

Electrical-noise measurements on chromium films

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Electrical-noise measurements on chromium films prepared by molecular-beam epitaxy revealed details of the dynamics of both the polarization (η) and wave-vector (Q) domains of the spin-density waves. Individual resistivity steps from Q domains were easily resolved, from which the domain sizes, the kinetics of individual domains, and domain interactions could be inferred. Similar measurements were made on η domains and statistical analysis of the variance in the octave noise power indicates they also switch between discrete stable orientations. The asymmetry in the noise magnitude along different crystal directions was measured and related to the underlying symmetry.

INTRODUCTION

The magnetic structures of the antiferromagnetic phases of single crystals of Cr are well known and, for the most part, well understood.¹ Below the Néel temperature T_N (311 K in an unstrained crystal) and above a second (spin-flip) transition temperature (123 K in an unstrained crystal) a linearly polarized, discommensurate transverse spin density wave (TSDW) forms, breaking the cubic symmetry. Both the wave vector Q and the orthogonal polarization direction η lie along (100) type axes, at least in the ideal case. If no special precautions are taken, such as field or stress cooling,² the Q -vector orientation breaks up into domains. Within the Q domains, the η -vector orientation breaks up into what are believed to be domains along the remaining two directions.

The Q domain walls are not thermodynamically stable, since once a single- Q sample has been formed by cooling in a magnetic field it requires a great deal of magnetic or mechanical stress to create new domains.² Thus the Q domain walls form in metastable configurations primarily for kinetic reasons. However, the behavior of the nonuniform polarization in the TSDW in Cr has been a long-standing mystery.³ Sluggish domain kinetics alone cannot account for the existence of multiple η domains because a multidomain structure reforms, with slight hysteresis, after the η is aligned by an external magnetic field.⁴

It is likely that in a pure crystal a continuous fourfold anisotropic term in the Hamiltonian of the form proposed by Steinitz *et al.*⁵ to fit susceptibility anisotropy data is present. The long-range strain interaction term⁵ favors alignment of the polarizations, which is unlike the magnetic field energy term for a ferromagnet, and so there is no reason for a multidomain sample to be more thermodynamically stable than a single domain sample in an ideal crystal.

An early proposal that the domain size was limited by the strain energy required to make a small 90°-rotated domain in a region that was macroscopically homogeneous⁵ does not seem to account for the reformation of an apparently random polarization structure after magnetic

alignment unless fixed strains cause different regions to prefer different polarizations at zero field. One could, in principle, account for a finite typical domain size by postulating a very low energy for domain-wall formation⁵ together with a repulsive interaction between domain walls, the latter of which would prevent the domain size from shrinking to zero. However, no physical justification for the existence of such an interaction has been proposed, to the best of our knowledge. Thus there is strong reason to suspect that the η domain structure is caused in some way by deviations from the ideal Hamiltonian, i.e., by defects, but neither the thermodynamics nor the kinetics of the η domains is clearly understood.

In this paper we use spontaneous fluctuations in the anisotropic resistivity tensor⁶ caused by domain rotations to study the dynamics of both Q and η domains in single crystal Cr films prepared by molecular beam epitaxy (MBE). The resistivity fluctuation technique is an unusual probe, in that it may be used on samples small enough to allow observation of individual domain dynamics. In addition, measurements of symmetry, similar to those which are obtainable with techniques such as internal friction, can be made, especially when single-crystal samples are used. The results indicate that both vector types (η and Q) show discrete switching; that different domains interact significantly with each other;⁷ that the Q configuration usually settles to a unique free-energy minimum in a given sample at a given temperature; the η configuration can end up in any of a large set of local free-energy minima; and that the principal axes of the anisotropic conductivity tensors are not completely aligned along the cubic axes.

BACKGROUND: PREVIOUS NOISE RESULTS

In most metal films the slow resistance fluctuations ($1/f$ noise) appear to be dominated by the effects of defect motions.^{6,8} The principal temperature dependence of such noise is due to the thermally activated kinetics. The temperature dependence of the magnitude is approximately described by a Dutta-Horn relationship,^{6,8} and is never very strong when the spectral form is close to $1/f$. In contrast, Scofield *et al.*⁹ noticed that in some well-

annealed Cr films the noise spectral density rose between one and two orders of magnitude below the approximate bulk Néel temperature (T_N), despite the lack of any interesting features in the spectral form, which suggested that the antiferromagnetic phase was somehow responsible for the large noise magnitude.

Subsequent experiments^{10,11} on a variety of polycrystalline films (all 90 to 220 nm thick) confirmed that the large noise was a property of the TSDW phase. Several facts led to that conclusion. First, both the small magnetic susceptibility anomaly near the transition temperature, which we denote T_N , which is quite difficult to measure in films, and the temperature of the noise rise, henceforth denoted T_n , (defined as the temperature with the maximum temperature derivative of the noise power) were measured on the same film and were found to coincide within experimental error. Second, two otherwise identical films with different strain were prepared by using substrates with different thermal expansion coefficients. The shift in T_N from the strain could be calculated,¹² and it corresponded very well with the shift in T_n . Third, in films with fewer defects, as determined by the residual resistivity, the noise rise was sharper and occurred at higher temperature, consistent with the expected effect on T_N . In MBE films the noise rise was extremely sharp and occurred close to the bulk T_N .¹¹

Measurements of a noise symmetry parameter,⁶ S , near 200 K, well below T_n , showed the noise is the result of η rotations. This parameter is defined by

$$S = 2 \langle \det[\delta \mathbf{R}] \rangle / \langle \text{Tr}[\delta \mathbf{R}^2] \rangle,$$

where $\delta \mathbf{R}$ is the 2D projection of the resistivity tensor fluctuation onto the sample plane. S is measured in cross-shaped samples, for which the different tensor components of the resistivity are simultaneously measurable. S ranges from 1 for purely scalar resistivity fluctuations, to -1 for traceless fluctuations, which correspond to rotations of a locally anisotropic resistivity tensor about an axis normal to the plane. The parameter S has an invariant definition, without reference to a particular set of axes, and is often measured in amorphous or polycrystalline samples in which the principal axes may be assumed to be randomly oriented with respect to the local current density vector. S was found to have negative values for temperatures well below T_n in Cr, indicating that the noise is the result of rotations of something.⁶ Further, when the tensile strain in the film plane was small, S was more negative. The \mathbf{Q} orientation is particularly sensitive to strain, and so \mathbf{Q} fluctuations out of the plane would be suppressed in high-strain samples, making S from \mathbf{Q} fluctuations more negative, which would be inconsistent with the results. However, rotations of the polarization vector η , of the TSDW around the \mathbf{Q} vectors would result in the observed change in S .

The results indicating that the resistivity noise at 200 K comes from η rotations were to be expected, since internal friction from η rotations is known to be present,^{1,3,13,14} which implies that strain fluctuations are present in thermal equilibrium through a fluctuation-dissipation argument. Barring an accidental equality of

two of the principal components of the resistivity tensor, these strain fluctuations would be accompanied by noise, although the magnitude could not be estimated *a priori* since there were no prior measurements of the resistivity anisotropy between the η direction and the $\mathbf{Q} \times \eta$ direction.

In samples of about 10^{-12} cm³ volume, the spectral density showed significant random deviations from a $1/f$ form, indicating that the noise came from a relatively small number of independent sites which each contribute a Lorentzian to the spectrum.^{6,9,15} (This result also contrasts with defect $1/f$ noise, which generally shows no features except in much smaller samples.)⁶ The 200-K noise was also slightly non-Gaussian, as would be expected if it came from a collection of independent systems fluctuating between discrete states.^{6,15} From a rather complicated argument, which is presented in the discussion of this paper, it was concluded that each polarization rotation encompassed a volume of about 10^{-16} cm³. Furthermore, the small-sample results showed, for the first time, that the fluctuating sites for η were stable for long periods at a given temperature but were randomly and irreversibly altered by very small temperature cycles (e.g., from 200 to 220 K and back).

