Manifestations of the electron-phonon interaction range in angle resolved photoemission spectra

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A theoretical study of the angle resolved photoemission spectra (ARPES) is presented for metallic systems with a low concentration of carriers interacting with phonons via the weak screened Fröhlich interaction. We show that for an efficient analysis of the screening one has to render the experimental ARPES intensity into the imaginary part of the electron self-energy. Several universal model-independent features of the phonon spectral component are revealed: (i) a universal screening dependence of the ARPES-intensity confinement inside the Fermi surface; (ii) similarity of the dispersion in the 1-st phonon sideband to that of the quasiparticle (QP) band at large Thomas-Fermi screening radii \( r_{TF} > 20 \) unit cells, and its anomalous shape at intermediate screenings \( 3 < r_{TF} < 20 \); (iii) the 2-nd phonon sideband is featureless, showing similarities to the QP dispersion only in its curvature. Manifestations of these predictions in the available ARPES data are discussed.

\textbf{Introduction.} It is well established both experimentally and theoretically that for low concentrations of carriers the electron-phonon coupling (EPC) produces phonon sidebands, appearing as satellites in ARPES below the QP peak \cite{1}. It has been argued \cite{2,3} that the energy and momentum intensity distribution of this phonon component depends on the spatial range of the EPC, the unscreened polar Fröhlich coupling having the longest and the Holstein on-site interaction the shortest ranges. However, despite extensive experimental data, we are not aware of any theoretical systematic study of ARPES in systems with the realistic screened polar Fröhlich interaction, which would consider the dependence of the spectra on the screening length. We can mention only a study of screened Fröhlich interaction for few particular cases \cite{2} in specific materials and the theoretical study \cite{3} of the EPC with a hypothetical forward scattering. Hence, search for universal manifestations of the screening dependence of the ARPES phonon component is of general interest for the experimental community. In this work we therefore perform a systematic analysis of the phonon sidebands for different screenings and show that there are several model- and dimension-independent properties featuring a universal relation to the screening length.

Recent experimental studies of the ARPES of polaronic materials encountered numerous cases when only a small part of the conduction band \( c_k \) at the bottom is filled and the concentration of itinerant charges is low \cite{4,12}. In this limit the Fermi energy, equal at zero temperature to the chemical potential \( \mu \) measured from the bottom of the conduction band, is considerably smaller than the total bandwidth. In particular, we consider the case \( \mu < \omega_0 \), where \( \omega_0 \) is the frequency of relevant optical phonons, observed in many realistic materials \cite{4,12}. This ensures a pattern of separated phonon sidebands, facilitating direct studies of the manifestations of the EPC screening.

\textbf{Theoretical description of phonon sidebands.} We use the standard EPC model, involving bare electrons in a band, \( \hat{H}_{el} = \sum_{k, \omega} f_k d^\dagger_k f_k a_k \), dispersionless optical phonons with frequency \( \omega_0 \), \( \hat{H}_{ph} = \omega_0 \sum \mathbf{q} b^\dagger_{\mathbf{q}} b_{\mathbf{q}} \), and the screened Fröhlich interaction,

\begin{equation}
\hat{H}_{el-ph} = \sum_{k, \mathbf{q}} V_d(\mathbf{q}) a^\dagger_{k+\mathbf{q}} a^\dagger_k (b^\dagger_{\mathbf{q}} b_{\mathbf{q}}) , \tag{1}
\end{equation}

\begin{equation}
|V_d(\mathbf{q})|^2 = a_{d=2} (|\mathbf{q}|^{d-1} + q_{TF}^{d-1}) . \tag{2}
\end{equation}

Here, \( a_{d=3} = 2\sqrt{2}\pi\alpha \) for three (3D) and \( a_{d=2} = \sqrt{2}\pi\alpha \) for two-dimensional (2D) systems \cite{13,17}, with \( \alpha \) characterizing the strength of the EPC. Within this model, it is assumed that the electron is embedded in \( d \)-dimensional lattice, with \( 1/r \) electron-polarization interaction \cite{16}, whose screening is static and characterized by the Thomas-Fermi wave number \( q_{TF} \). The direct space radius of the screening, \( r_{TF} = \pi/q_{TF} \), roughly gives the screening range in the lattice constant units. The unscreened Fröhlich interaction corresponds to \( r_{TF} \to \infty \), while the on-site Holstein coupling implies \( r_{TF} \to 0 \).

