

Alternative model for the structural modulation in NbSe<sub>3</sub> and *m*-TaS<sub>3</sub>

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A model is proposed for the sliding charge density wave mechanism in the isostructural quasi-one-dimensional compounds NbSe<sub>3</sub> and *m*-TaS<sub>3</sub>. It is based on an assumption that below either of the two onset temperatures  $T_1$  and  $T_2$  both modulation modes are present and form small statistically distributed domains along particular trigonal prismatic columns, i.e., along type III below  $T_1$  and in addition along type I below  $T_2$ . Presuming the domains are of comparable sizes to the coherence lengths in the diffraction experiments, their disordered distribution results in a selective contribution to the reciprocal space in both temperature regions, which is in good accord with the diffraction experiments. The model might form the basis for a better explanation of the twinkling domains and low temperature tunneling microscopic images, which were not fully understood on basis of the old model with a selective occupation of the two column types by the two modulation modes.

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## INTRODUCTION

NbSe<sub>3</sub> and the monoclinic form of TaS<sub>3</sub> (*m*-TaS<sub>3</sub>) are two very peculiar isostructural compounds (Fig. 1) (NbSe<sub>3</sub>:  $a = 10.009 \text{ \AA}$ ,  $b = 3.4805 \text{ \AA}$ ,  $c = 15.629 \text{ \AA}$ ,  $\beta = 109.47^\circ$ ,  $P2_1/m$ ,  $Z=6$ ),<sup>1,2</sup> (*m*-TaS<sub>3</sub>:  $a = 9.515 \text{ \AA}$ ,  $b = 3.3412 \text{ \AA}$ ,  $c = 14.912 \text{ \AA}$ ,  $\beta = 109.00^\circ$ ,  $P2_1/m$ ,  $Z=6$ ).<sup>3,4</sup> They belong to both families, the quasi one- as well as quasi-two-dimensional structures. The first are characterized by the trigonal prismatic (TP) columns along their **b** direction and the second by layers in the **b-c** plane, held together by weak van der Waals (vdW) forces. In the past considerable attention was paid to both compounds due to their nonlinear transport properties,<sup>5,6</sup> which were attributed to sliding charge-density waves (CDW's). It was generally accepted that in both cases two incommensurate (IC) modulation modes appear independently at different onset temperatures along two of the three available TP columns.<sup>7,8</sup> The only difference between NbSe<sub>3</sub> and *m*-TaS<sub>3</sub> is that in the first the IC component of the  $\mathbf{q}_1$  vector is slightly smaller and the one of  $\mathbf{q}_2$  slightly larger than  $\frac{1}{4}b^*$ , while in the second this is reversed [NbSe<sub>3</sub>:  $\mathbf{q}_1 = (0, 0.241, 0)$ ,  $\mathbf{q}_2 = (0.5, 0.260, 0.5)$ ,  $T_1 = 144 \text{ K}$ ,  $T_2 = 59 \text{ K}$ ;<sup>1,2,7</sup> *m*-TaS<sub>3</sub>:  $\mathbf{q}_1 = (0, 0.254, 0)$  and  $\mathbf{q}_2 = (0.5, 0.245, 0.5)$ ;  $T_1 = 240 \text{ K}$ ,  $T_2 = 160 \text{ K}$ ].<sup>3,4</sup> It appears that in both compounds the two components add to  $\frac{1}{2}b^*$  within the experimental error. However, Fleming *et al.*<sup>7,9</sup> concluded from high-resolution synchrotron experiments that the difference from  $\frac{1}{2}b^*$  in NbSe<sub>3</sub> is 0.006, which is three times their possible experimental error.

NbSe<sub>3</sub> was the subject of intensive studies. These included structural determination,<sup>11–20</sup> numerous theoretical

and experimental works on CDW transport and pinning,<sup>5,6,11–15,21–43</sup> nuclear magnetic resonance experiments,<sup>44–46</sup> transmission electron (TEM),<sup>47,48</sup> scanning tunneling (STM) of both pure<sup>49–55</sup> and doped NbSe<sub>3</sub> (Ref. 56) as well as atomic force microscopy (AFM) of doped NbSe<sub>3</sub>.<sup>57–61</sup> However, some questions are still a matter of debate: Wilson<sup>62</sup> explained the two IC components of  $\mathbf{q}_1$  and  $\mathbf{q}_2$  on the basis of bonding and electron transfer and Bruinisma and Trullinger<sup>63</sup> suggested a rather strong tendency of the two CDW's to be locked to each other and less to the lattice, but their apparent addition to  $\frac{1}{2}b^*$  in both compounds remains unclear. Another peculiar fact is that from a large number of known CDW compounds only a few were reported to show sliding, making it an exception rather than a rule; in addition to NbSe<sub>3</sub> (Refs. 5, 64–66) and *m*-TaS<sub>3</sub>,<sup>67</sup> these include NbS<sub>3</sub>,<sup>68</sup> (TaSe<sub>4</sub>)I,<sup>69</sup> (NbSe<sub>4</sub>)<sub>10</sub>I<sub>3</sub>,<sup>70</sup> A<sub>0.3</sub>MoO<sub>3</sub> with A = K, Rb, Tl (Ref. 71–74) and tetra-thio-fulvalene-tetracyanoquinomethane (TTF-TCNQ).<sup>75</sup> Further, STM stud-

