

Frequency splitting of intervalley phonons in graphene

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Abstract – We study the thermal distribution of intervalley phonons in a graphene sheet. These phonons have two components with the same frequency. The degeneracy of the two modes is preserved by weak electron-phonon coupling. A sufficiently strong electron-phonon coupling, however, can result in a splitting into an optical and an acoustic phonon branch, which creates a fluctuating gap in the electronic spectrum. We describe these effects by treating the phonon distribution within a saddle-point approximation. Fluctuations around the saddle point indicate a Berezinskii-Kosterlitz-Thouless transition of the acoustic branch. This transition might be observable in the polarization of Raman scattered light.

Graphene, a two-dimensional sheet of carbon atoms forming a honeycomb lattice, has highly unusual electronic properties [1–4]. This is due to the fact that there are two bands that touch each other at two Dirac nodes. Moreover, the low-energy quasiparticles of undoped graphene experience a linear dispersion around the Dirac nodes. Transport properties, characterized by the longitudinal conductivity at the Dirac nodes, are quite robust and do not vary much from sample to sample. Exactly at the Dirac point a minimal DC conductivity has been observed in a number of experiments [1–3]. Its value is not much affected by disorder [5,6]. A typical source of disorder in graphene are frozen lattice deformations (ripples) [7,8]. In a more realistic description, these deformations may not be frozen but fluctuate thermally due to the softness of the two-dimensional material.

In this paper we investigate effects of the electron-phonon interaction in graphene. Considering only in-plane displacements of graphene atoms, we have four different phonon modes. For low-energy electronic states electron-phonon interaction is efficient if the phonon wave vector is close to Γ , K or K' points. As was explained by Basko and Aleiner [9], the only modes which are effectively coupled to electrons are the pseudovector optical phonons corresponding to the irreducible representation E_2 of the group C_{6v} from the Γ -point, and the scalar phonons corresponding to the

irreducible representations A_1 and B_1 of the same group from the points K and K' .

Experimental information on the interaction is obtained by Raman spectroscopy [10–14] and by angle-resolved photoemission spectroscopy [15]. The above-mentioned theoretical analysis of the role of different phonons is confirmed by the fact that in the Raman spectrum of graphene only two two-phonon peaks are seen: the D^* peak near $2\omega_{A_1} = 2650 \text{ cm}^{-1}$ and the G^* peak near $2\omega_{E_2} = 3250 \text{ cm}^{-1}$, corresponding to scalar and pseudo-vector phonons, respectively. In our paper we will consider the interaction of electrons only with the scalar phonons.

The physics of the electron-phonon system is defined by the Hamiltonian with optical (Einstein) phonons at frequency ω_0 [16]:

$$H = \sum_{\mathbf{r}, \mathbf{r}'} \left[\omega_0 b_{\mathbf{r}, \mathbf{r}'}^\dagger b_{\mathbf{r}, \mathbf{r}'} + [h_{\mathbf{r}, \mathbf{r}'} + \alpha(b_{\mathbf{r}, \mathbf{r}'} + b_{\mathbf{r}, \mathbf{r}'}^\dagger)] c_{\mathbf{r}}^\dagger c_{\mathbf{r}'} \right]. \quad (1)$$

Here $c_{\mathbf{r}}^\dagger$ ($c_{\mathbf{r}}$) are the electron creation (annihilation) operators and $b_{\mathbf{r}, \mathbf{r}'}^\dagger$ ($b_{\mathbf{r}, \mathbf{r}'}$) are the phonon creation (annihilation) operators. This Hamiltonian describes an effective attraction between the fermions and a renormalization of the electronic hopping amplitude $h_{\mathbf{r}, \mathbf{r}'}$, reducing the hopping rate substantially (polaron effect). The attractive interaction may lead to the formation of Cooper pairs. In 2D this effect is not relevant due to strong fluctuations, preventing the system to become superconducting.

