Fluctuation effects in quasi-one-dimensional conductors: 
Optical probing of thermal lattice fluctuations

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We have studied the effect of thermal lattice fluctuations on the optical properties of the charge-density wave (CDW) condensates K\(_2\)MoO\(_3\) and (TaSe\(_4\))\(_2\)I. From the temperature dependence of the CDW gap absorption in the midinfrared frequency range, there is a strong indication of the important role played by the thermal lattice fluctuation and zero-point lattice motion. The latter removes the inverse square-root singularity expected for the case of a static distorted lattice. In fact, a considerable broadening (i.e., larger than \(k_B T\)) of the subgap tail absorption is found by increasing the temperature towards the transition temperature \(T_{CDW}\). We compare our experimental findings with the predictions of a theoretical model, which suggests that zero-point and thermal lattice motions have a similar effect on the electronic properties as disorder. Better agreement with the theory is obtained when the contribution to the disorder parameter from phase phonon modes is also taken into account.

I. INTRODUCTION

A central aspect of the charge-density-wave (CDW) phase transition is the appearance of the so-called fluctuation effects. In the preceding paper, we have extensively discussed the influence of the precursor effects on the CDW phase transition.\(^1\) Emphasis was given to the optical fingerprints related to the paraconductivity contribution, which develops in the so-called fluctuation regime below the mean-field critical temperature \(T_{MF}\), but above the temperature \(T_{CDW}\), where three-dimensional long-range order develops. The experiments reported in Ref. 1 give clear evidence for important deviations from conventional metallic behavior between \(T_{MF}\) and \(T_{CDW}\). The collective nature of the conductivity was established, while its frequency dependence along the chains suggests the presence of features such as a finite conductivity and a "quasipinned" response.\(^1\) Furthermore, a pseudogap-like excitation was also clearly identified.\(^1\)

In this paper, we focus our attention to the thermal lattice fluctuations below the transition temperature \(T_{CDW}\). In the great majority of one-dimensional compounds, the zero-point lattice motion \(Su = (\hbar/2M\omega_A)^{1/2}\) (where \(M\) is the mass displaced and \(\omega_A\) the amplitude mode frequency which determines the size of the actual zero-point motion) is of the same order of magnitude of the Peierls distortion and, consequently, one might expect important effects on the electronic density of states. One might also wonder why there should be a clearly developed gap excitation in the spectrum at all.\(^2\)

Indeed, several one-dimensional systems, e.g., KCP, (CH\(_3\))\(_x\), Pt-halogen chains,\(^3\)-\(^6\) showed a particular broadening of the single-particle excitation at \(2\Delta\) (which is opened at the Fermi surface, due to the periodic lattice distortion with twice Fermi wave vector \(2k_F\)).\(^2\) However, while there was no accepted theory of the temperature dependence of the lattice distortion and the gap parameter, most of the previous experiments were compared to empirical formulas or the mean-field (BCS) form with a renormalized temperature. Attempts have been also made to explain some of these facts in terms of solitons,\(^7\) disorder, or imperfect nesting.\(^8\) Furthermore, some of those materials (like, e.g., KCP) are plagued by a quite important extrinsic static disorder,\(^3\) which also may lead to states in the gap and, therefore, to broadening of the optical gap feature.

Recently, McKenzie and Wilkins demonstrated that, starting from the Peierls-Fröhlich Hamiltonian with electron-phonon coupling as introduced by Brazovskii and Dzyaloshinskii,\(^9\) the zero-point and the thermal lattice motions have a similar effect on optical properties as disorder.\(^2\) The appearance of substantial subgap-tail ab-
absorptions is predicted. First, these subgap tail absorptions, which broaden by increasing the temperature, determine a remarkable deviation from the expected inverse square-root singularity at $\Delta$, suggested by the static mean-field approach. Second, the density of states is nonzero for all energies. Third, when the temperature becomes of the order of a characteristic phonon frequency $\omega_A$, the disorder increases, and the smearing of the density of states increases considerably. In this paper, we report on our attempt to study these features in more depth by probing the effects of the thermal lattice fluctuations with optical methods on the model compound $K_{0.3}MoO_3$ and its alloys, and also on $(TaSe_4)_2$I. In these latter compounds, the static disorder, due to random positions of the counterions is absent, making the extrinsic disorder negligible.

