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Charge-Density-Wave Switching Behavior in an Applied Temperature Gradient

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We have investigated the effect of thermal gradients upon switching crystals of the charge-density-wave (CDW) conductor $NbSe_3$. We find that drastically different effects occur depending upon the zero gradient nature of the I-V characteristic. In samples which normally depin via a single switch a finite gradient tends to break this switch into a number of much smaller switches, while samples which normally depin via a number of small switches are unaffected by the gradient. From these results we determine the nature of the distribution of ultra-strong impurities which give rise to switching in $NbSe_3$.

The quasi-one-dimensional conductor $NbSe_3$ forms in long, thin needle-like fibers.[1] The material undergoes a pair of charge-density-wave (CDW) transitions at T_{c1}=144K and T_{c2}=59K. In both cases the CDW is only pinned to the lattice by relatively weak impurities and can be forced to slide along the length of the sample by applying modest (~10 mV/cm) electric fields along the crystal chain axis. The sliding CDW gives rise to nonlinear conductivity and a decrease in the crystal's differential dc resistance past the threshold field E_T. The depinning is usually evident in the current-voltage (I-V) relationship as a smooth, continuous change from the pinned linear state to the higher conductance sliding nonlinear state; well past E_T the CDW conductance saturates and the differential resistance becomes field independent. The sliding CDW has a number of intriguing properties associated with it, including narrow band and broad band noise oscillations[2] as well as mode locking[3].

In some samples of $NbSe_3$ the CDW depins in a sharp, hysteretic manner which gives rise to a strong discontinuity in the I-V curve. This phenomenon, referred to as *switching*, was first observed in the low temperature CDW state of $NbSe_3$.[4] Since its discovery, switching has been observed in nearly all materials which display CDW conduction, including TaS_3,[5] $K_{0.3}MoO_3$,[6] and $(NbSe_4)_{3.33}I$.[7] Switching can also be induced by iron doping $NbSe_3$[8] or by irradiating either TaS_3 or the blue bronzes.[9] This suggests that an impurity-based mechanism is responsible for switching. In $NbSe_3$, switching only occurs over a limited temperature range, extending from roughly 20K to 35K although in some rare instances it has been observed at temperatures as high as 40K.[10] The dynamic switching state is characterized by a temperature independent depinning field,[10] hysteresis,[11] negative differential resistance,[12] inductive ac response,[13] and period-doubling routes to chaos.[14]

The depinning process in switching samples can proceed via either single or multiple switches. We subdivide switching samples into two classes according to the number of switches evident in their I-V characteristics. In what we call type I samples the CDW depins largely via a single predominant switch although other, much smaller switches might also exist in the I-V characteristics. Type II samples depin via a number of smaller, roughly equal-sized switches. Both type I and type II behavior has previously been observed in switching samples of $NbSe_3$.[15]

In this Communication we report the first observation of temperature gradient effects on switching samples of $NbSe_3$. We find that the two classes of switching samples are affected by a thermal gradient in drastically different ways. Type I switches are broken apart by a gradient to form numerous small switches while type II switches are not further divided by a gradient. These very different results allow us to determine the nature of the distribution of the ultra-strong impurities which give rise to switching behavior in $NbSe_3$.

Recent work has indicated that switching in both $NbSe_3$ and Fe_xNbSe_3 is associated with the separation of the CDW into distinct, serially arranged macroscopic phase-velocity-coherent domains with independent CDW threshold fields and phase velocities.[15] These domains were found to be separated by localized phase-slip centers which form the interface between neighboring CDW domains. These phase-slip centers are presumed to consist of ultra-strong impurity sites.[16] The switching CDW depins locally when the dc bias exceeds the large phase-slip depinning field. Hence, switching crystals are comprised of a number of macroscopic current domains with independent threshold fields. This leaves open the question at to whether or not the pinning within a switching domain is non-uniform. Earlier work has shown that sublevels within hysteresis loops exist in crystals which display relatively strong switching behavior.[11] This suggests that non-uniform pinning within otherwise coherent domains may exist.

A powerful experimental technique for the examination of intra-domain structure consists of the study of CDW transport in a longitudinally applied temperature gradient. This method has proven to be useful in the study of phase-velocity-coherent domains in non-switching samples where temperature gradients cause the CDW to break up into separate and distinct current carrying domains.[17-21] This technique exploits the temperature dependence of the parameters which characterize the CDW condensate such as the low field resistivity and CDW carrier concentration. Unlike non-switching $NbSe_3$ crystals, the threshold field E_T in switching crystals is independent of temperature.[10] As a result, the temperature dependent threshold current $I_T \sim E_T/\rho_o$ will scale inversely with the changing low field resistivity ρ_o. In the low temperature CDW state of $NbSe_3$ ρ_o is an increasing function of temperature, and I_T will therefore be a decreasing function of temperature. Hence, a temperature gradient can be used to study the internal structure of a sliding switching domain.

