

## Effects of electron-electron interactions on the electronic Raman scattering of graphite in high magnetic fields

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We report the observation of strongly temperature ( $T$ )-dependent spectral lines in electronic Raman-scattering spectra of graphite in a high magnetic field up to 45 T applied along the  $c$  axis. The magnetic field quantizes the in-plane motion, while the out-of-plane motion remains free, effectively reducing the system dimension from 3 to 1. Optically created electron-hole pairs interact with, or shake up, the one-dimensional Fermi sea in the lowest Landau subbands. Based on the Tomonaga-Luttinger liquid theory, we show that interaction effects modify the spectral line shape from  $(\omega - \Delta)^{-1/2}$  to  $(\omega - \Delta)^{2\alpha-1/2}$  at  $T = 0$ . At finite  $T$ , we predict a thermal broadening factor that increases linearly with  $T$ . Our model reproduces the observed  $T$ -dependent line shape, determining the electron-electron interaction parameter  $\alpha$  to be  $\sim 0.05$  at 40 T.

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Electron-electron ( $e$ - $e$ ) interactions become progressively more important as the system dimension is lowered. One-dimensional (1d) systems, in particular, provide model environments in which to explore interaction effects [1]. Interacting 1d electrons are expected to form an exotic electronic state of matter, the Tomonaga-Luttinger liquid (TLL) [2–5]. A strong magnetic field,  $B$ , can suppress the kinetic energy of electrons, thus enhancing the effect of interactions, as exemplified by the fractional quantum Hall effect [6–8]. For a 3d material, an applied magnetic field creates an effective 1d system along the field, ideal for a systematic study of interaction effects in a highly controllable fashion [9]. Particularly promising are 3d metals with small electron and/or hole pockets near the Fermi energy,  $E_F$ , such as bismuth [10–14] and graphite [12,15–18], where the magnetic quantum limit can be readily reached with  $B \sim 10$  T.

Here we use Raman spectroscopy to study electronic states and correlations in graphite in a strong  $B$  up to 45 T applied along the  $c$  axis. The  $B$  quantizes the electronic motion in the  $ab$  plane while the motion along the  $c$  axis remains free, thus reducing the effective dimension from 3 to 1. Instead of the main Raman features related to phonons [19,20], in this work we focus on a series of electronic Raman features assigned to electronic inter-Landau-level (LL) transitions [21], whose  $B$  dependence can be explained through the Slonczewski-Weiss-McClure (SWM) model [22–24]. Each feature exhibits strongly temperature ( $T$ )-dependent shape. Our calculations show that scattering by thermally excited acoustic phonons [25–28] is too weak to explain the observations. Electron-electron interactions, on the other hand, are shown to be the cause for the observed  $T$  dependence, through the “shake-up” process, known in the problem of x-ray (or Fermi-edge) singularities [5]. Namely,

optically created electron-hole ( $e$ - $h$ ) pairs interact with, or shake up, the 1d Fermi sea in the lowest Landau subbands, resulting in line-shape deviations from single-particle densities of states (i.e., 1d Van Hove singularities). Based on the TLL theory [1–5], we show that  $e$ - $e$  interactions modify the Van Hove singularity to the form  $(\omega - \Delta)^{2\alpha-1/2}$  at 0 K, where  $\omega$  is the photon frequency,  $\Delta$  is the band-edge frequency, and  $\alpha$  is a dimensionless measure of the influence of  $e$ - $e$  interactions. At finite  $T$ , we predict a thermal broadening factor,  $\Gamma(T) \propto T$ . Our model reproduces the observed  $T$ -dependent line shape, determining  $\alpha$  to be 0.016, 0.026, and 0.048, at 20, 30, and 40 T, respectively.

Raman-scattering measurements were performed on natural graphite (NGS Naturgraphit GmbH) in a back-scattering Faraday geometry in  $B$  up to 45 T, as described in Ref. [21]. The excitation beam from a 532-nm laser was focused to a spot size  $< 20 \mu\text{m}$  with a power of  $\sim 13$  mW. Most of the data were collected with a spectral resolution of  $\sim 3.4 \text{ cm}^{-1}$ . For high- $B$ , low- $T$  ( $\leq 10$  K) measurements of the sharpest peaks, a spectral resolution of  $\sim 1.9 \text{ cm}^{-1}$  was employed. The  $T$  drift over an integration time of up to 7 min, measured by a  $T$  sensor installed below the sample, was  $< 1$  K at  $T = 7$  K and  $< 2$  K at  $T \geq 180$  K.

