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Scotch tape induced strains for enhancing superconductivity of $FeSe_{0.5}Te_{0.5}$ single crystals

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We investigated the superconducting transition temperature T_c of $FeSe_{0.5}Te_{0.5}$ single crystals, which can be enhanced up to 14% by attaching onto a commercial Scotch tape. The Scotch tape exhibits a large cooling shrinkage at low temperatures, which is considerably more pronounced than that of the metallic $FeSe_{0.5}Te_{0.5}$ single crystal, thus providing a compressive strain of 2.4×10^{-3} at 15 K. For such strain, we calculated that the lattice parameter of c/a can be increased to ~0.31%, which corresponds to the enhancement of the superconductivity. The present finding provides a rapid and simple method to examine the microstructure sensitive physical properties of the layered-structure materials by using the Scotch tape as strain generator. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4903922]

The newly discovered Fe-based superconductors absorbed increasing attentions for understanding the high temperature superconducting transition (high-T_c) mechanism and applications.[1–8] Like the high-T_c cuprate superconductors, the superconductivity of the Fe-based family is also generated by suppression of the antiferromagnetic ordering via tuning the parameters, including chemical doping and external pressure.[3,4] Among the Fe-based family, FeSe superconductors possess the simplest structure for the PbO-type binary element structure with stacks of Fe_2Se_2 layers. The highest T_c of FeSe was found as 15 K with half of Se substituted by Te.[2,5] Applying external pressure on $FeSe_{0.5}Te_{0.5}$ can enhance the T_c up to 23 K,[9] being considerably higher than that of chemical doping, while the maximum T_c and superconducting properties vary with the pressuring systems and the samples. The pressure provides isotropic compression along three dimensions, but neither a uniaxial nor a biaxial strain, which arouses arguments on the mechanism of lattice modification on the superconductors, mostly are two-dimensional. To uncover the phase diagram of T_c and lattice parameters, a uniaxial component of the pressure is of great important. The epitaxial growth thin films can be induced uniaxial pressures due to the mismatched lattice parameters between the crystals and the substrates.[10] For instance, T_c of $FeSe_{0.5}Te_{0.5}$ (15 K in bulk) can be increased to 17 and 21 K on the substrates of $LaAl_2O_4$ and $SrTiO_3$, respectively.[11,12] A recent report showed that a monolayer FeSe film ($T_c = 8$ K in bulk) on $SrTiO_3$ substrate observed a superconducting gap at a dramatically high temperature of 65 K.[13]

Up to now, the correlation between the microstructure and T_c of the $FeSe_{0.5}Te_{0.5}$ superconductors remains controversial, because various factors of the structural parameters may contribute to the superconductivity, including the in-plane lattice shrinkage,[11] the out-of-plane lattice constant c,[12] the anion height from the Fe-square planes,[14] or even the Fe vacancies.[15] Besides, superconductivity in the previous thin film could be influenced by some other extrinsic effects not just the lattice mismatch induced strains, such as incorporation of oxygen, inhomogeneous components, interface effects, film thickness, and particularly the growth conditions, all of which will inevitably confuse the intrinsic strain effect on superconductivity.[15] In addition, the lattice mismatch between the substrate and thin films was generally considered at room temperatures in previous investigations; another important factor has not yet be seriously taken account into the superconductivity modification, namely, the mismatch of thermal expansion between the crystals and substrates, which is normally different at low temperatures.

In this letter, we investigated the modification of superconductivity for the $FeSe_{0.5}Te_{0.5}$ thin crystal which was induced compressive strains by commercial Scotch tape. The T_c of a $FeSe_{0.5}Te_{0.5}$ ultrathin single crystal attached onto a piece of Scotch tape can be unexpectedly enhanced by about 2 K due to a mismatch of thermal strain. The results obtained from such a subtle treatment indicate the high sensitivity of superconductivity to the strain induced variations of the microstructures and arouse further researches on the strains inducing as-modified microstructure for the enhancement of

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superconductivity, which could be significant for future applications on strain controlled superconductivity.

High quality $FeSe_{0.5}Te_{0.5}$ single crystals were synthesized by the self-flux method.[16] As a typical layered material, the $FeSe_{0.5}Te_{0.5}$ single crystals can be exfoliated from bulk single crystals into ultrathin flakes using the commercial Scotch tape, as widely used on graphene.[17] For the thickness of the ultrathin flakes, we can estimate from the optical microscopy using the backlight, namely, the flakes could be a few hundred nanometers even less once it is transparent under backlight as can be seen in the inset of Fig. 1.

