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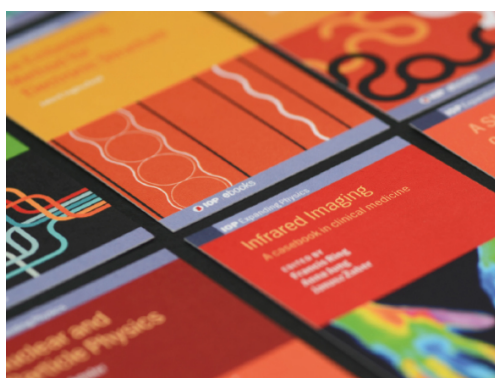
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Superconductivity at High Pressure in NbSe₃.

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Abstract. – We have measured the electrical resistivity of NbSe₃ between 2 K and room temperature up to a pressure of 7.2 GPa. At $P_1 = 3.5$ GPa we observe the extinction of the high-temperature charge density wave (T_1 -CDW) and the enhancement of the superconducting critical temperature T_c to ≈ 5 K. The logarithmic pressure slopes of T_1 ($P < P_1$) and T_c ($P > P_1$) are found to be practically equal. A similar behaviour had been reported previously at lower pressures for T_2 ($P < P_2$) and T_c ($P_2 < P < P_1$) in the distorted state. We discuss these results in terms of an anisotropic superconducting state in NbSe₃, with different gaps associated with different types of chains.

Charge density wave (CDW) transitions are a widespread phenomenon in low-dimensional materials [1]. The existence of a strong anomaly in the response function of the electron gas coupled to the lattice leads to a structural distortion accompanied by a gap in the electronic spectrum. In a mean-field calculation the transition temperature is given by

$$T_{\text{CDW}} = 5.43T_{\text{F}} \exp[-1/\lambda_c] \approx 5.43T_{\text{F}} \exp[-\omega_q/g^2N(E_{\text{F}})], \quad (1)$$

where T_{F} is the Fermi temperature, ω_q is the bare phonon energy corresponding to the softened phonon, g^2 is the coupling constant between carriers and phonons, and $N(E_{\text{F}})$ is the electronic density of states at the Fermi energy. The application of pressure has a strong effect on the CDW. The pressure hardening of the bare phonon energies can render the distortion less favourable energetically, resulting in a lowering of T_{CDW} as can be deduced from (1). Furthermore, the decrease in volume enhances the coupling between the existing 1D chains (or 2D layers), reducing the density of states $N(E_{\text{F}})$ that contributes to the charge density wave, and therefore T_{CDW} . In this way, a sufficiently high pressure can cause the destruction of the CDW. The disappearance of the distortion allows the development of a superconduct-

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ing state whose transition temperature is now given by [1]

$$T_c = 1.44\theta_D \exp[-1/\lambda_s] = 1.44\theta_D \exp[-1/N(E_F)V], \quad (2)$$

where V is the electron pairing potential. This description has been verified in, *e.g.*, 2D transition metal dichalcogenides [2] and 1D organic materials [3] (where, instead of a CDW, a spin density wave occurs). NbSe_3 is a particularly interesting case as it actually develops *two* CDWs at $T_1 = 145$ K and $T_2 = 59$ K, respectively [1]. In agreement with band structure calculations [4], nuclear magnetic resonance experiments [5] or measurements of the depinning electric fields [1] have evidenced that the two CDWs are essentially independent and noninteracting. The origin of this behaviour lies in the fact that NbSe_3 has three pairs of nonequivalent trigonal prismatic chains. From electronic-band theory it has been concluded that the high-temperature CDW with wave vector $\mathbf{q}_1 = (0, 0.24177, 0)$ develops in the bands evolved from the so-called type-III chains [4], while the low-temperature one with $\mathbf{q}_2 = (0.5, 0.26038, 0.5)$ originates in bands due to type-I chains. Previous experiments at moderate pressures [6] had shown that the T_2 -CDW is destroyed at a critical pressure P_2 of about 0.6 GPa while superconductivity appears, with T_c rising rapidly to 3.3 K. Recently, more detailed measurements [7] have emphasized that the logarithmic pressure slope of T_2 is approximately the same as that of T_c . Based on these observations, their authors suggested that both the T_2 -CDW and the superconducting states that develop above P_c occur on the same (type-I) chains. The question naturally arises whether the same conclusions apply to the T_1 -CDW. In order to answer it experimentally, it is necessary to completely suppress the latter distortion by going to higher pressures. This was achieved in the experiments described hereafter, allowing a comparison of the effects associated with each CDW.

