Dynamics of the charge-density wave. II. Long-range Coulomb effects in an array of chains

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In an array of one-dimensional conductors, fluctuations in the phase of the charge-density wave lead to changes in the electron density which are coupled by long-range Coulomb interactions. The Coulomb effect has a profound influence on the dynamics of the charge-density wave as observed by inelastic-neutron scattering. If the momentum transfer equals the Bragg vector of the three-dimensionally-ordered charge-density wave, it is predicted that almost all of the spectral weight is shifted from the pinning frequency to the plasma frequency ω_{pl}*. In $K_2Pt(CN)_4Br_{0.3}3.2H_2O$ (KCP) this effect is not observed due to the finite transverse correlation length that exists in this system even down to the lowest temperature. The organic conductor tetrathiafulvalene-tetracyanoquinodimethane (TTF-TCNQ), on the other hand, is an excellent candidate for this effect. By combining our results on the dynamics and the neutron scattering experiment on KCP, it is concluded that even the existence of short-range order in KCP is impossible to understand without including the long-range Coulomb effects.

I. INTRODUCTION

In a previous paper[1] (referred to as I) we have studied the dynamics of a pinned charge-density-wave (CDW) in a single chain. In reality we always have an array of such chains forming a three-dimensional crystal. As we shall see there are interesting complications in the dynamics of phase oscillations due to the existence of long-range Coulomb interaction. We shall address this question and the related question of the three-dimensional ordering of CDW in this paper.

II. LONG-RANGE COULOMB INTERACTION IN AN ARRAY OF CONDUCTING CHAINS

Let us denote the charge density on the ith chain by

$$\rho_i = \bar{\rho} + \rho_0 \cos[Qz + \phi_i(z)]. \tag{2.1}$$

There exist two sources of short-range interaction between these CDWs. The first is the electron-phonon interaction and the second is the Coulomb interaction between the CDWs. Due to the oscillatory nature of the CDW, it can be shown that such Coulomb interaction is in fact exponentially short range.[2] The sum of these two interactions can be parametrized by[3]

$$H' = \sum_{i\neq j} \mathcal{J}_{ij} \int dz \cos[\phi_i(z) - \phi_j(z)]. \tag{2.2}$$

This interaction will tend to lock the CDWs in a three-dimensionally-ordered state. We will separate $\phi_i(z)$ into two parts as in I, ie., $\phi_i(z) = \phi_i^0(z) + \psi_i(z)$ where $\phi_i^0(z)$ is the equilibrium phase and $\psi_i(z)$ is the small oscillation about it. We introduce the three-dimensional Fourier transform

$$\psi_i(z) = \frac{1}{(LN)^{1/2}} \sum_{\vec{q}} e^{iq_z z + \vec{q}_\perp \cdot \vec{R}_i} \psi_{\vec{q}}, \tag{2.3}$$

where R_i is the location of the chain in the perpendicular direction, L is the linear dimension in the z direction, and N is the number of chains. Equation (2.2) can be approximated by

$$H' \approx 4\pi v' \sum_{\vec{q}} v_\perp^2 q_\perp^2 |\psi_{\vec{q}}|^2. \tag{2.4}$$

The total elastic energy then takes the form

$$H = (4\pi v')^{-1} \sum_{\vec{q}} (v_\perp^2 q_\perp^2 + v_z^2 q^2) \psi_{\vec{q}} \psi_{-\vec{q}}. \tag{2.5}$$

In Eqs. (2.4) and (2.5) v' is defined by $v' = v_z^2/v_F$, where v_F is the Fermi velocity. We are interested in studying the Green's function including impurity averaging

$$\mathfrak{D}(\vec{q}, i\omega_n) = \int_{-\beta}^{\beta} d\tau\, e^{i\omega_n \tau} \langle T\psi_{\vec{q}}(\tau)\psi_{\vec{q}}(0)\rangle_{\text{av}}. \tag{2.6}$$

This can be written

$$\mathfrak{D}_1(\vec{q}, i\omega_n) = 4\pi v' (\omega_n^2 + v_\perp^2 q_\perp^2 + v_z^2 q_z^2 - 4\pi v' \Gamma)^{-1}, \tag{2.7}$$

where Γ is the self-energy term which takes into account the interaction with the impurities as discussed in I.