An entire bulk crystal of $FeSe_{0.5}Te_{0.5}$ and the following exfoliated flakes on the tape are measured on the magnetic properties under a magnetic field of 10 Oe by magnetic properties measurement system (MPMS, Quantum Design), as shown in Fig. 1. The magnetization has been measured in a zero-field cooling (ZFC) sequence. For the bulk sample, the magnetic field has been applied parallel to the *ab*-plane. However, for the flakes sample, it is hard to determine the field direction. The bulk crystal exhibits a T_c onset of 14.0 K, being consistent with the previous results,[16] while the T_c onset of the tape-attached flake is enhanced as 15.5 K. To confirm the effects of the tape, we separated the flakes from the tape using dichloromethane and then measured the magnetization of the collected flakes. The T_c was observed as to restore to its original bulk value, although the magnetization demonstrates slight degradation from both bulk and tape-attached flakes, which could be ascribed to the collection process from the tape. Note that the dichloromethane solvent has no effect on the sample quality, which gives clear evidence that the superconductivity of $FeSe_{0.5}Te_{0.5}$ flakes is purely enhanced by the tape. Actually, we repeated the experiment for several times and an increase of T_c by 1–2 K was reproducible.

We also measured the temperature dependent resistance with standard four-probe configuration by physical properties measurement system (PPMS, Quantum Design) for the bulk crystal and an individual flake on the tape. We selected the ultrathin crystals with thicknesses below 100 nm for full transmission of strain from the substrate to the crystals. Figure 2 shows the temperature dependent normalized resistance R/R_n of a bulk crystal and an individual ultrathin flake on Scotch tape. The T_c of the bulk crystal and the flake is observed as 14.7 and 15.9 K, respectively, being in accordance with the magnetization results. Note that the flake exhibits similar profile of the temperature dependent normal state resistance as that of bulk crystal.

The magnetic and transport properties results demonstrate that the superconductivity of $FeSe_{0.5}Te_{0.5}$ crystal can be modified by the tape during low temperatures. Since the Scotch tape is a typical organic material, it exhibits considerable large shrinkage at low temperatures. In contrast, the shrinkage of $FeSe_{0.5}Te_{0.5}$ single crystals should be much less than that of tape due to the metallic behavior. Thus, we studied the relative changes of the length $\Delta L/L_0$ with respect of temperature for the $FeSe_{0.5}Te_{0.5}$ single crystal and Scotch tape as shown in Fig. 3. The strain was monitored using a strain gauge adhered onto the tape and the crystal. Note that the strains of both tape and crystal are in-plane isotropic, thus we investigated any the linear changes of the length $\Delta L/L_0$ instead of the plane. The tape shows around two times of $\Delta L/L_0$ as that of $FeSe_{0.5}Te_{0.5}$. Therefore, once attaching the crystals onto the tape, it will provide a compressive strain onto $FeSe_{0.5}Te_{0.5}$ due to their mismatch in thermal contractions, which can be estimated from the differential $\Delta L/L_0$ between the tape and the $FeSe_{0.5}Te_{0.5}$ given as the curve of Scotch-$FeSe_{0.5}Te_{0.5}$ in Fig. 3. Since the $FeSe_{0.5}Te_{0.5}$ flakes are considerably thin, we considered that the strain from the tape can be fully transmitted to the crystals. The highest compressive strain reaches to about -2.4×10^{-3} at 15 K, which generated enhancement of T_c for 14%. According to the thin films experimentals,[11] there is a rough linear relation between $T_c \approx -450\alpha + 12$, where α is the strain from the lattice mismatch between the substrate and the thin films. For our case, we can estimate that the compressive strain from the Scotch tape (2.4×10^{-3}) can induce T_c

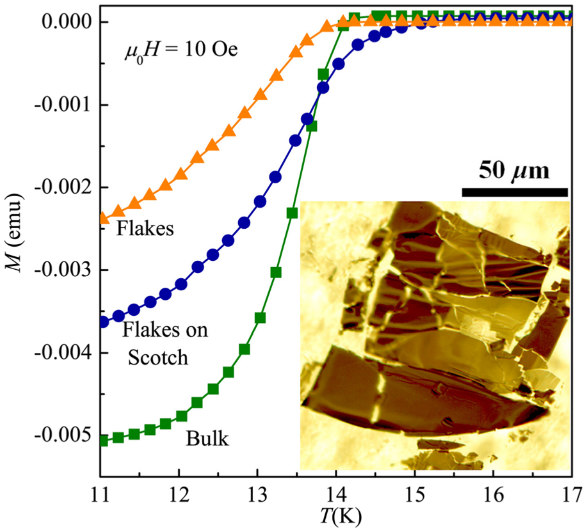

FIG. 1. Temperature dependence of the magnetization for a $FeSe_{0.5}Te_{0.5}$ bulk single crystal (squares), exfoliated flakes on Scotch tape (circles), and the flakes alone after separation from the tape by using dichloromethane (triangles). Inset shows an image of $FeSe_{0.5}Te_{0.5}$ flakes on Scotch tape obtained by mechanical exfoliation from a bulk crystal under backlight.