EXPERIMENTAL TECHNIQUES

We have used films made by several techniques¹⁰ but the new experimental results reported here concern films prepared by molecular beam epitaxy (MBE).¹⁶ The first film was polycrystalline, and thus differences in the noise behavior along different crystal directions could not be distinguished. The second film was a single crystal film 85 nm thick grown with a 2 nm thick Nb underlay on sapphire. The sample plane was normal to a (110) type direction. We designate that direction to be (1 $\bar{1}$ 0) so that the directions in the plane may be consistently described with positive indices.

The single-crystal film had a residual resistance ratio [$R(300 \text{ K})/R(4 \text{ K})$] of 9 and a room temperature resistivity of 17 $\mu\Omega$ cm. The normalized temperature coefficient of the resistivity above T_N was

$$\partial \ln \rho / \partial T = 0.0029 \text{ K}^{-1}$$

The crystallographic orientation of the film was verified to $\pm 4^\circ$ using x rays, and the FWHM mosaic was 0.4° . The films were grown at 800 K and thus a differential linear thermal expansion strain of 5×10^{-4} exists at room temperature.¹⁷

The films were patterned into small resistor networks using conventional photolithographic techniques. Mesoscopic samples were made using a shadow masking step edge technique developed for this experiment. The networks were two arm or four arm bridges similar to those used in AC resistance measurements.^{18,19} Such balanced bridge circuits are relatively immune to temperature fluctuation noise and to fluctuations in the current source. Typical sample volumes for our larger samples were 2×10^{-10} cm³.

One sample network was made from resistor arms oriented along different crystalline axes, as shown in Fig. 1. Two arms ran parallel to the [111] direction, one to

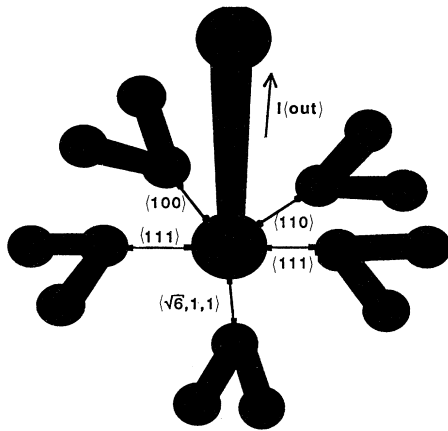


FIG. 1. The 85 nm thick MBE single-crystal film has a sample plane normal to a (110) type axis. The macroscopic pattern shown was patterned using standard photolithographic techniques. The dark regions represent areas where the Cr remains. The arms are the narrow constrictions ($7\ \mu\text{m} \times 350\ \mu\text{m}$) which are oriented along the $\langle 100 \rangle$ -, $\langle 110 \rangle$ -, $\langle \sqrt{6}11 \rangle$ -type axes, with two along a $\langle 111 \rangle$ -type axis. A [111] arm forms a bridge with one of the other four with current flowing out in the direction shown. Each arm has a voltage and a current lead.

[001], one to [110], and one to the $[\sqrt{6}11]$ (an arbitrary angle between [001] and [111] direction). After patterning, the angles were uncertain to 7° (4° systematic, 5° random). These arms could then be used in various combinations to make a two-arm bridge, allowing the relative noise magnitude along the different directions to be compared.

In order to make smaller samples, about $10^{-13}\ \text{cm}^3$, suitable for identifying individual fluctuation units, step-edge techniques were used.²⁰ Since it was not feasible to grow MBE films on preexisting substrate steps, a new step-edge technique was developed. After a photoresist step is made on the Cr film, ion milling and subsequent evaporation at slightly different angles produces a narrow line of SiO, under the shadow of the edge. The photoresist and any exposed Cr is then removed, leaving a narrow (e.g., 200 nm) line of Cr under the SiO.

The resistance fluctuations were measured using an ac driving current ($f=1\ \text{KHz}$) with current densities $\langle 6 \times 10^6\ \text{A/cm}^2$ to avoid excessive sample heating. The quadratic dependence of the spectral density on current was periodically verified. A Stanford Research 552 low-noise preamplifier was used followed by a lock-in amplifier for phase sensitive detection. The voltage was typically sampled at a rate of 500 Hz, and the voltage sweeps were processed by a Skymnk array processor in a Digital Equipment Corporation LSI 11/23 microcomputer. Typically 400 squared fast Fourier transforms (FFT's) were averaged together to obtain a noise power spectrum requiring about 20 min of data collection, which results in a statistical uncertainty in the noise power of less than 3% in each octave in the frequency band from 0.3–150

Hz. (The frequency where the signal to background white noise ratio is one was about 20 Hz for the typical sampling current used.) A Linear Research 130 temperature controller and a Lakeshore diode temperature sensor stabilized the temperature to 0.2 K. An Air Products continuous flow cryostat was used for the anisotropy portion of the experiment.

RESULTS AND DATA ANALYSIS

Macroscopic samples

A sharp change of more than two orders of magnitude appeared in the noise spectral density of the polycrystalline samples as illustrated in Fig. 2, where the sample-independent Hooge^{6,8} noise parameter α is plotted as a function of frequency. α is defined

$$\alpha = [S_v(f) f N_s] / (V^2),$$

where V is the RMS sample voltage, and N_s is the number of atoms in the sample. The single-crystal (100) data are also shown. These data represent eight separate temperature sweeps. The T_n is 323 K, close to the bulk T_N of 311 K. This difference can be approximately accounted for, by considering the strain on the crystal due to the differential thermal expansion of the film and the substrate.^{12,21} In the same films, the resistive anomaly at the transition was measurable, but quite small, as seen in the temperature derivative of the resistance also shown in Fig. 2. The extraordinary sensitivity of the electrical noise to such weak symmetry breaking as the Cr SDW is particularly evident in these clean films, both because the transition is sharp and because there is little defect noise above T_n .

In single-crystal samples, resistance fluctuations can be measured along different crystal axes, rather than averaged over orientations as in polycrystalline samples. If the η and Q vectors indeed lie only along the principal crystal axes, then the resistance fluctuations which result

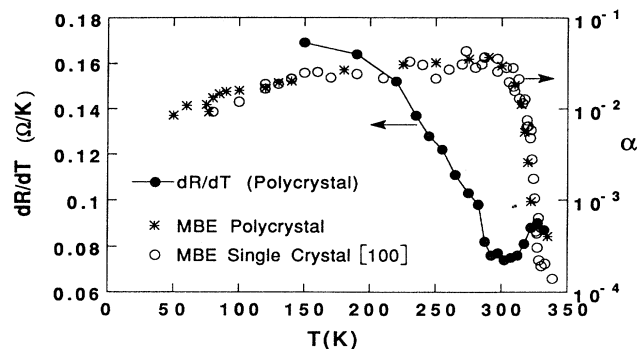


FIG. 2. The Hooge noise parameter, α increases by two orders of magnitude near the bulk Néel temperature. MBE polycrystal Cr and MBE single-crystal show the same behavior. The resistance anomaly is also observable in the same polycrystal sample, indicating the rise in the noise is a result of the magnetic ordering.

from switching between these orientations should show a distinctive directional symmetry. The resistance along the [111] direction is independent of which axes are occupied by Q and η . Thus if the current were completely along the [111] direction and the TSDW showed ideal behavior, no TSDW noise would be observed.

If the TSDW principal axes coincide with the cubic axes the variation in conductivity noise power for a direction in the $(1\bar{1}0)$ plane at angle Ψ from the [001] direction is proportional to $(3 \cos^2 \Psi - 1)^2$. Thus the [001] direction should show the most noise and there should be a minimum in the [111] direction. In addition, there should be a second peak, $\frac{1}{4}$ as large as the [001] peak, in the [110] direction.

Our measurements of the noise magnitude along different axes in a temperature region near the transition are shown in Fig. 3(a), and the expected qualitative anisotropy is evident. Figure 3(b) shows the noise power over our full temperature range, along with the ratios of the noise power in the [110], and the [111] directions to the [100] direction. The results show that the noise along the [111] direction is substantially reduced from the noise along the other directions measured. Just below T_n the magnetic noise along the [001] direction has about six

times the magnitude of the magnetic noise of an identical sample in the same crystal along the [111] direction. (The ratio of the overall magnitude is not quite this large, because the nonmagnetic noise which was present above the transition, whose temperature dependence we presume to be weak, can be treated as a background and subtracted out.) Since the field lines at the ends of the samples are not parallel to the sample itself, and since any lithographic imperfections also induce field lines along other directions, the ratio of 6 should be taken as a lower limit to the actual ratio of the resistivity noise along the [001] to the [111] direction. As the sample is cooled to 295 K, this ratio drops to about two, and remains approximately constant down to 80 K. Since the sample geometry is unchanged, the change of symmetry is a real property of the fluctuations.

