Coherent Voltage Oscillations Induced by Sliding Charge-Density Waves: Interface or Bulk Phenomenon?

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The coherent voltage oscillations (narrow-band noise), induced by sliding charge-density waves, were investigated in $NbSe_3$ at $T = 45$ K. By applying nonperturbing potential sensors on the surface of the sample we show that a significant portion of the narrow-band noise is generated in the bulk. We found evidences for domain structure in the velocity-velocity correlation function of the sliding charge-density waves. We attempt to resolve the contradiction with previous experimental studies.

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The presence of coherent-voltage oscillations (narrow-band noise, NBN) accompanying the nonlinear conduction observed in charge-density-wave (CDW) compounds like $NbSe_3$, TaS_3, $(TaSe_4)_2I$, or $K_{0.3}MoO_3$ is one of the most exciting phenomena observed in these materials.[1,2] Together with the unusually large frequency-dependent dielectric constant and sharp threshold of nonlinear conduction, the NBN is considered a signature of the collective motion of the electrons.[3] In spite of its fundamental importance in the understanding of sliding-mode conduction there is still no widespread agreement on the specific mechanism leading to the NBN generation.

At present there are two opposing points of view in the literature, both of them supported by experimental evidence sometimes contradictory to each other. Models based on the Fukuyama-Lee-Rice Hamiltonian[4] of deformable CDWs predict narrow-band noise generated in the bulk of the sample due to the interaction of CDWs with impurities.[5,6] Recently Fisher has shown that in this case the amplitude of current oscillations scales with $1/\sqrt{V}$, V being the volume of the sample.[5] Similar conclusions can be drawn from other impurity models,[6,7] taking into account that for randomly placed impurities the existence of a finite pinning force excludes an infinite phase-correlation length. The length dependence of the noise amplitude, measured by Mozurkewich and Gruner,[8] seems to indicate that indeed, there are multiple noise sources in the specimen with randomly distributed phases. On the other hand, Ong, Verma, and Maki,[9] and independently Gor'kov[10] argued that NBN is produced during the CDW–normal-electron conversion, inevitably happening when CDW electrons leave the sample. This view is also supported by measurements, performed on samples of different lengths[11] and in the presence of temperature gradient.[9] The situation calls for further experimental studies, preferably with methods less susceptible to the irreproducibilities (discussed in detail in Ref. 11) of the earlier measurements.

For the purposes of the present investigation we classify the different mechanisms of noise generation into four groups. For models belonging to class A there is no time variation in the current carried by the CDW, and NBN voltages are generated in a limited volume at the current contacts [Fig. 1(a)]. For class B models the CDW still moves as a rigid object, but the velocity (and consequently the CDW current) has an oscillating component in time [Fig. 1(b)]. The variations in the velocity may be induced by the contacts[5] or by the interaction with impurities.[12] For class C and D models the sample can be considered as a collection of oscillators. In case C these oscillators are not entirely independent as they have the same time-averaged frequency, but random fluctuation around the mean leads to dephasing between them. This picture may correspond to a deformable CDW sliding over pinning centers.[5] In case D we remove the restriction on the time-average frequency by allowing CDW–normal-electron conversion inside the sample. This would allow the possibility of the NBN spectrum to split into multiple peaks, each of them corresponding to a domain limited by surfaces of electron conversion.

In this Letter we attempt to distinguish between these four conceptually different possibilities of noise

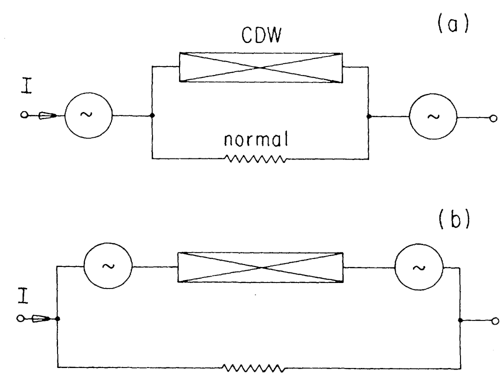

FIG. 1. Equivalent circuits representing two extreme cases of NBN generation by rigidly moving CDWs. The effect of sliding CDWs is replaced by a nonlinear resistor and the ac voltage sources. The circuits exhibit voltage oscillations when driven by constant current I.

