Magnetic-Field–Induced Carrier Conversion in a Charge-Density-Wave Conductor

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We have performed narrow-band noise and dc conductivity measurements on the charge-density-wave (CDW) conductor $NbSe_3$ in magnetic fields up to 75 kG. In the lower CDW state, the anomalous magnetoresistance is shown to result from a direct magnetic-field–induced conversion of normal to CDW carriers. The H and T dependence of the new CDW carrier concentration, and hence CDW order parameter, is explicitly determined.

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A growing number of synthetic inorganic quasi one-dimensional metals have been observed to be unstable towards $2k_F$ Peierls distortions, resulting in a charge-density-wave (CDW) ground state. Of particular interest in these systems is the dynamic nature of the CDW condensate itself, and a great deal of experimental and theoretical effort has been devoted to the understanding of the resulting collective-mode transport.[1] A simple Peierls distortion of the crystal lattice is associated (in a 1-D system) with a complete destruction of the Fermi surface (FS), through formation of a temperature-dependent gap in the (single-particle) excitation spectrum. Indeed, virtually all low-dimensional conductors which are known to display "sliding" CDW conduction undergo metal-insulator transitions. Below the transition temperature T_P, a semiconducting state obtains, with $T=0$ gaps on the order of $2\Delta_0 \approx 1000$ K.

A notable exception to this behavior is $NbSe_3$, which undergoes two CDW transitions ($T_{P1}=144$ K, $T_{P2}=59$ K), neither of which completely destroys the entire FS: $NbSe_3$ remains metallic (or semimetallic) at all temperatures.[2,3] Interestingly, $NbSe_3$ was the first material to display CDW conduction, and has been the most widely studied in terms of CDW dynamics. In general, the observed CDW behavior in $NbSe_3$ is qualitatively similar to that observed in true CDW semiconductors [e.g., TaS_3, $(TaSe_4)_2I$, $K_{0.3}MoO_3$], and hence in $NbSe_3$ the "normal" carriers (resulting from the remaining portions of the FS below T_P) are often assumed to represent a noninteracting parallel conduction mechanism.

Recently, Coleman *et al.*[4] reported unusual magnetoresistance effects in the lower CDW state ($T<59$ K) of $NbSe_3$. At the relatively high temperature of 25 K, for example, a relative increase in the resistivity, $[\rho(H)-\rho(0)]/\rho(0)$, on the order of 5 was observed at $H=227$ kG. Coleman *et al.* concluded that the large enhancement of the resistance anomaly was not caused by the dynamics of the CDW, but rather by the effect of the magnetic field on either the number or the mobility of the normal electrons. They further speculated that such a strong coupling between carriers and the applied magnetic field implies a spin-density-wave (SDW) component to the density-wave ground state.

Subsequent work by Balseiro and Falicov[5] has demonstrated that strong magnetic fields may lead to a destruction of electron-hole pockets in imperfectly nested anisotropic conductors, leading to an H-enhanced gap at the Fermi level for a system with an intrinsically stable density-wave ground state (either CDW or SDW). The mechanism of Balseiro and Falicov (which includes the effects of band broadening and tunneling between bands) would imply that, in $NbSe_3$ the field H induces a direct conversion of normal carriers to CDW (condensed) carriers, thus effectively modifying the CDW order parameter Δ.

We have performed careful noise and resistivity measurements on $NbSe_3$ in magnetic fields up to 75 kG, in an attempt to observe directly a possible H-induced increase in CDW carrier concentration n_c. We find H-induced changes in the narrow-band noise spectrum which indicate clearly that n_c increases with increasing H, the effect becoming greater at lower temperatures where the magnetoresistance increases. The observed changes in n_c correlate well to the percentage of FS destroyed by the magnetic field, as determined by the Ohmic and nonlinear dc conductivity.

Single crystals of $NbSe_3$ were mounted in a two-probe configuration with the chain (b) axis of the crystal perpendicular to the field H. A dc current was applied through the sample, and the voltage across the sample was amplified and detected with either a dc voltmeter or a high-frequency spectrum analyzer. For applied electric fields E exceeding the threshold E_T for the onset of CDW conduction, and with $H=0$, a clear narrow-band noise spectrum was observed, with a dominant fundamental frequency peak and numerous higher harmonics. Application of magnetic fields up to 75 kG were found to have no marked effect on the amplitude or quality of the narrow-band noise spectrum, but, for fixed dc bias, the fundamental noise