The paper is organized as follows: in the second section, we briefly describe the experimental techniques and present the results. In the third section, we first give an account for the theoretical model, which will be used for the interpretation and discussion of the experimental findings. The analysis of the data leads to the evaluation of the temperature dependence of the disorder parameter, which governs the thermal lattice fluctuation effects. We show that a full understanding of the experimental data is only achieved when considering the contribution of the phase-phonon modes to the disorder parameter. The theoretical approach is consequently extended in order to account for the latter contribution and good agreement between the data and the theoretical prediction is then obtained. A brief summary of the most important experimental and theoretical findings will conclude our presentation. Some of our results have been published earlier.11,12

II. EXPERIMENT AND RESULTS

Single crystals of $(TaSe_4)_2$I were grown using a vapor transport technique. Large crystals of up to $10 \times 4 \times 1 \text{ mm}^3$ grew within a period of three days.11 The $K_{0.3}MoO_3$ specimens used in this study have been grown by electrolytic reduction of the molten mixture of $K_2MoO_4$ and $MoO_3$. We also studied tungsten (W)-alloyed crystals grown by the same technique. Doping with $WO_3$ yields crystals in which the W concentration varies widely, but doping with $K_2WO_4$ results in a nearly uniform impurity distribution, as confirmed by electron microprobe analysis. The crystals can reach more than $10 \times 10 \times 3 \text{ mm}^3$. The dc conductivity, measured by a four-wire technique, gives results similar to those previously reported,13 from which we inferred a CDW phase-transition temperature $T_{CDW}$ of 183 and 263 K for $K_{0.3}MoO_3$ and $(TaSe_4)_2$I, respectively.13

Reflectivity measurements as a function of temperature and with light polarized parallel to the chain direction were performed in a broad photon energy spectral range from $10^5$ down to approximately $15 \text{ cm}^{-1}$, using several spectrometers with overlapping energy ranges. A full description of the experimental techniques can be found in Ref. 1. The reflectivity measured over such a broad energy interval has been used for the Kramers-Kronig transformations, from which we obtain the optical functions $[\epsilon(\omega)$ and $\sigma(\omega)]$.14 To this end, the spectra have been extrapolated at higher frequencies (i.e., above the UV spectral range) and below FIR with the standard extrapolations.14,14

The complete excitation spectrum of $K_{0.3}MoO_3$ and $(TaSe_4)_2$I has been presented and thoroughly discussed elsewhere.11,11-15 For the present discussion, we will concentrate our attention on the midinfrared spectral frequency window. In this respect, Fig. 1 displays the optical conductivity at low temperature for both compounds, where the frequency axis has been scaled by the gap value. No remarkable difference has been obtained in the W-doped alloys of $K_{0.3}MoO_3$.

By lowering the temperature below $T_{CDW}$, the optical conductivity of $K_{0.3}MoO_3$ at low frequencies drops and one observes the appearance of several peaks (see, e.g., Fig. 1 of Ref. 11), due to the collective excitation of the CDW condensate, to the phase-phonon absorptions,11,14,14 and particularly to the progressive formation of a broad excitation at $\omega=2\Delta \approx 0.2 \text{ eV}$ (Fig. 1). Furthermore, a strong absorption at about 500 cm$^{-1}$ also characterizes the excitation spectrum in the CDW ground state.14,15 This striking feature has been assigned either to a high-
frequency phason mode\textsuperscript{14} or to a midgap state, due to a topological soliton.\textsuperscript{15} The absorption at 0.2 eV is ascribed to the CDW gap absorption $2\Delta$. In fact, the peak in the absorption is in fair agreement with the gap estimated from the activation energy for dc transport.\textsuperscript{1,13,14} Reference 2 suggests that the subgap states do not contribute to the transport, because they are much more strongly localized than the states with energy larger than $\Delta$. We also observe that this gaplike absorption seems to persist above $T_{\text{CDW}}$.\textsuperscript{1,15,16} Moreover, a recent scanning tunneling microscopy experiment is indicative of a gap of $140\pm20$ meV in $K_0.3MoO_3$ in the electronic density of states.\textsuperscript{17}