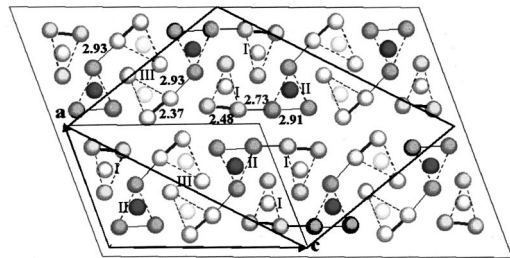


FIG. 1. The structure of NbSe<sub>3</sub>. Indicated are the average structure unit cell (small cell) with the three types of columns, the enlarged LT2 modulation unit cell and the shortest Se-Se distances.

ies of NbSe<sub>3</sub> at 4.2 K could not be unambiguously brought into accord with the accepted model.<sup>52</sup> Most striking is the fact that the modulation observed was present with a variable intensity along all three types of columns, which should not be the case if the type-III and type-I columns were selectively modulated by the  $\mathbf{q}_1$  and  $\mathbf{q}_2$  modes only.<sup>52,53</sup> Last not least, twinkling stringlike domains, composed of moirélike fringes, were observed in satellite dark field TEM images above the lower onset temperature. They remained a puzzle<sup>47,48</sup> although Fleming *et al.*<sup>9</sup> attributed these effects to radiation damage.

### THE MODEL

The basic assumption of the present approach is that a normal Peierls transition with a single  $\frac{1}{4}b^*$  modulation mode is not stable in the discussed cases. Instead, two modes with  $(\frac{1}{4} - \varepsilon)b^*$  and  $(\frac{1}{4} + \varepsilon)b^*$  appear simultaneously forming disordered domains, whose sizes are smaller than the coherence regions in the diffraction experiments.<sup>76</sup> In such experiments no clue can be found whether the disorder is established in a dynamical equilibrium between the two modes or as a spatial disorder. Although this approach might be realized in various ways, the presented model is verified as long-period commensurate rather than IC for both CDW's. It is assumed that an even number of  $\mathbf{q}_1$  and an odd number of  $\mathbf{q}_2$  modulation periods correspond within the experimental error to a certain even number of unit cells of the average structure, e.g., for NbSe<sub>3</sub> the IC components of  $\mathbf{q}_1 = (0, 0.241, 0)$  and  $\mathbf{q}_2 = (0.5, 0.260, 0.5)$  are approximated by  $q_{1b} = \frac{14}{58} = 0.241$  and  $q_{2b} = \frac{15}{58} = 0.259$ . The spatial and/or temporal disorder between the two modes is formally achieved by considering a superposition of the form  $\mathbf{q}_1 + \mathbf{q}_2$ , which on the other hand locks-in with the lattice of the basic structure. The suggested model can be described in a few steps:

(1) In the temperature region between  $T_1$  and  $T_2$  the modulation occurs only along type-III (in the literature denoted as “yellow”) columns (LT1 modulation). These are the columns with the shortest Se-Se bonds and with a close neighborhood between two columns of the same type forming a pair. They are arranged in slabs along the  $\mathbf{a}$  direction and the modulation is described in the small cell of Fig. 1. The modulation is composed of two modes ( $\mathbf{q}_1$  and  $\mathbf{q}_2$ ), both considered as long period commensurate ( $q_1 = 14 \text{ mod } 58b$ ,  $q_2 = 15 \text{ mod } 58b$ ). Both modes are in an equilibrium and can easily switch one into another, resulting in an average  $\mathbf{q}_1 + \mathbf{q}_2$  modulation. Only full periods are interchanged along each TP column and the  $\mathbf{q}_1$  mode locks-in with the underlying basic structure twice as often as the  $\mathbf{q}_2$  mode does.

(2) Important is the ordering perpendicular to and along the columns. Regardless of whether the type-III columns in the LT1 temperature range are modulated by the  $\mathbf{q}_1$  or  $\mathbf{q}_2$  modes, the domains these modes form are fully out-of-phase ordered along the  $\mathbf{a}$  direction. However, contrary to the  $\mathbf{q}_1$  domains, which are all in-phase along the columns, the domains with the twice longer  $\mathbf{q}_2$  modulation periods can appear with two possible phase shifts, dependent on whether the number of  $\mathbf{q}_1$  modes separating them, is even or odd.

These  $\mathbf{q}_2$  phases will in different domains of the same columns along the  $\mathbf{a}$  direction be either  $(0, \frac{1}{2}, 0, \frac{1}{2}, \dots)$  or  $(\frac{1}{2}, 0, \frac{1}{2}, 0, \dots)$  with regard to a single modulation length, whereas the  $\mathbf{q}_1$  domains will all adopt one of the two possibilities only. By a spatial (and/or temporal) averaging within the range of coherent scattering only the  $\mathbf{q}_1$  domains will contribute to the reciprocal space.

