Solid State Communications, Vol.57, No.1, pp.27-30, 1986.
Printed in Great Britain.

0038-1098/86 $3.00 + .00
Pergamon Press Ltd.

DISTINCT CURRENT-CARRYING CHARGE DENSITY WAVE STATES IN $NbSe_3$

R. P. Hall and A. Zettl

Department of Physics, University of California, Berkeley
Berkeley, Calif. 94720

(Received 16 September 1985 by A. Zawadowski)

We have performed detailed measurements of the dc current-voltage (I-V) characteristics of $NbSe_3$ in the hysteretic switching regime. Within the hysteresis loop, we observe a series of well-defined and quasi-stable current-carrying states, each with a unique I-V relationship. Transitions between the states, induced by both the applied electric field and thermal fluctuations, are observed. Rapid and random transitions between closely spaced levels are suggested to result in excessive current or voltage noise for the depinned charge density wave condensate.

A particularly interesting aspect of charge density wave (CDW) conduction in linear chain metals is that of switching, where only slight and smooth increases or decreases in the applied electric field E can lead to sudden discontinuous "jumps" in CDW current. The switching phenomenon has been observed in virtually all materials which display CDW conduction (e.g. $NbSe_3$, TaS_3, $K_{0.3}MoO_3$)[1-3], although it occurs only over limited ranges of temperature, and not in all crystals of a given material. Switching is particularly striking when it corresponds to the onset of CDW conduction. However, the observation of switching above the depinning field E_T, or multiple switchings for a given sample, suggests that switching and initial CDW depinning are in general to be regarded as independent processes[4]. In $NbSe_3$, switching has been observed at temperatures as high as 45 K, and is especially prevalent in the temperature range 20 K - 30 K.

Various models have been proposed to account for switching phenomena in CDW conductors. A single phase coordinate description[5] assumes a finite inertia for the dynamic CDW condensate, while other approaches invoke additional degrees of freedom for the CDW condensate. For example, the model of Joos and Murray[6] is an application of the kinetic Ising model, where the CDW crystal is composed of an array of domains, each being in a conducting or nonconducting state. CDW conduction through the crystal occurs only after a continuous channel of conducting domains has been established. An alternate many degree of freedom model[7] considers macroscopic polarization of the CDW condensate, and predicts switching when the local applied field everywhere exceeds the local threshold depinning field within the sample volume. Recently, localized phase slip centers have been suggested as the origin of switching[8]. There, switching corresponds to a break-up of the CDW phase, with different macroscopically coherent regions of the sample assuming different phase velocities for a given externally applied electric field.

In this Communication we report on detailed dc I-V characteristics of $NbSe_3$ in the switching regime. Within the overall hystersis loop associated with the switching process at low temperatures we observe a rich "sublevel" structure corresponding to many distinct current-carrying states. Hysteretic transitions, initiated either by slight perturbations in the externally applied electric field, or thermal fluctuations, are observed between closely spaced sublevel states. We interpret the sublevel structure as representing different portions of the CDW condensate being depinned, consistent with a macoscopic domain structure where phase slip centers comprise the interface between domains.

Our experiments employed single $NbSe_3$ crystals prepared by conventional vapor transport methods, with typical threshold electric fields E_T = 30 mV/cm at 48 K. Two probe contacts were made with conductive silver paint. Both current controlled and voltage controlled dc conductivity measurements were performed, with similar results. Here we report only on voltage-controlled experiments. For these experiments, a low output impedance voltage follower (bandwidth dc to 300 MHz) provided the drive voltage to the $NbSe_3$ crystal in series with a small current sensing resistor, with a resistance value typically two orders of magnitude smaller than the sample resistance. The drive voltage could be automatically ramped up or down, or adjusted manually.

Fig. 1 shows a series of voltage-driven I-V curves for $NbSe_3$ at selected temperatures in the lower CDW state, obtained by first monotonically increasing, and then monotonically decreasing, the bias voltage. At 42 K, switching at the threshold E_T for CDW conductivity is clearly observed. At 35 K, switching is again observed at E_T, but a well-defined hysteresis loop has developed for the single switching event into or out of the sliding CDW state. With decreasing temperature below 35 K, the hysteresis becomes more pronounced. The magnitude for the overall hysteresis loop is defined as H_V = Vmax - Vmin.

