

Onset of charge-density-wave conduction: Switching and hysteresis in NbSe₃

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(Received 21 May 1982)

We report the observation of a switching phenomenon associated with the development of the current-carrying charge-density-wave (CDW) state of NbSe₃. We suggest that this effect is associated with the coupling of CDW regions and with the establishment of a coherent sliding CDW state in the material.

The nonlinear conductivity observed in the charge-density-wave (CDW) state in NbSe₃ at very low electric field strengths is suggestive of a new collective transport phenomenon. Early observations indicated a field dependence of the dc conductivity,¹ and subsequent experiments demonstrated that there is a sharp threshold field E_T for the onset of nonlinear conduction.² This, together with current fluctuations in the nonlinear region,^{2,3} is highly suggestive that the current is carried by the sliding CDW through mechanisms suggested originally by Fröhlich.⁴

Two models^{5,6} were suggested recently to describe the observed field² and frequency⁷ dependences: A classical description⁵ treats the low-field ac response as that of an overdamped oscillator, and the field dependence is modeled by a classical particle moving in a periodic potential under the influence of an electric field. The model leads to a sharp threshold field E_T , and above the threshold the conductivity is given by

$$\sigma(E) = \frac{ne^2\tau}{m} \left[1 - \left(\frac{E_T}{E} \right)^2 \right]^{1/2}, \quad (1)$$

where m and e are the mass and charge of the particle, and $\tau = \Gamma/m$, where Γ is the friction coefficient. Equation (1) holds for a sinusoidal potential, but any regular periodic potential leads, in the case of an overdamped response, to a $[1 - (E_T/E)^2]^{1/2}$ behavior near E_T .

A tunneling model proposed by Bardeen⁶ assumes that CDW's can tunnel through the potential barriers. In the semiconductor model the CDW tunneling leads, for $E > E_T$, to the expression

$$\sigma(E) = A (1 - E_T/E) \exp(-E_0/E), \quad (2)$$

where A is a constant of proportionality and E_0 is a characteristic field related to the CDW gap, similar to Zener tunneling. Equation (2) is similar to an empirical expression suggested by Fleming.² The only difference in Fleming's expression is that E is replaced by $E - E_T$ in the exponential of Eq. (2). The tunneling formula leads to an excellent description of the nonlinear conductivity of NbSe₃ (Ref. 8) and also

of TaS₃,⁹ over a broad electric field region.

In this Communication we report the observation of a switching phenomenon which is associated with the development of the current-carrying CDW state. The switching shows up in direct I - V traces and leads to a hysteresis between increasing and decreasing driving currents. The switching can directly be observed by a pulse technique, and the conductivity due to both the pinned and current-carrying state can simultaneously be measured. The time T between the start of the pulse and the beginning of the switching is a sensitive function of the applied current, while the switch duration (i.e., actual transition time) τ is a few microseconds. The distribution of times before switching T for a fixed applied current suggests that the switching is a deterministic and not a stochastic phenomenon.

Figure 1 shows an I - V curve recorded at $T = 26.5$ K by driving the current from a current source. The conductivity is Ohmic (i.e., $I \sim V$) up to a point where a switching occurs to a smaller voltage, i.e., a higher-conductivity state. Beyond that applied current, nonlinear conduction is observed, and σ (defined as I/V) increases with increasing current, as observed in previous studies.^{1,2} Upon decreasing the driving current, the switching back to the low-conductivity state occurs at a lower current, resulting in a well-resolved hysteresis effect.

The sharp jump to the current-carrying state cannot be observed at high temperatures. With increasing temperature, the hysteresis becomes first progressively smaller for the same rate of current sweep dI/dt , and also the magnitude of the voltage jump is reduced. Above about 38 K no jump is associated with the development of the nonlinear conductivity.

We also note that although we have observed the hysteresis effects on various samples, it appears to be extremely sensitive to sample perfection. Simultaneously, with the hysteresis we have also measured the coherent voltage oscillations above threshold reported earlier by Fleming.¹⁰ These oscillations, and the associated narrow-band noise, depend sensitively on the quality of the specimens, as inhomogeneities and irregular sample cross sections tend to smear out the

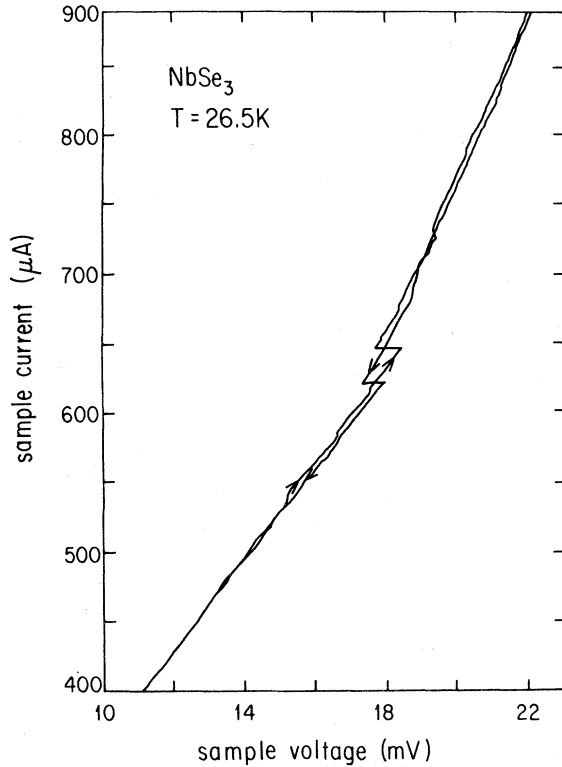


FIG. 1. I - V traces with increasing and decreasing driving current. The sweep time was 1 min for the full range shown in the figure. The sample length $l = 1.5$ mm.