The spectral function \( A(\mathbf{k}, \omega) \), measured for \( \omega < 0 \) (electron removal processes), is determined by the electron Green function \( G(\mathbf{k}, \omega) \), \( A(\mathbf{k}, \omega) = -\text{Im} G(\mathbf{k}, \omega)/\pi \). It may be expressed in terms of the electron self-energy \( \Sigma(\mathbf{k}, \omega) \), appearing due to the coupling \cite{1} to phonons,

\begin{equation}
A(\mathbf{k}, \omega) = \frac{-\text{Im} \Sigma(\mathbf{k}, \omega)}{\omega - \xi_\mathbf{k} - \text{Re} \Sigma(\mathbf{k}, \omega)} + |\text{Im} \Sigma(\mathbf{k}, \omega)|^2 , \tag{3}
\end{equation}

with \( \xi_\mathbf{k} = \epsilon_\mathbf{k} - \mu < 0 \), the hole energy measured from the Fermi level. The EPC leads to a complex structure of
\( A(k, \omega) \), where in addition to the QP peak one observes phonon sidebands, with the \( n \)-th sideband being shifted downwards from the Fermi level by \( n\omega_0 \). Since the ARPES seldom show more than two satellites \([8, 9, 11]\), we concentrate on the first two sidebands. Within the zero-temperature diagrammatic expansion (see \([18]\) and the Supplemental Material \([19]\)), the first-order contribution in \( \alpha \) to the imaginary part of the self-energy (\( \text{Im} \Sigma \)),

\[
\text{Im}\Sigma^{(n=1)}(k, \omega) = -\pi \sum_{BZ} |V_d(q)|^2 \Theta(-\xi_{k+q}) \delta(\omega - \xi_{k+q} + \omega_0), \tag{4}
\]

is the leading contribution to the 1-st phonon sideband, with the finite intensity restricted to the frequency window \([-\omega_0 - \mu, -\omega_0]\). The 2-nd sideband is in the frequency window \([-2\omega_0 - \mu, -2\omega_0]\), with the leading contribution given by second-order \( \alpha^2 \) (two-phonon) terms,

\[
\text{Im}\Sigma^{(n=2)}(k, \omega) = -\pi \sum_{BZ} \Theta(-\xi_q) \delta(\omega - \xi_q + 2\omega_0) \times \frac{\sum_{q'}^ {BZ} |V_d(q')|^2}{\xi_q - \xi_{k+q'}} \times \frac{1}{(\xi_q - \xi_{k+q'} - \omega_0)} . \tag{5}
\]

The approximate expression for the ARPES intensity corresponding to the first two sidebands is given by,

\[
A^{(n=1,2)}(k, \omega) \sim \frac{\text{Im}\Sigma^{(n=1,2)}(k, \omega)}{\omega - \epsilon_k}, \tag{6}
\]

where for weak couplings, in leading order approximation, we neglect self-energy contributions in the denominator of Eq. \((6)\). For stronger EPCs, following Ref. \([20]\), the real part of the self-energy may be absorbed into a renormalized dispersion \( \tilde{\epsilon}_k, \epsilon_k = \epsilon_k + \text{Re}\Sigma(k, \omega) \). This facilitates experimental analysis, because \( \tilde{\epsilon}_k \) is a measured quantity, whereas the bare \( \epsilon_k \) is not directly accessible. Note, the condition \( \mu < \omega_0 \) excludes singularities because the denominator in Eq. \((6)\) is nonzero in the frequency windows of phonon sidebands.

