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Cyclotron resonance study of the electron and hole velocity in graphene monolayers

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We report studies of cyclotron resonance in monolayer graphene. Cyclotron resonance is detected using the photoconductive response of the sample for several different Landau level occupancies. The experiments measure an electron velocity at the K (Dirac) point of $c_K^*=1.093\times10^6$ m s^{-1}, which is substantially larger than in thicker graphitic systems. In addition we observe a significant asymmetry between the electron and hole bands, leading to a difference in the electron and hole velocities of 5% by energies of 125 meV away from the Dirac point.

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The observation of two-dimensional electronic systems in monolayer graphene,[1] where the electrons behave as Dirac fermions and show a variety of novel quantum Hall effects,[2–4] has led to an explosion of interest in this system. As well as new basic science, the exceptionally high electron velocities also mean that graphene has considerable potential for applications in high-speed electronics.[5] The basis for this behavior is the nearly linear dispersion of the energy bands close to the K point, where the dispersion relations cross with the form $E=\pm c^*\hbar k$, where c^* is the electron velocity. This has been predicted for over 50 years,[6] but has been measured only recently for bulk graphite[7] and ultrathin graphite layers,[8] while the first direct absorption measurements for monolayer graphene have just been reported.[9] We describe here a photoconductance study of cyclotron resonance in a monolayer of graphene in which the application of a magnetic field leads to the formation of Landau levels given by[10]

$$E_N = \mathrm{sgn}(N)\times c^*\sqrt{2e\hbar B|N|}, \qquad (1)$$

where $|N|$ is the Landau quantum index and B is the magnetic field. This allows us to make a precise measurement of the electron velocity and to examine deviations from exact linear behavior, which show that the electron- and holelike parts of the band structure have significantly different masses and that the velocity is significantly larger than for thicker graphitic material.

The experiment studies the photoconductive response from a multiply contacted single-monolayer sample of graphene, which was prepared using the techniques that have been described earlier.[1,2] The graphene films were deposited by micromechanical cleavage of graphite with multiterminal devices produced by conventional microfabrication, with a typical sample displayed in Fig. 1(a). Shubnikov–de Haas oscillations were first studied at 1.5 K to establish the carrier densities as a function of gate voltage and to ensure that the film studied was a single monolayer of graphene, since bilayers and thicker films are known to have a completely different dispersion relation.[11–13]

Cyclotron resonance was measured by detecting the modulation of the conductivity of the samples produced by chopped infrared radiation from a CO_2 laser operating between 9.2 and 10.8 μm. The sample was illuminated normally with unpolarized light parallel to the magnetic field in the Faraday geometry. Typical power densities were $\sim3\times10^4$ W m^{-2}, corresponding to a total power incident on the

