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# Magneto-transmission as a probe of Dirac fermions in bulk graphite

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**Abstract**
Far-infrared magneto-transmission spectroscopy has been used to probe 'relativistic' fermions in highly oriented pyrolytic and natural graphite. Nearly identical transmission spectra of those two materials are obtained, giving the signature of Dirac fermions via absorption lines with an energy that scales as $\sqrt{B}$. The Fermi velocity is evaluated to be $\tilde{c} = (1.02 \pm 0.02) \times 10^6$ m s$^{-1}$ and the pseudogap at the $H$ point is estimated to $|\Delta| < 10$ meV.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

The rush, which started after the discovery of Dirac-like particles in graphene [1–3], naturally resulted in an intensified interest in the properties of bulk graphite with the apparent aim to prove directly the presence, and to investigate the nature, of massless holes in the vicinity of the $H$ point, where according to the standard band structure of graphite, formulated by Slonczewski, Weiss and McClure (SMW), these relativistic fermions are located [4, 5].

The first sign of such particles was reported by Toy *et al* [6] via the $\sqrt{B}$-dependent features in their magneto-reflection experiment, which is a direct fingerprint of Dirac particles. Further evidence did not come earlier than in the 'graphene age' with the reinterpretation of the Shubnikov–de Haas (SdH) and de Haas–van Alphen experiments. The analysis of the phase of these quantum oscillations, carried out by Luk'yanchuk and Kopelevich [7, 8], suggested the presence of normal (massive) electrons with a Berry phase 0 and Dirac holes with a Berry phase $\pi$ in bulk graphite. Another proof of particles with a linear dispersion around the $H$ point was offered by angular-resolved photoemission spectroscopy (ARPES) performed by Zhou *et al* [9] and later on also by Grüneis *et al* [10]. Both massive and massless particles were identified in scanning tunneling spectroscopy (STS) by Li and Andrei [11] and recently also in far-infrared (FIR) magneto-transmission [12].

Hence, many different experimental techniques indicate the presence of Dirac holes in bulk graphite. Nevertheless, the mutual consistency of individual reports represents a serious difficulty, which merits closer scrutiny. The analysis of quantum oscillations [7, 8] assumes that 'bulk' graphite is a system composed of graphene single- and few-layers [13] and thus no crystal ordering along the $c$ axis exists, which directly contradicts the commonly accepted SWM model. On the other hand, the available ARPES results are in relatively good agreement with the SWM model, but are characterized by a relatively low accuracy and probe the sample surface only. The sensitivity to the sample surface only is also a problem for STS experiments, which additionally failed to reveal Dirac holes in earlier equivalent measurements [14].

The available FIR experiments are also not trouble-free when compared, as the Dirac holes were observed in [6, 12], but not seen in recent magneto-reflection experiments [15]. In principle, these optical experiments support the validity of the SWM model. Nevertheless the relative strength of spectral features related to the $H$ point in comparison to the $K$ point is not in agreement with theoretical calculations of optical conductivity in magnetic fields [16]. Very recently, the SWM model was found by Kuzmenko *et al* [17] to be fully sufficient to explain the temperature dependence of the reflectivity of graphite. Note that the graphite band structure parameters estimated using optical methods [6, 12], for example the pseudogap at the $H$ point, differ from those obtained from