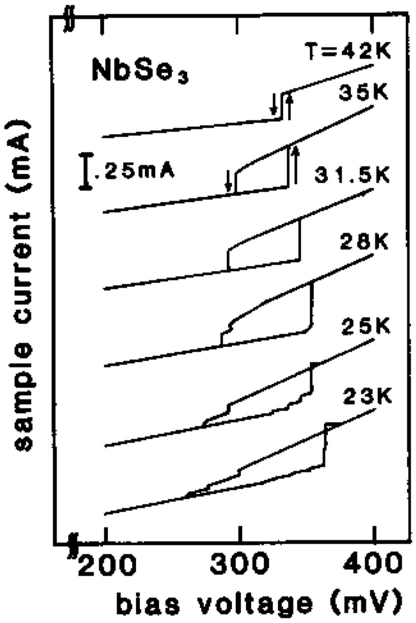


Fig. 1. Voltage-controlled I-V characteristics of $NbSe_3$ at selected temperatures in the switching regime. Additional switching structure is observed on the hysteresis loop at lower temperature.

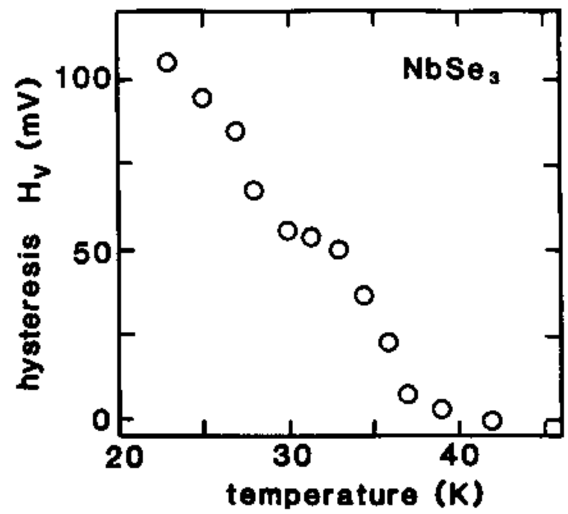
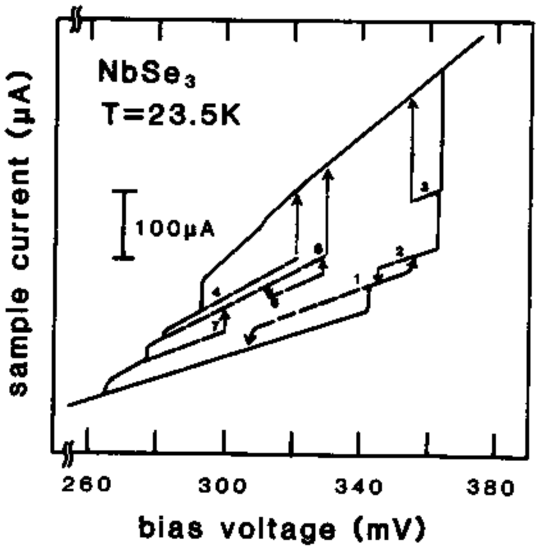


Fig. 2. Hysteresis magnitude H_V versus temperature for $NbSe_3$ in the switching regime.

Vmax is the upper closing point of the hysteresis loop, i.e. the voltage bias above which the I-V characteristics become independent of bias sweep direction. Similarly, Vmin defines the lower bias point for the closing of the hysteresis loop. In the present case, the CDW is always pinned below Vmin, irrespective of bias sweep direction. Fig. 2 shows H_V versus temperature, as determined from a complete set of I-V traces which includes the traces of Fig. 1. As is apparent from Fig. 2, H_V smoothly increases with decreasing temperature in the switching regime. Although an increase in hysteresis with decreasing temperature is not unexpected, it appears difficult to directly correlate the result of Fig. 2 to other temperature-dependent parameters of the CDW state, for example the order parameter or the intrinsic depinning field (in the absence of switching). It should also be noted that the curve of Fig. 2 is highly sample dependent, especially with regard to the x-axis intercept (point of zero hysteresis).

An important feature of the hysteretic response in Fig. 1, for temperatures below 30 K, is the appearance of additional switching structure. For example, at 28.3 K the transition back to the pinned CDW state for decreasing bias voltage sweep is comprised of a series of two smaller distinct switches, rather than a single large switch as occurs at 30 K. At temperatures below 28 K, additional switching structure is observed during increasing bias voltage sweep as well, and in general, more additional switching structure appears with decreasing temperature. At 23 K, for example, the transition to the "fully conducting" CDW state (where the hysteresis loop has closed and increases in dc bias result in no further switches) occurs by a series of at least four individual switches; the transition back to the fully pinned state occurs via at least three distinct switches.