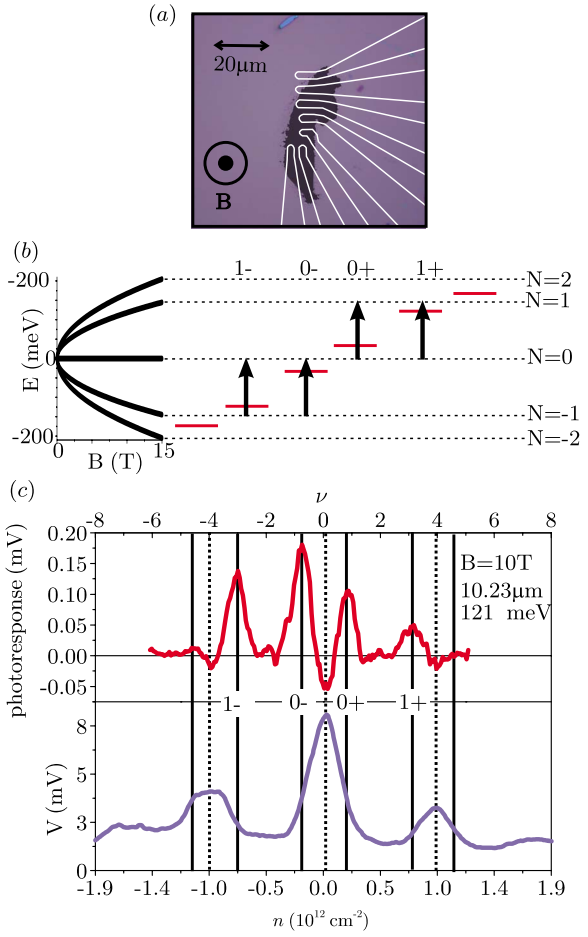

FIG. 1. (Color online) (a) Sample image with outline of the contacts used in the measurements. (b) Plot of the Landau energies as a function of magnetic field for Landau index $N=-2,\ldots,2$. Arrows indicate the resonant transitions probed in the energy range of the CO_2 laser. (c) Density dependence of the two-contact resistive voltage and photoconductive response of a typical graphene sample for infrared radiation of 10.23 μm at 10 T measured with a current of 100 nA.

samples studied of order 5 μW. The majority of experiments were performed in two-contact mode with a current of $I=100$ nA, since this gave the best signal-to-noise ratio, although similar spectra were also observed in a four-contact configuration. Figure 1(c) shows the photoconductive signal and the two-contact resistance of a graphene layer as a function of carrier density n, with the sample immersed in liquid helium at 1.5 K. This demonstrates that large positive photoconductive signals are observed at the edges of the conductance peaks, at the points where the resistivity is changing most rapidly with temperature and chemical potential. The response is proportional to the energy absorbed and thus provides an accurate relative measurement of the absorption coefficient. At resonance we observe voltage modulations as high as 3%. The peak response is detected when the Landau level occupancy $\nu=nh/eB$ is -3.0 $(1-)$, -0.76 $(0-)$, 0.88 $(0+)$ and 3.1 $(1+)$, where 0 corresponds to the Dirac point. A small negative response is also observed when the Landau levels are exactly half filled at occupancies of $\nu=-4,0,+4$. The two response peaks labeled $(1-)$ and $(1+)$ correspond to hole- and electronlike transitions from the Dirac point $(N=0)$ to the $N=\pm 1$ Landau levels, respectively. The $(0-)$ and $(0+)$ peaks both correspond to mixtures of the two transitions as the $N=0$ level is partially filled with either holes or electrons, but with either the hole or electron transition, respectively, predominant as indicated in Fig. 1(b). When $|\nu|>4$ no resonant absorption can occur in this field range, and we observe only some much weaker additional features caused by nonresonant bolometric response from the sample. This is greater at higher magnetic fields where localization of the carriers is increased.

In order to detect the resonances, we measure carrier density sweeps at each value of magnetic field, and compile a full map of the photoresponse as shown in Fig. 2 for a wavelength of 9.25 μm. This demonstrates that clear resonances can be detected for all four occupancies where strong photoresponse is seen. The immediate conclusion from this plot is that the resonances all occur in the region of 10 T, but that there is a significant asymmetry between the electron- and holelike transitions. A further negative photoresponse is observed at low magnetic fields (<2 T), which we attribute to interband photon absorption processes such as $-(N+1)\rightarrow N$ and $-N\rightarrow(N+1)$. In order to demonstrate the high-field resonances more clearly and to investigate the magnetic field dependence of the transition energies, we show traces in which the Landau level occupancy is held constant, by the simultaneous scanning of the gate voltage and magnetic field in order to follow the constant occupancy lines as shown in Fig. 2.

Sequences of resonances for the electronlike and holelike transitions are shown in Fig. 3. The resonances are plotted as a function of $\sqrt{B}$ and fitted with conventional Lorenzian line shapes with the addition of a linear correction to account for the increasing bolometric response at high fields. Some resonances show significant anisotropy, and we therefore quote an error for individual points of ±20% of the half width at half maximum. A typical fit is shown for each of the four resonances. The 0− resonances are particularly broad and therefore give higher errors. The resonance positions are

