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Phase transition induced by a magnetic field in $NbSe_3$

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Charge-density-wave transport in $NbSe_3$ is shown to be greatly affected by the application of a magnetic field. A (T,H) transition line can be drawn. The threshold electric field above which the charge-density-wave moves is strongly reduced when the magnetic field, applied perpendicular to the (b,c) plane, is increased beyond the critical line. Such an effect does not occur when the magnetic field is parallel to the (b,c) plane. At helium temperature the reduction of the threshold electric field occurs beyond a threshold magnetic field of 2.5 T.

The study of magnetic-field-induced transitions in materials with restricted dimensionality has been largely investigated in the last few years. A charge-density-wave (CDW) transition has been shown to occur in graphite[1] at high magnetic field, H, and low temperature and explained in the general context of electron-hole condensation of a two-dimensional electron gas.[2] Instability of the two-dimensional open orbit metal against spin-density-wave (SDW) formation[3] in the presence of a magnetic field has been recently found in the bis-tetramethyltetraselenafulvalene-X [$(TMTSF)_2X$] family of organic transfer salts. The SDW appears with a complex cascade of subphases.[4] However, only a few measurements have been performed on systems which already at $H=0$ undergo a CDW transition.

A phase transition has been observed at 0.35 T in the two-dimensional layered compound $2H$-TaS_2 by the method of time-differential perturbed angular correlation on the Ta nuclear quadrupole.[5] The phasing of the CDW relative to the lattice has been shown to be modified by the application of H indicating the coexistence of a CDW with a SDW.

Recently, magnetoresistance measurements of $NbSe_3$ have been reported.[6] $NbSe_3$ undergoes two CDW transitions respectively at $T_1=145$ K and $T_2=59$ K but however remains metallic below T_2. This metallic behavior indicates that the Fermi surface (FS) has not been totally destroyed by both CDW's probably because of the lack of perfect nesting between portions of the FS connected by distortion wave vectors. Below T_2 a large magnetoresistance has been measured; $\Delta\rho(H)/\rho$ shows a maximum in the range of $T\sim 20$–25 K at $H=20$ T. Moreover, T_2 has been found to be nearly independent of H up to 23 T. Balseiro and Falicov[7] have tentatively explained the enhancement of the resistivity anomaly[6] below T_2 by an improvement, resulting from the application of H, of the imperfect nesting of the FS, leading to a modification of the FS area left after CDW condensations and consequently increasing the CDW gap and the number of electrons in the CDW condensate. Moreover, $NbSe_3$ is the prototype of pseudo one-dimensional conductors exhibiting collective transport properties when an electric field larger than a threshold value, E_c, is applied.[8] This extra conductivity is ascribed to the Fröhlich-type conductivity when the CDW is depinned from impurity centers and moves through the crystal. It has been also shown that E_c is strongly reduced under H at helium temperature.[9] In this Rapid Communication we report on measurements of the threshold field E_c as a function of temperature when H is applied perpendicular to the chain direction. We show that the reduction of E_c only occurs when H is applied perpendicular to the more conducting plane. E_c is only reduced when a critical line $T(H)$ is crossed. At helium temperature a threshold magnetic field of 2.5 T is needed for detecting a variation on the value of E_c.

$NbSe_3$ has a monoclinic unit cell and crystallizes in ribbon form. The plane of the ribbon is the (b,c) plane which is the more conducting plane because van der Waals bondings between sheets occur along a^*. Great care has been used for selecting perfect single crystals. So we have measured the variation of the resistance of the specimen at helium temperature as a function of angle at H fixed. In this angular variation, orientations of **c**, **a**, and $\mathbf{a}^*$ axes are detected.[10] H was produced by a 10-MW Bitter magnet at the Service National des Champs Intenses at Grenoble. The samples used in this investigation were chosen from different batches and have a resistance ratio between room temperature and helium temperature in the range of 50 to 150. E_c is measured by recording the variation of the differential resistance dV/dI (at frequency of 33 Hz) as a function either of the applied dc current or of the applied dc voltage. Nonlinearity occurs at the threshold value[11] I_c or V_c. E_c is defined as RI_c/l or V_c/l with R the Ohmic resistance of the specimen and l its length, typically 1 mm. E_c is also measured by the onset of noise generation when the CDW starts to move. This noise is detected by a lock-in amplifier used as an ac voltmeter. For $H=0$ the temperature is measured with a germanium and a platinum resistance. The temperature regulation is achieved with a ceramic capacitance thermometer which has a negligible magnetocapacitance in the field range used. The temperature could be kept stable to ± 0.1 K for a period of 1 h. For a good thermal exchange experiments have been performed with the sample immersed in cryogenic liquids as hydrogen or neon or within a pressurized helium atmosphere (20 bars at room temperature).