For (TaSe$_4$I, on the other hand, the low-frequency optical conductivity, displayed in Fig. 1 of Ref. 12, is very low at all temperatures and the excitation spectrum is dominated by the huge and sharp absorption at about $\omega=2\Delta-0.4$ eV (Fig. 1), which is also clearly seen above $T_{\text{CDW}}$ and is ascribed to the CDW gap, as well.\textsuperscript{1,13,14} We note that our optical results on (TaSe$_4$I bear a satisfactory similarity with the findings of two previous investigations.\textsuperscript{18}

III. DISCUSSION

As clearly manifested in Fig. 1, the observed optical conductivity in the mid-IR frequency range is quite different from the mean-field expectation, for which at all temperatures below $T_{\text{CDW}}$ the absorption is predicted to be zero for $\omega<2\Delta$ and contains an inverse-square-root singularity at $\omega=2\Delta$ (see Fig. 1).\textsuperscript{16} Indeed, the singularity is absent, and there is a significant tail below the maximum. Furthermore, as the temperature increases, broadening of the spectrum occurs on an energy scale much larger than $k_B T$.

The broadening is particularly remarkable for temperatures larger than approximately 50 K, and it is larger than what might be expected from anisotropy effects or interchain coupling. Huang and Maki\textsuperscript{8} have calculated the effect of anisotropic band structure on the density of states (DOS). This is related to the imperfect nesting of the Fermi surface (FS). Let $t_i$ and $t_j$ be the hopping integral parallel and perpendicular to the chains, respectively. The key quantity describing the imperfect nesting of the FS is

$$\epsilon_0 = \frac{t_i^2 \cos(ak_F)}{4t_i \sin^2(ak_F)},$$

where $a$ is the lattice constant along the chain. Huang and Maki found that the DOS $\rho(E)$ is zero for $0<E<\Delta-\epsilon_0$ and had a logarithmic singularity at $E=\Delta+\epsilon_0$. This means, relative to the single chain case, that the DOS is broadened on an energy scale of order $2\epsilon_0$. Hence, $\sigma(\omega)$ should be broadened on a scale $4\epsilon_0$.

In $K_0.3MoO_3$, x-ray scattering data gives $k_F a \sim 0.75\pi$ (Ref. 19) and from band-structure calculations\textsuperscript{20} Pouget and co-workers have estimated that $t_i \sim 180$ meV and $t_j \sim 25$ meV and $t_{ji} \sim 10$ meV, where I and II correspond to the different bands.\textsuperscript{19} The deviation from the perfect nesting is determined by $|t_i + t_{ji}| = 15$ meV. Hence, this is the value we should use for $t_i$ in Eq. (1), and, consequently, we estimate $\epsilon_0 = 0.44$ meV. This predicts a broadening of $\sigma(\omega)$ at most 2 meV, whereas the observed broadening (Fig. 1) is about 50–80 meV (i.e., $\sim 400-600$ cm$^{-1}$). Thus, the interchain coupling cannot explain the broadening of $\sigma(\omega)$, observed experimentally.

Therefore, we proceed to compare the optical results with the consequence of the thermal lattice fluctuations, using the theoretical approach first suggested by Mckenzie and Wilkins.\textsuperscript{2} In the following two sections, there will be, first of all, a review of the most relevant theoretical aspects with particular emphasis to the optical conductivity, and afterwards, a quantitative comparison between theory and experiment will be performed. It will be shown that a full understanding of the experimental data can be achieved by extending the theoretical approach to the phase-phonon modes contribution to the thermal lattice fluctuations.

