Theory of Structural Variation in a Quasi-One-Dimensional Conductor

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Neutron-scattering studies of the quasi-one-dimensional conductor $K_2Pt(CN)_4Br_{0.30}\cdot H_2O$ demonstrate not only the existence of soft phonon modes but also the coexistence of central peaks and the development of a superlattice on cooling which, however, never becomes a true structural phase transition. We suggest that the central peak is due to bromine-induced, static, platinum-chain distortion and show that it affects the electronic motion sufficiently to create a strong enough fluctuation in the three-dimensional system to remove the expected phase transition.

Potassium cyano-platinide (KCP), $K_2Pt(CN)_4\times Br_{0.30}\cdot 3H_2O$, is a quasi-one-dimensional conductor[1] which has actually been shown to have giant Kohn anomalies in the phonon spectrum.[2] The temperature dependence of the phonons and the possibility of a structural phase transition in the deuterated KCP crystal have been investigated by neutron scattering.[3,4] There are two interesting findings: (1) A peak in neutron scattering intensity, "the central peak," appears at zero frequency and wave vector $(\zeta,\zeta,2k_F)$, where the c axis is along the direction of the platinum chains, even at temperatures where the soft phonon has not become unstable; (2) the intensity of the quasi-elastic peak near $\vec{q}_0=(\pi/a,\pi/a,2k_F)$ increases on lowering the temperature, but it never develops into a genuine Bragg peak. In other words, the long-range order perpendicular to the c axis is not established, and a three-dimensional structural phase transition is absent down to 6 K.

We present here a summary of our attempt to understand this behavior of KCP. Since the frequency width of the central peak at all temperatures and wave vectors $(\zeta,\zeta,2k_F)$ is within the instrumental resolution,[3,4] we may assume that the peak is elastic. We then suggest that it is induced by the bromine disorder in the following way. The conduction electrons are confined to move along the platinum chains and possess the one-dimensional character. Therefore, the density response is particularly strong near $2k_F$. This causes the giant Kohn anomaly in the phonon frequency spectrum.[5] The same effect can cause an impurity to induce a strong static Friedel oscillation of the electron charge density with wave vector $2k_F$ along the chains, which, in turn, induces a static lattice distortion of the same wave vector where the lattice is particularly soft. The random distribution of impurities[6] leads to a zero average of the net displacement of each platinum ion but leaves a finite mean-square displacement, which gives rise to the elastic central peaks in neutron scattering intensity.[7] Axe and Shirane[8] have briefly considered the impurity mechanism for the central peak in Nb_3Sn (though without specifically the Friedel oscillations) and rejected it in favor of an anharmonic-phonon mechanism. There are several reasons why KCP is different: (1) There is an unavoidable amount of disordered bromine ions. (2) The Kohn effect in the density response is shown experimentally to be strong. (3) A theory invoking anharmonicity would give the central peak a finite and temperature-dependent width, and this would have to be negligible compared with the 0.6-meV instrumental resolution. (4) The disorder-induced central peak forms an essential part of our theory to explain the observed structural variation with temperature. Without it, the absence of a structural phase transition would be difficult to understand.

In the mean-field theory for one dimension, the Kohn soft phonons become unstable on lowering the temperature, resulting in a phase transition to the Peierls distortion.[9] This phase transition is removed by the inclusion of large fluctuation effects in one dimension, but is reinstated at a lower temperature if the soft phonons are made three-dimensional[10] as a consequence of the interchain coupling, which arises partly from ionic and molecular interactions and partly from the Coulomb interaction of conduction electrons.[11] However, we shall show that the large disorder-induced static fluctuation is sufficient to remove the sharp phase transition, consistent with the ex-

perimental observation. A special consequence of the impurity-induced fluctuations is that the transverse and longitudinal coherence lengths remain finite at zero temperature in contrast to the case of one-dimensional dynamically induced fluctuations.[12]

The qualitative discussion above is based on an approximate calculation of the electron-phonon-impurity system. The phonon Green's function consists of a static part induced by the randomly distributed impurities,[13] Fig. 1(a), and a dynamic part, Fig. 1(b). The static part gives an elastic peak with intensity approximately proportional to ω_q^{-4}, where ω_q is the phonon frequency evaluated with the phonon self-energy at zero frequency. Strictly speaking, ω_q is not the true phonon frequency except at very low frequencies, but it does determine the stability of the lattice. ω_q is calculated with the phonon self-energy given by a "Hartree-Fock" modification[14] of the mean-field theory including interchain coupling [Fig. 1(c)]. Near $\vec{q}_0$, it is of the form

$$\omega_q^2 = \lambda \Omega_q^2 [\eta + \xi_\parallel^2 p_z^2 + V(p_\perp)], \tag{1}$$

where λ is the electron-phonon coupling constant and Ω_q the unrenormalized phonon frequency. p_z and $p_\perp$ are components of the wave vector $\vec{q}$ parallel and perpendicular to the platinum chains measured from $\vec{q}_0$. The dependence on the transverse wave vector, $V(p_\perp)$, is due to the interchain coupling, assumed to be the same as that