oscillations, and lead to a complicated noise spectrum.¹¹ The sudden onset of the non-Ohmic conductivity, as shown in Fig. 1, was observed on samples where the current oscillations were also clearly observed. Samples which did not show clear oscillation phenomena also did not show the hysteresis effect.

The switching phenomena between the nonconducting state and the conducting state can directly be observed by applying a current pulse to the sample and observing the subsequent voltage drop on an oscilloscope. Below a threshold current I_T a regular pulse response with no unusual features is observed. Increasing the current I above I_T , the observed voltage shows a well-defined jump from a voltage V_1 to a smaller voltage V_2 , as shown in Fig. 2(a). V_1 corresponds to the Ohmic conductivity observed below I_T , and we conclude that this corresponds to the pinned CDW state. After a time T switching, usually of duration τ approximately a few microseconds, occurs to a state with lower voltage V_2 , i.e., to a state which has a higher conductivity. V_1 and V_2 measured simultaneously by pulse techniques for various applied currents is shown in Fig. 2(b). The full line represents the Ohmic conductivity observed also below I_T , while the dotted line is a guide to the eye and represents the I - V behavior of the current-

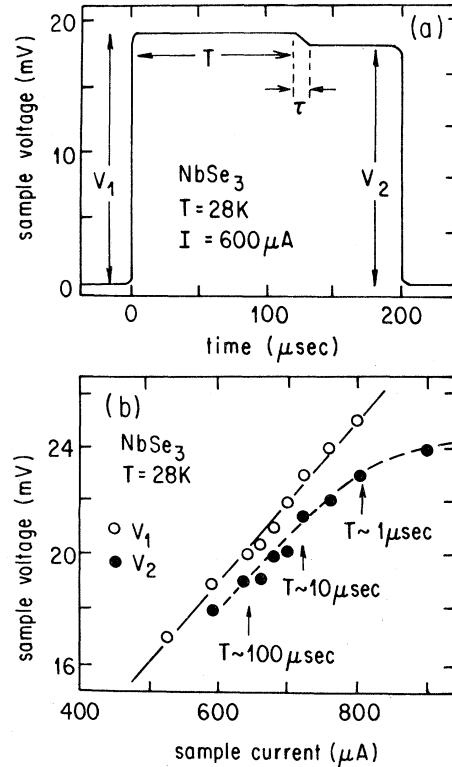


FIG. 2. (a) Digitally smoothed response waveform to a current pulse with I just above I_T . The switching phenomena is clearly seen at a time $120 \mu\text{sec}$ from the start of the pulse. Identified in the figure are the voltages V_1 and V_2 (corresponding to the nonconducting and conducting states), the time before switching T , and the switch duration τ . (b) I - V curves obtained from pulse measurements similar to the one shown in (a), where the voltages V_1 and V_2 are defined. The full line is the Ohmic conductivity, while the dotted line is a guide to the eye for the nonlinear conductivity. Typical values of the time to switch T , for a given sample current I , are also shown.

carrying state. The finite time before switching T fluctuates about a mean value \bar{T} , which strongly decreases with increasing I . Just above threshold, values of \bar{T} of the order of a few hundred microseconds were observed, while at $I \sim 1.25 I_T$ the switching occurs at such a rapid rate that only the current-carrying state is observed, as shown in Fig. 2(b). We also remark that the sudden onset of nonlinear conduction has also been observed recently by Monceau *et al.*¹² Hysteresis and switching phenomena, however, have not been reported by the authors.

In the absence of any theory which may account for our observations we give only a tentative discussion of the observed phenomena. We note first that neither the classical nor the tunneling model, in its present form, leads to a sudden onset of nonlinear

conduction at I_T . In the classical model, the frequency-dependent conductivity $\sigma(\omega)$ suggests an overdamped response,⁷ and consequently an inertial term is neglected. The consequence of this assumption is that the excess current associated with the moving CDW starts from $I_{CDW}=0$ at threshold, and only the differential conductivity dI/dV diverges at threshold. The inclusion of an inertial term in the classical model would lead to a sudden jump in the conductivity at I_T , and also to a hysteresis effect. Whether a model can be worked out which is compatible with the observed $\sigma(\omega)$, the switching phenomenon and hysteresis effect, and also with the detailed form of $\sigma(E)$ in the sliding conductivity region, remains to be seen.

