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The unusual charge-density wave in the monoclinic phase of NbS₃

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We are reporting on the charge-density wave (CDW-2) forming in NbS₃, phase II, at the temperature $T_{P2}=150$ K. The transition is found only in the sulfur deficient samples. The CDW-2 is clearly detected in the resistance and in the collective transport measurements, but does not show any lattice distortions at T_{P2} in the diffraction studies. However, features near T_{P2} are clearly detected in the temperature dependences of the ⁹³Nb NMR relaxation time and in XANES (X-ray absorption near-edge spectroscopy) spectra measured at S and Nb peaks. Anomalous photoresponse of CDW-2 is also reported. Among others, the concept of excitonic dielectric is considered as the origin of CDW-2.

Introduction

The monoclinic phase of NbS₃, known also as phase II, shows two coexisting charge-density waves (CDWs) at room temperature, T . The corresponding lattice distortions are characterized by the wave-vectors $\mathbf{q}_1 = (0.5, 0.298, 0)$ and $\mathbf{q}_0 = (0.5, 0.352, 0)$ [1]. The \mathbf{q}_1 -CDW (CDW-1) emerges at $T_{P1}=340-370$ K and shows the features typical of the collective electronic transport. Evidence for the \mathbf{q}_0 -CDW (CDW-0) transport has been also shown recently, T_{P0} has been tentatively estimated as 620-650 K [2].

Here we are reporting on the unusual features of the CDW forming at $T_{P2}=150$ K (CDW-2).

The results and discussion

The appearance of CDW-2 and its collective transport are clearly detected in the transport studies. The T dependence of conductivity, σ , shows a distinct feature at T_{P2} ; the voltage, V , dependences of σ below T_{P2} show a drop at the threshold value, V_1 [2]. However, no lattice distortions at T_{P2} have been detected in the diffraction structural studies (TEM and powder X-ray). Nevertheless, features near T_{P2} have been clearly detected in the

temperature dependences i) of the nuclear transverse magnetization relaxation time measured at the central ⁹³Nb NMR line and ii) of XANES (X-ray absorption near edge structure) spectra measured at two S and one Nb peak [2].

Under RF irradiation sliding CDW-2 shows Shapiro steps, – a typical feature of CDW transport. The RF interference shows that the charge density carried by CDW-2 per one wavelength can be from 3 to 1000 times lower than that of CDW-1. This is too low for a usual CDW formed through the Peierls transition. The charge density scales with the sample specific conductivity, σ_s , above T_{P2} . The composition studies of NbS₃ samples have shown that CDW-2 is observed only in S deficient samples, which show relatively high σ_s . Therefore, CDW-2 is concluded to be a condensate of electrons donated by S vacancies [2]. This is the first known case of doping-induced electrons condensation into a collective state.

One of the surprising features of CDW-2 is the variation of the $\sigma(T)$ curves with the change of σ_s (for $T>T_{P2}$). The value of T_{P2} does not depend on σ_s , and changes in σ_s within 1.5 orders of magnitude result

only in vertical shift of the $\sigma_s(T)$ curve in the logarithmic scale. Thus, the transition at T_{p2} is not directly coupled with the concentration of the electrons induced by doping [2].

We have also observed indications of the purely electronic nature of the CDW-2, while the lattice distortion, if any, plays a secondary role. In contrast to CDW-1, both T_{p2} value and the I-V curves of CDW-2 show very low sensitivity to the uniaxial strain, ε , of the samples [2]: $d\log(T_{p2})/d\varepsilon \sim 0.1$, $d\log(T_{p1})/d\varepsilon \sim 1.5$, which is a very low value for a CDW, while the non-linear conductivity does not show any regular changes for ε in the range 0–1%. The critical slowing-down of fluctuations revealed by NMR near T_{p2} is also in accord with the electronic nature of the distortion: the relaxation of the transverse magnetization can be stimulated by fluctuations of both magnetic and electric fields, and its freezing can reveal not only a lattice but also an electronic ordering. This would explain, why no lattice distortion is observed at T_{p2} with diffraction methods.

Another anomalous feature of NbS₃ is the effect of infra-red (IR) irradiation on the CDW-2 transport [3]. Earlier the impact of light on the CDW transport has been reported for K_{0.3}MoO₃ [4] and o-TaS₃ [5]. In both cases the threshold field, E_t , for the CDW depinning grows under optical irradiation. The effect was explained in terms of the CDW rigidity: the optically excited electrons are supposed to decrease the CDW elastic modulus and increase E_t .

To study photoconductivity in NbS₃ we used IR LED with the frequency corresponding to $h\nu=1.2$ eV, which is above the known energy gap, 0.43 eV [6]. In case of CDW-1 the V_t value is found to increase with the power of irradiation, W , like in [4,5]. The V_t variation was too small to establish the dependence of V_t vs. W .

In contrast, CDW-2 shows a *decrease* of V_t vs. W ; $\delta V_t \propto W^{1/2}$ (Fig. 1). For the maximum W values δV_t is of the order of V_t at $W=0$, while $V_t(W)$ is non-linear even for the lowest values of W , when $\delta V_t \ll V_t(0)$. Presumably, changes of the CDW-2 rigidity [4,5] result in a linear $V_t(W)$ dependence for small W . Thus, the behavior of CDW-2 under the irradiation indicates its specific interaction with the pristine lattice.

The increase of the linear conductivity under the IR irradiation also follows the $W^{1/2}$ dependence [3].

To explain the origin of CDW-2 we considered the mechanisms of Wigner crystallization and Keldysh-Kopaev transition (formation of excitonic dielectric)

[2]. These orderings are primarily of electronic nature.

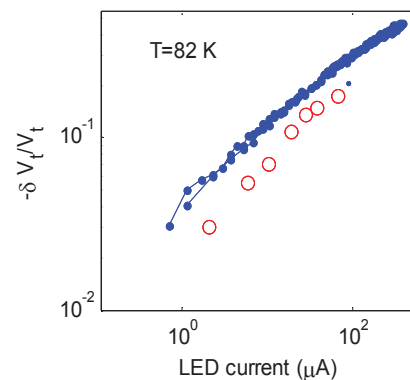


Fig. 1. The relative V_t drop vs. W . The sample dimensions are $45\mu\text{m} \times 0.014\mu\text{m}^2$. $100\mu\text{A}$ corresponds to $\sim 1\text{ mW/cm}^2$. ‘o’ – data taken from the I-V curves matching. ‘•’ – from $\sigma(W)$ at fixed V .

The concept of excitonic dielectric is more favorable, because it explains the low sensitivity of T_{p2} on the concentration of electrons condensed into CDW-2. In the limiting case of zero concentration of the donated electrons the transition persists: the CDW-2 is formed by coupling of equal numbers of electrons and holes, as is suggested for the “classical” Keldysh-Kopaev transition. Alternatively, the CDW-2 can emerge at stacking faults in NbS₃. This hypothesis seems to be supported by the first STM studies of this compound.

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