

Fluctuations and order in a one-dimensional system. A spectroscopical study of the Peierls transition in $\text{K}_2\text{Pt}(\text{CN})_4\text{Br}_{0.3} \cdot 3(\text{H}_2\text{O})$

P. Brüesch, S. Strässler, and H. R. Zeller

Brown Boveri Research Center, CH 5401 Baden, Switzerland

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Using optical spectroscopy in the energy range 1 meV–6 eV as a tool we have studied the Peierls transition in $\text{K}_2\text{Pt}(\text{CN})_4\text{Br}_{0.3} \cdot 3(\text{H}_2\text{O})$. We find a pseudogap at 0.2 eV at room temperature which transforms into a real gap as $T \rightarrow 0$ without significantly changing the gap energy. From this we infer that although no long-range order exists at room temperature the rms order parameter has already reached its zero temperature value. At $T \approx 0$ we find a strong structure in $\sigma(\omega)$ at $\omega \approx 15 \text{ cm}^{-1}$ which is assigned to an oscillation of the pinned charge-density wave (CDW). A simple phenomenological model allows us to estimate the contribution of the thermally unpinned CDW to the dc conductivity at $T > 0$. We conclude that the contribution is 50% or less of the total conductivity.

I. INTRODUCTION

Diffuse-x-ray and inelastic-neutron-scattering studies have established that $\text{K}_2[\text{Pt}(\text{CN})_4]\text{Br}_{0.3} \cdot 3(\text{H}_2\text{O})$ (KCP) undergoes a Peierls transition.¹⁻⁶ Within a simple band-structure model the metallic state of a one-dimensional (1-D) metal is unstable and mean-field theory predicts a second-order soft-mode phase transition into an insulating state.⁷⁻¹⁰

It is known that in a strictly 1-D system, the critical region is of order T_c ; in other words, there is no long-range order at $T \neq 0$, and at $0 < T \leq T_c$ the physical properties are dominated by fluctuations.^{11,12} In a real system, however, some residual coupling to the three-dimensional (3-D) world invariably exists, making a phase transition at finite temperature feasible.¹³⁻¹⁶ We thus expect an extremely interesting interplay between fluctuation and order phenomena. Recently, a large number of theoretical papers¹⁷⁻²³ have addressed themselves to this question and have resulted in intriguing predictions. No corresponding experimental investigations have been published, however.

In this paper, we will use optical spectroscopy as a tool to study both the parameters of the ordered state and fluctuation phenomena. A simple phenomenological model will be presented which offers intuitive insight with a minimum of mathematical formalism and which provides a unified picture of the experimental observations.

The electronic instability of the system manifests itself in the formation of a charge-density wave (CDW), which in the case of KCP has a period of 6.7 Pt-Pt distances.¹⁻⁶ Fröhlich has argued that in an ideal system, the CDW is not fixed in space. Under the influence of an electric field, the CDW travels and within Fröhlich's model is able to carry a supercurrent.⁸ As dis-

cussed by Lee, Rice, and Anderson,¹⁷ in a real system the translational degree of freedom is broken, and at $T=0$ the CDW is pinned by impurities, disorder, or the periodic potential of the lattice itself. (3-D coupling by itself does not lead to pinning but it greatly enhances the susceptibility of the system to other pinning mechanisms.)

In this paper, we will deal with the following questions: (i) Formations and properties of the CDW defined by its rms amplitude $\langle Q^2 \rangle^{1/2}$ and its relation to the optical conductivity, mobility, and density of states. These are rather intrinsic properties and may be related to theoretical results. (ii) Pinning of the CDW, electro-dynamics of the pinned CDW, pinning-unpinning fluctuations. An optical study of the oscillations of the pinned CDW yields important information concerning intrinsic properties of the CDW. The pinning-unpinning fluctuations which may lead to para-conductivity¹⁷⁻²² in the vicinity of the pinning temperature are determined by the nonideality of the system and correspondingly, will be discussed in the framework of a physically transparent phenomenological theory.

The over-all properties of the CDW can be studied in the infrared at $\sim 10^3 \text{ cm}^{-1}$, and the elementary excitations of the pinned CDW at about $10\text{--}100 \text{ cm}^{-1}$. Accordingly, we have performed an infrared (ir) and far infrared (fir) study of the optical properties of KCP.