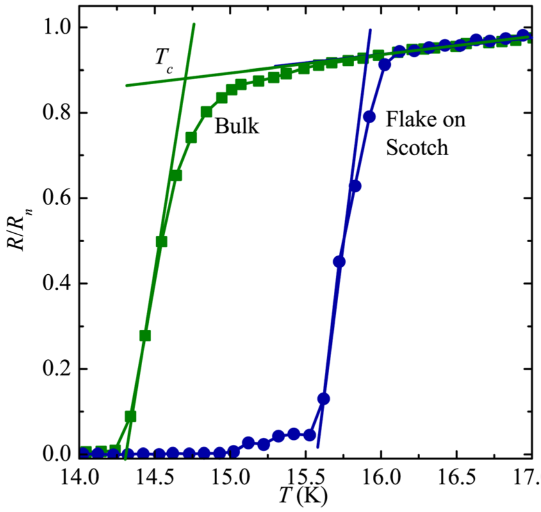

FIG. 2. Temperature dependence of the normalized resistance R/R_n (R_n corresponds to the normal state resistance at $T = 18$ K) for $FeSe_{0.5}Te_{0.5}$ bulk crystal (solid squares) and an individual flake on tape (solid circles).

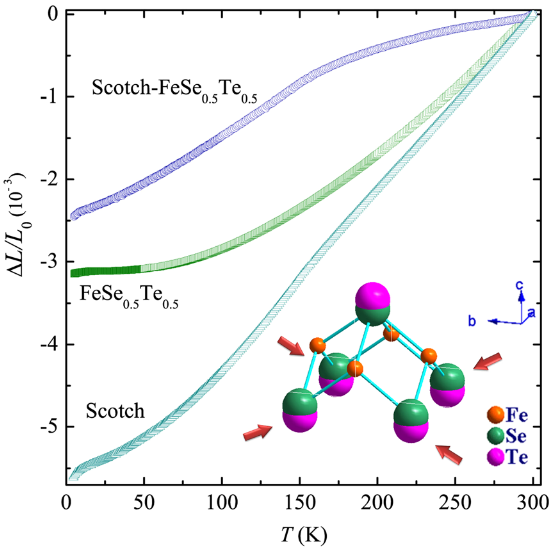

FIG. 3. Relative length change $\Delta L/L_0$ (L_0 is the initial length at $T = 300\,\mathrm{K}$) as a function of temperature for the $\mathrm{FeSe_{0.5}Te_{0.5}}$ single crystal and the Scotch tape. The Scotch-$\mathrm{FeSe_{0.5}Te_{0.5}}$ curve demonstrates the mismatch of thermal shrinking length between the Scotch tape and $\mathrm{FeSe_{0.5}Te_{0.5}}$. Inset gives a unit cell of $\mathrm{FeSe_{0.5}Te_{0.5}}$ which is subjected to compressive strains from the Scotch tape within the *ab*-plane.

enhancement of ∼1.08 K, which is in accordance with our experimental result (1–2 K).

