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10 gigahertz-range synchronization at room temperature in nano-crystals of the quasi-one-dimensional conductor NbS_3

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Nano-crystals of quasi one-dimensional conductor NbS_3, phase II, have been synthesized. The samples show a charge-density wave (CDW) transition at 365 K. The crystals show sharp threshold field and coherent CDW transport revealed by the AC-DC coupling – the Shapiro steps. The thinnest samples (cross-section down to $5 \cdot 10^{-4}$ μm^2) show the Shapiro steps under at least 16 GHz irradiation at room temperature.

NbS_3, phase II, is standing out for its extremely high Peierls transition temperature, T_P= 330 - 355 K [1]. Studies of NbS_3–II are few in number because of the difficulties with its synthesis. The number of NbS_3 polytypes is not quite clear. The most reproducible in synthesis is phase I (triclinic), high-ohmic at all T; it does nor show CDW transport. Phase II is monoclinic with 8 Nb chains per unit cell and shows approximate trimerisation below T_P= 340-355 K [1,2]. Its room temperature resistivity is of the order of 10^{-1} Ωcm. This phase is remarkable for the room-temperature CDW transport. Phase III [3] is characterized by slightly different crystalline structure and shows nonlinear conductivity below $T_P \approx 150$ K. It is not clear, however, if phases II and III are essentially different, as their resistivity was measured in different temperature ranges.

In the previous paper [4] we reported successful synthesis of the conducting phase (II). The samples were grown from the vapor phase by direct reaction of Nb and S in mole ratio 1:3 with a 10% excess of sulfur. The growth continued for two weeks in a quartz tube. The temperature gradient over the tube length (20 cm) was 665-715 C.

We studied transport properties of NbS_3 whiskers in wide ranges of temperatures (4 – 400 K) and samples cross-sections (10^{-3}-20 μm^2). For bulk samples (10^{-1}-20 μm^2) temperature dependences of resistance demonstrated two Peierls transitions at 365 K and 150 K. For thinner samples (10^{-3} -10^{-1} μm^2) the Peierls transition at 150 K tends to vanish.

Investigations of non-linear conductivity (differential resistance *vs.* voltage) also showed the existence of two temperature regions, where the onset of non-linear conduction is marked with a sharp threshold. The data argue for development of two CDWs: below 365 and 150 K. This is the 1st demonstration of so sharp depinning of the CDWs in NbS_3. For thick samples the lowest value of the threshold field, E_t, is about 2 V/cm, and for the thinnest ones E_t is above 100 V/cm, in consistence with size effects known for other CDW compounds [5]. For the low-T CDW E_t is somewhat larger.

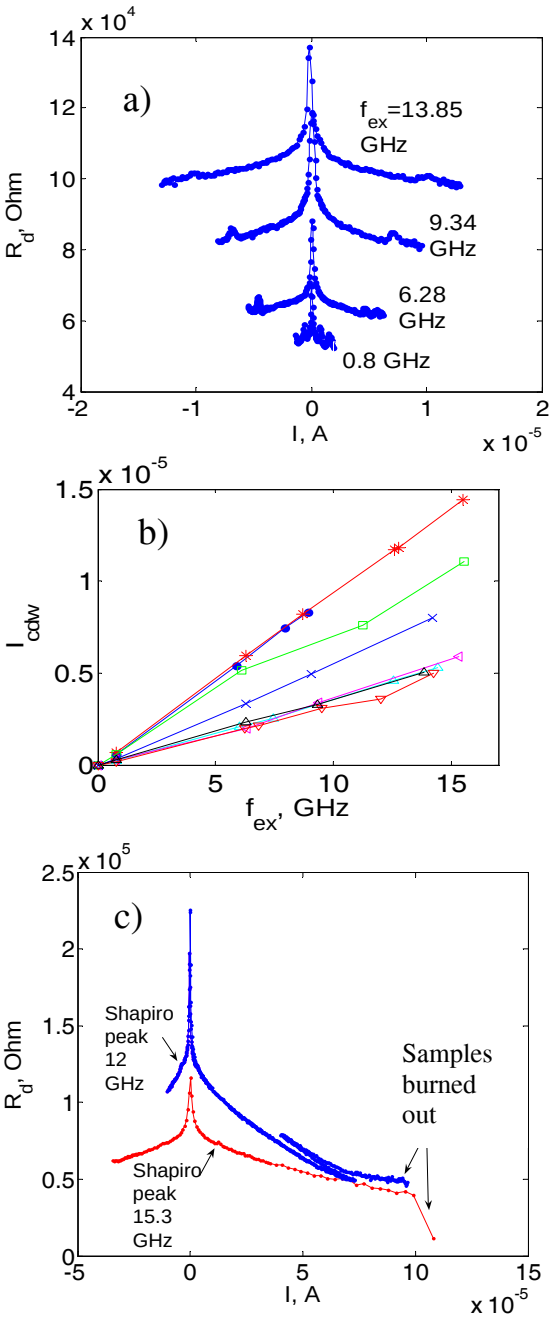

Figure 1. a)- R_d *vs.* I dependencies for NbS_3 nanobridge at different radiation frequencies and room temperature; b)- the dependences of the CDW current corresponding to the 1-st Shapiro step on the external frequency at room temperature; c) R_d vs. I dependencies at utmost currents at room temperature.