It would be extremely difficult to account for the increasingly strong symmetry signature near the transition with a single type of fluctuation, since one expects a particular TSDW vector to become more strongly constrained to lie along an easy direction as the SDW further condenses. A plausible interpretation is that near the Néel temperature Q fluctuations which are strongly tied to particular Fermi surface directions are dominant. As the temperature is lowered these tend to freeze out, leaving only η fluctuations. Magnetic field studies on bulk Cr show that η is not strongly tied to the cubic axes at room temperature,^{1,4} so the locally preferred orientations may be strongly influenced by random local strains. Thus η could switch locally between orientations not along principal cubic axes, which would then contribute conductivity fluctuations along a [111] axis. However, the $\langle 001 \rangle$ -type axes remain preferred, so that even for η noise the [111] direction remains relatively quiet. The interpretation in terms of two different types of domains also fits well with results on individual switching events observed in mesoscopic samples, discussed below.

That the magnetic noise magnitude along the [110] direction is intermediate between those along the [111] and [001] in our entire temperature range, is expected. However, at no temperature does the ratio of the noise along the [110] direction to that along [001] approach the expected value of $\frac{1}{4}$. In fact, the ratio never drops below $\frac{3}{5}$. Furthermore the ratio of the [110] magnitude to the [001] magnitude shows similar temperature dependence to the ratio of the noise magnitude along the [111] direction to that along the [100] direction, though the minimum is broader and less dramatic. This result also points toward a different noise generating mechanism just below T_n . The noise below 250 K, which we interpret as η fluctuations, shows relative magnitudes which are reasonably consistent with a picture in which η rotates about cubic axes but is not itself strongly tied to these axes.

There is a possible explanation for why, near T_n , the [110] magnitude is close to the [001] magnitude while the [111] magnitude is so small. The possible Q directions could be rotated toward the sample plane as a result of strain and/or surface effects. If this were the case, Q switching would give some noise along [111] and nearly as much noise along [110] as along [001]. An admixture

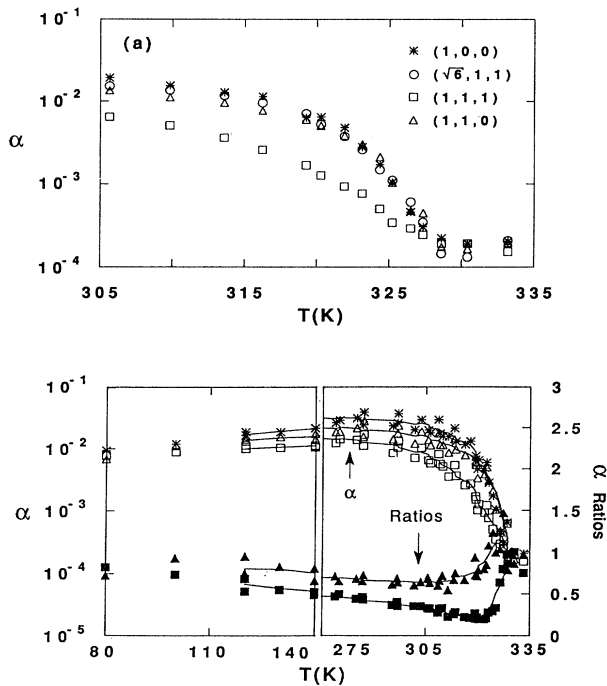


FIG. 3. The anisotropy in the noise along the different axes qualitatively reflects the underlying crystal symmetry. However, the quantitative values deviate significantly from the expected relationship. (a) The symbols represent the noise in the directions shown. The anisotropy persists down to 80 K as shown in (b). A fit to the data is shown to help guide the eye. The ratio $\alpha([111])/\alpha([100])$ has a distinct minimum at 320 K (solid squares). The ratio $\alpha([110])/\alpha([100])$ is also shown (solid triangles).

of η rotations about Q vectors preferentially located in the plane (hence along [001]), with η orthogonal to Q but otherwise not respecting the crystal symmetry, gives no noise along [001] and $\frac{3}{4}$ as much along [110] as along [111]. However, in view of anomalous features appearing in the magnitude ratios above 320 K and the very large temperature coefficient of the noise in that regime, we hesitate to place much weight on any interpretation of the detailed magnitude ratios in that regime.

The data displayed a $1/f^\delta$ form with $\delta=0.95\pm 0.05$ at all temperatures, except just below the transition, where δ dropped slightly to $\delta=0.90\pm 0.05$. No evidence of the spin-flip transition was observed, and it is assumed to be suppressed in these films.¹ Also, no evidence of an enhanced asymmetry is seen near 220 K, at which temperature the effect of the Q orientation on the lattice parameter changes sign in bulk crystals.²² Thus we do not believe that the Q vectors reorient in our samples at 220 K.

Mesoscopic samples

Figure 4 shows the resistance of one of the small samples at several temperatures as a function of time near the transition. It takes no special experience with noise

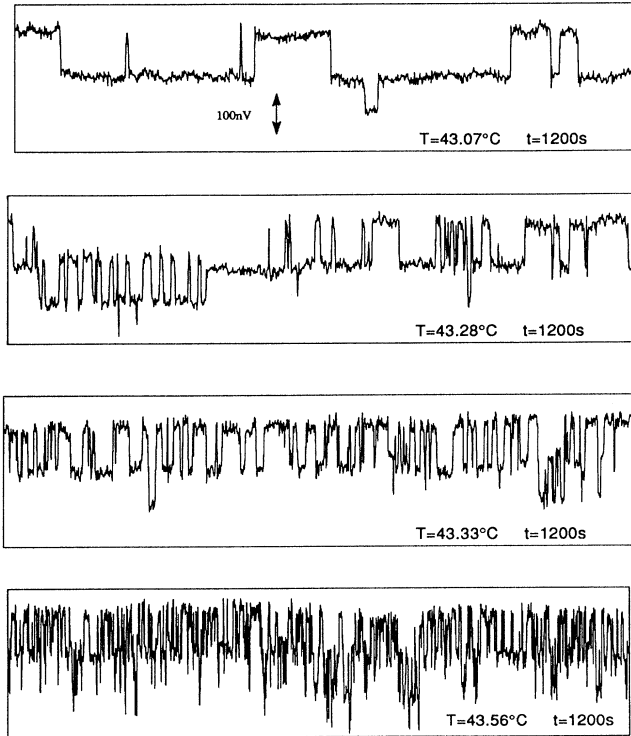


FIG. 4. In a single-crystal sample with volume $2 \times 10^{-13} \text{ cm}^3$ (sample A) switching behavior in the voltage near the transition could be tracked over about 1 K at 316 K. Voltage as a function of time is shown for 1200 sec at four temperatures. The arrow represents a 100 nV change in voltage, or fractional change of $(\delta V/V) \cong 10^{-5}$. The temperature dependence of the characteristic relaxation rates for the different levels is highly non-Arrhenius.

analysis to see that the resistance makes discrete switches between well-defined levels. All such switching events disappear above the Néel temperature and thus must be associated with switching of some degree of freedom of the TSDW.¹ One possibility is that the TSDW order is forming and breaking in small domains. Another is that the Q vectors of domains are switching between allowed states. A third possibility is that η is switching between different orientations. We shall explain why the second possibility—equilibrium Q domain switching—fits the data best.

The approximate domain size, or at least a minimum estimate, is easy to calculate from the size of the resistance changes. The fractional change in sample resistance during the discrete steps in Fig. 4 is 1.5×10^{-5} , so that the product $\beta_Q V_Q \approx (\delta V/V) V_s \approx 5 \times 10^{-18} \text{ cm}^3$, where β_Q is the fractional change in the diagonal component of the resistivity tensor due to Q domain orientation along the current direction, $\delta V/V$ is the fractional voltage change, V_Q is the volume of the domain or region over which the switch occurs, and V_s is the full sample volume. Since these steps were observed at 2 K below the apparent transition temperature, one does not expect that the anisotropy of the conductivity tensor for Q switching of the fully condensed TSDW ($\beta_Q \cong 6 \times 10^{-2}$) should be found, but rather $\beta_Q \cong 1 \times 10^{-2}$.²³ At 7 K below the transition β_Q is about 2×10^{-2} , or twice as large, in the bulk, and the steps are found to be larger by a factor of 1.7.