(3) Below the temperature  $T_2$  an additional modulation of the form  $\mathbf{q}_1 + \mathbf{q}_2$  occurs along the type-I (“orange”) columns with slightly less isosceleslike bases (LT2 modulation). The Se-Se bond lengths are slightly longer than in the type-III columns, but the adjacent columns forming a pair are at comparable distances. However, pairs of type-I columns are not arranged in slabs as the type-III columns are, but are separated by the type-II columns. Type-II (“red”) columns have much longer Se-Se bonds, they are isolated and are apparently not modulated. The LT2 modulation must be described in the larger cell of Fig. 1, which is a  $B$ -centered setting for the basic structure. The LT2 modulation destroys the  $B$  centering. Due to the shielding between type-I column pairs there is an important difference with regard to the ordering along the type-III columns. The  $\mathbf{q}_1$  modes along the two TP columns forming a type-I pair will again be out of phase. Due to shielding it is energetically equivalent to shift the two pairs with regard to the  $\mathbf{q}_1$  modes in the nearest type-III columns by  $\frac{1}{4}$  and  $\frac{3}{4}$  or vice versa (the phase shifts are again given with regard to a single modulation period). The resulting disorder is different from the one  $\mathbf{q}_2$  domains undergo along the type-III columns. All  $\mathbf{q}_1$  domains along a particular type-I column are again in phase, but the  $\mathbf{q}_1$  phase shifts between adjacent type-I column pairs are not fully correlated any more. They are statistically distributed between the two possible out-of-phase settings, giving no contribution to the reciprocal space.

(4) The  $\mathbf{q}_2$  mode along the type-I columns should as part of the LT2 modulation undergo an antiphase disorder for both mentioned reasons; the first would be related to the  $\mathbf{q}_1$  disorder between the type-I column pairs and the second to the disorder caused by the two possible phase shifts along particular columns, like as in case of the LT1 modulation. However, at these lower temperatures the type-III and type-I columns already tend to achieve the best possible phase relationships. If that was not so, the modulation unit cell in the LT2 temperature range would not be enlarged. Once established, the phases of the  $\mathbf{q}_1$  domains along particular type-I column pairs cannot be rearranged any more. This is because all  $\mathbf{q}_1$  domains along an entire pair of columns, separated by the  $\mathbf{q}_2$  domains, would have to be interchanged simultaneously. Contrary, the phases of the  $\mathbf{q}_2$  domains can be changed individually between  $\frac{1}{4}$  and  $\frac{3}{4}$ , because either of the two possible shifts will equally well lock-in with either of the two fixed  $\mathbf{q}_1$  phase shifts ( $\frac{1}{4}$  or  $\frac{3}{4}$ ). Consequently, the nearest four columns modulated by  $\mathbf{q}_2$ , i.e., the two type-III columns along the  $\mathbf{a}$  direction and the two type-I columns along the  $[101]$  direction, will become fully ordered with the corresponding phases 0 (type III),  $\frac{1}{4}$  (type I),  $\frac{1}{2}$  (type III), and  $\frac{3}{4}$  (type I) or alternatively with  $\frac{1}{2}$  (type III),  $\frac{1}{4}$  (type I), 0 (type III),  $\frac{3}{4}$  (type I). In both cases the same phases will be kept at

the type-I columns and their  $\mathbf{q}_2$  domains will contribute to the corresponding diffraction pattern.

### THE CALCULATION

In Fig. 1 a larger area of the average structure is shown. Two cells are indicated, the small cell of the average structure and the LT1 modulation with the lattice constants  $(\mathbf{a}, \mathbf{b}, \mathbf{c}, \beta)$  and the larger cell of the LT2 modulation, with the lattice constants  $(\mathbf{a} + \mathbf{c}, \mathbf{b}, \mathbf{c} - \mathbf{a}, \beta')$ . The shortest Se-Se bonds are also given.

The described model must work equally well for any amplitudes and IC modulation vectors, presuming the basic requirements described in the model are fulfilled. To prove it a simplified model was considered first. The reciprocal space of a superstructure with 14 Nb atoms (i.e., with the Se atoms omitted) fitting six and five modulation periods for  $\mathbf{q}_1$  and  $\mathbf{q}_2$ , was calculated. An arbitrary transversal modulation of two sine functions for  $\mathbf{q}_1$  and  $\mathbf{q}_2$  affecting the  $x$  coordinates only was applied to the type-III columns for the LT1 modulation and to both, type-III and type-I columns in case of the LT2 modulation:

$$\begin{aligned} x' &= x + 0.02 \sin[2\pi(6y - \Delta)] + 0.02 \sin[2\pi(5y - \Delta)], \\ y' &= ny/14, \quad n = 1, \dots, 14, \\ z' &= z, \end{aligned} \quad (1)$$

