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Frequency-Dependent Conductivity in NbSe3

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Frequency-Dependent Conductivity in $NbSe_3$

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The observation of a frequency-dependent conductivity (σ) and dielectric constant (ϵ) in $NbSe_3$ is reported. In both charge-density—wave phases a strong frequency dependence and huge dielectric constant are observed below 100 MHz, with greatest effects observed at 42 K. The conductivity σ increases smoothly from the dc value to the high-frequency ($f = 100$ MHz) limit; this increase is accompanied by the reduction of ϵ. A resistance-capacitance network model is suggested to account for the observed frequency dependence.

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Niobium triselenide, $NbSe_3$, shows two phase transitions,[1] at 145 K (T_1) and 59 K (T_2) associated with the development of incommensurate charge-density waves (CDW). These transitions lead to the increase of resistivity which is ascribed to the reduction of the Fermi surface;[2,3] the CDW condensate is assumed to be pinned by impurities to give negligible contribution to the dc conductivity at low electric fields. The conductivity increases when the electric field is larger than a threshold field E_T and then saturates at high electric fields.[4-6] This and the observation of noise in the nonlinear region[6,7] together with the x ray observation of the superstructure at high electric fields[8] demonstrate that the depinning of the CDW is responsible for the increase of the conductivity with increasing electric field. The resistive anomalies are also suppressed at microwave frequencies[2] suggesting that the pinning frequency is less than 10^{10} Hz.

In this paper we report the first observation of frequency-dependent conductivity associated with the incommensurate CDW states in $NbSe_3$. We show that the CDW anomalies are suppressed at extremely low frequencies and also lead to a gigantic dielectric constant. The transition between the low-frequency ($f<1$ MHz) and high-frequency ($f>100$ MHz) limit is smooth with no clearcut evidence for a well-defined pinned CDW mode.

The conductivity was measured between 10 and 300 K by a two-probe configuration. Contact resistances, measured by dc four-probe configuration, were found to be two orders of magnitude smaller than the sample resistance. Also the measured resistance (including the small contact resistance contribution) was found to be independent of frequency in the high temperature, metallic region showing that the measured frequency dependence at low temperatures is associated with the behavior of $NbSe_3$. A bridge method was used up to 100 MHz and a radiofrequency circulator circuit at high frequencies.[9] In both cases the sample impedance is represented by a parallel RC circuit which is balanced by variable resistance and capacitance components. The balance of the resistive component gives $\mathrm{Re}\sigma(\omega) = R^{-1}$, the balance of the capacitance gives $\mathrm{Im}\sigma(\omega) = C$ with the dielectric constant defined as $\epsilon(\omega) = \mathrm{Im}\sigma(\omega)/\omega$. Our measuring configuration allows the joint measurement of the dc and rf conductivity. $\mathrm{Re}\sigma(\omega)$ can be measured with an accuracy better than a few percent, but the balancing out of the phase component is more ambiguous: values of $\mathrm{Im}\sigma(\omega)$ have an error of about 30%. Measurements up to 100 MHz were performed on different samples and normalized to the room temperature data obtained on the sample measured between dc and 100 MHz. The out-of-phase component was calibrated using the relation $\mathrm{Im}\sigma(\omega)/\mathrm{Re}\sigma(\omega) = R\omega c$. In the light of the observed dc nonlinearity[4-6] care has been taken to avoid nonlinear effects, the applied rf voltage was always less than that corresponding to the threshold field E_T.

Figure 1 shows the temperature dependence of R measured at various frequencies. The capacitance C is shown in Fig. 2. We observe a positive dielectric constant and strong frequency dependence in both CDW states, with a more pronounced effect in the low-temperature ($T < T_2$) state. The overall temperature dependence of the dielectric constant $\epsilon(\omega)$ and conductivity $\sigma(\omega)$ is strongly related to the observed nonlinear current-voltage characteristics.[4-6] In the low-temperature phase the threshold field E_T of the onset of nonlinearity has a minimum around 42 K; we observe the largest dielectric constant and strongest frequency dependence at this temperature. Also, E_T is about a factor of 5 larger in the high-temperature ($T_2 < T < T_1$) phase, and we observe a smaller dielectric constant and weaker frequency dependence here. These observations are in broad agreement with that expected for a pinned CDW.[10-12] The onset of nonlinear conductivity is related to the pinning forces, provided by the impurities;[13] these restoring forces are also responsible for a positive dielectric constant. It is expected therefore that a small critical field is accompanied by a large dielectric constant as observed. Also while various models lead to somewhat different relations between the frequency- and field-dependent transport of a CDW condensate, all suggest that strongly nonlinear behavior is coupled to strong frequency dependence, a feature also observed here.