II. EXPERIMENTS AND RESULTS

The experimental procedure for the reflectivity measurements of KCP are described in detail in Refs. 24–27. All measurements are very tedious because extreme care has to be taken to avoid water loss.²⁸ The only satisfactory way we found to cool the sample without loss of water and with-

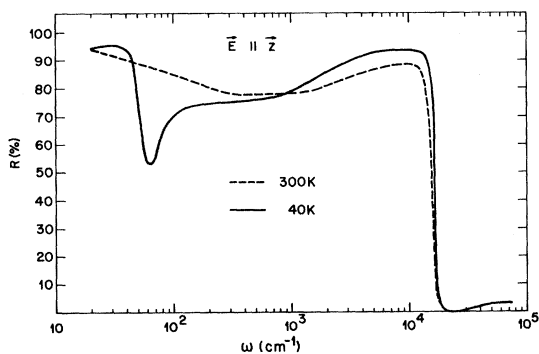


FIG. 1. Far-infrared to uv spectrum of KCP for light polarized parallel to the conducting axis at 300 K (dashed line) and at 40 K (solid line). The most drastic change in the two spectra is the strong structure in the far-infrared at 40 K. This structure is assigned to the oscillating charge-density wave (pinned Fröhlich mode) induced by the Peierls distortion.

out condensing moisture on the sample or the reference mirror was to cool it very slowly in an atmosphere of controlled humidity. At or below 200 K, the crystal could be kept for arbitrary times in dry He gas without any noticeable loss of water. In addition to the technique described in Ref. 27, the more recent measurements of the temperature dependence of the reflectivity in the fir were made by using 6- μ m-thick polyethylene foils as window material.²⁹ All data presented are given on virgin samples measured during a cool-down cycle. This was necessary because the optical behavior depends to some extent on the thermal history of the crystals.²⁸

Figure 1 shows the $\vec{E} \parallel \vec{z}$ reflectivity of KCP in the whole spectral range from the fir to the uv at 300 and 40 K. It should be pointed out that the measurements in the ir and fir were carried out on different crystals on different spectrometers using different measurement techniques (direct in

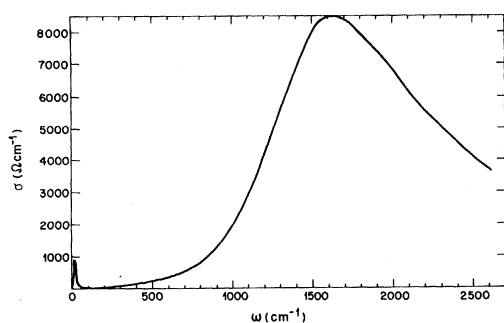


FIG. 2. Optical conductivity of KCP as determined by a Kramers-Kronig analysis of the reflectivity at 40 K. The very strong resonance at 1600 cm^{-1} is due to transitions across the Peierls gap while the small and sharp peak centered around 15 cm^{-1} is due to the pinned Fröhlich mode.

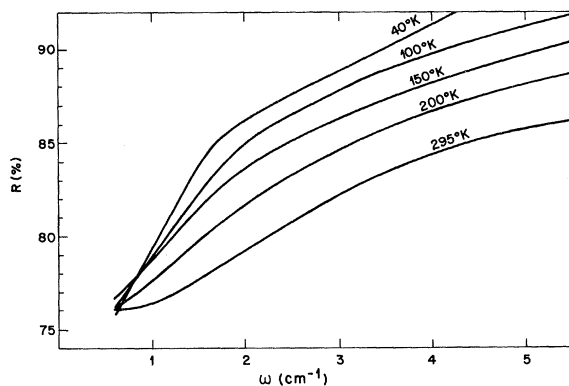


FIG. 3. Temperature dependence of the infrared reflectivity of KCP for light polarized parallel to the conducting axis.

the ir case and Fourier transforms for fir). This introduces a matching problem in the overlap region with discrepancies of up to 3% in the reflectivity. In order to avoid spurious results in the Kramers-Kronig analysis of R due to the overlap problem and due to the low-frequency extrapolation in the fir, we have treated the structure in the ir and in the fir separately. The fir data are analyzed in terms of a generalized oscillator fit, which will be discussed in detail in Sec. III. Subsequently, the ir data were extrapolated to small frequencies using the fir results as a guide and a Kramers-Kronig analysis was performed. Kramers-Kronig analysis and an oscillator fit yielded nearly identical results in the ir region, which gives confidence in the validity of the Kramers-Kronig analysis. Details of the high frequency extrapolation and the visible and uv part of the spectrum are given in Ref. 28.

