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Nonlinear magnetoresistance and charge-density-wave depinning at liquid-helium temperatures in $NbSe_3$

J. Richard, P. Monceau, and M. Renard

Centre de Recherches sur les Très Basses Températures, Centre National de la Recherche Scientifique, Boîte Postale No. 166 X, 38042 Grenoble Cédex, France

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The transverse magnetoresistance of $NbSe_3$ has been measured at liquid-helium temperatures as a function of the dc bias. The amplitude of the Shubnikov–de Haas oscillations is greatly suppressed by the electric field before the charge-density wave becomes depinned. As soon as the nonlinear state has been reached, the charge-density wave remains in a metastable state even after the electric field is reduced to zero. These metastable states give rise to a huge increase of the magnetoresistance, a shift of the extrema of the Shubnikov–de Haas oscillations, and a decrease of the Fermi-surface cross sections.

Two charge-density-wave (CDW) phases occur in $NbSe_3$ at $T_1=145$ K and $T_2=59$ K. Among quasi-one-dimensional conductors exhibiting collective nonlinear transport properties at any temperature below the Peierls transition,[1] $NbSe_3$ is the unique compound which remains metallic at low temperature.[2] The electrical conductivity increases when the applied electric field exceeds a threshold value E_T. This extraconductivity is ascribed to the Fröhlich-type conductivity which occurs when the CDW is depinned from impurity centers and moves through the crystal. The value of E_T, corresponding to the CDW phase below T_2, is minimum near 42 K and strongly increases when T is reduced. Because of heating problems, E_T cannot be measured below ~ 20 K. The metallic behavior at low temperature indicates that the Fermi surface (FS) has not been totally destroyed by both successive CDW's probably because of the lack of perfect nesting between portions of the FS connected by the distortion wave vector. Quantum oscillations in the magnetoresistance at liquid-helium temperature have revealed a rather simple FS.[3–5] If the surface is approximated by an ellipsoid, the volume occupied by this ellipsoid is about 0.1% of the Brillouin zone leading to a carrier concentration of $\sim 10^{18}$ cm^{-3}, which is close to the number obtained from the Hall constant,[6] namely, 3×10^{18} cm^{-3}.

Recently, the effect of a magnetic field on the linear and nonlinear properties of $NbSe_3$ has been investigated. The large magnetoresistance below T_2 has been tentatively ascribed[7] to an improvement of the nesting of the FS leading to an enhanced CDW gap.[8] An increase of 30% in CDW carrier concentration at $T=37$ K and $H=7$ T has been recently reported,[9] but similar experiments with H up to 20 T have not revealed such an effect.[10] Coleman, Everson, Eiserman, and Johnson[11] have also shown that E_T is strongly reduced at liquid-helium temperature when H is applied. The variation of the electrical resistance as a function of E was found to be bell shaped and not to exhibit a sharp variation when $E \gtrsim E_T$. Fits to the Zener tunneling model lead to a very small or nearly zero value of E_T. Finally, transitions to new metastable states have been measured for temperatures in the range of 1.2–4.2 K when the product of the current and the magnetic field is larger than a critical value.[12] The corresponding critical electric fields are found to be much higher than those required to depin the CDW. Moreover, these transitions have a strong effect on the Shubnikov–de Haas oscillations.[12] Here we report on resistivity measurements of $NbSe_3$ between 1.8 and 4.2 K as a function of electric field and magnetic field. We show that the amplitudes of the Shubnikov–de Haas oscillations are strongly suppressed by the application of E and that the noise generation which is currently associated with the CDW depinning only occurs at a finite threshold electric field. When a dc current higher that the threshold value for depinning the CDW is applied and reduced to zero, the magnetoresistance is shown to be greatly increased; then, extrema of the Shubnikov–de Haas oscillations are shifted to lower magnetic field values and a decrease in the FS cross sections is also observed.

