Physics And Chemistry Of Low-dimensional Inorganic Conductors

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Nanostructured and Modulated Low-Dimensional Systems

Albert Prodan, Herman J. P. van Midden, Erik Zupanič, Rok Žitko
Jožef Stefan Institute, Jamova 39, SI-1000 Ljubljana, Slovenia
albert.prodan@ijs.si

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Abstract. Charge density wave (CDW) ordering in $NbSe_3$ and the structurally related quasi one-dimensional compounds is reconsidered. Since the modulated ground state is characterized by unstable nano-domains, the structural information obtained from diffraction experiments is to be supplemented by some additional information from a method, able to reveal details on a unit cell level. Low-temperature (LT) scanning tunneling microscopy (STM) can resolve both, the local atomic structure and the superimposed charge density modulation. It is shown that the established model for $NbSe_3$ with two incommensurate (IC) modes, $\boldsymbol{q_1} = (0,0.241,0)$ and $\boldsymbol{q_2} = (0.5,0.260,0.5)$, locked in at T_1=144K and T_2=59K and separately confined to two of the three available types of bi-capped trigonal prismatic (BCTP) columns, must be modified. The alternative explanation is based on the existence of modulated layered nano-domains and is in good accord with the available LT STM results. These confirm i.a. the presence of both IC modes above the lower CDW transition temperature. Two BCTP columns, belonging to a symmetry-related pair, are as a rule alternatively modulated by the two modes. Such pairs of columns are ordered into unstable layered nano-domains, whose $\boldsymbol{q_1}$ and $\boldsymbol{q_2}$ sub-layers are easily interchanged. The mutually interchangeable sections of the two unstable IC modes keep a temperature dependent long-range ordering. Both modes can formally be replaced by a single highly inharmonic long-period commensurate CDW.

Introduction

The physical concepts for the stability of the charge density waves (CDW) in one-dimensional (1D) conductors were first given by Peierls [1], who predicted a semi-metallic modulated ground state. A small energy gap is opened at the Fermi level (E_F), accompanied by a loss of carriers and by anomalies in the electrical resistivity and the Pauli paramagnetism. The CDW is stabilized if the gain in electron energy more than compensates the increase in the elastic energy. Since $\boldsymbol{k_F}$, the wave-vector at E_F, depends only on the filling of the conduction band, the CDW will in general be incommensurate (IC) with the underlying lattice. Large flat and approximately parallel sheets in the Fermi surface, separated by $\boldsymbol{q} = 2\boldsymbol{k_F}$ are responsible for a so-called nesting instability and the formation of a CDW.

An interesting property connected with CDWs is their sliding under the application of an external electric field. The effect is attributed to the ability of a CDW to overcome its pinning to the lattice and to the impurities [2]. The relatively small group of 1D compounds exhibiting such sliding includes in addition to the most thoroughly studied $NbSe_3$ and its isostructural monoclinic polymorph m-TaS_3 [3] also NbS_3 [4], $(TaSe_4)I$ [5], $(NbSe_4)_{10}I_3$ [6] and the "blue bronzes" $A_{0.3}MoO_3$ with A = K, Rb, Tl [7-10].

The monoclinic room-temperature (RT) basic structure of $NbSe_3$ (a = 1.0006 nm, b = 0.3478 nm, c = 1.5626 nm, β = 109.30°, space group $P2_1/m$) [11] is reproduced in Fig. 1. The unit cell contains three types of symmetry-related pairs of bi-capped trigonal prismatic (BCTP) columns with all type-I and type-III Nb-Se distances very similar (between 0.26 nm and 0.27 nm), while the inter-column Nb-Se bonds of the type-II BCTPs appear slightly longer (about 0.29 nm). Nb chains in Se cages form columns, aligned parallel to the monoclinic $\boldsymbol{b_0}$-direction. Strongly bonded corrugated layers, parallel to the $\boldsymbol{b_0}$-$\boldsymbol{c_0}$ monoclinic plane, are separated by wide Van der Waals (VdW) gaps (with Se-Se inter-column distances between 0.37 and 0.41 nm). The $NbSe_3$ basic structure is very specific:

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