**Numerical calculations.** For numerical calculations we set \( \omega_0 = 1 \) and consider the dispersion \( \epsilon_k = 2t \sum_{\alpha=1}^2 (1 - \cos(k_i)) \) of hypercubic lattice in 3D/2D, where \( t \) is the nearest neighbor hopping. With the lattice constant \( a = 1 \), the effective mass at the bottom of the band is \( m_0 = 1/2t \). The intersection with \( \mu \) in the \([k'k'k'']/[k'00]\) direction of the 3D \((2D)\) models is at \( k' = \text{arccos}[1 - (\mu/(2dt))] \), representing the Fermi momentum \( k_F \) of the Fermi surface (FS). To search for general properties of the ARPES phonon components in various systems, we use two very different sets of parameters: \( S1 \) (\( S2 \)) denotes \( \mu = 0.5 \) (\( \mu = 0.1 \)) and \( t = 1 \) (\( t = 1/24 \)), with mass \( m_0^S = 0.5 \) (\( m_0^S = 12 \)). In both these cases, only a small fraction of the lowest band states is occupied.

The multidimensional integration in Eqs. \((4,5)\) is impossible to perform with a fixed mesh of momenta because the Van Hove singularities are present in the band density of states and sharp peaks coming from EPC vertices require a dense mesh. E.g., for 6-dimensional integration in Eq. \((5)\), even a sparse mesh with only 100 points per dimension invokes a summation over \( 10^{12} \) points, which is clearly outside of reasonable calculation times. To overcome these numerical limitations, the integration is performed by the importance sampling scheme similar to that used for the fixed diagram order integration in Diagrammatic Monte Carlo method \([21]\).

Our calculations elucidate several universal features of ARPES phonon sidebands that are independent of specific details of a measurement or material parameters, i.e. the direction \([klm]/[kl]\), the dimensionality \(3D/2D\) or the parameter set \(S1/S2\).

**Momentum-space confinement of phonon sidebands.** The first important finding concerns the confinement of the ARPES spectra in the Brillouin Zone (BZ) area, \( k \leq \pi \). Our calculations of the 1-st sideband of the ARPES intensity and of the corresponding \( \text{Im}\Sigma \) are presented in Fig. \((1)\) (a) and (b), respectively. It is often concluded in experimental papers that the higher intensity accumulated within the FS, \( k \leq k_F \ll \pi \), identifies the Fröhlich interaction with a weak screening. However, careful consideration of the Eq. \((6)\) leads to the renouncement of this delusion. Let us consider the example of extremely strong screening, equivalent to the Holstein-type of on-site coupling. In this case, \( \text{Im}\Sigma^{(n=1)}(k, \omega) \) is fully
momentum independent and uniformly spread over the whole BZ, as shown in Fig. 2(b). Indeed, the two energy distribution curves (EDCs) in the right panel of Fig. 2(b), exhibiting the energy dependence of the \( \text{Im}\Sigma \) in the BZ cut at fixed momentum, are almost indistinguishable: the energy dependence follows the density of occupied electron states at the bottom of the band, shifted by \( \omega_0 \). Assuming a momentum independent interaction \( V_0(q) \), this behavior is readily obtained from Eq. (4). However, \( A^{(n=1)}(k,\omega) \) in Fig. 2(a) exhibits strong momentum dependence. The cause is the denominator in Eq. (6), which highlights the area within the FS and easily camouflages the fact that the EPC might be over-screened. Comparison of the results for \( A^{(n=1)}(k,\omega) \) and \( \text{Im}\Sigma^{(n=1)}(k,\omega) \) in Fig. 2 represents a straightforward theoretical support for the experimental fact that the intensity of the ARPES sidebands is always confined in the momentum-space region near the band minimum \( \{ \{3|9|11|12 \} \). As demonstrated in Fig. 2, the \( \text{Im}\Sigma \) of the sideband component of ARPES intensity is the most relevant quantity to study the screening effects. Experimentally, \( \text{Im}\Sigma^{\exp}(k,\omega) \) can be obtained from the ARPES \( A^{\exp}(k,\omega) \) measured in the frequency range of the sidebands, and the \( k,\omega \) dependence of the QP spectrum. A general data processing protocol, valid even in the cases when QP spectrum is strongly broadened, is given in the Supplemental Material \( 19 \) while in the simplest case of sharp QP peak with dispersion \( \epsilon^{\exp}_k \) one can use the following expression

\[
\text{Im}\Sigma^{\exp}(k,\omega) \sim A^{\exp}(k,\omega) [\omega - \epsilon^{\exp}_k]^2 .
\] (7)