with  $|q_1| = 6/14 = 0.43$  and  $|q_2| = 5/14 = 0.36$ , with  $\Delta$  describing the phase shifts and with  $x, y, z$  and  $x', y', z'$  the average structure and the modulated structure Nb coordinates. The results of the calculations are shown in Figs. 2 and 3. In the case considered in Figs. 2 a modulation composed of the  $\mathbf{q}_1$  and  $\mathbf{q}_2$  modes, with all required characteristics of the described realistic model, was applied to the type-III columns only (LT1 modulation). The  $\mathbf{q}_1$  mode locks-in after every seven unit cells, which is twice as often as the  $\mathbf{q}_2$  mode does. Therefore, two possible phase shifts ( $\Delta = 0$  or  $\Delta = \frac{1}{2}$ ) occur between  $\mathbf{q}_2$  domains. Contrary, the  $\mathbf{q}_1$  blocks are all in phase, regardless of the number of  $\mathbf{q}_2$  modulation periods separating them. By a spatial (and/or temporal) averaging within the range of coherent scattering the unit cell of the superstructure appears as a superposition of domains with  $\Delta = 0$  and  $\Delta = \frac{1}{2}$  for the  $\mathbf{q}_2$  and with only  $\Delta = 0$  for the  $\mathbf{q}_1$  mode. In the model calculation split positions with an occupancy of 0.5 account for the superposition of the modes with different phases. In Fig. 2(a) the phase shifts are indicated by numbers in brackets for each split position, the first number being the phase for  $\mathbf{q}_1$  and the second the one for  $\mathbf{q}_2$ . Between two adjacent columns in a pair the modulation always differs by a phase shift of  $\pi$  (i.e.,  $\Delta = \frac{1}{2}$ ) leading to an ordered sequence along the  $\mathbf{a}$  direction. For convenience the large  $B$ -centered cell was chosen in case of LT1 as well, although the calculation could have been performed in the small cell of the basic structure. The calculated diffraction pattern in Fig. 2(b) shows that only  $\mathbf{q}_1$  satellites ( $|q_1| = 0.43$ ) appear in the plane  $(k, k, 0)$  between the positions of the main reflections. Since the chosen unit cell was  $B$ -centered, only reflections with even  $h$  values in the plane  $(h, k, 0)$  were expected.

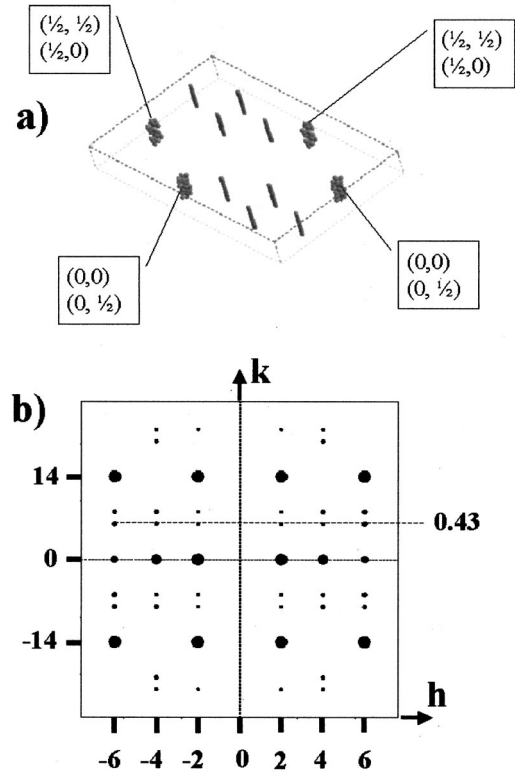


FIG. 2. The model calculation as described in the text for the LT1 temperature region. In (a) the modulated and nonmodulated Nb chains are depicted and the phases for both split positions (see text) are indicated. In (b) the calculated diffraction pattern with  $\mathbf{q}_1$  satellites only is shown.

Figures 3(a) and 3(b) exhibit the LT2 modulation, again for the simple model structure. By lowering the temperature type-I columns are also modulated in addition to those of type III, again with all important features, described for the realistic model. The same modulation function was applied to the columns of type I with the phases as indicated in Fig. 3(a). The  $B$  centering is destroyed by the modulation and satellites (but no main reflections) appear also at odd  $h$  values. The additional satellites are only of the  $\mathbf{q}_2$  type ( $|q_2| = 0.36$ ) and the calculated diffraction pattern is qualitatively in full agreement with the expectations, as shown in Fig. 3(b).

As a second step in our analysis the realistic model was calculated, with all Nb and Se atoms in the real cell present and displaced according to the modulation functions with  $|q_1| = \frac{14}{58}$  and  $|q_2| = \frac{15}{58}$ . All characteristic features of the described model were again maintained. The amplitudes were similar to those given by van Smaalen *et al.*<sup>10</sup> For simplicity reasons the calculations were carried out with an enlarged  $2a \times 58b \times 2c$  unit cell. A longitudinal modulation of the Nb chains, accompanied by a longitudinal and transversal adjustment of the corresponding Se TP columns was considered. In addition small adjustments of the Se(5) and Se(6) chains of the type-II columns were also considered, first to relax the modulation along the type-III and second that along the type-I columns. Like in the simplified model the calculated features of the diffraction pattern for the more realistic

