High-Pressure Studies on a Newly Discovered Spin-Ladder Iron-Based Superconductor $BaFe_2S_3$

Iron-based superconductors are well known as high-T_c superconductors, which have a two-dimensional iron lattice as a common feature. In copper-based superconductors, both two- and one-dimensional copper lattices have been found as a structural motif. We have discovered pressure-induced superconductivity in an iron-based quasi-one-dimensional spin-ladder-type material $BaFe_2S_3$ using a diamond anvil cell. The superconductivity emerges below 14 K, immediately after the appearance of metallic phase at ~11 GPa. The X-ray powder diffraction patterns indicate that the superconductivity appears in the ladder structure. Our findings indicate that this material provides useful insight into iron-based superconductivity.

The discovery of iron-based superconductors has had a significant impact on material science. Iron-based superconductors are well known as high-Tc superconductors, which have a two-dimensional iron lattice as a common feature, despite having different crystal structures. These materials exhibit characteristic magnetic phases next to the superconducting phase. A stripe-type magnetic order is observed in the 1111, 122, 111 and 11 types, and a block-type magnetic order is observed in the 245 type of iron-based superconductors. Recent studies on spin-ladder-type iron-based materials revealed that they exhibit various kinds of magnetic ordering phase, which are a one-dimensional analog of both the stripe and block magnetism observed in iron-based superconductors [1, 2]. Due to such similarity of magnetic property, superconductivity was expected to be found in spin-ladder-type iron-based materials, even though they have shown only insulating behavior. In the case of copper-based superconductors, the spin-ladder one had been theoretically predicted and was discovered in $(Sr,Ca)_{14}Cu_{24}O_{41}$ under high pressure above 4 GPa at 12 K [3]. This discovery shed light on studies on high-T_C superconductivity. Here we report the pressure-induced superconductivity of iron-based spin-ladder-type $BaFe_2S_3$, having the stripe order magnetic phase [4].

Electrical resistance measurements were carried out by a standard dc four-probe method up to 30 GPa using a diamond anvil cell (DAC) [4]. Powdered NaCl was used as the pressure-transmitting medium, and platinum ribbons were used as electrical leads with a thin BN layer acting as electric insulation between the leads and the rhenium gasket. High-pressure X-ray diffraction measurements were performed using synchrotron radiation, applying a wavelength of 0.621 Å to a sample in a DAC with a liquid pressure-transmitting medium (Daphne 7474 or helium).

Figure 1a shows the resistance under pressures up to 13 GPa. Upon the application of pressure, the insulating properties are gradually suppressed and the *R* curve exhibits a metal-insulator transition around 11 GPa and a sudden decrease at 11 GPa and 13 K. This feature is attributed to a superconducting transition, based on two observations for T_c suppressed by magnetic fields and electrical currents. The pressure dependencies of T_c obtained by several experimental trials are summarized in **Fig. 1c**, in which T_c shows a dome shape with a maximum T_c of 17 K at 13.5 GPa.

The X-ray diffraction patterns under high pressure did not show definite signs of structural transition, indicating that the superconductivity appears in the ladder structure, as shown in **Fig. 2a**. **Figures 2b** and **2c** show the pressure dependencies of lattice constants at 300 K and 10 K normalized to the ambient pressure values, respectively. The *b*-axis perpendicular to the ladder layer is seen to be the most compressible and it is likely that anisotropic compression makes the system more metallic by increasing the transfer of Fe 3*d* electrons across the inter-ladder bonds, which suggests the bandwidth-controlled-type Mott transition. Theoretical calculation confirmed the electronic structure revealed by our X-ray results under high pressure [5]. Although we could not evaluate the magnetic ordering under high pressures, the suppression of magnetic ordering with the application of pressure is a general trend in iron-based and copper-based superconductors. Therefore, it is reasonable to consider that the increased conductivity disturbs the magnetic ordering in $BaFe_2S_3$. A subsequent high-pressure study using a cubic anvil press reported the magnetic phase in the high-pressure region [7]. The superconductivity likely emerges in the vicinity of the quantum critical point associated with both charge and spin fluctuations.

From the structural point of view, $BaFe_2S_3$ is quite similar to the copper-based ladder material $Sr_{14-x}Ca_xCu_{24}O_{41}$. In the copper-based one the pressure-induced charge transfer from the CuO chain to the Cu_2O_3 ladder results in the appearance of superconductivity, which is categorized as a filling-control-type Mott transition, in contrast to the bandwidth-controlled-type Mott transition in $BaFe_2S_3$. In the ladder material, the *d* wave symmetry of the superconductivity is theoretically predicted, and is distinct from the common $s^{\pm}$ wave symmetry of two-dimensional square lattice structures [6]. Another difference is that no definite magnetic ordering has been observed adjacent to the superconducting phase in $Sr_{14-x}Ca_xCu_{24}O_{41}$, which is in contrast to the superconductivity emerging adjacent to the magnetic phase in $BaFe_2S_3$. We conclude that the properties of $BaFe_2S_3$ are related to those of various unconventional superconductors, and may clarify the interplay between magnetism and superconductivity. Further substitutional and high-pressure studies are expected to shift the transition pressure to a much more accessible range, possibly to ambient pressure.

