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Charge-density-wave narrow-band noise in $NbSe_3$ at $T < 4$ K

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We report observations and the first study of the properties of charge-density-wave (CDW) narrow-band noise, i.e., current oscillations, at $T < 4$ K in the CDW material $NbSe_3$. The CDW narrow-band noise (NBN) appears to exist only in certain ranges of magnetic-field strength and the amplitude tends to die out at high electric fields. The observation of NBN, which is unique to CDW systems, confirms that the non-Ohmic resistance in a magnetic field that is found in $NbSe_3$ at these temperatures, $T < 4$ K, is correlated with motion of a CDW. We also find that the CDW current versus NBN frequency relationship is nonlinear at $T < 4$ K, unlike that found at higher temperatures, $T > 25$ K and $B \leq 9.6$ T. The nonlinear current at a given frequency is also very large compared to that at higher temperatures.

INTRODUCTION

Recently a lot of research effort has been given to investigating the interaction and effect of an applied magnetic field on charge-density-wave (CDW) systems. Coleman *et al.*[1] found that when a magnetic field was applied perpendicular to the high-conductivity axis in $NbSe_3$ a very large magnetoresistance $\Delta\rho/\rho$ was observed at temperatures in the range $10\text{ K} < T < 50$ K. They found that at $B = 22.7$ T, $\Delta\rho/\rho$ was on the order of 2–4 in the temperature range 20–45 K and $\Delta\rho/\rho$ went to zero at the lower CDW transition temperature, 59 K. Balseiro and Falicov[2,3] have attributed this large increase of the resistive anomaly with magnetic field to an enhancement of the CDW gap through an improvement of the Fermi-surface nesting. This magnetic-field-enhanced gap would then lead to a direct conversion of carriers from the normal state to the CDW state and a consequent increase of the resistivity. From experiments on the CDW narrow-band noise (NBN) in the temperature range, $24\text{ K} < T < 55$ K, Tritt *et al.*[4,5] found that the magnetic field has very little effect on the CDW carrier concentration up to their highest field, 9.6 T, although like Coleman *et al.*, they found a large enhancement of the resistive anomaly with magnetic field. Thus, there appears to be some other mechanism which causes this large magnetoresistance at high temperatures, $20\text{ K} < T < 55$ K, other than the conversion of normal to CDW carriers.

A further concern is the interaction of the CDW state in $NbSe_3$ with an applied magnetic field at $T < 4$ K. Coleman *et al.*[1] found that in this temperature range the magnetoresistance is very large, i.e., at $B = 22.7$ T, $\Delta\rho/\rho$ was on the order of 15–20. As a semimetal with just a few small pockets of carriers,[6–8] $NbSe_3$ can be expected to have a large magnetoresistance. Nevertheless, $\Delta\rho/\rho$ on the order of 15–20 seems anomalously large. Coleman *et al.*[9] and Everson *et al.*[10] also found that in a magnetic field, at $T < 4$ K, the conductivity is enhanced at high electric fields, and they interpreted this enhanced conductivity as a depinning of the CDW from the lattice. Previously, nonlinear conduction and threshold electric fields in $NbSe_3$ had only been measured down to $T \approx 20$ K due to high threshold fields and sample heating problems at temperatures below 20 K. However, in a magnetic field at $T < 4$ K, a threshold electric field and nonlinear conduction exist and are assumed to be related to motion of a CDW, as it is at higher temperatures.

At high magnetic fields the Shubnikov–de Haas (S-dH) oscillations in $NbSe_3$ are very sensitive to slight changes in the metastable state configuration of the CDW. The S-dH oscillation frequency changes by approximately 10% when the CDW is initially depinned and then repinned.[11–14] As indicated by spontaneous changes in the magnetoresistance and in the $B = 0$ resistance, an abrupt transition with time in the state of the sample is observed in samples that have been rapidly quenched from higher temperatures to liquid-helium temperature.[15,16] When these spontaneous changes occur, the S-dH oscillation frequency may change by as much as 12%. In both cases, depinning-repinning and post-transition, the sample will remain in the latter state (equilibrium state) without further changes unless the temperature is raised and consequently the sample annealed. Then, a metastable state could be reachieved. Both of the above processes affect the metastable state configuration of the CDW and thus the normal carriers of the system.

At $T < 4$, the S-dH oscillation amplitude in $NbSe_3$ is depressed at high electric fields.[9,13,17] In one report Richard, Monceau, and Renard[13] find that the S-dH oscillation amplitude is depressed at high electric fields by suppressing the S-dH maxima whereas the minima are unchanged. They also report that the magnetoresistance is non-Ohmic before the onset of CDW motion. They detected the onset of CDW motion by observing a large increase in the broadband noise. They find threshold electric fields, at liquid-helium temperatures, to be between 300–850 mV/cm for the samples they report. Monceau and Richard also find that the threshold field as defined by an increase in broadband noise is angularly dependent on B and that a critical line (T,H) exists for a reduction in E_T (Refs. 18 and 19). At 4.2 K and $B = 7$ T, Monceau, Richard, and Laborde[19] observed NBN but, as they stated, were unable to measure I_{CDW} with enough accuracy to obtain the corresponding I_{CDW} vs F relationship.