Figure 1(a) shows Raman spectra taken at 10, 20, and 30 T at 7 K. The main band is the  $G$  peak at  $\sim 1580 \text{ cm}^{-1}$ , due to  $E_{2g}$  phonons [19,20]. In the presence of  $B$ , electronic Raman features appear, coming from inter-LL transitions, labeled (1,1), (2,2),  $\dots$ , etc., which we focus on in this work. Figure 1(b) shows a series of spectra taken at various  $B$  at 7 K, exhibiting electronic peaks that move with  $B$ . These peaks can be attributed to the “symmetric” inter-LL excitations in the vicinity of the  $K$  point [21,29]. The strongest, lowest-frequency transition among these is (1,1), which is from the  $n = -1$  level in the valence band to the  $n = 1$  level in the conduction band. Similarly, we observe the (2,2), (3,3), and (4,4) transitions; see also the zero-field in-plane dispersions and energy levels near the  $K$  point in the inset to Fig. 1(a). The symmetric inter-LL

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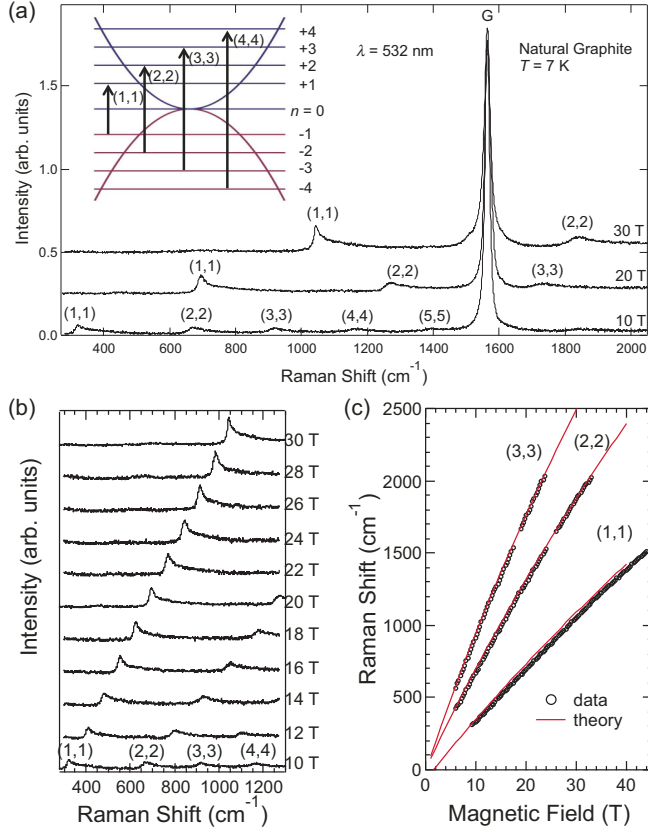


FIG. 1. (Color online) (a) Raman spectra at 10, 20, and 30 T. The feature at  $\sim 1580 \text{ cm}^{-1}$  is the  $G$  peak due to the  $E_{2g}$  phonons. Inset: schematic energy-level diagram showing the transitions responsible for the electronic peaks. (b) Data taken at various  $B$  at  $T = 7 \text{ K}$ , showing peaks due to (1,1) through (4,4) interband transitions. (c) Peak positions of the observed (1,1), (2,2), and (3,3) transitions as a function of  $B$ , together with calculations based on the SWM model.

excitations are nonresonant Raman processes and were theoretically investigated for single-layer graphene (SLG) [30] and bilayer graphene (BLG) [31]. The peak positions of the three lowest-energy transitions are plotted against  $B$  in Fig. 1(c); they agree well with our calculations [32] (solid and dashed lines) based on the SWM model [21].