The experimental set-up has been described in detail in ref. [8]. The pressure is generated by tungsten carbide anvils. The retaining gasket is made out of pyrophyllite and the sample is embedded in steatite, which provides «quasi-hydrostatic» conditions. The pressure is measured at low temperature by monitoring the superconducting transition of a lead strip connected in series with the sample. The pressure inhomogeneity within the cell is estimated to be $(0.3 \div 0.1)$ GPa, from the width of the latter transition. From independent measurements on ZrV_2 , a material extremely sensitive to uniaxial stress, we also estimate the latter to be typically less than 0.5% of the applied pressure at $P = 7$ GPa. The cell is loaded at room temperature, then slowly cooled down in a simple helium cryostat. The dimensions of the fiber-shaped samples were approximately $50 \mu\text{m} \times 200 \mu\text{m} \times 1 \text{mm}$. Due to this rather poorly defined geometry, no attempt was made to calibrate the resistivity in absolute units.

The resistance measured at different applied pressures is shown in fig. 1a). Note that the lowest pressure already exceeds the critical value for suppressing the T_2 -CDW. We observe that all the curves present a linear dependence down to T_1 . Up to 3.6 GPa, the transition at T_1 is signalled by a small bump corresponding to the condensation of carriers under the T_1 -CDW. This anomaly shifts rapidly to lower temperatures as P increases, to finally disappear above 4 GPa. Figure 1b) shows the detail of the low-temperature region, in particular the various superconducting transitions. The width of these transitions is imputable to the pressure inhomogeneity, as discussed above.

The resulting pressure variation of T_c is shown in fig. 2, together with previous data from ref. [7] at lower pressures. The corresponding values of T_1 and T_2 have also been plotted on the same, semi-logarithmic, scale. The main feature of the data is a strong enhancement of the superconducting critical temperature in the vicinity of P_1 , from a minimum of ≈ 2.2 K near 1.7 GPa (from [7]), up to ≈ 5 K at 4 GPa. Above this pressure, $\ln T_c$ decreases again with a slope of $\approx -0.16 \text{ GPa}^{-1}$, substantially weaker than that reported in ref. [7] for the distorted state ($P_2 < P < P_1$). This difference can be related to the slower pressure decrease of $\ln T_1$ as compared to $\ln T_2$. Recalling that, in the results of Ido *et al.*, the pressure slopes of $\ln T_2$

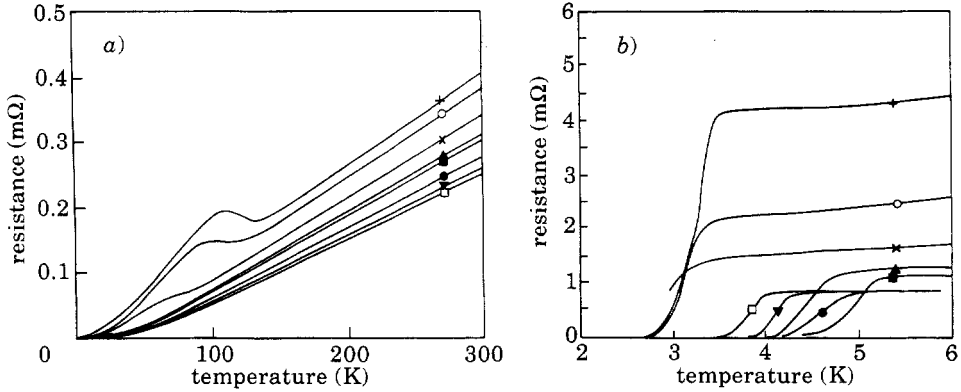


Fig. 1. - *a*) Resistivity of NbSe₃ as a function of temperature for different pressures. *b*) Resistivity measurements showing the superconducting transition of NbSe₃ for different pressures. In both *a*) and *b*) notations are: + 0.68 GPa; ○ 1.23 GPa; × 2.6 GPa; ▲ 3.6 GPa; ■ 4 GPa; ● 5.6 GPa; ▼ 7 GPa; □ 7.2 GPa.

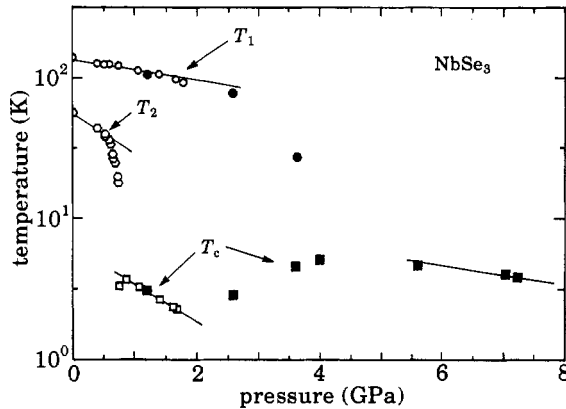


Fig. 2. - Pressure dependence of the transition temperature of the high- and low-temperature charge density waves, T_1 and T_2 , respectively, and of the superconducting transition temperature T_c . Full symbols, our data; open symbols, data of ref. [7].