The tunneling model result, Eq. (2), also leads to a smooth increase of the conductivity at $I = I_T$, and dI/dV has a small discontinuity at I_T . This discontinuity is absent in Fleming's empirical formula. With time-dependent phenomena not included, the tunneling model does not lead to the switching phenomena we report here. We note that if tunneling occurs between two macroscopically occupied states, leading to macroscopic quantum tunneling (MQT) as suggested by Leggett,¹³ then tunneling events may lead to switching phenomena. These were recently observed in Josephson junctions,¹⁴ where the distribution functions of tunneling and of thermally assisted transfer rates were measured. These lead to stochastic events, and the probability distribution of times before switching T , between T and $T + \Delta T$, is given by

$$P(T) \propto \exp(-T/T_0) , \quad (3)$$

where T_0 is a parameter dependent upon the current I .

In order to determine if the switching phenomenon is a stochastic or deterministic process, we have studied the response wave forms of a series of identical current pulses. We find that for a given current I , the time before switching T is not a uniquely defined quantity, but varies from one pulse to another. Since each switch leads to a sharp downstep in the measured sample voltage, one can obtain the distribution of T , for a given current I , by summing a substantial number of response waveforms and taking the derivative of the resulting sum. This is shown in Fig. 3 for two values of I , where we have taken the derivative of the sum of 64 successive pulse responses, recorded with a high-speed multichannel signal averager. In contrast to the exponential behavior suggested by Eq. (3), Fig. 3 shows a probability distribution for switching centered about a mean value \bar{T} . An increase in the pulse current I leads to a decrease in \bar{T} , and a sharpening of the distribution. This indicates that the switching behavior is a deterministic process. The full and dotted lines

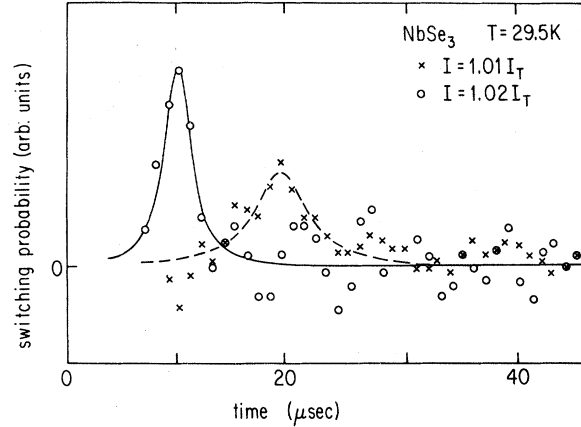


FIG. 3. Distribution of time to switch T for two different driving currents I . The full line is a fit to a normalized Lorentzian distribution with $\alpha = 6.32$, while the dashed line is for $\alpha = 3.07$ [see Eq. (4)].

in Fig. 3 are fits to the Lorentzian distribution

$$P(T) = \frac{\alpha}{\pi} \frac{1}{1 + \alpha^2(T - \bar{T})^2} , \quad (4)$$

with the parameter α given in the figure caption. For $I < I_T$, the probability distribution is zero everywhere, while for $I \rightarrow \infty$ a delta function $\delta(T=0)$ is expected. We conclude therefore that tunneling processes do not account for our experimental findings.

As remarked before, neither the classical, nor the quantum-mechanical description of the nonlinear conductivity is able to account for the sudden voltage jump associated with the development of the current-carrying CDW state. We believe that the reason for this, and the most likely explanation of our findings, is that a random distribution of CDW segments,¹⁵ which arise due to inhomogeneities, grain boundaries, etc., is neglected. The distribution of CDW regions leads *a priori* to a distribution of pinning energies. It has been suggested by Portis¹⁶ that such a distribution is important, and he has accounted for $\sigma(E)$ and $\sigma(\omega)$ by incorporating the effect of a distribution in the classical response of CDW segments.

We suggest that with the application of a current or electric field the various CDW regions have first to be coupled together before a coherent current-carrying state can develop. The switching phenomenon we observe is then associated with the finite time required for coupling after the application of a driving field. The situation is similar to that observed in coupled Josephson junctions,¹⁴ and in granular superconductors¹⁷ where Josephson coupled grains are observed. A single model which could account for our observations is the extension of the

classical model for weakly coupled particles moving in a periodic potential. A coupling term which depends on the relative position x of the particles (or on the relative phase of the CDW's) like $A \sin(x_i - x_j)$ describes the tendency of the CDW segments to be coupled. Due to the inherent nonlinearity of the problem coupled or uncoupled regions may be obtained depending on the driving forces. We remark that metastable states and hysteresis effects have recently been obtained in a model of coupled particles in a periodic potential.¹⁸

We also note that our observations are most probably closely related to the memory and long-time effects observed recently by Brill *et al.*,¹⁹ Gill,²⁰ and

Fleming.¹⁰ They have shown that there is a long exponential voltage buildup associated with the development of a steady-state current-carrying state, and the response depends on the direction of the applied electric field.

ACKNOWLEDGMENTS

We wish to thank T. Holstein, P. Chaikin, J. Bardeen, S. A. Wolf, and E. Ben-Jacob for useful discussions. This research was supported by the NSF under Grant No. DMR 81-03085. One of us (A.Z.) acknowledges support from an IBM fellowship.

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