There is, however, another type of Coulomb interaction which is long range. The existence of a spatial variation of the phase ϕ_i gives rise to a change in the *background* charge density

$$\frac{\delta\rho_i}{\bar{\rho}} = \frac{1}{Q} \frac{\partial}{\partial z} \phi_i. \tag{2.8}$$

This is because $\partial\phi_i/\partial z \neq 0$ is equivalent to a local change in the Fermi momentum, which in turn implies a local change in the electron density in

order for the energy gap to remain at the Fermi surface. This is in fact the mechanism which couples the short-wavelength excitation in the CDW to long-wavelength excitation and is responsible for the optical activity of the phase mode.[2] This long-wavelength charge density gives rise to the following long-range Coulomb contribution to the energy

$$H_c = \frac{1}{2}\sum_{i\neq j}\int dz'\,dz'\,\frac{1}{\pi^2}\frac{\partial}{\partial z}\phi_i(z)\frac{\partial}{\partial z'}\phi_j(z')v_{ij}(z-z'), \tag{2.9}$$

where $v_{ij}(z-z')$ is the Coulomb potential and we have used the relation $\bar{\rho}=Q/\pi$. By considering the case of a square lattice in the transverse direction we transform (2.9) into Fourier space

$$H_c = \frac{4\pi e^2}{2\pi^2 a^2}\sum_{\vec{q}}\frac{q_z^2}{\epsilon_z q_z^2+\epsilon_\perp q_\perp^2}\phi_{\vec{q}}\phi_{-\vec{q}}, \tag{2.10}$$

where a is the interchain spacing and $\epsilon_\perp$ and ϵ_z are the high-frequency (high compared with the pinning frequency) dielectric constant. In one-dimensional systems ϵ_z is usually of the order of several hundred due to virtual excitations across a relatively small energy gap, whereas $\epsilon_\perp$ is of the order of 2 or 3. Hence it is important to take this anisotropy into account.

In this section we solve the problem within a mean-field theory which treats the impurity pinning within one chain first and then couples the chains together using Eq. (2.10). We obtain

$$\begin{aligned}\mathfrak{D}(q,i\omega_n) &= \left(\mathfrak{D}_1^{-1}(q,i\omega_n)+\frac{2e^2}{\pi a^2}\frac{q_z^2}{\epsilon_z q_z^2+\epsilon_\perp q_\perp^2}\right)^{-1}\\ &= 4\pi v'\left(\omega_n^2+v_z^2q_z^2+v_\perp^2q_\perp^2-4\pi v'\Gamma\right.\\ &\quad\left.+\omega_{pl}^{*2}\frac{q_z^2}{q_z^2+(\epsilon_\perp/\epsilon_z)q_\perp^2}\right)^{-1},\end{aligned} \tag{2.11}$$

where ω_{pl}^* is the plasma frequency of phase oscillations and is given by

$$\begin{aligned}\omega_{pl}^{*2} &= (4\pi v')2e^2/\pi a^2\epsilon_z\\ &= 4\pi e^2(\bar{\rho}/a^2)/m^*\ .\end{aligned} \tag{2.12}$$

Here m^* is the Frohlich effective mass and we have used the relation[1] $v'=(m/m^*)v_F$. Note that due to the large effective mass in Eq. (2.12), ω_{pl}^* is of the order of the normal phonon frequency and not the dielectric plasma frequency.[2]

Equation (2.11) implies a pole at a frequency near ω_{pl}^* for $q_\perp=0$ and $q_z\to 0$. We also note the highly singular dependence of this pole on $\vec{q}$. In particular if the dielectric constant is isotropic ($\epsilon_z=\epsilon_\perp$), in the limit of $|q|\to 0$, the plasma frequency $\omega_{pl}(\vec{q})=\omega_{pl}^*\cos\theta$, where $\cos\theta=q_z/|\vec{q}|$. This is a well-known result for the case of a three-dimensional array of electron gas.[4] We note that the phase mode which is a kind of Goldstone mode has a finite energy in the limit $q\to 0$, provided $\lim_{q\to 0}|q_z/q_\perp|\neq 0$, even in the absence of impurity pinning. A similar situation is known[6] in the theory of superconductivity where the Goldstone mode has a nonvanishing eigenfrequency in the limit of $|q|\to 0$ due to long-range Coulomb forces.