A. Theory

The relevant theory has been worked out by Mckenzie and Wilkins, who showed that if the relevant phonon frequency at $q = 2k_F$, $\omega_4$, is much smaller than the optical frequency, as is the case in the optical absorption in most Peierls materials [e.g., for $K_0.3MoO_3$, $\omega_4 \sim 80$ K and for (TaSe$_4$I ($\omega_4 \sim 130$ K (Refs. 13 and 14)], the quantum and thermal lattice fluctuations can be modeled by static, Gaussian-random, backscattering potential, $\xi(x)$, with zero mean.\textsuperscript{2} Further, if the phonon dispersion near $q = 2k_F$ is ignored, $\xi(x)$ is fully characterized by the disorder-averaged correlator $\langle \xi(x)\xi(y) \rangle = \gamma \delta(x-y)$. In general, the dimensionless disorder parameter $\eta = \gamma / \hbar \omega_4 \Delta$, where $v_F$ is the Fermi velocity, has contributions from extrinsic disorder such as to impurities, $\eta_e$, as well as from the intrinsic disorder due to lattice fluctuations, $\eta_t$. The latter is related to the dimensionless electron-phonon coupling constant $\lambda$, the phonon frequency at $2k_F$, and temperature:\textsuperscript{2}

$$\eta = \eta_e + \eta_t = \eta_e + \lambda \frac{\pi \hbar \omega_4}{2\Delta} \coth \frac{\pi \hbar \omega_4}{2T}. \hspace{1cm} (2)$$

The strength of the disorder is proportional to the electron-phonon scattering rate for states well above the gap. The dimensionless disorder parameter of Eq. (2) fully determines the properties of the subgap states. At $T = 0$ it is also possible to write $\eta = (2\Delta / \hbar \omega_4) / \delta u / \mu_0^2$ (where, $\bar{\omega}$ is the bandwidth). Hence, when $\bar{\omega}$ is larger than the gap, as it usually is, the effect of the lattice fluctuations is reduced. This explains why a reasonably well-defined gap is still possible (i.e., measurable) even though $\delta u \sim \mu_0$ [e.g., for $K_0.3MoO_3$, $\delta u \approx 0.05$ A and for (TaSe$_4$I $\mu_0 \approx \delta u \approx 0.09$ A].\textsuperscript{2}

As stated above, there are significant modifications of the electronic density of states with respect to the rigid distorted lattice prediction.\textsuperscript{10} The calculation of the optical conductivity also reflects the basic features found in the calculation of the density of states.\textsuperscript{21} Moreover and even more interesting, it was found that the strong
subgap conductivity follows a universal scaling form.\textsuperscript{21} In fact, by scaling the calculated conductivity at different temperatures by the corresponding peak value \( \sigma_{\text{peak}} \) and the frequency by the half width \( (\Gamma) \) for the low-frequency side of the \( \sigma_0 \) peak, the scaled curves for all \( \eta \) values have a universal form independent of \( \eta \) below the peak frequency \( \omega_{\text{peak}} \).\textsuperscript{21}

However, in most quasi-one-dimensional materials, that undergo a CDW transition, there are a large number of atoms in the unit cell. For example, the unit cell of \( K\alpha_3\text{MoO}_3 \) contains 20 \text{MoO}_3 units. Consequently, there is a large number of distinct phonon modes that can couple to the electrons. Thus, the simple model of a single-phonon mode near \( 2k_F \), coupling to the electrons, considered in Refs. 2 and 21 is not realistic.

We consider then the generalization of this earlier work to the case where \( G \) phonons of frequency \( \omega_n(q) \) and coupling strength \( g_n \) couple to electrons with Fermi velocity \( v_F \), spin \( \sigma \) and are described by a spinor \( \Psi_j(x) \). The Hamiltonian of the system is therefore

\[
H = \int dx \Psi_j^+(x) \left[ -i v_F \sigma_j \frac{\partial}{\partial x} + \frac{1}{2} \left[ \Delta(x) \sigma_+ + \Delta^*(x) \sigma_- \right] \right] \Psi_j(x) + \sum_n \omega_n(q) b_n^+ b_n ,
\]

where \( b_n \) is the annihilation operator associated with the \( n \)th phonon mode. The field associated with the Peierls distortion is

\[
\Delta(x) = \sum_n g_n \sum_q \frac{\hbar}{2M \omega_n(2k_F+q)} \left[ b_n(2k_F+q) \right]^{1/2} \times \left[ b_n(2k_F+q) + b_n^+(2k_F+q) \right] e^{i q x} .
\]

For each phonon mode, we define a dimensionless electron-phonon coupling constant \( \lambda_n = N(0) g_n^2 / \omega_n(2k_F) \), where \( N(0) \) is the density of states at the Fermi level and \( g_n \) the linear coupling constant specifying the interaction of the conduction electron with the \( n \)th phonon band.