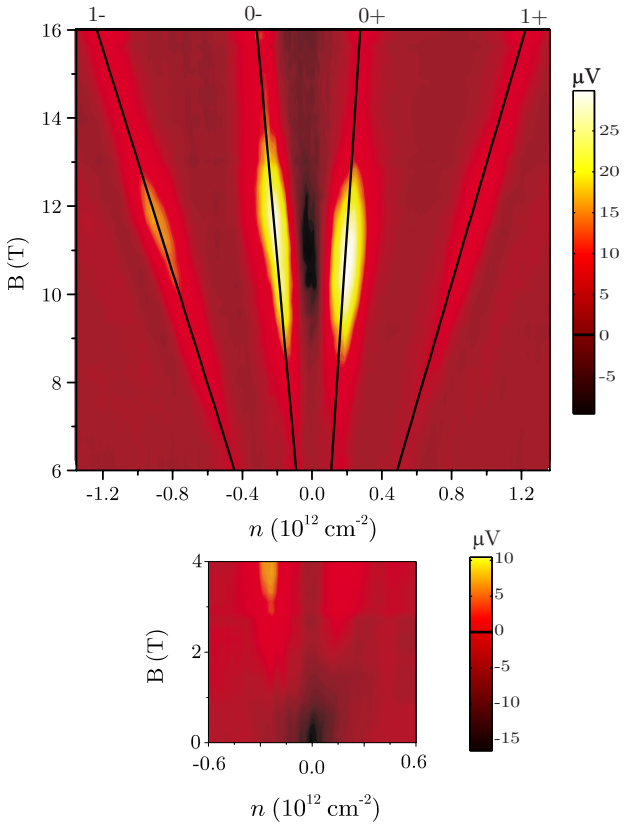

FIG. 2. (Color online) Photoconductive response as a function of gate voltage and magnetic field for 9.25 μm (134 meV). The low-field section of the map has an enhanced sensitivity to display the sharp negative resonance at zero field.

plotted as a function of $\sqrt{B}$ in Fig. 4. The resonance energies are expected to be given by Eq. (1), with a single value of the electron velocity c^*. Our results show clearly that this is not the case. Fitting velocities to each of the resonances separately gives values of $c^*=(1.117,1.118,1.105,$ and $1.069\pm0.004)\times10^6$ m s^{-1} for the 1+, 0+, 0−, and 1− resonances, respectively. The resonances measured for the 1− and 1+ occupancies show the lowest and highest values for c^*, as would be expected if the electron and hole masses are different, since these correspond to pure holelike and electronlike transitions, while the values for $(0-)$ and $(0+)$ are intermediate between the two extremes. Defining a single Fermi velocity averaged over the extremal values for electrons and holes in the region of the Dirac point gives $c^*=(1.093\pm0.004)\times10^6$ m s^{-1}. Interpreting the resonance positions in terms of the conventional cyclotron effective mass gives $m^*=0.009m_e$.

Values reported previously for the Fermi velocity suggest that it is quite strongly dependent on the number of graphene sheets in metallic systems. Angle-resolved photoemission on bulk graphite[7] gives 0.91×10^6 m s^{-1}, while the cylotron resonance measurements of Sadowski *et al.*[8] on thin (3–5) layers of epitaxial graphite give 1.03×10^6 m s^{-1}. A recent report on tunneling measurements in bilayer graphene[14] has found 1.07×10^6 m s^{-1}, while the results above and the cyclotron absorption by Jiang *et al.*[9] on monolayer graphene[3] give values of $\simeq1.1\times10^6$ m s^{-1}. By contrast, estimates