III. INELASTIC NEUTRON SCATTERING FROM CDW

A natural question to ask at this point is whether neutron scattering will measure the pinning frequency or the plasma frequency. The neutron is coupled to the lattice displacement and measures the density-density correlation function. The one-phonon creation structure factor is proportional to

$$S(\vec{q},\omega)=[n(\omega)+1]W(\vec{q},\omega)/\omega\,, \tag{3.1}$$

where

$$\begin{aligned}W(\vec{q},\omega) = \omega\,\mathrm{Im}\Big(\sum_{ij}\int dz\int_{-\beta}^{\beta}d\tau\langle\rho_i(z,\tau)\rho_j(0,0)\rangle\\ \times e^{iq_\perp\cdot(\vec{R}_i-\vec{R}_j)}e^{iq_zz}e^{i\omega_n\tau}\Big)_{i\omega_n\to\omega-i\delta}\end{aligned} \tag{3.2}$$

is the spectral weight function. In Eq. (3.1) $n(\omega)$ is the Bose factor and

$$\begin{aligned}\rho_i &= \rho_0\cos[Qz+\phi_i(z)]\\ &= \rho_0\cos[Qz+\phi_i^0(z)+\psi_i(z)]\,.\end{aligned} \tag{3.3}$$

Suppose we ignore the slow z variation in ϕ_i^0, we obtain

$$\begin{aligned}W(\vec{q},\omega)\\ =\frac{\rho_0^2}{4}\omega\,\mathrm{Im}\Big\{\sum_{ij}\int dz\int d\tau\,\langle e^{-i(\phi_i^0-\phi_j^0)-iQz}\psi_i(z,\tau)\psi_j(0,0)\rangle\\ \times e^{iq_\perp\cdot(\vec{R}_i-\vec{R}_j)}e^{iqz}e^{i\omega_n\tau}\Big\}_{i\omega_n\to\omega-i\delta}.\end{aligned} \tag{3.4}$$

Now let us assume that the chains have ordered three dimensionally, and for definiteness, assume that neighboring chains are out of phase. This condition is satisfied in KCP at low temperatures. Then $\phi_j=Q_\perp R_j$ and Bragg peaks will appear at $(\vec{Q}_\perp,2k_F)$ where $\vec{Q}_\perp=(2\pi/a)(\frac{1}{2}+h,\frac{1}{2}+k)$ and h and k are integers. Equation (3.4) becomes

$$W(\vec{q},\omega)=\omega\sum_{Q_\perp}\mathrm{Im}\mathfrak{D}\,(q_z-Q,\vec{q}_\perp-\vec{Q}_\perp,i\omega_n\to\omega-i\delta). \tag{3.5}$$

It is now clear that the only place where the plasmon effect is important is when the transverse momentum vector $\vec{q}_\perp$ obeys the Bragg condition of the new unit cell, i.e., $\vec{q}_\perp=\vec{Q}_\perp$. When $\vec{q}_\perp$ is dif-

ferent from $\vec{Q}_\perp$ the last term in Eq. (2.11) is unimportant, as the $q_\perp^2$ term in the denominator is large. In this case $S(\vec{q},\omega)\approx[n(\omega)+1]\,\mathrm{Im}\mathfrak{D}_1(\vec{q},\omega)$, and should show a peak only at the pinning frequency.

If $\vec{q}_\perp$ equals a transverse Bragg vector $\vec{Q}_\perp$, the situation is more complicated. Let us consider the case when the neutron has good resolution. Then the appropriate quantity to look at is a spherical average of the structure factor in the limit of $|\vec{q}-\vec{Q}_\perp|\to 0$

$$\mathrm{Im}\bar{\mathfrak{D}}(\omega)=\mathrm{Im}\int_{-1}^{1}\frac{d\mu}{2}\,\frac{4\pi v'}{-\omega^2-4\pi v'\Gamma+\omega_{p1}^{*2}[\epsilon_z/(\epsilon_z-\epsilon_\perp)][\mu^2/(\mu^2+\alpha)]}\,, \tag{3.6}$$

where $\alpha=\epsilon_\perp/(\epsilon_z-\epsilon_\perp)$ and $\mu=q_z'/|\vec{q}-\vec{Q}_\perp|$ where $q_z'=q_z-Q$. The μ integration can be done and in Fig. 1 we show the spectral weight $\omega\,\mathrm{Im}\bar{\mathfrak{D}}(\omega)$ for several values of α. We have used for definiteness the weak impurity pinning result of I and we have chosen $\omega_{p1}^{*2}\epsilon_z/(\epsilon_z-\epsilon_\perp)$ to be three times the pinning frequency γ. We see that for smaller α the spectral weight shifts increasingly to the plasma frequency. This is to be expected by inspection of Eq. (3.6) which shows that as long as $\mu>\alpha$ the pole is at the plasma frequency. While the above arguments are based on the limit $|\vec{q}-\vec{Q}_\perp|\to 0$ and the assumption of good neutron resolution, the qualitative feature should hold even if the neutron resolution is poor, as one can perform the spherical averaging for each finite $|\vec{q}|$ and then average over $|\vec{q}|$.

