for intraband and interband excitations, respectively. Moreover, the wave functions of occupied and unoccupied states offer important insights into the possible excitation channels. Since the LS wave functions of SHG are similar to those of monolayer graphene, the same selection rule $|\triangle n| = |m^{c,v} - n^{c,v}| = 1$ applies [28,42,43].