

Quantization of states and strain-induced transformation of charge-density waves in the quasi-one-dimensional conductor TaS₃

S. G. Zybtshev and V. Ya. Pokrovskii*

Kotel'nikov Institute of Radioengineering and Electronics of RAS, Mokhovaya 11-7, 125009 Moscow, Russia

(Received 4 March 2016; published 16 September 2016)

We report studies of low-field conductivity, σ , of nanosized samples of orthorhombic TaS₃ as a function of strain, ε . The $\sigma(\varepsilon)$ curves show steplike changes associated with the “quantization” of the wave vector, q , of the charge-density wave. Based on the effect we have revealed the change of the q -vector with strain: its in-chain component (normalized by the reciprocal lattice constant) is found to *increase* with sample stretch. A similar $q(\varepsilon)$ dependence results from the analysis of the $\sigma(\varepsilon)$ hysteresis for macroscopic samples. This means that the strain-induced anomalies cannot be explained by the transition of the CDW to fourfold commensurability with the pristine lattice (lock-in transition), as it was supposed earlier. We also discuss the metastable length states and the elastic anomalies in TaS₃ in light of the observed $q(\varepsilon)$ dependence.

DOI: [10.1103/PhysRevB.94.115140](https://doi.org/10.1103/PhysRevB.94.115140)

I. INTRODUCTION

The most remarkable feature of quasi-one-dimensional conductors with incommensurate charge-density waves (CDWs) is the nonlinear transport associated with the CDW sliding [1]. Some of the compounds which belong to this group show also unique properties, which, to a wide extent, can be called mechanical, namely, strain-induced features in conduction and thermopower [2–6], anomalous elastic [6–13], thermal-expansive [14], and electromechanical properties [15–19]. In particular, abrupt changes in nonlinear and linear transport under uniaxial stretch [3–6] accompanied with a feature in the stress-strain relation [13], drop of the Young’s modulus [7,8,10,11] and shear modulus [6,9,11,12] on the CDW depinning, hysteresis in thermal expansion [14], large electric-field-induced deformations (uniform [15] and nonuniform [16–18]) have been reported. All these features demonstrate interplay of the CDW and pristine-lattice properties.

The effects mentioned above are found in a number of compounds and clearly reveal general features of the CDW. At the same time, they reflect individual properties of each CDW compound. For example, a several-percent drop of the Young’s modulus on the CDW depinning has been reported for orthorhombic TaS₃ (in the text below, TaS₃) and (TaSe₄)₂I [1,2], —one to two orders of magnitude lower for NbSe₃ [11,20], and no drop (to the accuracy of 5×10^{-5}) for K_{0.3}MoO₃ [21]. Such a scattering of properties could be expected from general consideration (see the discussion in Ref. [12], p. 2970). If one stretches a sample and no CDW phase slippage occurs, the wavelength, λ , will grow proportionally to the length, L , or lattice constant, c . The condition $\lambda \propto c$ also implies conservation of the CDW phase with respect to impurities [12]. In principle, this value of λ can be different from the *equilibrium* λ for the strained sample. However, in the case of an ideal one-dimensional (1D) conductor the relation $\lambda \propto c$ coincides with the condition for the equilibrium value of λ . In fact, in the simple 1D model the CDW wave vector q is exactly equal to $2k_F$, or $\lambda \equiv \pi/k_F = 2/n_0$. Here k_F is the Fermi wave vector; n_0 is

the linear concentration of electrons over a conducting chain. With $n_0 \propto 1/c$ we come to $\lambda \propto c$. Thus, within the simple 1D model the CDW in a strained sample remains in equilibrium with respect to the strained lattice and does not contribute to the elastic energy (Eq. (2) from Ref. [14]). Then, the mechanical anomalies must originate from individual features of the compounds, such as charge transfer under strain [22–24], three-dimensional (3D) effects (nesting), and electron-hole asymmetry [25,26]. These effects can either violate the relation $n_0 \propto 1/c$, or make λ deviate from $2/n_0$ under strain [24,27]. In both cases $\lambda/c \neq \text{const}$, or, in terms of q , $q/c^* \neq \text{const}$. Below this condition is referred to as a “nontrivial” dependence of q (namely, of the in-chain component of the q -vector) on strain.

For the following it is reasonable to introduce the dimensionless value $q' \equiv q/c^*$. The dependence of q' on strain, ε , gives the most common interpretation of the anomalies, at least, if longitudinal strains are concerned. In this case the CDW-lattice coupling can be characterized in terms of the coefficient $g \equiv (\delta q'/q')/(\delta c^*/c^*) = -(\delta q'/q')/\varepsilon$ introduced in Ref. [27]. If $g = 0$, the strain dependence of q is trivial, and no anomalies are expected.

The most detailed mechanics-related studies have been performed for TaS₃ [3–10,12–20], a typical Peierls quasi-one-dimensional conductor with CDW forming at $T_P = 220$ K [1]. For this compound an integral picture of lattice-CDW interplay has been arranged. Basic for this picture is the assumption that under uniaxial stretch q' decreases and at a critical strain ε_c , achieves fourfold commensurability with the lattice, i.e., shows a lock-in transition. At $\varepsilon > \varepsilon_c$ q' is supposed to reduce further [3–6]. This inference is based on a number of experiments showing pronounced features in conductivity (both linear and nonlinear) and thermopower at ε close to ε_c . The $q'(T)$ dependence [28] seemed to confirm this picture: Around 220 K q' is slightly above 1/4, but approaches the fourfold commensurability with T decrease. The dependence $\varepsilon_c(T)$ was found to be qualitatively similar with $q'(T)$ [3,4,14]. Confronting the two dependences [14] gave $g = 6$. This inference implied that ε_c is the strain of the lock-in transition.