The result of such an analysis is shown in Fig. 2. There are two peaks in the optical conductivity $\sigma(\omega)$. The small peak which is centered around 15 cm^{-1} is due to the strong structure observed in the fir (Fig. 1) and is assigned to the pinned Fröhlich mode.^{27,29} The very strong peak centered around 1600 cm^{-1} is due to the decrease of the reflectivity in the ir and is due to transitions across the Peierls gap.^{28,30}

The temperature dependence of the reflectivity in the ir is shown in Fig. 3. (These measurements have been performed on the same crystal in a single cool-down cycle). The decrease in R between 1000 and 2000 cm^{-1} becomes more pronounced with decreasing temperature. Figure 4 shows the optical conductivity $\sigma(\omega)$ in the ir as obtained by means of a Kramers-Kronig analysis. With decreasing temperature, the peak in $\sigma(\omega)$ at 1600 cm^{-1} considerably sharpens while its position and oscillator strength remain constant.

Figure 5 gives the temperature dependence of the

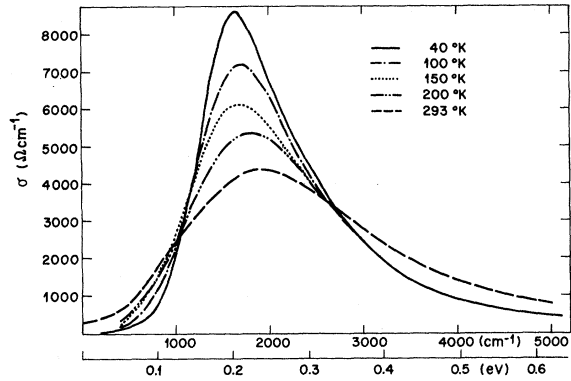


FIG. 4. Temperature dependence of the optical conductivity σ in the infrared. The resonance at high temperatures originates from a strong mobility pseudogap which transforms into the real Peierls energy gap as $T \rightarrow 0$ without significantly changing the gap energy. The resonance at 40 K is due to transitions across the Peierls gap.

structure observed in the fir. At 4.2 K, a very strong structure is observed which decreases in intensity with increasing temperature and disappears around 200 K. We have performed an oscillator fit with a frequency-dependent damping to the reflectivity data shown in Fig. 5. The details of the fit will be discussed in Sec. III. Figure 6 shows the resulting optical conductivity $\sigma(\omega)$. At 4.2 K, a sharp peak at about 15 cm^{-1} is ob-

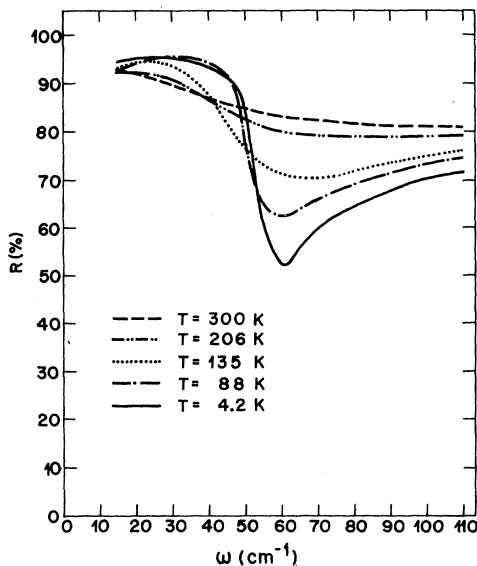


FIG. 5. Temperature dependence of the reflectivity observed in the far-infrared for light polarized parallel to the conducting axis. A strong structure with a very high oscillator strength is observed at 4.2 K which is assigned to the pinned Fröhlich mode. The increasing damping smears out the structure with increasing temperature.