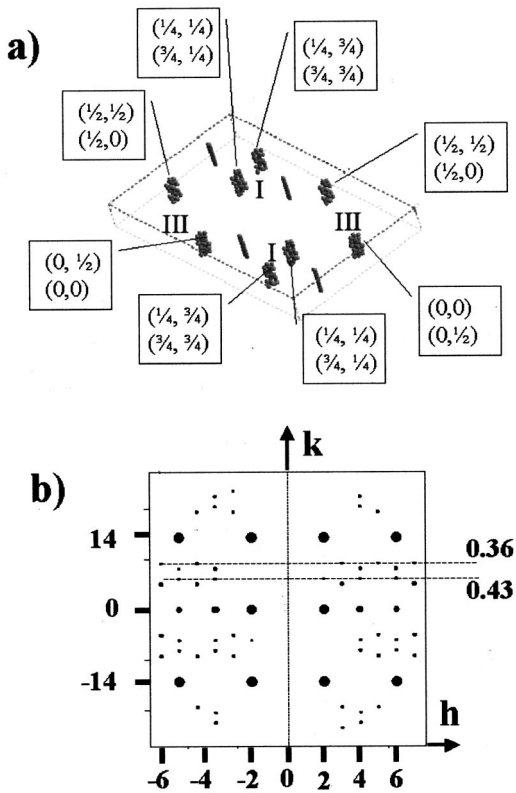


FIG. 3. The model calculation as described in the text for the LT2 temperature region. In (a) the modulated and non-modulated Nb-chains are depicted and the phases for the two modulated types of columns (see text) are indicated. In (b) the calculated diffraction pattern with  $\mathbf{q}_1$  and  $\mathbf{q}_2$  satellites is shown.

model agree qualitatively very well with the observed x-ray and electron diffraction patterns. Satellites appear only at positions where they are observed. Further, as expected, the intensities of the satellites, calculated on the basis of the model with the two modulation modes applied selectively to the type-III and type-I columns and those, calculated on the basis of the present model, gave identical values, if the same amplitudes were used in both cases. In our calculations the amplitudes were not refined further with the aim to achieve a better agreement with the experimental data, since no such accurate and reliable data were available.

## DISCUSSION

The essence of this paper is to show that in principle the DP's in both LT1 and LT2 temperature range can formally be explained on the basis of a superposition of the two modes  $\mathbf{q}_1 + \mathbf{q}_2$  and that the  $\mathbf{q}_2$  mode can indeed be present in the LT1 temperature range already, without giving any detectable contribution to the diffraction pattern. Thus, the observed phase transition may in this case mean that the same modulation affects the second type of columns just by rearranging the relative phases.

It appears that the phases between type-III and type-I columns are in fact not very important, since they hardly influence the calculated intensities. In the present model they were taken as  $\frac{1}{4}$  and  $\frac{3}{4}$  at the type-I columns as compared to 0

and  $\frac{1}{2}$  along the type-III columns, which seems to be the most plausible choice. Contrary to the  $\mathbf{q}_1$  ordering, the  $\mathbf{q}_2$  ordering must involve phase shifts smaller than  $\pi$ . As a net result of the possible phase relationships between various domains the  $\mathbf{q}_1$  periods in type-III columns must turn out to be fully ordered and the  $\mathbf{q}_2$  ones disordered, whereas in the type-I columns it should be vice versa. In addition to keeping both  $\mathbf{q}_1$  and  $\mathbf{q}_2$  modes along all columns forming a pair out-of-phase and in addition to keeping the type-III and type-I columns in proper phase relationships, an additional relationship between the type-I column pairs must be established in the LT2 range, which enlarges the modulation unit cell. The experiments show, that not only the  $\mathbf{q}_2$  contribution is present in the LT2 region, but also that its ordering *requires* a doubling of the modulation unit cell. The later is a clear proof that at lower temperatures interactions at distances, which involve the isolated type-I column pairs, *must* take place. Obviously, the  $\mathbf{q}_1$  and  $\mathbf{q}_2$  out-of-phase ordering between the type-III columns forming sheets represent the most stable ordering. The  $\mathbf{q}_2$  disorder in that case (LT1) is a disorder at relatively large distances, because it is generated by the number (even or odd) of the  $\mathbf{q}_1$  periods separating the  $\mathbf{q}_2$  parts along the columns. The ordering that appears at lower temperatures (LT2) between the type-I column pairs is of a different type. It involves ordering *perpendicular* to the columns, at distances only slightly larger as compared to those involved in LT1 ordering. Once the interaction between the type-III and type-I columns is established, both the  $\mathbf{q}_1$  and  $\mathbf{q}_2$  domains along the type-I column pairs tend to become ordered with regard to the nearest type-III pairs. The same ordering as in case of  $\mathbf{q}_1$  applies for the  $\mathbf{q}_2$  ordering as well, but the later already contain out-of-phase domains along the type-III columns. Thus, neither  $\mathbf{q}_1$  nor  $\mathbf{q}_2$  domains should contribute to the reciprocal space. However, an interaction between type-I pairs themselves results in full  $\mathbf{q}_2$  ordering at distances comparable to the enlarged modulation unit cell.

There are experiments, which were not satisfactorily explained on the basis of the model with the two modulation modes selectively occupying the type-III and type-I columns. First, the observed mobile and twinkling domains as well as the moirélike fringes in satellite dark field (SDF) TEM images of NbSe<sub>3</sub> (Ref. 47) are consistent with the appearance of fluctuating antiphase domains which are assumed as being characteristic for the  $\mathbf{q}_1 + \mathbf{q}_2$  modulation in the LT1 temperature region. Thus, there is a new interpretation of the twinkling beside the one proposed by Fleming *et al.*,<sup>9</sup> who attributed this twinkling to radiation damage. It should be noted that Fung and Steeds<sup>47</sup> already pointed out that the effects observed in SDF images might be attributed to an interference between CDW's with slightly different modulation vectors. However, the idea was not followed further, because the fringes were indeed observed at temperatures above  $T_2$  already, where the  $\mathbf{q}_1$  mode only was supposed to be present.