generation by investigating the narrow-band voltage oscillations in $NbSe_3$ at 45 K. We have developed a new technique to change the distance between potential sensors without removing the sample from the cyrostat. These are the first measurements where the length dependence of the NBN was studied without significantly changing any other parameters of the measurement.

Gold wires were attached to the two ends of the high-purity $NbSe_3$ needle crystals (typical dimensions $5\ \mu m \times 10\ \mu m \times 1000\ \mu m$ with threshold field, $E_T = 30$ mV/cm and residual resistivity ratio of 130 ± 10) by the standard silver painting method. These leads (denoted by 1 and 3) served as current contacts to drive constant current through the specimen. Potential probes were also applied along the sample but they were not fixed by silver paint. These sensors were made of impure $NbSe_3$ fibers, connected at one end to a gold lead and pressed to the side of the sample. The elasticity of the sensor fiber prevented any excessive mechanical stress on the sample but still ensured a reasonable contact resistance in the order of 50–500 Ω. In one set of experiments two of these touching probes (2 and 2′) were used, spaced approximately equidistant between the ends of the sample. In other experiments we applied only one touching probe (2), but we were able to move it along the sample while the sample was inside the He gas-flow cryostat. The position of probe 2 was deduced from low amplitude resistance measurements ($I_{ac} < I_T/20$ where I_T is the threshold current), assuming that the contact resistance is small. We estimate a 10% systematic error in the inferred contact position due to the finite contact resistances.

The sample was driven by a constant total current, and the voltage drop between the contacts was detected by the differential amplifiers of a Tektronix 7603 oscilloscope (input impedance 1 MΩ, bandwidth 1 MHz). By using two channels we simultaneously investigated two segments of the sample. The signal from one channel of the oscilloscope was fed to a HP 8557A spectrum analyzer in order to determine the intensity of the main harmonic component. A dedicated IBM personal computer performed the data evaluation.

We made several tests to check the performance of the device. The transmission function of the setup was investigated by mixing and analyzing known amplitude ac signals. We concluded that the most effective way to estimate the signal is to record several high-resolution (narrow bandwidth, usually 10 kHz) spectra, take the digital average, and calculate the integrated intensity above the background noise level. The result is somewhat less than the real amplitude, the correction being on the order of bandwidth times background noise level. Fortunately, the background noise was relatively low and did not change in our experiments and this allowed us to draw quantitative conclusions from the noise amplitude measurements.

We measured narrow-band noise between the current leads and attempted to observe a change in the spectrum when we removed, or put back the potential sensor(s). We did not see any measurable change even for samples where the noise spectrum had more than one fundamental frequency. This crucial test shows that, contrary to silver painted contacts,[13] the touching probe does not act as a strong perturbation. No phase shift was observed when low-amplitude ac signals were applied with frequencies comparable to the measured noise frequency, indicating that the capacitive coupling between the leads is negligible.

For class-A models there is a fluctuating $u_{13}(t) = u_1(t) + u_3(t)$ voltage between the ends (u_1 and u_3 being the noise voltage generated at the end contacts 1 and 3), but there is no noise between the potential probes 2 and 2′ as the CDW current I_{CDW} is independent of time and the voltage here is entirely determined by $u_{22'} = I_{norm} R_{22'} = (I_{tot} - I_{CDW}) R_{22'}$ where I_{norm} is the current carried by the normal electrons and $R_{22'}$ is the Ohmic resistance. Experiments on samples having two potential probes clearly demonstrated that the noise is not localized to the current contacts in this manner. We were always able to detect narrow-band noise between probes 2 and 2′ indicating that the CDW current oscillates in the entire volume of the sample. This test still leaves open the question of contact-induced oscillations in the CDW current which leads to oscillating normal current and NBN voltage on any segment of the sample (class-B models). Measuring the length dependence of the NBN intensity and comparing the phase of the NBN of different segments is the only way to distinguish between class-B and -C models.

