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Shift in the longitudinal sound velocity due to sliding charge-density waves

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The nonlinear conductivity observed for moderate electric fields in $NbSe_3$, TaS_3, $(TaS_4)_2I$, and $K_{0.3}MoO_3$ below the charge-density-wave-transition is believed to be due to the sliding of the charge-density waves. The sliding motion leads to a Doppler shift of the x-ray diffraction peaks, but this effect has not yet been resolved. We show here that besides the Doppler shift, a sliding incommensurate charge-density wave causes a change in the longitudinal sound velocity of the crystal that is linear in the charge-density-wave velocity. The resulting anisotropic shift is estimated in a mean-field approximation and found to be experimentally observable.

Some compounds exhibit charge-density waves (CDW's) below a transition temperature T_c and also display nonlinear conductivity for small electric fields (on the order of 1 V/cm).[1] This nonlinear conductivity has been interpreted as arising from "excess" current caused by sliding of the CDW. The small magnitude of the threshold field E_t supports this view because the electric field energy is small compared to the Fermi energy E_F, making a change in the number of free carriers unlikely. Experimentally, it has been shown that the x-ray diffraction peaks from the CDW do not lose intensity in the field, so that the nonlinearity is not caused by conversion of condensate electrons to normal electrons.[2]

However, to date there has not been an independent experimental verification that the excess conductivity for fields above E_t is due to sliding motion of the CDW. The most direct measurement would resolve the Doppler shift of the x-ray diffraction superlattice peaks when a CDW with wave vector $\vec{Q}$ moves with finite velocity $\vec{v}$, causing the elastic peak at $(\vec{Q},\omega=0)$ to change to an inelastic peak at $(\vec{Q},\omega=\vec{Q}\cdot\vec{v})$, but so far the shift is below the experimental resolution.[3]

In this Rapid Communication we show that for an incommensurate CDW the motion induces changes in the longitudinal sound velocity of the crystal that could be measured in ultrasonic experiments. The change in the sound velocity is proportional to the sliding velocity, and the resulting anisotropy should make the shift easier to resolve. The size of the effect is estimated and shown to be accessible to present ultrasonic techniques.

The effect is estimated using a very simple mean-field approximation in which the CDW amplitude is fixed and the phase ϕ varies sinusoidally, $\phi=\vec{Q}\cdot\vec{x}$. Impurities and thermal effects that induce fluctuations in the phase are ignored. It is straightforward to generalize the discussion to allow for harmonics of the wave vector $\vec{Q}$. One can start with a microscopic model involving electron-phonon coupling and solve for the equilibrium CDW distortion. For instance, in one dimension one could use the Fröhlich Hamiltonian[4]

$$H=\sum_k\left(\frac{\hbar^2k^2}{2m}b_k^\dagger b_k+\hbar ska_k^\dagger a_k\right)+\sum_{k,q}\lambda_k(a_k^\dagger b_q^\dagger b_{q+k}+\text{H.c.}) \qquad (1)$$

and solve the gap equation in the mean-field approximation to find the CDW amplitude at wave vector $\vec{Q}=2k_F$. Here, the $a^\dagger$'s and a's are phonon creation and annihilation operators, the $b^\dagger$'s and b's create and annihilate electrons, m is the electron mass, and s is the speed of sound. Regarding this amplitude as fixed, one then evaluates the resulting effective Hamiltonian for the low-frequency phonons. A static CDW thus induces a potential on the ions of the form $V_0\cos\vec{Q}\cdot\vec{x}$. One can evaluate V_0 for the one-dimensional jellium model, but here it will be estimated by using experimentally obtained values of the mean ionic displacements in the CDW state. The change in the phonon frequencies caused by a periodic potential *sliding* with velocity v is calculated to second order in perturbation theory for small V_0, and it is shown that there is a contribution linear in v.