The nonzero value of g is also a key for explaining the softening of the lattice with the CDW depinning [12,24,27] at the electric field $E = E_i$. As we concluded above, $g \neq 0$ means that the CDW in a strained sample appears out of

*pok@cplire.ru

equilibrium (with respect to the changed lattice constant) and contributes to the total elastic energy of a sample. However, in the sliding state the deformations of the CDW relax (through phase slippage and creep), their contribution drops out from the total energy, and the Young's modulus of the sample decreases.

Thus, by now, a consistent-looking picture of mechanical anomalies in TaS₃ has been arranged. However, an important element of this picture is lacking: The q' change under strain has not been studied. The experiments presented in this paper reveal the $q'(\varepsilon)$ dependence. Contrary to the expectations, q' is found to *increase* with ε . This result appears in contradiction with the picture described above and gives ground for a serious reconsideration of the mechanical anomalies in TaS₃ and, evidently, in other CDW compounds.

II. EXPERIMENTAL APPROACHES

Structural studies under strain, including those at low temperatures, require an unconventional setting of the experiment and can be rather complicated. However, studies of the transport properties of the CDW conductors in the linear regime of the current-voltage characteristics can give unambiguous and precise information about the q' change. The most direct technique is based on the effect of q -vector “quantization.” In nanosized samples discrete conducting states corresponding to different integer numbers of CDW periods can be resolved [29–32]. Transitions between the states are observed as steplike changes of conductivity, σ , and reveal production or annihilation of a CDW period through a phase slip (PS) event. One PS results in $\delta q'/q' = \pm\lambda/L$. Counting the number of steps in $\sigma(T)$ one can find the q' change in the corresponding temperature range. This approach provides an extremely high resolution in $q'(T)$ change, which can exceed that of the x-ray studies, as in the case of K_{0.3}MoO₃ [30,33]. Here we apply this technique to reveal $q'(\varepsilon)$.

Though steps for TaS₃ nanosamples were observed previously [29,34], discrete equidistant conducting states and regular switching between them with temperature were not clearly demonstrated. For observation of the “quantization” the sample must be thin enough for the CDW to be coherent in cross section, and short enough for the σ steps to be resolved. The dimensions required for TaS₃ are $\sim 10^{-3} \mu\text{m}^2$ in cross section and not more than tens of microns in length. In addition, it is important to impose certain boundary conditions for the CDW at the contacts [30,35]. This requirement can be fulfilled, if the contacts are deposited with the laser ablation technique [30]. We have prepared such samples and have observed stepwise $\sigma(T)$ curves for them [see Fig. 2(b)]. From the temperature distribution of the steps we have restored the $q'(T)$ dependence [36,37], which appeared in agreement with the diffraction experiment [28]. An *upwards*-directed step in σ corresponds with a new CDW period entrance, i.e., *increase* in q' , and vice versa [29,34,36,37], as it could be expected for the p -type conductivity of quasiparticles [38].

For studies of $\sigma(\varepsilon)$ curves we constructed a setup allowing closely continuous change of the sample length with high accuracy [39]. Uniaxial stretch is achieved by means of bending an epoxy-based substrate. The ends of the substrate are resting on two bearings. The technique has been applied to the samples with the distance between contacts $L \approx 10\text{--}1000 \mu\text{m}$

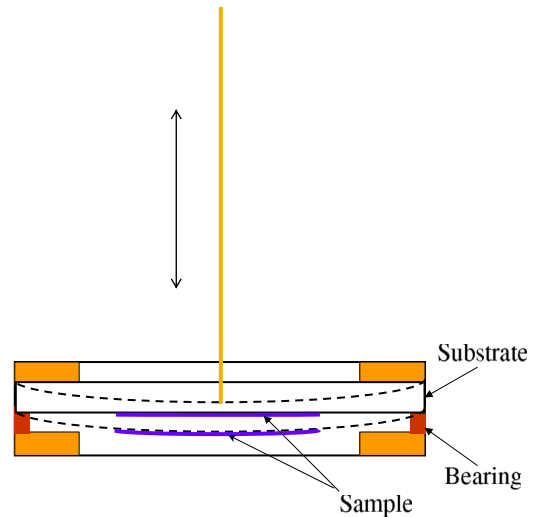


FIG. 1. A sketch of the substrate fixed over bearings (at the edges). Bending deformation is created by a bar. The bent substrate is drawn with broken lines. The sample is placed on the bottom (convex) surface of the substrate.

and thickness, $t \approx 10^{-3}\text{--}5 \mu\text{m}$. The sample is attached tightly to the substrate surface (Fig. 1). The bending is provided by a bar pushing the substrate from the opposite side. The bar is driven from outside the cryostat by means of a mechanical motion transducer. A micrometer screw driven with hands or with an electric motor allows vertical displacement of the bar. The resulting strain is

$$\varepsilon = 4\delta y d / L_{\text{sub}}^2, \quad (1)$$

where δy is the displacement of the bar, d the thickness, and L_{sub} the length of the substrate. Relation (1) implies the condition $t \ll d$; with $d = 0.4\text{--}0.5 \text{ mm}$ it is always fulfilled with a good reserve. With $\delta y < 0.6 \text{ mm}$ and $L_{\text{sub}} = 8 \text{ mm}$ the condition $\delta y \ll L_{\text{sub}}$ is also fulfilled. A thin (200–300 Å) gold film deposited on the substrate near the sample plays the role of a strain gauge. The strain-resistance coefficient (“gauge factor”) of the film was calibrated based on the relation (1). The gauge could resolve $\delta\varepsilon$ well below 10^{-4} . The maximum values of ε achieved 1%–1.5% and were usually limited by the substrate cracking.