The CDW motion can be synchronized by external RF or microwave irradiation. In the case 100% synchronization the CDW velocity is (nearly) constant for a certain voltage range, and the so-called Shapiro steps of (nearly) zero differential conductivity of CDW appear on the I-V curves. Under the synchronization the CDW current, I_{CDW}, and the irradiation frequency, f, are related as $f=f_0 n/p$ (n, p – integer), and f_o is the so-called fundamental frequency,

Equation 1. $f_o=I_{CDW}/MeN$,

where e is the elementary charge, $M\approx 2$ is the number of electrons per CDW wavelength, λ, and N is the total number of conducting chains. The Shapiro steps provide unambiguous evidence of the CDW transport, and we have observed them for both CDWs of NbS_3. For the upper CDW, i.e. at room temperature, synchronization up to 50% at frequencies up to 4 GHz was achieved [4]. It should be noted that 100% synchronization was observed for blue bronze [6] and $NbSe_3$ [7] only. Knowing cross-sections of the samples and the area of the unit cell of NbS_3 we can calculate the number of conducting chains per unit cell contributing to the CDW transport basing on the relation (1). This value was calculated for 11 samples and varied between 0.3 and 1.2 [4]. This result indicates that at most 2 chains contribute to the CDW transport – one for each CDW state.

Thus NbS_3 (II phase) has been successfully synthesized. The samples demonstrate the highest temperature of the Peierls transition, 365 K. Another Peierls transition, at 150 K, was shown to be the feature of the same phase of NbS_3, in contrast to the earlier conception [4].

Preparation of 1 μm-long microbridges with cross-section down to $5\cdot10^{-4}$ μm^2 allowed us to study Shapiro steps in the 10 GHz range since the short bridges provide better load matching of the high-frequency generator with samples. All measurements were done at room temperature. Figure 1a shows typical dependencies of differential resistance vs. current at different radiation frequencies. Well-defined Shapiro steps (pronounced maxima of R_d) can be seen. Figure 1b justifies validity of the fundamental ratio, f_0/I_{CDW}, (Eq. 1) up to 16 GHz for 8 samples. To our knowledge, this is the 1st observation of CDW synchronization at such high frequencies and room temperature, and we have no indications of approaching a limit [8]. This result proves that the periodic pinning potential does not vanish at high electric fields corresponding to the CDW velocities of the order of 10 m/s. Technically, it is very difficult to apply the utmost synchronizing frequencies to the nano-bridges. However we can evaluate them from the maximal currents (Eq. (1)) flowing through nanobridges. R_d vs I dependencies at utmost currents are shown in fig. 1c. Simultaneously we have irradiated these nanobridges. One can see Shapiro steps under irradiation at 12 and 15.3 GHz. Then, the highest current density, ~ $6\ 10^6$ A/cm^2, corresponds to the CDW velocities ~200 m/s and fundamental frequencies ~200 GHz.

What are the advantages of NbS_3 which allow observation of the synchronization at the utmost frequencies? This compound is unique in the sense that it shows highly coherent CDW transport with low density of conducting chains and, consequently, low density of energy dissipation. The highly anisotropic structure of NbS_3 allows synthesis of whiskers down to nanometer dimensions retaining their CDW properties. Both features enable passing extremely high current densities through the samples without heating. Another reason of high synchronization frequencies may be the intrinsic properties of NbS_3, that is, the high characteristic frequencies. According to the rigid overdamped oscillator model [9] the high characteristic frequencies $\sim E_t e\tau/(\lambda m^*)$ can be associated with the high threshold fields ($E_t \geq 100$ V/cm for the nanometer samples) and low friction of CDW ($1/\tau$). Indeed, one can see that Shapiro peaks on differential curves (Fig. 1a)) at frequencies above 10 GHz begin to smear out. It means that characteristic frequencies of NbS_3 could be on the order of 10 GHz.

In conclusion, we have presented the 1st demonstration of highly coherent CDW transport for nano-sized samples at room temperatures. These samples detect GHz-range radiation showing Shapiro steps. The results argue in favor of single-phase composition of the NbS_3 samples, though structural studies are desirable. In general, the room-temperature CDW at our disposal can be the ground for new consideration of all the numerous potential applications of this unique collective electronic state.

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