We review the features of the charge density wave (CDW) conductor NbS$_3$ (phase II) and include several additional results from transport, compositional, and structural studies. Particularly, we highlight three central results: (1) In addition to the previously reported CDW transitions at $T_{P1} = 360$ K and $T_{P2} = 150$ K, a third CDW transition occurs at a much higher temperature $T_0 \approx 620 – 650$ K; evidence for the nonlinear conductivity of this CDW is presented. (2) We show that the CDW associated with the $T_{P2}$ transition arises from S vacancies acting as donors. Such a CDW transition has not been observed before. (3) We demonstrate the exceptional coherence of the $T_{P1}$ CDW at room temperature. The effects of uniaxial strain on the CDW transition temperature and transport are reported.

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I. INTRODUCTION

Since Peierls transitions, at which electrons condense into charge density waves (CDWs), usually occur well below room temperature (RT), studies of CDWs in quasi-one-dimensional (1D) conductors have been usually considered a branch of low-temperature (LT) physics [1,2]. The formation of a CDW is accompanied by dielectrization (i.e., gapping) of the electronic spectrum with a corresponding drop in the electrical conductivity. The periodic lattice distortion accompanying the CDW can be studied with diffraction techniques in momentum space and scanning tunneling microscopy in real space. A notable feature of quasi-1D CDWs is their ability to slide in a sufficiently high electric field, resulting in nonlinear conductivity. This sliding is accompanied by the generation of narrowband and wideband noises. The quasi-1D CDWs also are featured with an enormous dielectric constant and metastable states originating from their deformability. In addition to these basic properties, a number of other aspects of CDW behavior has been investigated, including synchronization of CDW sliding with an external radio frequency (RF) field (the so-called Shapiro steps), coherence stimulation of CDW sliding by asynchronous RF irradiation [3], the effects of pressure [2] and uniaxial strain [4–7] on the Peierls transitions and CDW transport, and enormous electric-field-induced crystal deformations [2,8].

Several trichalcogenides of the group V metals ($MX_3$), namely NbSe$_3$, TaS$_3$ (orthorhombic and monoclinic), and phase II NbS$_3$ (hereafter NbS$_3$-II), constitute a family of typical quasi-1D CDW conductors [1,2]. Their crystal structures are formed of metallic chains surrounded by trigonal prismatic cages of chalcogen atoms. Though these compounds are apparently isoelectronic, their properties are rather diverse. For example, they display very different CDW wave vectors, indicating different degrees of filling in conduction electronic bands. A plausible reason for this variety may be the relative positioning of the chalcogen atoms [9]. Depending on their interatomic distances, chalcogen atoms can either be isolated from each other or form bonded pairs. Correspondingly, one valence electron from a chalcogen atom can either belong to the conduction band or to a localized bond.

The monoclinic polymorph NbS$_3$-II exhibits some fascinating features. Two CDW transitions have been reported for NbS$_3$-II. One CDW has a wave vector $q_1 = (0.5a^*,0.298b^*,0)$ [10,11] and forms at $T_{P1} = 330–370$ K [11,12], a temperature much beyond the traditional realm of LT physics. While several basic experimental results on NbS$_3$-II were published in the 1980s [1,2,10,11,13–16], little subsequent work was undertaken until 2009. This gap was largely due to difficulties with synthesizing the compound: initially, NbS$_3$-II whiskers were only found as rare inclusions accompanying the growth of the semiconducting phase of NbS$_3$ (hereafter NbS$_3$-I) [11], which was more extensively studied [1,2,17,18]. A new phase III NbS$_3$ with a phase
transition at 150 K was also reported [13], but was later suggested to be a subphase of NbS3-II [8,12,19]. The synthesis conditions of NbS3-II were established from studies in the Kotel’nikov Institute of Radioengineering and Electronics of Russian Academy of Sciences (RAS) in 2009 [8] and were successfully reproduced in the National Taiwan University. A detailed description of NbS3-II growth conditions is presented in Ref. [8].

The most notable feature of NbS3-II is its nonlinear transport at RT [1,2,11,12,19], associated with the CDW formed at $T_{P1}$. The RT CDW shows exceptionally high transport velocities and sliding coherence. The corresponding fundamental frequencies $f_r$, as revealed by the RF interference technique [12,19], exceed 15 GHz. The coherence of this CDW can be further improved by external asynchronous RF irradiation [3] and by uniaxial strain [20].

The Peierls transition at $T_{P1}$ is clearly detected in transport and structural studies: a pronounced increase of resistance $R$ with decreasing temperature is observed near $T_{P1}$, while its I-V curve becomes nearly linear above 340 K. The intensities of the $q_1$ satellites decrease significantly above 350 K [11,21], while the satellites of the second modulation wave vector $q_0 = (0.5a^*, 0.552b^*, 0)$ remain detectable to at least 450 K.

Within NbS3-II, low-Ohmic and high-Ohmic subphases [8,19,22] have been identified. As described previously [8], the low-Ohmic samples are prepared at 670–700 °C, and the high-Ohmic ones at 715–740 °C. Both subphases can be grown during the same run if a temperature gradient is present within the synthesis ampoule. Electron diffraction reveals a doubling of the lattice constant along the $a$ axis ($\sim$9.65 Å) for the high-Ohmic crystals, which is absent in the low-Ohmic samples [19]. In addition to the CDW transition at $T_{P1}$, the low-Ohmic samples undergo a further CDW transition at $T_{P2} = 150$ K [13] as detected in the temperature-dependent resistance $R(T)$ curves [8,12,19]. Below $T_{P2}$ nonlinear conduction with a pronounced threshold field $E_t$ is observed. This indicates a charge transport coupled with this LT CDW. The presence of Shapiro steps definitively reveals sliding of the LT CDW [12,19]. However, RF synchronization studies reveal a surprisingly low charge density of this LT CDW. The so-called “fundamental ratio” $j_f/j_s$ appears very low and is sample dependent [12,19] (here $j_s$ is the CDW current density at the first Shapiro step). Attempts to find structural changes below $T_{P2}$ using electron [10] and x-ray diffraction techniques were unsuccessful.

The LT CDW state emerges from a dielectrized state following the two CDWs formed at higher temperatures and remains a rather enigmatic charge-ordered state, which might be considered as a generalization of the usual CDW. The emergence of a new CDW in this rather resistive state is unusual, and a Keldysh-Kopaev transition [23] (the formation of an excitonic dielectric [24]) has been proposed as a possible mechanism [19].

