Charge-density-wave conduction noise in blue bronze: Bulk-generated Gaussian noise

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The conduction noise associated with the sliding charge-density wave in $K_{0.3}MoO_3$ is investigated by measuring the total noise power and its fluctuations. The statistical properties of the fluctuations show the conduction noise to be Gaussian, which indicates an underlying randomness in the sliding process. The size of the noise increases above threshold and then saturates for large bias. This saturation occurs when the fundamental frequency of the conduction noise is comparable to the dielectric relaxation frequency. The saturated value of the noise increases with sample length, indicating that the noise is produced in the bulk. The linear variation found suggests that the conducting domains extend the full length of the crystal.

I. INTRODUCTION

Nonlinear transport in materials undergoing a Peierls transition[1] has excited much interest and controversy since its discovery in 1976.[2] In particular, the origin of the radio-frequency conduction noise that accompanies the sliding charge-density wave (CDW) remains unclear. The noise has been attributed to the CDW's interaction with a pinning potential,[3-8] or, alternatively, to phase slip at an interface between pinned and sliding-CDW regions.[9-11] Efforts to discriminate between these mechanisms of noise generation by measuring the dependence of the conduction noise on specimen size have reached contradictory conclusions.[10,12-16]

While some investigators have referred to the radio-frequency signal as "coherent current oscillations," several others have remarked[10,13,17-19] on large temporal fluctuations in the amplitude of the signal. Recently these fluctuations themselves have been the topic of investigation. We showed[20] that the conduction noise in $NbSe_3$ has Gaussian statistics, and Bhattacharya and coworkers[19] found that the fluctuations in $NbSe_3$ persist even in the mode-locked state. Clearly the fluctuations are a key feature of CDW conduction, but their origin remains unexplained.

Most noise experiments have been performed in $NbSe_3$, which is unusual among sliding CDW compounds in that it retains its metallic character within the CDW state. Its noise spectrum consists of a narrow peak (with overtones), leading to the term "narrow-band noise." Noise studies are somewhat scarcer in the compounds which undergo a complete metal-semiconductor transition, for which conduction-noise spectra tend to be quite broad.[15,21,22]

This work reports investigations of the conduction noise in one of the semiconducting CDW compounds, the blue bronze $K_{0.3}MoO_3$. In Sec. II the experimental details are discussed. Section III presents the statistics of the CDW noise. Section IV explores the bias dependence of the size of the noise and its temperature dependence. In Sec. V the dimension dependence of the size of the noise is reported. The results are discussed in Sec. VI.

II. SAMPLES AND METHODS

Blue bronze exhibits a depinnable CDW below the Peierls transition temperature $T_P = 180$ K. It is a convenient material for CDW studies because it grows in relatively large crystals, which facilitates handling. In addition, the method of making good electrical contacts to the material is known.[23] The crystals of blue bronze used for these experiments were grown by electrolytic reduction and were x rayed to determine the orientation of the conducting (**b**) axis. They were ground to rectangular geometry, typically $4\times1\times\frac{1}{2}$ mm^3, with **b** parallel to the long dimension. After cleaning with a dilute NH_4OH solution, copper contacts were electroplated on the ends using a 15:1 $CuSO_4/H_2SO_4$ solution. The sample was suspended from two 0.025-mm-diameter gold wires (which provided electrical contact as well as pliant mechanical support during cooling) using silver paint (DuPont compound No. 4929). After the painted contacts dried for at least 12 h, the sample was cooled slowly and immersed in liquid nitrogen to ensure a constant temperature of 77 K. Conduction-noise spectra were visually inspected on Hewlett-Packard HP-8557A spectrum analyzer.

For the statistical portion of the experiment the sample was current biased above threshold, and the amplified conduction noise (at frequencies between 50 and 500 kHz) was frequency mixed up to 30 MHz. A 30-kHz band at the fundamental frequency was extracted by using a band-pass crystal filter, the power in this frequency window was amplified and rf-detected (RHG EST3010 amplifier and square-law detector), and the time series of the detected power was digitized. From this time record the distribution of power and the spectrum of the fluctuations in the power were calculated.