In this paper, we present data concerning the NBN in $NbSe_3$ at $T<4$ K in the presence of a magnetic field. We find the existence of NBN, which is a signature that is unique to a system exhibiting CDW motion.[20] Thus, this confirms that the non-Ohmic conductivity under these conditions is due, at least in part, to motion of the CDW. We find that the NBN exists only in certain regions of magnetic-field strength and that the amplitude of the NBN dies out at high electric fields. We also find that I_{CDW} is very large and does not follow a linear relation with the NBN frequency F as it does at higher temperatures.[4,5,21]

EXPERIMENTAL PROCEDURE

We have measured the NBN frequency F, the CDW current density J_{CDW}, and the sample magnetoresistance $\Delta\rho/\rho$ for $T<4$ K in magnetic fields up to $B=9.6$ T. Single crystals of $NbSe_3$ with a residual resistance ratio of approximately 80 were mounted perpendicular to the magnetic field (with $\mathbf{B}\parallel\mathbf{c}$) and in a four-probe resistance configuration. The samples were attached to four copper wires with Dupont silver paint (No. 5007). The samples used were roughly $0.001\times0.005\times0.5$ mm^3 in size. A spectrum for NBN, at $T=1.5$ K and $B=9.6$ T, is shown in Fig. 1 at an applied electric field of 600 mV/cm. We see that the amplitude of the current oscillation is large and the fundamental frequency F is approximately 3 MHz and at least two harmonics are observable. This spectrum is of somewhat less quality than a spectrum for the same sample at higher temperatures.

The threshold electric fields at these temperatures, $T<4$ K, are high, usually $E_T\approx600$–900 mV/cm, and therefore heating, especially at the contacts, is a potential problem. We performed all the NBN experiments at these temperatures with the sample immersed in superfluid helium in order to minimize this problem. After the temperature was stabilized, we typically performed a magnetic-field sweep, thus establishing the S-dH oscillations and the metastable state configuration in which the sample was residing. At the highest magnetic field we would then apply a large electric field to the sample to depin the CDW. We then lowered the electric field below threshold and performed another magnetic-field sweep down and back up at $E\ll E_T$. This essentially locked the sample into the equilibrium state,[15] which is relatively stable. The intent is to perform the NBN experiments starting from the equilibrium state, which we did. After the equilibrium state was established, we applied a specific magnetic field and increased the electric field until NBN was observed with a spectrum analyzer. A dc current was passed through a standard resistor in series with the sample for determination of the total current. The Ohmic resistance of the sample was measured with the four-probe configuration for electric fields much less than the threshold field. A computer simultaneously measured the frequency, the sample resistance, the total current, the temperature, and then calculated the nonlinear current or I_{CDW} for each data point.

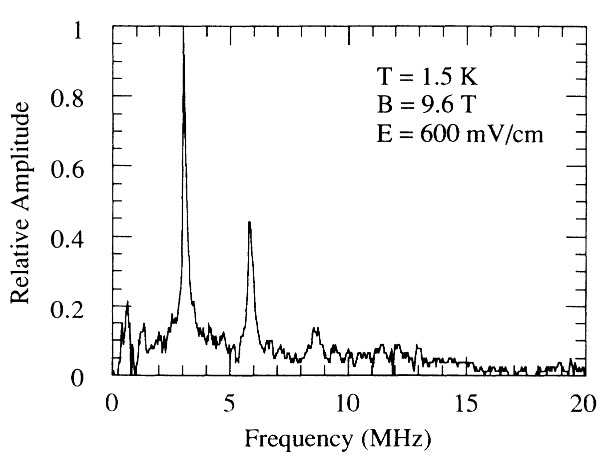

FIG. 1. A plot of the amplitude of the NBN in $NbSe_3$ vs the frequency at $T=1.5$ K and $B=9.6$ T. The fundamental frequency is seen to be 3 MHz and at least two harmonics are observable. The applied electric field E is 600 mV/cm.

RESULTS AND DISCUSSION

At $T>20$ K, when the CDW begins to slide, NBN may be observed whose frequency has been found to be linearly dependent on the current that is carried by the charge-density wave. The slope of the I_{CDW} vs F curve is proportional to the CDW carrier concentration n_{CDW}.[21] The CDW current I_{CDW} is given by the following relationship: $I_{CDW}=I_T-V_S/R_\Omega$, where I_T is the total current, V_S is the sample voltage, and R_Ω is the Ohmic sample resistance. I_{CDW} as calculated from this relationship is really a measure of the non-Ohmic current and is assumed to be due totally to CDW conduction.

