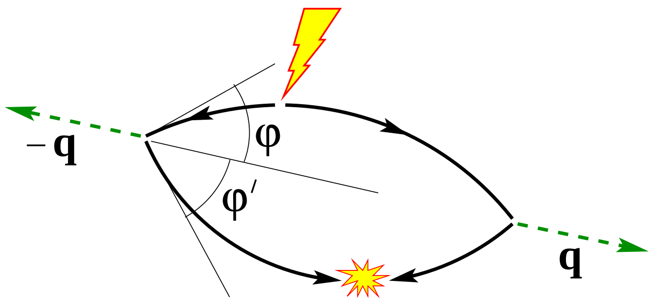

FIG. 3: (Color online) Schematic of the electron and hole motion during the Raman scattering process. The lightening represents the incident photon which creates the electron-hole pair. The solid arcs denote the propagation of the electron and the hole in the magnetic field. The flash represents the radiative recombination of the electron-hole pair. The dashed arrows denote the emitted phonons.

the D peak in the vicinity of an edge[31] (details of this calculation are presented in the Appendix). An essential ingredient of the calculation is the semiclassical electronic Green's function for graphene in a magnetic field which was calculated in Ref. 32. Under the assumption $\omega_c \ll (\gamma, \omega_{\rm ph}) \ll \omega_{\rm in}$, the result of the calculation can be represented as:

$$\mathcal{M}(q) \propto \int\limits_0^\infty dz\,\sqrt{z}\,e^{-[i(q-2p)+2\gamma/v]z - i[p/(12R^2)]z^3} = \\ = 2(i\pi)^{3/2}\frac{R}{\sqrt{p}}\frac{d}{du}\left[\mathrm{Ai}^2(u) - i\,\mathrm{Ai}(u)\,\mathrm{Bi}(u)\right], \quad (1)$$

$$u \to \left(q - 2p - \frac{2i\gamma}{v}\right)\left(\frac{R^2}{p}\right)^{1/3}. \quad (2)$$

For vanishing magnetic fields, the cyclotron radius $R \to \infty$ and the relation $\mathcal{M}(q) \propto (q - 2p - 2i\gamma/v)^{3/2}$, which is Eq. (64) of Ref. 29, is recovered. For finite values of the magnetic field, the matrix element is expressed in terms of Airy functions $\mathrm{Ai}(u)$ and $\mathrm{Bi}(u)$[33]. Note also the similarity with the expression for the polarization operator in Ref. 30.

Since the density of the final phonon states is practically q-independent[34], $|\mathcal{M}(\omega_{\rm in}/v + \Omega/(2v_{\rm ph}))|^2$ describes the Raman scattering intensity as a function of Ω, the Raman shift measured with respect to the center of the $2D$ peak at zero field. To illustrate the change of the peak shape in magnetic fields, we present in Fig. 4(a) $|\mathcal{M}(\omega_{\rm in}/v + \Omega/2v_{\rm ph})|^2$ for $B = 0, 10, 30$ T, considering $\hbar\omega_{\rm in} = 1.7$ eV, $\hbar v = 7$ eV $\cdot$ Å ($v = 1.06 \times 10^8$ cm/s), $v_{\rm ph}/v = 50$ cm^{-1}/eV, $\hbar\gamma = 27$ meV. This value for γ is chosen in order to reproduce the observed shift of the peak maximum for increasing magnetic fields. However, the resulting FWHM for the $2D$ band is about twice smaller than the one observed experimentally at zero field. Most likely, this indicates the presence of an additional broadening mechanism besides the electronic scattering. As we cannot determine the nature of this mechanism, we model it by introducing phenomenologically an additional broadening of the $2D$ band through a convolution of $|\mathcal{M}|^2$ with a Gaussian curve of width σ:

$$I_{2D}(\Omega) \propto \int\limits_{-\infty}^{\infty} \frac{d\Omega'}{\sqrt{2\pi}\sigma}\,e^{-\frac{(\Omega'-\Omega)^2}{2\sigma^2}}\left|\mathcal{M}\left(\frac{\omega_{\rm in}}{v} + \frac{\Omega'}{2v_{\rm ph}}\right)\right|^2. \quad (3)$$

There is no simple relation between the FWHM of the original $|\mathcal{M}|^2$ peak ($8\gamma(v_{ph}/v)\sqrt{2^{2/3}-1}$ at zero field), the FWHM of the broadening Gaussian ($2\sigma\sqrt{\ln 4}$), and the FWHM of the resulting peak. The result of the convolution for $\sigma = 9.5\,\mathrm{cm}^{-1}$ is shown in Fig. 4(b).

In the following, we assume γ and σ to be independent of the magnetic field. This approximation is reasonable as long as the magnetic field is non-quantizing.[35] At $B = 30$ T, electrons with energy $\epsilon = 0.85$ eV measured from the Dirac point, have a cyclotron frequency $\omega_c = (v^2\hbar/\epsilon)(eB/\hbar c)$ of about 27 meV, so the strongest magnetic fields in this experiment seem to be close to the limits of validity of this approximation. Beyond this regime, Landau quantization of the electronic spectrum should manifest itself as oscillations of the peak intensity due to periodic modifications of the resonance conditions with increasing magnetic field, while in the experiment no such oscillations are observed.

The results of this calculation in terms of Raman shift and of FWHM of the peak, as described by Eq. (3), are presented as red solid lines in Fig. 2 (a)–(d) for the two components of the observed 2D band feature. The following parameters were adjusted: (i) the central frequencies of the two components at zero field, giving the overall vertical offset for the curves in Fig. 2 (b), (d); (ii) $\sigma = 9.5\,\mathrm{cm}^{-1}$ and $\sigma = 11.9\,\mathrm{cm}^{-1}$ were taken for the low-frequency and high-frequency components, respectively, in order to reproduce the FWHM at zero field, in combination with (iii) $\hbar\gamma = 27$ meV which determines *all four* slopes in Fig. 2 (a)–(d). Clearly, without precise knowledge of the origin of the two $2D$ band components we cannot account for the slight difference in their zero-field widths. Nevertheless, the fact that the calculation reproduces the red shift of the $2D$ band energy as a function of the magnetic field which is quadratic for fields up to 5–10 T and linear at higher fields, with a single value of γ for both components, shows that the electrons have the same dynamics in the parts of the sample, responsible for the two components.

Besides, the deduced value of $\hbar\gamma = 27$ meV is in good agreement with that deduced from the doping dependence of the $2D$ peak intensity in exfoliated graphene[36,37] (given the energy dependence of γ and the fact that the latter measurements were performed at higher excitation energy, the electron scattering in those samples is somewhat weaker than in ours). We also note that the found value $\hbar\gamma = 27$ meV is in reasonable agreement with the line width of electronic transitions in high magnetic fields measured in this range of energy on similar samples.[38] This fact is remarkable because those measurements were