(below P_2) and $\ln T_c$ (between P_2 and P_1) were practically equal, we can see in fig. 2 that the sample holds approximately true here for those of $\ln T_1$ (below P_1) and $\ln T_c$ (above P_1). It is thus tempting to assume that T_2 and T_c (distorted) on the one hand, T_1 and T_c (undistorted) on the other hand have a common physical origin and that, above P_1 , the superconducting phase arises from the electron states of the same (type-III) chain that is involved in the T_1 -CDW formation, just as superconductivity in the distorted state was associated with the T_2 -CDW (*i.e.* type-I chains). If this is true, the equality of the logarithmic pressure coefficients of T_{CDW} and T_c follows directly from the mean-field equations (1) and (2), provided the dominant terms in their derivatives,

$$\frac{d \ln T_{CDW}}{dP} = \frac{1}{T_F} \frac{dT_F}{dP} - \frac{1}{\lambda_c^2} \frac{d\lambda_c}{dP}, \quad (3)$$

$$\frac{d \ln T_c}{dP} = \frac{1}{\theta_D} \frac{d\theta_D}{dP} - \frac{1}{\lambda_s^2} \frac{d\lambda_s}{dP}, \quad (4)$$

be those corresponding to the electron-phonon coupling. This assumption is plausible since neither T_F , nor θ_D , normally exhibit such large pressure variations like those reported here. However, one should keep in mind that the presence of a CDW distortion can affect both the phonon spectrum and, even more likely, the electron density of states near E_F , so that the pressure variations of the different terms cannot be regarded as uncorrelated. A more fundamental shortcoming of using eqs. (1) and (2) is that both of them apply to the weak-coupling limit of the electron-phonon interaction. There is growing evidence that intermediate-coupling ($\lambda \sim 1$) polaronic descriptions might be more relevant to the crossover regime implied by the CDW-to-superconducting transition [9]. Whereas this theoretical issue presently remains unsettled, its solution will not affect the main result of this study, namely the continuity in the pressure coefficients of T_{CDW} and T_c on each side of the transition.

Coming back to the results in fig. 2, we wish to point out that there exists a major difference between the transitions at P_1 and P_2 ; the enhancement of $T_c(P)$ begins already around 2.5 GPa, *i.e.* substantially below P_1 , whereas the appearance of superconductivity took place quite suddenly just above the critical pressure P_2 , where T_2 vanished [7]. Material and/or pressure inhomogeneity may cause some broadening, but the present effect is too large to be explained on this basis only. It can be remarked that the pressure drop of T_2 was also much steeper than is found here for T_1 , which brings additional support to our view that both CDW and superconducting phenomena go hand in hand. One possible explanation is that the portions of the Fermi surface involved in the formation of the higher CDW have a less pronounced one-dimensional character than those affected by the lower one, allowing carriers to be gradually released from the gap prior to the transition. This assumption is based on recent X-ray results of Moudouen *et al.* [10], which showed the existence of 2D correlations on the type-III chains.

Finally, we note that there is initially a strong pressure decrease of the residual resistivity R_0 , then the variation seems to level off above P_1 . This effect cannot be attributed to electrical contact problems, nor to changes in the geometrical factor (*e.g.*, due to cracks within the sample closing gradually as P increases), since the room-temperature resistivity exhibits a distinct, more continuous behaviour. The decrease of R_0 can basically result from: i) an increase of the carrier density and/or ii) a decrease of their effective masses. The density of carriers gapped by the lattice distortion at T_1 can be estimated from the relative size of the resistivity anomaly $\alpha_1 = (R_M - R_N)/R_N$, where R_M is the value of the resistance at the maximum and R_N is the value of the normal resistance at the same temperature extrapolated from above T_{CDW} . The values that we obtain for $\alpha_1(P)$ are constant almost up to the critical pressure, and coincide with those of Ido *et al.* at lower pressures. The final drop of α_1 very close to P_1 (no gapped carriers are left above this pressure) might be due to interchain couplings as suggested by the latter authors. Hall effect measurements under pressure would be useful to confirm this interpretation.

In conclusion we observed that high pressures destroy the T_1 -CDW of NbSe₃ at about 3.5 GPa, causing a rise of its superconducting transition temperature T_c to about 5 K. The analysis of the pressure variation of T_c suggests that this new superconducting state is closely related to the T_1 -CDW, *i.e.* arises from the same set of chains where the corresponding distortion takes place. Recalling that the superconducting state below 3.5 GPa had been shown to arise primarily from the T_2 -CDW chains, we can describe NbSe₃ as an anisotropic superconductor, with possibly two weakly interacting superconducting states associated with the two CDWs. In order to better understand the effect of pressure in the CDW state, in particular to confirm the absence of carrier release from the CDW gap, it is desirable to get a direct estimate of the P -dependence of k_F , for instance by following the incommensurate satellites

as a function of pressures by neutron diffraction. Experiments are in preparation to clarify this point.

* * *

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