A sample cross-section area was estimated based on its room-temperature resistance and resistivity $3 \times 10^{-4} \Omega \text{ cm}$, but was also controlled with the help of rf interference (Shapiro steps) [39,40]. For the case of TaS₃ the ratio of the CDW current density at the first step to the irradiation frequency, j_c/f , was taken to be $69 \text{ A}/(\text{MHz cm}^2)$. The control was especially important for the thinnest samples, whose resistivity can be larger than of the normal-sized ones [41].

III. RESULTS

Figure 2(a) shows a repeatedly recorded $\sigma(\varepsilon)$ dependence obtained for a nanosized TaS₃ sample. In this experiment an electric motor was used to provide smooth rotation of the micrometer screw and, consequently, gradual enough bar displacement. The dependence has an appearance of a hysteresis loop and looks very similar to the $\sigma(T)$ loops for

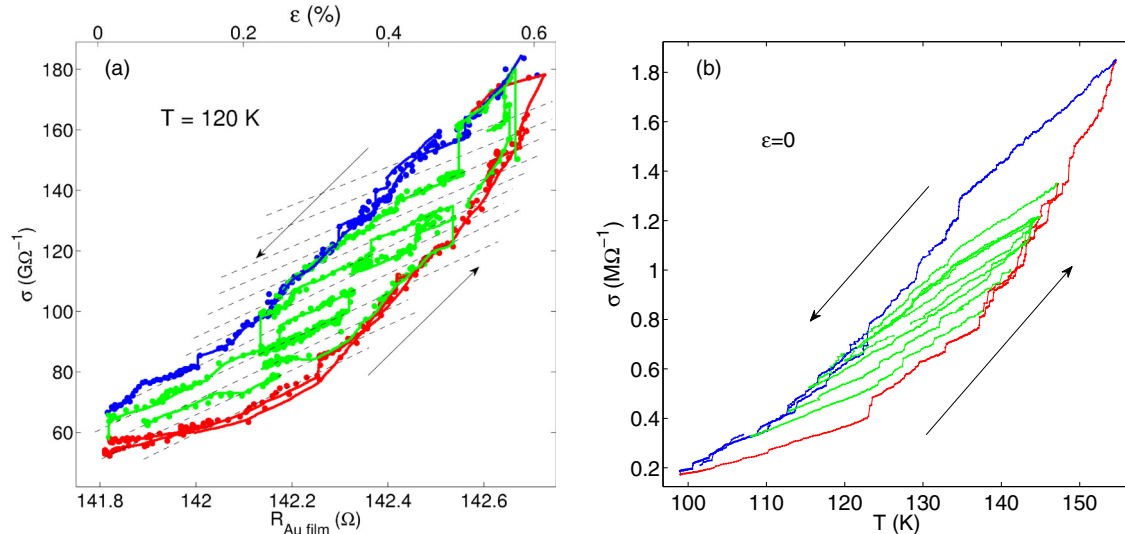


FIG. 2. (a) Repeatedly recorded $\sigma(\varepsilon)$ dependences for a TaS₃ nanosample with dimensions $23 \mu\text{m} \times (2 \times 10^{-3}) \mu\text{m}^2$. The bottom scale shows the resistance of the strain gauge. The upper scale shows the approximate strain. The solid lines are guides for the eye. (b) Similar dependences vs T for a nanosample with dimensions $24 \mu\text{m} \times (3.6 \times 10^{-3}) \mu\text{m}^2$.

nanosized samples [Fig. 2(b)]. One can distinguish discrete conducting states corresponding to $q' = \text{const}$, which are approximately shown with broken lines in Fig. 2(a). They are separated by $\delta\sigma_0 \approx 7 \text{ G}\Omega^{-1}$. Within these states $\sigma(\varepsilon)$ is reversible in ε ; note that though q' is constant, the CDW strain changes with ε [42]. Some steps can be seen not only at the edges, but also close to the center of the loops. This means that the mechanical or electric impacts sometimes perturb the CDW metastable states.

The transitions between the states are stepwise. Growth of ε gives rise to upward-directed steps of σ . From this we can conclude that q' grows with ε , in contrast with the assumption made in Refs. [3–6].

The change of ε by about 0.25% results in ten steps of σ , i.e., in $\delta q'/q' = 10\lambda/L = 5.6 \times 10^{-4}$. From this we find $g = -0.22$. This value of g appears about 30 times below the value suggested earlier [14] and is of the opposite sign. As an illustration, 1% strain results in a decrease of λ/c by 0.2%. This only drives the CDW away from fourfold commensurability. Given this $q'(\varepsilon)$ dependence one cannot attribute the anomalies in $\sigma(\varepsilon)$ to a lock-in transition, at least, if fourfold commensurability is implied.

A similar conclusion can be made from studies of the $\sigma(\varepsilon)$ hysteresis loops for macroscopic TaS₃ samples. Qualitative analysis of the loops and the conclusion about the direction of q' change with ε has been made in Ref. [39].

Figure 3 shows a typical $\sigma(\varepsilon)$ dependence for a relatively large sample. Alike loops were reported earlier, but they were not discussed [6,43]. Evidently, the hysteresis can be attributed to q' falling behind its equilibrium value with ε change. When ε increases, σ is below its equilibrium value (Fig. 3), and the relaxation of the CDW strain must result in an increase of σ . This was seen experimentally, after applying a pulse of $E > E_t$. From this one can conclude once again, that q' grows under the sample stretch.