These inter-LL transitions have strong  $T$  dependence, as shown in Fig. 2, where Raman spectra at various  $T$  are plotted for (a) 20, (b) 30, and (c) 40 T. At the lowest  $T$ , the peaks exhibit sharp and asymmetric line shapes, reminiscent of a 1d Van Hove singularity, as expected from the effective dimension reduction from 3 to 1 in a  $B$ . As  $T$  increases, there is significant peak broadening and blueshift. The blueshift is expected from the thermal expansion of the carbon-carbon bonds, which changes the tight-binding parameters [28]. On the other hand, the thermal broadening cannot be explained within the tight-binding model. To quantify it, we first fit the spectra within a single-particle model using the joint density of states for interband transitions, obtained from the SWM model, with  $T$ -dependent Lorentzian broadening [32]. Figure 2(d) plots the extracted Lorentzian FWHM  $\Gamma$  as a function of  $T$  for 20 and 30 T. Apart from a small finite linewidth at  $T = 0$ ,

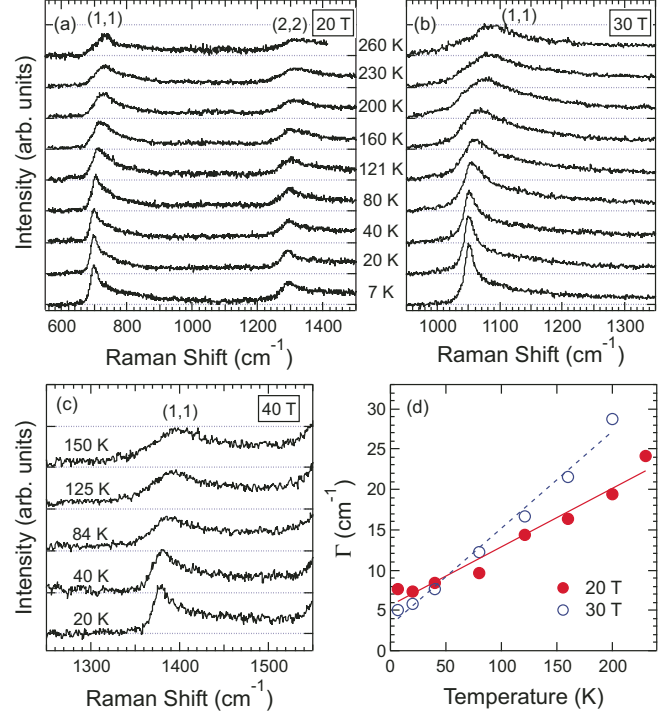


FIG. 2. (Color online) Temperature-dependent electronic Raman scattering of graphite at (a) 20 T, (b) 30 T, and (c) 40 T. (d) Temperature dependence of the broadening factor  $\Gamma$  of the (1,1) line at 20 T (solid circles) and 30 T (open circles). The lines are fits to the data.

$\Gamma_0 \approx 5 \text{ cm}^{-1}$ , possibly due to disorder,  $\Gamma$  linearly depends on  $T$ .

Within the single-particle picture,  $T$  only appears in the Fermi-Dirac distribution function, but this is a negligible effect since both the initial and final states of the Raman process are far away from  $E_F$ , which resides in the  $n = 0$  bands. For example, for the (1,1) transition at 30 T, the electron and hole bands are  $\sim 65 \text{ meV}$  (or  $\sim 750 \text{ K}$ ) away from  $E_F$ . Thus, we need to take into account the interactions of the photoexcited  $e$ - $h$  pairs with some low-energy modes that would significantly change when  $T$  changes from 4 to 300 K. Specifically, since the linear- $T$  broadening in Fig. 2(d) implies a Bose-Einstein distribution at an energy scale much smaller than  $k_B T$ , we only consider bosonic excitations whose characteristic energies are  $\ll 100 \text{ K}$ . Hence, we consider two types of low-energy modes: (i) particle-hole ( $p$ - $h$ ) excitations across  $E_F$  in the  $n = 0$  bands [Fig. 3(a)] and (ii) acoustic phonons. We find that interactions with (i) explain the observed  $T$ -linear broadening while interaction with (ii) is too weak to explain it.