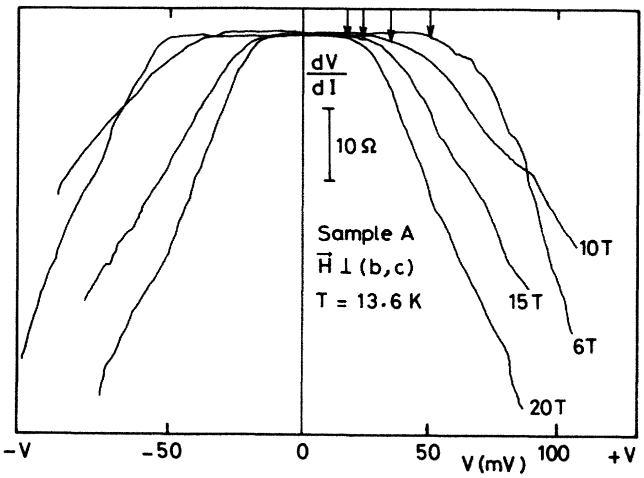

FIG. 1. Variation of the differential resistance dV/dI as a function of the dc applied voltage at $T=13.6$ K for fixed values of the magnetic field, H, applied perpendicular to the (b,c) plane of a $NbSe_3$ specimen (sample A). The arrows indicate the threshold voltage.

The variation of dV/dI as a function of the applied dc voltage for sample A at $T=13.6$ K is drawn in Fig. 1 for fixed values of H applied perpendiuclar to (b,c). V_c (indicated by an arrow) is largely reduced when H is increased. The variation of E_c with temperature for sample A is plotted in Fig. 2 for $H=0$ and $H=20$ T. For $H=0$, E_c strongly increases when T is reduced which is commonly observed[8,12] although poorly understood. For $H=20$ T, E_c is independent of H at high temperature but strongly decreases below 20 K. The inset of Fig. 2 shows similar results for sample B at $H=0$, 10 T, and 20 T. The electrical behavior of this particular sample can be ascribed to so-called "switching" samples[13-15] for which two threshold fields have to be defined:[13] the first one, E_c, at which noise first occurs without a significant decrease in dV/dI and the second one at which dV/dI exhibits a strong discontinuity. We have found that this latter threshold, independent of T[14], is also independent of H while the smaller threshold follows the variation plotted in the inset. E_c is reduced below 20 K for $H=20$ T and below 18 K for $H=10$ T. Moreover, the effect of H yields an enlargement of the extraconductivity for $E>E_c$. So the influence of H on the depinning process is quite general and is not restricted to specimens with particular impurities, probably acting as strong pinning impurities,[15] which exhibits "switching" properties.

We have performed measurements of dV/dI and of noise either at fixed H as a function of T or at fixed T as a function of H. The lower part of Fig. 3 shows the variation of E_c for sample B as a function of H applied perpendicular to (b,c) at fixed values of T. It is seen that E_c only decreases above a critical value of H. The upper part of Fig. 3 demonstrates the effect of the orientation of H on the variation of E_c at $T=1.8$ K for sample C. When H is applied parallel to (b,c), E_c is nearly independent of H. The reduction of E_c only occurs when H is perpendicular to (b,c) beyond a threshold magnetic field which at helium temperature is ~ 2.5 T. At the fixed value of $H=17$ T we have plotted the angular variation of E_C as a function of the angle θ between H and the plane (b,c). We have found[16] that, similar to the angular dependence of the anisotropic critical field H_{c2} for a two-dimensional su-

