Tunable harmonic vibrations of quasi one-dimensional conductors induced by sliding charge-density waves

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We demonstrate that samples of quasi one-dimensional compounds show torsional vibrations induced by the sliding charge-density wave (CDW). Narrowband noise at the fundamental frequency is found in the vibrations spectra. The result suggests an approach to studies of the CDW dynamics and presents CDW conductors as tunable (ultra)sonic acoustoelectronic generators—unique solids vibrating under dc voltage.

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I. INTRODUCTION

The condensed electronic state, charge-density wave (CDW), is a periodic modulation of both the lattice-site positions and electron density forming at the Peierls transition temperature, T_P.[1–3] The transition converts a quasi one-dimensional conductor into a dielectric due to the gap 2Δ opening at the Fermi surface. Below T_P the conductivity of the samples falls down, but it strongly increases at electric fields, E, above the threshold value, E_t, when the CDW begins to slide, providing the Fröhlich mode of charge transfer. The sliding is accompanied by low-frequency broadband electric noise (BBN) and high-frequency narrowband noise (NBN).

The BBN and NBN generation is widely studied in a number of quasi one-dimensional conductors. Generally speaking, NBN reflects sliding in a periodic pinning potential, while BBN arises from the inhomogeneity of sliding in space or in time. The NBN frequency, f_f, equals to the inverse time of the CDW advancement by one wavelength, λ, or in other words, of $\approx 2e$ charge transfer per CDW chain: $f_f = i/2e$, where $i = I_{cdw}s_0/s$ is the current through a CDW chain, I_{cdw}—the total CDW current, s—sample cross-sectional area, s_0—area per chain. Thus, f_f is directly related to the CDW periodicity and, therefore, is called "fundamental." Although the origin of noise is not unambiguous,[4–9] it is clear that the generation of noise is distributed over the whole volume of the sample.[10–12]

CDW as an electronic crystal is an elastic medium.[2,3] Electric-field-induced CDW deformation is known to reveal itself in enormous nonuniform strains of the samples.[13] The kinds of nonuniform strain are likely to be various,[14,15] but the most studied one is the torsional strain (TS).[13,16,17] The threshold and hysteretic field dependences of torsional angle, $\delta\varphi(E)$, clearly show that TS is coupled with the CDW deformation. We have observed enormous TS for $K_{0.3}MoO_3$, $(TaSe_4)_2I$, NbS_3, but the most detailed studies were performed for TaS_3, which has been taken for the present studies.

In the experiments cited above, the CDW was deformed by *external* action, namely, voltage variation, and this deformation was transmitted to the crystal. Here, we consider a CDW sliding in a constant field. In this case its time-dependent deformations reflect *internal* dynamics of the CDW. In view of the mechanical coupling of the CDW and the lattice, one can expect transfer of these deformations to the crystal. If so, a promising goal is to search for such vibrations, which can be expected at the same frequencies as the electric noise.[18]

In this paper we report studies of vibrations of the quasi one-dimensional conductor TaS_3 under dc electric field $E > E_t$. The samples, arranged as multimode mechanical resonators, demonstrate harmonic oscillations at a number of resonant modes. We show that these oscillations are excited at the fundamental frequencies of the CDW sliding and thus can be tuned by the electric field. BBN vibrations are also observed; the details are reported elsewhere.[20] The CDW conductors appear as unique examples of solids vibrating under a dc field.

II. EXPERIMENTAL DETAILS

We concentrated our studies on the *torsional* vibrations, because they appeared especially convenient in study:[13] our optical technique resolves torsional angles on the order of 10^{-7} rad, which corresponds to the shear at the surface $\sim 10^{-(9-10)}$. TaS_3 samples ($T_P = 220$ K, $2\Delta \approx 120$ meV) from a high-quality batch with $E_t \sim 0.3$–0.4 V/cm were selected. Figure 1 illustrates the idea of the experiment. One end of the whisker (1) is tightly fixed at the substrate with an indium piece (the black oval) serving as a contact. Thus, the whisker appears suspended over the substrate. The contact to the second end is provided by means of a long, thin (a high-temperature superconductor (HTSC) $Ba_2Sr_2CaCu_2O_x$ whisker) wire (2) soldered with conducting epoxy. The configuration allows nearly free TS of the samples. A micromirror (3)—a laser-cut $Ba_2Sr_2CaCu_2O_x$ whisker covered with a golden film— is stuck to the sample near the free end.[13] A two-sectional photodiode (4) is fixed near the cryostat; thus, the output signal from the amplifier (5) is proportional to the beam deflection, i.e., the torsional angle.