In this paper, we present a number of new experimental results for NbS3-II. Section II focuses on the features of the RT CDW. We report unprecedentedly high values of the CDW fundamental frequencies, as revealed by Shapiro steps. A high coherence of the RT CDW sliding is shown by the nearly complete CDW synchronization under RF power and by Bessel-type oscillations of the Shapiro steps’ width vs RF power. In addition, we demonstrate that the coherence can be further improved by applying strain $\varepsilon$ parallel to the chains, i.e., along the $b$ axis. The strain also strongly affects the CDW transition temperature $T_{P1}$, which decreases by approximately 80 K for $\varepsilon \approx 1.5\%$. In Sec. III, we report transport measurements at temperatures up to about 650 K. A new feature in $R(T)$ is found near 620–650 K and is attributed to the onset of an ultrahigh temperature (UHT) CDW. The evidence of nonlinear transport provided by the UHT CDW will be given. We also demonstrate that, on heating above $\sim$800 K, the high-Ohmic subphase transforms gradually into the low-Ohmic subphase and further into a metallic-like compound. Section IV focuses on the LT CDW. Unlike the transition at $T_{P1}$, the LT CDW transition appears nearly insensitive to strain, as is the nonlinear conduction associated with this LT CDW. Electron-probe microanalyses (EPMA) reveal a shortage of S in the low-Ohmic samples. This suggests a coupling between the relatively high conductivity of the low-Ohmic samples and the presence of S vacancies. Though electron diffraction patterns show no changes below 150 K, the $T_{P2}$ transition is detected by scanning transmission electron microscopy-based electron energy loss spectroscopy (STEM-EELS) and x-ray absorption near edge spectroscopy (XANES). These techniques indicate charge transfers between states coupled with S and Nb atoms, as well as with S vacancies. Nuclear magnetic resonance (NMR) studies suggest “freezing” of the condensed electronic state near $T_{P2}$. In Sec. V, we present an overview of the results and discuss various possible origins of the LT CDW transition. The Keldysh-Kopaev (excitonic insulator) transition [23,24] appears more consistent with the experimental data than other mechanisms of electronic condensation. In the concluding Sec. VI, we summarize the main features of the CDW states of NbS3-II.

II. RT CDW: EFFECTS OF RF IRRADIATION AND UNIAXIAL STRAIN

The RT CDW in NbS3-II is remarkable not only for the occurrence of sliding, but also for its extremely high sliding co-
Large area per CDW chain, harmonic) are observed at frequencies as high as 20 GHz. The highest fundamental frequencies are obtained in these samples. As Fig. 1 shows, Shapiro steps (the first dimension) require special arrangement of the samples for a better synchronization of the CDW at frequencies above 20 GHz. In comparison to similar compounds, this value appears unusually low; an adaptation of the conventional model may be required in the case of NbS₃-II, like for the monoclinic phase of TaS₃ [25]. Nonetheless, the universality of this “fundamental ratio” allows a precise determination of the cross-sectional area of the samples [26]. A very high coherence of the RT CDW sliding is observed in samples with nanoscale transverse dimensions. The experimentally determined sample cross-sectional area (determined by 80–90% (Fig. 2). At a sufficiently large RF power in NbS₃-II nanodimensional samples, the RT CDW shows nearly complete synchronization; the differential conductivity σₐ of the CDW is reduced by 80–90% (Fig. 2). At the same time, the widths of the Shapiro steps show a nonmonotonic dependence on the RF power with Bessel-type oscillations. To observe such oscillations, the CDW must be reduced by the effect of RF synchronization. At a fixed RF power, the Shapiro steps are more pronounced at increased strain, becoming visible directly in the I-V curves (Fig. 5). The effect of strain appears similar to that of asynchronous RF irradiation, which also improves the coherence of the CDW sliding in a number of compounds [3,32], including NbS₃-II [3]. However, the origins of enhanced coherence in these two cases are likely different. While RF is believed to periodically draw CDW back to its starting state before it loses coherence [3], strain can change the crystal defect structure [20]. As shown in Fig. 4, the irreversible growth of coherence after strain removal may be explained by a reduction in the crystal defect density after the uniaxial deformation. For example, it is known that twin-free yttrium barium copper oxide (YBCO) crystals can be obtained by applying uniaxial pressure at 420°C in flowing oxygen [33]. However, the major width and the threshold voltage Vₜ (half-width of the “zeroth step”) on the RF power. A nonmonotonic suppression of Vₜ has been previously reported for NbSe₃ [30].

We recently developed techniques to apply controlled uniaxial strain to whiskerlike samples, including nanosized ones [20]. The NbS₃-II samples were stretched by bending epoxy-based substrates [20]. Studies of NbS₃-II at RT [20] and of orthorhombic TaS₃ [20,31] have demonstrated that strain can improve the CDW coherence. Figure 4 presents a set of curves showing the differential conductivity of NbS₃-II as a function of voltage V. Unlike the case of orthorhombic TaS₃ in which an ultracoherent CDW emerges through a phase transition at a critical strain [20,31], CDW coherence in the case of NbS₃-II grows gradually with ε (Fig. 4). The thresholds become sharper, the growth of σₐ above the thresholds occurs faster, and the value of the maximum CDW conductivity increases [20]. After strain removal, the threshold voltage decreases and the threshold becomes slightly sharper, while the value of resistance at V = 0 indicates that the sample is unstrained.

The growth of coherence with strain is also demonstrated by the effect of RF synchronization. At a fixed RF power, the Shapiro steps are more pronounced at increased strain, becoming visible directly in the I-V curves (Fig. 5). The effect of strain appears similar to that of asynchronous RF irradiation, which also improves the coherence of the CDW sliding in a number of compounds [3,32], including NbS₃-II [3]. However, the origins of enhanced coherence in these two cases are likely different. While RF is believed to periodically draw CDW back to its starting state before it loses coherence [3], strain can change the crystal defect structure [20]. As shown in Fig. 4, the irreversible growth of coherence after strain removal may be explained by a reduction in the crystal defect density after the uniaxial deformation. For example, it is known that twin-free yttrium barium copper oxide (YBCO) crystals can be obtained by applying uniaxial pressure at 420°C in flowing oxygen [33]. However, the major coherence. The “fundamental ratio” jₑ/fₑ is sample independent within the experimental error (defined by the uncertainty in the sample cross-sectional area). The experimentally determined value jₑ/fₑ = 18 A/MHz/cm² [19] corresponds to one CDW chain per unit cell when considered within the commonly applied model of CDW sliding mode conductivity [12]. In comparison to similar compounds, this value appears unusually low; an adaptation of the conventional model may be required in the case of NbS₃-II, like for the monoclinic phase of TaS₃ [25]. Nonetheless, the universality of this “fundamental ratio” allows a precise determination of the cross-sectional area of the samples [26]. A very high coherence of the RT CDW sliding is observed in samples with nanoscale transverse dimensions. The highest fundamental frequencies are obtained in these samples. As Fig. 1 shows, Shapiro steps (the first harmonic) are observed at frequencies as high as 20 GHz. The large area per CDW chain, s₀ = 2e/(jₑ/fₑ) = 180 Å² [1,27], compared to other quasi-1D CDW compounds, is consistent with the relatively small amount of Joule heating of the NbS₃-II samples. They show the highest density of CDW current before burnout [19]. Based on the highest current densities ever passed through these samples, it is suggested [19] that fundamental frequencies as high as fₑ = 200 GHz are attainable. However, synchronization of the CDW at frequencies above 20 GHz requires special arrangement of the samples for a better impedance match with RF radiation.