The conventional approach to determine the properties of the phonons and the electrons is based on a self-consistent evaluation of the self-energy (Migdal approximation) [17,18]. The latter provides an effective (or renormalized) energy and its imaginary part an effective scattering rate. Such a static approximation might be insufficient in a two-dimensional system, since it does not take into account thermal fluctuations. This was already discussed in an experimental study of graphene [13]. To avoid this problem, we include thermal fluctuations in our approach. To this end, we replace the phonon operators $b_{\mathbf{r},\mathbf{r}'}$, $b_{\mathbf{r},\mathbf{r}'}^\dagger$ by their quantum average: $b_{\mathbf{r},\mathbf{r}'} \approx \langle b_{\mathbf{r},\mathbf{r}'} \rangle \equiv v_{\mathbf{r},\mathbf{r}'}$ and $b_{\mathbf{r},\mathbf{r}'}^\dagger \approx \langle b_{\mathbf{r},\mathbf{r}'}^\dagger \rangle \equiv v_{\mathbf{r},\mathbf{r}'}^*$. In this approximation we can keep thermal fluctuations but ignore quantum fluctuations of the phonons. The electrons, on the other hand, are studied in full quantum dynamics. This reduces the grand-canonical ensemble at inverse temperature β , defined by the generating function $\text{Tr}e^{-\beta H}$ to a functional integral with respect to thermal fluctuations of the lattice distortations $v_{\mathbf{r},\mathbf{r}'}$ and a trace with respect to the electrons. It should be noticed that only the real part $\text{Re}(v)$ couples to the electrons. After performing the trace over the electrons we get

$$\text{Tr}e^{-\beta H} \approx \int \det(\mathbf{1} + e^{-\beta h}) e^{-\beta S_0} \mathcal{D}[\vec{u}] \equiv Z \quad (2)$$

with

$$S_0 = \frac{\omega_0}{2\alpha^2} \sum_{\mathbf{r},\mathbf{r}'} u_{\mathbf{r},\mathbf{r}'}^2$$

for the rescaled phonon field $u_{\mathbf{r},\mathbf{r}'} = \alpha v_{\mathbf{r},\mathbf{r}'}$.

The graphene unit cell contains two atoms, each of them has one π -orbital. Its band structure has two bands with two nodes (or valleys). It is convenient to introduce a sublattice representation for the tight-binding Hamiltonian h and to expand it around the two valleys K and K' . This expansion leads to a wave function that is represented by a 4-component column vector. We shall work in a direct product vector space of the valley space and the sublattice space. For the phonons we consider here only the intervalley contribution, which play a major role in the electron-phonon interaction [9]. This gives for the low-energy Hamiltonian

$$h = -it\Pi_3\vec{\partial} + \vec{u} \cdot \vec{\Pi}\Sigma_0. \quad (3)$$

The parameter t is the bandwidth. Here we have used a coordinate system that refers to one sublattice and one valley in the notation of ref. [19]: the indices of the Pauli matrices Σ_j ($j = 0, \dots, 3$) are acting on the A-B sublattices and the indices of the Pauli matrices (Π_j ($j = 0, \dots, 3$)) are acting on the two valleys K and K' . Moreover, we have $\vec{\partial} = \vec{\Sigma} \cdot \vec{\partial}$, where $\vec{\partial}$ is a lattice difference operator. For graphene in a homogeneous magnetic field this electron-phonon interaction was considered in ref. [20]. Then the

intervalley phonon field has two components in the valley space:

$$\begin{pmatrix} 0 & u_{\mathbf{r}} \\ u_{\mathbf{r}}' & 0 \end{pmatrix} = u_{1,\mathbf{r}}\Pi_1 + u_{2,\mathbf{r}}\Pi_2,$$

where $u_{1,\mathbf{r}} = (u_{\mathbf{r}} + u_{\mathbf{r}}')/2$ and $u_{2,\mathbf{r}} = i(u_{\mathbf{r}} - u_{\mathbf{r}}')/2$ [16].

