counted for by the instrumental resolution of 0.7 –0.8 eV. Another part can be attributed to the instrumental angular resolution coupled to the rapidity of the energy dispersion; a reasonable estimate of the angular resolution would be 5° full width at half maximum, implying a further energy spread of ~0.8 eV. It is therefore quite possible that the residual width due to the inherent lifetime of the surface state is consistent with the above expectations. Clearly, experimental work at higher angular and energy resolution would be desirable to resolve this point.

In summary, we have extended the technique of KRIPES to the study of surface states. We have mapped the $E(\boldsymbol{k}_{\parallel})$ relations for an unoccupied surface state on Pd(111)—information unobtainable in any other technique. In general, KRIPES promises to deliver the half of the information on surface states not accessible with use of ARUPS. Some particular questions concerning the energy width and dispersion of the Pd(111) surface state would be worthy of further investigation.

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Sliding Conductivity of Charge-Density Waves

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A classical model of charge-density wave motion at large velocities is used to describe the nonlinear response of the charge-density wave in fields well above the threshold. The charge-density wave is regarded as a charged, deformable medium and its interaction with pinning centers is treated in perturbation theory. The results fit the available high-field data on $NbSe_3$ well. The observed interference effects between a large dc field and an ac field are also explained.

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Peierls and Fröhlich[1] showed how the coupling between conduction electrons and the phonons of an anisotropic metal could lead to a charge-density wave (CDW): a periodic modulation of the electron density and a corresponding distortion of the lattice. Fröhlich also suggested that a CDW could slide and carry electrical current. This phenomenon has now been observed[2,3] in $NbSe_3$; the CDW is pinned at small electric fields but moves in the presence of fields larger than a threshold depinning field.[4] This results in a nonlinear current-voltage curve,[2,3] which has to date been analyzed in terms of both a single classical degree of freedom[5] (or "particle") and a Zener-tunneling model.[6] The latter theory involves the quantum mechanical tunneling of macroscopic portions of the CDW, which we believe implausible. Neither theory is consistent with all of the data.[7]

The purpose of this Letter is firstly to present a model which we believe is a more realistic and complete model of a moving CDW than has been given to date. It is classical and includes the internal degrees of freedom of the CDW. Secondly we calculate its properties, within perturbation theory, at large dc electric fields.

The first result is that the leading terms in the current density, j, for high electric field, E, are

$$j = \sigma E - C\sqrt{E} \tag{1}$$

with σ the conductivity in the absence of pinning and C a constant. This form fits the available data very well. We have also investigated the effects of an alternating current superposed on the dc current. The results explain the observed interference effects.[8] The characteristic field, $(C/\sigma)^2$, determines the range of validity of the perturbation theory and is equal, to within numerical factors, to the weak pinning threshold field of Lee and Rice.[4] The voltage oscillations measured[3] with a dc current are, however, not present in the theory. This indicates that either the model, or its leading order perturbative solution, is incomplete, or that finite-size effects are being observed. This issue is the subject of continuing study.

We consider a CDW interacting with impurities, which are treated perturbatively following a method[9,10] which has been successfully used to study vortex lattice motion in superconducting films. Dimensionality is crucial. The exponent in the second term of (1) is $(d-2)/2$ where d is the dimensionality of phase space for coherent excitations of the internal degrees of freedom of the CDW. These classical internal degrees of freedom are thus of central importance, the fit of the experiments to Eq. (1) indicating the three-dimensionality of the CDW, which is consistent with x-ray studies.[11] The impurities distort the CDW as it moves over them, resulting, at second order in the pinning potential, in a net retarding force. The important normal modes of the CDW turn out to be those of long wavelength and low frequency, and so macroscopic equations of motion may be used. At large fields, the CDW is moving rapidly over the pinning centers, causing the local distortions of the CDW to be small. The leading order correction is then expected to dominate the nonlinear transport.