As we mentioned earlier, for most one-dimensional conductors, α is very small. For KCP we estimate that $\alpha\approx 0.02$. However, the neutron scattering data[6] show a peak at 2.5 MeV at both $(0,0,2k_F)$ and $(\frac{1}{2},\frac{1}{2},2k_F)$ and no sign of any plasmon excitation (which should be at 7.19 MeV according to the optical data[7]) at $(\frac{1}{2},\frac{1}{2},2k_F)$. For an explanation we have to go back to Eq. (3.3). We recall that KCP is never ordered three dimensionally even at the lowest temperature. It is then reasonable to factorize the $\langle\ \rangle$ term in Eq. (3.4) into

$$\begin{aligned}&\langle e^{i(\phi_i^0-\phi_j^0)}\psi_i(z,\tau)\psi_j(0,0)\rangle\\ &\quad\approx\langle e^{i(\phi_i^0-\phi_j^0)}\rangle\langle\psi_i(z,\tau)\psi_j(0,0)\rangle\\ &\quad\propto(-1)^{i-j}(|\vec{R}_i-\vec{R}_j|/\lambda)^{-1/2}\exp(-|\vec{R}_i-\vec{R}_j|/\lambda)\\ &\qquad\times\langle\psi_i(z,\tau)\psi_j(0,0)\rangle\,,\end{aligned} \tag{3.7}$$

where λ is the transverse correlation length. Upon Fourier transform we obtain a convolution of $\mathrm{Im}\mathfrak{D}$ with a Lorentzian. Let us specialize to the case when $\vec{q}_\perp=\vec{Q}_\perp$. We obtain

$$W(q_z+Q,\omega)=\omega\left[\ln\left(\frac{\gamma^2+q_0^2}{\gamma^2}\right)\right]^{-1}\mathrm{Im}\int_0^{q_0}dq_\perp^2\,\frac{1}{\gamma^2+q_\perp^2}\,\frac{4\pi v'}{-(\omega-i\delta)^2+v_z^2q_z^2+v_\perp^2q_\perp^2-4\pi v'\Gamma+\dfrac{\omega_{p1}^{*2}q_z^2}{q_z^2+(\epsilon_\perp/\epsilon_z)q_\perp^2}}\,. \tag{3.8}$$

Here we have introduced a cutoff q_0 in the integral and a corresponding normalization factor because a Lorentzian is not convergent in two dimensions. Since the important $\vec{q}_\perp$ dependence is in the plasma frequency term, we ignore the $v_\perp^2q^2$ term in Eq. (3.8) and the $q_\perp$ integration can be performed analytically. Roughly speaking the effect of the convolution is to replace the last term in the denominator in Eq. (3.8) by $\omega_{p1}^{*2}q_z^2/[q_z^2+(\epsilon_\perp/\epsilon_z)\lambda^{-2}]$. The result is a reduction in the spectral weight at ω_{p1}^*. To make comparison with experiment we note that whereas correlation in the transverse direction is resolved in the neutron scattering experiment, the opposite is the case in the chain direction. It is therefore necessary to average over q_z

$$\bar{W}(\omega)=\int_0^{\xi_0^{-1}}dq_z\,W(q_z+Q,\omega)\,. \tag{3.9}$$

We cut off the averaging at q_z equal to the inverse of the coherence length ξ_0 because that is the limit of validity of the small q_z expansion. We again choose Γ corresponding to the weak pinning solution given in I and $\omega_{p1}^*(\epsilon_z/(\epsilon_z-\epsilon_\perp))^{1/2}=3\gamma$. We also know that at $q=\xi_0^{-1}$, v_zq should approximately equal the bare phonon frequency which is also close to the plasma frequency. We have quite arbitrarily chosen $v_z\xi_0^{-1}=2.5\gamma$. From discussions below Eq. (3.8) it is clear that the relevant dimensionless parameter is $\Omega=(\epsilon_\perp/\epsilon_z)(\xi_0/\lambda)^2$. In Fig. 2, $\bar{W}(\omega)$ is plotted for several values of this parameter. We can see the change of the spectral weight from the pinning frequency to the plasma frequency as this parameter decreases. For KCP at 80 K, $\lambda\approx 50$ Å, $\epsilon_\perp/\epsilon_z\approx 0.02$. The coherence length ξ_0 is unknown but an estimate that satisfies the lower bound could be 500 Å. We obtain $(\epsilon_\perp/\epsilon_z)(\xi_0/\lambda)^2\approx 2$. For this value we see from Fig. 2 that there is relatively little spectral weight at the plasma frequency. The considerable width is due mainly