The integral Z in eq. (2) serves as a generating function that allows us to get, for instance, the static electronic Green's function by differentiation of $\ln Z$ as

$$G_{\mathbf{r},\mathbf{r}'} = \frac{1}{Z} \int (\mathbf{1} + e^{-\beta h})_{\mathbf{r},\mathbf{r}'}^{-1} e^{-\beta S} \mathcal{D}[\vec{u}] \quad (4)$$

with

$$S = S_0 - \beta^{-1} \log \det(\mathbf{1} + e^{-\beta h}), \quad \vec{u} = (u_1, u_2). \quad (5)$$

It is useful to notice that $e^{-\beta S} = e^{-\beta S_0} \det(\mathbf{1} + e^{-\beta h})$ is a non-negative function. Therefore, $e^{-\beta S}/Z$ is a probability density for the phonon field, and the static one-particle Green's function then can also be written as an average $\langle \dots \rangle_{ph}$ with respect to the distribution $e^{-\beta S}/Z$ [21]:

$$G_{\mathbf{r},\mathbf{r}'} = \langle (\mathbf{1} + e^{-\beta h})_{\mathbf{r},\mathbf{r}'}^{-1} \rangle_{ph}. \quad (6)$$

The distribution $e^{-\beta S}/Z$ is invariant under a unitary transformation. For the special four-dimensional transformation

$$h \rightarrow U h U^\dagger, \quad U = \begin{pmatrix} 1 & 0 \\ 0 & e^{-i\varphi} \end{pmatrix} \Sigma_0 \quad (7)$$

with $0 \leq \varphi < 2\pi$ the electron-phonon coupling term in eq. (3) satisfies the relation

$$U \vec{u} \cdot \vec{\Pi} U^\dagger = O \vec{u} \cdot \vec{\Pi}, \quad (8)$$

where O is the orthogonal transformation (*i.e.* rotation by angle φ) in the space of the matrices Π :

$$O = \cos \varphi \Pi_0 + i \sin \varphi \Pi_2 = \exp(i\varphi \Pi_2).$$

Since the gradient part of the Hamiltonian h is invariant under a global rotation of the phonon field \vec{u} , the distribution $e^{-\beta S}/Z$ is also invariant. As a consequence, the phonon field can produce spontaneously a dimerization $\langle \vec{u} \rangle \neq 0$ with massless fluctuations.

Second-order perturbation theory is the standard approach for evaluating the change of the phonon frequency by the electron-phonon interaction [7,22,23]. In our model, defined by the action (5), second order in α gives us for the renormalized frequency of the intervalley mode,

$$\alpha^2 \frac{\partial^2 S}{\partial u_i \partial u_j} \Big|_{u=0} = (\omega_0 - a_2) \delta_{ij} \quad (9)$$

with

$$a_2 = \frac{\alpha^2}{\pi \beta t^2} \ln \left[\frac{\cosh(\beta E_F) + \cosh(\beta t \Lambda)}{1 + \cosh(\beta E_F)} \right], \quad (10)$$

where $\Lambda = 2\sqrt{\pi}$ is the momentum cutoff. For $\beta t \gg 1$, which is satisfied even at room temperature due to $t \approx 2.7$ eV, the renormalized phonon frequency has the asymptotic behavior

$$\omega_u \sim \omega_0 - \frac{\alpha^2}{t^2\pi} (2\sqrt{\pi}t - |E_F|).$$

Thus, the intervalley phonons are softened by the electron-phonon coupling. Moreover, the phonons harden as we go away from the Dirac point $E_F = 0$, in agreement with recent experiments [13,14,24]. However, this frequency becomes negative if $\omega_0 < a_2$, indicating an instability of the electron-phonon system. In particular, the vanishing frequency reveals a phonon softening, where the optical phonon mode becomes acoustic. In the following we shall discuss that this instability is associated with a splitting of the degenerate phonon modes, where instead of a single phonon frequency two different frequencies appear.