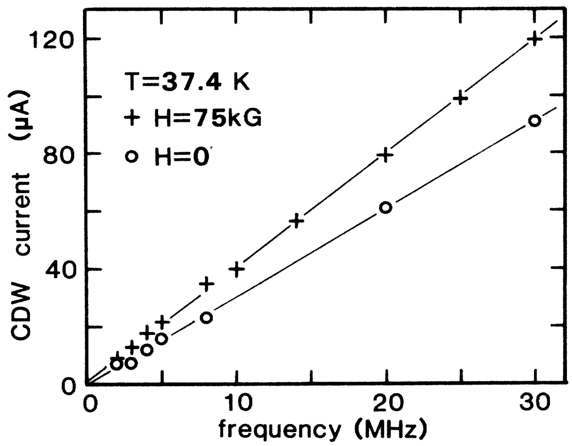

FIG. 1. I_{CDW} vs narrow-band noise frequency in $NbSe_3$ with and without an applied magnetic field.

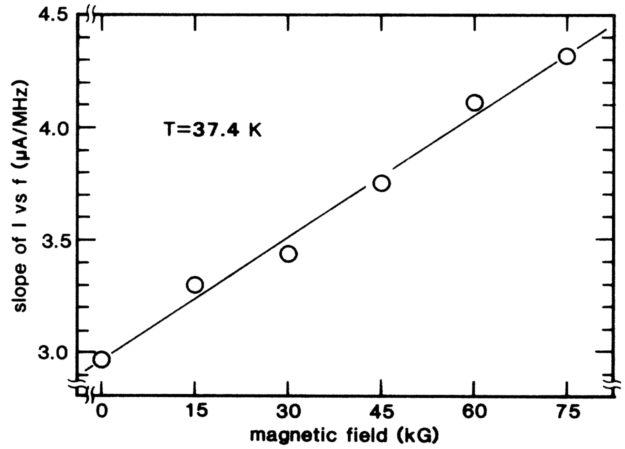

FIG. 2. $I_{CDW}/f \approx n_c$ vs H in $NbSe_3$. An increasing magnetic field results in an increase in CDW carrier concentration. Up to $H=75$ kG, the carrier conversion rate is linear in H.

frequency f was highly magnetic-field dependent.

In a simple model,[6,7] the narrow-band noise frequency is related to the excess CDW current I_{CDW} through

$$I_{CDW} = n_c e v_d A = n_c e f \lambda A, \qquad (1)$$

where $I_{CDW} = I - V/R_0$ with I the total sample current, V the time-averaged sample voltage, and R_0 the low-field (Ohmic) sample resistance. v_d is the CDW drift velocity, A the sample cross-sectional area, and λ a constant, usually taken to be the CDW wavelength. Hence for fixed n_c, I_{CDW} is directly proportional to f, and the ratio I_{CDW}/f reflects directly n_c.

Figure 1 shows I_{CDW} vs f for $NbSe_3$ at $T=37.4$ K, for $H=0$ and for an applied field $H=75$ kG. For $H=0$, the linear relationship is in accord with Eq. (1) and consistent with previous narrow-band noise studies in $NbSe_3$.[7-9] With $H=75$ kG, a linear dependence of f on I_{CDW} is again observed, but with a different slope. Figure 1 demonstrates clearly that the ratio I_{CDW}/f is *increased* in the presence of an applied magnetic field. With e, λ, and A independent of H in Eq. (1), Fig. 1 demonstrates that *the effect of a magnetic field in* $NbSe_3$ *is to directly enhance the CDW carrier concentration.* At 37 K and $H=75$ kG the increase in CDW carrier concentration is approximately 30% over the $H=0$ value, a fairly dramatic change. As we show below, the magnetic-field–induced carrier conversion (normal to CDW electrons) appears to account entirely for the spectacular (low electric field) magnetoresistance in $NbSe_3$, and hence the present experiments rule out normal-carrier–mobility effects as being the source of the magnetoresistance.

Data such as those shown in Fig. 1 were taken at various values of magnetic field strength and temperature in the lower CDW state. Figure 2 shows the slope I_{CDW}/f vs H for T fixed at 37.4 K. Up to $H=75$ kG, a virtually linear dependence of I_{CDW}/f (and hence n_c) on H is obtained, with $d(I_{CDW}/f)/dH = 1.8\times10^{-2}$ μA/MHz·kG. With $\lambda = 14$ Å, this corresponds to a magnetic-field–induced carrier conversion rate of $dn_c(H)/dH = 3.6\times10^{18}$ carriers/kG·cm^3.