At a sufficiently large RF power in NbS₃-II nanodimensional samples, the RT CDW shows nearly complete synchronization: the differential conductivity σₐ of the CDW is reduced by 80–90% (Fig. 2). At the same time, the widths of the Shapiro steps show a nonmonotonic dependence on the RF power with Bessel-type oscillations. To observe such oscillations, the CDW must exhibit a high coherence. Previously, the Bessel-type oscillations were observed for NbSe₃ only [28]. The evolution of the I-V curves with increasing RF power is shown by a video clip in the Supplemental Material [29]. It shows the screen of a digital oscilloscope displaying the rapidly recorded I-V curves of a NbS₃-II sample. Figure 3 shows the normalized dependence of the first Shapiro step.

![FIG. 2. An example of σₐ vs V dependence under RF irradiation at RT. The sample dimensions are 50 μm × 0.011 μm².](image-url)
FIG. 4. The RT voltage dependencies of differential conductivity $\sigma_d$ for a sample under uniaxial strain. The strain increases from $\varepsilon = 0$ (the lowest curve) to $\varepsilon \approx 1\%$ (the uppermost curve) in approximately equal steps. The curve marked with red circles was obtained after the strain was removed. The sample length $L$ is 24 $\mu$m. Part of the coherence growth is reversible (Fig. 4). This could be attributed to an alignment of the metallic chains under strain. Increased velocities of the internal acoustic modes and their reduced friction in the strained samples can also stimulate coherence of the CDW sliding through CDW-lattice coupling [8,34].

Studying strain effects of CDW transitions is important to better understand how the CDW condensate forms. Uniaxial strain decreases the anisotropy; as the interatomic distances along the conducting chains increase, the interchain distances decrease due to the Poisson contraction. The reduction in anisotropy corrugates the Fermi surfaces and decreases $T_p$. At the same time, 1D fluctuations are suppressed, which increases $T_p$. The actual changes in $T_p$ result from the competition between these two effects. Applied uniaxial strain reduces $T_p$ in orthorhombic TaS$_3$ [5,6] (but with a tendency to increase it after exceeding a critical value of $\varepsilon$ [31]), NbSe$_3$ [5], and K$_{0.3}$MoO$_3$ [31]. In the monoclinic TaS$_3$, the lower transition is shifted downwards, while no shift was observed for the upper transition temperature for strains up to 1.5% [35].

The effects of uniaxial strain on CDW compounds have been examined less thoroughly than those of hydrostatic pressure, which is in part similar to the effect of stretching. In fact, a general feature of quasi-1D CDW compounds is also a lowering of $T_p$ under pressure, indicating that the corrugation of Fermi surfaces has a dominant influence on $T_p$ [2,36]. The only known exception is the monoclinic TaS$_3$, whose upper transition temperature increases at small pressures. It was proposed that the suppression of 1D fluctuations dominates the $T_p$ variation in this strongly anisotropic TaS$_3$ polytype.

In the case of NbS$_3$-II, strain strongly affects the RT CDW: $\varepsilon \sim 1.5\%$ results in lowering $T_{p1}$ from 360 K to below 280 K (Fig. 6). It is also obvious that strain sharpens the CDW transition. The decrease of $T_{p1}$ in NbS$_3$-II means that, in spite of the large anisotropy [37], it is primarily dominated by Fermi-surface nesting conditions and less by the suppression of the 1D fluctuations.

The reduction of $T_{p1}$ with $\varepsilon$ must be taken into account in the I-V curves under strain (Fig. 4). For larger applied strains, the $\sigma_d(V)$ dependencies were measured close to $T_{p1}$. However, the evolution of the curves with $\varepsilon$ cannot be attributed to the proximity of $T_{p1}$ only. In the absence of strain, the I-V curves are smeared at temperatures close to the CDW transition [12]. Therefore, the growth of CDW coherence observed in Fig. 4, as well as the increased sharpness of the Peierls transition at $T_{p1}$ [Fig. 6(b)], are a direct consequence of a tensile strain.

The “inverse” electromechanical effects, i.e., the impact of electrical field on the dimensions and the form of the samples, have been less studied for NbS$_3$-II. Similarly to orthorhombic TaS$_3$, K$_{0.3}$MoO$_3$, and (TaSe$_4$I, NbS$_3$-II also shows electric-field-induced torsional strain. It can be observed at RT. However, the torsional angles are relatively small and are one to two orders of magnitude smaller than those observed in TaS$_3$. The most reliable measurement of the torsional angle as a...
problems with the epoxy substrates at elevated temperatures. The torsional angle was measured optically [8]. Without RF irradiation, a threshold current for the onset of torsion was observed, like in the cases of the orthorhombic TaS₃, K₀.₂₇MoO₃, and (TaSe₄)₂I [8,38]. Radiofrequency irradiation (45 MHz) suppresses the thresholds and increases the torsional angles by an order of magnitude. This effect is in line with the coherence stimulation by an asynchronous RF irradiation [3].

The power of the RF irradiation applied at 45 MHz is sufficient to suppress the threshold down to zero, as is the case for the most of interference experiments [39] (see also Fig. 3). This is also seen in the torsional curve (Fig. 7), which has almost lost threshold features. At the same time, the torsional angles have increased by an order of magnitude. This effect seems to be in line with the coherence stimulation by an asynchronous RF irradiation [3]. In fact, coherent CDW shows larger tensile strength [31,32], i.e., larger critical strain before phase slippage. The larger CDW strain must result in a larger torsional strain of the sample.

III. THE ULTRAHIGH Tp CDW: INDICATIONS OF A PEIERLS TRANSITION AND A FRÖHLICH MODE

As mentioned in the Introduction, the diffraction patterns of NbS₃-II at 300 K show two incommensurate satellites at two CDW q vectors: \( q₀ = (0.5a^*, 0.352b^*, 0) \) and \( q₁ = (0.5a^*, 0.298b^*, 0) \) [10,11]. While the first set of satellites remain visible up to 450 K, the second set vanishes gradually above 360 K, which is close to \( Tₚ \) [11]. When heated above 450 K under vacuum or ambient conditions, NbS₃-II crystals start to degrade.