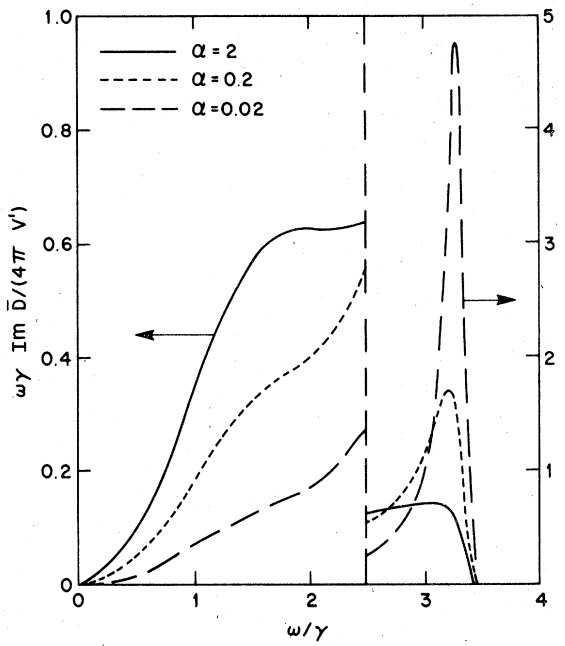

FIG. 1. Spectral weight at a momentum transfer close to a new Bragg vector and spherically averaged as described by Eq. (3.6). The parameter $\alpha = \epsilon_\perp/(\epsilon_z - \epsilon_\perp)$ is the anisotropy in the dielectric constant. The plasma frequency $\omega_{pl}^*(1+\alpha)^{1/2}$ is chosen to be 3γ. Note the shift in the spectral weight to the plasma frequency for increasing anisotropy.

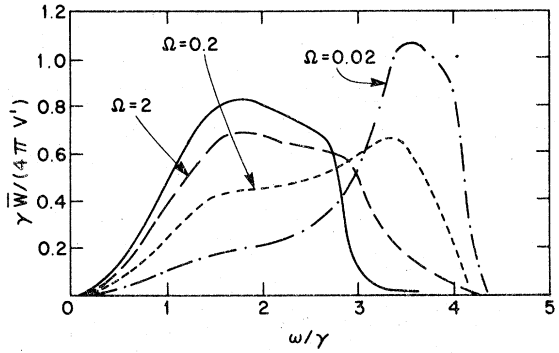

FIG. 2. Spectral weight $\bar{W}$ taking into account the finite transverse correlation length λ and the finite resolution in the chain direction. The parameter $\Omega = (\epsilon_\perp/\epsilon_z)(\xi_0/\lambda)^2$ and $\epsilon_\perp/\epsilon_z$ is chosen to be 0.02. The solid line is the averaged spectral weight in the absence of the Coulomb effect. Note the shift in spectral weight toward the plasma frequency (chosen to be 3γ) as Ω decreases, or as λ increases.

to the averaging over q_z. This is clear upon comparison with the solid curve in Fig. 2 which is the averaged spectral weight in the absence of Coulomb effects (i.e., $\omega_{pl}^* \to 0$). It is difficult to make quantitative comparison with experiment apart from the fact that the experiment [Fig. 9 of Ref. 6] shows a broad structure around 2.5 meV and no feature at $\omega_{pl}^* = 7.19$ meV. (We interpret the broad feature around 5 MeV as due partly to the amplitude mode and partly to the normal phonons with $q_z \gtrsim \xi_0^{-1}$ that are measured due to the finite resolution.) Note that the scattering intensity $\sim kT\omega^{-2}\bar{W}$ using Eq. (3.1) and the ω^{-2} factor tend to shift the weight to lower frequency.

Thus the absence of the plasma mode in the neutron scattering of KCP is due to finite range of the three-dimensional ordering. Physically this is a consequence of the fact that neutrons couple to the lattice position and are therefore sensitive to the phase on each chain. If the chains have random phase with respect to each other, an excitation of the lattice position at a definite transverse q vector must imply a random displacement of the CDWs from chain to chain. The dipoles induced at the ends of the sample are incoherent, and the plasmon is not excited. The situation is different in the optical excitation of the phase mode. The electromagnetic field is coupled to the long-wavelength charge density. From Eq. (2.8) this charge density is proportional to the gradient of the phase and is independent of its absolute value. Thus the plasma frequency is observable in optical reflectivity[7] even if the CDWs are completely disordered in the transverse direction.

