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Superconductivity under pressure in $Ba_{1-x}K_xBiO_{3-y}$

V G Tissen, E G Ponyatovskii, M V Nefedova, L A Klinkova and N V Barkovskii
Institute of Solid State Physics, 142432 Chernogolovka, Moscow district, Russia

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Abstract. Measurements of the superconducting transition temperature T_c of $Ba_{0.62}K_{0.38}BiO_{3-y}$ with $T_c = 27.5$ K (sample 1) and $Ba_{0.61}K_{0.39}BiO_{3-y}$ with $T_c = 17.6$ K (sample 2) have been performed under a pressure up to 10 GPa by the inductive method. With increasing pressure, T_c of sample 1 initially increases, then has a maximum and finally decreases down to 6 K at 9.6 GPa. An irregularity in the dependence of T_c versus P is observed at about 8 GPa. For sample 2, $T_c(P)$ demonstrates a qualitatively similar behaviour with an additional dip in the low-pressure range. The suppression of T_c is explained within the framework of the Bilbro–McMillan model.

1. Introduction

The discovery of superconductivity in $Ba_{1-x}K_xBiO_3$ has received considerable attention since the compound with $x \simeq 0.4$ exhibits the highest superconducting transition temperature $T_c \simeq 30$ K yet observed for oxides not containing copper [1–3]. There are several crystal structures in this system, depending on x and temperature, but superconductivity occurs only in the cubic phase while the others demonstrate non-metallic behaviour [4]. Pressure as an additional variable is often used to obtain more information about various properties of solids at ambient temperature through their variations when subjected to compression. The increase in T_c with increasing P for samples of $Ba_{1-x}K_xBiO_3$ with different compositions was found within the pressure range below 1 GPa [5–7]. At the same time, the optical reflectivity in the near-infrared and visible regions for $Ba_{0.6}K_{0.4}BiO_3$ was reported to decrease rapidly with increasing P up to 40 GPa, thereby indicating broadly the loss of metallic character of the sample [8]. Therefore the depression of T_c could be expected at a sufficiently high pressure. Indeed, the change in the pressure coefficients of T_c from positive to negative values for $Ba_{0.62}K_{0.38}BiO_3$ has recently been revealed at about 4 GPa in AC susceptibility measurements up to 8 GPa [9]. The aim of the present work is to study the pressure dependence of T_c in single crystals of $Ba_{1-x}K_xBiO_{3-y}$ up to such a pressure which shifts T_c close to the low-temperature limit (4.2 K) of the experimental set-up. The data on the destruction of superconductivity may shed light on the conditions for its occurrence in this system.

2. Experimental details

Single crystals of $Ba_{1-x}K_xBiO_{3-y}$ were grown by the electrochemical method as described previously [10]. In this work, $Ba_{0.62}K_{0.38}BiO_{3-y}$ with $T_c = 27.5$ K (sample 1) and

$Ba_{0.61}K_{0.39}BiO_{3-y}$ with $T_c = 17.6$ K (sample 2) were examined. The difference between T_c for the two samples is thought to be due to the considerable oxygen vacancies contained in sample 2. High pressures were generated in the diamond anvil cell whose body and other metallic parts were made of non-magnetic alloy. Anvils with 0.5 mm culet diameter are mounted on the sapphire backing plates in order to decrease the inductive coupling between the coil system used for AC susceptibility measurements and the metallic surroundings. Gaskets pre-indented to 0.06 mm from an initial thickness of 0.3 mm were used. The sample with typical dimensions 0.08 mm $\times$ 0.08 mm $\times$ 0.03 mm and small ruby chips were loaded in the hole of 0.15 mm diameter. A methanol–ethanol mixture in the ratio 4:1 served as a pressure-transmitting medium. The cell can be cooled to 4.2 K in a glass Dewar vessel. The temperature was measured with a (Cu–Fe)/Cu thermocouple. We estimate the accuracy to be ± 0.2 K. The pressure was determined at room temperature by the ruby fluorescence method after cooling the cell under pressure to 4.2 K followed by heating. The superconducting transition was detected by measuring an AC magnetic susceptibility χ as a function of temperature. Data were taken during the heating process. The coil system consists of two coils. The measuring coil system is positioned symmetrically around the anvils and the reference system is placed near it. The two coil systems are parts of a bridge. After additional compensation the signal from the secondary coils is detected with a lock-in amplifier.