The instability cannot be treated within second-order perturbation theory but requires a self-consistent approach. Here it is natural to perform the integration in eqs. (2) and (4) in a saddle-point (SP) approximation. This leads to the SP equation $\delta_u S = 0$ which determines an average lattice dimerization $|\vec{u}| = \bar{u}$. Assuming a uniform solution \bar{u} , the SP equation $\partial S / \partial u_i = 0$ reads

$$\bar{u} = 2 \frac{\alpha^2}{\omega_0} \bar{u} \int \frac{1}{\epsilon_k} \frac{\sinh(\beta\epsilon_k)}{\cosh(\beta E_F) + \cosh(\beta\epsilon_k)} \frac{d^2k}{(2\pi)^2} \quad (11)$$

with the dispersion $\epsilon_k = \sqrt{t^2k^2 + \bar{u}^2}$ of the electrons. A nonzero \bar{u} opens an electronic gap $\Delta = 2\bar{u}$. This equation fixes only \bar{u} , not the direction of the vector $\vec{u}_0 \equiv (\bar{u} \cos \varphi, \bar{u} \sin \varphi)$. For a nontrivial solution $\bar{u} \neq 0$ we can perform the integration in eq. (11) and obtain the equation

$$\frac{\cosh(\beta E_F) + \cosh(\beta\sqrt{\Lambda^2 t^2 + \bar{u}^2})}{\cosh(\beta E_F) + \cosh(\beta\bar{u})} = e^{\beta\pi t^2 \omega_0 / \alpha^2}. \quad (12)$$

Then the critical point β_c of the instability is determined from this equation for $\bar{u} = 0$. For $\beta t \gg 1$ and $|E_F| < \bar{u}$ we get from eq. (12)

$$\bar{u} \sim \frac{2\alpha^2}{\omega_0} \left(1 - \frac{\pi t^2 \omega_0^2}{4\alpha^4} \right) \quad (13)$$

and for $\beta t \gg 1$, $|E_F| > \bar{u}$

$$\bar{u} \sim \sqrt{\left(\frac{\pi t^2 \omega_0}{\alpha^2} + |E_F| \right)^2 - 4\pi t^2}. \quad (14)$$

While an increase of the electron-phonon coupling α increases \bar{u} for $|E_F| < \bar{u}$, it decreases \bar{u} for $|E_F| > \bar{u}$. This result indicates that the instability of the $\bar{u} = 0$ solution is supported by the electron-phonon interaction near the Dirac point, whereas the instability is suppressed further away from the Dirac point.

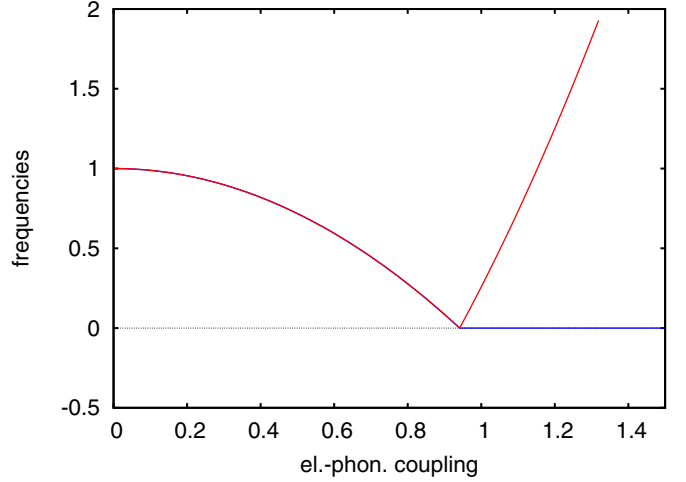


Fig. 1: (Colour on-line) Splitting of the phonon modes (from eqs. (17), (18)). The red (blue) curve represents ω_1 (ω_2) in units of ω_0 , indicating that for the dimensionless electron-phonon coupling $\alpha/\sqrt{t\omega_0} > a_c \approx 0.94$ there exist two different frequencies.