In case B, $I_{norm} = I_{tot} - I_{CDW}$ oscillates coherently along the sample and therefore the NBN amplitude U_{12} scales linearly with the distance between contacts 1 and 2. The relationship $u_{13}(t) = u_{12}(t) + u_{23}(t)$ holds at every time t, which means that the oscillation amplitudes add up, $U_{13} = U_{12} + U_{23}$, and there is a strong phase correlation between $u_{12}(t)$ and $u_{23}(t)$. In case C there is no phase correlation between U_{12} and U_{23} and the time-averaged noise amplitude $U_{13} \leq U_{12} + U_{23}$.

Figure 2 shows the result of the length-dependence study on samples having a single fundamental noise frequency. Altogether three samples were investigated, with the NBN frequency set to the vicinity of 500 kHz by the proper choice of dc current. The vertical scale in the lower portion of the figure gives the noise amplitude from each side of the sample (normalized to the sum $U_{12} + U_{23}$ in order to easily see the overall behavior). The general trend is that the noise amplitude increases with contact separation. We emphasize

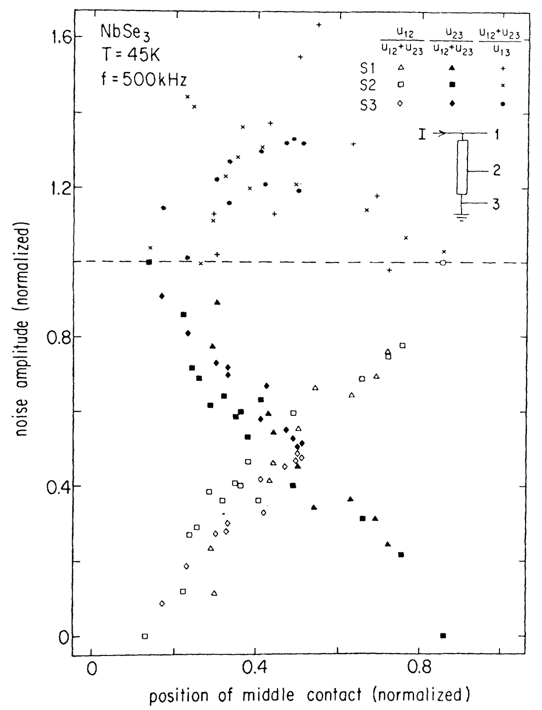

FIG. 2. Time-averaged noise amplitude vs position of contact 2. The total length of each sample ($S1,S2,S3$) is normalized to 1. The open symbols correspond to NBN amplitude between probes 1 and 2 (U_{12}, see inset) normalized to $U_{12}+U_{23}$, the solid ones denote $U_{23}/(U_{12}+U_{23})$. The sum $U_{12}+U_{23}$, normalized to the measured total NBN amplitude U_{13}, is also plotted. Note that (i) the NBN amplitude changes along the sample excluding noise generation corresponding to Fig. 1(a), and (ii) the amplitude does not scale linearly (because the sum/total is systematically greater than 1), excluding the case presented in Fig. 1(b). The latter conclusion is further supported by investigation of the phase of the oscillations (see text).