3. Results and discussion

The temperature dependence of χ is displayed at several pressures for sample 1 in figure 1. With the exception of the $\chi(T)$ curve for $P = 1.9$ GPa not shown here for clarity, the superconducting transition is clearly shifted downwards with slight broadening by pressure. It is also obvious that measurements were terminated at 9.6 GPa due to the incompleteness of the transition. As can be seen by a comparison of the data in figures 1 and 2, sample 2 exhibits a narrower width of the superconducting transition at atmospheric pressure than sample 1 does. The application of pressure, however, considerably increases the width and reduces the amplitude of the transition. One of the possible interpretations for these results is that the oxygen distribution over sample 2 becomes inhomogeneous under pressure, thereby leading to a gradual formation of the non-superconducting phase. Such an explanation is in line with the assumption about a possible deficiency of oxygen in this sample.

T_c is defined by the onset of the superconducting transition as indicated by an arrow in figure 1. The pressure dependences of T_c for both samples together with the data published previously [9] are shown in figure 3. Despite a more complicated behaviour of T_c versus P for our samples, it appears that no discrepancies exist between these and the reported data. Taking into account the difference in T_c, the occurrence of an anomaly such as the pronounced dip observed at about 8 GPa on the $T_c(P)$ curve for sample 1 could be anticipated at still higher pressure for the sample examined previously. For sample 2, the anomaly on the $T_c(P)$ curve in the lowest-pressure range can be explained as the result of competitive processes: one increasing T_c with increasing pressure in the manner usually observed for superconducting $Ba_{1-x}K_xBiO_3$ and the other decreasing T_c due to a formation of a non-superconducting phase. Other features observed for sample 2 at high pressure are, in principle, similar to those for sample 1, although the multiphase state of the sample naturally distorts the picture.

The non-metallic behaviour of $Ba_{1-x}K_xBiO_3$ over a wide range of x has been explained by the formation of charge density waves (CDWs), either long range or local, depending on the lattice structure [4]. The suppression of superconductivity and metal-like reflectivity

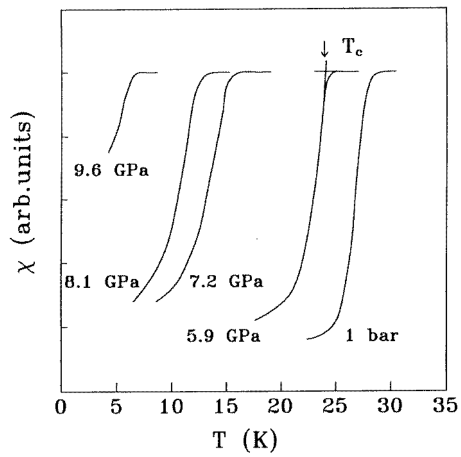

Figure 1. Temperature dependence of AC susceptibility for sample 1 of $Ba_{0.62}K_{0.38}BiO_{3-y}$ at different pressures. The onset of the superconducting transition is indicated by an arrow.

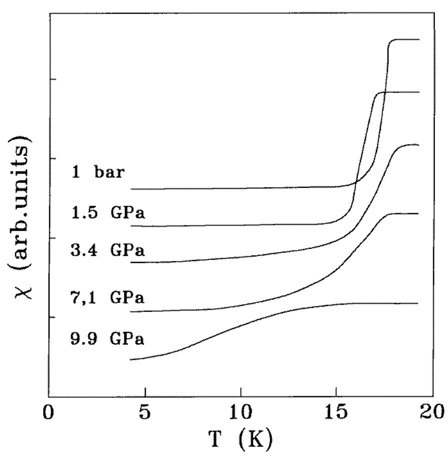

Figure 2. Temperature dependence of AC susceptibility for sample 2 of $Ba_{0.61}K_{0.39}BiO_{3-y}$ at different pressures.