For small \bar{u} eq. (12) can be expanded. This allows us to study the properties of the phonon renormalization for nonzero \bar{u} , *i.e.* in the transition regime where $\bar{u} \sim 0$ and the perturbation theory is unstable. The SP equation then reads

$$\alpha^2 \frac{\partial S}{\partial u_j} \Big|_{u=\bar{u}} \approx (\omega - a_2 + a_4 \bar{u}^2) \bar{u}_j = 0. \quad (15)$$

Besides the trivial solution $\bar{u} = 0$ this equation has the nonzero solutions

$$\bar{u} = \pm \sqrt{a_2 - \omega_0} / \sqrt{a_4} \quad (a_2 - \omega_0 \geq 0).$$

The stability matrix is

$$\alpha^2 \frac{\partial^2 S}{\partial u_i \partial u_j} \Big|_{u=\bar{u}} = 4(a_2 - \omega_0) \frac{\bar{u}_i \bar{u}_j}{\bar{u}^2} \quad (16)$$

instead of (9). Since the SP solution \vec{u}_0 is on a circle with radius \bar{u} , the eigenvalues of the matrix on the right-hand side are $\omega_1 = 4(a_2 - \omega_0)$ and $\omega_2 = 0$. For a stable solution with $\bar{u} = 0$, on the other hand, the stability matrix agrees with the result of the second-order perturbation theory of eq. (9). Combining (9) (for $a_2 < \omega_0$) and (16) (for $a_2 \geq \omega_0$), we obtain the renormalized phonon frequencies as

$$\omega_1 \approx |a_2 - \omega_0| [1 + \Theta(a_2 - \omega_0)], \quad (17)$$

$$\omega_2 \approx (\omega_0 - a_2) \Theta(\omega_0 - a_2). \quad (18)$$

Thus, the ω_2 mode, which is related to the A_1 mode with frequency 1325 cm^{-1} of ref. [9], vanishes for the solution $\bar{u} \neq 0$, as plotted in fig. 1.

After having solved the SP equation for a uniform phonon field \vec{u} , we study the fluctuations around this solution. Fluctuations may play an important role and should modify the uniform solution due to the two-dimensionality of graphene. In particular, the fluctuations

are crucial for the vanishing phonon frequency ω_2 in eq. (18).

With the special SP solution \vec{u}_0 also any rotated phonon mode $O\vec{u}_0$ is a SP solution. Therefore, we can choose the special solution $\vec{u}_0 = (\bar{u}, 0)$ and, using the identity (8), obtain the action at the SP as

$$S \approx S_0 - \frac{1}{\beta} \log \left[\det(\mathbf{1} + e^{-\beta(-it\Pi_3\phi + \bar{u}U\Pi_1U^\dagger)}) \right],$$

which is degenerated with respect to a global unitary transformation U . In order to study fluctuations around the SP solution we introduce a spatially fluctuating unitary matrix U_r . This can be cast into a nonlinear matrix field

$$\Phi_{\mathbf{r}} = U_{\mathbf{r}}\Pi_1U_{\mathbf{r}}^\dagger = \begin{pmatrix} 0 & e^{i\varphi_{\mathbf{r}}} \\ e^{-i\varphi_{\mathbf{r}}} & 0 \end{pmatrix} \Sigma_0, \quad (19)$$

such that the action becomes

$$S \approx S_0 - \frac{1}{\beta} \log \left[\det(\mathbf{1} + e^{-\beta(-it\Pi_3\phi + \bar{u}\Phi)}) \right]. \quad (20)$$

This action can be expanded, either for $\bar{u}/t < 1$ or for $\bar{u}/t > 1$.