To understand the microstructure variation of $\mathrm{FeSe_{0.5}Te_{0.5}}$ under strain, we investigated the biaxial in-plane strain induced modification of the lattice parameters using the first principles calculation based on density functional theory (DFT) as implemented in the ABINIT code.[18] The $2 \times 2 \times 1$ super-cell is used to model $\mathrm{FeSe_{0.5}Te_{0.5}}$. For simulation of strains within the *ab*-plane, the $a(b)$-axis is controlled by the strains of the substrate, tape in the present case. On the other hand, the c-axis and atomic positions are fully relaxed until the force acting on each atom is less than $10^{-4}\,\mathrm{eV}^{-1}$. The Brillouin zones are sampled with a $10 \times 10 \times 12$ Monkhorst-Pack k-mesh. The cutoff energy for the plane wave expansion is set to be 800 eV. The local density approximation (LDA) proposed by Perdew and Wang (PW92) is used for the exchange-correlation energy.[19] We found that the in-plane compressive strain results in a decrease of the lattice parameter a but a increase of c. Margadonna and co-workers found that decreasing anion height from the Fe-square planes results in enhancement of T_c for the α-FeSe.[14] However, the high-pressure experiments in $\mathrm{FeSe_{0.5}Te_{0.5}}$ seem unlikely to obey the relation between the anion height and T_c.[20,21] On the other hand, the neutron diffraction study on the atomic configurations of $\mathrm{FeSe_{0.5}Te_{0.5}}$ exhibited that the reducing Te height from the Fe-square planes can contribute to the suppression of superconductivity of the annealed samples.[22] For the strain of -0.25% in our present case, we estimated the anion height from d_{Fe-Te} and d_{Fe-Se} as -0.016% and -0.03%, respectively, being considerably less than the value of Δa (-0.25%) and Δc (0.24%). Therefore, we can hardly attribute the anion height variation to the enhancement of superconductivity in the present case. Since one can hardly identify if the T_c modification is originated from a separated variation of a or c constant, the ratio constant c/a which corresponds to the Se-Fe-Te angle is a promising way. Based on calculation results, a strain variation of 0.25% from the tape gives rise to a modification of 0.31% on c/a or the Se-Fe-Te angle.

To further study the effects of the strains on the physical properties, we also studied the parent compound FeTe. The magnetic properties of the FeTe were investigated for both bulk crystal and the ultrathin flakes attached on tape (see Fig. 4). The data were collected under a magnetic field of 1 T, first upon cooling from room temperature down to 5 K field-cooled cooling (FCC) and then warming back as field-cooled warming (FCW). The bulk crystal exhibits an anti-ferromagnetic transition T_N at around 70 K and thermal hysteresis between FCC and FCW due to the corresponding structural transition, being consistent with the previous studies.[16] For the ultrathin flakes, however, the anti-ferromagnetic transition is broadened. The differential of the magnetization curves demonstrates that the mid-peaks of the ultrathin flakes are about 1.9 K higher than those bulk crystals, regardless of FCC or FCW. The enhancement of T_N for FeTe is consistent with the T_c of $\mathrm{FeSe_{0.5}Te_{0.5}}$, suggesting that the tape substrate effects on microstructure of both $\mathrm{FeSe_{0.5}Te_{0.5}}$ and FeTe single crystals, and consequently the intrinsic physical properties.

In conclusion, we investigated the compressive strains effects on superconductivity of the $\mathrm{FeSe_{0.5}Te_{0.5}}$ superconductor using a commercial Scotch tape. The magnetic and transport measurements demonstrated that the T_c of the $\mathrm{FeSe_{0.5}Te_{0.5}}$ single crystals can be enhanced up to 14%, via adhering the $\mathrm{FeSe_{0.5}Te_{0.5}}$ ultrathin flakes onto the tape. Once separating the flakes from the tape by using dichloromethane, the T_c turns back to the value of the bulk crystal. We also calculated the modification on the lattice parameter from the compressive strains of 2.4×10^{-3} at 15 K and found that the strain results in increase of c/a to 0.31%. The magnetic properties of the parent compound FeTe also demonstrated an enhancement of the anti-ferromagnetic transition T_N for ∼1.9 K than the bulk crystals by attaching the crystals onto the tape. This approach can be employed to explore the physical properties of the layered materials sensitive to

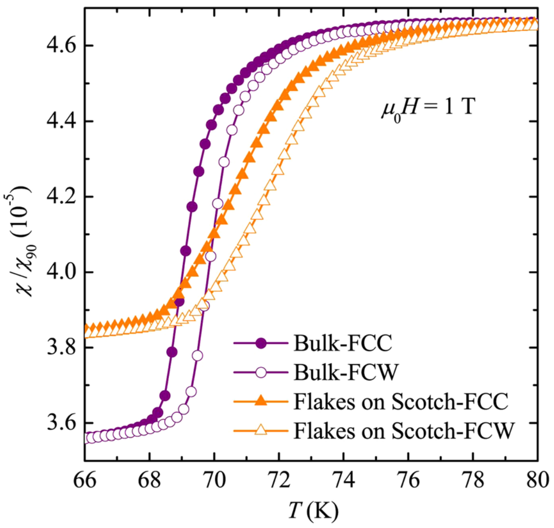

FIG. 4. Temperature dependence of the magnetization for a FeTe bulk single crystal (cycles) and flakes on tape (triangles) under magnetic field parallel to *ab*-plane.

microstructure alterations and find many practical applications.

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