For a quantitative estimate of $q'(\varepsilon)$ we are applying an analysis of the $\sigma(\varepsilon)$ dependence very similar with that of the

$\sigma(T)$ hysteresis loop in TaS₃ [26]. To take into account the metastability, we present conductivity as a function of two variables, ε and q' . The slope of line 2 corresponds to the equilibrium change of q' with ε . In this case one can distinguish two terms contributing to σ change:

$$d\sigma/d\varepsilon = \partial\sigma/\partial\varepsilon|_{q'} + \partial\sigma/\partial q'|_{\varepsilon} dq'/d\varepsilon. \quad (2)$$

The first term is directly associated with ε change (at $q' = \text{const}$). The second one is to be attributed to $\delta q'$ induced by $\delta\varepsilon$. After reversal of straining from decrease to increase (or vice versa) the slope of the $\sigma(\varepsilon)$ curve reduces several times (compare the slopes of the straight lines 1 and 2 in Fig. 3). Slope

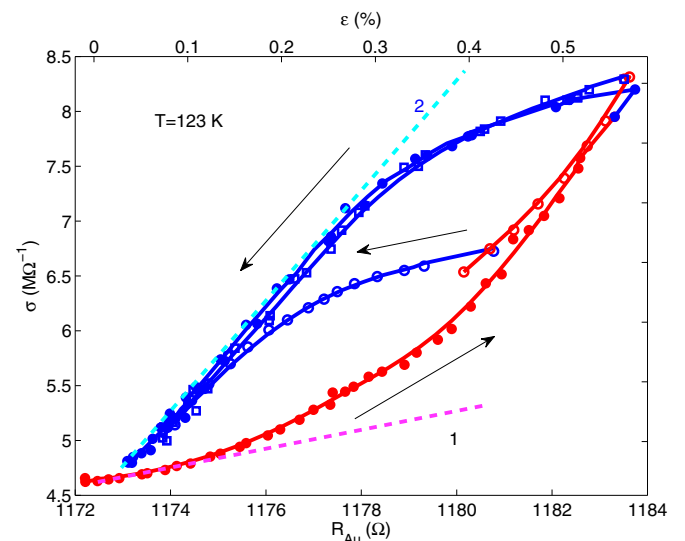


FIG. 3. Repeatedly recorded dependence of conductivity for a TaS₃ sample on strain. The bottom scale shows the resistance of the strain gauge. The upper scale shows the approximate strain. Different markers correspond to different sweeps. The solid lines are guides for the eye. The sample length is $250 \mu\text{m}$; cross-section area is $0.4 \mu\text{m}^2$.

of line 1 corresponds to the condition $q' = \text{const}$ (no PS); i.e., only the first term in (2) contributes to $\delta\sigma$. This minor part of σ growth with ε could be attributed to T_p lowering under stretch [44] through a reduction of the Peierls gap. The major part of the equilibrium σ change (60%–80% of it) appears to be coupled with $q'(\varepsilon)$ dependence. Neglecting $\partial\sigma/\partial\varepsilon|_{q'}$ one can write for the slope of line 2

$$d\sigma/d\varepsilon \approx \partial\sigma/\partial q'|_{\varepsilon} dq'/d\varepsilon, \quad (3)$$

or, in normalized units,

$$(d\sigma/\sigma)/d\varepsilon \approx (\partial\sigma/\sigma)/(\partial q'/q')|_{\varepsilon} (dq'/q')/d\varepsilon. \quad (3')$$

Relation (3') allows an estimate of $(dq'/q')/d\varepsilon$. From Fig. 2(a) we can find $(\partial\sigma/\sigma)/(\partial q'/q')|_{\varepsilon}$ from the height of the steps, as $(\delta\sigma_0/\sigma)/(\lambda/L)$, which makes about 1200. One can also roughly estimate this ratio without referring to the nanosized samples, based on the semiconductor model [26]. A simple way is to present $(\partial\sigma/\sigma)/(\partial q'/q')$ as $(\mu/\mu_{300})(\sigma_{300}/\sigma)$ [29,34]. Taking $(\mu/\mu_{300}) = 5$ [38], $(\sigma_{300}/\sigma) = 120$ we obtain a close value, about 600. From Fig. 3 we find $(d\sigma/\sigma)/d\varepsilon \approx 200$. With the given estimates of $(\partial\sigma/\sigma)/(\partial q'/q')|_{\varepsilon}$, g appears in the range 0.17–0.33, in agreement with the value obtained for the nanosized sample.

Qualitatively similar $\sigma(\varepsilon)$ hysteresis loops were obtained in the temperature range 100–140 K. This means that, at least, the sign of g is the same for all the temperatures in the range.

A more detailed analysis of the $\sigma(\varepsilon)$ curves for TaS₃ is presented in Refs. [36,37]. We have also observed $\sigma(\varepsilon)$ hysteresis for K_{0.3}MoO₃ [36]. A similar treatment of the hysteresis loop gives a $q'(\varepsilon)$ dependence for K_{0.3}MoO₃ with $g \approx -0.4$. As we mentioned above, no drop of Young's modulus at the CDW depinning was detected for K_{0.3}MoO₃ [21]. As an explanation, it had been assumed that, in contrast to trichalcogenides [22,23], q' in K_{0.3}MoO₃ is independent of ε [12,24]. In other words, it had been suggested that for the case of K_{0.3}MoO₃, $g = 0$.

This contradiction with our experiment [36] means that another interpretation for the behavior of elasticity in K_{0.3}MoO₃ [21] needs to be proposed.