The magnetoelectronic Raman scattering matrix was previously calculated for SLG [30] and BLG [31] and can be readily generalized to graphite in the presence of  $B$ :

$$\hat{R} = \Lambda \sum_{\vec{k}} \Psi_n^\dagger(k_y, k_z) \Psi_{-n}(k_y, k_z), \quad (1)$$

where  $\Lambda$  is the scattering amplitude,  $k_y$  ( $k_z$ ) are electron momenta in the  $ab$  plane (along the  $c$  axis),  $\Psi_n^\dagger$  creates an electron in the  $n = 1, 2, 3, 4, \dots$  bands, and  $\Psi_{-n}$  creates a

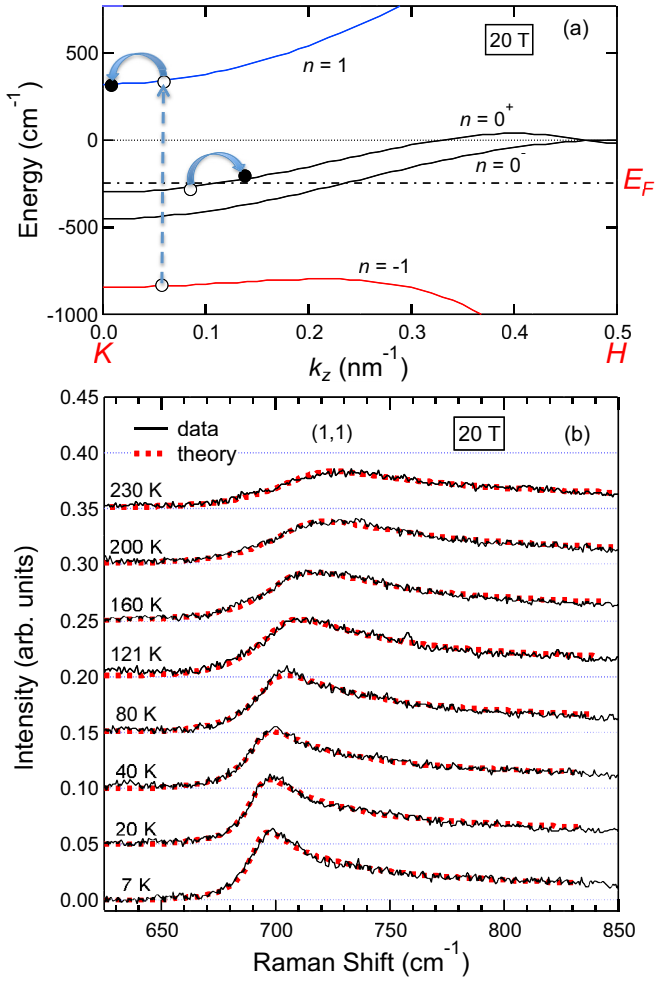


FIG. 3. (Color online) (a) The shake-up process. The (1,1) electron-hole pairs shake up the 1d Fermi sea in the lowest-energy Landau subbands, creating particle-hole pairs across  $E_F$ . (b) Temperature dependence of the line shape for the (1,1) transition at 20 T, together with fits (dashed lines) based on the model shown in (a).

hole in the  $n = -1, -2, -3, -4, \dots$  bands. Both electrons and holes are massive at the bottom of the bands at the  $K$  point, i.e.,  $m_n \neq 0$  for all  $n$ 's, similar to BLG, but there is  $e$ - $h$  asymmetry, i.e.,  $m_1 \neq m_{-1}$ .

Figure 3(a) depicts the basic ingredients involved in the  $e$ - $e$  interaction process we consider here, together with dispersions calculated via the SWM model for the  $n = 0^\pm$  and  $\pm 1$  bands at 20 T. The two lowest-energy bands ( $n = 0^\pm$ ) cross  $E_F$ , and the carriers near  $E_F$  have approximately linear dispersions. In the (1,1) process, an  $e$ - $h$  pair is created in the  $n = \pm 1$  bands, which interact with, and are thereby dressed with, multiple  $p$ - $h$  excitations in the  $n = 0^\pm$  bands near  $E_F$ . As  $T$  is raised, the thermal smearing of the Fermi edge leads to stronger interaction between the massive  $e$ - $h$  pair and the massless  $p$ - $h$  pairs, and the peak broadens. This type of shake-up process was theoretically studied in carbon nanotubes at 0 K [33,34]: a 1d Van Hove singularity,  $(\omega - \Delta)^{-1/2}$ , is predicted to become  $(\omega - \Delta)^{2\alpha-1/2}$  with  $\alpha \sim 0.1$  once the shake-up process is taken into account.