As we now demonstrate, the additional switching structure which develops in the hysteretic regime reflects transitions between well-defined current-carrying states. Fig. 3a shows the detailed I-V characteristics within the hysteresis loop at 23.5 K, for the same $NbSe_3$ sample as was used for Figs. 1 and 2. The solid lines represent the overall hysteretic I-V structure, consistent with that displayed previously in Fig. 1. The striking features of Fig. 3a are the dashed lines, which represent distinct, repeatable, and quasi-stable current-carrying states. The direct mapping of the first of these states (labeled 1 in the figure)was achieved by slowly advancing the dc bias voltage from zero into the hysteretic regime, until a small switch occured, and then immediately reversing the direction of bias sweep. The I-V characteristics of this sublevel state were then traced out by slowly varying the dc bias and recording the resulting current. Observed vertical transitions from sublevel 1 to a second sublevel, labeled 2, are indicated by vertical arrows in Fig. 3a. This second sublevel, and subsequent sublevels, were traced out in a fashion similar to that described for level 1. Since the sublevels 1, 2, and 3 were achieved by first starting the dc bias voltage from zero, we classify these structures as "lower" sublevel states. As shown in Fig. 3a, distinct sublevel structure is also observed if the dc bias is started from a high value, exceeding Vmax. Sublevels thus achieved are labeled 4, 5, 6, and 7 in the Figure. We classify these states as "upper" sublevel states, since they are arrived at by first decreasing the dc bias voltage from above Vmax.



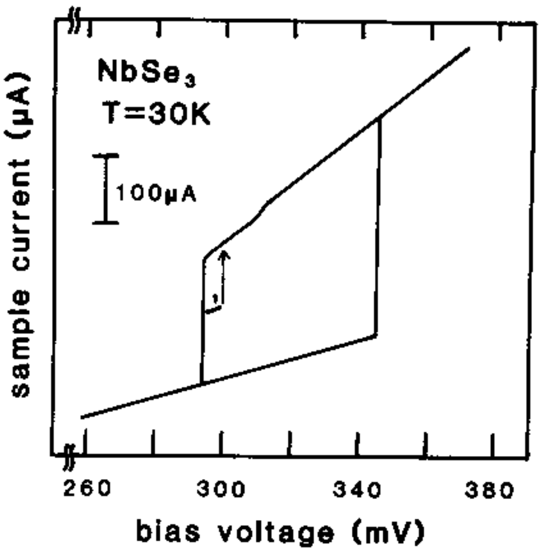


Fig. 3. a) Detail of I-V structure of $NbSe_3$ at 23.5 K. The numbered dashed lines represent distinct sublevel states; the vertical arrows correspond to electric field-induced transitions between the states (see text).

b) Detail of I-V structure at 30 K. Only one stable sublevel state is observed within the hysteresis loop.

The substructure indicated in Fig. 3a was found to be entirely reproducible. However, upon tracing out the various sublevels, transitions between sublevels did not always occur at the same dc bias value. The vertical arrows represent repeatable transition points. Since these transitions are determined soley by the dc bias voltage magnitude, we identify them as elecric field-induced. As indicated in Fig. 3a, both upward and downward electric field-induced transitions are observed. Other non-repeatable transition points, not indicated on Fig. 3a, were also obtained. These transitions occured if the system was left unperturbed for a sufficiently long time on a particular sublevel, with a (fixed) dc bias voltage relatively close to the indicated vertical arrows. The timescale for waiting for such transitions to occur was often on the order of seconds to minutes, and we identify these transitions as induced by thermal fluctuations. Because of this sensitivity to thermal fluctuations, we term the substates in Fig. 3a as quasi-stable.

The importance in distinguishing between "lower" and "upper" sublevel states in Fig. 3a lies in that no transitions were found to occur between a state in the lower group to a state in the upper group, or vice versa. It is thus possible that the two groups of sublevel states do not coexist. The lower sublevel states might be established only as the dc bias voltage V_{dc} is increased past Vmin, while the upper sublevel state would seem to be established only as V_{dc} is decreased past Vmax. Increasing V_{dc} past Vmax, or decreasing V_{dc} past Vmin, would then effectively eradicate the lower and upper sublevel states, respectively.

The strong temperature dependence of the overall hysteresis loop shown in Fig. 2 is reflected also in the sublevel structure. Fig. 3b shows the complete sublevel structure observed at 30 K. Only one sublevel state is clearly resolved, in contrast to the six defined at 23.5 K. Above 30K, no sublevel structure was observed within the overall hysteresis loop. Clearly, the formation of stable sublevel structure is a highly temperature-dependent process.

Our observations of sublevel structure within the response hysteresis loop have serious implications in regard to the theory of switching. It is apparent that sublevel structure is inconsistent with a single-phase coordinate approach[5], even with inertial terms included, and hence we exclude such a model. The overall hysteresis behavior observed in Fig. 1 and further analyzed in Fig. 2 might be compatible with a model of macroscopic phase polarization and subsequent depinning[7], but since in that model there exist no metastable states for a depinned CDW condensate, it cannot explain the distinct current-carrying states we observe.