Rice has performed an analysis of this model in the rigid lattice (or mean-field) approximation, which neglects the effects of quantum and thermal lattice motions.\textsuperscript{22} Below a common transition temperature, all the phonon modes soften and each operator \( b_{n2k_F} \) has a nonzero expectation value \( \langle b_{n2k_F} \rangle = a_n \exp(i \phi_n) \), where \( \phi_n \) is the phase associated with the CDW corresponding to the \( n \)th phonon mode. The energy gap \( \Delta \) of the system is \( 2e_F \exp(-1/\lambda') \), where \( \lambda' = \sum_n \lambda_n \) is the total electron-phonon coupling. The key point of Rice's model is that in addition to the single-electron contribution to the optical conductivity, collective contributions associated with oscillations in the phases \( \phi_n \) of the combined lattice and charge distributions about their zero equilibrium values will also be observed. Such oscillations involve a bodily displacement of an appropriate component of the condensed charge and are, therefore, optically active along the chain direction. The situation closely parallels that found by Lee, Rice, and Anderson for the collective mode of an acoustic-phonon-stabilized Fröhlich CDW state.\textsuperscript{10}

In other words, the electron-phonon coupling interaction causes those phonons (which would be inactive without electron-phonon coupling, and which are essentially due to Raman-active intramolecular modes), renormalized as collective modes, to become FIR active in the chain direction. These latter excitations are called "phase phonons." A quantitative comparison of Rice's theory with experiment makes it possible to extract values for \( \lambda_n \) and \( \omega_n \) for each of the phonon modes.

We now assume that \( \omega_n \ll \Delta \) and that the phonon dispersion near \( 2k_F \) is negligible (i.e., the temperature is not close to the transition temperature where there is anomalous phonon dispersion near \( 2k_F \)). It is then possible to repeat an analysis similar to that in Ref. 23 and replace the quantum field \( \Delta(x) \), given by Eq. (4), with a random static potential \( \Delta_0 + \xi(x) \), where \( \Delta_0 = \langle \Delta(x) \rangle \) is the magnitude of the order parameter and \( \xi(x) \) has Gaussian correlations,

\[
\langle \xi(x) \rangle = 0, \quad \langle \xi(x) \xi(x') \rangle = \gamma \delta(x-x') .
\]

It turns out that the dimensionless disorder parameter \( \eta = \gamma / \pi \omega_n \Delta \) is then given by

\[
\eta(T) = \eta_0 + \sum_n \frac{\lambda_n \pi \omega_n}{2 \Delta} \coth \left( \frac{\omega_n}{2T} \right), \quad \omega_n \ll 2 \Delta ,
\]

where \( \eta_0 \) is the temperature-dependent contribution of Eq. (2). Therefore, Eq. (6) represents the generalization of Eq. (2) from a single- to a multiple-phonon model.

**B. Comparison with the experimental results**

One of the most striking outcomes of the theoretical model\textsuperscript{21} is the universal scaling form of the strong subgap conductivity. Figure 2 shows the scaling of the experimental optical conductivity (using the data shown in Fig. 1 of Refs. 11 and 12) for the pure blue bronze and for \( (\text{TaSe}_2)_2\text{I} \) at several temperatures. As suggested theoretically, we recover also experimentally the universal behavior. Similar results can be found by rescaling the optical conductivity of other one-dimensional systems as well, like, e.g., \text{KCP}\textsuperscript{3}, \text{Pt}-halogen chain\textsuperscript{4} (\text{CH}_2)\textsuperscript{4}.