To evaluate from experimental data the intensity confinement of the \( \text{Im}\Sigma \) within the FS, we define few relevant estimators. The first one is \( R_n(k) = \int_{\omega_0}^{\omega_0 + \mu} d\omega \text{Im}\Sigma^{(n)}(k,\omega) \), corresponding to \( \text{Im}\Sigma \) integrated over energies within the \( n \)-th phonon sideband. Then, using \( R_n(k) \), the confinement within the FS may be given in terms of the ratio of the intensity within and outside the FS,

\[
R_n = \left( \int_0^{k_F} dk \frac{R_n(k)}{k_F} \right) / \left( \int_{k_F}^{\pi} dk \frac{R_n(k)}{\pi - k_F} \right) .
\] (8)

Since \( R_n \) is normalized per unit momentum, \( R_n \to 1 \) when \( r_{TF} \to 0 \), while for weak screenings \( (r_{TF} \gtrsim 20) \) saturating to its maximal value. Although for different systems this maximal value may differ by an order of magnitude, for all 8 cases considered (parameter sets S1/S2, 3D/2D, diagonal \([k'k'k']/[k'k']\) and nondiagonal \([k'00]/[k',0] \) directions) and for the both sidebands, the scaled measure of the confinement as a function of \( r_{TF} \),

\[
C_{n}(r_{TF}) = \frac{R_n(r_{TF}) - 1}{R_n(r_{TF} = \infty) - 1} + 1 ,
\] (9)

shown in Fig. 3 demonstrates a fairly universal behavior. Thus, assuming that a spectrum of a reference material is measured near maximal \( r_{TF} \) values \( (r_{TF} \gtrsim 20) \), the universal behavior of \( C_{n}(r_{TF}) \) may be used to determine through series of measurements how \( r_{TF} \) changes in similar materials as a function of some material parameter, e.g. doping. The map of \( \text{Im}\Sigma \) in Fig. 3 shows that the weak screening regime \( (r_{TF} \gtrsim 20) \) can be unambiguously identified using a certain universal feature of the 1-st phonon sideband. Namely, in this regime, the EDCs and the momentum distributions curves (MDCs, providing the intensity dependence in the BZ cut at fixed energy), have to reveal maxima whose momentum dependencies almost exactly follow the QP dispersion.
FIG. 4. Contour plots of (a) ImΣ^{(n=2)}(k, ω) and (b) its second derivative over momentum ∂^2 ImΣ^{(n=2)}(k, ω)/∂k^2 for the 3D parameter set S1 at r_{TF} = 100. Right panels in (a) and (b) are the EDCs and upper panels are MDCs along the cuts shown in the left-bottom panels of (a) and (b).

On the other hand, we never observe the EDC and MDC maxima in ImΣ of 2-nd phonon sideband, with its typical intensity distribution shown in Fig. 3 and zoomed in Fig. 4(a). However, by taking the second derivative ∂^2 ImΣ^{(n=2)}(k, ω)/∂k^2, which is in line with the conventional experimental ARPES-data processing using the curvature analysis, one recovers a dispersion resembling that of the QP peak. This universal property is well-illustrated by Fig. 4 comparing the intensities of ImΣ^{(n=2)}(k, ω), ∂^2 ImΣ^{(n=2)}(k, ω)/∂k^2, and the corresponding EDCs and MDCs.