If we assume that Q switching is the mechanism, then $V_Q > 5 \times 10^{-16} \text{ cm}^3$. If polarization switching is involved, larger domain sizes would probably be required, since β_P is probably smaller than β_Q . If fluctuations in and out of the TSDW phase are involved, slightly larger resistivity changes (about 4×10^{-2}) could be involved,²³ allowing for somewhat smaller domain sizes.

Since a particular switcher is characterized by a step size, and an up rate and a down rate, all of which change continuously as a function of temperature, it is possible to study the properties of a particular switcher over a range of temperatures. The latent heat for the formation of the TSDW phase is known,¹ so the temperature range over which phase fluctuations occur is calculable for a given domain volume. The result is that a single domain should show appreciable phase fluctuations over a range of some 10 mK, which is much less than the 1-K range over which individual fluctuators were tracked. Therefore the steps are produced by rotations of the TSDW order, not by fluctuations out of the TSDW phase.

Since η rotations are believed to be responsible for the 200-K noise,¹⁰ one might suspect that they are also responsible for the switching events near T_n . However, several arguments suggest that this may not be the case. First, the temperature signature of the bulk internal friction in Cr changes just below T_N ,^{1,14} and suggests that two mechanisms with different temperature dependence are present. Second, while elevated noise levels persist in the MBE Cr samples well below T_n , the discrete steps are much less prominent. Thus a small number of large fluctuators dominates the noise near T_n . At lower temperatures a large number of small fluctuators dominates the

noise yielding the same overall noise magnitude.

The switcher shown in Fig. 4 was studied in detail as a function of temperature over a range of 0.7 K at 313 K. The characteristic switching rate (the sum of the up and down switching rates) changed far more rapidly than could be explained by an Arrhenius relation. The resistance step size also increased with cooling, in approximate quantitative agreement with the known temperature dependence of β_Q for Q orientations in the bulk.^{1,23} As shown in Fig. 5, the dependence of the characteristic switching rate on the step size was approximately exponential, suggesting that the temperature dependence of the rate is primarily due to the temperature dependence of the potential barrier to rotation, which depends on the extent of the TSDW condensation in roughly the same way as does β_Q . An extrapolation of the rate to zero step size, and hence zero barrier height, gives an attempt rate of about 10^6 Hz, which does not seem wildly unphysical for the coherent rotation of a domain.

The Q domains are not, for the most part, simple two-state systems. Particular fluctuators can be trapped in one of their states for long times compared with the typical time for leaving that state. Such effects in general indicate fluctuators with at least three states and often indicate interactions between different fluctuating sites.^{6,8} The variability in the noisiness of a given switcher can sometimes be seen in a time series of octave power measurements. The resistance as a function of time in a 33-Hz bandwidth at 315.9 K is dominated by a single apparently two-state switcher as shown in Fig. 6(a). The resultant power spectrum, resembling a Lorentzian but slightly broader, is shown in Fig. 6(b). Figure 6(c) shows noise power as a function of time for the two neighboring frequency octaves indicated in (b). Highly correlated slow modulations can be seen clearly above the usual (uncorrelated) Gaussian statistical variations. In this case, switching, reminiscent of the resistance versus time, is observed. These slow modulations can be characterized

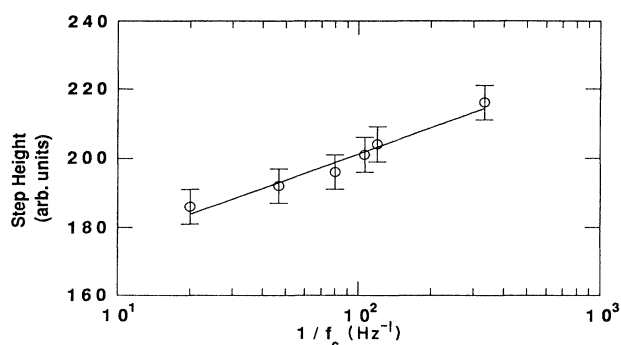


FIG. 5. Near the transition, the step height of a switcher in sample A changes exponentially with inverse switching frequency (which is the frequency of the peak in the power spectrum) as the sample is cooled from 316.8 to 316.1 K. The temperature dependence of the step height and that of the fractional change in the diagonal component of the resistivity tensor due to the orientation of the Q vector (β_Q) are roughly the same.

by a “second spectrum” obtained from the Fourier transform of the octave power time series.⁶ The resultant second spectrum is only slightly broader than a Lorentzian, as shown in Fig. 6(d). The clear interpretation is that the activity of this site is modulated by an interaction with some other single degree of freedom. Other second spectra are more complicated, apparently indicating domain interactions with a larger number of degrees of freedom. A strain mediated domain-domain interaction which has been proposed by Pink and Steinitz⁷ to explain

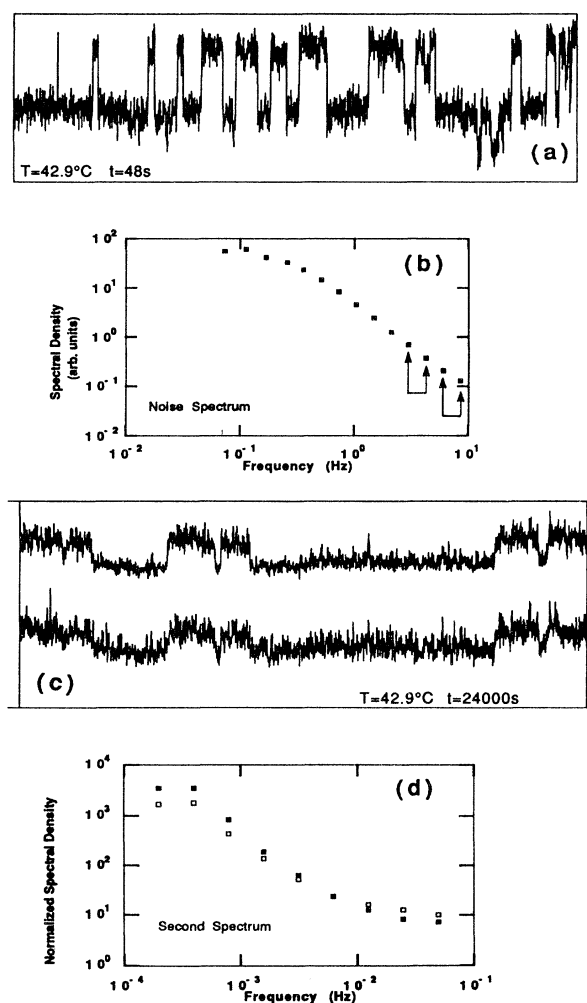


FIG. 6. Detailed statistical analysis of a particular switcher in sample A reveals complicated series dynamics. (a) Resistance vs time at 315.9 K is shown. (b) The corresponding power spectrum is shown. The noise power in the indicated octaves was repeatedly sampled to produce the octave-power time series in (c) which have been normalized to the Gaussian expectation and offset for clarity. Averaging the squared Fourier transform of such power vs time records produce the “second spectra” shown in (d). The filled squares represent the lower octave and the open squares represent the higher octave.