At fixed dc current (7), we could simultaneously measure the time-dependent angle, $\varphi(t)$ and voltage across the sample, $V(t)$. The two devices marked as "V" (6 and 8) could be either selective ac nanovoltmeters (Unipan-237) or two channels of a digital oscilloscope (Acute DS 1002).

All the experimental data presented were acquired near liquid-nitrogen temperature.

III. EXPERIMENTAL RESULTS

We started a search of the vibrations with BBN: the amplitude of BBN voltage is at least an order of magnitude higher than that of NBN.[21] Besides, below the first torsional resonance ($\sim$1 kHz), the TS is not affected by the mechanical characteristics of the suspended structure. Time domains of

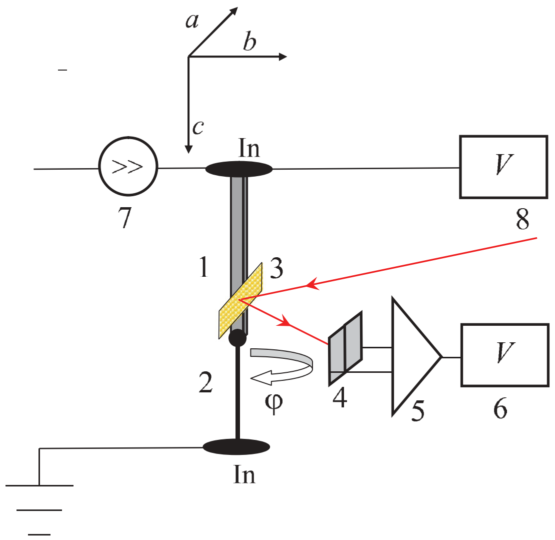

FIG. 1. (Color online) The scheme of the experimental setup. 1, the sample; 2, thin conducting wire; 3, micromirror; 4, two-sectional photodiode; 5, differential amplifier; 6 and 8, ac voltage-monitoring devices in the optical and voltage channels, respectively; and 7, source of dc current. The arrows above point out the crystallographic axes of TaS_3.

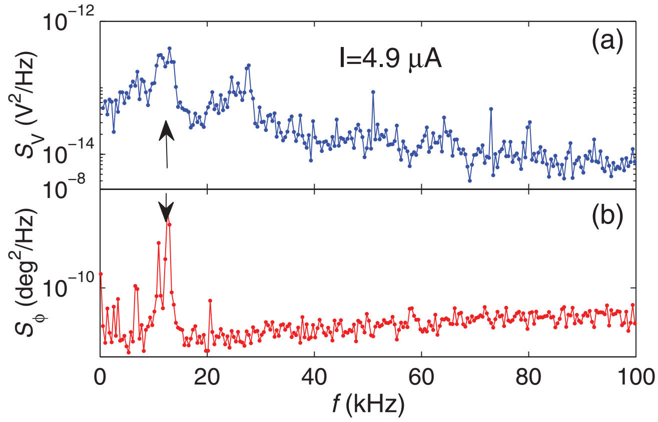

FIG. 2. (Color online) The spectra $S_V(f)$ (a) and $S\varphi(f)$ (b) measures at $I = 4.9\ \mu$A. At this current f_f equals 12.3 kHz (see the arrows) and coincides with the first resonance (see Fig. 4).

the ac voltages across the sample and across the photodiode were recorded simultaneously at different I values. Power spectral densities of the TS and of the voltage noise, $S_\varphi(f)$ and $S_V(f)$, respectively, were obtained at the frequency range 0.025–200 Hz. Similar features in the evolution of both spectra with current were found. Fluctuations of voltage and, especially of the angle, grow sharply at currents around I_t and saturate approximately at $3I_t$. For ∼3-mm-long sample $S_\varphi(f)$ achieves 10^{-6} deg^2/Hz at low frequencies, i.e., $\delta\varphi \sim 10^{-3}$ deg. Both spectra can be approximated as $1/f^\alpha$, with α decreasing from 1.5 to 0.5 with I growth. This means that the spectral densities begin to grow at first at low frequencies. Qualitatively similar behavior of torsional vibrations was found for another compound, $(TaSe_4)_2I$. The details of BBN in TS are presented elsewhere.[20]

The central goal of this paper was to search for the NBN vibrations. First, we made sure that the selected samples demonstrate distinct NBN in voltage. Typically, we could distinguish two or three maxima over each $S_V(f)$ curve [Fig. 2(a)] whose frequencies were shifting upwards with increasing I. The splitting of the NBN maxima indicates a domain structure of the CDW sliding, characteristic of TaS_3 below 100 K.[5] The frequency of the main maximum appears linear in I_{cdw}, (Fig. 3, the squares) with the ratio $I_{cdw}/sf \approx 26$ A/(MHz cm^2), typical of TaS_3,[22] although reduced because of the multidomain structure.