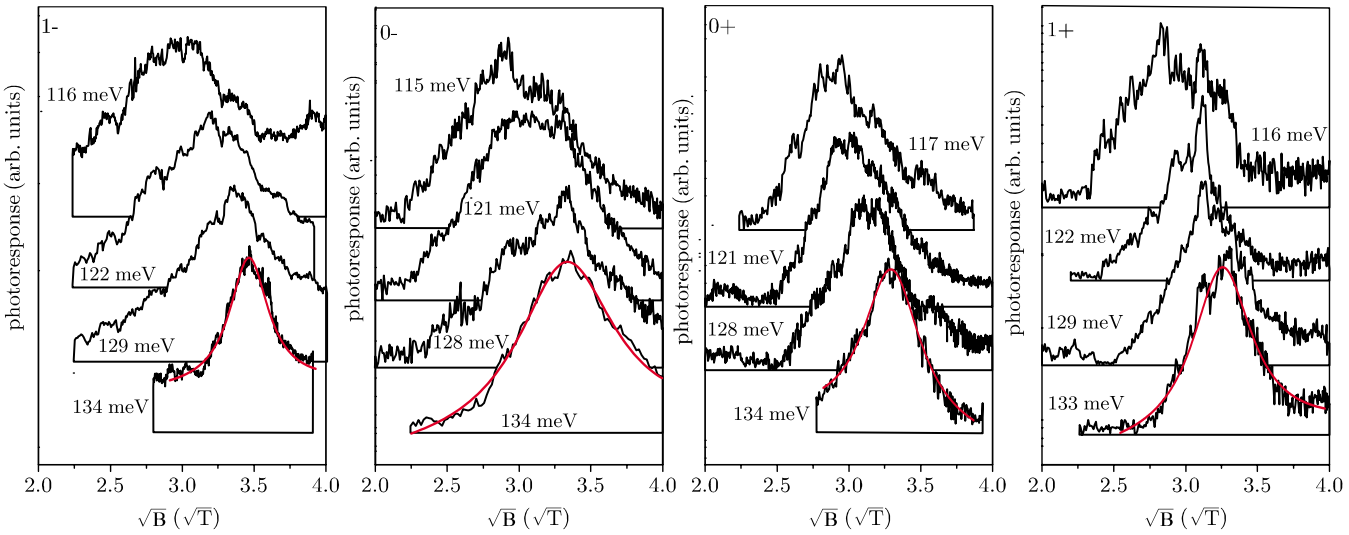

FIG. 3. (Color online) Photoconductive response as a function of $\sqrt{B}$ with the carrier densities scanned to keep the occupancies constant at $\nu=-3.0$ (1−), −0.76 (0−), 0.88 (0+), and 3.1 (1+) for wavelengths from 9.2 to 10.7 μm. The red (gray) lines show fits using Lorenzian line shapes combined with a linear background response.

based on the electronic properties of semiconducting carbon nanotubes deduce $c_K^*=0.94\times10^6$ m s^{-1}, corresponding to values of γ_0, the transfer integral, of order 2.9 eV.[15,16]

Theoretically, nearest-neighbor tight binding theory[17] predicts electron energies in terms of γ_0 and s_0, the nearest-neighbor overlap integral, of

$$E=\frac{\epsilon_{2p}\mp\gamma_0\sqrt{\omega(k)}}{1\mp s_0\sqrt{\omega(k)}}. \tag{2}$$

Setting $\epsilon_{2p}=0$ to give a correct description of the bands close to the K point, and with $\sqrt{\omega(k)}=\left(\frac{\sqrt{3}}{2}\right)ka_0$, where $a_0=0.246$ nm is the graphene lattice parameter, gives the electron velocity as

$$c_\pm^*=c_K^*\frac{1}{1\mp s_0E/\gamma_0}, \tag{3}$$

where $c_K^*=\frac{\sqrt{3}\gamma_0a_0}{2\hbar}$. Typical values for the parameters of $\gamma_0=3.03$ eV and $s_0=0.129$, which have been derived from first-principles calculations[18] and found to give good agreement with experiment,[17] give values for $c_K^*=0.98\times10^6$ m s^{-1} but predict only a very small asymmetry of the velocity of ±0.5%. More complex calculations such as those including up to third-nearest neighbors[19] give values that lead to even lower values of γ_0 (2.7 eV) and hence c^*. This suggests, therefore, that the currently accepted values of the transfer integral are consistent with the graphite results, but there is a progressive increase in the electron velocity as the graphite is thinned down to the single-monolayer graphene result. The changes in the transfer integral are probably related to the screening or changes in the details of the π bonds perpendicular to the graphene surface, which are also responsible for the band structure at the K point. These bonds are directly linked to the interlayer coupling of the graphene sheets and to their coupling to the SiO_2 insulator, suggesting that this coupling leads to an enhancement of the electron velocity, as has been suggested recently for carbon nanotubes,[20] where filling of the nanotubes with crystalline material leads to changes in the transfer integral. Using a value of $c_K^*=1.093\times10^6$ m s^{-1} leads to the deduction of a value of $\gamma_0=3.38$ eV.

