Field-Induced Magnetostructural Transitions in Antiferromagnetic Fe$_{1+y}$Te$_{1-x}$S$_x$

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Abstract Transport and structural properties of Fe$_{1+y}$Te$_{1-x}$S$_x$ were studied in pulsed magnetic fields. Application of high magnetic fields induces first order transitions showing positive magnetoresistance effects in the antiferromagnetic phase. Polarizing microscope images taken at high magnetic fields revealed the concomitant melting of the orbital order. These results indicate the importance of crossed coupling between spin and lattice or orbital degrees of freedom in this compound.

Keywords Superconductivity · Magnetism · Phase transition · Fe$_{1+y}$Te$_{1-x}$S$_x$

1 Introduction

Since 2008, iron-based superconductors have attracted considerable attention because of the second highest transition temperatures next to the cuprate superconductors [1]. The superconductivity in the cuprates emerges when antiferromagnetic (AFM) states of the parent compounds are removed by chemical substitution, whereas that in the ironates is induced also by application of pressure to the AFM states. Thereby, we are naively interested in the states in which the AFM order is suppressed by application of high magnetic fields.
As one of the main differences between the both systems, all of the five 3d bands are involved in the electronic states near the Fermi level in the ironates [2] making sharp contrast with the single-band nature of the cuprates. In particular, the degeneracy of the $d_{yz}$ and $d_{zx}$ orbitals in the tetragonal phase causes orbital fluctuation, which may play a crucial role to realize the high temperature superconductivity in the ironates [3]. When the AFM order, and also the accompanying structural transition, emerges, break of the four-fold symmetry results in the static real-space order of the 3$d$ orbitals. Therefore, if we can remove the spin order by application of magnetic fields, whether the orbital order survives or not is an important issue for the electronic states in the field-induced phase.

The effects of magnetic fields on the AFM states on the ironates have been less studied. Although we reported the occurrence of field-induced transitions in EuFe$_2$As$_2$ [4], the phenomena were discernible only in the vicinity of $T_N$, and hence, could not be discussed in detail. In this work, we studied the effects of magnetic fields on Fe$_{1+y}$Te$_{1-x}$S$_x$ crystals in pulsed high magnetic fields. Since these iron chalcogenides have lower $T_N$ than the iron arsenides exhibit, we expect prominent changes in the physical properties over wide range of temperature.

2 Experimental Details

The crystals of Fe$_{1+y}$Te$_{1-x}$S$_x$ were synthesized by the flux method [5, 6]. The ratios between Te and S determined by the electron-probe microanalyses were $x_F = 0$, 0.03, and 0.048 for the nominal values of $x = 0$, 0.05, and 0.10, respectively [6]. Pulsed magnetic fields up to 65 T were generated using non-destructive magnets installed at the International MegaGauss Science Laboratory of ISSP. Magnetoresistance was measured using the ac four-probe method. Polarizing microscope images in pulsed fields were obtained with using our newly developed high-speed imaging system [7].

3 Results

Figure 1(a) shows the temperature ($T$) dependence of the in-plane resistivity ($\rho_{ab}$) of Fe$_{1+y}$Te$_{1-x}$S$_x$ crystals. As a common feature of these compounds, the $\rho_{ab}$ exhibits slight increase upon cooling in the paramagnetic state, and then steeply decreases below $T_N$. The $T_N$ monotonically decreases as the $x$ increases. Application of high magnetic fields to the AFM states causes gradual increase in the $\rho_{ab}$, and then leads to the saturation at certain fields marked by the closed and open triangles in Fig. 1(b). Similar transitions are observed also for $H \parallel ab$-plane in all the samples studied [8]. Figures 1(c) and 1(d) are the phase diagrams for $H \parallel ab$-plane and $H \parallel c$-axis, respectively. The horizontal axes were normalized by the $T_N$ of each compound. The transitions appear to be induced by smaller fields for $H \parallel ab$-plane and also for the larger $x$ although the origin of the anisotropy and compositional dependence are not clear at the present. The extrapolation of the phase boundary of the $x = 0.10$ sample suggests the occurrence of the transition at around 70 T in the low temperature limit.
Fig. 1 (a) Temperature dependence of in-plane resistivity ($\rho_{ab}$) in Fe$_{1+y}$Te$_{1-x}$S$_x$ crystals for $x = 0$, 0.05, and 0.10. (b) Magnetoresistance of the $x = 0.10$ crystal in magnetic fields applied parallel to the $c$-axis. The data were vertically offset for clarity. The transition fields were defined by the crossing points of the extrapolated magnetoresistance curves in high- and low-field regions (dotted lines), and marked by closed and open triangles. Phase diagram of Fe$_{1+y}$Te$_{1-x}$S$_x$ for (c) $H \parallel ab$-plane and (d) $H \parallel c$-axis. The horizontal axis is normalized by the $T_N$ of each compound (Color figure online)