For the portion of the experiment characterizing the bias and dimensional dependence of the conduction noise, specimens were current biased, and the amplified noise voltage was measured with a true-rms voltmeter (Fluke 8920A—bandwidth 10 Hz–20 MHz). This instrument accurately analyzes nonsinusoidal signals by measuring heating in a thermal-rms converter circuit, which is equivalent to measuring the total integrated power. The CDW conduction-noise power was calculated from the total noise power by subtracting the noise power for zero bias from the total. (This correction was small; at $E=2E_T$ the correction was typically less than 3%.) To account for variation in sample impedance (which changes with bias as well as among samples), the noise, referenced to the 50-Ω input impedance of the amplifier, was converted into the open-circuit value. For the length dependence, specimens were shortened by grinding the samples from both ends and electroplating fresh contacts.

Samples reported upon here had two-probe low-field resistivities at 77 K of 0.12–0.16 Ω cm and sharply defined threshold fields E_T with values ranging from 59 to 180 mV/cm. Although the features in the power spectra of the conduction noise are broad (unlike $NbSe_3$), there is still a well-defined peak frequency f_0, which is found to increase proportionally with CDW current:

$$f_0=\alpha J_{CDW}=\alpha(I_{tot}-V_s/R_0)/A \ . \quad (1)$$

Here I_{tot} is the total current, V_s is the sample voltage, R_0 is its resistance below threshold, and A is its area. f_0 is generally interpreted to be the ratio of the velocity of the CDW to its wavelength, λ: $f_0=J_{CDW}/(ne\lambda)$. Thus, from the number of condensed electrons and the observed wavelength of the distortion, α can be calculated[24] to be 13 kHz/(A/cm^2). Our experimental values ranged from 21 to 44 kHz/(A/cm^2). Such upward departures from 13 kHz/(A/cm^2) are frequently reported in the literature and are attributed to a reduction of the sample's effective cross section due to incomplete conduction.

Many samples were rejected for failing to have sharply defined thresholds or reasonable resistivities or both, or for having values of α larger than approximately 50 kHz/(A/cm^2). We speculate that the rejected specimens were damaged by handling, or had bad contacts caused by either inadequate cleaning before electroplating or contaminated electroplating solution. While the characteristics of acceptable samples were reproducible, those of rejected specimens were not.

III. STATISTICAL PROPERTIES OF CONDUCTION NOISE

Previous experiments[20] showed that the CDW conduction noise in $NbSe_3$ is Gaussian and thus shows a fundamental randomness. $NbSe_3$ is an unusual CDW material in that its Fermi surface is not completely gapped. Blue bronze, however, undergoes a complete metal-semiconductor transition and thus presents an opportunity to search for potential nonrandomness in a more typical CDW material. To test for non-Gaussian effects, the fluctuations in the conduction-noise power spectrum of blue bronze were studied.

Figure 1(a) shows the distribution of power, $p(P)$, in the band-passed CDW conduction noise, and the theoretical fit to a random process of the same average power. To illustrate what happens to an indubitably random signal, the distribution of Johnson noise from a resistor was measured in the same way. The Johnson noise was amplified so that the average noise power was the same as that of the CDW noise. Figure 1(b) shows the distribution of power in that band-passed Johnson noise. In each case, the solid line is the theoretical fit[25]

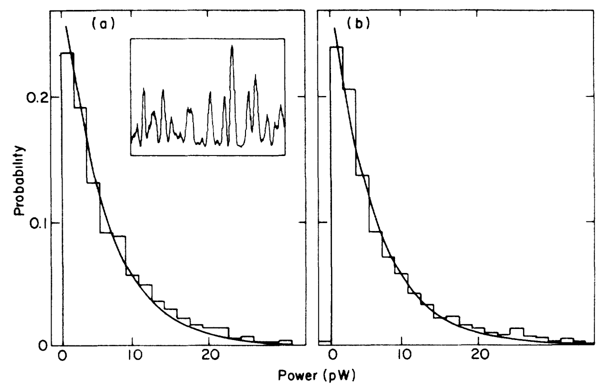

FIG. 1. Histograms of power in (a) band-passed CDW conduction noise and (b) band-passed Johnson noise. The solid line is the theoretical probability distribution for narrow-band Gaussian noise. The inset shows a short part of a time record of the fluctuating power used to calculate distribution in (a) and spectrum in Fig. 2(a).