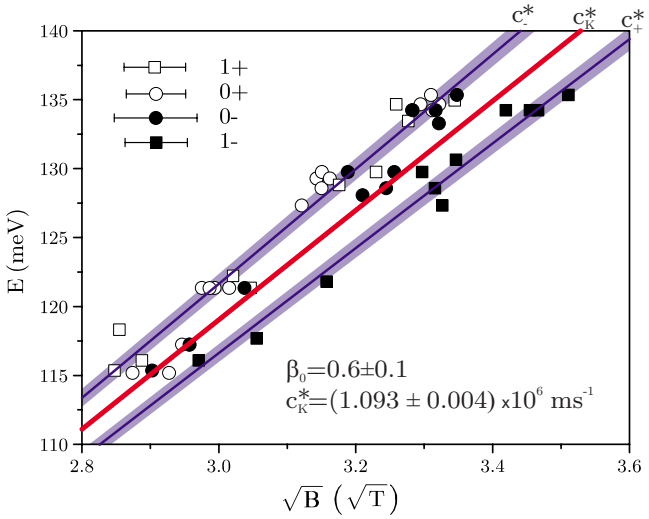

FIG. 4. (Color online) Resonance positions for the four resonances as a function of $\sqrt{B}$, together with a single fitted value of the electron velocity c_K^* [red (gray) line]. The outer lines show fits to Eq. (3), with the shaded bands covering the error limits from c_K and β_0. The individual resonance positions have errors as shown of ~±2%, corresponding to $0.2\Delta\sqrt{B}$, where $\Delta\sqrt{B}$ is the half width at half maximum absorption.

The second conclusion from Fig. 4 is that the asymmetry between electron and hole is considerably larger than that predicted by the simple tight binding theory. We model this by replacing the overlap integral s_0 with an empirical factor β_0 in Eq. (3) and refitting the data shown in Fig. 4 with the modified equation

$$c_{\pm}^{*} = c_{K}^{*} \frac{1}{1 \mp \beta_0 E/\gamma_0}. \quad (4)$$

The best fits to the data are shown in Fig. 4 with values of $c_K^*=(1.093\pm0.004)\times10^6$ m s^{-1} and $\beta_0=0.6\pm0.1$. These values give velocities for the electrons and holes of $c_{\pm}^{*}$ 1.118×10^6 and 1.069×10^6 m s^{-1} in the energy range close to ±125 meV. We therefore have clear evidence for the breaking of particle-antiparticle symmetry in the graphene system at the level of $\pm2.5\%$, approximately five times larger than expected for simple tight binding theory.[17] This may be linked to the intrinsic single-particle band structure, with some indications of this in the comparison of *ab initio* and tight binding dispersions,[19] although these calculations suggest values of c_K^* as low as 0.87×10^6 m s^{-1}. By contrast, the magnitude of the asymmetry is comparable, but of the opposite sign to that predicted ($\sim\mp3\%$) using random phase approximation methods, which take account of dynamical screening,[21] and which also predict an overall $\sim13\%$ enhancement of the velocity. It is also possible that the gating process itself will lead to some changes in the π bonding, due to the changes in surface field, and that this is linked to the velocity enhancement in thinner layers.

In addition to conventional single-particle effects, it may also be possible that many-body corrections could influence the value and asymmetry of the electron velocity. Kohn's theorem[22] has long been known to exclude the influence of electron-electron interactions on long-wavelength excitations for conventional parabolic systems. Calculations for graphene[23] suggest, however, that, although there are several similarities with the normal electron case, the linear dispersion may lead to finite Coulomb contributions to the cyclotron resonance transition energies, and that these will be strongly dependent on the level occupancy, although these are based on perfect particle-hole symmetry.

The resonance linewidths (half width at half maximum) deduced from fitting the data in Fig. 3 are all in the region of $(0.27-0.37)\sqrt{T}$ (1.5–2.5 T). Using our measured value of c_K^* gives an energy broadening $\hbar/\tau\simeq12$ meV, corresponding to a simple momentum relaxation time of $\sim5.5\times10^{-14}$ s, a mean free path $\lambda=c^*\tau\sim0.06$ μm, and a mobility $\mu\sim1.1$ m^2 V s^{-1}. The linewidths are significantly smaller than those observed by Jiang *et al.*[9], which may explain why these authors did not observe the electron-hole asymmetry.

In conclusion, therefore, we have measured cyclotron resonance in a monolayer graphene system, which demonstrates that the electron velocity is significantly enhanced relative to the value expected from previous calculations and measurements for thicker graphitic systems. In addition, we have demonstrated a considerable asymmetry in the carrier velocity for the electron- and holelike parts of the dispersion relation close to the K point of the Brillouin zone. These measurements suggest that there are still considerable uncertainties in understanding the band structure of monolayer graphene, which may lead to significant changes in any theories[24] based on perfect particle-antiparticle symmetry.

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