We describe the  $n = 0^-$  electrons as a TLL with the Hamiltonian [1–4] given by

$$H_0^c = v_F \int dz [\psi_R^\dagger i \partial_z \psi_R - \psi_L^\dagger i \partial_z \psi_L], \quad (2)$$

where  $v_F$  is the Fermi velocity and  $\psi_{R(L)}^\dagger$  creates a particle near the right (left) Fermi point. The  $n = 0^+$  band can be described by a similar Hamiltonian but with a different  $v_F$ . By approximating the energy dispersion near  $E_F$  as  $E \propto k_z$ , we can rewrite Eq. (2) via bosonization as

$$H_0^c = \frac{v_F}{2\pi} \int dz [(\nabla\phi)^2 + (\nabla\theta)^2], \quad (3)$$

where  $\nabla\phi = -2\pi[\rho_R + \rho_L]$ ,  $\nabla\theta = 2\pi[\rho_R - \rho_L]$ , and  $\rho_R$  ( $\rho_L$ ) is the density operator for right-moving (left-moving) electrons.

We assume that the photogenerated electrons ( $n = 1$ ) and holes ( $n = -1$ ) interact with the  $n = 0^-$  conduction electrons separately. For the  $n = 1$  band, where electrons are massive, we can treat the electrons through

$$H_1 = \int dz \Psi_1^\dagger \left[ -\frac{1}{2m} \partial_z^2 + \Delta_1 \right] \Psi_1, \quad (4)$$

where  $\Delta_1$  is the band-edge frequency and  $\Psi_1^\dagger$  ( $\Psi_1$ ) is the creation (annihilation) operator for the  $n = 1$  band. We also assume that the interaction Hamiltonian only involves the total charge density, thus neglecting any backscattering and umklapp scattering:

$$H_{\text{int}} = \frac{V}{2} \int dz \left[ \Psi_1^\dagger \Psi_1 - \frac{1}{2\pi} \nabla\phi \right]^2. \quad (5)$$

We write the effective Hamiltonian for the system as the sum of Eqs. (3)–(5):  $H = H_0^c + H_1 + H_{\text{int}}$ .

The diagonalization of the Hamiltonian is a unitary transformation  $U^\dagger H U$  and has been previously solved by many authors [33–36]:

$$U^\dagger = \exp \left[ -i \frac{\gamma^+}{\pi} \int dy \theta(y) \Psi_1^\dagger(y) \Psi_1(y) \right]. \quad (6)$$

Under this transformation, the original interacting system can be mapped to a noninteracting one, and the massive-electron operator acquires an additional string operator,  $\Psi_1(z) = \exp[-i\gamma^+\theta(z)/\pi] \tilde{\Psi}_1(z)$ , where  $\tilde{\Psi}_1^\dagger$  creates a free electron in the  $n = 1$  band. The massive  $n = 1$  electron then gets dressed by the additional string operator, i.e., the  $n = 0^-$  conduction electrons adiabatically adjust to the massive electrons. Similarly, we can obtain a dressed expression for the massive hole.