NBN was observed only above ∼1 kHz. At such frequencies the amplitudes of torsional oscillations are affected by the amplitude-frequency characteristic of the suspended structure (Fig. 4).[23] The oscillating system is complex; therefore, a series of resonant peaks of different height can be seen in Fig. 4; some of the resonances could be attributed to the flexural oscillations of the micromirror.[15]

Studies of the spectra $S_\varphi(f)$ showed that harmonic torsional oscillations at f_f could be excited only when f_f coincides with one of the resonances [Fig. 2(b) shows an example]: we distinguished peaks on the $S_\varphi(f)$ curves at the resonant frequencies that could "flare up" and "dim" with dc current change. We interpreted this as $f_f(I)$ passing a given resonance. This observation, however, is difficult for documentation, because the peaks heights strongly fluctuate. Therefore, we decided to go another way.

Instead of measuring frequency dependences of voltage from the photodiode at given currents, we recorded *current* dependences of noise at fixed frequencies with the help of a selective nanovoltmeter. The filter of the nanovoltmeter was tuned so that its frequency coincided with one of the resonances (Fig. 4). The output signal was rescaled into the "noise angle," $\delta\varphi_n$. If at some current the sample showed torsional oscillations at a frequency coinciding with the settings of the filter, we would observe a peak of $\delta\varphi_n$ at this current. The quality factor

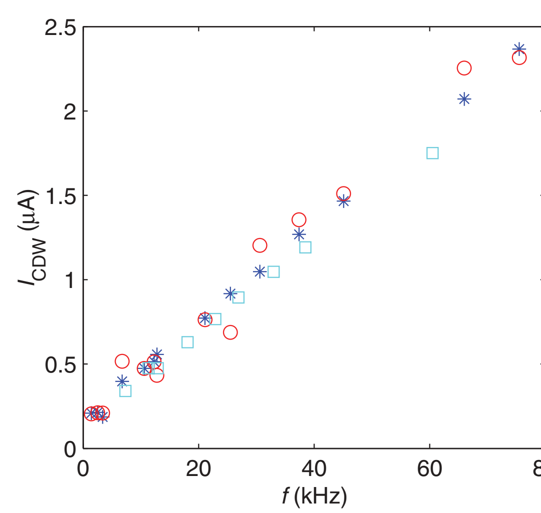

FIG. 3. (Color online) The relation between I_{cdw} and f_f. □, I_{cdw} corresponding to the frequency of the main maximum of $S_V(f)$ curve [Fig. 2(a)]. ✳, I_{cdw} corresponding to the main peak of $V_n(I)$ vs filter frequency [Fig. 5(a)]. ○, I_{cdw} corresponding to the main peak of $\varphi_n(I)$ vs filter frequency [Fig. 5(b)]. To reduce the dynamical error, the current of the peaks in $V_n(I)$ and $\varphi_n(I)$ were taken as half difference of the positions of the peaks at I and $-I$.

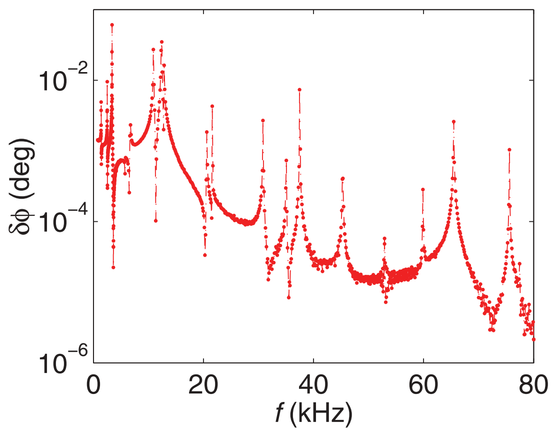

FIG. 4. (Color online) The amplitude of TS ($\delta\varphi$ rms) under ac voltage 185 mv vs frequency (Ref. 23). $V_t = 125$ mV. The peaks reveal different resonant modes.

of the filters was about 30, and the averaging time constant was ~1 s. A similar nanovoltmeter tuned to the same frequency was measuring noise, δV_n, in the voltage channel (Fig. 1). The voltages from the analog outputs of the voltmeters were recorded simultaneously as a function of current through the sample.

