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Charge-density-wave magnetodynamics in $NbSe_3$

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We report complex ac conductivity measurements of the charge-density-wave (CDW) conductor $NbSe_3$ in applied magnetic (H) fields to 75 kG. Increasing H results in an increase in the characteristic crossover frequency, and an apparent increase in the CDW carrier concentration. A corresponding decrease in the dc threshold field E_T with increasing H suggests a breakdown of the conventional impurity-pinning mechanism in intense magnetic fields.

Niobium triselenide $NbSe_3$ is one of several quasi-one-dimensional materials which display charge-density-wave (CDW) conduction.[1] $NbSe_3$ is unique in that when it undergoes the Peierls distortion to the CDW state, only a portion of the Fermi surface is destroyed, leading to pseudometallic, rather than semiconducting, behavior below the transition. This behavior refers to the normal carriers, i.e., those carriers whose states on the Fermi surface are unaffected by the transition, and which act as a conduction mechanism "in parallel" with the CDW condensate.

Previous work[2] on $NbSe_3$ has demonstrated unusual magnetoresistance effects in the lower $(T < 59 \text{ K})$ CDW state. Magnetic (H) fields oriented perpendicular to the chain (b) axis of the crystal enhance the Ohmic resistivity anomaly and shift the maximum in the anomaly to lower temperature. The strong magnetoresistance has been suggested[3] to result from a "one-dimensionalization" of the band structure by the magnetic field, leading to an effective conversion of normal to condensed electrons. Such a process is consistent with the observation that the electric-field- (E) dependent dc conductivity $\sigma_{dc}(E)$ is independent of H in the saturated $E \rightarrow \infty$ limit.[2] Indeed, recent experiments[4] on the narrow-band noise spectrum of $NbSe_3$ have clearly demonstrated H-induced enhancement of the CDW carrier concentration n_c.

In this Rapid Communication, we report on complex ac conductivity $\sigma(\omega)$ measurements of $NbSe_3$ in the lower CDW state, in the frequency range 4 MHz–1 GHz, and for applied H fields 0–75 kG. Our results indicate that, in the presence of magnetic field, $\sigma(\omega)$ remains overdamped, and the conductivity lost at low frequency due to the magnetoresistance is fully recovered in the high-frequency limit. At both low and high frequencies, the dielectric response is consistent with an H-induced conversion of normal to CDW carriers. The characteristic ac "crossover frequency" ω_{co} increases with increasing H, whereas the dc threshold field E_T decreases with increasing H; this suggests a possible breakdown in the conventional impurity-pinning mechanism which dictates the dc and ac responses.

Our experiments were performed on single crystals of $NbSe_3$ grown by conventional vapor-transport methods, with typical minimum threshold fields $E_T = 30$ mV/cm at 48 K. The crystals were mounted in a two-probe configuration with silver paint contacts, and formed the termination of a microcoaxial cable. $\sigma(\omega)$ was measured parallel to the chain (b) axis, and H was applied perpendicular to b. Both the real and imaginary parts of $\sigma(\omega)$, $\text{Re}\sigma(\omega)$, and $\text{Im}\sigma(\omega)$ were determined by a computer-controlled network analyzer (Hewlett-Packard model No. 87454A), using an ac sinusoidal excitation amplitude more than two orders of magnitude smaller than E_T. Our experimental setup allowed simultaneous measurement of the electric-field-dependent dc conductivity $\sigma_{dc}(E)$, and observation of the narrow-band noise spectrum for $E > E_T$.

Figure 1 shows $\sigma(\omega)$ for $NbSe_3$ at 25 K, both with and without an applied H field. From the general behavior of $\sigma(\omega)$, it is apparent that the CDW continues to be overdamped in the presence of H. Important features of the data are found at the frequency extremes. In the low-frequency limit there is a large H-induced reduction in $\text{Re}\sigma$, consistent with the Ohmic dc magnetoresistance. In the high-frequency limit, $\text{Re}\sigma$ becomes H independent, and both $\text{Re}\sigma(\omega, H=0)$ and $\text{Re}\sigma(\omega, H=75 \text{ kG})$ appear to approach the same saturation value.[5] Simultaneous (pulsed) measurements of $\sigma_{dc}(E)$ confirmed that this saturation value corresponds to $\sigma_{dc}(E \rightarrow \infty)$, independent of H. The crossover frequency ω_{co} may be defined as that frequency for which $\text{Re}\sigma(\omega)$ has attained one half its maximum value. ω_{co} is identified for the two data sets