For comparison, Figs. 1 and 2 also present the model calculation with total disorder parameter \( \eta = 0.66 \) (\( T = 0 \) K) and \( \eta = 1.0 \) (\( T = 140 \) K). Moreover, Fig. 1 gives clear evidence of the differences between the static mean-field theory result of Lee, Rice, and Anderson (\( \eta = 0 \)) (Ref. 10) and the approach followed in this paper (\( \eta = 0.66 \)).\textsuperscript{21} The deviations of the experimental optical conductivity from the theoretical calculation above the gap (Fig. 1 and 2) are due to the electronic interband transitions, which are not considered in the model of Ref. 21. Bandstructure calculations show that \( K\alpha_3\text{MoO}_3 \) has several bands within 0.1 eV of the conduction band, while the
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FIG. 2. Scaling plot of the experimental optical conductivity for (a) $K_{0.3}MoO_3$ and (b) (TaSe$_4$)$_2$I in the midinfrared spectral range. A calculation with the model of Ref. 21 is also presented for the disorder parameter $\eta=0.66$ ($T=0$ K) and $\eta=1.0$ ($T=140$ K).

upper bands of (TaSe$_4$)$_2$I are at much higher energies. The thermal excitations due to the electronic interband transitions in the upper bands will affect the temperature dependence of the gap absorption. This is because the CDW wave vector shifts due to thermal population of the upper band. Therefore, we expect that the gap absorption is more temperature dependent in blue bronze than in (TaSe$_4$)$_2$I. Indeed, this is in agreement with the experimental findings (Figs. 1 and 2).

The results for (TaSe$_4$)$_2$I (Fig. 2) follow the general trend set by the theoretical prediction, which leads to the universal scaling form of $\sigma_2(\omega)$. However, there are some remarkable deviations from the theory of Refs. 2 and 21, which are much more important than those for $K_{0.3}MoO_3$. In particular, it looks like $\sigma_2(\omega)-\omega$ for frequencies $\omega<\Delta$. It is not clear what the origin of this discrepancy is. Nonetheless, we should remark that the linear frequency dependence of $\sigma_2(\omega)$ for $\omega<\Delta$ was calculated in a pseudogap model by Sadovskii.

From the evaluation of the data leading to the universal scaling form presented above, it is then possible to extract the temperature dependence of the disorder parameter. This is obtained by choosing $\eta$ such that the computed $\Gamma(\eta)/\omega_{peak}(\eta)$ is equal to $\Gamma(T)/\omega_{peak}(T)$. The experimental values of $\eta(T)$ for the two compounds are shown in Fig. 3.

As already pointed out in the theoretical section, a comparison of the experimental values of $\eta(T)$ with the theoretical prediction of Eq. (2) using reasonable parameters would not be realistic or very satisfactory, unless one assumes an additional and very large extrinsic disorder contribution ($\eta_s$), for which it would be difficult to find a plausible justification. However, it is possible to take into account the effect of the so-called phasons within the same theoretical framework, which leads, as shown in Sec. III A, to Eq. (6) for $\eta(T)$.

As extensively described in our previous work, the parameters $\lambda_n$ for the dimensionless electron-phonon coupling constants, and $\omega_n$ for the frequencies and $\Gamma_n$ for the dispersions of the bare phonons, which are coupled to the electronic density fluctuation of wave vector $q$, were obtained by a fit of the optical conductivity in the CDW ground state, using the phason model of Rice. The full set of the fit parameters for (TaSe$_4$)$_2$I is summarized in Table I. We refer to Table I of Ref. 11 for the parameters used for $K_{0.3}MoO_3$. For instance, Fig. 9 of Ref. 14 shows

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<th>$\omega_n$ (cm$^{-1}$)</th>
<th>$\lambda_n$</th>
<th>$\Gamma_n$ (cm$^{-1}$)</th>
<th>$\omega_n$ (cm$^{-1}$)</th>
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a comparison between the experimental data and the corresponding theoretical calculation in the case of K$_{0.3}$MoO$_3$. The reproducibility of the data is rather satisfactory and similar results are also obtained for (TaSe$_4$)$_2$I. For the blue bronze it is possible to recognize the importance of the phonon modes activity by comparing its excitation spectrum with that of the so-called red-bronze (K$_{0.3}$MoO$_3$). The latter one is isostuctural to K$_{0.3}$MoO$_3$, but does not have a CDW ground state. The red bronze has very little and weak phonons in FIR, making clear that the huge FIR activity in K$_{0.3}$MoO$_3$ must have origin in the phonon modes coupling to the CDW condensate.