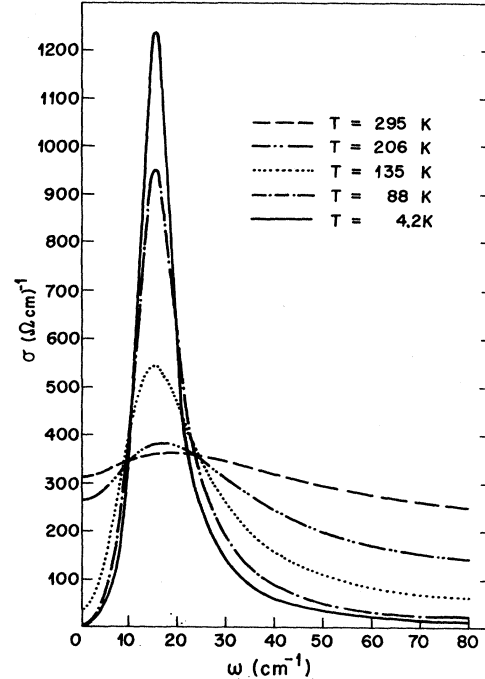


FIG. 6. Temperature dependence of the optical conductivity σ in the far infrared. The dc conductivity for $T \geq 135 \text{ K}$ is mainly due to the single-particle contribution which has been added to the oscillator term of the pinned Fröhlich mode.

served. With increasing temperature, the peak gets broader and eventually becomes overdamped.

III. DISCUSSION

The first paragraph of this section deals with the ir data which yield information about the fluctuating Peierls gap. In the second paragraph, the fir data are discussed in terms of vibrational and translational excitations of the CDW, and it is shown that alternative models can be ruled out. Finally, the importance of the Br^- potentials with respect to CDW formation is evaluated in the light of the new experimental findings.

The optical-conductivity data of Fig. 4 indicate a constant temperature-independent Peierls gap of $2\Delta \approx 0.2 \text{ eV}$, corresponding to a mean-field transition temperature T_p^{MF} of $(600-700)^\circ\text{C}$. At all experimentally accessible temperatures ($T \leq 300 \text{ K}$), one is sufficiently below T_p^{MF} for $2\Delta(T) \approx 0.2\Delta(0)$ to hold. On the other hand, it is known that in a strictly 1-D system no long-range order exists for $T > 0$.^{11,12} Accordingly, instead of interpreting the optical results in terms of an order parameter $\langle Q \rangle$ (defined as a Pt distortion coordinate), it is more appropriate to introduce the rms order parameter $\langle Q^2 \rangle^{1/2}$. The optically observed gap is now interpreted as a pseudogap, i. e., the finite correlation length in Q rounds off

the singularities at the gap boundaries and introduces a finite density of states in the gap.^{12,16} The experimental fact that the rms amplitude $\langle Q^2 \rangle^{1/2}$ of the CDW is temperature independent greatly simplifies the physical interpretation which, in principle, thus reduces to a discussion of the temperature dependence of the correlation length and of the phase of Q .

From NMR data³¹ it is known that the density of states $N(E_F)$ at 295 K is only slightly depressed from its free-electron value. At the same temperature, we observe a strong structure in $\sigma(\omega)$ which, in view of the NMR data, has to be explained by a strong mobility pseudogap which apparently shows up before the density-of-states pseudogap is formed. Although we were not able to give a rigorous proof, it seems intuitively clear that the optical conductivity is more strongly affected by fluctuation effects than the density of states for dimensionality reasons. This has to do with the strong relation between scattering and gap formation in a 1-D system.

The remainder of this section deals with the structure in the fir and their relation to the elementary excitations of the CDW.

At $T = 0$ the response of the pinned CDW can be discussed phenomenologically in terms of an oscillator model,

$$\epsilon = \epsilon_\infty + \epsilon_\infty (\omega_p^2 - \omega_T^2) / (\omega_T^2 - \omega^2 - i\Gamma\omega) \quad (3.1)$$

where ϵ_∞ is the high-frequency dielectric constant due to transitions across Peierls gap, ω_T the oscillator frequency defined by the pinning force constant, and Γ a damping term. ω_p , the plasma frequency of the CDW, is related to the effective mass m^* of the collective excitations by $\omega_p^2 = 4\pi N e^2 / m^* \epsilon_\infty$, where N defines the density of condensed electrons.