$$p(P)=(a\sigma^2)^{-1}e^{-P/a\sigma^2}, \quad P\geq 0 \quad (2)$$

obtained by considering the action of a square-law device $y=ax^2$ and low-pass filter on a Gaussian input voltage

$$p(V)=(2\pi\sigma^2)^{-1/2}e^{-V^2/2\sigma^2} \ . \quad (3)$$

The shapes of the distributions in Fig. 1 are the same—each is the distribution of the detected, filtered power appropriate to a Gaussian process. The fluctuations extend down to zero power, reflecting the fact that the voltage fluctuations are as large as the mean voltage.

To search further for possible non-Gaussian properties, the spectrum of fluctuations in the CDW noise power was calculated. A short example of the fluctuating power is shown in the inset to Fig. 1. The band-passed spectral power as a function of time was Fourier analyzed to yield the "second spectrum" of the noise.[26] In general, such a second spectrum of a band-passed Gaussian signal is flat up to the bandwidth of the filter. A representative second spectrum for CDW conduction noise is shown in Fig. 2, along with the second spectrum of Johnson noise of the same average power. Both are flat up to a frequency of 30 kHz, which is just the bandwidth of the crystal filter. Second spectra were studied at several different sampling rates to investigate a large frequency range, and at fields both less than and greater than those at which the conduction noise saturates (see Sec. IV). In all cases studied (including results from several different specimens), the second spectrum of CDW conduction noise was identical to that of band-passed Johnson noise: it was flat over 4 orders of magnitude in frequency (from 1 Hz to 30 kHz), and its level was within 1 dB of the Johnson-noise second spectrum.

Thus, judging from the statistical perspectives of the distribution of power and the second spectrum, the CDW conduction noise in blue bronze is Gaussian.

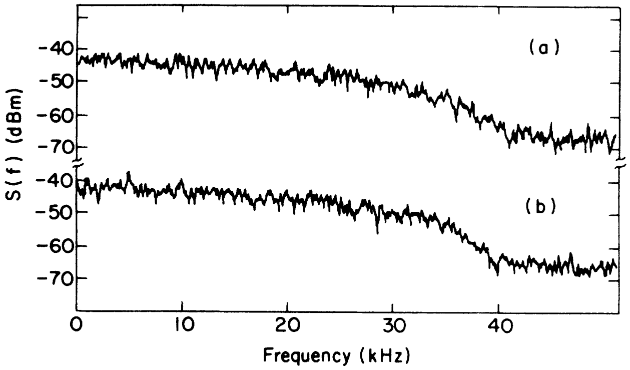

FIG. 2. Representative second spectra of (a) band-passed CDW conduction noise and (b) band-passed Johnson noise. The falloff for $f \gtrsim 30$ kHz reflects the bandwidth of the filter.

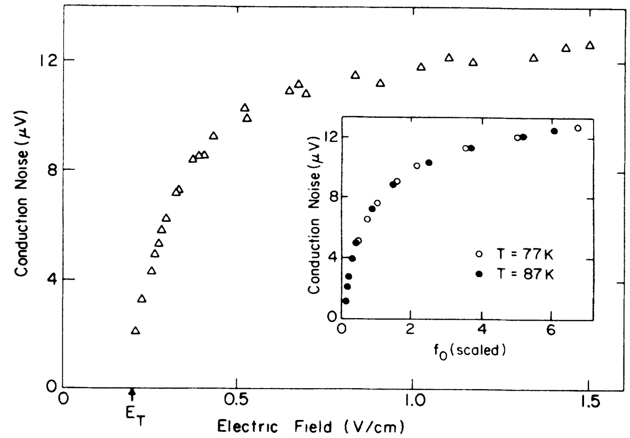

FIG. 3. Total conduction noise voltage vs electric field. Noise was measured with true rms broadband voltmeter. Inset shows conduction noise vs scaled frequency of the fundamental for two temperatures. Frequencies at 77 K are actually 3.2 times smaller than those at 87 K (e.g., at a scaled f_0 of 4, $f_0^{77\,\mathrm{K}} \approx 90$ kHz; $f_0^{87\,\mathrm{K}} \approx 290$ kHz).

