High-T_c superconducting phases of $FeSe_{1-x}S_x$ at high pressure

In condensed matter physics, it is one of the important problems to understand the mechanism of unconventional superconductivity which is not described by the standard BCS theory. The phase diagrams of such unconventional superconductors have a common feature, i.e. the superconducting phase exists near the magnetic order. Recently-discovered Fe-based superconductors also have this feature. From this point, it is thought that the spin fluctuations enhanced near the verge of the magnetic order are related to the mechanism of the unconventional superconductors. On the other hand, recent studies suggest that the nematic order that spontaneously breaks rotational symmetry of the system exists in the Fe-based superconductors and its quantum fluctuations may play an essential role for the superconductivity. However, this remains unclear because the nematic order usually coexists with the magnetic order [1]. To solve this issue, FeSe which uniquely exhibits a nonmagnetic nematic order is a key system. FeSe has the simplest crystal structure among Fe-based superconductors and shows the tetragonal-to-orthorhombic structural transition (nematic transition) at T_s~90 K as well as the superconductivity at T_c~9 K. It is reported that by partially substituting Se with isovalent S, a nematic quantum critical point can be reached, at which the electronic nematic transition temperature T_s is suppressed to absolutely zero [2]. By applying physical pressure, T_s is suppressed as in the case of S-substitution system. However, the magnetic order is induced before the nematic order is completely suppressed. This pressure-induced magnetic order has a dome shape in the pressure phase diagram. When the magnetic order is suppressed in high-pressure region, high-T_c superconductivity at T_c = 38 K is realized [3]. These results indicate that pressure and S-substitution have different effects on the electronic states of FeSe. Thus, it is important to explore how the ground state of FeSe changes when chemical pressure by S-substitution and physical pressure is controlled as independent parameters. We have performed high-pressure studies in high-quality single-crystalline $FeSe_{1-x}S_x$ up to 8 GPa. We find a systematic change of the pressure phase diagram in FeSe by the S-substitution. Our results imply that the respective role of nematic and magnetic fluctuations can be elucidated from the precise control of pressure and substitution in this system [4].

High-pressure resistivity measurements

First, the transport measurements under high pressure up to 8 GPa in high-quality single crystals of $FeSe_{1-x}S_x$ (x=0.04, 0.08, 0.12 and 0.17) are conducted with a constant-loading type cubic anvil apparatus invented by Uwatoko group at Institute for Solid State Physics in The University of Tokyo. Each sample is grown by the chemical vapor transport technique. From the anomalies in the resistivity curve, we assigned the phase transition temperatures, T_s, T_c, as well as the magnetic transition temperature T_m. From this we establish the temperature(T)-pressure(P)-S-substitution(x) 3-dimensional (3D) electronic phase diagram. As shown in Fig. 1, the electronic nematic

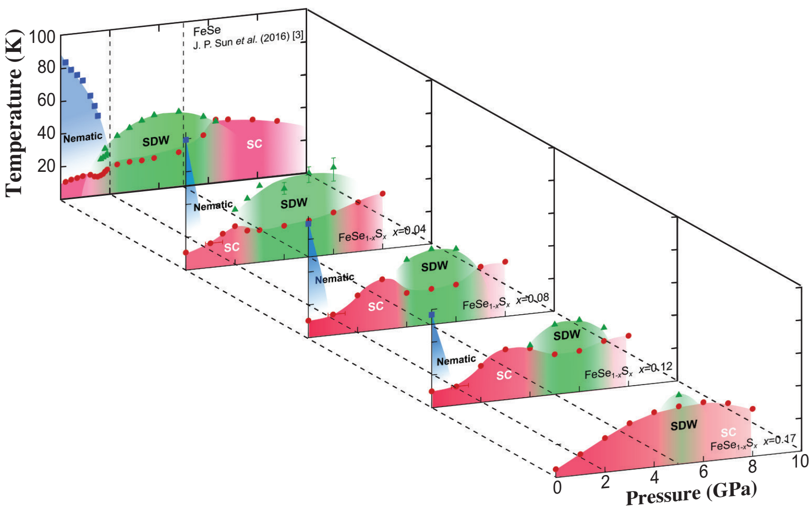

Fig. 1. The T-P-x 3D electronic phase diagram of $FeSe_{1-x}S_x$ established by this work. The overlap of the nematicity and magnetism is separated by S-substitution. Furthermore, it is also shown that the high-T_c superconductivity occurs near the magnetic phase.

phase and pressure-induced magnetic phase overlap in low-pressure region of the electronic phase diagram of FeSe, but these two orders separate with each other by increasing the S-content. Besides, at the region where both two orders are absent, a new high-T_c superconducting phase is observed. Highest T_c is achieved close to the verge of magnetic phase, which suggests the pressure-induced magnetic phase has an intimate link with high-T_c superconductivity in this system.