The samples used in these experiments were single crystals of $NbSe_3$ produced by conventional vapor-transport methods. The experimental arrangement consisted of single switching crystals suspended in vacuum between two large copper mounting posts anchored to peltier heater chips. Electrical contacts were made to the samples with silver conductive paint. This two probe mounting arrangement allowed the temperature of both ends of the sample to be independently varied. Thermometry was accomplished by a diode sensor and several miniature differential thermocouples. All I-V curves presented in this paper were measured using a current driven configuration.

The switch-depinning threshold current, I_T, is a strong function of temperature in the switching regime. Fig. 1 displays the I-V characteristics of a type I switching sample of $NbSe_3$ measured at selected temperatures between 24.6K and 29.3K. In all cases there is no temperature gradient on the sample. The crystal shows negative differential resistance at 29K and clear switching below 27K. Below 26K the CDW depinning proceeds via two independent switches, labeled S_1 and S_2 in the figure, indicating that two macroscopic current carrying domains exist within the crystal. These two domains are possibly separated by a phase-slip center. The two switches become hysteretic below 25.5K, and no additional switches are observed below 24K.

The effects of a temperature gradient on this same crystal are shown in Fig. 2. The temperature of one end of the sample was held fixed at T_0=24K, while the other end was warmed to a higher temperature, $T_0+\Delta T$, ranging between 24K to 28K. The curves in Fig. 2 are labeled by the temperature difference ΔT. The length of the sample was L_s=0.3 mm and the largest gradient applied was therefore 13.3 K/mm. The data in Fig. 2 demonstrates that applied thermal gradients have a strong effect upon the switching characteristics of type I $NbSe_3$. As the temperature gradient is increased the

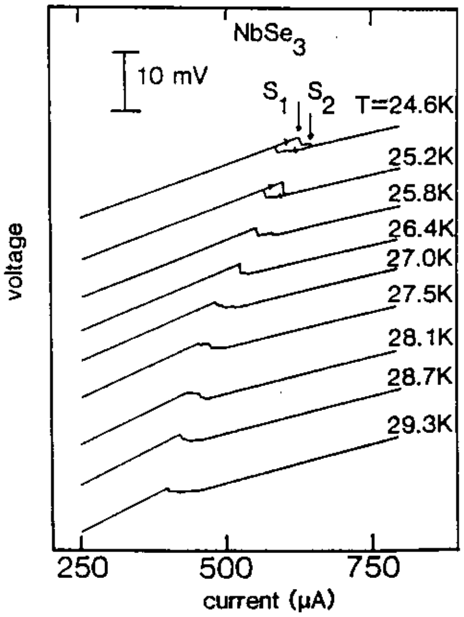

Fig.1: Zero gradient current driven I-V characteristics for a type I $NbSe_3$ switching crystal at temperatures between 24K and 29K. At low temperatures two distinct hysteretic switches, S_1 and S_2, occur and are identified by the vertical arrows.

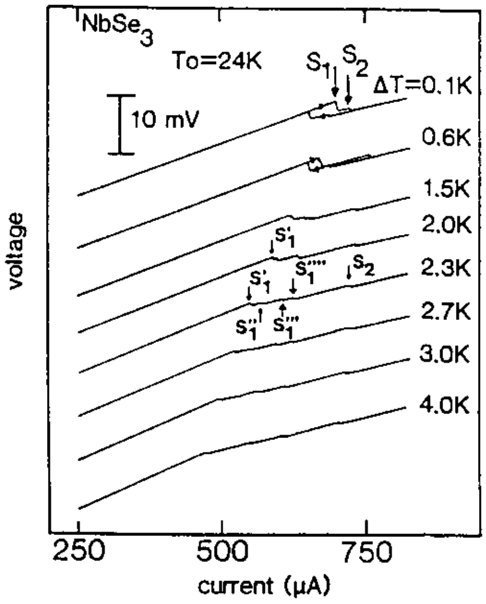

Fig.2: Current driven I-V characteristics for the same type I $NbSe_3$ sample used in Fig. 1, but in the presence of a temperature gradient. The cold end of the sample is held at T_0=24K while the hot end is at $T_0+\Delta T$. With increasing ΔT, switch S_1 breaks up into a series of smaller switches S_1', S_1'', etc., as identified in the ΔT=2.3K trace.

dominant switch, S_1, appears to break up into a series of smaller switches, labeled S_1', S_1'', and S_1''' in the ΔT=2.3K trace of Fig. 2. These smaller switches continue to break up and move to lower current biases as the temperature gradient is further increased. The smaller switch, S_2, appears essentially unaffected by the temperature gradient. This general behavior is not unique to this sample; it has been observed in all type I switching samples which we have investigated in a temperature gradient.