IV. VARIATION OF CONDUCTION NOISE WITH BIAS

Many studies of conduction noise in CDW materials report changes in the power spectra as the sample bias is varied.[12,15,27–31] The amplitude of the fundamental, the relative harmonic content, and the widths of the spectral peaks all depend on bias. In order to compare fairly the size of the conduction noise under different conditions (different biases, temperatures, or specimen lengths, for example), an unambiguous measure of the size of the noise must be employed. The statistical characterization of the CDW noise provides just such a measure: a consequence of the Gaussian statistics of the conduction noise is that its distribution may be completely characterized by its second moment, the rms voltage. Using this property avoids confusion between changes in spectral shape and variations in total noise amplitude, facilitating comparison between different experimental conditions.

Figure 3 shows the total CDW conduction-noise rms voltage as a function of electric field, measured with the true-rms voltmeter. For fields less than threshold there is no increase in noise above the background level. As E is increased above E_T, the noise grows abruptly, until at fields above approximately 5 times threshold the conduction noise saturates.

At that knee in the noise versus bias curve the fundamental frequency of the conduction noise is approximately 100 kHz. This value is comparable to the dielectric relaxation frequency f_R in blue bronze, measured[32] to be approximately 70 kHz at 77 K. f_R is a strong function of temperature, so to test whether the knee frequency should be identified with f_R the bias-dependence experiment was repeated at 87 K by immersion in liquid argon. We found that the knee frequency increased by a factor of approximately 3.2, in rough agreement with the reported 3.5-fold increase in the dielectric relaxation frequency between these two temperatures.[32] The saturated value of the noise did not change. Furthermore, as the inset to Fig. 3 shows, the shape of the noise versus frequency curve is the same at these temperatures. However, α was larger by a factor of approximately 1.6 at the higher temperature. Theoretically α should not change at temperatures well below T_P, as neither the wavelength of the distortion nor the number of condensed electrons change. Consequently, the rough agreement between the actual knee frequency observed and the dielectric relaxation frequency is suggestive only.

We also used the traditional spectrum-analyzer technique of digitally averaging the conduction-noise spectra to estimate the bias dependence of the conduction noise. Figure 4 shows conduction-noise voltage spectra $S^{1/2}(f)$

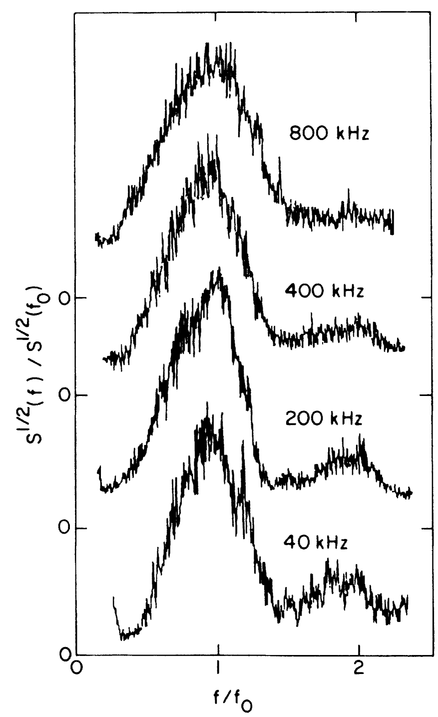

FIG. 4. Blue bronze voltage spectra at different frequencies, plotted as $S^{1/2}(f)/S^{1/2}(f_0)$ vs f/f_0, where f_0 is the frequency of the fundamental. Note that the scaled shape of the fundamental is approximately independent of f_0.