IV. DISCUSSION

The central result of this paper is the effect of uniaxial strain to the CDW wavelength in TaS₃. The q' increase with a sample stretch appears unexpected, but does not contradict the previous results in the sense that there had been no attempts so far to find the $q'(\varepsilon)$ dependence. Though the techniques proposed here to reveal the q' change are not direct, as diffraction techniques could be, the result is quite reliable. The relation between the σ and q' jumps is in agreement with the widely accepted semiconductor model of the low-field conductivity of a CDW system [26,29]: Entrance of a CDW period ($\delta q' > 0$) denotes creation of two extra condensed electrons on each chain at the expense of the quasiparticles' charge. Because the low-field conductivity of TaS₃ is p type [38], the growth of positive charge results in growth of σ . The relation between $\delta\sigma$ and $\delta q'$ can be checked also without involving particular models of conductivity. Once the steps and hysteresis in σ are attributed to those in q' , the *negative*

sign of $d\sigma/dq'$ would be in contradiction with the $q'(T)$ dependence [28]. Alternatively, the estimated *positive* values of $d\sigma/dq'$ allow recovery of the $q'(T)$ dependences [26,30,36], whose quantitative agreement with the diffraction data is especially obvious for the case of K_{0.3}MoO₃ [26,30].

On the basis of our results, we can propose the following qualitative picture of the CDW transformations with stretch increase. For $\varepsilon < \varepsilon_c$, q' grows slightly. The change of q' explains the major part of $\sigma(\varepsilon)$ dependence, as well as its hysteresis. The q' increase goes until a sudden change of the q -vector at $\varepsilon = \varepsilon_c$. This means that at $\varepsilon > \varepsilon_c$ a new CDW state forms. It shows extremely high coherence both in the sliding state [36,37,39] and at rest [36,37]. Onset of the new CDW is also supported by an increase in T_p and sharpening of the Peierls transition, when ε increases above ε_c [36,37].

Let us consider the possible effects of strain on the CDW. Stretching a quasi-one-dimensional sample makes it more 3D: Both the increase of the lattice constant along the chains and decrease in the transversal directions due to Poisson contraction reduce the anisotropy of structure. An obvious consequence of this is the reduction of 1D fluctuations. The $\sigma(T)$ dependences above T_p for stretched TaS₃ samples confirm this conclusion: The metallic behavior, i.e., the negative derivative $d\sigma/dT$ becomes more pronounced with strain [36,37]. Another consequence of the anisotropy reduction concerns Fermi surfaces: Their nesting becomes worse. This effect governs the initial reduction of T_p in the stretched samples [36,37,44]. It is natural to couple the $q'(\varepsilon)$ for small ε values with nesting as well. Assuming that for a perfect 1D case $q' = 0.25$, one can attribute the deviation of q' from 0.25 with the imperfect nesting. For unstretched samples, $q' > 0.25$ [28]. Then stretching will result in a further growth of q' deflection from 0.25 with ε increase. Thus, the obtained $q'(\varepsilon)$ dependence is likely to reflect the nesting modifications under strain. Also, the q' value can be affected by a charge transfer between the valence and conducting bands due to S atoms coupling-decoupling under strain [22–24]. However, we consider this contribution to $q'(\varepsilon)$ to be less probable, taking into account a similar behavior of $q'(\varepsilon)$ in K_{0.3}MoO₃ [36], where the bonds are ionic and charge transfer is unlikely.

The next question to be discussed is the origin of the anomalies at ε_c [3–6,39]. Stretching a TaS₃ sample up to $\varepsilon \approx 0.5\%$ – 1% results in a two to six times growth of σ , but then the growth saturates, and at ε_c the conductivity shows a drop [3,39,44], which is rather sharp for the high-quality samples [39]. To explain the anomaly at the critical strain one can suppose that at $\varepsilon = \varepsilon_c$ a different q -vector (mapping the corrugated Fermi surfaces) becomes more effective in gapping electrons [36,37,39]. A first-order transition of the CDW into a different phase was suggested in Ref. [13], where a feature in the stress-strain dependence, namely, a drop of Young's modulus, has been observed at $\varepsilon = \varepsilon_c$. The authors noted strongly different properties of the new CDW state forming above ε_c , which could scarcely be explained in terms of a lock-in transition [13]. One can suppose that formation of the new CDW [39] is accompanied with a gain of electronic energy. This could explain the drop of Young's modulus of TaS₃ around ε_c [13]. The nearly steplike reduction of σ vs ε [39] can indicate an increase of the Peierls gap, Δ [44]. Alternatively, it can be explained with a decrease of

the electron-hole misbalance, i.e., the difference of hole and electron concentrations, $p-n$, in the new CDW.

An additional reason for the formation of the new CDW can be rooted in the complicated dependence of the electron energy gain on the CDW (lattice distortion) amplitude. Decrease of 1D fluctuations in a stretched sample promotes formation of a higher-amplitude CDW [44]. As it has been noted in Ref. [45], for large-amplitude CDWs the nesting is not so crucial: The electrons near $k = q/2$ would be gapped by the CDW distortion even if their energy deviates from E_F , but the deviation is less than Δ . In this case the electronic energy gain will spread over the entire Brillouin zone [45]. Formally this means that while at low Δ the energy gain is proportional to $-\Delta^2 \ln \Delta$, for high Δ it becomes proportional to Δ with a relatively large factor [45]. At $\varepsilon = \varepsilon_c$ the minimum of the total energy, i.e., the sum of the positive elastic and the negative electronic energies, can be achieved at two different values of Δ . Above ε_c the larger Δ becomes energetically favorable. One can expect that the switching of q -vector at $\varepsilon = \varepsilon_c$ will have features of a first-order transition. In a narrow ε range around ε_c coexistence of the two CDW phases has been reported [39].