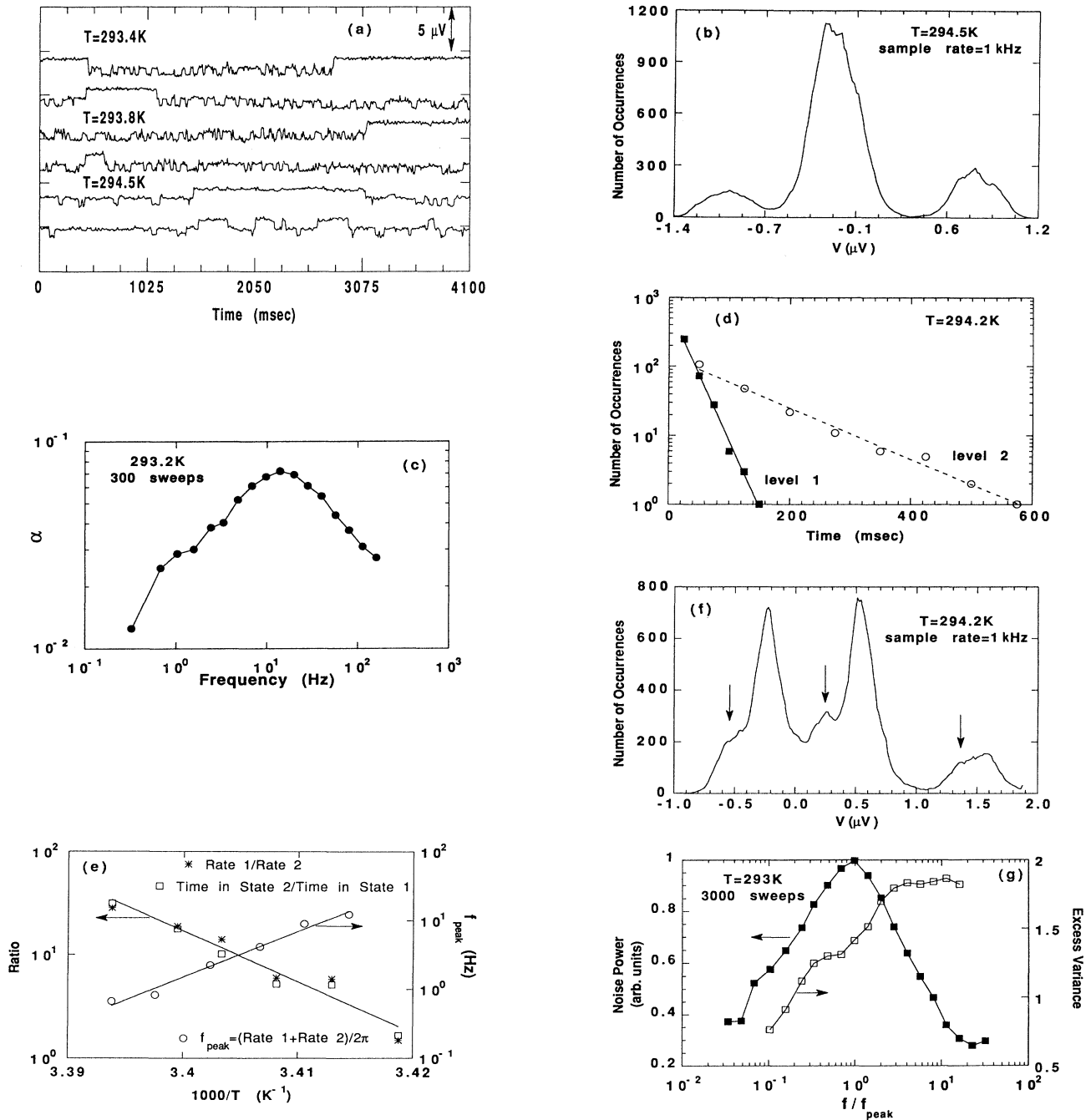


FIG. 7. (a) Voltage traces vs time are shown for a three-state switcher in a single-crystal sample with volume $1 \times 10^{-13} \text{ cm}^3$ (sample B) with applied voltage of 40 mV. Data shown were taken at three temperatures near 293 K. This switcher was tracked as temperature was cycled over a range of about 3 K. (b) A histogram of the voltages shows that the three states can be distinguished and the time spent in each state determined. Typically, over 10^5 voltage points were measured at 2 kHz. (c) The power spectrum (from a 300 FFT average) is dominated by a Lorentzian peak. The peak frequency and height help to characterize the domain. (d) Histogram of the time spent before switching is shown for two of the states. The characteristic relaxation rates are determined by fitting to an exponential. (e) The ratio of the relaxation rates and the essentially equivalent ratio of the time spent in the two principal levels are shown vs inverse temperature, along with the characteristic peak frequency. The frequency of the peak in noise power increased as the sample was cooled. (f) A histogram of voltages at 294.2 K shows each of the three principal levels split into two sublevels, indicating a second independent two state system. (g) Both the noise power per octave and the variance in the octave noise power in excess of the Gaussian expectation are shown vs normalized frequency. The frequency is normalized to the frequency of the peak in the noise power, so that many spectra near the temperature shown could be combined in order to improve the statistics. The excess variance peaks a few octaves above the peak in the noise power, characteristic of discrete switching behavior.

anomalous ultrasonic attenuation peaks could account for these results although an interaction between domains and mobile defects cannot be ruled out.

This interesting domain behavior is also clearly visible in the voltage traces shown in Fig. 7(a) taken near 290 K. The random telegraph signal shows switching between two lower resistance levels and an occasional switch to a higher third resistance level in which the other switching is not present. No reasonable defect could produce such large resistance changes. The three distinct levels are easily identifiable in the histogram of the resistance values shown in Fig. 7(b). The dramatic effect on the power spectrum is shown in Fig. 7(c) where a peak dominates the data. Here, there are two domains which are highly interactive or a single domain having three stable configurations each with a unique resistance value with respect to the current direction, and each with a unique characteristic switching rate. It is also possible that the two lower levels indicate an η domain orientation which has equal resistance when its encompassing Q domain is in one of its two orientations. In any case the switching shows complicated series kinetics.

The characteristic switching rate is determined by fitting a histogram of the time spent in each level before switching to another level to an exponential as shown in Fig. 7(d). This domain behavior was tracked as temperature was cycled over about 3 K. There was significant hysteresis in the frequency of the peak in the power spectrum, as well as the peak magnitude, though the step sizes did not change with temperature. Figure 7(e) shows the ratio of the switching rates for level 1, to level 2, which is equivalent to the ratio of the time spent in each state as determined from the resistance histogram, versus inverse temperature. These indicate how the energy difference (ΔE) between the two states in a two-level-system model changes with temperature. The Arrhenius fit to the data results in $\Delta E \approx 560k_B T$, an entirely unreasonable result. The sum of the switching rates, which reflects the corner frequency in the power spectrum, and indicates the barrier height between the two states, was weakly temperature dependent on cooling, and actually decreased with increasing temperature on warming. Figure 7(e) shows the peak frequency as a function of inverse temperature on cooling.

Clearly neither the thermodynamics nor the kinetics of a two-level-system model with a constant energy difference and a constant barrier height describe this behavior. The state energies and the barrier height must themselves be temperature dependent, due to strain effects and/or interactions between fluctuating regions.

The paucity of switching events into the upper-third voltage level in Fig. 7(a) hindered detailed analysis of its switching characteristics. However it is clear, from the long lifetimes, and the infrequent switching, that the energy barrier between this state and the other two is large and increases with decreasing temperature. This behavior is also likely to be the result of temperature-dependent stresses in the crystal influencing the energy barrier and the energy difference between the domain orientations.

At the highest temperatures in this cycling series, near 295 K, a second domain can be seen in the resistance his-

togram shown in Fig. 7(f). The switching rate of this domain does not depend on the state of the other domain system, as shown by the constant ratio of its height to that of the neighboring peaks. In other words, this switcher shows parallel kinetics. This domain is possibly removed spatially from the others and is thus uninfluenced by them.