We associate the break up of the original isothermal switch S_1 in Fig. 2 with the subdivision of a switching domain near the "hotter" end of the sample (see below). This large switching domain gives rise to the switch S_1 in the absence of the gradient. As the gradient is increased the domain breaks up into a number of subdomains, each depinning at a current equal to the average threshold current in that subdomain. The depinning current drops with an increase in subdomain temperature because the low field resistance is an increasing function of temperature. The initial, lowest threshold switch S_1' must correspond to a small domain adjacent to the "hot" end of the sample. This is demonstrated in Fig. 3 where the depinning threshold current of switch S_1' is plotted as a function of temperature [assuming T(domain S_1')=T_0+ΔT] along with the temperature dependence of the threshold current of the zero gradient switch S_1 as deduced from Fig. 1. The two thresholds match closely throughout the temperature range from 24K to 28K. This demonstrates that the thermal gradient causes the original switching domain, which gave rise to switch S_1, to break apart.

We have performed similar thermal gradient experiments on type II switching samples of $NbSe_3$. As displayed in Fig. 4, thermal gradients do not further subdivide these samples into more independent subdomains. The thermal gradient only acts to move the switching onset to lower threshold currents, as expected when the local switching domain is raised in temperature. Thus, it would appear that type II switching samples, already under the influence of an internal chemical "gradient", are unaffected by an externally applied thermal gradient. We note that in some cases samples which show type I behavior at low temperatures display type II behavior (multiple switches in their zero gradient I-V characteristics) at elevated temperatures. In all cases the I-V characteristics of these samples in their type II state are unaffected by a thermal gradient and are qualitatively in accord with Fig. 4.

We now analyze these results in order to determine the nature of the distribution of ultra-strong impurities which give rise to switching. The data in Fig. 2 indicate that a temperature difference as small as 2K is sufficient to break a phase-velocity-coherent switching domain into four subdomains over a distance of only 300 μm. This length is far smaller than the sample length required for a thermal gradient to divide this type of domain in a non-switching sample of $NbSe_3$.[17-21] The much larger effect in switching $NbSe_3$ clearly indicates that CDW impurity pinning is non-uniform within switching domains. These apparently coherent switching domains must actually consist of a number of coupled

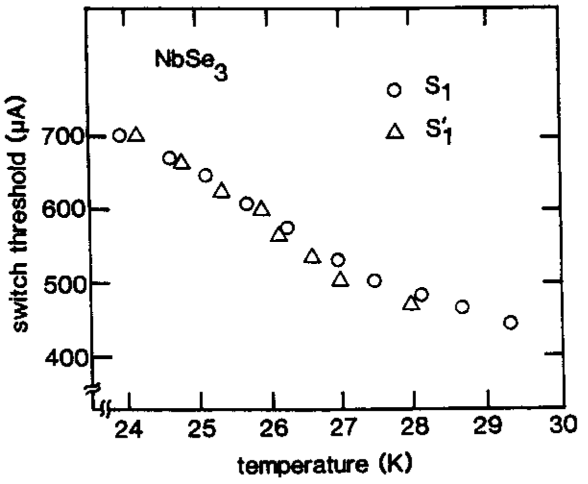

Fig.3: Threshold current versus temperature for switching to occur in the same type I $NbSe_3$ crystal used in Figs. 1 and 2. The open circles refer to switch S_1 of Fig. 1 (zero gradient) while the triangles refer to the first switch S_1' of Fig. 2 (with a gradient). The temperature used when plotting $I_T(S_1')$ is T_0+ΔT. The close correspondence of the two data sets indicate that S_1' is associated with a switching current domain very near the "hot" end of the sample.

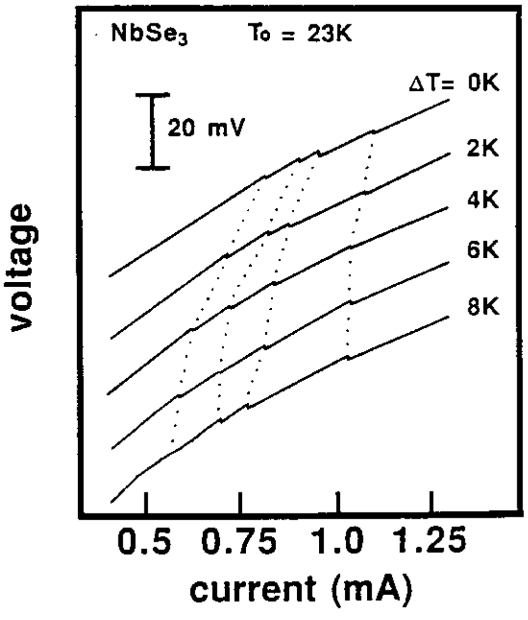

Fig.4: Current driven I-V characteristics in a temperature gradient for a type II $NbSe_3$ crystal. In this sample the original four switches do not divide into smaller switches when the gradient is applied. The dotted lines are guides to the eye showing the progression of switching onsets as the gradient is varied.