Though the scenario proposed gives an idea of the dramatic changes of the CDW properties at ε_c , it does not explain the several-times drop of E_t [4,36,39] and the ultrahigh coherence of the new CDW [36,39]; the low dissipation of the new CDW is especially pronounced in the four-probe studies [36,37]. Within the weak pinning models [1] the reduction of E_t can be attributed either to a growth of the CDW elastic modulus (coupled with the Δ increase) or to a reduction in the defects concentration. Within the semiconductor model the elastic modulus of the CDW is proportional to $d\zeta/dq'$ [46], where ζ is the shift of the chemical potential of the electrons. In the unipolar approximation $d\zeta/dq'$ is proportional to $T\delta\sigma_0/\sigma$, which appears approximately the same for the unstrained and the ultracoherent CDWs at given T [36,37]. Thus, no growth of CDW elasticity happens at ε_c , which could explain the reduction of E_t up to five times. Evidently, this means that the effect of Δ increase is roughly compensated by T_P decrease. Alternatively, the transformation of the CDW properties can be attributed to a change in the defect structure of the crystal under the uniaxial stretch [39]. However, the reversibility of the basic CDW transformation at ε_c would mean the reversibility of the defects annihilation production, which does not look reasonable.

The preferable reason for the formation of the ultracoherent CDW, as we think, routes in the *intrinsic* defect structure of the CDW. The known models attribute pinning and deformations of a CDW to impurities and/or defects of the pristine lattice. However, a certain defect structure can exist in a CDW even in a perfect pure crystal, as it exists in almost any solid, even if it is perfectly pure. Then the CDW can be more perfect in a stretched sample than in an unstrained one. The studies of the room-temperature CDW in NbS₃ (phase II) with the help of high-resolution transmission electron microscopy (TEM) give evidence for such intrinsic defects, which can even affect the pristine lattice [37].

The growth of the CDW coherence under stretch can be a universal phenomenon for different CDW conductors: A similar tendency, though without features of a phase transition, was observed for NbS₃ (phase II) at room temperature [39].

Presumably in NbS₃ the changes of the CDW defect structure go gradually with ε , while in TaS₃, through a steplike transition [39].

As we mentioned in the beginning, the $q'(\varepsilon)$ dependence is supposed to be the key for understanding the Young's modulus drop on the CDW depinning and of the metastable length states of TaS₃. We first consider the $L(T)$ hysteresis [14]. It was found that in the overcooled state the TaS₃ samples are shorter than in the overheated one. Therefore, the hysteresis in $L(T)$ looks somewhat unusual: The length seems to "outrun" its equilibrium value, L_{eq} . This is not surprising in itself: The length change results from superposition of regular thermal expansion and the effect of CDW strain on top of it. In Ref. [14] the deviation of L from L_{eq} , δL , was attributed to the impact of longitudinal CDW strain, i.e., to a metastable value of q' . Within this model $\delta L/L \approx -g(Y_c/Y_L)(\delta q'/q')$, where $\delta q'$ is the deviation of q' from equilibrium; Y_c/Y_L is the ratio of the Young's moduli of the CDW and the lattice (Eq. (3) from Ref. [14]). With heating q' increases [28], so, in view of the $q'(T)$ hysteresis, for the overheated state $\delta q'$ must be negative. For $g > 0$ the positive value of δL is obtained, in agreement with the experiment. The negative sign of g breaks this agreement: With $g < 0$ it looks as if L under CDW strain readjusts itself so that the CDW longitudinal strain grows [47], in an apparent contradiction with Le Chatelier's principle [48]. From this one can conclude that the simple explanation of length hysteresis does not work.

The most reasonable way to overcome this contradiction is to go beyond the 1D model of CDW-lattice interaction. In Ref. [28] the b^* component of the q -vector was found to be temperature dependent, as well as the c^* component. One can suppose that metastability in sample dimensions is driven by the CDW-lattice interaction in the directions normal to the chains. Then the length change is to be calculated with the help of the Poisson coefficient. It is known that transverse CDW strains exceed the longitudinal ones (see, e.g., [49]). For K_{0.3}MoO₃ it has been found that CDW formation at T_P has the largest effect on the dimensions in the [102] direction, perpendicular to the chains' direction [50]. It is also clear that the enormous voltage-induced torsional strain observed in TaS₃ [19] cannot be reduced to longitudinal CDW strains.

Unlike the metastable length states, metastable states in conductivity can be described in terms of longitudinal CDW strains. This point of view is justified, e.g., by the distribution of $\delta\sigma$ along the sample after application of electric field [51], which appears consistent with the idea of q' increase near one contact and decrease near the other one. The semiconductor model describing correlation of $p-n$ and $\delta q'$ [26,29,34] also looks consistent: The correspondence of the steps of conductivity with the production/annihilation of a CDW period [29,30,34], the reasonable values of mobility found from the conductivity steps [30,31,36,37], and the recovered $q'(T)$ dependences [26,30,36,37] confirm the correlation between $\delta q'$ and $\delta\sigma$. If this is the case, one can expect violation of scaling between metastabilities in length and in conductivity. This expectation is confirmed by Ref. [15], in which metastable length states induced by electric field were reported. A correlation between the $L(E)$ and $\sigma(E)$ hysteresis loops was observed. However, in contrast with Ref. [14], larger L corresponded to higher σ . It was also noticed [15]

that the scaling between L and σ was rough; particularly, substantial length changes below E_t were not accompanied by any changes in σ .

Once the transversal components of the CDW strain dominate metastability in length, they should also be taken into account in the treatment of the elastic anomalies in Young's modulus. Particularly, in the model [27] the coefficient g should be considered as a tensor. One more relevant complication can be connected with the possible coexistence of commensurate and incommensurate CDWs [52].