Second, the low-temperature STM image of NbSe<sub>3</sub>, which could not be unambiguously explained on basis of the previous model may indeed show disordered  $\mathbf{q}_1$  and  $\mathbf{q}_2$  modes with phase shifts as suggested. There is an additional interesting point connected with the low temperature STM and AFM works.<sup>52-54,56-61</sup> The images consistently show a

comparable modulation along *all three* column types. This can easily be brought into accord with the present model, presuming the *same* modulation modes are present along the observed type-III, type-I, and type-II columns, but seems to be in contradiction with the model, which allows only *different* modes along the type-III and type-I column types.

In conclusion, the proposed model seems to be in certain cases in better agreement with the experimental observations than the models considered so far. However, other solutions based on similar principles cannot be simply ruled out until the satellite intensities in both temperature regions are re-determined as accurately as possible and the modulation struc-

ture refined within models, which take into account a statistical distribution of the two modes.

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- <sup>1</sup>A. Meerschaut and J. Rouxel, *J. Less-Common Met.* **39**, 197 (1975).
- <sup>2</sup>J. L. Hodeau, M. Marezio, C. Roncan, R. Ayroles, A. Meerschaut, J. Rouxel, and P. Monceau, *J. Phys. C* **11**, 4117 (1978).
- <sup>3</sup>C. Roucau, R. Ayroles, P. Monceau, L. Guemas, A. Meerschaut, and J. Rouxel, *Phys. Status Solidi* **62**, 483 (1980).
- <sup>4</sup>A. Meerschaut, L. Guemas, and J. Rouxel, *J. Solid State Chem.* **36**, 118 (1981).
- <sup>5</sup>P. Monceau, *Electronic Properties in Inorganic Quasi-One-Dimensional Materials*, edited by P. Monceau (Reidel, Dordrecht, 1985), p. 139.
- <sup>6</sup>*Physics and Chemistry of Low-Dimensional Inorganic Conductors*, edited by C. Schlenker, J. Dumas, M. Greenblatt, and S. van Smaalen, Vol. 354 of *NATO Advanced Studies Institute Series B: Physics* (Plenum, New York, 1996).
- <sup>7</sup>R. M. Fleming, D. E. Moncton, and D. B. McWhan, *Phys. Rev. B* **18**, 5560 (1978).
- <sup>8</sup>J. A. Wilson, F. J. DiSalvo, and S. Mahajan, *Adv. Phys.* **24**, 117 (1975).
- <sup>9</sup>R. M. Fleming, C. H. Chen, and D. E. Moncton, *J. Phys. (France)* **44**, C3-1651 (1983).
- <sup>10</sup>S. Van Smaalen, J. L. De Boer, A. Meetsma, H. Graafsma, H.-S. Sheu, A. Darovskikh, P. Coppens, and F. Lévy, *Phys. Rev. B* **45**, 3103 (1992).
- <sup>11</sup>*Proceedings of the International Workshop on Electronic Crystals*, Carry-le-Rouet, 1993, edited by S. Brazovskii and P. Monceau [*J. Phys. IV* **3** (1993)].
- <sup>12</sup>*Proceedings of the International Conference on Science and Technology of Synthetic Metals*, Göteborg, 1992, edited by S. Stafström, W. R. Salaneck, O. Inganäs, and T. Hjertberg [*Synth. Met.* **55–57**, (1993)].
- <sup>13</sup>*Proceedings of the 20 International Conference on Low-Temperature Physics*, Eugene, 1993, edited by R. J. Donnelly [*Physica B* **194–196** (1994)].