In our phenomenological calculation, we have assumed that the sharp absorptions, found in the FIR optical conductivity, are somehow effective for the phase-phonon mechanism.\textsuperscript{14} However, from the theoretical point of view, only the symmetric (Raman active) intramolecular phonon modes are suitable for the coupling to the CDW condensate.\textsuperscript{22} In the Raman spectra of K$_{0.3}$MoO$_3$ for the experimental configuration, where the symmetric $\Delta_x$ modes are allowed by the selection rules, almost 70 sharp excitations are found.\textsuperscript{26}

Even though the large number of symmetric intramolecular phonon modes in both compounds\textsuperscript{26,27} prevents the establishment of a reliable one-to-one correspondence between the bare Raman modes and the FIR phonons, we model the FIR activity in both compounds using a limited set of modes, which are allowed to interact with the CDW condensate (see, e.g., Table I). The scope of this analysis is mainly to show the importance of the effect, also with respect to the thermal lattice fluctuation, rather than to numerically produce all the details of the excitation spectrum.

Using our fits for the phase-phonon modes (Table I and Ref. 11) and the values reported in Table II for $\omega_A$, $\eta_x$, $\lambda$, and $2\Delta$, a good fit to the experimental values of $\eta(T)$ is achieved (Fig. 3). The set of parameters also satisfies several experimental constraints. For instance, $2\Delta$ and $\hbar\omega_A$ are in fair agreement with the gap evaluated from the activated behavior of the dc resistivity and with the measured amplitude mode frequency (see values in brackets of Table II).\textsuperscript{13,14,27-29} Furthermore, from the expression $\omega_A = \sqrt{\lambda_{\text{tot}}\omega_{2k_F}}$ and $\lambda_{\text{tot}} = \lambda + \sum_n \lambda_n$, we can calculate the unrenormalized phonon frequency $\omega_{2k_F}$ responsible for the Kohn anomaly. The values of $\omega_{2k_F}$ for both compounds are summarized in Table II and for K$_{0.3}$MoO$_3$ this quantity is comparable to the value of $\hbar\omega_{2k_F} = 5$ meV, arrived at with neutron-scattering investigation.\textsuperscript{26}

Moreover, it has been shown by Kim, McKenzie, and Wilkins\textsuperscript{21} that the peak of the optical conductivity is the product of $\sigma_0 = e^2 v_F / 4 \Delta$ and the dimensionless ratio $\sigma_{\text{peak}}(\eta) / \sigma_0$ determined from fitting the experimental curves. Accordingly the peak conductivity yields $\sigma_0$ and hence the Fermi velocity $v_F$, since the gap $2\Delta$ and cross-sectional area $A$ are known (Table II). At low temperatures, we can assume $\eta = 0.7$ and $\sigma_{\text{peak}}(\omega) / \sigma_0 = 0.35$ for both compounds.\textsuperscript{21} Therefore, using the experimental values of $\sigma_{\text{peak}}(\omega)$, we obtain values for $v_F$ given in Table II. For (TaSe$_4$)$_2$I, our value of $v_F$ is in fair agreement with other evaluations arrived at by using different experimental results, like, e.g., previous optical results\textsuperscript{18} and neutron-scattering investigation,\textsuperscript{20} but is about a factor of ten smaller than the value derived from the magnetic susceptibility\textsuperscript{13} (see Table II). The value of $v_F$ for the blue bronze is also a factor ten smaller than the evaluation of Poug"{e}r\textsuperscript{28} from Wangbo's band structure calculation,\textsuperscript{20} or from previous optical\textsuperscript{15} and magnetic susceptibility investigations.\textsuperscript{13} Similar evaluations for other compounds [like, e.g., NbSe$_3$ (Ref. 13) and KCP (Ref. 21)] are at variance as well, resulting in deviations of a factor between two and five with respect to the evaluations from band-structure calculation. It is striking that for all the compounds the optical $v_F$ is always less, and often substantially less, then the band-structure estimation of $v_F$. This tends to suggest that correlation effects play an important