Anomalous phonon sideband dispersions. As we already mentioned, for all 8 studied cases we found that at small screenings r_{TF} > 20, e.g. like in Fig. 3, the dispersion obtained from the EDCs and MDCs of ImΣ^{(n=1)}(k, ω) of the 1-st sideband follows the QP dispersion shifted by ω_0. However, this simple behavior preserves only as long as the screening is not too strong. Namely, we found an anomalous behavior of ImΣ^{(n=1)}(k, ω) for the screening radii 3 ≤ r_{TF} ≤ 20. In this screening range, as illustrated by Fig. 5 the curve that follows the EDC maxima may appear shifted upwards by ε_0 from the sideband position at r_{TF} ≥ 20, i.e. from the bottom of the 1-st phonon sideband. Moreover, in comparison to the QP dispersion, the fits of EDCs maxima with a parabola give different effective masses, e.g. m^* = 0.76 m_0 in the example presented in Fig. 5(a). These differences are nonuniversal: Figs. 5(c-d) show that m^* is smaller (larger) in 3D (2D) and ε_0 varies (does not vary) in 3D (2D). Furthermore, while in 3D the EDC maxima follow the parabolic dispersion over the whole 1-st sideband, in 2D the parabolic dispersion at small momenta transforms into a steep dispersion at large momenta. In any case, we note that all the anomalous behaviors found here are restricted to the specific screening range, 3 ≤ r_{TF} ≤ 20, for all considered models and dimensionalities.

As shown in Fig. 6 this anomalous sideband dispersion is observed in the ARPES spectra as well, either from the EDCs or from the standard curvature analysis. The values of ε_0 and m^* obtained from the ARPES and ImΣ are different due to denominator in Eq. (6). In particular, the comparison of Fig. 6(a) and Fig. 6(b) theoretical supports the experimental observation that the intensity of the ARPES sidebands is always mostly confined to momenta near the band bottom, k < k_r, where k_r is considerably smaller than k_F [4, 8, 11, 12].

Conclusions. In terms of the leading corrections in the EPC, we theoretically modeled the ImΣ and the ARPES in the frequency ranges of the 1st- and 2nd phonon side-
bands. Our results apply to metallic systems with a low concentration of carriers that interact with dispersionless optical phonons through a weak screened Fröhlich interaction. The static screening is characterized by the Thomas-Fermi radius \( r_{TF} \). We have shown that the role of the screening may be most efficiently analyzed in terms of \( \text{Im}\Sigma \)-intensity, obtained by transformation of the measured ARPES-intensity. We found that regardless of the model, dimensionality and the electron momentum direction \( [klm] \), the ARPES bear several universal features: (i) phonon sidebands involve well defined energy windows in the spectra; (ii) the ARPES intensity shows confinement within the Fermi surface as a universal feature; in addition, we have theoretically explained the experimental fact that the intensity of phonon sidebands tends to confine to momenta which are considerably smaller than the Fermi momentum \( k_F \); (iii) the 1-st phonon sideband repeats the shifted shape of the QP band for weak screenings \( r_{TF} \geq 20 \). This behavior undergoes significant changes at larger screenings, \( 3 \leq r_{TF} \leq 20 \): its shape and \( m^* \) change in 3D and 2D cases, whereas the bottom of its energy \( \epsilon_0 \) changes only in 3D; (iv) we never observe the EDC and MDC maxima in the 2-nd phonon sideband, which is fully in agreement with the experimental data; however, in accordance with experiments, the second derivative (curvature) \( \partial^2 \text{Im}\Sigma^{(n=2)}(k,\omega)/\partial k^2 \) reveals maxima of the corresponding EDCs and MDCs, which dispersion resembles the QP dispersion.

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FIG. 6. Contour plot of (a) ARPES component \( A^{(n=1)}(k,\omega) \) and (b) \( \text{Im}\Sigma^{(n=1)}(k,\omega) \) for the 3D parameter set S2 at \( r_{TF} = 5 \). The circles are maxima of the EDCs at corresponding cross sections of the BZ. These EDC maxima are fitted by parabolas, giving corresponding masses \( m^* \) and onsets \( \epsilon_0 \) of the phonon sidebands determined from the (a) ARPES and (b) \( \text{Im}\Sigma \).
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