- <sup>14</sup>*Proceedings of the International Conference on Science and Technology of Synthetic Metals*, Seoul, 1994, edited by Y. W. Park and H. Lee [*Synth. Met.* **70** (1995)].
- <sup>15</sup>*Proceedings of the 22 International Conference on Low-Temperature Physics*, Helsinki, 1999, edited by V. Gantmakher, P. Hakonen, E. Thuneberg, J. Pekola, and F. B. Rasmussen [*Physica* **284–288** (2000)].
- <sup>16</sup>J. D. Brock, A. C. Finnefrock, K. L. Ringland, and E. Sweetland, *Phys. Rev. Lett.* **73**, 3588 (1994).
- <sup>17</sup>S. Rouzière, S. Ravy, J. P. Pouget, and R. Thorne, *Solid State Commun.* **97**, 1073 (1996).
- <sup>18</sup>H. Requardt, F. Ya. Nad, P. Monceau, R. Currat, J. E. Lorenzo, S. Brazovskii, N. Kirova, G. Grübel, and Ch. Vettier, *Phys. Rev. Lett.* **80**, 5631 (1998).
- <sup>19</sup>V. Kiryukhin, D. Casa, B. Keimer, M. J. Higgins, and S. Bhattacharya, *Phys. Rev. B* **57**, 1332 (1998).
- <sup>20</sup>Y. Li, D. Y. Noh, J. H. Price, K. L. Ringland, J. D. Brock, S. G. Lemay, K. Cicak, R. E. Thorne, and M. Sutton, *Phys. Rev. B* **63**, R041103 (2001).
- <sup>21</sup>R. E. Thorne, W. G. Lyons, J. W. Lyding, J. R. Tucker, and J. Bardeen, *Phys. Rev. B* **35**, 6348 (1987); **35**, 6360 (1987).
- <sup>22</sup>A. Jánossy, C. Berthier, and P. Ségransan, *Phys. Scr.* **19**, 578 (1987).
- <sup>23</sup>J. C. Gill, *Phys. Scr.* **T25**, 51 (1989).
- <sup>24</sup>D. A. DiCarlo, J. McCarten, T. L. Adelman, M. Maher, and R. E. Thorne, *Phys. Rev. B* **42**, 7643 (1990); J. R. Tucker, *ibid.* **47**, 7614 (1993); D. A. DiCarlo, J. McCarten, and R. E. Thorne, *ibid.* **47**, 7618 (1993).
- <sup>25</sup>H.-A. Lu, C. G. Slough, R. V. Coleman, A. Maiti, and L. M. Falicov, *Phys. Rev. B* **44**, 6037 (1991).
- <sup>26</sup>S. K. Srivastava and B. N. Avasthi, *J. Mater. Sci.* **27**, 3693 (1992).
- <sup>27</sup>S. Ramakrishna, M. P. Maher, V. Ambegaokar, and U. Eckern, *Phys. Rev. Lett.* **68**, 2066 (1992).
- <sup>28</sup>M. P. Maher, T. L. Adelman, S. Ramakrishna, J. P. McCarten, D. A. DiCarlo, and R. E. Thorne, *Phys. Rev. Lett.* **68**, 3084 (1992).
- <sup>29</sup>J. C. Gill, *Phys. Rev. Lett.* **70**, 331 (1993); P. M. Maher, S. Ramakrishna, R. E. Thorne, and V. Ambegaokar, *ibid.* **72**, 3915 (1994).
- <sup>30</sup>T. L. Adelman, S. V. Zaitsev-Zotov, and R. E. Thorne, *Phys. Rev. Lett.* **74**, 5264 (1995).
- <sup>31</sup>T. L. Adelman, M. C. de Lind van Wijngaarden, S. V. Zaitsev-Zotov, D. Di Carlo, and R. E. Thorne, *Phys. Rev. B* **52**, R5483 (1995).
- <sup>32</sup>I. Bâldea, H. Köppel, and L. S. Cederbaum, *Phys. Rev. B* **52**, 11 845 (1995).
- <sup>33</sup>M. P. Maher, T. L. Adelman, D. A. DiCarlo, J. P. McCarten, and R. E. Thorne, *Phys. Rev. B* **52**, 13 850 (1995).
- <sup>34</sup>I. Bloom, *Phys. Rev. B* **53**, R10 449 (1996).
- <sup>35</sup>S. G. Lamay, R. E. Thorne, M. C. Saint-Lager, and P. Monceau, *Phys. Rev. B* **54**, 16 341 (1996).
- <sup>36</sup>J. P. Sorbier, H. Tortel, P. Monceau, and F. Lévy, *Phys. Rev. Lett.* **76**, 676 (1996).
- <sup>37</sup>J. C. Gill, *Phys. Rev. B* **53**, 15 586 (1996).
- <sup>38</sup>T. L. Adelman, M. C. de Lind van Wijngaarden, S. V. Zaitsev-