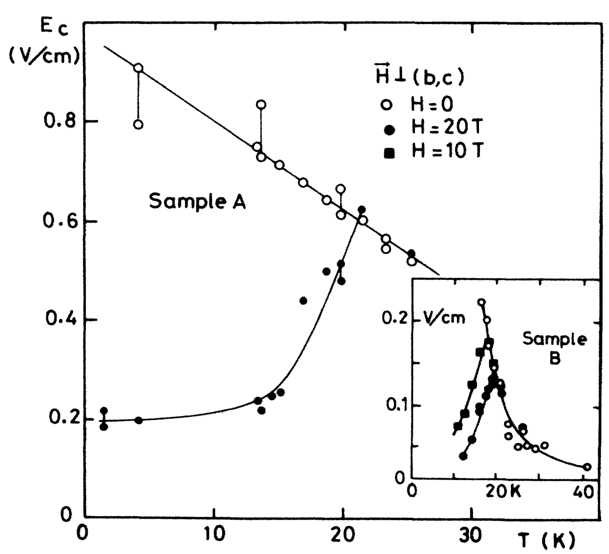

FIG. 2. Variation of the threshold electric field, E_c, as a function of temperature at $H=0$, and $H=20$ T for sample A. The inset shows the similar variation for sample B at $H=0$, 10, and 20 T.

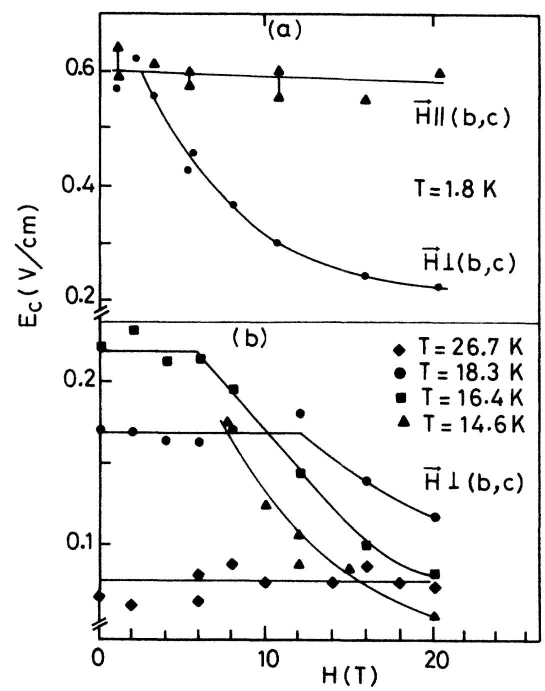

FIG. 3. Lower part: Variation of the threshold electric field, E_c, as a function of the magnetic field, H, perpendicular to the (b,c) plane of $NbSe_3$ (sample B) at fixed temperatures. Upper part: Variation of E_c at $T=1.8$ K as a function of the magnetic field applied parallel and perpendicular to the (b,c) plane of $NbSe_3$ (sample C).

perconductor, the angular variation of E_c fits relatively well with the relation

$$\frac{E_c[\mathbf{H}\parallel(b,c)]}{E_c[\mathbf{H}\perp(b,c)]}=(\cos^2\theta+\epsilon^2\sin^2\theta)^{-1/2}$$

with $\epsilon\sim3$ at $H=17$ T.

In Fig. 4 we have drawn the critical line $T(H)$ for observing the reduction of E_c by the application of H deduced from measurements on samples A, B, and C. The sharp variation of the critical line in the helium temperature range seems to indicate a first-order transition. The inset shows the variation of the linear magnetoresistance $\Delta R/R$ as a function of T for fixed values of H. We interpret this nonmonotonous behavior (not so pronounced as in Ref. 6) as resulting from a jump between two regular monotonic variations of magnetoresistance with different amplitudes. As usual the transition temperature has to be taken at the inflection point in the $\Delta R/R$ variation which is indicated by arrows in the inset. When reported on the (T,H) diagram these data fit well the critical line indicating the connection between anomalous properties in the linear state and observation of the influence of H on the depinning process.

As known, the magnetic field has a spin and an orbital effect. When H is applied, the electron band is split into two subbands with spin up and spin down separated by