Synchrotron X-ray diffraction measurements

To understand more about the nature of the pressure-induced high-T_c superconducting phase, the crystal structure of $FeSe_{1-x}S_x$ at high pressure needs to be clarified. Synchrotron X-ray diffraction measurements of $FeSe_{1-x}S_x$ ($x = 0.08$) under pressure have been performed at SPring-8 **BL22XU** by using a diamond anvil cell. In this content, the transport measurements reveal the magnetic order between 4 and 6 GPa (Fig. 2(a)). The upper parts of Figs. 2(b) and 2(c) show the results of the temperature dependence of (331) Bragg peak at 3 and 4.9 GPa, respectively. At 4.9 GPa, the split of the Bragg peak by the tetragonal-orthorhombic structural transition is clearly observed near the temperature where the resistivity anomaly by magnetic transition appears (Fig. 2(c)). This result is consistent with the previous synchrotron X-ray diffraction measurements of FeSe [5]. This confirms that the dome-shaped magnetic order in the pressure phase diagram of $FeSe_{1-x}S_x$ is accompanied with the tetragonal-orthorhombic structural transition. In contrast, at 3 GPa where the new high-T_c superconductivity is observed by the transport measurements, no split of the Bragg peak is observed, indicating that the crystal structure remains tetragonal down to the lowest temperature (Fig. 2(b)). This demonstrates that in $FeSe_{1-x}S_x$ nematic and magnetic phases are separated completely and that the high-T_c superconductivity appears in the non-magnetic tetragonal phase.

These results show that in FeSe-based system it is possible to control the electronic nematicity and the magnetism in the electronic phase diagram by the combination of S-substitution (chemical pressure) and physical pressure. These two different pressures have different effects on the electronic state of FeSe. In this system, the high-T_c superconductivity appears in the vicinity of pressure-induced magnetic phase, which suggests that the non-magnetic electronic nematic fluctuations alone cannot induce high-T_c superconductivity, but the pressure-induced magnetism with orthorhombicity has an intimate relationship with the high-T_c superconductivity.

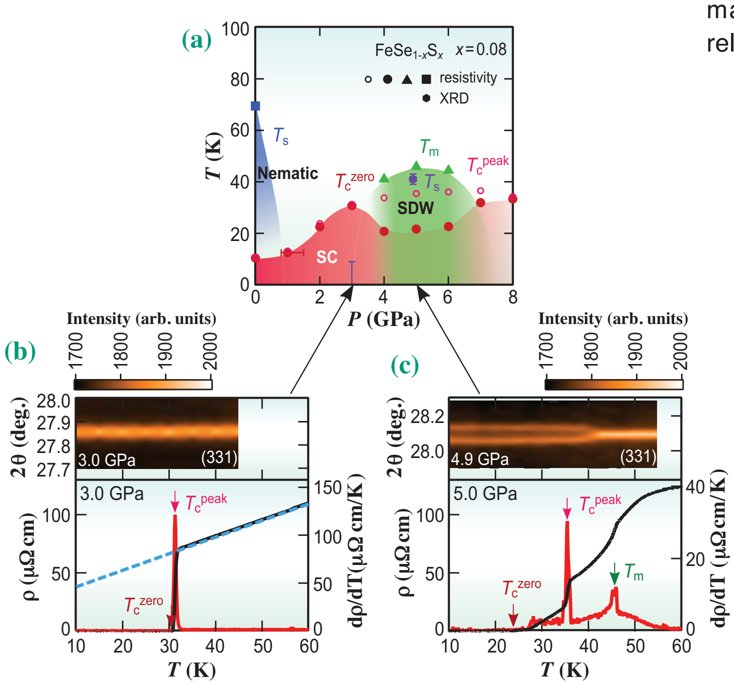

Fig. 2. Temperature-pressure phase diagram for $x = 0.08$. **(a)** T-P phase diagram of $FeSe_{1-x}S_x$ ($x = 0.08$) together with T_s determined by the high-pressure synchrotron X-ray diffraction (XRD) in a diamond anvil cell (purple hexagon with error bars). **(b,c)** Temperature-dependence of Bragg intensity as a function of 2θ angle is indicated in color scale for 3.0 GPa (b) and 4.9 GPa (c). Temperature dependence of resistivity $\rho(T)$ and $d\rho/dT$ are also shown with the same horizontal axis. The red (green) arrows indicate T_c (magnetic transition temperature, T_m). The blue dashed line in (b) is a T-linear fit to the normal-state $\rho(T)$ at 3.0 GPa.

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