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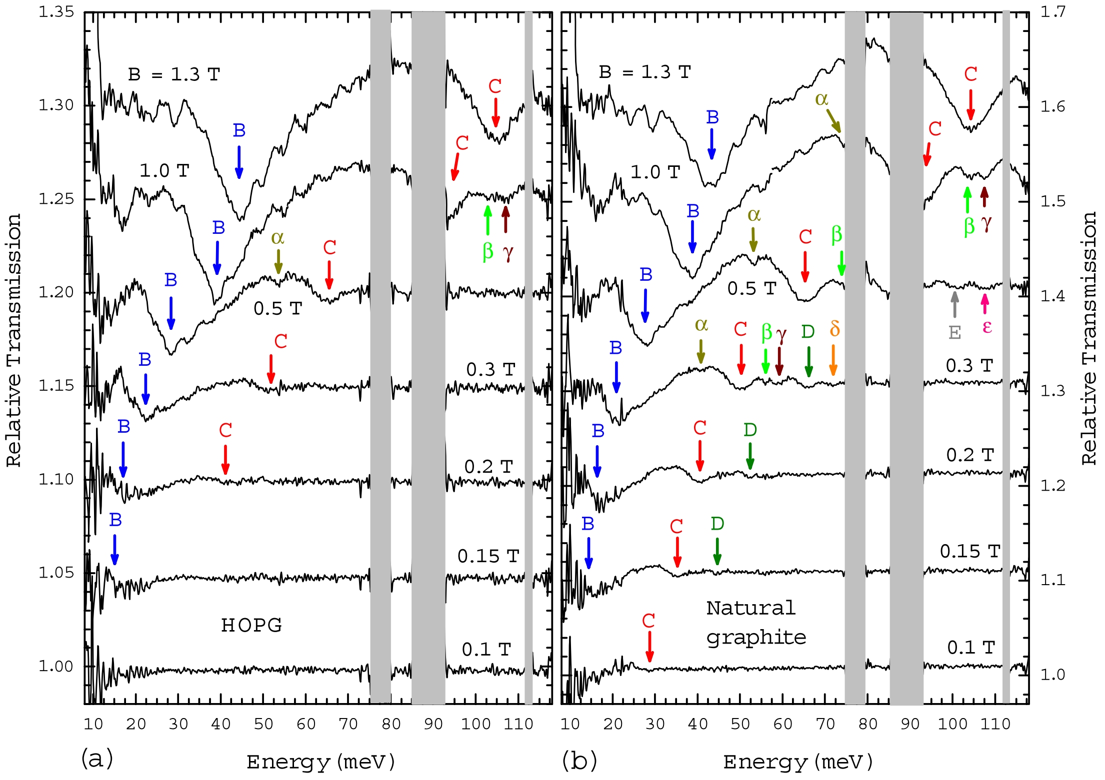

**Figure 1.** Magneto-transmission spectra of HOPG (a) and NG (b) taken in the 10–120 meV spectral window. The individual spectra in (a) and (b) were shifted vertically with a step of 0.05 and 0.1, respectively.

ARPES experiments [10]. A similar problem occurs for the position of the Fermi level, as the hole density inferred from the analysis of quantum oscillations [7, 8] is almost an order of magnitude higher than that obtained from FIR spectroscopy [12].

To explain the contradictory results obtained using different experimental methods, among others, material properties of various types of bulk graphite are nowadays under discussion. For instance, highly oriented pyrolytic graphite (HOPG) is assumed in [7, 8, 13, 18] as being a more anisotropic, nearly 2D material, in comparison with Kish graphite or natural graphite (NG). Similar behavior was reported in STS experiments [14] performed on HOPG and Kish graphite.

In this paper, we present FIR magneto-transmission measurements performed on two types of bulk graphite, HOPG and NG. We focus on spectral features exhibiting $\sqrt{B}$ dependence, which serve as a fingerprint of Dirac-like particles. We find no significant difference in the optical response of both materials and evaluate the same Fermi velocity $\tilde{c} = (1.02\pm0.02)\times10^6$ m s$^{-1}$ as well as the pseudogap at the $H$ point $|\Delta| < 10$ meV.

## 2. Experiment

Thin samples for transmission measurements were prepared by exfoliation of HOPG (ZYA grade) and NG in the way described in [12] and characterized using micro-Raman. Probing the sample prepared from HOPG [12], the detected Raman signal corresponded to bulk graphite [19]. More complex results were obtained on the NG sample, where the shape of the 2D feature in the Raman spectrum varied on scanning across both the sample and the NG crystal from which the sample was prepared, indicating the presence of bulk graphite as well as few-layer graphite stacks [20]. Graphite flakes covered around 50% and up to 70% of the tape surface in the case of HOPG and NG, respectively. Transmission experiments were carried out on a macroscopic circular-shaped sample with a diameter of several millimeters.