The differential resistance dV/dI is measured with four contacts made with silver paint by an ac bridge working at 33 Hz as a function of a superposed dc current. Voltage noise is detected with a lock-in amplifier used as an ac voltmeter. The length between voltage probes is typically 1 mm and the cross section of the samples between 10 and 50 μm^2. The resistance ratio between room temperature and liquid-helium temperature (RRR) is 80 for sample 1, 100 for sample 2, and 18 for sample 3. Measurements are only performed with H perpendicular to the chain b axis and up to 20 T. H is produced by a 10-MW Bitter magnet at the Service National des Champs Intenses at Grenoble. $NbSe_3$ has a monoclinic unit cell and crystallizes in the form of a ribbon. The plane of the ribbon is in the (b,c) plane. The position of this plane was located by rotating the specimen perpendicular to H and by measuring the angular variation of the dc magnetoresistance at a small (2–3 T) constant field. As shown in Fig. 1 of Ref. 4(b), the magnetoresistance shows a deep minimum at $\theta=0$ when H is parallel to the (b,c) plane; the angular position where H is perpendicular to the (b,c) plane $(\theta=\pi/2)$ is detected by a shallow minimum surrounded by equidistant maxima at $\theta=109°$ and $\theta=\pi-109°$, where 109° is the angle in the monoclinic unit cell between the c and a axes. Specimens which did not exhibit this angular dependence,

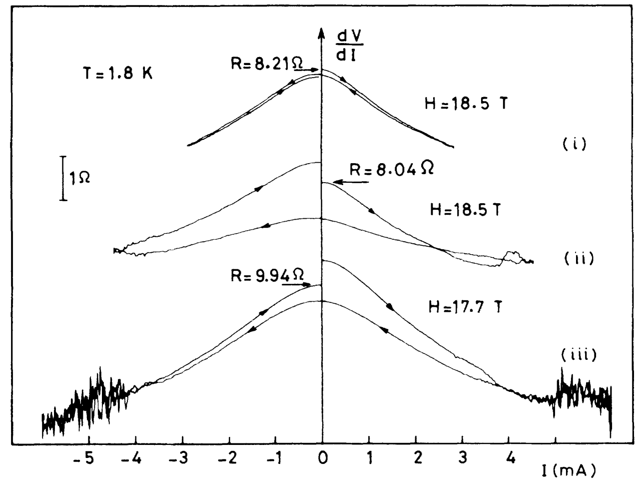

FIG. 2. Variation of the differential resistance dV/dI of $NbSe_3$ (sample 1) at $T=1.8$ K as a function of the applied current with H parallel to the (b,c) plane. (i) $H=18.5$ T and the maximum applied current (2.8 mA) does not exceed the threshold value. (ii) $H=18.5$ T and the CDW is depinned above 3.5 mA. (iii) $H=17.7$ T and with a larger current excursion.

the signature of single-crystal behavior, were rejected.

The variation of the linear dc magnetoresistance of sample 1 with $\mathbf{H}\parallel(b,c)$ and $\mathbf{H}\perp(b,c)$ at $T=1.8$ K is drawn in Fig. 1. Curves α correspond to the virgin state measured just after the sample has been cooled down from room temperature. The magnetoresistance exhibits the typical Shubnikov–de Haas oscillations, as already reported.[3-5] Figure 2(i) shows the variation of the differential resistance with $\mathbf{H}\parallel(b,c)$ at $H=18.5$ T as a function of the applied dc current up to a value of 2.8 mA. It can be seen that dV/dI decreases when I is increased but in a reversible manner when I is reversed. Next the applied dc current is increased up to 4 mA. As shown in Fig. 2(ii), dV/dI exhibits a strong hysteresis when I is cycled up and down and noise (at 33 Hz) is detected above $I=3.5$ mA. The dc current is then swept down to zero and variations of the linear magnetoresistance (curves β) are measured, as in Fig. 1. The resistance at $H=0$ does not change, but the magnetoresistance is greatly increased and the Shubnikov–de Haas oscillations are strongly modified. It can be seen that oscillations are detected at lower values of H for both orientations. Then H is fixed at the value of 17.7 T, where the magnetoresistance is now maximum and the variation of dV/dI as a function of I is measured, as shown in Fig. 2(iii) with I up to 6 mA: A strong noise is seen when I is higher than 3.5 mA but dV/dI only shows hysteresis for $|I|<3.5$ mA. This hysteresis loop can be stabilized by cycling I many times between equal positive and negative values. The linear magnetoresistance does not undergo further changes when the excursion of I is increased from 4 to 6 mA and that is still true when I is increased beyond 6 mA. To recover the virgin state (curves α in Fig. 1) the sample has to be warmed up and cooled down again. The temperature at which the "annealing" occurs has to be determined more precisely but it is below 77 K (temperature to which our samples have been warmed).