It is natural to look for systems where the three-dimensional ordering is much longer range than in KCP. An obvious candidate is the organic conductor TTF-TCNQ where neutron[8] and x-ray scattering[9,10] indicate a low-temperature ordered state with the transverse correlation length beyond the experimental resolution. In the temperature interval $47 < T < 54$ CDWs are expected only on TCNQ chains and the ordering wave vector is $(q_a, q_c) = (a^*/2, 0)$. At lower temperatures, especially if $T < 38$, the transverse Bragg vector is $(q_a, q_c) = (a^*/4, 0)$. Thus the situation is very complicated due to the large size of the new unit cell in the a direction ($4a$) and to the existence of two types of chains in the a direction which are oppositely charged. There exist eight phase modes in the new Brillouin zone ($0 < q_a < a^*/4, 0 < q_b < Q, 0 < q_c < c^*$), one of which is the sliding mode ϕ_+, i.e., all chains in a unit cell are moving in phase. We focus our attention on the combination of modes in which the positively charged chains and the negatively charged chains move exactly out of phase and denote it by ϕ_-. In Fig. 3(a) $q_\perp$ is away from the transverse Bragg vector. The lower mode ϕ_+ is one in which the CDWs on the two chains move in phase and it is expected to be pinned by impurities. The upper mode ϕ_- is one in which the CDWs move out of phase and it is split from ϕ_+ by the inter-

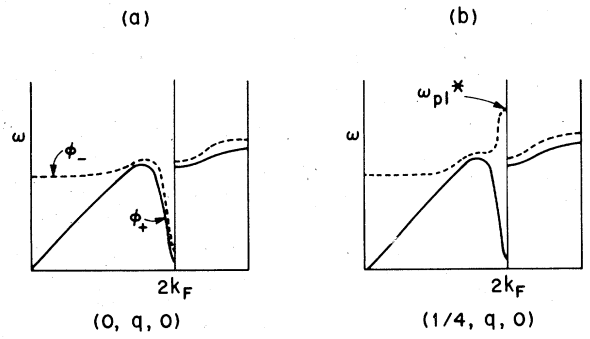

FIG. 3. Schematic picture of the expected phonon dispersion in TTF-TCNQ at low temperature. Solid line is the mode in which the TTF and the TCNQ chains move in phase. Dashed line is the optic mode in which they move out of phase. (b) shows that when the momentum transfer is near a Bragg peak [in this case $(\frac{1}{4}, 2k_F, 0)$], the ϕ_- mode is pushed up to the plasma frequency ω_{pl}^*. In practice the dispersion curves will have to be broadened by the neutron resolution function.

chain coupling energy. The other two modes at $q_z = 2k_F$ are the amplitude modes. Since the chains are oppositely charged, it is the ϕ_- mode which is optically active. Thus at $q_\perp$ equal to the transverse Bragg vector the ϕ_- mode is expected to be pushed up to the plasma frequency. This is shown schematically in Fig. 3(b).

IV. THREE-DIMENSIONAL ORDERING OF CDWs

In this section we briefly discuss the question of three-dimensional ordering in the presence of impurities in the light of the information on the dynamics that we have obtained so far. Let us first assume that we are in the weak pinning regime for the individual chain, i.e., the phase is slowly varying over many impurity sites. Furthermore, we assume that a short-range three-dimensional ordering is established, i.e., the phase is slowly varying in the transverse direction over several interchain spacings. First let us ignore the long-range Coulomb interaction. Then the argument for the domain size and pinning frequency given in I for a single chain can directly be generalized to the three-dimensional anisotropic problem. Let us suppose that the domain size is given by $L_z L_\perp^2$. We have to minimize the sum of the elastic energy and the impurity potential per unit volume

$$F = \frac{1}{4\pi v' a^2}\left(\frac{v_z^2}{L_z^2} + \frac{2v_\perp^2}{L_\perp^2}\right) - \frac{V_0\rho_0\sqrt{n}}{(L_z L_\perp^2)^{1/2}}\,, \tag{4.1}$$

where n is the number of impurities per unit volume. It is clear that F is minimized by the relations

$$v_z/L_z = v_\perp/L_\perp \tag{4.2}$$

and

$$v_z/L_z = (V_0\rho_0/\pi v' a^2)^2 n/v_\perp^2\,. \tag{4.3}$$

This result is in agreement with the expression for the range of the exponential decay derived by Sham and Patton[11] when generalized to the anisotropic situation.[12]