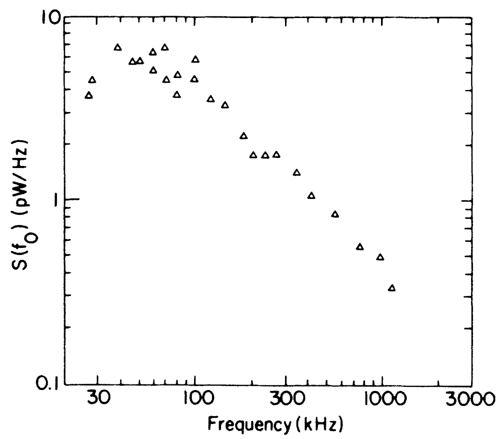

FIG. 5. Log-log plot of the digitally averaged power spectral density $S(f_0)$ of CDW conduction noise vs f_0, the frequency of the fundamental.

(in units of V $\mathrm{Hz}^{-1/2}$) at several different biases. The spectra have been plotted in linear units and scaled in both amplitude $[S^{1/2}(f)/S^{1/2}(f_0)]$ and frequency (f/f_0) so that their shapes may be compared easily. Because the shapes of the fundamental frequency components are similar, the power in the fundamentals may be crudely approximated by $S(f_0)f_0$, where $S(f_0)$ is the digitally averaged power spectral density at f_0. $S(f_0)$ is plotted against f_0 in Fig. 5. Above roughly 100 kHz, $S(f_0)$ falls off approximately as $1/f_0$, indicating that the total power in the fundamental saturates. This technique has several drawbacks, however. While in some samples the shapes of the scaled fundamental at different biases are similar, the *overtones* consistently disappear at high bias (see Fig. 4), so this measure cannot be expected to reflect accurately the total power. Furthermore, in many samples the shape of the fundamental is *not* the same at different biases, making this technique of approximating the total power useless. A more robust measure of the total conduction noise power, such as the true-rms technique, gives more trustworthy results.

V. VARIATION OF CONDUCTION NOISE WITH SAMPLE DIMENSION

Experiments in several CDW materials have addressed the origin of the conduction noise, but this fundamental question in the physics of sliding CDW's remains controversial. Typically these experiments have used spectrum analyzers to measure the change in the size of the fundamental as the size of the sample is reduced, to distinguish whether the noise is generated throughout the sample or only at the contacts. In $NbSe_3$, noise-voltage length dependences of $\sqrt{l}$ (Ref. 12) and $1-e^{-l/l_0}$ (Ref. 13) have been taken as evidence of bulk- and contact-generated noise, respectively. However, most CDW materials are relatively fragile: the inevitable bending and cutting that is associated with shortening thin fibers,[18] along with the uncertainties of spectrum-analyzer measurement, may be partly responsible for the contradictory results. On the other hand, as the large, solid crystals of blue bronze are not as likely to be damaged by handling and reduction, using the true-rms technique on this material provides an opportunity to avoid these complications.

The inset to Fig. 6 shows the total conduction noise V_N versus field (true-rms technique) for several different lengths of one specimen. The saturated value of V_N for each length is plotted against length in the main figure for two samples of different cross-sectional area. The rms conduction noise increases with length, and the data are in approximate agreement with a linear variation.

In the few cases where the shapes of the scaled spectra remained the same *at different lengths* as well as at different frequencies, the length dependence was also investigated by comparing S ($f_0 = 100$ kHz) at different lengths. In addition, the noise variation with length was studied in several samples by squaring digitally averaged voltage spectra $S^{1/2}(f)$ (taken at high bias) and then numerically integrating. While neither of these estimates of the total power is as trustworthy as the true-rms technique, they were consistent with a linear variation of conduction-noise voltage with length.