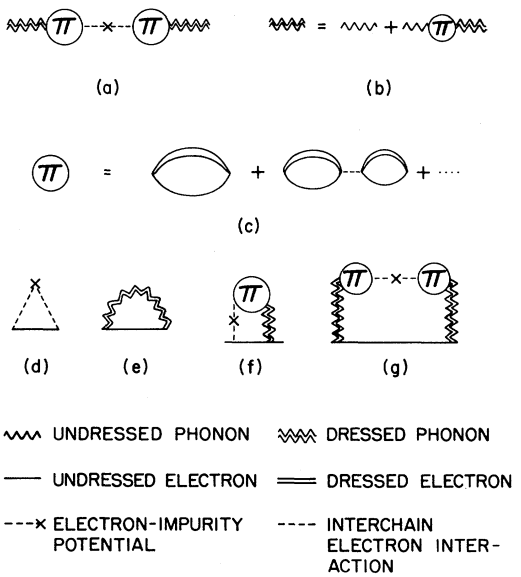

FIG. 1. (a) Static part of phonon Green's function. (b) Dynamic part of phonon Green's function. (c) Phonon self-energy. (d)–(g) Electron self-energy.

due to interchain Coulomb interaction of conduction electrons alone, which is calculated by the method of Sham,[15] taking the electrons to be free along the c axis[16] and tightly bound perpendicular to it. The shape of the $V(p_\perp)$ curve is almost independent of the transverse extent of the electronic wave function although the absolute magnitude varies greatly. The interaction of electron charge-density waves at $\vec{q}_0$ (out of phase from chain to chain) is attractive, but the interaction at $(0,0,2k_F)$ (in phase from chain to chain) is repulsive. Thus, we may take $V(p_\perp)$ to vanish at $p_\perp = 0$, letting η be a measure of the softest Kohn mode at $\vec{q}_0$:

$$\eta = \frac{\mu}{\lambda(\lambda+\mu)} + \frac{\lambda}{\lambda+\mu} \ln\left(\frac{T}{T_P}\right) + P\left(\frac{\Delta}{T}\right), \tag{2}$$

where μ is the single-spin electron density of states times the interchain energy of the density wave $\vec{q}_0$, assumed to be much smaller than λ here, T_P is the mean-field Peierls transition temperature, and Δ is the electron fluctuation energy gap. P vanishes for the zero gap. $\xi_\parallel$ is given by

$$\xi_\parallel = (V_F/T) Q(\Delta/T), \tag{3}$$

where V_F is the Fermi speed. P and Q are monotonic functions of Δ/T.

The conduction electron is confined to move in a platinum chain. The self-energy terms are given by Figs. 1(d)–1(g). The impurity scattering contributes two different kinds of terms, one, Fig. 1(d), giving the electron a finite lifetime τ which tends to lower the mean-field transition temperature.[17] The other terms give the electron a fluctuation energy gap Δ, which consists of

$$\Delta^2 = \Delta_0^2 + \Delta_1^2 + \Delta_2^2. \tag{4}$$

Δ_0 comes from the soft-mode fluctuations, and Δ_1 and Δ_2 from the impurity-induced lattice distortion, Figs. 1(f) and 1(g), respectively. If the soft phonons were one-dimensional, all the terms in the gap equation (4) would diverge as $\eta \to 0$. The large gap modifies η via the fluctuation term P in Eq. (2), resulting in a self-consistent solution of η which is finite at all temperatures, thus suppressing the mean-field phase transition. As the soft phonons are three-dimensional, Δ_0 and Δ_1 are only weakly dependent on η and by themselves would lower the transition temperature. But as $\eta \to 0$, Δ_2 remains divergent, thus suppressing the phase transition via a static (impurity-induced) fluctuation.[18]

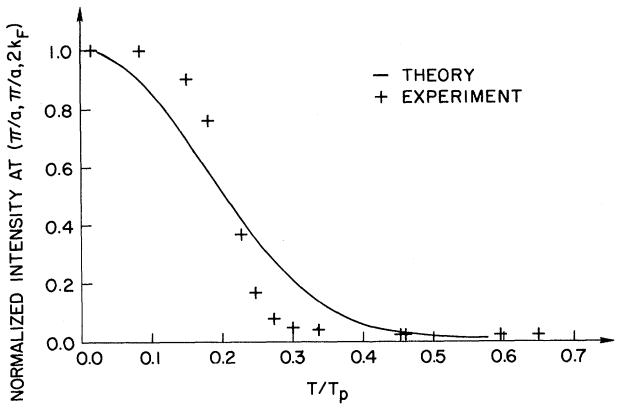

FIG. 2. Temperature dependence of the central peak intensity at $(\pi/a, \pi/a, 2k_F)$.