The spectral function can be obtained by calculating the imaginary part of the retarded Green's function [5],

$$G^R(z, t) \equiv -i\theta(t) \langle [\Psi_{-1}^\dagger(z, t) \Psi_1(z, t), \Psi_1^\dagger(0, 0) \Psi_{-1}(0, 0)] \rangle. \quad (7)$$

At zero  $T$ , Eq. (7) can be evaluated directly in real space. However, at finite  $T$ , one has to follow a different route. As the Green's function for the massive electron/hole and that for the conduction electrons are both straightforward to obtain,

the total Green's function can be written as a convolution of three Green's functions,

$$\begin{aligned} G^R(z,t) &\approx -i\theta(t)[-iG_{-1}^<(-z,-t)][iG_1^>(z,t)]F(z,t), \\ G^R(0,\omega) &= -i \int \prod \frac{dp_i}{2\pi} \frac{d\omega_i}{2\pi} G^0(p_2,\omega_2)F(p_1,\omega_1) \\ &\quad \times \delta(0-p_1-p_2)\delta(\omega-\omega_1-\omega_2), \\ G^0(p,\omega) &= \int \frac{dp_1}{2\pi} \int \frac{d\omega_1}{2\pi} G_1^>(p_1,\omega_1) \\ &\quad \times G_{-1}^<(p_1-p,\omega_1-\omega), \end{aligned} \quad (8)$$

where

$$F(z,t) = \langle \exp[-i\gamma\theta(x,t)] \exp[i\gamma\theta(0,0)] \rangle. \quad (9)$$

We can express the spectral function in a universal form as

$$A(\omega) = \Lambda T^{2\alpha-0.5} \tilde{F}\left(\frac{\omega/T}{4\pi}, \alpha\right), \quad (10)$$

where

$$\begin{aligned} \tilde{F}(z,t) &= \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} B[n+\alpha, 1-\alpha] B[m+\alpha, 1-\alpha] \\ &\quad \times \text{Re} \left[ \frac{(2i)^{2\alpha}}{\sqrt{z - \frac{i}{2}(m+n+\alpha)}} \right]. \end{aligned} \quad (11)$$

In Eq. (11) there are two dimensionless parameters:  $\omega/T$  and  $\alpha$ . The first parameter implies that the spectral width linearly depends on  $T$  for a fixed  $\alpha$ . The meaning of  $\alpha$  can be understood by studying the  $T=0$  asymptotic behavior of Eq. (11), and comparing it with the previous zero- $T$  results [33,34]. It then becomes clear that

$$A(\omega) \propto \frac{\Theta(\omega - \Delta)}{(\omega - \Delta)^{2\alpha-1/2}}, \quad (12)$$

where  $\Theta$  is the Heaviside function. For metallic carbon nanotubes,  $\alpha$  was estimated to be  $\sim 0.1$  [33,34]. To fit our experimental data with our model, we use the true band structure instead of a parabolic approximation, by fitting the tail up to  $\sim 0.2(\pi/c)$  from the  $K$  point. Figure 3(b) shows how well our model fits the data, determining  $\alpha(20 \text{ T}) \approx 0.016$ ,  $\alpha(30 \text{ T}) \approx 0.026$ , and  $\alpha(40 \text{ T}) \approx 0.048$ . These values are smaller than the value estimated for nanotubes, as expected, but there is a trend that  $\alpha$  increases with  $B$ , as this tends to make the system more 1d.

We now consider acoustic phonons, which can also couple to the massive electrons and holes. We use the approximation that in-plane and out-of-plane modes are separated. This approximation leads to a slight numerical modification of the following equations but greatly simplifies our understanding of electron-acoustic phonon interactions in graphite. The properties of acoustic phonons can be described by five elastic constants [37]:  $C_{11} = 1109 \text{ GPa}$ ,  $C_{66} = 485 \text{ GPa}$ ,  $C_{33} = 38.7 \text{ GPa}$ ,  $C_{13} = 0 \text{ GPa}$ , and  $C_{44} = 5 \text{ GPa}$ . Unlike the case of optical phonons [19,26,38,39], coupling with acoustic phonons vanishes at the  $\Gamma$  point since the electron-acoustic-phonon interaction Hamiltonian  $H_{\text{ep}} \sim \sqrt{q}$  [25,40,41], where  $q$  is the phonon wavenumber. We then evaluate the thermal

broadening of Raman peaks by calculating the imaginary part of the self-energy:

$$\begin{aligned} H_{\text{ep}} &= \sum_{\vec{k}, \vec{k}', \vec{q}} g_{\vec{q}} h(\vec{q}) \Psi_1^\dagger(k_y + q_y, k_z + q_z) \Psi_1(k_y, k_z) (b_{-\vec{q}}^\dagger + b_{\vec{q}}), \\ g_{\vec{q}} &= \frac{\eta \kappa q \sin \theta}{2} \sqrt{\frac{\hbar}{2NM\omega_{\vec{q}}}} \frac{\sqrt{2}}{2} \frac{\Delta_B^2}{\Gamma\gamma_1}, \\ h(\vec{q}) &= \left( 4 - 2l_B^2 q^2 \sin^2 \theta + \frac{l_B^4 q^4 \sin^4 \theta}{8} \right) e^{-(l_B^2 q^2 \sin^2 \theta)/4}, \end{aligned} \quad (13)$$

where  $l_B = (\hbar/eB)^{1/2}$  is the magnetic length,  $\eta \sim 2$ , and  $\kappa \sim 1/3$  [26]. To first order, we estimate the scattering rate through Fermi's "golden rule":

$$W_i = \frac{2\pi}{\hbar} \sum_f |\langle f | H_{\text{ep}} | i \rangle|^2 \delta(E_i - E_f). \quad (14)$$

When the momentum transfer during the scattering process is small (i.e.,  $vq \ll k_B T$ ), the phase space for phonon modes is  $q^2 (\frac{1}{e^{vq/kT}} + \frac{1}{2} \pm \frac{1}{2}) \sim qT \rightarrow 0$ , and when the momentum transfer is large, the overlap between initial and final states has a factor  $\exp(-q_{\perp}^2 l_B^2)$ . For  $B = 30 \text{ T}$ ,  $l_B \sim 5 \text{ nm}$ , which is at least one order larger than the carbon-carbon bond length. Thus, the contribution to scattering drops exponentially as the phonon modes move away from the  $\Gamma$  point (or, equivalently, with increasing energy). The calculated momentum-dependent scattering rate is then given by

$$\begin{aligned} W(k_z) &= \Lambda' \int_0^\pi d\theta \frac{\tilde{q}^2 \sin^3 \theta}{\sqrt{\sin^2 \theta + \frac{V_4^2}{V_1^2} \cos^2 \theta}} \frac{h^2(q, \theta)}{\cos^2 \theta} \\ &\quad \times \left( n_{\omega_{\vec{q}}} + \frac{1}{2} \pm \frac{1}{2} \right), \end{aligned} \quad (15)$$

where

$$\begin{aligned} \tilde{q} &= \frac{2ml_B V_1}{\hbar} \frac{\hbar k_z}{m V_1} \cos \theta \mp \sqrt{\sin^2 \theta + V_4^2 / V_1^2 \cos^2 \theta}, \\ \hbar \Lambda' &= \frac{\eta^2 \kappa^2 m}{16\pi M} \frac{V_{\text{unit}}}{l_B^3} \frac{\Delta_B^2}{\gamma_1^2} \frac{\sqrt{2}v}{V_1} \Delta_B \approx 4.1 \times 10^{-6} \text{ cm}^{-1}. \end{aligned}$$

This value leads to  $W(k_z) \sim 10^{-4} \text{ cm}^{-1}$  at 30 T and 200 K, too small to explain the observed broadening, which requires the scattering rate to be  $\sim 10 \text{ cm}^{-1}$ . There are two reasons for the small  $\hbar \Lambda'$ : one is  $m/M \sim 10^{-3}$ , and the other is  $V_{\text{unit}}/l_B^2$ . The latter, i.e., the magnetic length suppression, is a unique aspect of this work, made possible by a high  $B$ .

In summary, we studied electronic Raman scattering in graphite in a strong magnetic field up to 45 T, applied along the  $c$  axis. We observe a series of spectral lines, ascribed to inter-Landau-subband transitions, and each line exhibits strongly  $T$ -dependent line shape. We developed a microscopic model based on the Tomonaga-Luttinger theory, with which we show that the shake-up process can explain the observed results. Specifically, electron-electron interactions modify the Van Hove singularity to the form  $(\omega - \Delta)^{2\alpha-1/2}$  at  $T=0$ . Our model accurately reproduces the observed  $T$ -dependent line shape, determining  $\alpha$  to be 0.016, 0.026, and 0.048, at 20, 30, and 40 T, respectively.

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