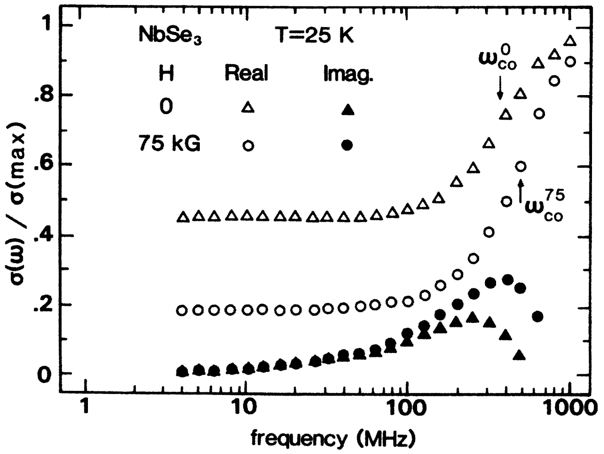

FIG. 1. Complex ac conductivity of $NbSe_3$ with and without a 75-kG H field. The crossover frequencies are indicated by vertical arrows.

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in Fig. 1. At 25 K, $\omega_{co}(H=0)/2\pi=350$ MHz, while $\omega_{co}(H=75\ \text{kG})/2\pi=480$ MHz, an increase over the $H=0$ value of approximately 37%. Figure 1 shows that $\text{Im}\sigma(\omega)$ is also affected by application of H. At low frequency no H-induced change was detected [within our experimental resolution of $\text{Im}\sigma(\omega)/\omega=\pm 1$ pF] in $\text{Im}\sigma$, while at high frequency $\text{Im}\sigma(\omega,H=75\ \text{kG})$ clearly exceeds $\text{Im}\sigma(\omega,H=0)$. At $\omega/2\pi=300$ MHz, for example, application of the $H=75$ kG field increases $\text{Im}\sigma$ by a factor of 1.7. In addition, the peak in $\text{Im}\sigma$, which corresponds roughly to the crossover frequency in $\text{Re}\sigma$, increases with increasing H.

Experimental data, such as that presented in Fig. 1, was obtained for the same NbSe$_3$ crystal at other temperatures in the lower CDW state. The same basic features described above were observed. Figure 2 shows the characteristic crossover frequency extracted from such data, as a function of temperature. It is apparent that $\omega_{co}(H=75\ \text{kG})$ always exceeds $\omega_{co}(H=0)$, with the relative increase becoming larger at lower temperature where the Ohmic magnetoresistance increases. Just below the transition temperature $T_P=59$ K, no appreciable difference was found in ω_{co} for $H=0$ and $H=75$ kG.

The main features of our experimental results appear consistent with an H-field-induced conversion of normal to CDW carriers. At low frequencies, only the normal carriers contribute to $\text{Re}\sigma$, and, as observed in Fig. 1, $\text{Re}\sigma$ is significantly reduced by application of H. At high frequencies, $\text{Re}\sigma$ has significant contributions from both CDW and normal carriers. Previous microwave and radio-frequency studies^6 have shown that in NbSe$_3$ the high-frequency conductivity attains a saturation maximum which corresponds to the (normal) conductivity expected in the absence of CDW formation. This indicates that, with $H=0$, the CDW carriers have the same high-frequency ac response as do the normal carriers, and the high-frequency conductivity thus depends only on the total carrier concentration $n=n_n+n_c$, with n_n the normal carrier concentration and n_c the CDW carrier concentration. In Fig. 1, the high-frequency conductivity $\text{Re}\sigma(\omega)$ is again observed to be equal to that expected in the absence of CDW formation [determined equivalently by extrapolating the Ohmic temperature-dependent resistance from high temperature to low, or by direct measurement of the dc conductivity $\sigma_{dc}(E\rightarrow\infty)$], indicating a conserved total carrier concentration in the presence of H, and an insensitivity to H of the high-frequency behavior of normal and CDW conduction states. For example, if the Ohmic dc magnetoresistance were strictly due to a normal-carrier mobility effect (assumed frequency independent), then $\text{Re}\sigma(\omega,H=0)$ and $\text{Re}\sigma(\omega,H=75\ \text{kG})$ in Fig. 1 would differ only by a frequency independent constant, in sharp contrast to the experimentally observed convergence in the data at high frequency.

A shift in carriers between normal and CDW states can also be inferred from $\text{Im}\sigma(\omega)$ in Fig. 1. Normal electrons do not contribute to $\text{Im}\sigma$ in the frequency range here being considered (well below the metallic plasma frequency), and hence $\text{Im}\sigma$ is dominated by the contribution from CDW carriers. As seen in Fig. 1, $\text{Im}\sigma$ increases with increasing H, in particular near the ac crossover frequency. As discussed below, the low-frequency behavior of $\text{Im}\sigma$ is also consistent with CDW carrier enhancement.