It turns out that a fit to the 4.2 K data of Fig. 5 is rather insensitive to the value of ω_T . We have thus determined ω_T by using the known value of the static dielectric constant, $\epsilon_0 \approx 3000$ at 4.2 K (Ref. 32) in the fit. By this procedure, we obtain $\omega_p = 58 \text{ cm}^{-1}$, $\omega_T = 15 \text{ cm}^{-1}$, $\epsilon_{ib} = 190$, and $\Gamma = 7.5 \text{ cm}^{-1}$ at 4.2 K.

Assuming all electrons to be condensed, an effective mass $m^* = 980$ is deduced.

Mean-field theory allows determination of the electron phonon coupling in two independent ways, $2\Delta = 16E_F e^{-1/\lambda}$ (Ref. 8, 19) with $E_F = 3.25 \text{ eV}$ yields $\lambda \sim 0.2$. Independently, λ is given by^{17,21}

$$m^*/m = 1 + (4\Delta^2 / \lambda \omega_0^2).$$

With $m^*/m \sim 10^3$, and ω_0 the unrenormalized phonon frequency $\omega_0 \sim 10 \text{ meV}$, we obtain $\lambda \sim 0.4$. The discrepancy between the two values for λ is partly due to the assumption of an energy-independent

electron-phonon interaction which, for instance, in the special case of a tight-binding band can be explicitly shown to be incorrect.³³

The value of $\lambda = 0.2$ obtained from the energy gap is very model dependent and strongly affected by such effects. In the determination of m^* we have neglected local-field effects. For an incommensurate CDW, there are no local-field effects for symmetry reasons. Also, for other modes polarized in the strand direction, local-field effects are very weak due to the fact that the conduction electrons are nearly free.^{25,34}

Before going on with the discussion, it is appropriate to point out that no other interpretation of the experimentally observed structure than that in terms of a pinned CDW is possible. The phenomenological Eq. (3.1) not only describes bound electronic systems but also lattice vibrations and coupled electron-phonon systems such as the Peierls CDW. From the effective mass $m^* \sim 10^3$, all lattice vibrations (except possibly proton motion) and also purely electronic excitations can be excluded. Protons are present in the system but at low symmetry sites.^{35,36} If they were responsible for the structure, they would exhibit a component in the transverse direction, which is not observed. Furthermore, several studies on deuterated samples show no change in the relevant physical properties. In particular, ω_p of the CDW and hence m^* remains the same.³⁷ Thus the effective mass or oscillator strength can only be caused by a coupled electron-phonon system, which in our case is the CDW.

Except for the oscillation around its pinned position, the CDW can also fluctuate into an unpinned state and carry a dc current. There has been a great deal of discussion as to whether this mechanism can lead to a large enhancement of the dc conductivity. Most authors calculated the formation of the CDW with the result $\sigma_{dc}^c \sim \langle Q^2 \rangle$, where σ_{dc}^c is the collective-mode contribution.^{18,19,21} In our case, $\langle Q^2 \rangle$ has already reached saturation, and the whole problem reduces to a discussion of the scattering rate.²² By definition, pinning implies a divergent scattering rate. If pinning occurs by disorder, etc., then long-range order is never established, T_p is not exactly defined, and around T_p (pinning temperature) the CDW can fluctuate between a pinned and an unpinned state.

In the following we give a generalization of Eq. (3.1) which allows us to include such fluctuations in a unified and conceptually simple picture. We start from a simple Drude-type equation and omit any pinning effects,

$$\epsilon = \epsilon_\infty + \epsilon_\infty [\omega_p^2 / (-\omega^2 - i\Gamma'\omega)] \quad (3.2)$$

Next we notice that fluctuations into the pinned

TABLE I. Results of the oscillator fit to the far-infrared reflectivity data (see text).

T (K)	ϵ_∞	ω_p (cm $^{-1}$)	δ (cm $^{-1}$)	Γ_0 (cm $^{-1}$)	γ (cm $^{-1}$)	σ_{sp} (Ω cm) $^{-1}$	σ_{obs} (Ω cm) $^{-1}$
4.2	190	58	15	8	0	0	0
88	225	55	15	11	0	1	1
135	240	53	15	20	0.1	30	30
206	320	46	15	30	7	100	260
295	350	42	15	50	7	200	300

state strongly enhance the low-frequency scattering rate. This is accounted for by introducing a frequency-dependent damping, which in its simplest possible form is given by

$$\Gamma' = \Gamma_0 + \delta^2/(\gamma - i\omega). \quad (3.3)$$

γ is a characteristic frequency which defines the transition from high-scattering to low-scattering rate and is related to the jump frequency of the CDW. Γ_0 is the scattering rate in the absence of pinning, and δ measures the effectiveness of pinning. The imaginary part of Γ is required by causality. The specific form of (3.3) implies that the unpinned CDW ($\delta = 0$) and the pinned CDW ($\gamma = 0$) have the same Γ . Within the framework of our model, pinning fluctuations only affect the low-frequency scattering rate but do not change the intrinsic properties of the CDW.