in $Ba_{1-x}K_xBiO_{3-y}$ appears to occur continuously with increasing pressure, in contrast with the abrupt disappearance of superconductivity with decreasing x. Therefore, it is reasonable to assume that the destruction of superconductivity under pressure is due to a continuous phase transition accompanied by the CDW. We further use the Bilbro–McMillan [11] model which gives some predictions concerning the coexistence of the superconductivity and the CDW. The structural transition, which is considered to be of second order, opens a Peierls-like energy gap in the electronic spectrum, thereby reducing the density of states of the Fermi level and T_c. According to this model, the equation, which relates T_c to the structural transition temperature T_m, is

$$T_c^{N_2(\epsilon_F)/N(\epsilon_F)} T_m^{1-N_2(\epsilon_F)/N(\epsilon_F)} = T_{C0} \quad (1)$$

where $N_2(\epsilon_F)$ and $N(\epsilon_F)$ are the densities of states at the Fermi level in the presence of the Peierls gap and in the absence of the Peierls gap, respectively. T_{C0} is the superconducting

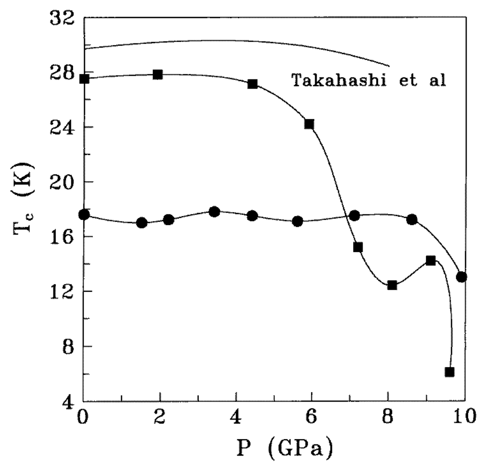

Figure 3. Pressure dependence of T_c for sample 1 (■), sample 2 (●) and a crystal of $Ba_{0.62}K_{0.38}BiO_{3-y}$ from [9]: ——, guides to the eye.

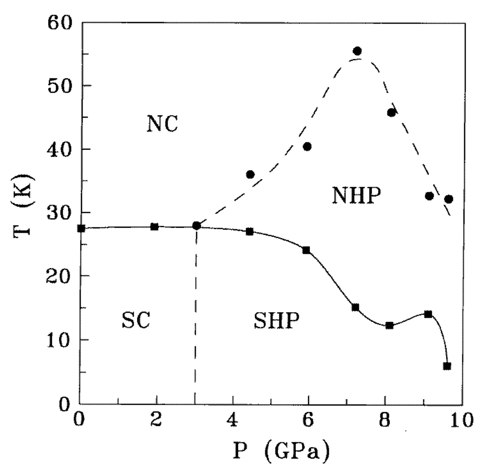

Figure 4. Tentative phase diagram of $Ba_{0.62}K_{0.38}BiO_{3-y}$ (sample 1): NC, normal cubic phase; SC, superconducting cubic phase; NHP, normal high-pressure phase; SHP, superconducting high-pressure phase.

transition temperature in the absence of the Peierls gap. To obtain numerical results for sample 1, the following assumptions are used.

(1) T_{C0} is pressure independent.

(2) The structural transition occurs first at pressure P_0 where T_c begins to drop. We take $P_0 = 3$ GPa.

(3) The expression for the pressure dependence of $N_2(\epsilon_F)/N(\epsilon_F)$ has the simplest parabolic form given by

$$N_2(\epsilon_F)/N(\epsilon_F) = (P_c^2 - P^2)/(P_c^2 - P_0^2) \tag{2}$$

where P_c is the pressure at which the sample becomes insulating. Our rough estimation gives $P_c = 10$ GPa.

Calculated values of T_m as a function of pressure are included in the tentative P–T phase diagram for $Ba_{0.62}K_{0.38}BiO_{3-y}$ shown in figure 4. It seems that the irregularity on the $T_c(P)$ curve at about 8 GPa and the maximum of T_m may be related to each other. X-ray diffraction measurements of this compound in the high-pressure low-temperature range would be very desirable to check the proposed phase diagram. At present, we can only speculate about the possible crystal structure of a hypothetical high-pressure phase. As mentioned above, all the phases in $Ba_{1-x}K_xBiO_3$, except for the cubic phase, are insulating at ambient pressure. However, there is a system closely related to $Ba_{1-x}K_xBiO_3$, namely $Ba_{1-x}Pb_xBiO_3$, in which a phase with tetragonal structure, which is absent in the former system, exhibits a superconducting behaviour [12]. Therefore, we suspect that $Ba_{0.62}K_{0.38}BiO_{3-y}$ has a tetragonal structure inside the T–P region delineated by the dashed line in figure 4.

Acknowledgments

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