Figures 5(a) and 5(b) show sets of $\delta V_n(I)$ and $\delta\varphi_n(I)$ curves, respectively, for different settings of the filter. One can see common features of the noise in voltage and in TS. In both cases a series of two to three peaks of different heights is observed. Tuning the filters to higher frequencies results in a shift of the peaks rightward along the current axis.

The main peak at each of the $V_n(I)$ curves [indicated by arrows in Fig. 5(a)] corresponds with the current, at which the fundamental frequency, f_f, coincides with the filter setting. This is clear, because the values of the correspondent nonlinear current vs filter frequency (Fig. 3, the stars) follow the same straight line as the squares. The central point is that the $\varphi_n(I)$ curves show peaks [see arrows in Fig. 5(b)] at the same currents, as the $V_n(I)$ curves. This means that the torsional vibrations are excited by the sliding CDW at the washboard frequencies f_f (Fig. 3, circles). To the best of our knowledge, this effect, i.e., excitation and tuning of harmonic mechanical oscillations by means of a dc current, is unique in solids. The closest analog we have found is the observation of flexural oscillations of a nanotube under periodic force in the Coulomb blockade mode.[24]

The vibrations cannot be reduced to the effect of the NBN voltage on the lattice: the contribution of the NBN voltage to TS appears two to three orders of magnitude below the value of $\delta\varphi_n$ observed. This can be easily seen from Figs. 4 and 5 [e.g., at 12.3 kHz under ac voltage 185 mV, the $\delta\varphi$ amplitude is $\sim 3.5 \times 10^{-2}$ deg (Fig. 4)]. Therefore, the NBN voltage 14 μV [Fig. 5(a)] would induce $\delta\varphi_n \sim 3.5 \times 10^{-2}$ deg $\times$ 14 μV/185 mV $= 2.6 \times 10^{-6}$ deg, while the observed value is 10^{-3} deg. Thus, NBN in TS results from immediate mechanical effect of the CDW on the crystal. This suggests another approach to the studies of the CDW dynamics: it reveals itself directly through the dynamics of the crystal, not mediated by the noise voltage.

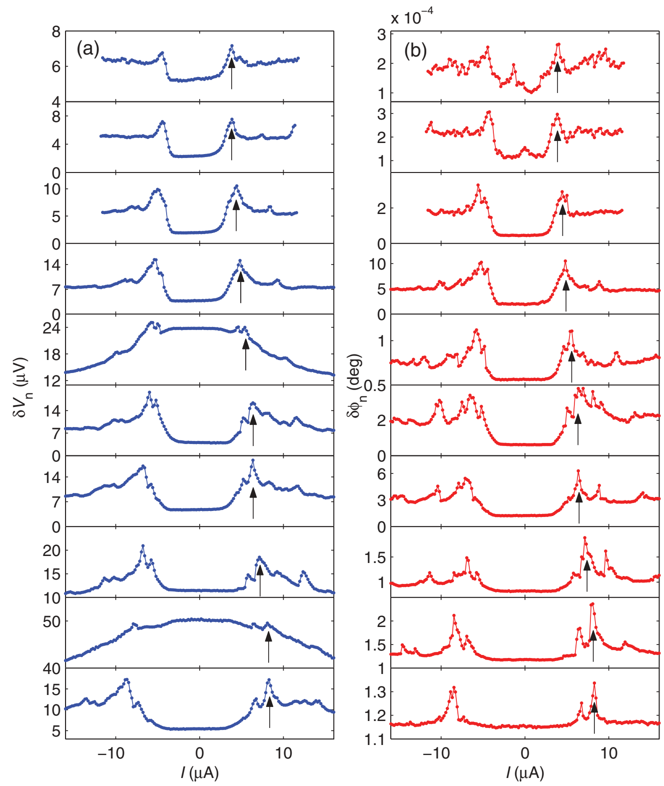

FIG. 5. (Color online) Current dependences of the noise voltage (a) and the torsional angle (b) at fixed filter frequencies: 1.45 (upper curves), 2.54, 6.77, 12.3, 21.1, 30.6, 37.4, 45.1, 66, and 75.5 kHz (lower curves). Several curves are omitted. The frequencies are selected to match the resonant modes (Fig. 4). $T = 84$ K. The arrows indicate the currents of the main peaks. The sample dimensions are $4500 \times 17 \times 7\ \mu m^3$.