Inserting (3.3) in (3.2) we obtain

$$\epsilon = \epsilon_\infty + \epsilon_\infty \omega_p^2 / \left[\left(\frac{\omega^2}{\omega^2 + \gamma^2} \right) \delta^2 - \omega^2 - i\omega \left(\Gamma_0 + \frac{\delta^2 \gamma}{\omega^2 + \gamma^2} \right) \right] \quad (3.4)$$

Comparison with (3.1) shows

$$\omega_T^2 = [\omega^2 / (\delta^2 + \gamma^2)] \delta^2, \\ \Gamma = \Gamma_0 + \delta^2 \gamma / (\omega^2 + \gamma^2).$$

Introduction of a frequency-dependent scattering rate Γ automatically also leads to a frequency-dependent restoring force ω_T . At $\omega \ll \gamma$, $\omega_T \approx 0$, the CDW can propagate but with a high damping due to fluctuations into the pinned state, and σ_{dc} becomes

$$\sigma_{dc} = \frac{1}{4\pi} \frac{\epsilon_\infty \omega_p^2}{\Gamma_0 + \delta^2/\gamma}$$

At high frequencies, $\omega \gg \gamma$, $\omega_T \sim \delta$. On this time scale, the CDW oscillates around its pinned position.

In order to get meaningful information from a fit to experimental results, we have to reduce the number of parameters. At $T \ll T_p^{MF}$ it is reasonable to assume that ω_p^2 is nearly independent of temperature. If pinning occurs by random Coulomb potentials provided by disorder, etc., then δ^2 is also temperature independent. ϵ_∞ is obtained from the asymptotic value of R at high frequencies,

and we are thus left with two strongly temperature-dependent parameters Γ_0 and γ .

An attempt to fit the optical data including the measured dc conductivity by this procedure completely failed at high temperature. The reason is that the values of Γ_0 and γ required to fit the optical data produce too small a σ_{dc} . In other words, the model cannot account for the observed dc conductivity. This can be corrected by adding a term to (3.4) which describes the single-particle conductivity and which for $\hbar\omega \ll 2\Delta$ can be written as

$$\epsilon = 4\pi i \sigma_{sp} / \omega.$$

The results of such fits are summarized in Table I, and the over-all quality can be judged from Figs. 7–8. For $\sigma_{dc} \neq 0$, the fits are not too sensitive to changes in the ratio of single-particle conductivity to total conductivity. However, σ_{sp} has to be in the range of (50–100)% of σ_{dc} , i. e., the contribution of the collective mode to σ_{dc} is 50% or less.

The *Ansatz* 3.3 allows us to discuss the pinned ($\gamma = 0$) and unpinned ($\delta = 0$) CDW in a unified and conceptually simple picture. The parameters ϵ_∞ , ω_p , and Γ_0 can in principle be calculated by theory; pinning effects, which in our case are expected to be caused by disorder, impurities, and 3-D coupling, are treated by the phenomenological parameters γ and δ . The pinning temperature T_p above which γ is different from zero can be related with a self-consistent phonon calculation

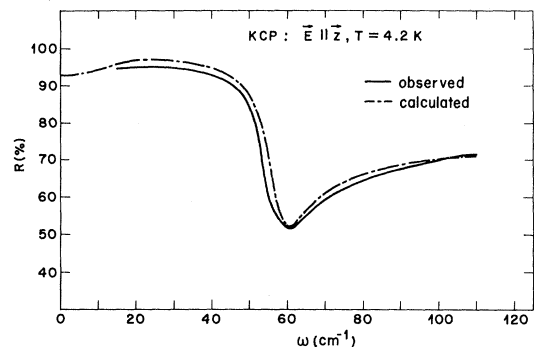


FIG. 7. Oscillator fit to the observed reflectivity in the far infrared at 4.2 K.