- Zotov, D. DiCarlo, and R. E. Thorne, *Phys. Rev. B* **53**, 1833 (1996).
- <sup>39</sup>I. Bâldea, H. Köppel, and L. S. Cederbaum, *Synth. Met.* **86**, 2225 (1997).
- <sup>40</sup>Yu. I. Latyshev, O. Laborde, P. Monceau, and S. Klaumünzer, *Phys. Rev. Lett.* **78**, 919 (1997).
- <sup>41</sup>J. Kuh, Y. T. Tseng, K. Wagner, J. Brooks, G. X. Tessema, and M. J. Skove, *Phys. Rev. B* **57**, 14 576 (1998).
- <sup>42</sup>S. G. Lemay, M. C. de Lind van Wijngaarden, T. L. Adelman, and R. E. Thorne, *Phys. Rev. B* **57**, 12 781 (1998).
- <sup>43</sup>J. H. Ross, Z. Wang, and C. P. Slichter, *Phys. Rev. Lett.* **56**, 663 (1986).
- <sup>44</sup>F. Devreux, *J. Phys. (France)* **43**, 1489 (1982).
- <sup>45</sup>J. Shi and J. H. Ross, *Phys. Rev. B* **45**, 8942 (1992).
- <sup>46</sup>K. K. Fung and J. W. Steeds, *Phys. Rev. Lett.* **45**, 1696 (1980).
- <sup>47</sup>J. W. Steeds, K. K. Fung, and S. McKernan, *J. Phys. (Paris), Colloq. C3* **44**, 1623 (1983).
- <sup>48</sup>R. V. Coleman, B. Drake, B. Giambattista, A. Johnson, P. K. Hansma, W. W. McNairy, and G. Slough, *Phys. Scr.* **38**, 235 (1988).
- <sup>49</sup>C. G. Slough, B. Giambattista, A. Johnson, W. W. McNairy, and R. V. Coleman, *Phys. Rev. B* **39**, 5496 (1989).
- <sup>50</sup>C. G. Slough, B. Giambattista, W. W. McNairy, and R. V. Coleman, *J. Vac. Sci. Technol. A* **8**, 490 (1990).
- <sup>51</sup>G. Gammie, J. S. Hubacek, S. L. Skala, J. R. Tucker, and J. W. Lyding, *J. Vac. Sci. Technol. B* **9**, 1027 (1991).
- <sup>52</sup>Z. Dai, C. G. Slough, and R. V. Coleman, *Phys. Rev. Lett.* **66**, 1318 (1991); S. Van Smaalen, J. L. De Boer, P. Coppens, and H. Graafsma, *ibid.* **67**, 1471 (1991); Z. Dai, C. G. Slough, and R. V. Coleman, *ibid.* **67**, 1472 (1991).
- <sup>53</sup>R. V. Coleman, Z. Dai, W. W. McNairy, C. G. Slough, and C. Wang, *Appl. Surf. Sci.* **60/61**, 485 (1992).
- <sup>54</sup>J. Ren and M.-H. Whangbo, *Phys. Rev. B* **46**, 4917 (1992).
- <sup>55</sup>A. Prodan, N. Ramšak, V. Marinković, S. W. Hla, F. W. Boswell, J. C. Bennett, and H. Böhm, *Phys. Rev. B* **54**, 10 370 (1996).
- <sup>56</sup>Z. Dai, C. G. Slough, and R. V. Coleman, *Phys. Rev. B* **45**, 9469 (1992).
- <sup>57</sup>R. V. Coleman, Z. Dai, Y. Gong, C. G. Slough, and Q. Xue, *J. Vac. Sci. Technol. B* **12**, 1801 (1994).
- <sup>58</sup>Q. Xue, Z. Dai, Y. Gong, C. G. Slough, and R. V. Coleman, *Phys. Rev. B* **48**, 1986 (1993).
- <sup>59</sup>Y. Gong, Q. Xue, Z. Dai, C. G. Slough, and R. V. Coleman, *Phys. Rev. Lett.* **71**, 3303 (1993).
- <sup>60</sup>Y. Gong, Q. Xue, D. L. Drake, J. Quian, and R. V. Coleman, *Phys. Rev. B* **51**, 12 975 (1995).
- <sup>61</sup>Q. Xue, Y. Gong, D. L. Drake, J. Quian, and R. V. Coleman, *Phys. Rev. B* **53**, 1068 (1996).
- <sup>62</sup>J. A. Wilson, *Philos. Trans. R. Soc. London, Ser. A* **314**, 159 (1985).
- <sup>63</sup>R. Bruinsma and S. E. Trullinger, *Phys. Rev. B* **22**, 4543 (1980).
- <sup>64</sup>N. P. Ong and P. Monceau, *Phys. Rev. B* **16**, 3443 (1977).
- <sup>65</sup>N. P. Ong, *Can. J. Phys.* **60**, 757 (1983).
- <sup>66</sup>G. Grüner, *Comments Solid State Phys.* **10**, 173 (1983).
- <sup>67</sup>A. H. Thompson, A. Zetti, and G. Grüner, *Phys. Rev. Lett.* **47**, 64 (1981).
- <sup>68</sup>A. Zettl, C. M. Jackson, A. Jánossy, A. Grüner, A. Jacobsen, and A. H. Thompson, *Solid State Commun.* **43**, 345 (1982).
- <sup>69</sup>Z. Z. Wang, M. C. Saint-Lager, P. Monceau, M. Renard, P. Gressier, A. Meerschaut, L. Guemas, and J. Rouxel, *Solid State Commun.* **46**, 325 (1983).
- <sup>70</sup>Z. Z. Wang, P. Monceau, M. Renard, P. Gressier, L. Guemas, and A. Meerschaut, *Solid State Commun.* **47**, 439 (1983).
- <sup>71</sup>J. Dumas, C. Schlenker, J. Marcus, and R. Buder, *Phys. Rev. Lett.* **50**, 757 (1983).
- <sup>72</sup>J. P. Pouget, S. Kagoshima, C. Schlenker, and J. Marcus, *J. Phys. (France) Lett.* **50**, L113 (1983).
- <sup>73</sup>M. Ganne, A. Boumara, M. Dion, and J. Dumas, *Mater. Res. Bull.* **20**, 1297 (1985).
- <sup>74</sup>B. T. Collins, K. V. Ramanujachary, and M. Greenblat, *Solid State Commun.* **56**, 1023 (1985).
- <sup>75</sup>R. C. Lacoë, H. J. Schulz, D. Jerome, K. Bechgaard, and I. Johansen, *Phys. Rev. Lett.* **55**, 2351 (1985).
- <sup>76</sup>H. Lüth, *Surfaces and Interfaces of Solid Materials* (Springer, Berlin, 1995), p. 207.