The simplest case involves phonons parallel to the CDW wave vector, for which one can model the phonons as arising from a one-dimensional chain of ions. For a CDW sliding with velocity v, the classical equation of motion[5] for the displacement x_j of the jth ion can be written

$$mx_j''=-\sum_k D_{jk}x_k+QV_0\sin Q(x_j+vt) \quad . \qquad (2)$$

The dynamical matrix D_{jk} describes the ion-ion interactions in the harmonic approximation, and m is the ion mass. We assume D_{jk} describes phonons so that it is a function of $j-k$, it falls off sufficiently quickly with distance, and that it is symmetric. This equation is nonlinear, so we proceed by assuming that V_0, and hence the distortions, are small. However, one must allow for the fact that the lowest-energy state of the chain is distorted; so one writes the position x_j of the jth ion as $x_j=aj+\delta_j(t)+u_j(t)$, where a is the lattice constant, $\delta_j(t)$ is the forced distortion, and $u_j(t)$ describes the phonons. To first order in V_0 the distortion $\delta_j^{(1)}(t)$ is

$$\delta_j^{(1)}(t)=\frac{QV_0}{m(Qv)^2-D(q=Q)}\sin Q(aj+vt) \quad . \qquad (3)$$

Here, $D(q)=\sum_j D_{oj}e^{iqaj}$. To second order in V_0, one finds that the

$$u(q,\omega)=\frac{1}{2\pi N}\sum_j\int dt\,\exp i(qaj-\omega t)u_j(t)$$

obey

$$m\omega^2(q)u(q,\omega)=D(q)u(q,\omega)-\tfrac{1}{2}Q^4V_0^2\frac{u(q,\omega)}{m(Qv)^2-D(q=Q)}+\tfrac{1}{2}V_0Q^2[u(q+Q,\ \omega-Qv)+u(q-Q,\ \omega+Qv)]\ . \tag{4}$$

Equations (4) are coupled linear equations that can be solved by iteration for small V_0 to yield

$$m\omega^2(q)=D(q)-\tfrac{1}{2}V_0^2Q^4\frac{1}{m(Qv)^2-D(q=Q)}+\tfrac{1}{4}V_0^2Q^4\left[\frac{1}{m(\omega+Qv)^2-D(q-Q)}+\frac{1}{m(\omega-Qv)^2-D(q+Q)}\right]\ . \tag{5}$$

We assume the unperturbed system has reflection invariance, so that $D(k)=D(-k)$. This expression can be evaluated in the limit $q\to 0$, $\omega\to sq$ (s is the speed of sound), $v\to 0$ to yield

$$s=s_0-\frac{Q^4V_0^2}{D^3(Q)}QD'(Q)v+O(v^2)\ . \tag{6}$$

Changes in s that are independent of v are accounted for in s_0, which is the speed of sound when $v=0$, and $D'(q)=dD(q)/dq$.

Alternatively, one can evaluate the phonon frequencies using Green's function techniques. It is again necessary to allow for the distortions that occur in the lowest-energy state, Fig. 1(a), in order to ensure stability. The diagrams that lead to Eq. (5) are all represented in Fig. 1. The first-order scattering process, not shown, from (q,ω) to $(q\pm Q,\ \omega\mp Qv)$ is responsible for the Doppler shift in the Bragg peak mentioned earlier but does not affect the sound velocity. A pictorial description of the mechanism is shown in Fig. 2; the phonon at (q,ω) scatters off the distortion at $[q+Q,\ \omega(q+Q)-Qv]$ and $[q-Q,\ \omega(q-Q)+Qv]$. The usual denominators of second-sound perturbation theory are then slightly shifted in different directions.

In the calculations discussed above the charge-density wave is assumed to slide as a rigid body for fields above threshold. This assumption is only valid well above threshold. Near threshold the internal degrees of freedom of the charge-density wave can not be ignored. The qualitative aspect of our result, the linear shift, which arises from symmetry breaking due to a moving density wave will persist, although there may well be enhanced damping of the sound mode.