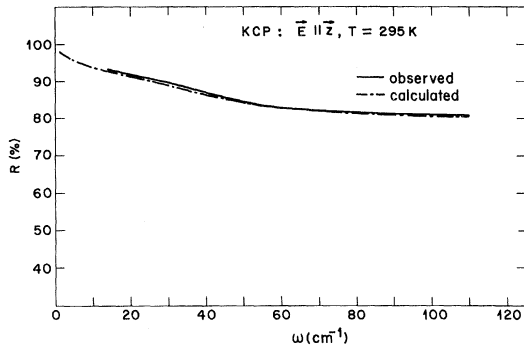


FIG. 8. Oscillator fit to the observed reflectivity in the far infrared at 295 K.

to the pinning frequency.³⁸ The result is

$$T_p = 2\sqrt{\lambda}/\pi e (\omega_0\omega_F/k_B\Delta) m^*/m).$$

With the values of the parameters as defined above, we obtain $T_p \approx 200$ K, in good agreement with the experimental value.

Before concluding this section, we will show that based on the above information, the role of the Br^- potentials on the Pt strands can be evaluated. It has been pointed out that the Br^- potentials, even if the Br are disordered, have a strong Fourier component with wave vector $2k_F$ along the Pt strands.^{1,36,39} The question thus arises whether the pseudogap in the Pt conduction band is really due to interaction with the LA phonons as discussed in this paper or to the influence of the Br^- potential. The Br^- distribution could be determined trivially by crystallography, or if the Br are mobile the coupling between Br^- and Pt could lead to a generalized Peierls distortion with the Br^- occupation number as generalized distortion coordinate. Since the Br sites are incommensurate with the CDW and the observed correlation length of the CDW is very large even at room temperature, such possibilities appear unlikely. If the Br^- were dominant, then the CDW would be rigorously pinned to the Br^- positions, the phase transition would lose its 1-D character, and the effect of fluctuations would be drastically reduced.

There is a very simple relation between the effective mass of the CDW and the distortion of the Pt ions. This relation is based exclusively on kinematic considerations. For a phase oscil-

lation of the CDW, the kinetic energy consists of two parts

$$E_{\text{kin}} = \frac{1}{2} Nm_e V^2 + \frac{1}{2} N_i M \frac{1}{2} (\dot{\phi} u)^2 = \frac{1}{2} Nm^* V^2,$$

where u is defined by the lattice distortion $\delta R = u \cos[(2\pi/\lambda)z + \phi]$. The first part is the kinetic energy of the electrons, and the second term the kinetic energy of the ions. The right-hand side defines m^* . With $N_i = 1/c$ ($c = \text{Pt-Pt distance}$), $V = (1/2k_F)\dot{\phi}$, $N = (2/\pi)k_F$, and $k_F = \nu(\pi/c)$ where ν gives the band filling, we find for the Pt distortion amplitude u

$$u = (c/\pi) \{ (1/\nu) [(m^* - m_e)/M] \}^{1/2}.$$

With the experimentally determined effective mass $m^* \sim 10^3$, $\nu = 0.85$, and M the platinum mass, we predict a Pt distortion amplitude $u \sim 0.05 \text{ \AA}$ at $T=0$. Experimentally, a value of 0.015 \AA is found⁴⁰ at room temperature by inelastic neutron scattering. Probably the discrepancy of about a factor of 3 is not primarily due to the temperature dependence of u but to the oversimplified assumption that the Pt is the only ion which moves. In order to perform a realistic calculation of u , the detailed kinematics of the phase mode would have to be known, and possibly coupling of the CDW to other vibrational modes with $q = 2k_F$ should also be included.

To conclude, we have shown that optical spectroscopy is a very useful tool to study order and fluctuations of the Peierls transition. Both the gap and the Fröhlich mode are strongly affected by fluctuations, but below the mean-field transition temperature the rms amplitude of the order parameter is nearly temperature independent, which greatly facilitates a theoretical discussion. The optical data indicate that the dc conductivity is predominately given by single-particle excitations. The low mobility of charge carriers in the fluctuation regime is a fundamental consequence of the dimensionality.

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