Our results appear most consistent with a macroscopic domain structure, where the sublevel structure, corresponding to distinct current-carrying states, results from a particular (relatively stable) configuration of pinned and current-carrying CDW domains. The same basic model has been used to account for negative differential resistance (NDR) in $NbSe_3$, where chaotic instabilities are attributed to rapid hopping between distinct current-carrying states[9]. In fact, the only significant difference between the states assumed in that analysis, and those directly observed here (at lower temperatures), is the relative energy spacing between the sublevels. In the NDR region, the energy spacing is significantly less, allowing for an increased transition rate (assuming a temperature independent attempt frequency), and hence chaotic response in the kHz and MHz frequency range[9].

The domain structure we associate with $NbSe_3$ in the switching regime is different from that suggested by Joos and Murray[6]. Recent experiments[8] have demonstrated the existence of bulk phase slip centers in $NbSe_3$ which segment the CDW crystal into only a few distinct macroscopic domains along the length of the crystal, with each domain having a unique depinning field and hence unique I-V characteristic. In contrast to the Joos Murray model, nothing in the phase slip case prevents a "middle" domain, near the center of the crystal, from depinning first and immediately thereafter conducting a real CDW current, despite the existance of domains which remain pinned near the ends of the crystal. We identify the sublevel structure of Fig. 3 with different configurations of pinned and depinned macroscopic domains within the sample. As an independent test of our model, one could attempt observation of the narrow-band noise spectrum associated with each sublevel, and correlate it to the noise spectra of individual depinned domains. This experiment has been performed and supports the model[10]. For example, on sublevel 3 in Fig. 3a, the noise spectrum indicates three depinned domains, while on sublevel 2, the noise spectrum indicates only two depinned domains, etc.

The successive disappearance of the stable sublevel structure in NbSe3 with increasing temperature above 23 K suggests that the strength of the phase slip centers (i.e. the ability of the phase slip centers to break the CDW phase locally) is strongly temperature dependent. As the temperature is increased, weak phase slip centers are annealed out, effectively phase-linking domains together, and reducing the number of possible macroscopic switches in the I-V characteristics. At relatively high temperatures (still in the switching regime), only the strongest phase slip center will remain active, permitting at most two distinct successive switches. Should one of the domains have a substantially greater depinning voltage than the other (as might occur, for example, if one domain is relatively small and located close to a potential contact), then only one distinct switch into and out of the "fully conducting" region will occur, as demonstrated in Fig. 1.

Finally, we comment that our model of distinct current-carrying states might well be applied to other CDW materials, for example TaS_3 and $K_{0.3}MoO_3$, where both random and regular (i.e. oscillatory) transitions between the pinned and depinned states are observed [2,11], often with strong hysteresis behavior. A detailed investigation of possible macroscopic domain structure and phase slip centers in these materials is presently underway.

This research was supported under NSF Grant DMR 84-00041. One of us (A.Z.) is an Alfred P. Sloan Foundation Fellow. R.P.H. acknowledges support from an IBM Pre-doctoral Fellowship.

References

1. A. Zettl and G. Grüner, Phys. Rev. B 26, 2298 (1982).
2. L. Mihály and G. Grüner (to be published); G. Kriza, A. Jánossy, and G. Mihály, in Charge Density Waves in Solids, Gy. Hutiray and J. Sólyom, eds., Springer Verlag Lecture Note Series in Physics, v. 217, p. 426 (1985).
3. J. Dumas, C. Schlenker, J. Marcus, and R. Buder, Phys. Rev. Lett. 50, 757 (1983).
4. R. P. Hall and A. Zettl, Solid State Commun. 51, 63 (1984).
5. R. P. Hall and A. Zettl, Solid State Commun. 55, 307 (1985); G. Grüner, A. Zawadowski, and P. M. Chaikin, Phys. Rev. Lett. 46, 511 (1981).
6. B. Joos and D. Murray, Phys. Rev. B 29, 1094 (1984).
7. A. Jánossy, G. Mihály, and L. Mihály, in Charge Density Waves in Solids, Gy. Hutiray and J. Sólyom, eds. Springer Verlag Lecture Note Series in Physics, v. 217, p. 412 (1985).
8. R. P. Hall, M. F. Hundley, and A. Zettl (unpublished).
9. R. P. Hall, M. Sherwin, and A. Zettl, Phys. Rev. Lett. 52, 2293 (1984). see also A. Maeda, M. Naito, and S. Tanaka, J. Phys. Soc. Japan, 54, 1912 (1985)
10. R. P. Hall and A. Zettl (unpublished).
11. J. Dumas and C. Schlenker, in Proceedings of the International Symposium on Nonlinear Transport and Related Phenomena in Inorganic Quasi-one-dimensional Conductors, Sapporo, Japan (1983), p. 198; A. Maeda, T. Furuyama, and S. Tanaka, Solid State Commun. (in press)