The expression (6) for $\delta s=s-s_0$ can be written in terms of the magnitude of the distortion $|\delta_j|\simeq\dfrac{QV_0}{D(Q)}$ as

$$\delta s=|\delta_j|^2Q^3\frac{D'(Q)}{D(Q)}v\ . \tag{7}$$

This expression for the shift displays several interesting features. First, it is proportional to $D'(Q)$, which is finite for an incommensurate CDW but is zero for a commensurate CDW. In the commensurate case, umklapp scattering within the unit cell must be considered, rendering the treatment described here inadequate, but we do not expect a shift linear in v to appear. Assuming that $D'(Q)$ is not anomalously large or small [so $QD'(Q)\sim D(Q)$], one finds δs is on the order of $Q^2|\delta_j|^2v$. Note that $|\delta_j|^2Q^2$ is the dimensionless measure of the lattice distortion due to the charge-density wave. The velocity shift obtained here can then be looked on as arising from the motion of this distortion at velocity v. Experimentally, $|\delta_j|$ is found to be approximately 5% of the lattice constant,[6] so for $Q\sim\pi$, drift velocities on the order of 10 cm/sec and sound velocities of about 10^5 cm/sec, one finds

$$\delta s/s\approx 10^{-6}\ , \tag{8}$$

which is large enough to be resolved experimentally.[7]

Since the shift is linear in v, changing the direction of the electric field driving the CDW should alter the shift. It is emphasized that this shift is added to the change induced by a static CDW.

In summary, we have shown that a moving CDW causes an anisotropic shift in the longitudinal sound velocity of the crystal that is linear in the CDW velocity. The effect provides a means to obtain independent experimental evidence for sliding CDW conductivity in crystals such as $NbSe_3$, $K_{0.3}MoO_3$, TaS_3, and $(TaS_4)_2I$.

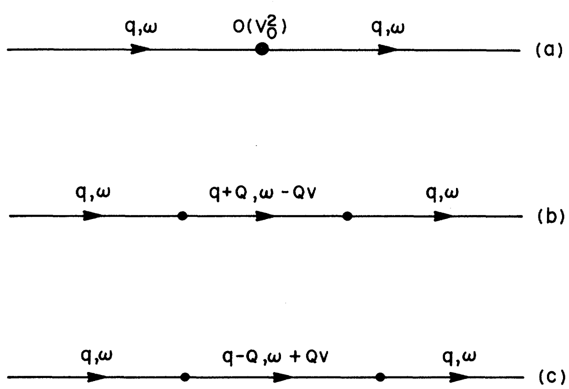

FIG. 1. Second-order diagrams that contribute to the sound velocity shift. Diagrams (b) and (c) contribute the linear term in the CDW velocity.

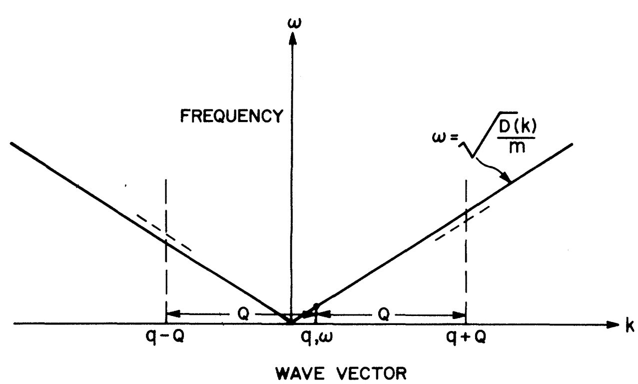

FIG. 2. Schematic representation of mechanism causing linear velocity shift. There are momentum transfers from q to $q+Q$ and $q-Q$, and the finite velocity causes different energy shifts for the two terms.

At a conference where preliminary results of this work were reported,[8] we learned of experiments[9,10] in which a change of the elastic modulus due to sliding charge-density wave was observed. However, the magnitude of the observed effects are much larger than estimated here because of geometry and phason distortion effects. Detailed comparison with experiment must await a more complete understanding of these factors.

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