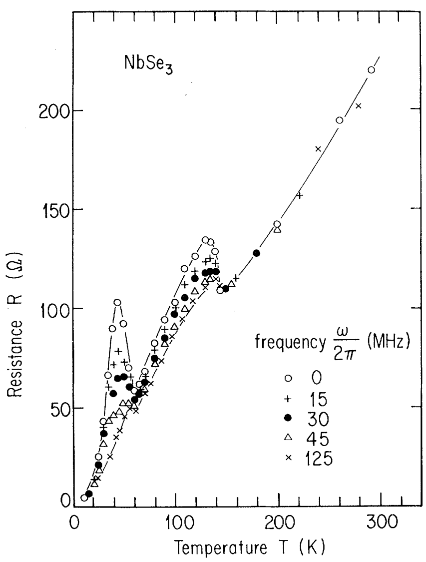

FIG. 1. Temperature dependence of the sample resistance R at various frequencies. The experimental data at 250 MHz follow closely those measured at 125 MHz and are omitted from the figure.

The frequency dependence of Reσ and Imσ, measured at 42 K, is shown in Fig. 3. The conductivity smoothly increases from the dc limit and saturates above 100 MHz at the value observed and also at microwave frequencies. At low frequencies (below 100 MHz) the observed frequency dependence closely follows the measurements by Longcor and Portis.[14] The dielectric constant $\epsilon = \mathrm{Im}\sigma/\omega$ is independent of frequency below about 50 MHz, and using the relation $\epsilon = 2\pi(1.8)10^{12}(RC) \times \mathrm{Re}\sigma$, we obtain $\epsilon = 2\times10^8$, an enormous value when compared with values measured in other highly anisotropic conductors, tetrathiafulvalinium-tetracyanoquinodimethane (TTF-TCNQ) ($\epsilon \sim 10^4$)[15] and $K_2Pt(CN)_4Ba_{0.3}\cdot 3H_2O$ (KCP) ($\epsilon \sim 5\times10^3$)[16] where incommensurate CDW's are believed to have a significant contribution to the dielectric response.

The classical model of a pinned CDW condensate in an anisotropic metal[17] leads to, in the absence of interaction between the CDW and normal electrons, two separate contributions to the conductivity. The normal (unnested) electrons lead to a frequency-independent background at frequencies smaller than the bandwidth, while the pinned mode shows up in a strongly peaked Re$\sigma(\omega)$

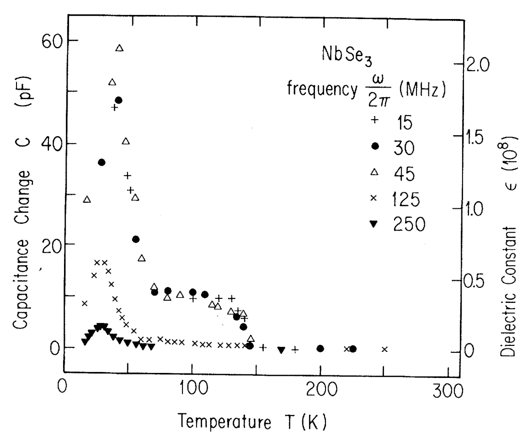

FIG. 2. Temperature dependence of the capacitance C at various frequencies.

around the pinning frequency ω_p, with the concomitant zero crossing of the dielectric constant at ω_p. Evidence for this feature comes from far-infrared studies in (TTF-TCNQ)[17] and KCP.[15] The frequency dependence shown in Fig. 3 is inconsistent with such a simple model of the pinned CDW; we do not see any evidence for a maximum of $\mathrm{Re}\sigma(\omega)$, and the dielectric constant remains positive in the whole measured temperature range.

$NbSe_3$ differs in many ways from that of a one-dimensional CDW state. The main difference perhaps is that the material retains parts of the Fermi surface in the CDW phase[3] and both the normal (unnested) and condensed electrons may contribute to the conductivity. The normal electrons lead to a frequency-independent conductivity at low frequencies covered by our experiment, and $\sigma(\omega)$ reflects the frequency-dependent transport due to electrons in the CDW state. We suggest that the pinned CDW condensate can be represented by a resistance (determined by the friction of the *unpinned* CDW) and by a capacitance which represents the pinning forces which act on the CDW. The normal electrons have only resistive components. This leads to a resistive-capacitive network shown in Fig. 3, where R_C and R_N describe the contribution of the condensed and normal electrons. This phenomenological description leads to the appropriate frequency dependence, $\mathrm{Re}\sigma(\omega) = R_N$, at low frequencies, and the CDW does not contribute to the conductivity. At high frequencies $\mathrm{Re}\sigma(\omega) = R_N^{-1} + R_C^{-1}$, and we recover the joint contribution of the normal and condensed electrons. The conductivity rises smoothly from the dc to the high-frequency limit, and $\mathrm{Im}\sigma(\omega)$ displays a maximum in the crossover frequency, $\omega \sim 1/C(R_N + R_S)$. The above network model, however, leads to a crossover between the low-frequency and high-frequency limits in a narrow frequency range, and to arrive at the smooth crossover (from 1 MHz to 100 MHz) a broad distribution of components, representing the distribution of pinning energies and of the length of the CDW segments, is required.

