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Magneto-transmission of multi-layer epitaxial graphene and bulk graphite: A comparison

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ABSTRACT

The magneto-transmission of a thin layer of bulk graphite is compared with spectra taken on multi-layer epitaxial graphene prepared by thermal decomposition of a SiC crystal. We focus on the spectral features evolving as $\sqrt{B}$, which are evidence for the presence of Dirac fermions in both materials. Whereas the results on multi-layer epitaxial graphene can be interpreted within the model of 2D Dirac fermions, the data obtained on bulk graphite can only be explained taking into account the 3D nature of graphite, e.g. by using the standard Slonczewski–Weiss–McClure model.

1. Introduction

Within the last few years, the properties of massless and massive "relativistic" particles have been probed in magneto-optical experiments carried out on several carbon-based systems: graphene monolayers [1,2] and bilayers [3], multi-layer epitaxial graphene [4–7], and bulk graphite [8–11]. These experiments offer an important insight into the electronic structure of these materials; in particular, they directly demonstrate the unusual $\sqrt{B}$-scaling of Landau levels (LLs) of Dirac fermions, where B is the magnetic field. Naturally, these experiments stimulated intensive theoretical work in this area [12–16].

Recently, remarkable experiments have been performed on the same systems without an applied magnetic field. A universal value of optical conductivity was demonstrated in graphene [17] as well as in bulk graphite [18]. The optical response of graphene monolayer and bilayer samples as a function of the gate voltage was reported in several experimental works (see Refs. [19–22]), and this topic has also been addressed theoretically [23–25]. The first results of time-resolved optical experiments on multi-layer graphene are also available [26,27].

In this paper, we present a direct comparison of the magneto-transmission spectra taken on a thin layer of bulk graphite and multi-layer epitaxial graphene. We focus on spectral features exhibiting a $\sqrt{B}$-dependence, which corresponds to the optical response of Dirac fermions, and show that there are essential differences in the spectra of both materials.

2. Experimental details

The investigated multi-layer graphene sample was prepared by thermal decomposition on the carbon face of a 4H-SiC substrate [28,29], and it contains around 100 graphene layers. The thin layer of bulk graphite was prepared by simple exfoliation as described in Ref. [9]. As no significant difference in the FIR optical response of natural graphite and HOPG was found [10], we present only results taken on natural graphite due to its higher crystalline quality. Both samples were characterized using micro-Raman. In the case of multi-layer epitaxial graphene, the Raman signature

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of decoupled layers, equivalent to a single flake of exfoliated graphene [30], as well as of additional graphite residuals were found; see Ref. [31]. The Raman spectra taken on a thin graphite layer dominantly showed a multi-component 2D band typical of many Bernal-stacked sheets, but some minor traces of decoupled layers were found; see the discussion in Ref. [10].

To measure the FIR transmittance of the sample, the radiation of a globar, delivered via light-pipe optics to the sample and detected by a Si bolometer placed directly below the sample, was analyzed by a Fourier transform spectrometer. All measurements were performed in the Faraday configuration with the magnetic field applied normal to the graphene/graphite layers. All the spectra were taken with non-polarized light in the spectral range 5–350 meV, limited further by several regions of low tape transmissivity or the SiC opacity; see the grey areas in Fig. 1.

3. Results and discussion

A comparison of the transmission spectra taken on multi-layer epitaxial graphene and a thin layer of bulk graphite is presented in Fig. 1. Starting with the results taken on multi-layer layer graphene in Fig. 1(a), a series of absorption lines is observed and denoted by Roman letters, following the notation introduced in Ref. [4]. Assuming the LL spectrum of graphene, $E_n = \tilde{c}\sqrt{2e\hbar Bn}$, together with the corresponding selection rules for dipole-allowed transitions $\Delta n = \pm 1$, the observed absorption lines B, C, D, E and F can be clearly identified as inter-LL transitions $L_{-m} \rightarrow L_{m+1}$ and $L_{-(m+1)} \rightarrow L_m$ with $m = 0, 1, 2, 3$ and 4, respectively. The Fermi velocity is found to be $\tilde{c} = (1.02 \pm 0.01) \times 10^6$ m s^{-1}. Hence, these results are fully consistent with a model of 2D Dirac fermions, and individual sheets in multi-layer epitaxial graphene indeed behave as if they are electronically decoupled. This was recently explained by a random mutual rotation of adjacent sheets in this material [32]. Similar spectra were also measured on a single sheet of exfoliated graphene [1]; however, the Fermi velocity seems to be enhanced by ≈10%, and these data also suggest some influence of electron–electron interaction, not observed in the transmission spectra of multi-layer epitaxial graphene.

The results obtained on a thin layer of bulk graphite (see Fig. 1(b)) are more complex. Basically, all the absorption lines observed in multi-layer graphene are also found in spectra of bulk graphite with practically the same Fermi velocity $\tilde{c} = (1.02 \pm 0.02) \times 10^6$ m s^{-1}. This justifies the same notation using Roman letters. In addition to these lines, another series of transitions, denoted by Greek letters and clearly exhibiting the $\sqrt{B}$-dependence, is present in the transmission spectra. These lines cannot be assigned to any dipole-allowed transitions between LLs in graphene. Nevertheless, the energies of the additional lines α, γ, δ and ε exactly match transitions that are symmetric around the Dirac point, $L_{-m} \rightarrow L_m$, with indices $m = 1, 2, 3$ and 4, respectively. The β line can be identified as transitions $L_{-1(-3)} \rightarrow L_{3(1)}$.