In order to extend the measuring range and prevent crystal degradation, the \( R(T) \) measurements at elevated temperatures reported herein were performed in an argon atmosphere. The temperature was monitored with a thermocouple. Heating to \( \sim 700 \) K and subsequent cooling was performed within several minutes. Figure 8 shows the \( R(T) \) curve in a wide temperature range and very clearly reveals the transitions at \( Tₚ₁ \) and \( Tₚ₂ \). The dependence at high temperatures, above \( Tₚ₁ \), was added from a different sample during a heating cycle. Above \( \sim 600 \) K, a notable degradation of sample properties begins, resulting in a growth of conductivity \( \sigma \). However, during fast cooling from \( \sim 700 \) K, the feature in the \( \sigma(T) \) reappears in the same temperature range (the dynamic error in temperature determination was tens of kelvin). Therefore the \( \sigma(T) \) feature around 620–650 K is attributed to a transition at \( Tₚ₀ \).

As shown in Fig. 9, heating to above \( \sim 800 \) K gradually transforms the high-Ohmic subphase into the low-Ohmic one. Further heating transforms NbS₃-II into a compound with metallic conductivity. As discussed below, the increased conductivity may be attributed to loss of S atoms and formation of S vacancies at elevated temperature.

Figure 10 shows a series of \( \sigma(\nu)V \) curves for a NbS₃-II sample, recorded below and above \( Tₚ₁ \). At \( T > Tₚ₁ \), a gradual growth in conductivity at fields above 0.3 kV/cm is observed (e.g. the curve at 450 K). It is difficult to separate the effects of CDW sliding and Joule heating in these curves. However, sliding of the UHT CDW can be checked by the effect of RF
irradiation on the $\sigma_d(V)$ curves [3]. This effect was studied at RT, where it was possible to place the sample sufficiently close to the RF generator output for better matching. Figure 11 shows the $\sigma_d(V)$ curves recorded at RT over a wide voltage range. At low voltages, the increase of $\sigma_d$ is attributed to RT CDW sliding, which shows saturation for values above about 1 V. However, at voltages above about 5 V ($E > 10$ kV/cm), the Shapiro steps under the 4 GHz irradiation. The sample dimensions are $5 \mu m \times 5.5 \times 10^{-3} \mu m^2$. 

FIG. 8. A wide-range temperature dependence of NbS$_3$-II resistance. Data from two whiskers (100 $\mu m \times 0.09 \mu m^2$-high $T$, 126 $\mu m \times 0.06 \mu m^2$-lower $T$) are combined into a single graph. The high-temperature points were obtained during a heating cycle in an Ar flow.

FIG. 9. Normalized RT differential resistance vs electric field of an as-grown high-Ohmic sample and after three subsequent heating/cooling cycles up to 800-850 K in an Ar atmosphere. The contact-separation $L = 1050 \rightarrow 630 \rightarrow 252 \mu m$ (becoming shorter after each heating cycle because of new contacts).

FIG. 10. The $\sigma_d(V)$ curves for a NbS$_3$-II sample below and above $T_{P1}$. The sample length is 44 $\mu m$.

FIG. 11. RT $\sigma_d(V)$ curves with and without RF irradiation. Note the Shapiro steps under the 4 GHz irradiation. The sample dimensions are $5 \mu m \times 5.5 \times 10^{-3} \mu m^2$. 

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σd grows rapidly again above the initial saturation level. To check whether this second rise comes from sliding of the UHT CDW, the effect of coherence stimulation by RF irradiation [3] was employed. One can see that, when the RF field is applied (with all other conditions kept fixed), σd grows faster and at lower electric fields (Fig. 11). Thus, the σd(V) diagram exhibits typical features of a CDW conductor with a threshold voltage and a saturation at higher voltages. Radio frequency voltage superimposed onto a slowly sweeping dc voltage would give a trivial opposite effect, i.e., smearing out the σd(V) curve (see Fig. 1 in Ref. [3]). Consequently, the observed increase of σd at higher voltages should be attributed to sliding of the UHT CDW. The largest current density jc of this sliding CDW, as estimated from Fig. 11, is ~5 × 106 A/cm²; it corresponds to a fundamental frequency of ~300 GHz (with jc/f = 18 A/MHz/cm²).

IV. THE UNUSUAL CDW FORMED BELOW 150 K

The transition at Tp2 = 150 K (Fig. 8) remains the least understood. It is only observed in the low-Ohmic subphase, and the drop in specific conductivity Δσs near 150 K is sample dependent [12,19]. These samples show a threshold in the I-V curves and Shapiro steps above the threshold voltage below Tp2 (Figs. 12 and 13), indicating the formation of a new CDW [12,13,19]. However, the “fundamental ratio” jc/f will correspond to about 1/3 of a chain per unit cell carrying the CDW. However, the lowest ratios measured below 150 K were two orders of magnitude smaller [12,19]. The difference between the RT CDW and LT CDW transport is clearly illustrated by Fig. 12, where σd vs nonlinear current is plotted for both CDWs under the same irradiation frequency, 400 MHz. For the first Shapiro step, the current of the LT CDW is nearly three orders of magnitude lower than that of the RT CDW. The CDW current density of the LT CDW is simply too low for a classical CDW formed through a Peierls transition.

Figure 14 shows an Arrhenius plot of specific conductivities σs for a number of NbS3-II samples. Shapiro steps at RT allow precise determination of their cross-sectional areas and consequently their specific conductivities. For comparison reasons, the dependence for a NbS3-I sample [17] is added. There is a large variation between the specific conductivities of different samples. The upper group of curves corresponds to the low-Ohmic subphase and clearly reveals the 150 K transition. These samples show RT specific conductivities ranging from 10 to 3 × 10²(Ω cm)⁻¹. This range is somewhat wider than reported earlier [8,19]. Despite the large differences in the actual conductivities, which vary by over an order of magnitude, the temperature dependence σs(T) for the low-Ohmic samples appears very similar on the logarithmic scale. Correspondingly, the drops in σs at Tp2 appear approximately proportional to their values above Tp2. Thus, all the excess electrons, not gapped at Tp0 and Tp1, are evidently dielectrically at Tp2, regardless of their actual concentration.

The conclusion is supported by the result presented in Fig. 15, where the fundamental ratio jc/f of the LT CDW is shown as a function of the drop in specific conductivity Δσs at Tp2 [41]. The CDW current density at fixed f will thus appears approximately proportional to Δσs.