According to the early neutron experiments on Fe$_{1.068}$Te [9], this class of iron chalcogenites exhibit transitions from the tetragonal to the monoclinic structure simultaneously with the magnetic transition. This monoclinic structure accommodates to the order of the $d_{yz}$ and $d_{zx}$ orbitals. Since this orbital order results in the anisotropy of the optical conductivity, we can identify the occurrence of the orbital order with using a polarizing microscope. We can visualize the domains of orbital order as the regions showing different contrast in the polarizing microscope images on the $ab$-plane surface [10]. The polarizing microscope images of the $x = 0.10$ crystal captured by a cooled charge-coupled-device camera taken at various temperatures are shown in Figs. 2(a)–2(d). In the paramagnetic state, the polarizing microscope image shows up homogeneous except for some steps and scratches as shown in Fig. 2(a). At 46.0 K, stripe-like structures emerge from the top-right corner of the Fig. 2(b) indicating the occurrence of the orbital order. Upon further cooling, the twin structure spreads over the sample. As an indicator for the degree of optical anisotropy derived by the orbital order, standard deviation of the optical intensity ($\sigma_I$) was calculated for the region marked by the dotted rectangle in Fig. 2(a), and plotted in Fig. 2(e) as a function of temperature after normalization by average intensity ($I_{avg}$). The $\sigma_I$ smoothly decreases with increasing $T$, and falls on the value of background at $T_N$.

Next, let us move to the effect of magnetic fields on the orbital order. To clarify this effect, we observed polarizing microscope images in pulsed magnetic fields up to 35 T. Figure 3(a) shows the profile of the pulsed field for the imaging experiment. The open circles represent the points at which the images were taken by the high-
Fig. 2 Polarizing microscope images of the cleaved ab-plane surface of the $x = 0.10$ crystal at temperatures of (a) 50.0 K, (b) 46.0 K, (c) 40.0 K, and (d) 8.8 K. $E_{\text{in}}$ and $E_{\text{out}}$ represent the directions of polarization in the polarizer and analyzer, respectively. (e) Temperature dependence of the normalized standard deviation of the optical intensity ($\sigma I/I_{\text{avg}}$) in the area marked by the dotted rectangle in (a) (Color figure online)

speed camera at a rate of 5,000 frames per second. Figure 3(b) shows the polarizing microscope image at 30 K before application of magnetic field. Figure 3(c) shows the image at 35.2 T. In this field, the stripe-like structures in the lower half of the image disappear indicating the local melting of the orbital order. The partially melted orbital order does not recover after the removal of the field as shown in Fig. 3(d). This irreversible change coincides with the hysteretic nature observed in the magneto-resistance measurements (Fig. 1(b)). We calculated the $\sigma I/I_{\text{avg}}$ in the area shown by the dotted rectangle in Fig. 3(c) and plotted in Fig. 3(e) as a function of magnetic field. This result evaluates partial reduction of the optical anisotropy derived by the orbital order. The qualitative difference between Figs. 3(e) and 1(b) is not surprising because the $\sigma I/I_{\text{avg}}$ represents the local information of the orbital order, whereas the $\rho_{ab}$ reflects the resistance in the whole sample. Our results demonstrate that magnetic field induced transitions in the AFM state of Fe$_{1+y}$Te$_{1-x}$S$_x$ coincide with the melting of the orbital order. The direct coupling between the spin and lattice/orbital degrees of freedom can be the key ingredient to realize the unique bicollinear spin order in the iron-calchogenides [11, 12], and should be investigated in detail in future experiments.
4 Conclusions

Field-induced transitions in Fe_{1+y}Te_{1-x}S_x crystals were studied through measurements of magnetoresistance and polarizing microscopy in pulsed high magnetic fields. The results demonstrate the occurrence of first order transitions in the antiferromagnetic states that are characterized by the positive magnetoresistance effects and by the disappearance of the optical anisotropy. Occurrence of the field-induced melting of the orbital order indicates strong coupling between spin and lattice/orbital degrees of freedom in this class of ironates.

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