We show in Fig. 3 for sample 2 at $H=19.7$ T and $\mathbf{H}\parallel(b,c)$ the variation of dV/dI and of the noise-voltage amplitude measured through voltage leads with a broadband pass frequency (a few Hz–100 kHz) as a function of the applied current. It can be clearly seen that the amplitude of the broadband noise increases strongly above 1.5 mA which is currently interpreted as resulting from the CDW depinning.[2] However, dV/dI is highly nonlinear before the onset of noise. In Fig. 4, the magnetoresistance

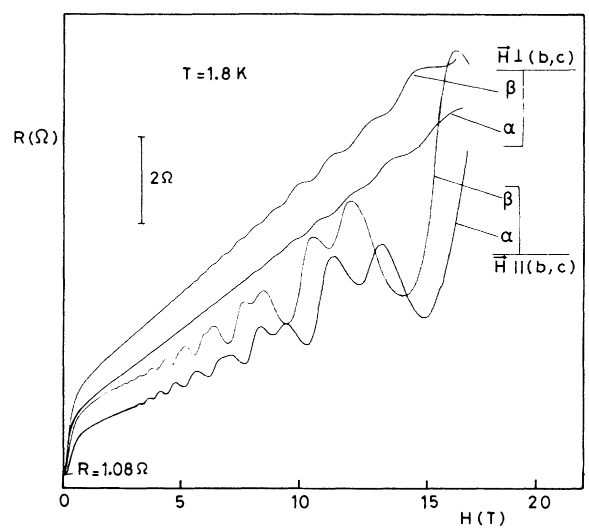

FIG. 1. Variation of the magnetoresistance of $NbSe_3$ (sample 1) at $T=1.8$ K with $\mathbf{H}$ parallel and perpendicular to (b,c) plane. Curves α correspond to the virgin state when the sample has just been cooled from room temperature. Curves β are recorded after a current (4 mA) large enough to depin the CDW has been swept up and reduced to zero.

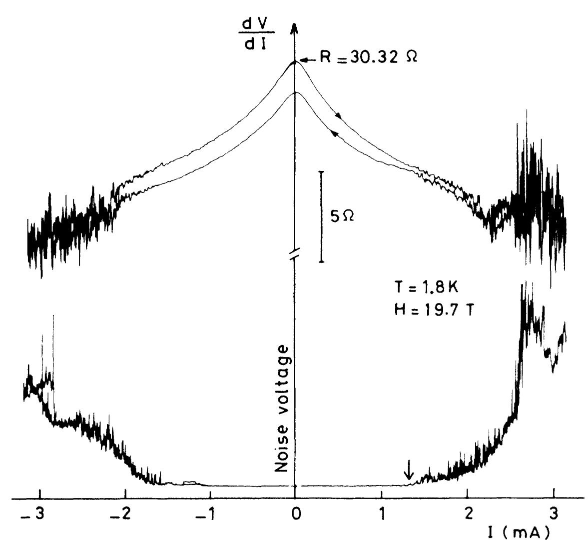

FIG. 3. Variation of the differential resistance dV/dI and of the noise voltage amplitude of $NbSe_3$ (sample 2) at $T=1.8$ K and $H=19.7$ T applied parallel to the (b,c) plane as a function of the applied current. The conductivity shows a nonlinear behavior before the noise generation and the CDW depinning.