When the noise powers, integrated over a frequency octave, of the individual FFT's are compared, fluctuations about the average octave noise power are found. For any Gaussian process, all such higher-order moments can be expressed in terms of the two-point correlation function, or equivalently the noise power spectrum. Specifically, in our case, this means the fluctuations of the power in one frequency bin are statistically nearly independent of those in any other frequency bin. (There is some correlation due to the discrete Fourier transform.) Further, the variance in the noise power of one frequency bin is equal to the square of the mean value of the noise power in that bin.²⁶ Finally, the variances in the octave noise power is just the sum of the variance in the noise powers of each bin in the octave, which is the sum of the squared noise power in each bin in the octave. (Again, there are small corrections due to the finite bin size.) The variance in our octave noise power data is computed and normalized to this Gaussian expectation. The excess variance, which is a measure of the non-Gaussian charac-

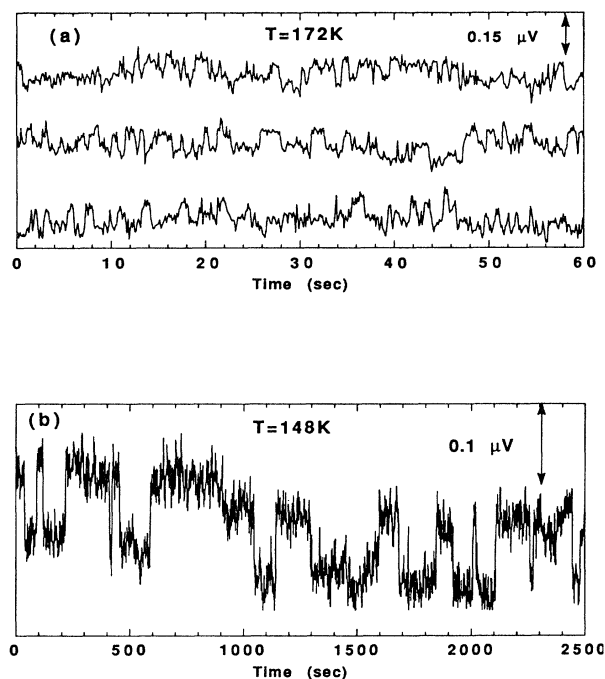


FIG. 8. Voltage vs time records for sample D at $T=172$ K, with an applied voltage of 10-mV rms are shown in (a). At this temperature, the sample resistance is 12Ω . Switching events are visible, but obscured by the background of other events. In (b) an extremely slow but clearly visible switcher is active at $T=148$ K. The switchers represent fractional resistance changes of 1.1×10^{-5} and 1.3×10^{-5} , respectively.

ter in the signal, is plotted as a function of normalized frequency in Fig. 7(g). Simulations and previous experiments have verified that the excess variance in a random telegraph signal peaks a few octaves in frequency above the noise power peak, at which frequency the white and $1/f$ Gaussian background noises begin to dominate the non-Gaussian fluctuations.^{24–26} The normalized noise power is also plotted in Fig. 7(g), where the behavior is verified for this set of switching data at 293 K. This same pattern will help to identify switching in lower temperature data where the distinct switching levels are difficult to distinguish in the resistance versus time traces.

Typical low-temperature resistance (voltage) traces are shown in Fig. 8(a). The voltage steps are not as large as in the high-temperature data. Though switching events can be found, they are more difficult to characterize because of the background of other events. The voltage traces shown in Fig. 8(a) were taken at $T = 172$ K on the smallest of the four mesoscopic samples. The fractional change in resistance for the steps visible in these sweeps is 1.1×10^{-5} , leading to $\beta V = 6 \times 10^{-19}$ cm³. This value is an order of magnitude smaller than that of the Q domains observed near T_n , and indicates these switching events are the result of some other process. The extreme-

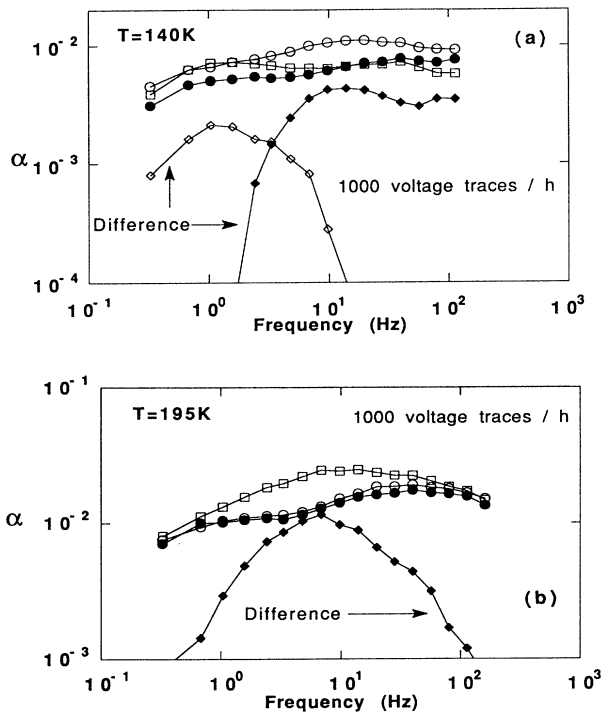


FIG. 9. The noise power vs frequency makes discrete changes over the course of the 8-h observation time. (a) $\alpha(f)$ is shown for successive averages in sample B held at 140 K. The open circles represent the first data (5 h), followed by the solid circles (2 h), and then the open squares (1 h). The open and solid diamonds show the differences between the first data and the two subsequent sets. (b) Data similar to (a) is shown. The open circles were first (2 h), followed by the solid circles (2 h), and then by the open diamonds (1 h). The solid diamonds show the differences between the first set and the remainder.

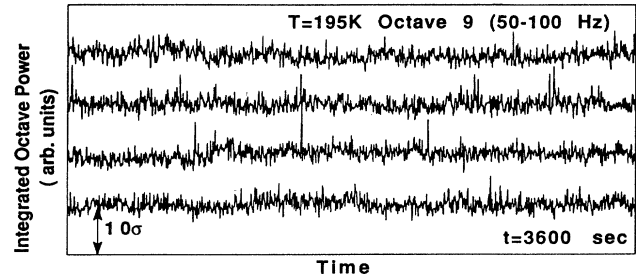


FIG. 10. Four records of octave noise power in the ninth octave (50–100 Hz) offset for clarity and normalized to the expected Gaussian standard deviation, are shown as a function of time for sample B. The arrow shows ten times the expected Gaussian standard deviation. Probable isolated switches between discrete power levels can be found, which appear similar to the switches in the resistance vs time.

ly slow but well isolated switcher shown in Fig. 8(b) was observed at $T = 148$ K, and represents a fractional resistance change of 1.3×10^{-5} . These data clearly show the discrete nature of the resistance fluctuations at these temperatures.

The low-temperature power spectra show small peaks as shown in Figs. 9(a) and 9(b). They appear and disappear as the active switching domain is modulated by another slower domain, which is not visible in our frequency window. The contribution of the switching domain to the power spectrum can be isolated by subtracting the average of the spectra in the upper state from the average of those in the lower state. The result is seen to be approximately Lorentzian, as expected for a two-state switcher.

The slower domains, which modulate those in the power spectrum, are often visible in the plot of octave power versus time as shown in Fig. 10. Distinct switching levels can be isolated, and at lower temperatures they resemble the R -vs- t traces in that the switching is not as persistent nor are the levels as separated as at higher temperatures. The resulting second spectrum, however, is approximately Lorentzian, as is the case at higher temperatures near the transition where the switching is more evident, which indicates slow two-state processes are modulating the noise power at low temperatures as well.

Further evidence that different mechanisms are responsible for the noise in the two-temperature regions is seen in the plot of Lorentzian peak sizes in the power spectrum, along with the average excess variance, as a function of temperature in Fig. 11. The two temperature regions are distinguishable where the typical values differ by about an order of magnitude. In this same region, the noise power in the large samples has reached a plateau and remains essentially constant. The rise in excess variance above 290 K reflects the reduced number of fluctuators in the frequency band. This, combined with the constant noise power, indicates the switchers are much larger in this temperature region, which is supported by the typical size of the Lorentzians observed. At lower temperatures, there are a larger number of small fluctuators.