The increase in ω_{co} with increasing H, as shown in Fig. 2, is in striking contrast to the observed decrease in E_T. We have carefully measured $E_T(H)$, for the same NbSe$_3$ crystal as was used for the data of Figs. 1 and 2, by applying a pulsed dc conductivity technique, and by observation of the onset of narrow-band noise for applied steady-state dc fields. For any given H field, the two methods yielded identical values of E_T. The open circles in Fig. 3 show the ratio $E_T(H)/E_T(H=0)$ for NbSe$_3$ at selected H-field values and temperatures. For a given field H, the relative reduction in E_T becomes greater at lower temperature,

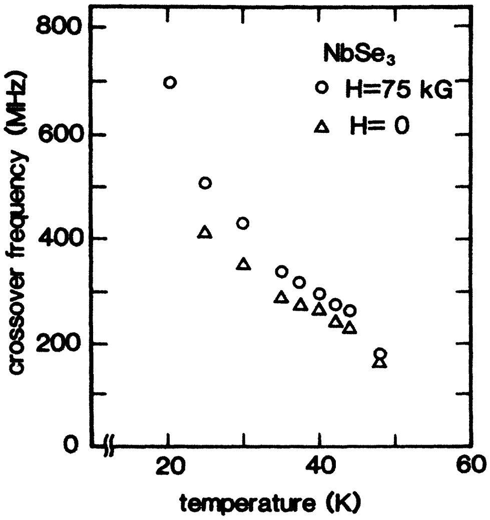

FIG. 2. Crossover frequency ω_{co} vs temperature in the lower CDW state of NbSe$_3$. At fixed temperature, an H field increases ω_{co}.

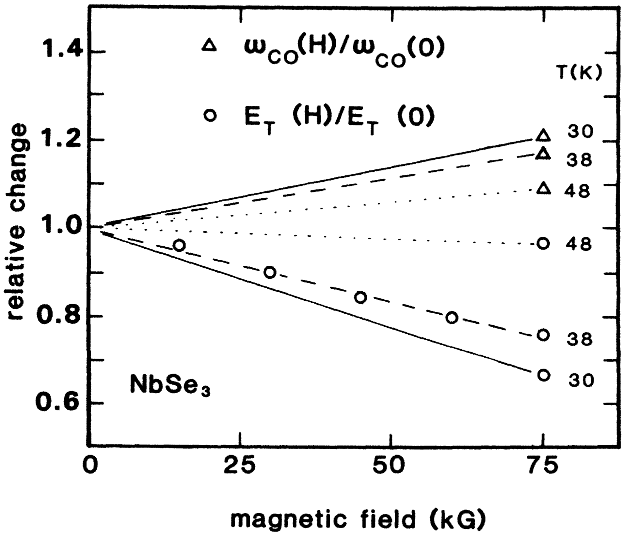

FIG. 3. Normalized crossover frequency and threshold electric field vs H in NbSe$_3$.

consistent with previous studies.[2] Figure 3 also contrasts the $E_T(H)$ behavior to that of $\omega_{\mathrm{co}}(H)$; the two parameters have dramatically opposite H-field dependences.

A number of models of CDW transport have been advanced, including rigid-particle classical motion,[7] quantum tunneling,[8] and deformable medium theories based on the Fukuyama-Lee-Rice Hamiltonian.[9] In a very simple classical description set forth by Grüner, Zawadowski, and Chaikin,[7] the CDW is treated as a charged rigid object in a periodic pinning potential. Although this model has some severe deficiencies, it allows us to readily identify important H-dependent CDW parameters. In the model, the limiting dc conductivity of the CDW condensate is given by $\sigma_{\mathrm{dc,CDW}}(E \rightarrow \infty) = n_c e^2 \tau_c / m^*$, while the ac conductivity is

$$\mathrm{Re}\,\sigma_{\mathrm{CDW}}(\omega) = (n_c e^2 \tau_c/m^*)[1+(\omega_0^2\tau_c/\omega)^2]^{-1} \ , \tag{1}$$

$$\mathrm{Im}\,\sigma_{\mathrm{CDW}}(\omega) = (n_c e^2 \tau_c/m^*)(\omega_0^2\tau_c/\omega)[1+(\omega_0^2\tau_c/\omega)^2]^{-1} \ , \tag{2}$$

where τ_c is the CDW characteristic scattering time, m^* is the effective mass of electrons condensed in the CDW state, and ω_0 is the CDW pinning frequency. $\omega_0^2\tau_c$ is identified with the ac crossover frequency. We assume the total sample conductivity to be given by $\sigma_{\mathrm{tot}} = \sigma_{\mathrm{CDW}} + \sigma_{\mathrm{normal}}$, where σ_{normal} represent the contribution of the normal carriers. Allowing for normal to condensed carrier conversion, the independence of σ_{tot} on H in the high-frequency or high electric field limit implies

$$n_c e^2 \tau_c/m^* + n_n e^2 \tau_n/m = n e^2 \tau_n/m = \mathrm{const} \ ,$$

where const here means an H-independent quantity. With τ_n, n, and m independent of H, we find $\tau_c/m^* = \mathrm{const}$, independent of H.