We also observed that the conduction noise increases strongly with decreasing sample cross section. In an attempt to quantify this, the two length dependences shown in Fig. 6 were extrapolated to 3.6 mm. This was a length that was reasonably well represented in several other samples. Using additional data from three other specimens, we obtained rough estimates of conduction noise for five cross sectional areas. Figure 7 shows the results of this rough assessment: $V_N \approx A^{-\beta}$, where A is the cross-sectional area and $\frac{1}{2} < \beta < 1$. The uncertainties of extrapolation prevent a confident verdict on the exact form of the variation of V_N with area, but we nevertheless observe that the smaller a specimen's cross section, the larger the size of its conduction noise. Other work in

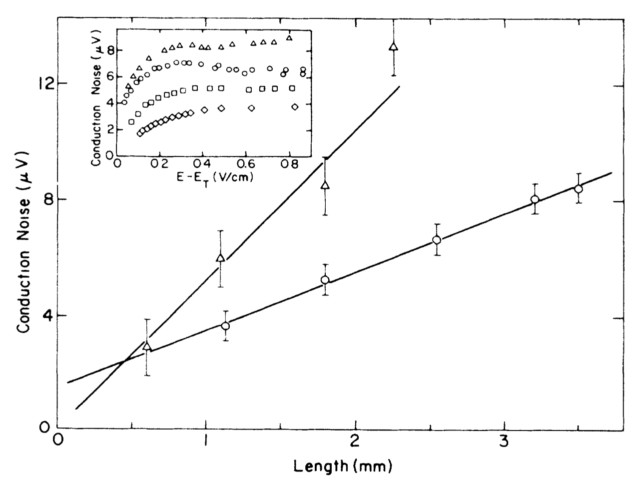

FIG. 6. Total saturated conduction noise vs length for two different specimens [areas of 1.8×10^{-3} cm^2 (triangles) and 5.3×10^{-3} cm^2 (circles)]. Lines are guides to the eye. Saturated value was calculated by averaging noise values for $5E_T < E < 12E_T$. Inset shows total noise vs $E-E_T$ for several lengths of the sample with larger area.

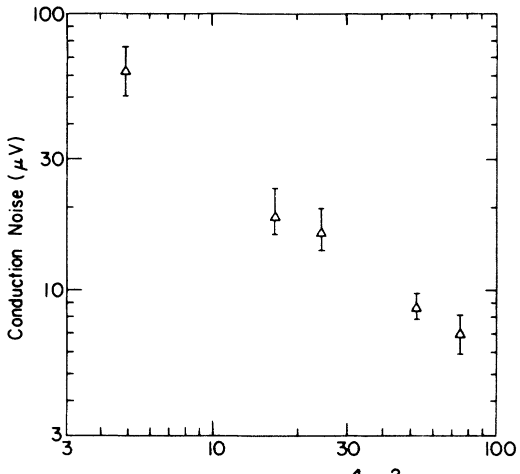

FIG. 7. Log-log plot showing total saturated conduction noise vs cross-sectional area of sample.

blue bronze[33] reported that the *height* of the fundamental spectral peak in blue bronze increased with decreasing cross section. However, peak widths narrowed at the same time, and the published data are insufficient to draw conclusions about the total noise power.

VI. DISCUSSION

This statistical investigation of blue bronze shows that the conduction noise associated with the sliding CDW is indistinguishable from Gaussian noise. This conclusion was previously reached[20] for the CDW conduction noise in $NbSe_3$. Blue bronze and $NbSe_3$ differ outwardly in several respects. While $NbSe_3$ grows in small ribbons, blue bronze crystals are much larger and tend to grow as chunks. Although $NbSe_3$ is metallic at all temperatures, the Fermi surface in blue bronze becomes completely gapped by the CDW phase transition. Furthermore, the conduction-noise spectra in $NbSe_3$ are much narrower, with ratios of width to center frequency as small as[34] 1/30 000. Our investigation finds that these differences are irrelevant to the materials' common statistical property, suggesting that the underlying source of the randomness is inherent to the sliding CDW. Evidently the Gaussian fluctuations are a central aspect of CDW transport.