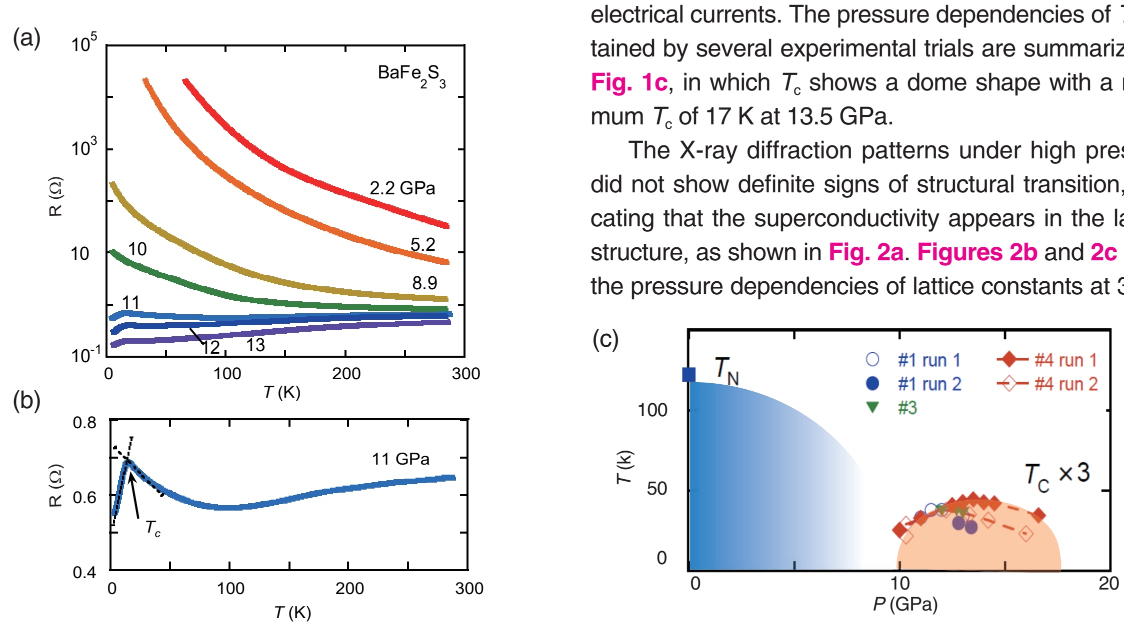

Figure 1: (a) Electrical resistance of $BaFe_2S_3$ for each pressure. The insulator-metal transition is observed between 10 and 11 GPa. The resistance drop due to superconductivity can be seen above 11 GPa. **(b)** Electrical resistance at 11 GPa on a linear scale, showing superconducting transition. **(c)** *P-T* phase diagram of $BaFe_2S_3$, which shows antiferromagnetic phase at ambient pressure. Superconducting transition appears above 10 GPa and disappears above 17 GPa. The maximum T_c is ~17 K.

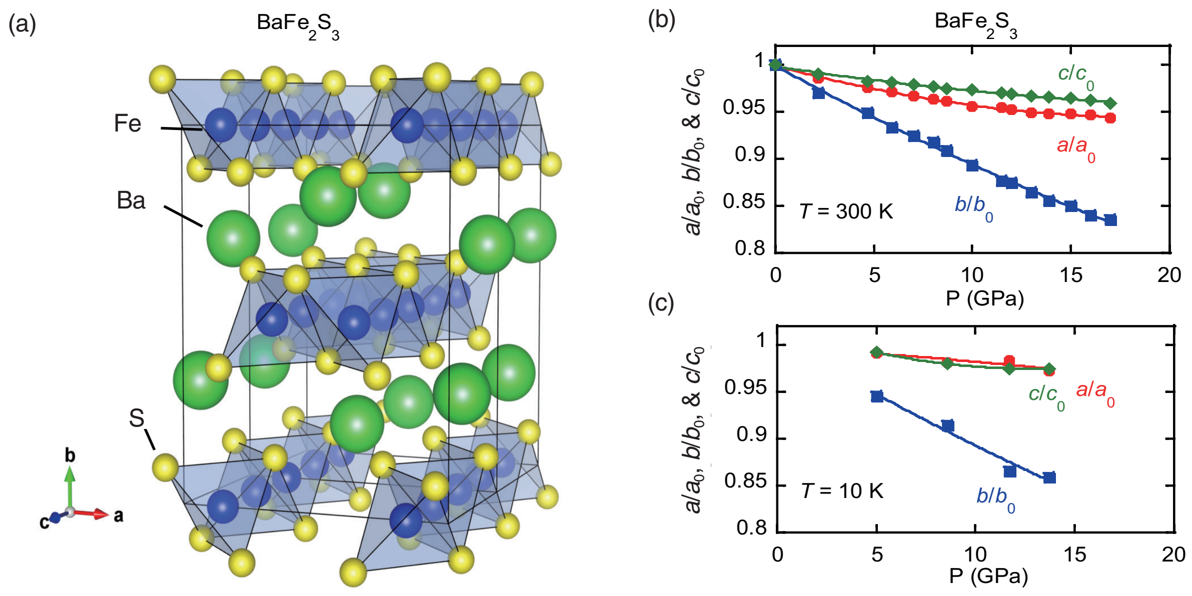

Figure 2: (a) The crystal structure of $BaFe_2S_3$, consisting of edge-shared FeS_4 tetrahedra extending along the *c*-axis and channels occupied by Ba atoms, in which Fe atoms form a ladder structure, depicted by VESTA software [8]. **(b)** Pressure dependence of lattice constants normalized to the ambient pressure values, obtained by high-pressure X-ray diffraction at ambient temperature. The layer direction (*b*-axis) is seen to be the most compressible. **(c)** Compression data obtained at 10 K, showing similar behavior to the data at 300 K.

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