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 photoco nductive response of the sample for several different Landau level occupancies. The experiments measure an electron velocity at the K- (Dirac) point of \( c_K = 1.093 \times 10^6 \text{ ms}^{-1} \) 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, where the electrons behave as Dirac Fermions and show a variety of novel quantum Hall effects, has led to an explosion of interest in this system. As well as new basic science, the exceptionally high velocities also mean that graphene has considerable potential for applications in high speed electronic systems. The basis for this beh aviour is the nearly linear dispersion of the energy bands close to the K point, where the dispersion relations cross with the form \( E = \pm c^* h k \), where \( c^* \) is the electron velocity. This has been predicted for over 50 years, but has only been measured recently for bulk graphite and ultrathin graphite layers, while the first direct absorption measurements for monolayer graphene have just been reported. We describe here a photocon ductance 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

\[
E_N = \text{sgn}(N) \times c^* \sqrt{2e h B |N|},
\]

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 behaviour which show that the electron and hole-like 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 by using the techniques which have been described earlier. The graphene films were deposited by micromechanical cleavage of graphite with multi-terminal devices produced by conventional microfabrication, with a typical sample displayed in figure 1 (a). Shubnikov-de Haas oscillations were first studied at 1.5K 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.

Cyclotron resonance was measured by detecting the modulation of the conductivity of the samples produced by chopped infrared radiation from a CO2 laser operating between 9.2 and 10.8 \( \mu \text{m} \). The sample was illuminated normally with unpolarized light parallel to the magnetic field in the Faraday geometry. Typical power densities were \(~3 \times 10^4 \text{ W m}^{-2}\) corresponding to a total power incident on the samples studied of order 5 \( \mu \text{W} \). The majority of experiments were performed in two-contact mode with a current of I=100mA 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 2 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 labelled (1-) and (1+) correspond to hole and electron-like 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 transition respectively predominant as indicated in figure 1(b). When \(|\nu| > 4 \) no resonant absorption can occur in this field range and we only observe some much weaker additional features caused by non resonant bolometric response from the sample. This is greater at higher magnetic fields where localisation 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 Figure 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 10T, but that there is a significant asymmetry between the electron and hole-like transitions. A further negative photoresponse is observed at low magnetic fields (< 2 T) which we attribute to inter-band photon absorption processes such as −(N + 1) → N and −N → (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 electron-like and hole-like transitions are shown in Fig. 3. The resonances are plotted as a function of \( \sqrt{B} \) and fitted with conventional Lorenzian lineshapes 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 plotted as a function of \( \sqrt{B} \) in Fig. 4. The resonance energies are expected to be given by equation 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 \( c^* = (1.117, 1.118, 1.105 \text{ and } 1.069 \pm 0.004) \times 10^6 \text{ ms}^{-1} \) 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 hole-like and electron-like 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 \( c^* = 1.093 \pm 0.004 \times 10^6 \text{ ms}^{-1} \).

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 gives \( 0.91 \times 10^6 \text{ ms}^{-1} \), while the cyclotron resonance measurements of Sadowski et al. on thin (3-5) layers of epitaxial graphite give \( 1.03 \times 10^6 \text{ ms}^{-1} \). A recent report on tunnelling measurements in bilayer graphene has found \( 1.07 \times 10^6 \text{ ms}^{-1} \) while the
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 $\mu$m. The red lines show fits using Lorenzian lineshapes combined with a linear background response.

results above and the cyclotron absorption by Jiang et al on monolayer graphene give values of $\approx 1.1 \times 10^6$ ms$^{-1}$. By contrast estimates based on the electronic properties of semiconducting carbon nanotubes deduce $c^*_K = 0.94 \times 10^6$ ms$^{-1}$ corresponding to values of $\gamma_0$, the transfer integral, of order 2.9 eV$^{15,16}$. Theoretically nearest neighbour tight binding theory predicts electron energies in terms of $\gamma_0$ and $s_0$, the nearest neighbour overlap integral, of

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

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

$$c^*_\pm = c^*_K \frac{1}{1 \pm \frac{s_0 \Delta \sqrt{B}}{\gamma_0}},$$  \(3\)

where $c^*_K = \frac{\sqrt{3}}{2a_0} \gamma_0$. Typical values for the parameters of $\gamma_0 = 3.03$ eV and $s_0 = 0.129$ which have been derived from first principles calculations give values for $c^*_K = 0.98 \times 10^6$ ms$^{-1}$ but predict only a very small asymmetry of the velocity of $\pm 0.5\%$. More complex calculations such as those including up to third nearest neighbours conclude values which lead to even lower values of $\gamma_0 (2.7$ 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 $\pi$ 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 inter layer 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 $c^*_K = 1.093 \times 10^6$ ms$^{-1}$ leads to the deduction of a value of $\gamma_0 = 3.38$ eV.

The second conclusion from Figure 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 $\beta_0$ in equation $3$ and re-fitting the data shown in Figure 4 with the modified equation

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 line). The outer lines show fits to equation $3$ with the shaded bands covering the error limits from $c^*_K$ and $\beta_0$. The individual resonance positions have errors as shown of $\sim \pm 2\%$, corresponding to $0.2 \Delta \sqrt{B}$, where $\Delta \sqrt{B}$ is the half width at half maximum absorption.
The best fits to the data are shown in Fig. 3 with values of $c_K^* = (1.093 \pm 0.004 \times 10^6 \text{ ms}^{-1}$ and $\beta_0 = 0.6 \pm 0.1$. These values give velocities for the electrons and holes of $c_\pm^* = 1.118$ and $1.069 \times 10^6 \text{ ms}^{-1}$ in the energy range close to $\pm 125 \text{ meV}$. We therefore have clear evidence for the breaking of particle-antiparticle symmetry in the graphene system at the level of $\pm 2.5\%$, approximately five times larger than expected for simple tight binding theory. 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. In addition to conventional single particle effects it may also be possible that many-body corrections could influence the value of the asymmetry in the electron band structure, with some indications of this in the comparison of ab initio and tight binding dispersions. Kohn’s theorem has long been known to exclude the influence of electron-electron interactions on long wavelength excitations for conventional parabolic systems. Calculations for graphene 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 Figure 3 are all in the region of $0.27 - 0.37 \sqrt{T}$ (1.5-2.5 Tesla). Using our measured value of $c_K^*$ gives an energy broadening $\hbar/\tau \simeq 12 \text{ meV}$, corresponding to a simple momentum relaxation time of $\sim 5.5 \times 10^{-14}\text{s}$ and a mean free path $\lambda = c^*\tau \sim 0.06\mu\text{m}$ and a mobility $\mu \sim 1.1m^2\text{Vs}^{-1}$. The linewidths are significantly smaller than those observed by Jiang et al., 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 hole like 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 based on perfect particle-antiparticle symmetry.

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