When the average excess variance in the octave power

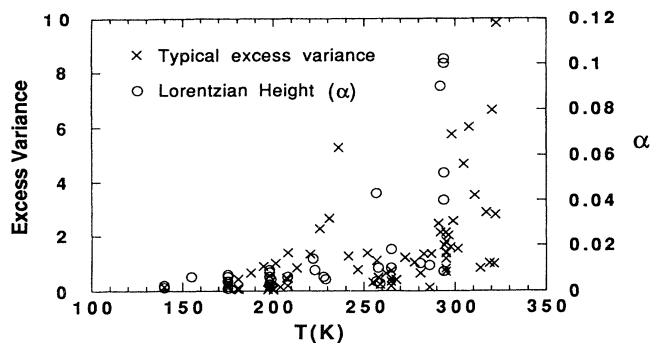


FIG. 11. The excess variance (see text) and the heights of the Lorentzian-like peaks seen in the power spectrum are shown as a function of temperature. At lower temperatures, the excess variance and the Lorentzian heights are small, indicating a larger number of smaller fluctuators are active than at high temperature.

is again plotted as a function of normalized frequency, together with the average power spectrum, for data over small temperature ranges near $T=175$, 195, and 265 K in Fig. 12, it is seen to peak a few octaves above the noise

power peak. These observations convince us that the η domains responsible for the noise at low temperature switch between two or a few stable orientations, which are determined by the Q domain direction, the stresses in the crystal, and the principal crystal axes. This settles a long standing question as to whether the η dynamics really consist of switching events, or some kind of continuous redistribution.

In some systems in which a large number of fluctuators interact, the particular set of active fluctuators changes randomly upon thermal cycling, apparently as the system falls into one of many local minima of a collective Hamiltonian.²⁵ In the MBE Cr films near T_n , this behavior was not observed. For example, cycling from a base temperature of $T=314$ K above the transition to $T=325$ K and back reproduced the same switcher as shown in Fig. 13, which was verified by comparing the relaxation rates, and the step size before and after cycling. Further, the same broad temperature regions near the base temperature, where no switching and very low noise levels were present, were also reproduced. Additional comparisons (in another sample) of striking features in the power spectral density at a range of base temperatures between room temperature and the transition, before and after cycling above T_n , reinforces this conclusion. Thus the set of al-

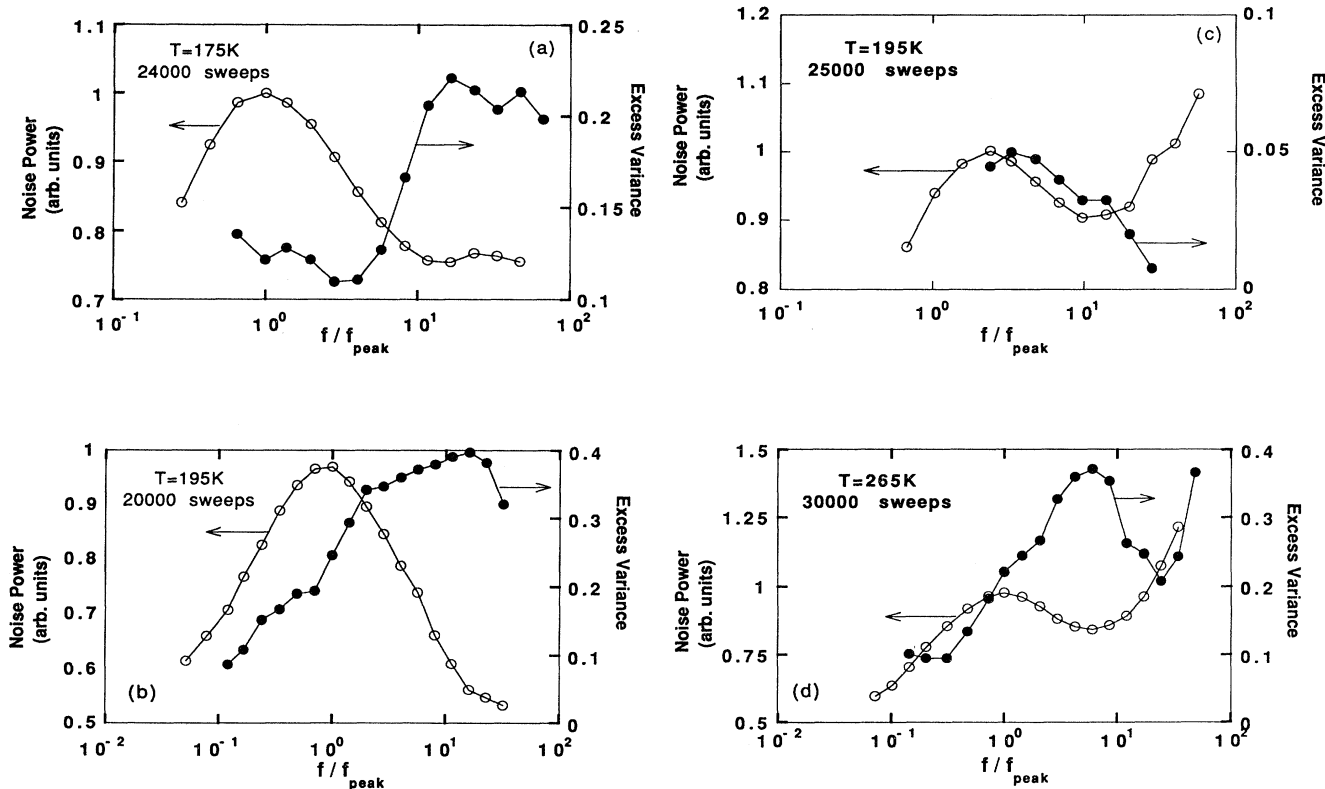


FIG. 12. Both the noise power per octave and the variance in the octave noise power in excess of the Gaussian expectation are shown vs normalized frequency in the low-temperature regime for sample B. As in Fig. 7(g), the frequency is normalized to the frequency of the peak in the noise power, so that many spectra near the temperature shown could be combined, in order to improve the statistics. The excess variance peaks a few octaves above the noise power peak at all four temperatures shown, indicating that discrete switching is present at lower temperatures as well as at higher temperatures.

lowed Q states apparently is almost uniquely set by local strains at least near T_n .

The effect of temperature cycling at lower temperatures was difficult to determine. The domain structure responsible for the features in the power spectrum seemed to be stable for times on the order of hours, at which time the features change, often disappearing, due to the influence of some other unseen switcher, only to reappear hours later. Cycling the temperature by +20 K at 175 K did not fully randomize the shape of the power spectrum as seen in Fig. 14(a), although significant changes on cycling are visible. Similar data at 199 K seem to show significant scrambling of the domains responsible for the features in the power spectrum, and data at 265 K showed an even more dramatic effect in Fig. 14(b). When the power spectra from the cycling data at 265 K were averaged together, a nearly featureless $1/f$ spectrum results with a magnitude very close to that of the larger crystal sample. This was also the case in general when many spectra from temperatures differing by a few percent were averaged, indicating that these processes randomize the statistics, and allow the system to occupy a large set of configurations. When the cycling data from 199 K were averaged together, some features were still present, and significant features were still apparent in the 175 K data, indicating that the cycling returns the system at these temperatures to the same limited set of configurations. It is difficult to draw conclusions from these few observations; however, it appears that the polarization configurations can settle into one of a number of possible states when the sample is cooled. Apparently the cycling temperature increment required to effect a rearrangement increases with decreasing base temperature, and thus 20 K is enough to affect a significant change at 200 K, but not at 175 K.

Calculation of the size of the apparent Q domains is straightforward, since the relevant resistive anisotropy β_Q is already known,²³ and since the actual resistive step size is directly observable. The principal complications are that β_Q is temperature-dependent near T_n , that the step size depends on the relative orientation of the field lines and the crystal axes, and that the current density is not known to high precision. Assuming that the largest

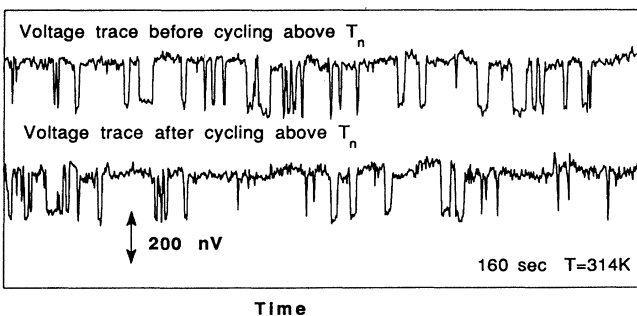


FIG. 13. In sample A, similar switching at 314 K was reproduced after cycling to 325 K, $T_n=323$ K. The upper trace shows the voltage sampling before cycling, and the bottom trace shows it afterward.

steps are from domains with one Q state parallel to the current, we find $V_Q=3\times 10^{-17}$ cm³, with only about a factor of 2 uncertainty.