IV. DISCUSSION

Let us discuss the origin of these (ultra)sonic vibrations of the lattice. First, the vibrations clearly demonstrate that the CDW conduction is accompanied by periodic in time displacements of the lattice sites. Evidently, the vibrations are not directly coupled with the oscillations on the λ length scale but reveal long-range strains of the CDW. The CDW, when sliding, is periodically deformed. This is clear both for the so-called weak pinning[4,5] and phase-slip[6–9] (PS) models of NBN. Within the weak-pinning model, the CDW motion occurs via local jumps of a wavelength in different places in the sample.[25] Each jump results in a phase gain 2π between adjacent domains (phase-coherence volumes). One can estimate the coherence volume as a product of longitudinal phase coherence length, $L_{2\pi} \sim 10\ \mu$m and square of perpendicular coherence length, $\sim 1\ \mu m^2$.[26] Within each volume, the CDW deformation (longitudinal, for simplicity), $\lambda/L_{2\pi}$, is on the order of 10 Å/10 μm $= 10^{-4}$. This results in a longitudinal sample strain, which (below 100 K) is an order of magnitude less than that of the CDW, $\sim 10^{-5}$ [Fig. 2(d) in Ref. 27]. In the representative sample the number of coherence volumes, N, is about $3500 \times 16 \times 10\ \mu m^3/10\ \mu m^3 \approx 6000$, and the macroscopic deformation should be $\sim 10^{-5}/\sqrt{N} = 4 \times 10^{-8}$. The typical amplitude of BBN in TS is of the order of 10^{-3} deg, and the correspondent shear is $\sim 4 \times 10^{-8}$, in agreement with the estimate. Here, we suppose that the shearing and uniaxial components of the strain can be of the same order; take notice also of strong transverse effects for the sliding CDW, which indicate dephasing of the CDW chains, i.e., CDW shear.[28]

The PS interpretation of the vibration is even more transparent. Near a PS center, the CDW periodically releases phase by 2π and periodically deforms, again, by $\lambda/L_{2\pi}$. For quantitative estimates of the sample deformation, one needs to know the distribution of PS centers in the sample and, probably, take into account their synchronization. Spatial distribution of phase slippage has been studied in detail in Refs. 11 and 12. Clearly, the number of PS centers in the sample volume can be great. However, we do not delve into the qualitative interpretation of the PS-induced torsional vibrations, because this would be too uncertain. In either treatment of the vibrations, CDW deformations should be involved. In contrast, NBN voltage is present in the rigid CDW model, while the CDW elasticity can suppress it.[2]

The NBN vibrations nominate the CDW conductors as acoustoelectronic generators for microelectromechanical-nanomechanical systems (MEMS-NEMS)—unique single-crystalline solids vibrating under dc voltage. The vibration frequency can be tuned by the bias. In our experiment the highest frequency (about 75 kHz) is set by the simple experimental technique. As one can see from Fig 5(b), the NBN torque does not tend to reduce with the growth of $I_{\rm cdw}$. The highest fundamental frequency observed is 16 GHz.[29] Clearly, in the case of mechanical oscillations, one can also advance along the frequency scale, especially for nanocrystals.[24,30,31] Besides, thinner samples are expected to show higher vibration amplitudes.[13] Note that detection of high-frequency noise in *voltage* is complicated because of the parasite capacitance of the amplifier, especially for high-ohmic samples. This problem does not concern the mechanical oscillations, as the transfer of the dc current into ac mechanical oscillations is direct.

V. CONCLUSION

In conclusion, we have found that the CDW conductors show a unique effect—tunable mechanical oscillations under dc current. The oscillations are observed at the fundamental frequency of the sliding CDW, and this demonstrates that the CDW conduction is accompanied by periodic in-time displacements of the lattice sites. In contrast to the NBN voltage, which, in principle, could exist even in the case of a rigid CDW,[2] the vibrations can be explained only involving internal degrees of freedom of the CDW. The effect opens an approach in the studies of CDW dynamics and can suggest ideas in construction of MEMS-NEMS and acoustoelectronic devices.

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