The same argument also produces a pinning frequency

$$\gamma \approx v_z/L_z\,. \tag{4.4}$$

When combined with Eq. (4.2) and the initial assumption of short-range order, i.e., $L_\perp > a$, we obtain

$$\gamma = v_\perp/L_\perp < v_\perp/a\,. \tag{4.5}$$

This inequality implies that the pinned mode must have observation dispersion in the transverse direction, since the frequency at a transverse momentum $q_\perp$ is roughly given by $\omega^2(q_\perp) = \gamma^2 + v_\perp^2 q_\perp^2$. Equation (4.5) then implies that $\omega(q_\perp = \pi/a) \gtrsim 2\gamma$. In other words we conclude that an interchain coupling $v_\perp$ that is strong enough to give short-range order must also be strong enough to give observable transverse dispersion. The problem is of course that exactly the opposite is seen experimentally in KCP: the mode at 2.5 meV is flat up to the transverse zone boundary.[6] While the above argument is made only for the weak impurity pinning case, we expect that the general conclusion would hold in the strong pinning regime, since three-dimensional ordering would be even more difficult in that case. Indeed in that case the pinning frequency is given by v_z/l, where l is the average distance between impurities. The observed pinning frequency for KCP is 2.5 meV or $\approx\frac{1}{3}$ of the normal phonon frequency. This implies that $l \approx 3\xi_0$. Since ξ_0 is the characteristic length over which the phase can vary, it is even more difficult to envision how three-dimensional ordering is possible if the impurity pinning were in the strong regime.

From the above discussions we conclude that it is impossible to understand the short-range ordering in KCP without including the effect of the long-range Coulomb interaction. This problem has been studied by Bergman, Rice, and Lee,[12] who concluded that the Coulomb effect tends to cancel the disruptive effect of the impurities. In the limit where the Coulomb effects dominate they obtained a power-law behavior for the correlation function instead of an exponential decay. Since a power-law decay has no intrinsic length scale, the domain type argument given earlier will no longer be applicable in this limit. This also indicates

that mean-field treatment of the Coulomb effect given in Sec. II of this paper is incomplete because the Coulomb effect also affects the single chain correlation and pinning frequency. We shall examine this question further in the next section.

V. COULOMB EFFECT ON THREE-DIMENSIONAL ORDERING

We shall include the Coulomb potential for the estimate of a domain size, i.e., instead of Eq. (4.1) we will minimize

$$F=\frac{1}{4\pi v' a}\left[\frac{V_z^2}{L_z^2}+2\frac{V_\perp^2}{L_\perp^2}\right]-\frac{V_0\rho_0\sqrt{n}}{(L_z L_\perp^2)^{1/2}}+\frac{2e^2}{\pi a^4}\frac{1}{\epsilon_z+\epsilon_\perp(L_z/L_\perp)^2}\,. \tag{5.1}$$

By introducing $r\equiv v_\perp L_z/v_z L_\perp$ and $\eta=\epsilon_\perp v_z^2/\epsilon_z v_\perp^2$ we rewrite (5.1) as

$$F=\frac{V_F}{4\pi a^2 L_z^2}(1+2r^2)-V_0\rho_0\frac{v_z}{v_\perp}\sqrt{n}\,rL_z^{-3/2}+\frac{2e^2}{\pi a^4\epsilon_z}(1+\eta r^2)^{-1}\,. \tag{5.2}$$

Minimization of Eq. (5.2) with respect to L_z and r yields the following two equations

$$(V_F/\pi a^2)(1+2r^2)-3V_0\rho_0(v_z/v_\perp)\sqrt{n}\,rL_z^{1/2}=0\,, \tag{5.3}$$

$$\frac{V_F}{\pi a^2 L_z^2}r-V_0\rho_0\frac{v_z}{v_\perp}\sqrt{n}L_z^{-3/2}-\frac{4e^2}{\pi a^4\epsilon_z}\eta\frac{r}{(1+\eta r^2)^2}=0\,. \tag{5.4}$$

Solving (5.3) for L_z and inserting the solution into Eq. (5.4) we obtain an equation for r

$$\frac{r^2(r^2-1)}{(1+2r^2)^4}=\frac{A}{(1+2r^2)^2}\,, \tag{5.5}$$

$$A=12e^2L_0'^2\eta/v_F a^2\epsilon_z\,, \tag{5.6}$$

where L_0' is given by

$$L_0'=(3\pi^2a^2V_0\rho_0v_z\sqrt{n}/v_\perp v_F)^{-2}=\left(\frac{1}{\pi^4}\frac{v_\perp}{v_z}\frac{L_0}{a}\right)^2L_0\,, \tag{5.7}$$

where L_0 is the domain size in the case of single chain as defined by Eq. (4.7) of I. In terms of L_0' and r, L_z and $L_\perp$ are given as follows