If the single-particle conductivity above Tp2 and the CDW conductivity below Tp2 are provided by the same electrons, the mobility in the normal state can be estimated from the relationship between Δσs and the “fundamental ratio” (Fig. 15). By multiplying Δσs/(jc/f) with an estimated value of λ = 10 Å, which corresponds to the wavelengths of the RT

![Graph](image_url)
Samples with $\sigma_s(T) < 10(\Omega\text{cm})^{-1}$ belong to the high-Ohmic subphase. Their pronounced dielectric behavior of the $\sigma_s(T)$ curves at RT and below (Fig. 14) indicates that the free carriers arise from thermal excitations across the Peierls gap formed at $T_P1$. A pure high-Ohmic sample shows an activation energy of about 2000 K below $T_P1$. This value is close to 2500 K, the half-value of optical gap recently reported for NbS$_3$-II [42]. Evidently, the number of free electrons in these samples is insufficient to condensate into a collective state. Alternatively, the transition can just become invisible in $\sigma(T)$ because of the low electron concentration.

To better understand the properties of NbS$_3$-II, a few further experiments were performed. The chemical composition of these samples was determined with EPMA. Reliable compositions were only obtained for samples with transverse dimensions larger than 1 $\mu$m, which all belonged to the low-Ohmic subphase. The difference in composition between the two subphases was not yet established. From a total of 15 measurements at several sample locations, we obtained a S:Nb ratio of 2.87 ± 0.04 for the low-Ohmic subphase. Independent studies, performed at National Taiwan University (NTU; Taiwan) and Moscow Institute of Physics and Technology (MIPT, Russia) gave comparable results. The nonstoichiometry indicates either the presence of S vacancies or excess Nb in the low-Ohmic samples. It has been reported previously that NbS$_3$ is susceptible to loss of S with heating [43]. It is therefore likely that the variation in specific conductivity found in the NbS$_3$-II samples is a result of S vacancies acting as donors, similar to the observed behavior of TiS$_3$ [44,45] and TiSe$_2$ [46,47]. Consequently, the higher concentration of S vacancies would account for the reduced electrical resistivity observed in samples, belonging to the low-Ohmic subphase. Although the EPMA measurements were not performed on the high-Ohmic samples, it is reasonable to assume that these samples are closer to the stoichiometric composition. Their transformation into the low-Ohmic phase under high-temperature treatment (Fig. 9) then can be attributed to a loss of S.

The LT CDW seems to further condense electrons from a state already dielectricized by the UHT and RT CDWs. This is rather unusual. For the conventional RT CDW, condensation of Nb $d$ state electrons leads to a Peierls gap. To further condense electrons in the Nb $d$ state at $T_P2$, some additional electrons can be transferred from the S $p$ state into the Nb $d$ state by forming (S$_2$)$_2^{2-}$ pairs. We have used XANES to probe the hole occupancy of the Nb $d$ state and S $p$ state by studying fluorescence-detected Nb L$_3$ edge (at 2375 eV) and S K edge (at 2476 eV) absorption lines. From RT to 150 K, the Nb-L$_3$ peak intensity decreases due to reduced hole occupancy of the Nb 4$d$ state, as shown in Fig. 16(a). Similarly, the pre-edge feature (2470 eV) of the S K edge also decreases in intensity between these temperatures, as shown in Fig. 16(b). This pre-edge arises from the transition from 1$s$ to a $p$-$d$ mixed empty bound state when the S 3$p$ state takes on a “hole” character by mixing with the Nb 4$d$ states. A reduction of the S pre-edge intensity is consistent with the expectation that the Nb 4$d$ hole occupancy is reduced. However, we also found that the S K edge main peak decreases in intensity by lowering the temperature from 300 to 150 K. This means that a simple scenario of electrons transferred from S 3$p$ to Nb 4$d$ states is questionable. Instead, some electrons are transferred to both the S 3$p$ and Nb 4$d$.
states. Figure 17 shows the evolutions of the Nb $L_3$ edge peak, the S $K$ edge peak, and the S $K$ edge pre-edge intensities as functions of temperature. The behavior shown in Fig. 17 is reversible with temperature.

A source of electrons to occupy Nb 4$d$ and S 3$p$ states could be the S vacancies acting as electron donors. The concentration of S vacancies is much larger than typical doping concentrations in semiconductor materials and therefore likely makes NbS$_3$-II a degenerate semiconductor. If the electronic structure of NbS$_3$-II is such that an electron pocket (from the Nb 4$d$ state) and a hole pocket (from the S 3$p$ state) coexist at the Fermi energy, a slight shift of it can either decrease or increase the hole occupancies of both the Nb 4$d$ and S 3$p$ states. Such an electronic structure is depicted in Fig. 1(a) of Ref. [48] and is believed to exist in WTe$_2$ [49].

As NbS$_3$-II is cooled from 300 to 150 K, thermal generation of carriers becomes less important, and the Fermi level tends to move upwards, towards the donor impurity band. This is revealed by the decrease of both the Nb $L_3$ edge and S $K$ edge peak intensities observed in XANES.

By reducing the temperature from 150 to 50 K, the Nb $L_3$ edge and the S $K$ edge peak intensities recover, resulting in a broad minimum at $T_{P2}$. While the exact cause of this phenomenon is still uncertain, the removal of electrons from Nb 4$d$ and S 3$p$ states could be related to exciton formation. Reduced screening is expected in low-dimensional materials and at lower temperatures, where the reduced number of free carriers leads to enhanced exciton binding energy. If the LT CDW has indeed an excitonic insulator nature, exciton formation would remove electrons below $T_{P2}$ from the Nb 4$d$ and S 3$p$ states. This would also be in accord with the recovery of Nb $L_3$ edge and S $K$ edge peaks below $T_{P2}$ in XANES spectra.

We also observed a similar minimum of the Ta $L_3$ edge XANES in the orthorhombic phase of TaS$_3$ near $T_T = 220$ K, which is believed to be a “classical” Peierls transition [1]. However, in o-TaS$_3$ the S $K$ edge was not simultaneously measured with the Ta $L_3$ edge. We therefore do not yet know whether o-TaS$_3$ and NbS$_3$-II exhibit similar XANES behaviors. This comparative measurement should be undertaken in the future.

In addition to XANES, preliminary data from STEM-EELS suggest that the local environments of S atoms change at the transition. At 105 K, the L edge of S is characterized by a single peak at 162 eV, while at 290 K, an additional peak is formed at 167 eV. Meanwhile, the $M_2$ and $M_3$ edges of Nb do not show noticeable changes. The STEM-EELS data support the suggestion that there is a change in the local environment of sulfur atoms. It is possible that changes in the S positions lead to a change in the number of S-S bonds forming (S$_2$)$^{2-}$ instead of 2S$^{2-}$ states, or to a partial activation of S donor vacancies at low temperatures. These processes would provide another “degree of freedom” for the S atoms to change the number of free electrons without altering the Nb : S ratio.