The calculation of a maximum estimate for the polarization domain size, used but not included in our previous work,¹⁰ is presented here. This calculation is based on S measurements and noise magnitude measurements made on polycrystalline films with different tensile strains. We argue first that a lower bound for the polarization resistivity anisotropy, β_P , can be calculated independently of the typical domain size V_P , using only the typical noise magnitude. By integrating over all orientations of the current with respect to the crystal axes, one finds

$$\alpha = \frac{4}{15} \beta_P^2 n_A n_P V_P^2,$$

where n_A is the concentration of atoms, and n_P is the concentration of active polarization domains per log unit of characteristic frequency and per $k_B T$ of energy asymmetry. We then use the fact that the range of the energy asymmetries is set by random strain interactions, including miscellaneous defect effects. Systematic strains (ϵ_S) produced by the differential thermal expansion of the film

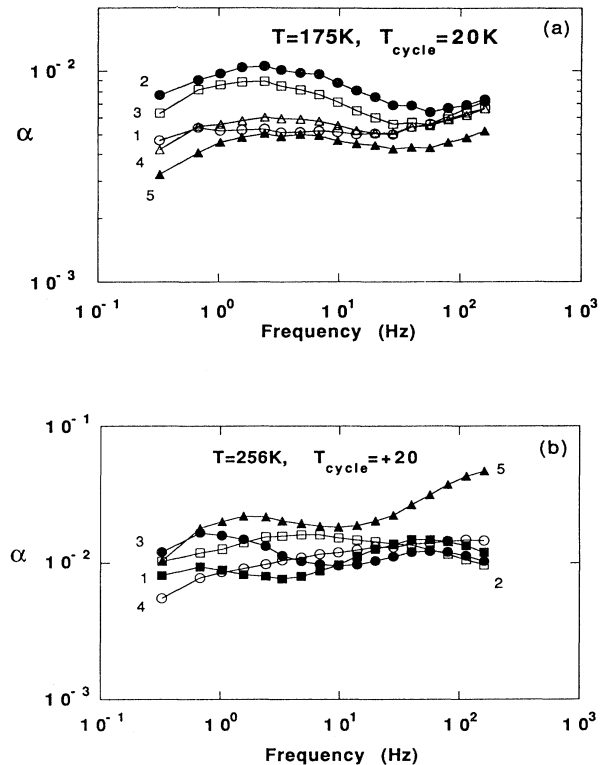


FIG. 14. (a) Spectra from sample B taken at 175 K after cycling the temperature to 195 K are shown. The numbers indicate the time order of the spectra observed after each of five cycles. The cycling obviously only partly randomized the features. (b) Similar data are shown with a measuring temperature of 256 K. Cycling the temperature to 276 K nearly completely randomized the power spectrum.

and the substrate should produce systematic energy differences between the polarization states in the plane and out of the plane, with an energy difference equal to $V_p a_p \epsilon_S$, where a_p is a known magnetoelastic constant.²⁷ If the net energy difference ΔE between orientations for a given domain is dominated by the systematic strain, the noise would be reduced in high-strain samples, for which the domains would tend to get thermodynamically stuck in their low-energy configurations. No such effect was found. This indicates the random strain term is large enough to give a number of domains with $\Delta E < k_B T$ nearly independent of the systematic strain. Further, while the value of S , which is sensitive to the orientation of the Q domain, does change as a function of systematic strain, the range of measured values shows that even on the larger volume scale of the Q domains the random strain is a significant fraction of the largest systematic strains. We also estimate that, given the $1/f$ spectra observed over a range of temperature, it is unlikely that the range of characteristic times for the domain relaxation spans less than 25 factors of e . Then

$$n_p < \frac{1}{25} k_B T / (\epsilon_S a_p V_p^2) .$$

This leaves us with

$$\alpha < 1.1 \times 10^{-2} \beta_p^2 n_A k_B T / (\epsilon_S a_p) ,$$

requiring $\beta_p > 2 \times 10^{-3}$, when numerical values are substituted for the variables.

The previous estimates of V_p were the result of a determination of $(\beta_p V_p)$ from an estimate of n_p based on the deviations of the spectral shape from its average value in small samples.¹⁰ Since the small samples studied here were small enough to allow determination of the magnitude of individual Lorentzian contributions to the noise, $(\beta_p V_p)$ could be measured directly. The total mean square fractional resistance fluctuations from a single domain in a two-level system model will be

$$\langle (\delta R / R)^2 \rangle = \beta_p^2 (V_p / V_S)^2 (\cos^2 \theta - \sin^2 \theta \sin^2 \phi)^2 \\ \times \text{sech}^2(\Delta E / k_B T) / 4 ,$$

where θ and ϕ describe the relation of the current flow to the two axes between which η switches. For the largest resistive steps, we presume that both the trigonometric and thermodynamic factors are close to unity. We measure $(\delta R / R) = 7 \times 10^{-6}$, and in turn $\beta_p V_p > 7 \times 10^{-19} \text{ cm}^3$ but not much larger, in very good agreement with the previous estimate of $12 \times 10^{-19} \text{ cm}^3$ for the typical $\beta_p V_p$. The maximum estimate for a typical V_p is then $6 \times 10^{-16} \text{ cm}^3$ which is in agreement with the range determined by Steinitz *et al.*²⁸ from the magnetic susceptibility anisotropy.

It is highly unlikely that the polarization would remain coherent over a volume larger than a Q domain, and the Q domains which were switching near T_n had $V_Q = 3 \times 10^{-17} \text{ cm}^3$. Whether this means that the actual

V_p is $< 3 \times 10^{-17} \text{ cm}^3$, which would require $\beta_p > 10^{-2}$, or that the Q domains whose switching is observed are atypically small is difficult to determine at this time.

DISCUSSION

The principal conclusion from these experiments is that the slow dynamics of the SDW polarization in Cr are in fact due to discrete switching events of domainlike objects with a range of relaxation rates, clearing up this long standing question. This conclusion is based on the combination of discrete Lorentzian-like components in the spectrum together with non-Gaussian statistics which are very similar to those obtained for two-state systems, along with marginally detectable steps in the resistance versus time, and clear switching behavior in the noise power versus time.

Since the noise along the [111] direction is substantial in the polarization noise regime, the polarization in these samples must not be constrained to lie exactly along cubic axes. Thus the polarization domains may actually be two-state systems in a disordered polarization configuration, rather than regions with sharp boundaries and an internally uniform polarization. The stability of the polarization, given that it is not tightly constrained to the cubic axes, certainly requires that defects and strains play an essential role. This overall picture is quite reasonable for a Hamiltonian consisting of a uniformly anisotropic term plus local strain fields.^{3,5}

At least in the MBE samples, many of both the Q and η domains do not form isolated two-state-systems. Three-state systems and more complicated coupled behaviors are common. A plausible interpretation is that the domains interact strongly with their nearest neighbors, possibly through strain,⁷ so that both the energies of the different SDW orientations and the barrier heights between them depend on the state of the neighbors. Similar strain-mediated interactions have previously been proposed to account for anomalies in field-dependent ultrasound absorption.⁷

The difference between the Q domains, which find a nearly unique stable configuration, and the η domains, which reach different metastable configurations on different cool-downs, is not fully understood. A reasonable interpretation would be that the Q domains have fewer states to pick from because they are more tightly constrained to lie along crystal axes, as shown by the noise anisotropy.

We do not have an explanation for why the SDW polarization noise spectral density is so nearly independent of sample preparation. The size of the observed domains is about the same in defect-laden evaporated films as in single-crystal MBE films. This approximate equality may suggest that the typical rotation rate is mainly a function of domain size, not defect concentration. The inferred polarization domain size is quite close to those that have been estimated for the domains in bulk single crystals. The near equality of the spectral densities, however, indicates that the fraction of domains within a given size

range and range of energy asymmetry is independent of the defect concentration, another unanticipated result.

In short, the reason why the TSDW polarization breaks up into domains of a particular size remains unknown. The actual existence of such switching regions, together with the importance of the interactions between them, now at least rests on a firm empirical base.

ACKNOWLEDGMENTS

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