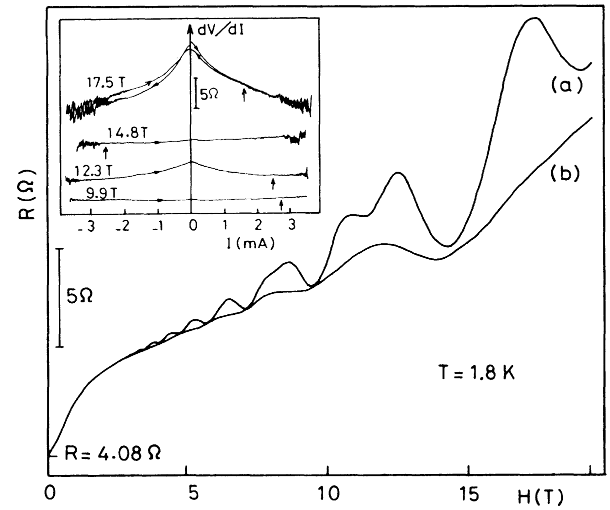

FIG. 4. Variation of the magnetoresistance of $NbSe_3$ (sample 2) at $T = 1.8$ K with the magnetic field parallel to (b,c) plane: curve (a) with a nearly zero dc applied current; curve (b) with the dc applied current of 2.3 mA lower than the critical current above which noise is generated in the sample, as indicated by arrows in the inset. The latter shows the variation of the differential resistance dV/dI as a function of the applied current for values of the magnetic field where $R(H)$ is maximum ($H = 17.5$ and 12.3 T) and minimum ($H = 14.8$ and 9.9 T).

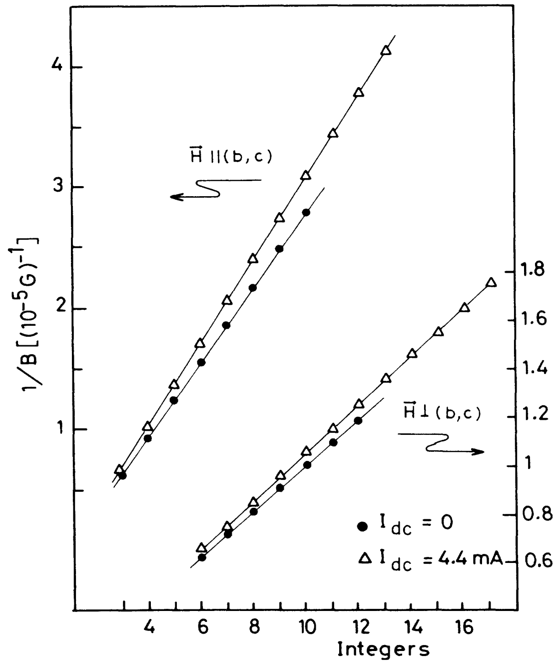

FIG. 5. $1/B$ values of the minima in the magnetoresistance of $NbSe_3$ (sample 1) as a function of quantum numbers for B applied parallel and perpendicular to (b,c) plane when the sample is in the virgin state (curves α in Fig. 1) and when the CDW has been depinned and repinned (curves β in Fig. 1).

of sample 2 with $\mathbf{H} \parallel (b,c)$, measured after the CDW has been depinned and repinned, is plotted when the applied dc current is nearly zero (curve a is equivalent to curve β for sample 1 in Fig. 1) and when the applied dc current is 2.3 mA. The inset of Fig. 4 shows the variation of dV/dI as a function of the applied dc current at the values of H where $R(H)$ is maximum at ($H = 17.5$ and 12.3 T) and minimum ($H = 14.8$ and 9.9 T). For each value of H the critical current above which noise is generated is indicated by an arrow. Figure 4 shows that the amplitude of the Shubnikov-de Haas oscillations is greatly suppressed by the application of a dc current lower than the critical current for depinning the CDW (with the exception of H higher than 17 T, where a small contribution to the reduction of R comes from the sliding CDW). Heating effects for explaining the reduction of the amplitude of the oscillations can be ruled out because the power dissipated in the sample is one order of magnitude lower than the maximum heat exchange between superfluid liquid helium and the surface of the sample.