In summary, we have observed steps of conductivity measured as a function of strain, ε , for nanosized TaS₃ samples. The steps are associated with "quantization" of the CDW wave vector. The "quantization" has been employed for detection of fine rearrangement of the CDW structure. In particular, it is found that sample expansion results in a growth of q/c^* , which means moving of q away from the fourfold commensurability, $q = 0.25c^*$. The parameter g characterizing q/c^* variation with ε [27] is found to be about -0.2 . This value is of the opposite sign and about 30 times smaller in absolute value than was suggested earlier. A similar value of g follows from the analysis of the $q(\varepsilon)$ hysteresis loops in macroscopic

TaS₃ samples. This result necessitates an alternative treatment for strained-induced anomalies and other mechanical effects found for TaS₃.

The result appears peculiar not only to TaS₃. For example, the analysis of the $\sigma(\varepsilon)$ hysteresis loop in K_{0.3}MoO₃ [36,37] also grants an unexpected $q(\varepsilon)$ dependence [21–23]. Evidently, to solve the contradictions with the previous concepts, one should take into account the transverse components of the CDW and lattice strains.

ACKNOWLEDGMENTS

We are grateful to R. E. Thorne for donating the high-quality TaS₃ samples, to A. A. Sinchenko for the help with the experiment, and to I. G. Gorlova and S. V. Zaitsev-Zotov for fruitful discussions. The support of RFBR (Grants No. 14-02-01240, No. 14-02-92015, No. 14-02-01236, No. 16-02-01095, and No. 14-02-31710) and the program "New materials and structures" of RAS are acknowledged. The elaboration of the "bending technique" for bulk samples, allowing both uniaxial expansion and contraction, was supported by the Russian Scientific Foundation (Grant No. 14-19-01644).

-
- [1] P. Monceau, *Adv. Phys.* **61**, 325 (2012).
- [2] J. W. Brill in *Handbook of Elastic Properties of Solids, Liquids, and Gases, Vol. II. Elastic Properties of Solids: Theory, Elements and Compounds, Novel Materials, Alloys, and Building Materials*, edited by M. Levy and L. Furr (Academic Press, San Diego, 2001), Chap. 10, p. 143.
- [3] V. B. Preobrazhensky, A. N. Taldenkov, and I. Yu. Kal'nova, *Pis'ma Zh. Eksp. Teor. Fiz.* **40**, 183 (1984) [*JETP Lett.* **40**, 944 (1984)].
- [4] V. B. Preobrazhensky, A. N. Taldenkov, and S. Yu. Shabanov, *Solid State Commun.* **54**, 399 (1985).
- [5] T. A. Davis, W. Schaffer, M. J. Skove, and E. P. Stillwell, *Phys. Rev. B* **39**, 10094 (1989).
- [6] Z. G. Xu and J. W. Brill, *Phys. Rev. B* **43**, 11037 (1991).
- [7] J. W. Brill and W. Roark, *Phys. Rev. Lett.* **53**, 846 (1984).
- [8] L. C. Bourne, M. S. Sherwin, and A. Zettl, *Phys. Rev. Lett.* **56**, 1952 (1986).
- [9] Z. G. Xu and J. W. Brill, *Phys. Rev. B* **45**, 3953 (1992).
- [10] R. L. Jacobsen, M. B. Weissman, and G. Mozurkewich, *Phys. Rev. B* **43**, 13198 (1991).
- [11] X.-D. Xiang and J. W. Brill, *Phys. Rev. B* **39**, 1290 (1989).
- [12] X.-D. Xiang and J. W. Brill, *Phys. Rev. B* **36**, 2969(R) (1987).
- [13] K. Das, M. Chung, M. J. Skove, and G. X. Tessema, *Phys. Rev. B* **52**, 7915 (1995).
- [14] A. V. Golovnya, V. Ya. Pokrovskii, and P. M. Shadrin, *Phys. Rev. Lett.* **88**, 246401 (2002).
- [15] S. Hoen, B. Burk, A. Zettl, and M. Inui, *Phys. Rev. B* **46**, 1874 (1992).
- [16] V. Ya. Pokrovskii, S. G. Zybtssev, and I. G. Gorlova, *Phys. Rev. Lett.* **98**, 206404 (2007).
- [17] V. Ya. Pokrovskii, S. G. Zybtssev, V. B. Loginov, V. N. Timofeev, D. V. Kolesov, I. V. Yaminsky, and I. G. Gorlova, *Physica B* **404**, 437 (2009).
- [18] S. G. Zybtssev, M. V. Nikitin, and V. Ya. Pokrovskii, *Pis'ma Zh. Eksp. Teor. Fiz.* **92**, 448 (2010) [*JETP Lett.* **92**, 405 (2010)].
- [19] V. Ya. Pokrovskii, S. G. Zybtssev, M. V. Nikitin, I. G. Gorlova, V. F. Nasretdinova, and S. V. Zaitsev-Zotov, *Phys.-Usp.* **56**, 29 (2013).
- [20] G. Mozurkewich, P. M. Chaikin, W. G. Clark, and G. Gruner, *Solid State Commun.* **56**, 421 (1985).
- [21] L. C. Bourne and A. Zettl, *Solid State Commun.* **60**, 789 (1986).
- [22] A. Meerschaut, *J. Phys. (Paris)* **44**, C3-1615 (1983).
- [23] A. Meerschaut and J. Rouxel, in *Crystal Chemistry and Properties of Materials with Quasi-One-Dimensional Structures*, edited by J. Rouxel (D. Reidel Publishing Company, Dordrecht, 1986), pp. 205–279.
- [24] V. Ya. Pokrovskii, *Pis'ma Zh. Eksp. Teor. Fiz.* **86**, 290 (2007) [*JETP Lett.* **86**, 260 (2007)].
- [25] C. Noguera and J.-P. Pouget, *J. Phys. I (Paris)* **1**, 1035 (1991).
- [26] S. N. Artemenko, V. Ya. Pokrovskii, and S. V. Zaitsev-Zotov, *Zh. Eksp. Teor. Fiz.* **110**, 1069 (1996) [*JETP* **83**, 590 (1996)].
- [27] G. Mozurkewich, *Phys. Rev. B* **42**, 11183 (1990).
- [28] C. Roucau, *J. Phys. (Paris)* **C3 44**, 1725 (1983); Z. Z. Wang, H. Salva, P. Monceau, M. Renard, C. Roucau, R. Ayroles, F. Levy, L. Guemas, and A. Meerschaut, *J. Phys. (Paris), Lett.* **44**, L315 (1983).
- [29] D. V. Borodin, S. V. Zaitsev-Zotov, and F. Ya. Nad', *Zh. Eksp. Teor. Fiz.* **93**, 1394 (1987) [*Sov. Phys. JETP* **66**, 793 (1987)].
- [30] S. G. Zybtssev, V. Ya. Pokrovskii, and S. V. Zaitsev-Zotov, *Nat. Commun.* **1**, 85 (2010).
- [31] S. G. Zybtssev and V. Ya. Pokrovskii, *Phys. Rev. B* **84**, 085139 (2011).