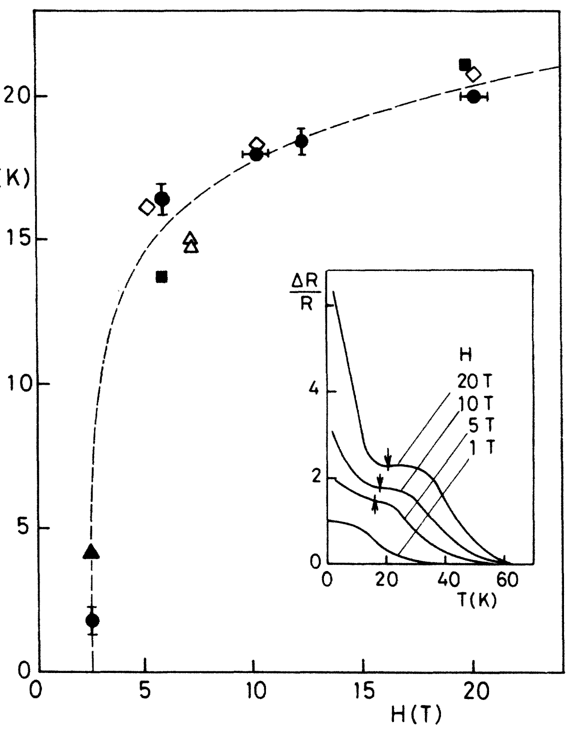

FIG. 4. Temperature T, as a function of the amplitude of the magnetic field, H, applied perpendicular to the (b,c) plane of $NbSe_3$ for the observation of a reduction of the threshold electric field (the line is only a guide for eye). Sample A (■); sample B, data from H fixed, T variable (⊢●⊣); T fixed, H variable (↕●); sample C, H fixed, T variable (△); T fixed, H variable (▲). The inset shows the variation of the linear magnetoresistance as a function of T at fixed values of H. Arrows indicate reflection points reported as ◇ in the $T(H)$ diagram.

$2\mu_B H$ with μ_B the Bohr magneton. It has been calculated that the Peierls transition temperature is reduced[17] by a quantity δT equal to $\gamma(\mu_B H)^2/4k_B T_c$ with $\gamma\sim1$ and k_B the Boltzmann constant. As a consequence of the band splitting, electrons now have different Fermi momentum: $2k_F$ for spin up being different from $2k_F$ for spin down. Such a situation of two kinds of bands with parallel distortion wave vectors has already been theoretically studied.[18] Moreover, in relation with properties of orthorhombic TaS_3,[8] it has been suggested that the variation with temperature of the longitudinal distortion wave vector and its locking to a nearly commensurate value near $T=130$ K results from coupling between the CDW's on two sets of chains with slightly different distortion vectors.[19] At the locking between CDW's, the threshold field E_c or orthorhombic TaS_3 shows a large decrease,[8] exactly similar to the variation shown in Fig. 2 for $NbSe_3$ under a magnetic field. This problem has been recently studied[20] in the limit of weak-bilinear coupling between CDW's on different chains leading to a possibility of a soliton lattice in the CDW phase. So the decrease of E_c might result from a doubly periodic superstructure induced by the magnetic field which would be more easily depinned as the CDW itself. However, while a reduction of the Peierls transition temperature of tetrathiafulvalene-tetracyanoquinodimethane has been measured[17] with modest values of H ($H\sim5$ T), T_2 in $NbSe_3$ is nearly not affected by H up to 23 T when H is applied perpendicular to the chain axis as reported by Coleman *et al.*[6] and confirmed in our study.[21] A reason for such a fact is probably the role of spin-orbit scattering which destroys the spin-up and spin-down states as good eigenstates for spin. The spin-orbit scattering time is also proportional to the atomic number and therefore must be much larger for transition-metal atoms as for elements constituting organic materials. It has to be noted that in layered two-dimensional compounds the critical field H_{c2} is not affected by the paramagnetic effect and it has been shown that the suppression of the paramagnetic effect is due to spin-orbit interaction.[22]

The anisotropy of E_c with the orientation of the magnetic field perpendicular to the chain axis as the angular dependence of E_c, similar to that of an anisotropic type-II superconductor, seems to favor the role of orbital effect of the magnetic field on the spin susceptibility in the CDW condensate. We speculate that the nesting vector varies with H along the critical line. The change in the nesting conditions under H is also detected in the linear magnetoresistance and will provide the nonmonotonous variation of $\Delta R/R$ as shown in the inset of Fig. 4. Observation by neutron scattering of any change in the superlattice reflections as eventual observation of a double superstructure resulting from a soliton lattice generation under magnetic field would be very useful for further developments.

In conclusion, we have shown that the threshold electric field for depinning the CDW in $NbSe_3$ is strongly reduced only when H is applied perpendicular to the (b,c) plane and when a critical line $T(H)$ is crossed. At helium temperature this effect occurs when H is beyond a threshold magnetic field of 2.5 T. We estimate that this transition results from a change in the nesting conditions when H is applied.

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