In the low-frequency limit, Eq. (2) yields a dielectric constant

$$\varepsilon = 4\pi \,\mathrm{Im}\,\sigma(\omega)/\omega = 4\pi n_c e^2/m^* \omega_0^2 \ . \tag{3}$$

Experimentally, we find no H-field effect on the very-low-frequency dielectric constant, hence $\omega_0^2 m^* \propto n_c$, or, equivalently, $m^*/\tau_c \propto n_c/\omega_0^2\tau_c$. With constant m^*/τ_c and the experimentally determined ratio

$$\omega_0^2\tau_c(H = 75\ \mathrm{kG})/\omega_0^2\tau_c(H=0) = 1.2$$

(measured at $T = 30$ K), we find

$$n_c(H = 75\ \mathrm{kG})/n_c(H=0) = 1.2$$

at $T = 30$ K. This H-induced increase in CDW carrier concentration is in good agreement with that determined from previous narrow-band noise studies,[4] which found $n_c(H = 75\ \mathrm{kG})/n_c(H=0) \approx 1.3$ at $T = 37.4$ K.

The above analysis unfortunately does not allow independent determination of $m^*(H)$, $\tau_c(H)$, and $\omega_0(H)$. In Bardeen's tunneling theory,[8] the dc and ac conductivities scale over a wide range of electric field and frequency, such that $\omega = 2\pi e^* LE/h$, with $e^*/e \approx m/m^*$ and L a CDW correlation length. Although the ac and dc conductivities for the NbSe$_3$ sample used for Fig. 1 did not show excellent scaling between $\sigma_{\mathrm{dc}}(E)$ and $\mathrm{Re}\,\sigma(\omega)$ at all temperatures investigated in the lower CDW state (although proper scaling was observed at selected temperatures), a rough correspondence between E and ω could always be established by matching $\mathrm{Re}\,\sigma(\omega)$ at the crossover frequency to $\sigma_{\mathrm{dc}}(E \rightarrow \infty)/2$, both with and without applied magnetic field. At $T = 30$ K, ω/E was found to correspond to 3 MHz/mV ($H = 0$), and $\omega/E = 3.1$ MHz/mV ($H = 75$ kG). Assuming L to be H independent, this leads to $m^*(H = 75\ \mathrm{kG})/m^*(H=0) = 0.97$. This implies that H does not appreciably change m^* (or τ_c), and hence the H-induced increase in $\omega_{\mathrm{co}} = \omega_0^2\tau_c$ is primarily due to an effective increase in ω_0, the CDW pinning frequency.

The increase in ω_0 with increasing H is in sharp contrast to the observed decrease in E_T with increasing H. Rigid particle, tunneling, and elastic medium models based on impurity pinning all predict that the ac and dc behaviors go hand in hand. For example, increases in impurity concentration generated by chemical doping or sample irradiation are predicted[10] to increase both ω_0 and E_T, and indeed this is observed experimentally in NbSe$_3$ and TaS$_3$.[10,11] In the context of impurity pinning (which clearly dominates at $H = 0$), our results would suggest that application of H effectively reduces "dc impurity pinning" but enhances "ac impurity pinning." We therefore conclude that important H-sensitive interactions, previously ignored by CDW theories, come into play when an H field is present in NbSe$_3$. Such an effect could have its origin in interactions between CDW carriers and normal electrons, or unusual CDW-phonon scattering at intermediate frequencies. Indeed, recent work on NbSe$_3$, TaS$_3$, and K$_{0.3}$MoO$_3$ have shown important couplings between the limiting CDW and normal carrier conductivities.[12] Whether these effects are related to the H-field phenomena reported here remains to be seen.

Finally, we remark that the only theory which discusses the anomalous magnetoresistance effects in NbSe$_3$, that of Balsiero and Falicov,[3] does not address the dynamics of the CDW condensate. An extension of this theory, which incorporates condensate magnetodynamics, may help clarify the role of impurity pinning in the presence of applied magnetic fields.

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