To explain the absorption lines denoted by the Greek letters we have to abandon the simplified model of 2D Dirac fermions and consider the full band structure of bulk graphite. According to the standard SWM model [33,34], Dirac fermions are located in the vicinity of the H point, where the bands E_1, E_2 and the doubly degenerate E_3 are close to the Fermi level. If the magnetic field is applied along the c-axis of the crystal, Landau levels or more accurately, Landau bands are created, see Fig. 2, having at the H point ($k_z = 0.5$) an analytical form [35,36] ($n \geq 1$):

$$E_3^{-1} = 0$$
$$E_3^0 = \Delta$$
$$E_{3\pm}^n = E_{1,2}^{n-1} = \frac{\Delta}{2} \pm \sqrt{\frac{\Delta^2}{4} + \xi Bn}, \quad (1)$$

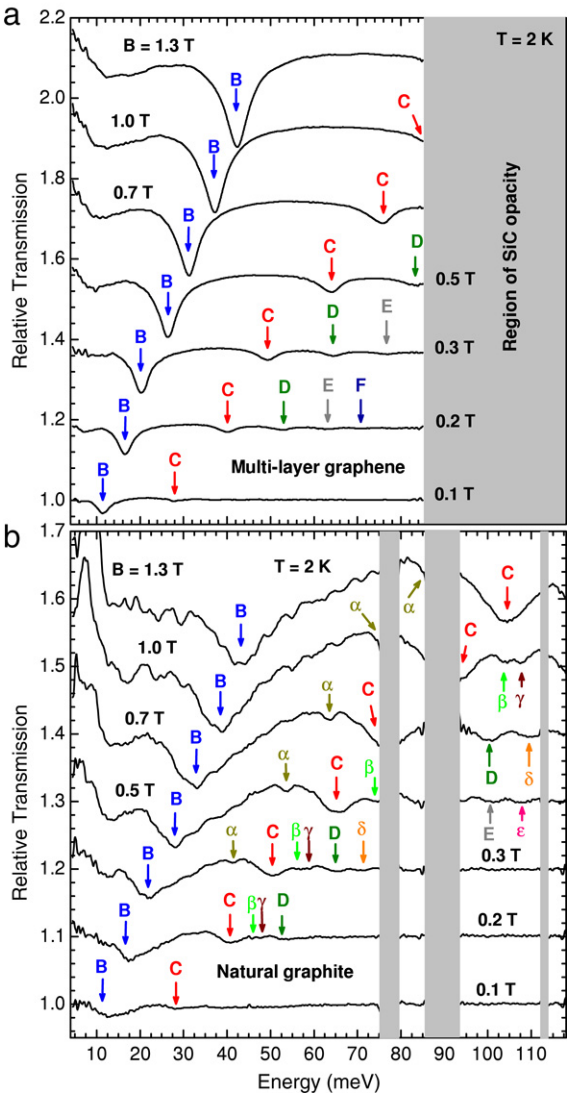

Fig. 1. Transmissions spectra of multi-layer epitaxial graphene (a) and bulk graphite (b) for selected magnetic fields at $T = 2$ K. The absorption lines corresponding to dipole-allowed transitions in graphene are denoted by Roman letters. Greek letters are used for additional transitions which scale as $\sqrt{B}$ and are only found in spectra taken on bulk graphite. For clarity, the spectra in part (a) and (b) were shifted by 0.18 and 0.10, respectively.

where Δ denotes the pseudogap, i.e. the separation between E_1 (and also E_2) and E_3 bands at $k_z = 0.5$ and $B = 0$. The parameter ξ is related to the Fermi velocity as $\xi = 2\tilde{c}^2 e\hbar$.

The form of LLs (1) implies several important H point-related optical properties of graphite. It suggests that the LL energy spectrum typical of graphene is present also at the H point in graphite, when the pseudogap $|\Delta|$ is small in comparison to the energies of the LLs. Experimentally, $|\Delta|$ is found to be significantly below 10 meV (see Refs. [9,36,37]), and therefore magnetic fields above 100 mT are sufficient to ensure this condition. On the other hand, each LL in Eq. (1) is (with the exception of E_3^0 and E_3^{-1}) doubly degenerate: $E_{3+}^n = E_1^{n-1}$ and $E_{3-}^n = E_2^{n-1}$; see Fig. 2. This double degeneracy is in addition to the spin and valley degeneracies present in graphene. Obviously, taking account of the same selection rules $\Delta n = \pm 1$, we obtain a considerably richer set of possible dipole-allowed transitions for graphite in comparison to graphene. For instance, the transition $L_{-1} \rightarrow L_1$ is strictly forbidden in graphene; nevertheless, the absorption line at this energy (α transition) is observed in bulk graphite at the H point due to the dipole-allowed transitions $E_{3-}^1 \rightarrow E_1^0$ and $E_2^0 \rightarrow E_{3+}^1$;