The FIR experiments have been performed using the experimental set-up described in [12, 21]. To measure the transmittance of the sample, the radiation of globar, delivered via light-pipe optics to the sample and detected by an Si bolometer, placed directly below the sample and cooled down to a temperature of 2 K was analyzed by a Fourier transform spectrometer. All measurements were performed in the Faraday configuration with the magnetic field applied along the $c$ axis of the sample. All the spectra were taken with non-polarized light in the spectral range of 10–350 meV, limited further by several regions of low tape transmissivity (see gray areas in figures 1–3). The transmission spectra were normalized by the transmission of the tape and by the zero-field transmission, thus correcting for magnetic-field-induced variations in the response of the bolometer.

## 3. Results and discussion

The FIR transmission spectra of HOPG and NG, taken at a temperature of 2 K, are presented in figures 1 and 2 as a

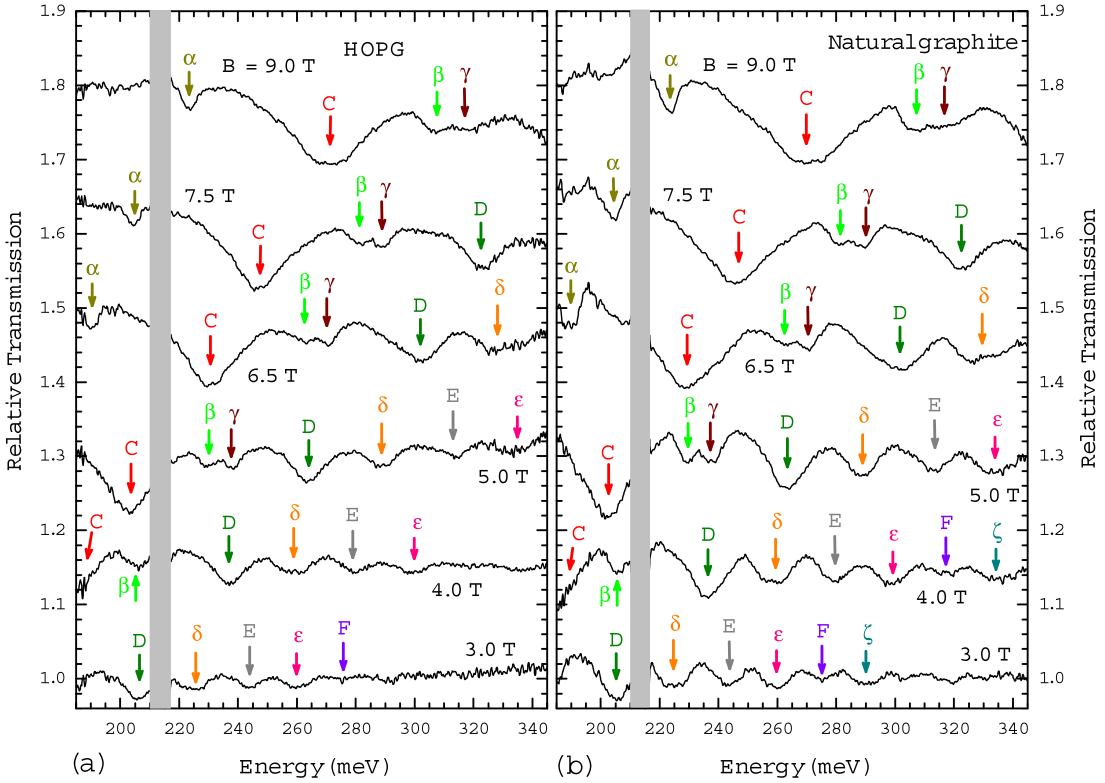

**Figure 2.** Comparison of magneto-transmission spectra taken in the spectral range 180–350 meV on HOPG and NG in parts (a) and (b), respectively. For clarity, the successive spectra in both parts were shifted vertically by an amount of 0.15.

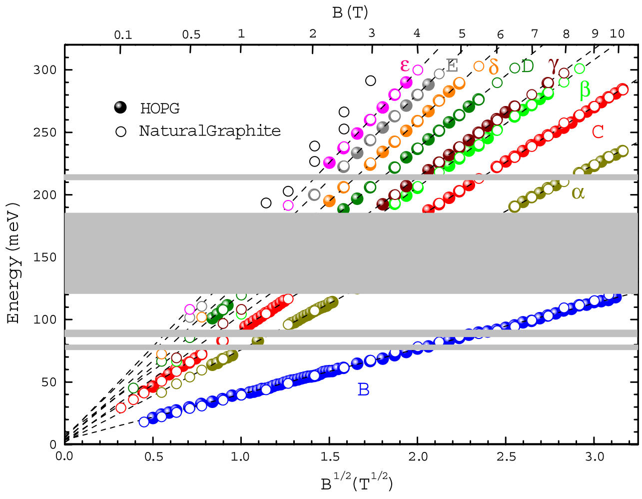

**Figure 3.** Positions of inter-LL transitions as a function of $\sqrt{B}$ for natural graphite (open circles) and HOPG (solid circles). The data for HOPG are taken from [12].