Finally, NMR studies provide additional insight into the $T_{P1}$ and $T_{P2}$ transitions. The $^{93}$Nb spectrum of nonoriented NbS$_3$-II powder samples in a fixed magnetic field of $B_0 = 7.5$ T clearly shows the presence of many inequivalent Nb sites in this phase. The temperature dependence of the most intense peak of the central line (the $-1/2 \leftrightarrow 1/2$ transitions) allows clear detection of the CDW transition at $T_{P1}$. In Fig. 18(a), the variation of the Knight shift, $K = (f_f - 1) \times 100\%$, of this central peak intensity as a function of temperature.
corresponding transverse magnetization relaxation time (tens of microseconds). In this case, decoherence of the correlation time becomes comparable to the spin-echo time causing relaxation is increasing as temperature decreases. That the characteristic time of such microscopic fluctuations of transverse nuclear magnetization happens due to different field. The right scale in Fig. 18(a) shows the estimated values of the conduction electrons in the presence of an external magnetic field. The Knight shift corresponds to an “extra” effective field at the nuclear site from the polarization of the conduction electrons in the presence of an external field. The right scale in Fig. 18(a) shows the estimated values of this “extra” field. Therefore, Fig. 18(a) reveals a new ordered state below $T_{P1}$. The most obvious reason for this is a lattice distortion coupled with CDW formation, as observed in NbSe$_3$.

There is no change in the Knight shift around $T_{P2} = 150$ K [Fig. 18(a)]. However, studies of the nuclear relaxation do reveal a feature in this temperature region. The temperature dependence of the nuclear transverse magnetization relaxation rate, $T_2^{-1}$, measured on the same most intense peak of the central $^{93}$Nb NMR line, is shown in Fig. 18(b). A maximum of $T_2^{-1}(T)$ appears at about 130 K. The loss (decoherence) of transverse nuclear magnetization happens due to different time-dependent local magnetic fields at nuclear sites. The $T_2^{-1}(T)$ dependence can be explained by the assumption that the characteristic time of such microscopic fluctuations causing relaxation is increasing as temperature decreases. At low temperatures the fluctuations slow down and their correlation time becomes comparable to the spin-echo time (tens of microseconds). In this case, decoherence of the transverse nuclear spin magnetization becomes faster and the corresponding transverse magnetization relaxation time $T_2$ becomes shorter. At even lower temperatures the fluctuations become very slow and their characteristic time appears much longer in comparison with the time of spin-echo formation. Consequently, the fluctuations do not contribute anymore to the transverse magnetization relaxation and $T_2^{-1}$ begins to decrease again. Thus, the maximum in $T_2^{-1}$ at 130 K corresponds to a “freezing” of one of the fluctuation sources causing relaxation [51,52]. The most probable candidate for such a source is an electronic or a lattice distortion, which exists at $T_{P2}$ or even above it. Such a behavior can be expected in case of strong 1D fluctuations, where the transition signifies a 3D ordering of the CDW fluctuations (see, e.g., Ref. [53]). A similar behavior of $T_2^{-1}$ was reported for the well-known CDW conductor NbSe$_3$ near 130 K [51], i.e. somewhat below the first CDW transition at 144 K.

Unlike the Knight shift, the relaxation of the transverse magnetization can be stimulated by fluctuation of both magnetic and electric fields. Therefore, the maximum in $T_2^{-1}(T)$ at 130 K can reveal not only a lattice but also an electronic ordering. Moreover, one can suppose that the LT CDW is mainly an electronic ordering, with corresponding lattice distortion remaining a small, secondary effect. This is consistent with the absence of changes in the Knight shift at $T_{P2}$, as well as the failed attempts to detect a lattice distortion at 150 K with electron diffraction [10].

Uniaxial strain can be considered a probe for the Peierls transition. Unlike $T_{P1}$, $T_{P2}$ appears much less sensitive to strain [Fig. 6(a)]. Some lowering of $T_{P2}$ is only detected at the highest applied strains. Given the same strain, the relative reduction of $T_{P2}$ is an order of magnitude smaller than that of $T_{P1}$. Since the suppression of the fluctuations increases $T_{P2}$ and the corrugation of Fermi surfaces has the opposite influence, a balance of these two effects in LT CDW might explain why $T_{P2}$ is less sensitive to strain than $T_{P1}$. We note that different behaviors with respect to strain were observed in other trichalcogenides with multiple CDW transitions. In the monoclinic TaS$_3$, the lower transition appears to be more sensitive to strain [34] and pressure [2,35] as compared to the upper one. In NbSe$_3$, the effects of pressure and strain are different; while under pressure, the lower transition temperature decreases faster than the upper one [2,35]; the rate of decrease is much slower under uniaxial strain, especially for small strains [5]. In the case of NbS$_3$-II, we can suppose that either $T_{P2}$ is more dominated by fluctuations than $T_{P1}$, or the transition at $T_{P2}$ has a different origin.

The same effect of strain is observed for the nonlinear conduction of the LT CDW (Fig. 13): the I-V curves do not show notable changes up to about 1.5% strain (the apparent slight improvement of coherence was not repeated in other samples). The weak dependence of the LT CDW on strain can indicate weak coupling of this CDW with the lattice.

V. DISCUSSION

The results presented show that NbS$_3$-II appears to be a unique quasi-1D compound. It shows two high-temperature CDW transitions at $T_{P1} = 330−370$ K and $T_{P2} ≈ 620−650$ K. Both CDWs show a Fröhlich-mode transport. The LT CDW formed below 150 K also demonstrates sliding, but its nature is not fully understood.