Now we assume that $\bar{u} > t$. Formally, t can be chosen independently of the real bandwidth of the system. But this choice means that the physics is restricted to quasiparticles up to energy t . Then the expansion of the action S of eq. (20) in powers of $1/\bar{u}$ provides a $U(1)$ nonlinear sigma model:

$$S \approx S'_0 + F(\beta, \bar{u}) Tr(\partial\Phi\partial\Phi) \quad (21)$$

with the prefactor

$$F(\beta, \bar{u}) = \frac{t^2}{\bar{u}^2} \left[\frac{\bar{u}}{4} + \frac{1}{\beta} \ln(1 + e^{-\beta\bar{u}}) + \frac{1}{2} \frac{\bar{u}}{1 + e^{\beta\bar{u}}} \right]. \quad (22)$$

Using the result of \bar{u} for $\beta\bar{u} > \beta t \gg 1$, the β dependence drops out of the prefactor:

$$F \sim \frac{t^2}{4\bar{u}} \sim \frac{t^2\omega_0}{32\pi\alpha^2} \frac{1}{1 - t^2\omega_0^2/16\alpha^4}. \quad (23)$$

This result can be considered as a renormalization effect for the temperature, where the renormalized dimensionless temperature reads

$$\tau = \frac{\bar{u}}{t^2} T \sim \frac{2\alpha^2}{\omega_0 t^2} \left(1 - \frac{\pi t^2 \omega_0^2}{4\alpha^4} \right) T. \quad (24)$$

Here we have assumed $|E_F| < \bar{u}$ and have used the expression of \bar{u} in eq. (13). As the electron-phonon coupling α increases, the renormalized temperature τ increases as well. This reflects the fact that the electronic fluctuations enhance the phonon fluctuations. The case $|E_F| > \bar{u}$ is rather unrealistic here, since we also have assumed $\bar{u} > t$.

The fluctuating term in eq. (21) represents an XY model for the angular fluctuations φ_r . Thus, the phonon fluctuations undergo a Berezinskii-Kosterlitz-Thouless (BKT)

transition if τ_c is of order unity [25]. For temperatures below the BKT transition point τ_c the fluctuations are strongly correlated whereas above this temperature the correlations of the fluctuations decay exponentially due to the proliferation of vortex pairs.

We conclude from our calculation that second-order perturbation theory with respect to electron-phonon interaction reveals a phonon softening. This result is in agreement with other calculations [7,22,23]. For sufficiently large electron-phonon coupling the perturbative approach breaks down and a self-consistent calculation is necessary. We have used an SP approximation for the phonon fluctuations and found an instability of the perturbative approach due to a splitting of the optical phonon mode into an optical and an acoustic branch. The latter is related to a massless mode caused by the rotational symmetry of the system. It is characterized by long-ranged correlations of the phonon fluctuations, in contrast to the short-range fluctuations of the optical phonons. However, at sufficiently high temperatures these long-range correlations can undergo a BKT transition by the creation of vortex pairs, resulting again in short-range correlated fluctuations. The average lattice dimerization vanishes for all regimes as a consequence of the Mermin-Wagner argument. A possible way to observe the BKT transition experimentally is to measure the polarization in Raman scattering, since the photon polarization couples to the direction of the lattice distortions.

The instability of the intervalley phonons is very different from the instability of the Holstein (out-of-plane) phonons studied recently, where the phonons undergo an Ising transition [26]. In the latter case only short-range correlated fluctuations appear (except for the critical point) but the average distortion has a nonzero value.

To understand the effect of the thermal phonon fluctuations on the electronic transport properties, we can return to eq. (6). Then $\vec{u}_{\mathbf{r}}$ produces a random gap in the electronic spectrum which has to be averaged with respect to the phonon distribution $e^{-\beta S}/Z$. Thus, the effect of thermal phonons is similar to the effect of frozen correlated disorder, depending on the temperature of the sample though. Since an uncorrelated random gap with vanishing mean does not affect the minimal conductivity [27–29], we expect a similar result for thermally fluctuating intervalley phonons, at least near the Dirac point.

In conclusion, our calculation reveals that the electron-phonon interaction in graphene leads to a substantial renormalization of the intervalley phonons. For sufficiently strong electron-phonon coupling this causes an instability, where the phonon frequency vanishes and a new pair of phonons appears, consisting of an optical and an acoustic branch.

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