function of the applied magnetic field. The spectra of HOPG, presented in part (a) of both figures, are taken from [12]. Individual absorption lines are denoted by Roman and Greek letters, following the notation used in [12], and their $\sqrt{B}$ dependence is demonstrated by the fan chart in figure 3. Therefore, these lines are related to the $H$ point of graphite, where the energy spectrum in the presence of the magnetic field can be, with reasonable accuracy, described by the expression

$$E_n = \operatorname{sgn}(n)\tilde{c}\sqrt{2e\hbar B|n|} = \operatorname{sgn}(n)E_1\sqrt{|n|}$$
$$n = 0, \pm 1 \pm 2\ldots, \qquad (1)$$

typical of LLs in graphene. The validity of this formula is limited to the case of the vanishing pseudogap $\Delta \to 0$ at the $H$ point, which is consistent with no splitting of the B line [12] observed in our spectra. Assuming the LLs in the form of (1), the absorption lines B, C, D, E and F can be assigned to the inter-LL transitions $L_{-m} \to L_{m+1}$ (or $L_{-(m+1)} \to L_m$) with $m = 0, 1, 2, 3$ and 4, respectively. Similarly, $\alpha, \gamma, \delta, \varepsilon$ and $\zeta$ lines can be interpreted as transitions symmetric around the Dirac point $L_{-m} \to L_m$ with $m = 1, 2, 3$ and 4, respectively. Finally, the $\beta$ line corresponds to transitions $L_{-1(-3)} \to L_{3(1)}$. Note that, whereas the transitions denoted by Roman letters have a direct counterpart in absorption lines observed in both epitaxial and exfoliated graphene [21, 22], the lines denoted by Greek letters do not obey the graphene-like dipole selection rule $|n| \to |n| \pm 1$, but are dipole-allowed when the LL structure around the $H$ point of bulk graphite is treated properly within the SWM model [12, 23].

Comparing figures 1 and 2, no significant differences are found in spectra taken on both types of bulk graphite investigated here. In both materials, the same set of absorption lines is observed, characterized by the same positions in the spectra, giving a Fermi velocity of $\tilde{c} = (1.02 \pm 0.02) \times 10^6$ m s$^{-1}$. The spectra also exhibit very similar lineshapes. In NG as well as HOPG, the B line shows no signs of splitting and thus, based on its width, we can estimate the pseudogap to be $|\Delta| < 10$ meV, in agreement with the value of $\approx$8 meV found by Toy *et al* [6]. This estimation is also supported by recent theoretical calculations performed by Grüneis *et al* [24], estimating the pseudogap $|\Delta| \sim 5$ meV in the TB-GW approximation.

Data obtained on NG exhibit more pronounced spectral features, which likely indicate a higher quality of the NG crystal in comparison to polycrystalline HOPG. The NG sample allows us to resolve transitions with higher LL indices at very low magnetic fields, cf spectra at $B = 0.3$ in figure 1. Similarly, the $\zeta$ line is clearly resolved in the transmission spectra of NG and not HOPG at higher magnetic fields. This finding is in agreement with our magneto-transport experiments performed on the same samples, where Shubnikov–de Haas oscillations are more pronounced and start at lower magnetic fields for NG than in the case of HOPG [25].

## 4. Conclusion

To conclude, the magneto-transmission spectroscopy has been used to probe the nature of Dirac fermions in highly oriented pyrolytic and natural graphite. Both types of bulk graphite exhibited a very similar, if not identical, optical response, giving the Fermi velocity $\tilde{c} = (1.02 \pm 0.02) \times 10^6$ m s$^{-1}$ and the pseudogap $|\Delta|$ below 10 meV.

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