The Gaussian statistics associated with CDW conduction require that a significant element of randomness be included in the explanation of the sliding phenomenon. Origins of randomness in CDW's have already been suggested theoretically. To explain the voltage noise accompanying sliding CDW's, Bleher and Wonneberger[35] have proposed "selectively amplified background noise" in the context of a single-coordinate model driven by a noise source. Miyashita and Takayama's investigation of the 1D Fukuyama-Lee-Rice model with heat bath effects[36] also proposes to account for the fluctuation in the noise spectrum. They suggest that "the stochastic aspects of the CDW current spectra . . . originate from the activated random process between quasistationary configurations of the sliding CDW."

The observed bias dependence of the noise agrees with the predictions of most, if not all, models of sliding CDW's. These include the single-coordinate classical model, including its extension to incorporate random noise;[37] Matsukawa's analysis[38] of the classical deformable model; the strong-pinning phase-slip theory of CDW dynamics by Tucker, Lyons, and Gammie;[39] and Bardeen's tunneling theory.[40] Each of these models predicts that the amplitude of the conduction noise increases abruptly when the bias is increased above threshold, and then saturates for large bias.

The crossover from increase to saturation can be understood as follows. When the CDW is sliding rapidly, its velocity is determined almost entirely by the externally applied electric field. The velocity is nearly constant because the pinning force is only a perturbation. Consequently, the temporal variation of the voltage reflects the spatial variation of the pinning force, the amplitude and shape of the wave form are independent of velocity, and the power saturates. This situation applies whenever the noise frequency f_0 exceeds the dielectric relaxation frequency f_R. When the CDW is sliding more slowly, the velocity is no longer roughly constant, small velocities are favored over larger velocities, and the size of the noise is less than the saturated value.

The increase of the size of the conduction noise with length implies that the noise is generated in the bulk of the material. Models that explain the conduction noise as a contact effect[9,10] predict that the noise voltage is independent of the length of the specimen (except perhaps for very short lengths). Consequently, noise generation at the contacts (e.g., by phase slip at sliding-pinned interfaces) cannot account for our observations.[41]

Furthermore, the observation that the size of the noise depends linearly on the length indicates that the domains extend the full length of the specimen. This is a surprising result that contrasts with behavior in $NbSe_3$, where square-root-of-length behavior[12] has been taken as an indication of noise generation at many randomly acting sources distributed throughout the volume of the specimen. This difference between the compounds might be due to the presence or absence of metallic electrons. While in $NbSe_3$ any inhomogeneous change in the CDW current could be compensated by a change in normal current, this alternate conduction channel is absent in blue bronze.

Together with the strong inverse dependence of the noise on cross-sectional area, the above interpretation suggests that CDW conduction in blue bronze involves the motion of many independent, parallel domains, or strands, each extending the full length of the sample. This strand model is consistent with other studies in blue bronze. The work of Mihály *et al.*[42] showing that CDW currents can run in opposite directions on different sides of a crystal demonstrates that the CDW conduction occurs along independent longitudinal paths. Jánossy *et al.*[43] found that a large thermal gradient applied along the length of a specimen of blue bronze failed to broaden the conduction-noise spectrum. That indicates that the velocity of the CDW is not affected by local conditions along the sample, but rather that there is a single velocity along a strand which is determined by the average conditions. Furthermore, a recent investigation by Csiba *et al.*[16] at biases very close to threshold shows that CDW conduction occurs by pulses propagating along the sample. Since the spatial extent of these pulses is the length of the crystal, their results also suggest that CDW domains stretch the entire length of the sample.

In conclusion, this investigation suggests the following picture of charge-density-wave conduction in blue bronze and the accompanying noise. The CDW conduction occurs along independent strands that extend the complete length of the crystal. The conduction noise is generated throughout the volume of the crystal by the sliding CDW's interaction with the pinning potential. The bias dependence of the size of the noise indicates the importance of dielectric relaxation in the ac motion of the CDW. Finally, the conduction noise fluctuates randomly and is not a temporally coherent oscillation. Instead, the genuine "noisiness" in the conduction process reflects the importance of underlying randomness in CDW dynamics.

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[41] Contacts could contribute a background value, which would be reflected as a nonzero intercept at zero length in the noise-versus-length measurements (see Fig. 6). Our data do not resolve this question.

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