The LT CDW is observed only in samples belonging to the low-Ohmic subphase and is not detected in the diffraction
experiments. Both the amplitude of the feature in $\sigma(T)$ at $T_{P2}$ and the density of the CDW current (at given $f_1$) depend strongly on the sample specific conductivity at RT, which can vary within the low-Ohmic subphase by more than an order of magnitude. Evidently, the variations in specific conductivity are connected with S deficiency. The excess free electrons, induced by “doping” from S vacancies, do not condense into a CDW until below 150 K. The concentration of these electrons is compatible with metals with a degenerate electronic gas. Consequently, the electronic structure can be treated in terms of new Fermi surfaces, which survived the two upper CDW transitions. The 150 K transition thus corresponds to the condensation of these free electrons and, presumably, of additional electrons released during redistribution of S bonding, into a CDW or a CDW-like formation. However, even for the low-Ohmic samples, the specific conductivity above $T_{P2}$ is far from being metallic. The free carriers are gapped by two already existing CDWs, and the resistivity is two to three orders of magnitude above the estimated value above $T_{P0}$ (Fig. 8). Both the $T_{P2}$ value and the form of the feature in $\sigma(T)$ at $T_{P2}$ (Fig. 14) show no obvious correlation with $\sigma_1$. Consequently, the transition at $T_{P2}$ is similar for the electron concentrations varying between samples by up to 1.5 orders of magnitude. In other words, the concentration of electrons condensed into the LT CDW can vary substantially, while the characteristics of this CDW remain similar. The transition disappears or becomes invisible only if $\sigma_1$ is less than $\sim 10(\Omega cm)^{-1}$. The possibility of forming a separate CDW by electrons from dopants has not been considered yet. In NbSe$_3$ and in the monoclinic phase of TaS$_3$, where multiple Peierls transitions are also observed, different transitions dielectrize the electrons belonging to different types of chains. In the case of NbS$_3$-II, some electrons originate from the S vacancies acting as donors, and they are not expected to occupy a separate band. In another case of CDW compound K$_{0.3}$MoO$_3$, doping with V results in extra holes, but these are gapped by the same CDW [54]. Contrary to NbS$_3$-II, doping in this case only results in a variation of the $q$ vector.

No hysteresis was found in the case of NbS$_3$-II in the $R(T)$, $R(V)$, and $R(\epsilon)$ measurements. Similar curves for nanosized samples did not show steps coupled with the addition/removal of a CDW wavelength ([7,55] and references therein). This could mean that the $q$ vectors of the two upper CDWs do not vary. NbS$_3$-II might have features in common with (TaSe$_3$I)$_2$, where the CDW also shows no metastable states (see [2], pp. 357–360 and 364–367). Probably for topological reasons [56], the CDWs cannot deform in NbS$_3$-II like they do in K$_{0.3}$MoO$_3$, TaS$_3$, and a number of other compounds. If this is the case, extra free electrons cannot be incorporated into the UHT or RT CDWs. Thus, “doping” with S vacancies results in a growth of conductivity at $T > T_{P2}$, while at $T_{P2}$, the extra electrons become condensed into a separate CDW.

While the structure of NbS$_3$-I is well known [57], the structure of NbS$_3$-II has not been determined yet. To analyze possible lattice instabilities of known phases of NbS$_3$, ab initio density functional theory (DFT) calculations of a band structure were performed for a model structure consisting of symmetrized monoclinic unit cells of NbS$_3$-I. That is, we manually removed the known dimerization of NbS$_3$-I and placed the atoms at the mean positions of a dimerized NbS$_3$-I unit cell [57]. This converted the compound into a metallic state. The DFT calculations were performed in both the local density approximations by the projector augmented-wave (PAW) method [58], and in the generalized gradient approximation [59] as implemented in the ABINIT simulation package [60]. Four bands were found to cross the Fermi level in the initial filling. Two of them have relatively large flat regions corresponding approximately to $\frac{1}{2}$ and $\frac{3}{2}$ filling of the respective bands. The corrugation of Fermi surfaces appears sensitive to the electron concentration: a reduced filling of these bands flattens the Fermi surfaces and makes the compound susceptible to CDW instability. These bands may be responsible for two transitions with two $q$ vectors. Two other bands form small near-cylindrical pockets around the Y point of the Brillouin zone and are aligned in the $Y-H$ direction. They are more sensitive to doping and may eventually disappear in the case of excessive doping. Extra electrons belonging to these pockets will not be condensed into the RT or UHT CDW, but may form a new condensed CDW state.

Several possibilities for the formation of the LT CDW can be considered. The first is the Peierls transition. This would require electrons, not condensed by the CDW transitions at $T_{P1}$ and $T_{P0}$, to be in nested sections of the Fermi surface. This is not a very likely scenario because of the strong dependence of the “fundamental ratio” on the electron concentration. There is a bigger probability that electrons belonging to these pockets will condense into a state in which the distance between the electrons depends on their concentration, both along and perpendicular to the chains. A candidate case is a Wigner crystal (WC), which is stabilized by the repulsive Coulomb forces. Unlike conventional CDWs, a WC is relatively weakly coupled with the lattice. This might be the reason why the LT CDW was not detected by TEM [61].

However, $T_{P2}$, as well as the form of the $\sigma(T)$ curve in the logarithmic scale (Fig. 14), remains stable over a wide range of electronic concentrations (nearly 1.5 orders of magnitude). Since the temperature of a Wigner crystallization depends on the concentration of electrons as a power law, the observation argues against a WC formation at 150 K.

The stability of $T_{P2}$ might indicate that the transition at 150 K, forming a periodic potential, is not directly coupled with the electrons induced by doping. Nonetheless, these electrons, irrespective to their concentration, are accommodated into this potential and form a CDW electronic crystal. In the case of the high-Ohmic samples, this condensation remains invisible in the $\sigma(T)$ curves. The transition at $T_{P2}$ can however show up in some other measurements, e.g., in $\sigma(\epsilon)$ studies at sufficiently high frequencies (see Fig. 4 in Ref. [15]). A kind of metal-dielectric transition was also observed close to 150 K in the NbS$_3$-I polytype, where the transition temperature appears stable in a wide range of pressures [18]. Thus, the 150 K transition might be an intrinsic feature of both NbS$_3$ polytypes and appears rather robust against pressure, strain, and doping.

Apart from the Wigner crystallization and the Peierls transition, the Keldysh-Koppev transition [23] (known also as the formation of excitonic dielectric [24]) has been suggested as a possible mechanism for condensation of electrons in NbS$_3$-II at $T_{P2}$ [19]. This transition represents a generalization of the Peierls transition, which can occur if the electron’s and hole’s Fermi surfaces have shapes that allow nesting. Such a
VI. CONCLUSIONS

Our studies reveal that NbS$_3$-II is an interesting member of the M$X_3$ group. It has three CDWs: a near RT CDW at $T_{P1}$ = 360 K, a LT CDW at $T_{P2}$ = 150 K, and another CDW at a much higher temperature $T_{P0} \approx 620$–650 K. Each CDW presents peculiar salient features. First, the RT CDW has exceptionally high coherence and shows extremely high velocities of CDW sliding. This suggests that this RT CDW can be considered for practical applications. Second, the threshold fields and currents of the UHT CDW are impressive. Third, the LT CDW is most unusual, and a complete physical picture of its formation remains elusive. However, the available evidence suggests that a novel mechanism linked to a Keldysh-Kopaev (excitonic insulator) transition could be involved in the formation of the LT CDW state.