On the basis of previous reports[5,11–16] there does not appear to be any magnetic-field dependence of n_{CDW} at temperatures below 4 K, although this conclusion is inferred from a measurement of the S-dH oscillations. Therefore, it was our desire to measure n_{CDW} in a more direct way, i.e., through the slope of I_{CDW} vs F. We found it is very difficult to detect the NBN at $T\approx1.5$ K even at our highest magnetic field, 9.6 T, and we have yet to observe the NBN below $B=6$ T. We observed the NBN at $T<4$ K in four samples. The spectra of two of these samples were of sufficient quality to perform detailed studies. In fact, we found that we could observe and study the NBN in only about one out of every twenty samples we ran, although all the samples exhibited non-Ohmic conductivity in a magnetic field at high electric fields at these temperatures. The NBN was prominent in all the samples we checked at higher temperatures, $T>30$ K. This, therefore, made the investigation of the NBN phenomena very difficult. Nevertheless, the observation of the NBN, as in Fig. 1 at $T=1.5$ K and $B=9.6$ T, confirms that the non-Ohmic magnetoconductivity can be associated with the motion of the CDW since the NBN is unique to CDW motion.

In Fig. 2, we show I_{CDW} vs F at $T=1.5$ K for various magnetic fields. It can be seen that the CDW current is not a linear function of the frequency as it is at higher

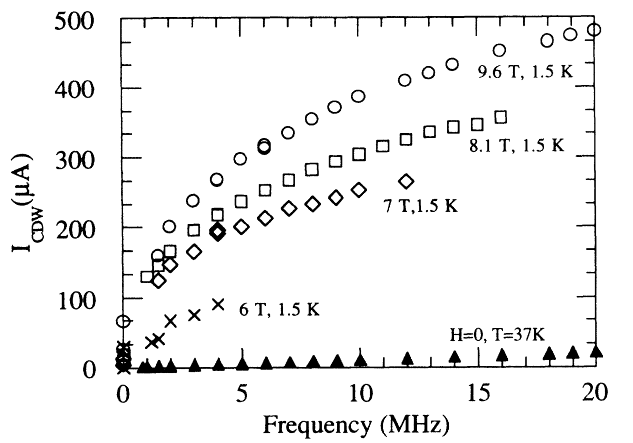

FIG. 2. A plot of I_{CDW} vs F at $T=1.5$ K for four magnetic fields. The sample parameters are length $l=0.51$ mm, R_S(300 K) $=345$ Ω, and an estimated cross-sectional area $A=3.7$ μm^2. The data shows the nonlinear dependence of I_{CDW} vs F. The amplitude of the NBN dies out at high electric fields and this occurs at successively lower frequency as the magnetic field is lowered. For comparison, I_{CDW} vs F for the same sample at $T=37$ K is also shown.

temperatures. It should be noted that from the definition of the CDW current it is strictly the non-Ohmic current. Thus, if the magnetoconductivity is a function of E, for reasons other than motion of the CDW, then our relating it to I_{CDW} is inappropriate. At this point there is no reason to believe this is the case. We also found that as the electric field was increased the amplitude of the NBN seemed to die away. The electric field was increased well beyond the point where the NBN amplitude died away without the NBN reappearing. As the electric field was reduced the NBN reappeared without hysteresis. The highest-frequency data point as shown in Fig. 2 for each of the four magnetic fields corresponds approximately to the highest electric field at which the NBN spectrum could be detected. Notice that the highest observable frequency occurred at successively lower frequency as the magnetic field was reduced. After taking this data, we increased the temperature to $T=37$ K and found that I_{CDW} vs F was linear and in perfect agreement with previous measurements on other samples at this temperature.[4,5] This data is also shown in Fig. 2. It is striking that at a given frequency I_{CDW} at $T=37$ K is approximately 20 times smaller than I_{CDW} at 1.5 K and 9.6 T. For another sample, at $T=1.3$ K, we found similar behavior in that I_{CDW} vs F was nonlinear and the amplitude of the NBN seemed to die away at high electric field. The NBN for this sample was observable around only two magnetic fields, 9.6 and 8 T, and not in between.

We first thought that the existence of the NBN would be related to maxima and minima in the S-dH oscillations similar to the nonlinear magnetoconductivity reported by Richard *et al.*,[13] but from the data we have taken we can find no such direct relationship. On the other hand, our own work emphasizes that the details of the phenomena under study are somewhat sample dependent. We had also hoped that we could get a measure of n_{CDW} at $T<4$ K and thus compare it to the value measured at higher temperatures. This would have allowed us to address two outstanding questions. First, if n_{CDW} were changed little from higher temperature, then it could be concluded that this CDW is the same as the higher temperature CDW; and second, we would know something about the temperature dependence of n_{CDW} down to lower temperatures. On the other hand, had n_{CDW} at $T<4$ K been well defined but very different than the higher temperature, then n_{CDW}, its nature and origin (possibly magnetic-field induced as in graphite[22]), would have warranted further study.

In conclusion, we have found NBN in $NbSe_3$ at $T<4$ K and $B>6$ T, which confirms that the non-Ohmic magnetoconductivity in this range is correlated with motion of a CDW. It appears that the NBN exists only in certain regions of magnetic field and that the amplitude dies out at high electric fields. We also find that I_{CDW} does not follow a linear relation with the NBN frequency as it does at higher temperatures.[4,5,21] Thus a direct measure of n_{CDW} is not accomplished. Finally, I_{CDW} is much larger at a given NBN frequency than is found in the same sample at higher temperatures.

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