$$L_z=L_0'[(1+2r^2)/r]^2\,, \tag{5.8a}$$

$$L_\perp=(L_0'v_\perp/v_z)(1+2r^2)^2/r^3\,. \tag{5.8b}$$

From Eq. (5.5) we see that $r>1$ if Coulomb forces are present, i.e., $A\neq 0$. Equation (5.5) is simplified if we note $\eta\gg 1$, which Bergman, Rice, and Lee have estimated to hold for KCP

$$A/\eta^2=r^6(r^2-1)/(1+2r^2)^4\,. \tag{5.9}$$

The right-hand side of Eq. (5.9) is an increasing function of r^2 and we easily find the following

$$r^2\simeq 1+81A/\eta^2,\ \text{for } A/\eta^2\ll 1\,, \tag{5.10a}$$

$$r^2\simeq 3(1-16A/\eta^2)^{-1},\ \text{for } A/\eta^2\lesssim\tfrac{1}{16}\,. \tag{5.10b}$$

Equations (5.10b) and (5.8a) and (5b) imply that if Coulomb forces are strong enough so that $A/\eta^2\geq\frac{1}{16}$, the domain size is infinite. This is in agreement with the result of Bergman, Rice, and Lee who showed that the correlation function behaves as a power law

$$\langle\phi(z)\phi(0)\rangle\sim(z/\xi_z)^{-\alpha}\ \text{ for } z\gg\xi_z\,, \tag{5.11}$$

where the coefficient α is equal to $(A/\eta^2)^{-1/2}$ apart from a numerical constant. For large α the correlation function is very small by the time we enter the power-law regime. This means that the correlation function is well approximated by an exponential decay over a large range in z. In this same regime our domain argument produces finite estimates for the domain size. On the other hand, for small α, or large A/η^2, the power-law behavior dominates. The system has no characteristic length scale. This explains why there exists no finite solution for the domain size in this regime.

We now examine the case $A/\eta^2<\frac{1}{16}$ when the domain size is defined. The pinning energy γ' is given by

$$\gamma'^2=4\pi v' V_0\rho_0\sqrt{n}/(L_z L_\perp^2)^{1/2}\,. \tag{5.12}$$

Using Eqs. (5.7) and (5.8) we obtain

$$\gamma'^2=\frac{4}{3\pi}\frac{v_z^2}{L_0'^2}\frac{r^4}{(1+2r^2)^3} \tag{5.13a}$$

$$=\frac{4}{3\pi}\frac{v_z^2}{L_z^2}(1+2r^2) \tag{5.13b}$$

$$=\frac{4}{3\pi}\frac{v_\perp^2}{L_\perp^2}\frac{1+2r^2}{r^2}\,. \tag{5.13c}$$

We note that Eq. (5.13b) is substantially different from the result in the absence of Coulomb interaction as given by Eq. (4.4), particularly $r\gg 1$, and that the scaling given by Eq. (4.2) no longer holds. However, from Eq. (5.13c) we see that we have the same difficulty as before, namely that γ' is still smaller than the zone boundary energy $v_\perp/a$ and consequently dispersion in the perpendicular direction is predicted. The pinning frequency in the case when the domain size is not defined is much more complicated and cannot be obtained by the present technique. Hopefully in this limit it is possible to have short-range order and simultaneously a pinned mode that does not exhibit dispersion.

VI. CONCLUSION

We have shown that in a three-dimensional crystal consisting of one-dimensional conducting chains, fluctuations in the phase of the CDWs induce long-wavelength charge densities which are in turn coupled by long-range Coulomb interactions. We have studied the implication of this observation for neutron scattering and concluded that if the CDWs are relatively well ordered three dimensionally, phase mode should be observed at the plasma frequency at a transverse q vector equal to a transverse Bragg vector in the ordered state. In particular, TTF-TCNQ is a good candidate for the observation of this effect. The absence of this effect in KCP is explained in terms of the finite transverse correlation length even at the lowest temperature.

We also found that the existence of the short-range order in KCP is impossible to understand without including the long-range Coulomb effects. Furthermore, if the Coulomb effect is included in the estimation of the domain size, the domain size may go to infinity for sufficiently strong Coulomb coupling. This is consistent with the results of Bergman, Rice, and Lee, who found a power-law decay of the correlation function in this limit. The description of the dynamics in this limit is too complicated to be treated by the present technique. However, it seems reasonable to think that if the self-energy Γ appearing in Eq. (2.11) is properly renormalized, the conclusions we have reached concerning the observability of the plasma frequency in neutron scattering should remain qualitatively correct.

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