For readers’ references, we opt to summarize the features of NbS$_3$-II as a list:

1) The properties of NbS$_3$-II depend strongly on the growth conditions. The RT conductivity $\sigma_d$(300 K) of the samples varies from $3 \times 10^2$ to $2(\Omega$cm)$^{-1}$.

2) The low-Ohmic samples [$\sigma_d$(300 K) = 10–3 $\times 10^2(\Omega$cm)$^{-1}$] are sulfur deficient with the S vacancies acting as electron donors. The gradual transformation of the high-Ohmic into the low-Ohmic ones under heating above 600 K is consistent with this conclusion.

3) The high-Ohmic subphase [$\sigma_d$(300 K) = 2–10($\Omega$cm)$^{-1}$] of NbS$_3$-II shows two CDW transitions at $T_{P1} \approx 340$–370 K and at $T_{P0} \approx 620$–650 K.

4) Apart from the transitions at $T_{P0}$ and $T_{P1}$, the low-Ohmic samples show a CDW transition at $T_{P2} = 150$ K. The specific conductivity drop at $T_{P2}$ is proportional to the specific conductivity above $T_{P2}$. This LT CDW is a condensate of the electrons donated by the S vacancies.

5) All three CDWs exhibit sliding at $E > E_t$.

6) Radiofrequency interference shows that the fundamental frequency of the RT CDW sliding can be at least 20 GHz. The extremely high coherence of this CDW is manifested in Bessel-type oscillations of $E_t$ and of the Shapiro steps’ width as a function of the RF power.

7) At RT, the NbS$_3$-II samples show torsional strain. The strain grows abruptly for $E > E_t$ and can be increased by an order of magnitude with RF irradiation.

8) Under uniaxial stretching, the RT CDW of NbS$_3$-II samples demonstrates many features of coherence enhancement, i.e., the threshold decreases and becomes sharper, the growth of $\sigma_d$ above $E_t$ is faster, the value of the maximum CDW conductivity increases, and the Shapiro steps under RF irradiation become more pronounced.

9) $T_{P1}$ is extremely sensitive to uniaxial stretching: $\varepsilon \sim 1\%$ can reduce $T_{P1}$ to below RT. The transition at $T_{P1}$ becomes sharper with strain, in line with the growth of the CDW coherence.

10) Sliding of the UHT CDW can be observed below and above $T_{P1}$. This is proven by the effect of RF-induced coherence stimulation. The $E_t$ value for this CDW can be on the order of 10 kV/cm at RT.

11) The LT CDW is nearly insensitive to tensile strain. At equal strain, the relative decrease of $T_{P2}$ is 10 times lower than that of $T_{P1}$. The I-V curves show no regular changes under strain up to 1%.

12) The charge density of the LT CDW transport, i.e. the $j_c/f_r$ value revealed by RF interference, is from 3 to 1000 times smaller than that of the RT CDW and scales with the specific conductivity above $T_{P2}$. The value $j_c/f_r$ appears to be well below $2e/s_0$, which is impossible for a normal CDW.

13) Unlike the cases of RT and UHT CDWs, no lattice distortion is observed at $T_{P2}$ by means of diffraction techniques. The $^{93}$Nb NMR study reveals a Knight shift at $T_{P1}$, but not at $T_{P2}$.

14) A clear maximum near $T_{P2}$ is observed in the temperature dependence of the nuclear transverse magnetization relaxation rate $T_2^{-1}$ measured at the central $^{93}$Nb NMR line. The feature in $T_2^{-1}(T)$ is interpreted by a freezing of the electronic density distortion (ordering) with cooling.

15) Near $T_{P2}$, minima are observed in XANES spectra of the S K edge, K line pre-edge, and the Nb L$_3$ edge. The result reveals electron transfer to both S 3p and to Nb 4d states down to $T_{P2}$. Further cooling below $T_{P2}$ reverses the trend of XANES intensity variation. A plausible cause for the observed XANES below $T_{P2}$ is a formation of excitons.

16) The condensation of the excess electrons in the low-Ohmic samples into a separate LT CDW can be explained with the rigidity of the RT and UHT CDWs, which does not allow changes of their $q$ vectors and condensation of extra electrons. In addition, ab initio calculations show that these electrons may belong to additional small pockets in the Fermi surfaces and a formation of a new condensed phase.

17) The nature of the LT CDW is not completely understood. The concept of an excitonic dielectric might explain the low sensitivity of $T_{P2}$ to the concentration of electrons condensed into this CDW.
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[19] From this, one can conclude that it is the $q_i$ CDW that provides the nonlinear transport at RT.
[20] Evidently, in Ref. [11], the high-Ohmic samples were reported, while in Ref. [13], the low-Ohmic ones (see below).
[25] Alternatively, the charge per chain transferred in time $1/f_e$ can be $1/e$, as it was reported for the monolcnic phase of TaS$_2$ [25]. This could be the case if the CDW current in a cell is carried by two chains with opposite phases.
[35] This is seen from easy cleavage of the samples, from their high aspect ratio (length/width $\sim 10^3$), from the elongated shape of the superlattice satellites [11]. The weak growth of the threshold field with cross-section reduction is also an indication of high anisotropy.
Here, $\Delta \sigma$ corresponds approximately to the difference in $\sigma$, between temperatures marking the half-height of the peak in the $d\log(\sigma)/dT$. 

In [11], it was noticed that, within the experimental accuracy, the $b^*$ component of $q_1 + 2q_0$ equals $b^*$. It was suggested that $q_1$ could be the second harmonic of $q_0$. Alternatively, this relation could imply an extra condition for $q_0$ and $q_1$, forbidding their variation.

Another probable reason for this is the large wavelength of the LT CDW. To estimate the $q_3$ vector, we must first establish the electronic structure of NbS$_3$. The two IC components of the two upper $q$-vectors are close to $1/3b^*$. If we assume that at $T_{\text{PT}}$, two chains per unit cell are gapped, and at $T_{\text{PT}}$, one (as found by the RF interference [19,12]), the relation $(q_{1b} + 2q_{0b})/3 = 1/3b^*$ gives the value of the “average” $q$ vector and can be interpreted as an indication of $1/3$ filling of the electronic band. Then the detailed formula of NbS$_3$ can be written as $2N$b$^b+N$b$^b+2(S$_2$)$^2-5S$.$\cdots$. In this case, there are 2/3 free electrons per Nb atom. Sulfur accepts either one or two electrons per atom. Then 5% S vacancies can provide 0.15 to 0.3 electrons per Nb atom. In this case, the CDW wavelength can range from 6.7 to 13.3 periods along the $b$ axis. It is a good challenge to find such a distortion.

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