Distortions $u(\vec{r})$ of the CDW are acted on by various forces. Firstly, there is an elastic restoring force per unit volume $-\int D(\vec{r}-\vec{r}')u(\vec{r})d^3r'$, where $\vec{r}'$ labels the position of the undistorted CDW at time 0 and $u(\vec{r}')$ is the local distortion in the incommensurate z direction. (Since commensurability energies are much larger than pinning energies due to dilute impurities, only distortions in the z direction need be considered.) Secondly, the CDW is acted on by the applied uniform electric field E, which we take to be along the z axis. In the long-wavelength limit the field couples to the full mobile charge of the CDW given by an effective charge density[4] ρ_c. The pinning forces $-\nabla\Phi(\vec{r})$, which have structure on short length scales, will couple to the spatially dependent CDW charge density $\rho(\vec{r})$.

The coupling of the CDW to the other degrees of freedom in the solid can be represented by a phenomenological damping force $-\lambda\dot{u}(\vec{r})$. In considering long-wavelength distortions of the CDW, it is necessary to consider the uncondensed electrons which act to screen the long-range Coulomb interactions. In the absence of this screening, the low-frequency phase modes with finite q_z components will be shifted to high frequencies and become plasmonlike. However, for $NbSe_3$ the uncondensed electrons are sufficiently mobile to screen out the Coulomb forces at the frequencies of interest, and the result of including this effect is an enhanced λ of the CDW. We will ignore for simplicity other scattering between normal conduction electrons and the CDW.

When we combine all the terms, the equation of motion for u becomes

$$m\ddot{u}(\vec{r},t) + \int D(\vec{r}-\vec{r}')u(\vec{r},t)d^3r' + \lambda\dot{u} - \rho_c E(t) = -\rho(\vec{r})\Phi_z(\vec{r}+\vec{u}(\vec{r},t)), \tag{2}$$

where m is the effective mass density of the CDW and the Cartesian subscript on Φ denotes differentiation.

For a given dc field E_0, and with a superposed ac field of frequency ν, the average velocity v of the CDW can be computed[9] in perturbation theory about the weak-pinning or high-velocity limit. We subtract off the CDW displacements in the absence of pinning[12]:

$$u(\vec{r},t) = vt + (a/\nu)\sin\nu t + \tilde{u}(\vec{r},t), \tag{3}$$

where the applied field is

$$E(t) = E_0 + [(a/2\rho_c)(\lambda + \mathrm{Im}\nu)e^{i\nu t} + \mathrm{c.c.}]. \tag{4}$$

The CDW distortion may then be written

$$\tilde{u}(\vec{r},t) = \int d^3r' \int dt'\, G(\vec{r}-\vec{r}', t-t')[(\rho_c E_0 - \lambda v) - \rho(\vec{r}')\Phi_z(\vec{r}' + \vec{u}(\vec{r}',t'))], \tag{5}$$

where the Fourier-transformed Green's function is given by

$$G^{-1}(\vec{k},\omega) = -m\omega^2 + D(\vec{k}) + i\omega\lambda. \tag{6}$$

The solution $\tilde{u}$ can be obtained in perturbation theory with contributions u_0, u_1, and u_2 given by replacing the quantity in square brackets in Eq. (5) by $\rho_c E_0 - \lambda v$, $-\rho(\vec{r}\,')\Phi_z(\vec{r}\,'+\vec{v}t' + (\vec{a}/\nu)\sin\nu t')$, and $-\rho(\vec{r}\,')u_1(\vec{r}\,', t')\Phi_{zz}(\vec{r}\,'+\vec{v}t' + (a/\nu)\sin\nu t')$, respectively. We obtain to second order an expression for the volume-averaged incremental dc velocity,

$$\langle\dot{\tilde{u}}\rangle = \lambda^{-1}[(\rho_c E - \lambda v) + \sum_{g,n}|\rho_g|^2\int d^3q(2\pi)^{-3}q_z^{\,3}\Lambda(\vec{q})J_n^{\,2}(q_z a/\nu)\mathrm{Im}G(q-g, q_z v - n\nu)], \tag{7}$$

where ρ_g is the structure factor of the CDW density $\rho(\vec{r})$, Λ is the impurity potential correlation function

$$\Lambda(\vec{q}) = \int d^3r\, e^{-i\vec{q}\cdot\vec{r}}\frac{1}{V}\int d^3s\,\Phi(\vec{s}+\vec{r})\Phi(\vec{s}), \tag{8}$$

and J_n is the Bessel function of order n. The condition that $\langle\dot{\tilde{u}}\rangle$ must vanish then leads to an expression for v, the self-consistent dc velocity, in terms of the field E_0.