- [32] R. Jaramillo, T. F. Rosenbaum, E. D. Isaacs, O. G. Shpyrko, P. G. Evans, G. Aeppli, and Z. Cai, *Phys. Rev. Lett.* **98**, 117206 (2007); R. K. Kummamuru and Y.-A. Soh, *Nature* **452**, 859 (2008).
- [33] S. Girault, A. H. Moudden, J. P. Pouget, and J. M. Godard, *Phys. Rev. B* **38**, 7980 (1988); J. P. Pouget, S. Girault, A. H. Moudden, B. Hennion, C. Escribe-Filippini, and M. Sato, *Phys. Scr.* **T25**, 58 (1989).
- [34] D. V. Borodin, S. V. Zaitsev-Zotov, and F. Ya. Nad, *Pis'ma Zh. Eksp. Teor. Fiz.* **43**, 485 (1986) [*JETP Lett.* **43**, 625 (1986)].
- [35] If the CDW phase at the contacts is not fixed, some phase slips can perturb sample volume from both sides of a contact. Then the resistivity changes both within the studied sample segment and beyond it. In this case fractional steps in $\sigma(T)$ are expected, and counting the number of steps can give only an approximate value of q' change.
- [36] S. G. Zytsev and V. Ya. Pokrovskii, [arXiv:1602.08123](https://arxiv.org/abs/1602.08123) [cond-mat.str-el].
- [37] S. G. Zytsev, V. Ya. Pokrovskii *et al.* (unpublished).
- [38] Yu. I. Latyshev, Ya. S. Savitskaya, and V. V. Frolov, *Pis'ma Zh. Eksp. Teor. Fiz.* **38**, 446 (1983) [*JETP Lett.* **38**, 541 (1983)].
- [39] S. G. Zytsev and V. Ya. Pokrovskii, *Physica B* **460**, 34 (2015).
- [40] E. Slot, M. A. Holst, H. S. J. van der Zant, and S. V. Zaitsev-Zotov, *Phys. Rev. Lett.* **93**, 176602 (2004).
- [41] S. V. Zaitsev-Zotov, V. Ya. Pokrovskii, and P. Monceau, *Pis'ma Zh. Eksp. Teor. Fiz.* **73**, 29 (2001) [*JETP Lett.* **73**, 25 (2001)].
- [42] The strain is proportional to the deviation of q' from its equilibrium value. Therefore, it can change even if $q' = \text{const}$, but the equilibrium q' value changes (with T or with ε).
- [43] A. N. Taldenkov (unpublished).
- [44] R. S. Lear, M. J. Skove, E. P. Stillwell, and J. W. Brill, *Phys. Rev. B* **29**, 5656 (1984).
- [45] K. Rossnagel, *J. Phys.: Condens. Matter* **23**, 213001 (2011).
- [46] V. Ya. Pokrovskii and S. V. Zaitsev-Zotov, *Synth. Met.* **32**, 321 (1989).
- [47] To check this, let us again consider the overheated state, for which $q' < q'_{\text{eq}}$, where q'_{eq} is the equilibrium value of q' . According to [14], $\delta L > 0$, and, as we know now, q'_{eq} is larger than at $\delta L = 0$. Thus, while q' appears below q'_{eq} , the length changes so that q'_{eq} increases; i.e., the difference $|q' - q'_{\text{eq}}|$ grows.
- [48] When a system at equilibrium is subjected to change in concentration, temperature, volume, or pressure, then the system readjusts itself to counteract the effect of the applied change and a new equilibrium is established.
- [49] K. Tsutsumi, T. Tamegai, S. Kagoshima, and M. Sato, Charge density waves in solids, *Lect. Notes Phys.* **217**, 17 (1985).
- [50] M. R. Hauser, B. B. Plapp, and G. Mozurkewich, *Phys. Rev. B* **43**, 8105 (1991).
- [51] S. E. Brown, L. Mihály, and G. Grüner, *Solid State Commun.* **58**, 231 (1986).
- [52] K. Inagaki, M. Tsubota, K. Higashiyama, K. Ichimura, S. Tanda, K. Yamamoto, N. Hanasaki, N. Ikeda, Y. Nogami, T. Ito, and H. Toyokawa, *J. Phys. Soc. Jpn.* **77**, 093708 (2008).