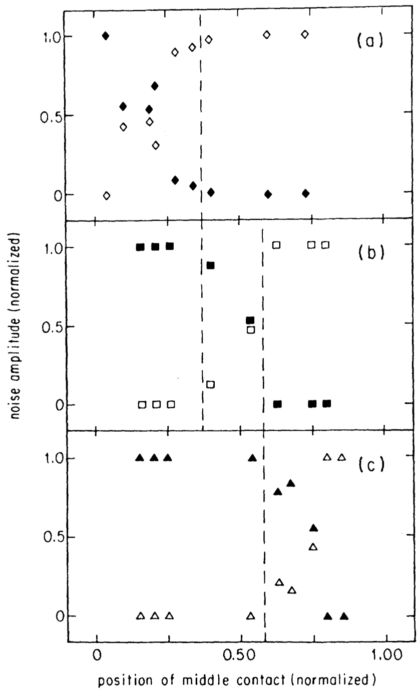

FIG. 3. Length dependence of NBN amplitude for a sample having multiple fundamental frequencies. The amplitude of the fundamentals are normalized separately similar to Fig. 2. Each frequency is found to be confined to a limited volume of the sample with boundaries estimated by the dashed lines.

that this increase is not linear, as can be seen from the data in the upper portion of the figure. This shows that the sum of the amplitudes from the two segments is systematically greater than the amplitude U_{13} between the ends, the difference being more pronounced when the touching probe is at the middle of the specimen. Since the noise amplitude is not linear with distance, case B (corresponding to rigid CDW motion) can be excluded. On the other hand, the observations are completely consistent with case C [for entirely random phases one expects $(U_{12}+U_{23})/U_{13}=\sqrt{2}$ when the potential probe is at the middle].
generated in the bulk of the sample is further supported by investigation of the relative phase of the NBN on the two sides of the sample. Oscilloscope photographs were taken by triggering the oscilloscope from one channel and simultaneously (chopped mode) detecting the signal in the two channels. If there is a phase correlation then the sine-wave-type structures should appear in both traces. On the other hand, phase fluctuations tend to cover up the structure for the nontriggered trace. Only a blurred trace is actually observed.[14]

It is well known that many of the samples, picked at random, have more than one fundamental noise frequency. After establishing that in single-mode samples a major portion of the NBN is generated far from the contacts, we attempted a search for longitudinal domain structure in these multiple peak samples. Figure 3 shows the length dependence of noise amplitude for a specimen having three well separated fundamental NBN frequencies. The results show that for a given noise peak the amplitude U_{12} depends on the position of sensor 2 only over a range smaller than the sample length. Complementary length dependence is observed for amplitude U_{23}. Outside of this range there is no length dependence and one of the amplitudes completely disappears. This observation can be explained by domain structure in the velocity-velocity correlation function. The remarkable aspect is that one of these domains [associated to the fundamental plotted in Fig. 3(b)] is confined completely to the interior of the sample.

In conclusion, all evidence we gathered by measuring the narrow-band noise amplitude and phase on different segments of $NbSe_3$ crystals seems to point to the same direction: A significant part of the NBN is generated in the bulk of the sample and the phase of the voltage oscillations is randomly fluctuating in time and space. Our observations are consistent with the results obtained by Mozurkewich and Gruner. On the other hand, in multidomain samples we observed noise amplitude independent of length. This may resolve the contradiction between Ref. 8 and Ref. 11.

We believe that in a "single-mode" sample, domain structure can be generated by application of a temperature gradient along the specimen. Moderate gradients leave the sample in the single domain state and do not induce splitting in the noise spectrum.[15] Medium gradients split the sample into two domains leading to doubling of the fundamental.[9] Preliminary results indicate that higher-temperature gradients produce multiple splitting in the noise spectrum,[16] corresponding to several domains in the specimen.

Our findings can be qualitatively interpreted in terms of a deformable CDW sliding over pinning centers. The basic assumption is that the noise frequency is proportional to the local CDW velocity. If there is no CDW–normal-electron conversion inside the sample then the mean velocity of CDWs is uniform along the specimen and there is a well-defined fundamental in the noise spectrum. However, it is quite possible that the deformation state of the CDW fluctuates in time. This leads to fluctuations in the local CDW velocity, and finally it results in dephasing for the NBN generated at different portions of the specimen. The length scale of this dephasing process may well be temperature and field dependent,[5] but under the circumstances of our study it is certainly shorter than the sample length.

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