The evaluation of the integral in Eq. (7) can be simplified by using bounds on the characteristic frequencies obtained from frequency-dependent Ohmic (i.e., small field) conductivity measurements.[13] If we consider replacing the right-hand side of Eq. (2) by $-K_p u$ where K_p is an average stiffness due to pinning, then we obtain for the ac Ohmic conductivity the simple harmonic-oscillator result

$$\sigma(\omega) = i\omega\rho_c^{\,2}(-m\omega^2 + i\omega\lambda + K_p)^{-1} + \sigma_n, \tag{9}$$

where σ_n is the conductivity of the normal electrons. Comparison of the parameters with experiment yields the bounds $\lambda/m \geq 10^{10}$ Hz, $\sigma_n \gtrsim 10^{15}$ Hz, and $(\rho_c^{\,2}/m)^{1/2} \gtrsim 10^{12}$ Hz. The characteristic frequency $g_z v$ which enters the argument of the Green's function is believed to be in the range[9] $\sim 10^6$–10^7 Hz, and we may hence take it to be small in comparison to the other characteristic frequencies.

The dominant contribution to the integral in Eq. (7) comes from long-wavelength, dissipative modes, $\omega_p(\vec{k}) \sim -i\lambda^{-1}D(\vec{k})$ with $\vec{k} = \vec{q} - \vec{g} \to 0$, for which we may neglect the inertial term in Eq. (6). Since the frequency $g_z v$ is small, the integral is dominated by small k and is given by replacing q with g everywhere except where it occurs as $q - g$, or $q_z v - n\nu$ when this is small. To obtain the dc current in the absence of an ac field, we set $a = 0$ in Eq. (7), whence only the $n = 0$ term appears in the sum over n and we obtain

$$j = (\sigma_n + \rho_c^{\,2}/\lambda)E_0 - (4\sqrt{2}\pi)^{-1}(K_x K_y K_z)^{-1/2}(\rho_c/\sqrt{\lambda})\sum_g|\rho_{\vec{g}}|^2|g_z|^3\Lambda(\vec{g})|g_z v_0|^{1/2}, \tag{10}$$

where for small k we have assumed[14] that $D(k) = K_x k_x^{\,2} + K_y k_y^{\,2} + K_z k_z^{\,2}$. The velocity in the absence of pinning, v_0, is given by $v_0 = \rho_c E_0/\lambda$ and we thus see that Eq. (10) has the form of Eq. (1) with C second order in the pinning. The nontrivial term in Eq. (10) is the first term in an expansion, valid for high velocities, in powers of $\Lambda E_0^{-1/2}$.

To test the form, Eq. (1), for the non-Ohmic transport, the data of Fleming and Grimes[3] are plotted in Fig. 1 as j/E vs $1/\sqrt{E}$. The high-field data are very well fitted by a straight line, as required by Eq. (1).

It is instructive to compare our result with the predictions of two other theories which yield results for the high-field dc response. Grüner, Zawadowski, and Chaikin[5] model the CDW by a single particle moving in a viscous medium in a periodic potential. Expanding their results about the high-field limit, one obtains

$$j = \sigma E + O(1/E); \tag{11}$$

with this form the tangent to the j vs E curve at large E intersects the $j = 0$ axis at the origin. This is clearly at variance with both Eq. (1) and the data of Ref. 3. Physically the deviation from the $j \propto E$ behavior at high fields is due to the excitation of internal modes of the CDW, which are not taken into account by a model which represents the CDW as a point particle.