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<th>TABLE II. Parameters used for the calculation of $\eta(T)$: the single-particle gap $2\Delta$, the electron-phonon coupling constant $\lambda$, the amplitude mode $\omega_A$, the extrinsic disorder $\eta_x$, the total electron-phonon coupling $\lambda_{\text{tot}}$, the cross-sectional area $A$, the peak value of $\sigma_{\text{peak}}(\omega)$, the Fermi velocity $v_F$, and the phonon frequency of the Kohn anomaly $\omega_{2k_F}$. In brackets, the experimental values where known.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$2\Delta$ (eV)</td>
</tr>
<tr>
<td>----------------</td>
</tr>
<tr>
<td>K$_{0.3}$MoO$_3$</td>
</tr>
<tr>
<td>(0.12)$^a$</td>
</tr>
<tr>
<td>(TaSe$_4$)$_2$I</td>
</tr>
<tr>
<td>(0.37)$^a$</td>
</tr>
</tbody>
</table>

$^a$References 1, 13, and 14.
$^b$References 13, 14, and 29.
$^c$Reference 28.
$^d$Reference 13.
$^e$References 13 and 15.
$^f$References 14 and 27.
$^g$References 13 and 18.
role. It might be that correlation effects increase the effective mass so that the optical sum rule will be less than expected. This is an important issue that will be explored in detail in a future work.

Moreover, we must observe that $\lambda_{\text{tot}}=2.22$ for (TaSe$_4$)$_2$I is fairly large, implying a kind of strong coupling. This also means that the weak-coupling model used in the theory of Ref. 2 is somehow invalidated. We have previously remarked that for (TaSe$_4$)$_2$I, the model calculation only gives the general trend, but does not reproduce the optical conductivity in the same satisfactory way as in K$_{0.3}$MoO$_3$. It remains to be seen whether this deviation from the calculated universal behavior of $\sigma_1(\omega)$ is somehow related to the strong-coupling envisaged here.

From the above analysis, it is nonetheless clear that the phasons add an important contribution to the temperature dependence of the disorder parameter. With this contribution the extrinsic disorder parameter $\eta_\pi$ can be considerably reduced compared to the anomalously large value, which should have been used by only applying Eq. (2). Indeed, the values of $\eta_\pi$ are now comparable to what has been found in other one-dimensional compounds. However, the calculation made use of phason modes for which the condition $\alpha_\pi \ll \Delta$ is not fully satisfied. Further theoretical work is needed in order to overcome this difficulty. Moreover, it is still to be established whether all the phonons involved in the phason mechanism really soften at the phase transition, as assumed in Rice’s model, and thus contribute to the thermal lattice fluctuation effects.

In addition, the model of lattice fluctuations in terms of Gaussian disorder is only valid for $T < T_{\text{CDW}}$, since it neglects the dispersion of the phonon near the wave vector $2k_F$. It is a challenge to provide a single theoretical model that can describe the crossover from the broad subgap tail studied here to the pseudogap feature seen above $T_{\text{CDW}}$.

IV. CONCLUSION

The optical conductivity on K$_{0.3}$MoO$_3$ and (TaSe$_4$)$_2$I reported here, gives convincing evidences that thermal lattice fluctuations have a significant effect on the dynamical conductivity and more generally the electronic density of states in the CDW ground state (i.e., below $T_{\text{CDW}}$). We found that the excitation associated with the CDW single-particle gap is progressively broadened with increasing temperature, where the broadening is larger than $k_B T$. The striking feature is the universal scaling form of the subgap conductivity, from which we have evaluated the temperature dependence of the disorder parameter. Moreover, we found that the temperature dependence of this latter quantity is strongly influenced by the phase-phonon modes. Their contribution is essential and plays a remarkable role in the thermal lattice fluctuation effects.

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The dependence of \( \Gamma / \omega_{\text{peak}} \) on the disorder parameter \( \eta \) can be fitted to \( \Gamma(\eta) / \omega_{\text{peak}}(\eta) = \eta^{0.62}(0.414 + 0.077\eta) \) (Ref. 21).

S. Donovan (private communication).