These measurements lead one to conclude that the CDW depinning under H at liquid-helium temperature occurs at a finite threshold field: E_T is 290 mV/cm at $H = 17.7$ T for sample 1 in Fig. 2, and $E_T = 870$ mV/cm at $H = 19.7$ T for sample 2 in Fig. 3. This threshold field is angularly dependent. It was reported that E_T strongly decreases with H when H is applied perpendicular to (b,c), but that E_T is not affected by H when H is parallel to (b,c).[10] (E_T shown by arrows in the inset of Fig. 4 is nearly independent of H for $10\ \text{T} < H < 20\ \text{T}$.) Then before the apparent onset of the CDW depinning there is another nonlinearity without threshold and without noise. The amplitude of the quantum oscillations is suppressed by E for both orientations of H, and high-purity samples (measured by RRR) show a greater degree of suppression than low-purity samples. Measurements on sample 3 with RRR ~ 18 have shown a very small variation of $R(H)$ before the CDW depinning. So if dc measurements are performed on pure samples both types of nonlinearity cannot be separated and, consequently, it can be estimated, as reported by Coleman *et al.*,[11] that depinning occurs at a vanishing threshold value. In Fig. 5, we have plotted the $1/B$ values of the minima of $R(H)$ for sample 1 as a function of quantum numbers. The oscillations are periodic in $1/B$ and the slope gives the frequency of the oscillations and therefore the area of the FS perpendicular to the orientation of H. For the virgin state (curves α in Fig. 1) we find a period of 0.33 MG for $\mathbf{H} \parallel (b,c)$ and 1.06 MG for the higher frequency when $\mathbf{H} \perp (b,c)$, in agreement with previous measurements.[3-5] When a current higher than the threshold is applied, not only are the quantum oscillations shifted, but also the period of oscillations is decreased (curves β in Fig. 1): 0.296 MG for $\mathbf{H} \parallel (b,c)$ (a decrease of 10%) and 1 MG for $\mathbf{H} \perp (b,c)$ (a decrease of 6%). Similar results were obtained on all the samples studied.

The depinning and the repinning of the CDW at liquid-helium temperature under a magnetic field has two main effects: an increase in the magnetoresistance $R(H)$ but no effect on the value of R at $H = 0$, and a change in the FS. Electric-field-induced disorder has been recently studied at low temperature in blue bronze.[13] It has been shown that the application of $E > E_T$ below 90 K leads to a transverse broadening of the superlattice peak. This broadening is metastable, it remains when E is reduced to zero, and it results from finite-range order of the CDW in the transverse directions. The experiment we have described may have a similar interpretation except that the disorder induced by the electric field primarily affects the magnetotransport properties of the small electron pocket which remains after both CDW condensations. Application of E higher than E_T, especially if the CDW displacement is not homogeneous, may lead to microdomains with domain walls parallel to **b**. The resistance at $H=0$ is not affected because electrons flow along **b**, but when $H\neq 0$ electrons in their cyclotron motion may be scattered by these domain walls leading to the equivalence of a size effect which would be responsible for the increase of R. Quantum oscillations take place when Landau levels cross the Fermi level. A shift in the extrema of the Shubnikov–de Haas oscillations means that the Fermi surface has been modified during the depinning-repinning process. One might think that the Fermi level has suffered local variations required to screen local displacements of the CDW frozen into metastable states after the CDW has been put into motion, but if that were the case, then, because of the randomness of pinning, the quantum oscillations should have been washed out. Two other possibilities can be considered: Either the number or the effective mass of carriers in the electron pocket has been modified. The decrease of 10% in the electron pocket (10^{17} cm^{-3}) would induce an increase of the CDW wave vector of 10^{-4} (there are 10^{21} electrons per cm^3 in the band affected by the CDW). No variation in the longitudinal CDW wave vector of $NbSe_3$ has been detected by synchrotron-radiation measurements within an accuracy of a few 10^{-5} when $E=3E_T$ has been applied at 45 K.[14] However, as explained above, the temperature at which the metastable states are frozen still has to be determined. A change in the FS can originate from the release of internal stresses resulting in the virgin state from random pinning. The variation of the frequency of the quantum oscillations has been measured as a function of hydrostatic pressure.[5] With the variation of 10% shown in Fig. 5, these stresses have to be equivalent to a pressure of 400 bars, which seems to be unphysical. Then the close interaction between metastable states and conduction band has to be further elucidated.

In summary, we have shown that the application of an electric field suppresses the amplitude of the Shubnikov-de Haas oscillations of $NbSe_3$ before the depinning of the CDW. The depinning-repinning of the CDW strongly increases the magnetoresistance and reduces the cross sections of the FS. These modifications result from metastable states induced by the application of the electric field. These metastable states can be released only if the sample is warmed and cooled down again.

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