Bardeen[6] has presented a theory of non-Ohmic conduction of a CDW, based on Zener-like tunneling. Expanding his result about the high-field limit gives

$$j = \sigma E - j_0 + O(1/E), \tag{12}$$

where j_0 is independent of E. However, it also disagrees with Eq. (1). If Eq. (12) were plotted in Fig. 1, the resulting curve would be a concave downward curvature. While data at higher fields are required to determine which theory, if any, is correct, the result of the present theory fits the available dc data very well, and rather better than the tunneling theory.[6]

In addition to the high-field dc response, we can compute from Eq. (7) the dc response to an ac field added to a large constant dc field by retaining

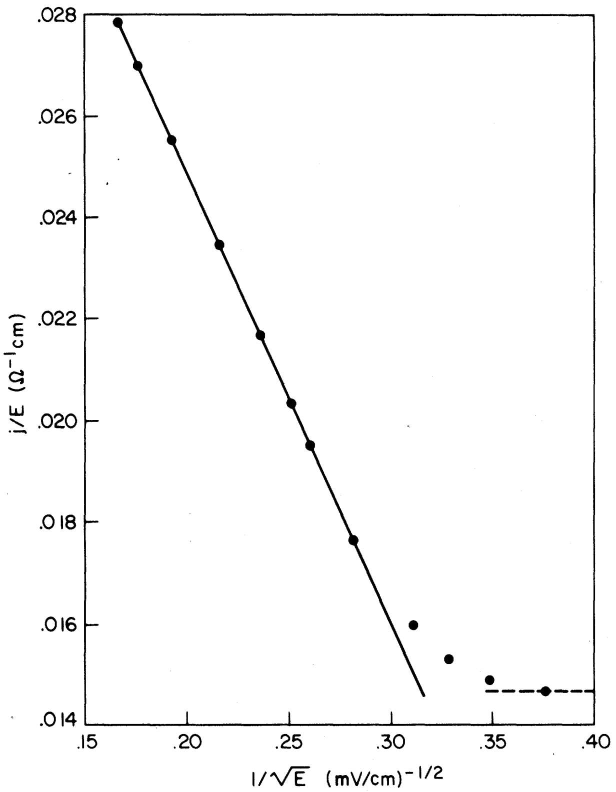

FIG. 1. The high-field current-voltage data on $NbSe_3$ at $T = 51$ K from Ref. 3. The theory predicts a straight line as shown in this plot of j/E vs $1/\sqrt{E}$.

the sum on all n and performing the integrals over $\vec{q}$ near reciprocal-lattice vectors. For a given large dc field E_0, there will be rather sharp steps in the dc current as a function of ν when $g_z v = n\nu$ for some n and g_z, that is, when the CDW moves n wavelengths in one period of the applied ac field. The differential conductivity near these steps is found to be

$$\frac{dj}{dE_0} = \frac{-\rho_c^2}{\lambda} \sum_{g_x g_y} \frac{|\rho_g|^2 g_z^4 \Lambda(\vec{g})}{4\pi\lambda v_0 (K_x K_y)^{1/2}} J_n^2\left(\frac{g_z a}{\nu}\right) \times P\left(\frac{4K_z}{\lambda v_0^2}(g_z v - n\nu)\right), \qquad (13)$$

where $P(x) = (|x|/\sqrt{2})(1+x^2)^{-1/2}[(1+x^2)^{1/2} - 1]^{-1/2}$ is a function peaked at $x = 0$ with $P(0) = 1$ and $P(x) \sim (2|x|)^{-1/2}$ for $|x| \gg 1$. For small ac field, a, and *fixed* E_0, the height of the current step at $\nu_s(n, g_z)$ and the peak height in dj/dE_0 will be proportional to $[(\frac{1}{2}na/v)^n/n!]^2$. We may identify $g_{z0}v$ as the fundamental frequency of the voltage fluctuations in the dc experiments,[3,8] with g_{z0} the z component of the smallest reciprocal-lattice vector. The steps corresponding to g_{z0} and various n have been observed experimentally by Monceau, Richard, and Bernard[8] although at this stage a detailed analysis of the amplitudes and step shapes has not been carried out.

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