subregions, with an average length of not more than 60 μm. Neighboring subregions are linked by ultra-strong impurity sites which cause switching. These impurity sites must have isothermal depinning thresholds which are roughly equivalent so that the entire set of subregions depin in a synchronized fashion when $\Delta T=0$. In this isothermal configuration, the ultra-strong impurity sites will not form phase-slip centers because each subregion moves at the same domain velocity. These subregions can only be synchronized if no thermal gradient is present. Because of the internal strain brought on by a temperature gradient, the domain will break into subdomains. In this situation, phase-slip centers will form on ultra-strong impurity sites in order to link neighboring CDWs together. In a non-switching crystal, the absence of ultra-strong impurities allow large velocity-coherent domains to stay coupled in the presence of relatively large thermal gradients.

A chemical "gradient" must exist within type II switching samples in order to account for the many switches evident in the zero gradient I-V characteristics of these samples. It is this gradient which causes CDW subregions to depin at widely varying threshold currents and fields. As a result, the depinning of the subregions will not be synchronized even in the absence of a thermal gradient.[22] Because the crystal is already separated into multiple domains, an applied thermal gradient is unable to break the sample into a greater number of subdomains. The chemical gradient which gives rise to this behavior could be in the form of large variations in either the local impurity or lattice defect concentrations.

The data from type II switching samples, such as those depicted in Fig. 4, indicate that the domains in these samples cannot be broken into subdomains even in gradients which break type I samples into far more domains. As an example, the data shown in Fig. 2 indicates that this type I sample is divided into roughly 10 domains by a 13 K/mm gradient, while the data shown in Fig. 4 indicates that this type II sample consists of only four domains even in thermal gradients as large as 20 K/mm. This indicates that the impurity sites may not be as widely and continuously distributed in type II samples as they are in type I samples. This would suggest that a continuous distribution of ultra-strong impurity sites may be an important requirement for the synchronized switch depinning of a large domain. A continuous distribution of pinning sites may force the local threshold fields to vary only slightly along the domain length, and in so doing link together the subregions to form a larger, more coherent domain. This situation could explain why the local threshold currents and fields seem to widely vary in multi-domained type II samples that are also unaffected by a thermal gradient.

We now consider the mechanism by which large switching domains in type I samples break up into smaller subdomains under the influence of a temperature gradient. To more clearly understand the form of the strain on a phase-velocity-coherent switching domain in a thermal gradient we must consider the elastic nature of the CDW phase. In the presence of a thermal gradient the local parameters which characterize the CDW within a domain will change. In order to keep the domain coupled, the CDW phase must deform to account for the variation in the domain parameters. As the thermal gradient is increased, the CDW parameters will vary over a wider range of values and the resulting strain on the CDW phase must increase. When the phase strain energy exceeds the energy required to form a phase-slip in the CDW, the domain will divide and form two subdomains with the new domain velocities more closely matching the local conditions. Because the sub domains can match local conditions more closely, the total phase strain energy of the two domains can be lower than that of the single domain. A phase-slip center will form at an ultra-strong impurity site, and will be located at the interface between the two new subdomains. Presumably the existence of many ultra-strong impurities in switching samples explains why these samples are much more readily affected by a thermal gradient than are non-switching samples. The non-switching samples will contain no ultra-strong impurities and the phase-slip energies in these samples will be much larger as a result. Hence, much larger thermal gradients will be needed to build up phase strain energies sufficient to energetically require the splitting of a domain and the formation of a phase-slip center.

In conclusion, temperature gradient studies indicate that type I and type II switching samples differ only in the distribution of ultra-strong pinning sites within them. Type I samples contain closely spaced impurity sites which give rise to synchronized depinning and the formation of *large* switching domains. These domains are very susceptible to the influence of external perturbations which tend to break them into a number of subdomains. Type II samples, on the other hand, contain relatively few ultra-strong impurity sites and do not undergo synchronized depinning. As a result these samples display multiple switches and form *small* switching domains. Due to the sparsity of impurity sites, these small domains are unaffected by external perturbations.

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