Figure 3 shows the relative increase in CDW carrier concentration, induced by an $H=75$ kG field, as a function of temperature in the lower CDW state of $NbSe_3$. Near the CDW transition temperature $T_P=59$ K, no increase in n_c is observed, while $n_c(H=75\text{ kG})/n_c(H=0)$ increases substantially with decreasing temperature. Figure 3 effectively defines the new CDW order parameter $\Delta(H,T)$. Previous studies[8,9]

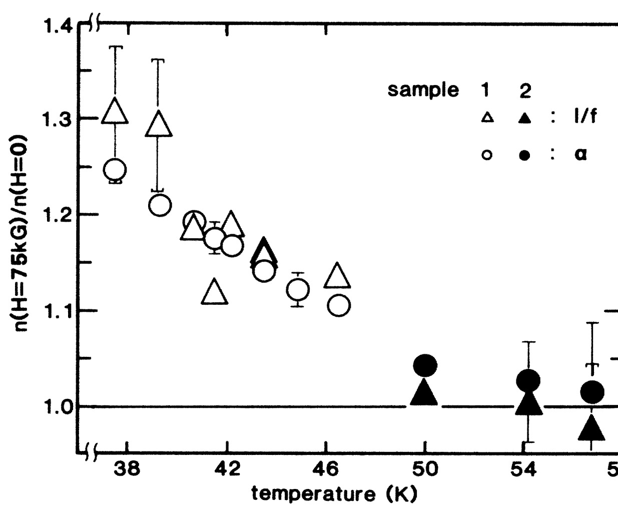

FIG. 3. Relative increase in CDW carrier concentration due to a 75-kG field in $NbSe_3$, as a function of temperature. The triangles refer to n_c determined directly from noise spectra, while the circles refer to the fraction of Fermi surface destroyed (see text).

have shown that the temperature dependence of $\Delta(H=0,T)$ is borne out directly in measurements of $I_{CDW}/f \approx n_c$. The CDW carrier concentration is related to the order parameter by[10]

$$n_c(T)/n_c(T=0) \approx \Delta(T), \quad (2)$$

valid for T close to T_P. For $T << T_P$, $\Delta(T)$ may again be related to $n_c(T)$,[10] and, as in superconductors, at $T=0$, $n_c(T)$ equals unity; i.e., all carriers are condensed in the CDW state, *if* the system displays perfect Fermi surface nesting.

The magnetic-field enhancement of n_c (and hence the enhancement of the CDW order parameter) shown in Fig. 3 for the narrow-band noise study may be compared to the percentage of FS removed by the CDW transition and the applied field H. In addition to measurements of the narrow-band noise just described, we have performed pulsed low- and high-electric-field I-V studies on $NbSe_3$ in the presence and absence of magnetic fields. Consistent with the results of Coleman *et al.*,[4] we find that although the low-field (Ohmic) resistance R_0 of the crystal is strongly influenced by H, the high-electric-field (saturated) resistance R_{sat} is independent of H. In a simple approximation, the fraction of FS removed by the CDW transition and H field may be expressed as[11]

$$\alpha(H,T) = \sigma_c(H,T)/[\sigma_0(H,T)+\sigma_c(H,T)], \quad (3)$$

with

$$\sigma_0(H,T) = 1/R_0(H,T),$$

$$\sigma_c(H,T) = [R_{sat}(H,T)]^{-1} - \sigma_0(H,T).$$

With $H=0$, approximately 60% of the remaining FS is destroyed by CDW formation at $T_P = 59$ K; with $H = 75$ kG this percentage is significantly greater, and temperature dependent. The circles in Fig. 3 represent the fractional increase in α due to application of a 75-kG magnetic field. The ratio $\alpha(H=75\ \text{kG},T)/\alpha(H=0,T)$ is seen to be in good agreement with carrier concentration increases deduced from the narrow-band noise studies. It should, however, be noted that Eq. (3) represents only a crude approximation to the fraction of FS removed; a more quantitative evaluation of α would require detailed information.

In conclusion, we find that magnetoresistance effects in $NbSe_3$ are well accounted for by a straightforward enhancement in the CDW order parameter $\Delta(H,T)$ induced by the applied magnetic field, consistent with the theoretical predictions of Balseiro and Falicov.[5] A detailed comparison of our results for $\Delta(H,T)$ with this theory would necessitate a comprehensive knowledge of the semimetallic band structure of $NbSe_3$.

Finally, we note that we have also performed extensive complex ac conductivity studies of $NbSe_3$ in applied magnetic fields up to $H = 80$ kG.[12] Those studies indicate again a direct conversion of normal to CDW carriers, resulting in an enhanced complex dielectric response, consistent with the dc transport and noise studies described here.

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