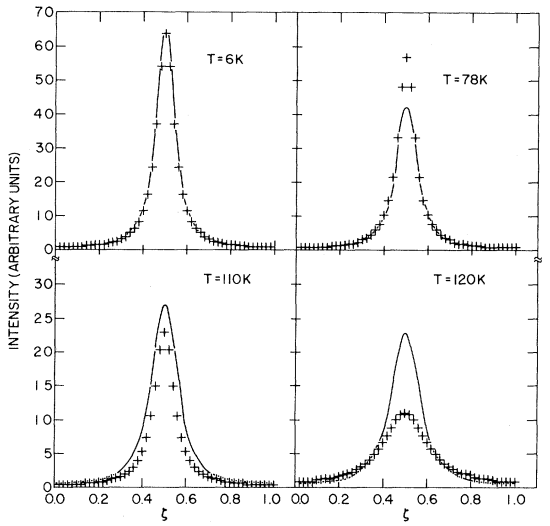

FIG. 3. Central peak along $(\zeta 2\pi/a, \zeta 2\pi/a, 2k_F)$ at various temperatures. —, calculated; +, Lorentzian fit to experiment (see Ref. 3).

If the instrumental resolution is taken into account and assumed to be larger than $\eta^{1/2}/\xi_{\parallel}$ but smaller than $\eta^{1/2}/\xi_{\perp}$, then the intensity of the bromine-induced central peak is proportional to $(\xi_{\parallel}\omega_q)^{-3}$. The latter is computed by taking the electron-phonon coupling constant $\lambda = 0.25$, the interchain interaction $\mu = 0.01$, and the disorder parameter[19] $\alpha \equiv 1/4\pi\tau T_P = 10^{-3}$. $T_P = 486$ K taking the Fermi energy to be 3.25 eV and the bandwidth as 4.5 eV.[16]

Figure 2 shows the comparison of the calculated intensity of the elastic peak at $\vec{q}_0$ as a function of temperature with experiment. The calculated intensity reproduces qualitatively the lack of a sharp phase transition and the saturation at low temperatures but does not drop as sharply around 100 K as observed experimentally. This may be due to our "Hartree-Fock" approximation for the fluctuation effects. The variation of the intensity with the wave vector $(\zeta 2\pi/a, \zeta 2\pi/a, 2k_F)$ along the (110) direction is fairly well reproduced, as shown in Fig. 3 where the measured curve has been made symmetric[3] about $\zeta = 0.5$. The asymmetry of the measured curve is due to the structure factor of the $Pt(CN)_4$ complex.[20] The calculated longitudinal coherence length ranges from $25c$ (c being the Pt-Pt distance along the chain) at room temperature to $200c$ at 6 K, in order-of-magnitude agreement with experimental estimates.[3,4]

The phonon spectrum derived from neutron-scattering experiments is at present uncertain in the region $q \sim 2k_F$. We find, however, that the dip should become extremely narrow at low temperatures where the solution to the equations of Fig. 1 leads to a relatively well-defined propagating collective mode. Narrowing of the dip near $2k_F$ occurs if the fluctuation energy gap is larger than the phonon energies.

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Observation of Transverse Zero Sound in Normal ^{3}He†

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The direct transmission of transverse zero sound between two AC-cut quartz transducers is observed in the vicinity of 3 mK; at higher temperatures the transition to classical viscous shear-wave behavior is observed through the effect on the damping of a single transducer.

Following the development of his theory of a Fermi liquid in 1957,[1] Landau predicted the existence of two collective excitations in normal ^{3}He at very low temperatures[2]; these excitations are referred to as longitudinal (symmetric) and transverse (asymmetric) zero sound. The onset of the longitudinal zero-sound mode was first observed by Keen and co-workers[3] while the complete temperature dependence of the velocity and attenuation was first studied by Abel, Anderson, and Wheatley[4]; the transverse mode has not been observed previously.

A requirement for the propagation of zero sound is that its velocity, V, be greater than the Fermi velocity, V_F. The strength of the Landau-quasiparticle interaction parameters, F_i, determines V and for the transverse mode the condition $V > V_F$ requires $F_1 > 6$ for the case $F_i = 0$, $i \geq 2$. Experimental data indicate $F_1 > 6$ for all pressures, but reliable data on higher F_i are not available.

The zero (or collisionless) sound regime is characterized by the condition $\omega\tau \gg 1$, where ω is the angular sound frequency and τ is a quasiparticle collision time; in the degenerate regime $\tau \propto T^{-2}$. As the temperature is increased, a hydrodynamic regime is entered where $\omega\tau \ll 1$. The modification of the transverse mode at high temperatures is radically different from the longi-