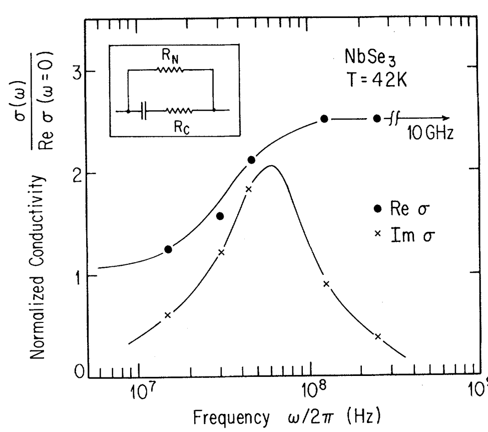

FIG. 3. Frequency dependence of Re $\sigma(\omega)$ and Im $\sigma(\omega)$ at $T = 42$ K. The microwave conductivity data are taken from Ref. 10.

We emphasize that, while the above network model incorporates the basic features of a pinned CDW (i.e., friction and restoring force), the relation to the underlying microscopic description of the CDW condensate is unclear. The model predicts a frequency-dependent transport closely analogous to a description in terms of overdamped harmonic oscillators[14] where, with appropriate distribution of characteristic lengths of the CDW segments, both the frequency and field dependence can be accounted for.

The temperature variation of the observed frequency dependence, in particular the strong maximum of the dielectric constant at temperatures near to the resistivity maximum below T_2, remains an unsettled question. The problem is closely analogous to the observation of minimum threshold electric field E_T at this temperature. Both features may signal a temperature-dependent scattering between the normal electrons and electrons in the CDW condensate[2] or a crossover from a temperature region where individual CDW segments have different field- and frequency-dependent response to a low-temperature limit where large regions of the CDW condensate exhibit collective motion.[7]

Finally we comment on the effect of combined application of dc and rf excitation in the nonlinear region. In experiments to be reported elsewhere we find that the application of a large dc electric field leads to a suppression of the frequency dependence and $\sigma(\omega)$ follows the temperature dependence of $\sigma(0)$ observed at large dc currents.[4,5] It also leads to the suppression of the capacitive component, showing that in the high-dc-field limit the sample can be represented by a resistive component only. This is just what is expected of pinned CDW which are depinned by an electric field. The effect of large-amplitude rf excitation on the dc conductivity is more complicated: with low-frequency ($f < 1$ MHz) excitation large dc nonlinearity can be induced, while at high ($f > 100$ MHz) frequencies the rf excitation has no effect on the dc transport.

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Multiple-$\vec{q}$ Structure or Coexistence of Different Magnetic Phases in $CeAl_2$?

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Neutron diffraction experiments under perturbations, such as uniaxial stress and hydrostatic pressure, suggest strongly that $CeAl_2$ exhibits two magnetic phases: a single-$\vec{q}$ collinear modulated structure and a type-II antiferromagnetic structure.

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The intermetallic Laves-phase compound $CeAl_2$ exhibits sinusoidally modulated magnetic structure,[1,2] which implies a moment reduction attributed to a Kondo-like coupling of the cerium $4f$ electrons to the conduction band. This structure develops with a propagation vector $\vec{q}_i = (\frac{1}{2}+\tau, \frac{1}{2}-\tau, \frac{1}{2})$; τ is found to be temperature independent and its value is $\tau = 0.110$ ($\pm$ 0.002).[2] The Fourier component of the magnetization $\vec{m}_{\vec{q}_i}$ is parallel to the [111] direction. Because of the symmetry of paramagnetic space group ($Fd3m$), there are twenty-four nonequivalent $\vec{q}_i$ vectors in the Brillouin zone (see Fig. 1) which leads to the coexistence of twenty-four Fourier components $\vec{m}_{\vec{q}_i}$. Orig-

