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Chalcogen (O_2, S, Se, Te) atmosphere annealing induced bulk superconductivity in $Fe_{1+y}Te_{1-x}Se_x$ single crystal

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ABSTRACT

We reported a detailed study of $Fe_{1+y}Te_{0.6}Se_{0.4}$ single crystals annealed in the atmosphere of chalcogens (O_2, S, Se, Te). After annealing with appropriate amount of chalcogens, $Fe_{1+y}Te_{0.6}Se_{0.4}$ single crystals show T_c higher than 14 K with a sharp transition width ~1 K. Critical current density J_c for the annealed crystals reach a very high value ~2–4 × 10^5 A/cm^2 under zero field, and is also robust under applied field at low temperatures. Magneto-optical imaging reveal that the J_c is homogeneously distributed in the annealed crystals and isotropic in the *ab*-plane. Our results show that annealing in the atmosphere of chalcogens can successfully induce bulk superconductivity in $Fe_{1+y}Te_{0.6}Se_{0.4}$.

1. Introduction

Among the family of iron-based superconductors (IBSs), $FeTe_{1-x}Se_x$ has received special attention due to its simple crystal structure, which is composed of only Fe(Te,Se) layers [1]. The simple crystal structure is preferable for probing the mechanism of superconductivity. On the other hand, its less toxic nature makes $FeTe_{1-x}Se_x$ a more suitable candidate for applications among the family of IBSs. However, superconductivity and magnetism of this system are not only dependent on the doping level, but also sensitive to Fe non-stoichiometry, which originates from the partial occupation of excess Fe at the interstitial site in the Te/Se layer [2,3]. The excess Fe with valence Fe^{+} will provide an electron into the system, and it is also strongly magnetic, which provides local moments that interact with the adjacent Fe layers [4]. The magnetic moment from excess Fe will act as a paring breaker and also localize the charge carriers [5,6]. To probe the intrinsic properties of $FeTe_{1-x}Se_x$ without the influence of excess Fe, some previous works have been performed to remove the effect of excess Fe by annealing in different conditions [5,7–15]. In this paper, we reported that bulk superconductivity in $Fe_{1+y}Te_{1-x}Se_x$ can be successfully induced by annealing in the atmosphere of all chalcogens, such as O_2, S, Se, and Te. The well-annealed samples show a large and homogeneous critical current density, $J_c \sim$ 2–4 × 10^5 A/cm^2 under zero field, which manifests that the chalcogens annealing is promising to be applied in fabricating high-quality iron chalcogenide wires and tapes.

2. Experimental

Single crystals with nominal compositions $FeTe_{0.6}Se_{0.4}$ were prepared from high purity Fe (99.99%), Te (99.999%), and Se (99.999%) grains [9]. More than 10 g of stoichiometric quantities were loaded into a small quartz tube with $d_1 \sim 10$ mm ϕ, evacuated, and sealed. Then we sealed this tube into a second evacuated quartz tube with $d_2 \sim 20$ mm ϕ. The whole assembly was heated up to 1070 °C and kept for 36 h, followed by slow cooling down to 710 °C at a rate of 6 °C/h. For annealing, the obtained crystals were cut and cleaved into thin slices with dimensions about 2.0 × 1.0 × 0.03 mm^3, weighed and loaded into a well-baked quartz tube ($d \sim 10$ mm ϕ) separately with appropriate amount of O_2, S, Se or Te. The quartz tube was carefully evacuated to the pressure better than 10^{-2} Torr, and sealed into the length of ~100 mm. Then the crystals were annealed at a fixed condition, 400 °C for 20 h, followed by water quenching. Magnetization measurements were performed using a commercial superconducting quantum interference device (SQUID). Microstructural and compositional investigations of the sample were performed using a scan-

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ning electron microscope (SEM) equipped with an energy dispersive X-ray spectroscopy (EDX). Magneto-optical (MO) images were obtained by using the local field-dependent Faraday effect in the in-plane magnetized garnet indicator film employing a differential method [16].

3. Results and discussion

As-grown $Fe_{1+y}Te_{0.6}Se_{0.4}$ single crystal usually shows no superconductivity or very weak diamagnetic signal below 3 K. After annealing in the atmosphere of O_2, S, Se or Te, superconductivity can be successfully induced. For O_2 annealing, the optimal condition is the molar ratio of O to the sample is ~0.015. While, for the sample annealed in S, Se, or Te vapors, the best quality sample can be obtained when the molar ratio of S/Se/Te to the sample is ~0.1. The amount of O_2 needed in annealing to obtain the best quality sample is much less than that of S, Se or Te. This difference may come from the small size of O_2, which make it possible to be easily intercalated between the layers of the single crystal, and induce the superconductivity quickly. On the other hand, for S, Se or Te, the large atom sizes hinder their intercalation into the center of the crystal. Thus, the annealing effect mainly happens from the surface of the sample. During this process, parts of the chalcogen vapor may be consumed by reacting with the surface layers. Temperature

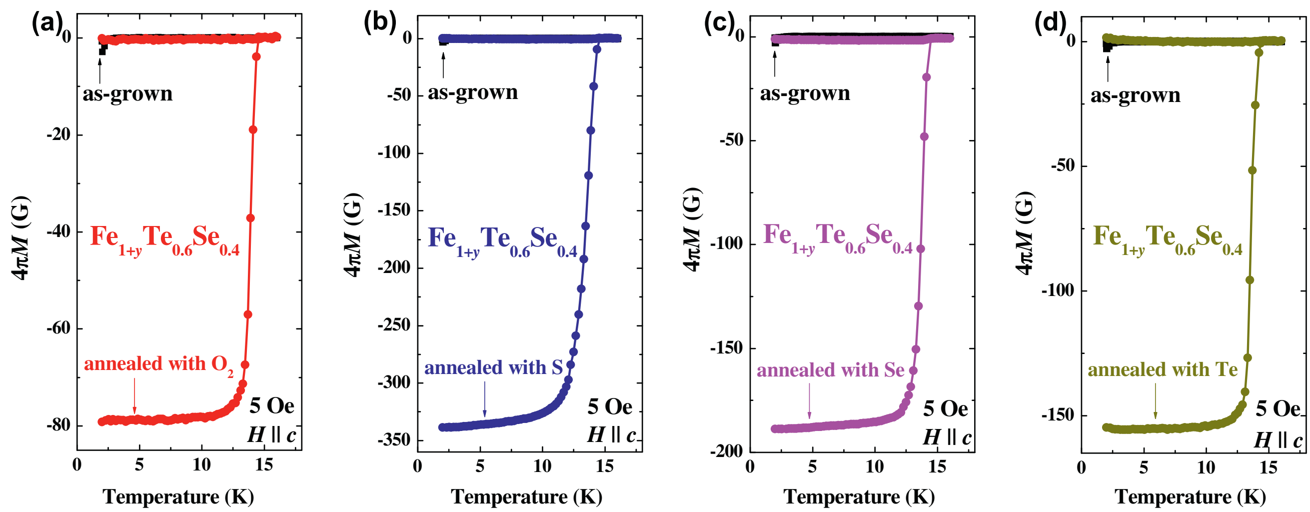

Fig. 1. Temperature dependence of zero-field-cooled (ZFC) and field-cooled (FC) magnetization at 5 Oe for $Fe_{1+y}Te_{0.6}Se_{0.4}$ single crystals annealed at 400 °C in the atmosphere of (a) O_2, (b) S, (c) Se, and (d) Te.

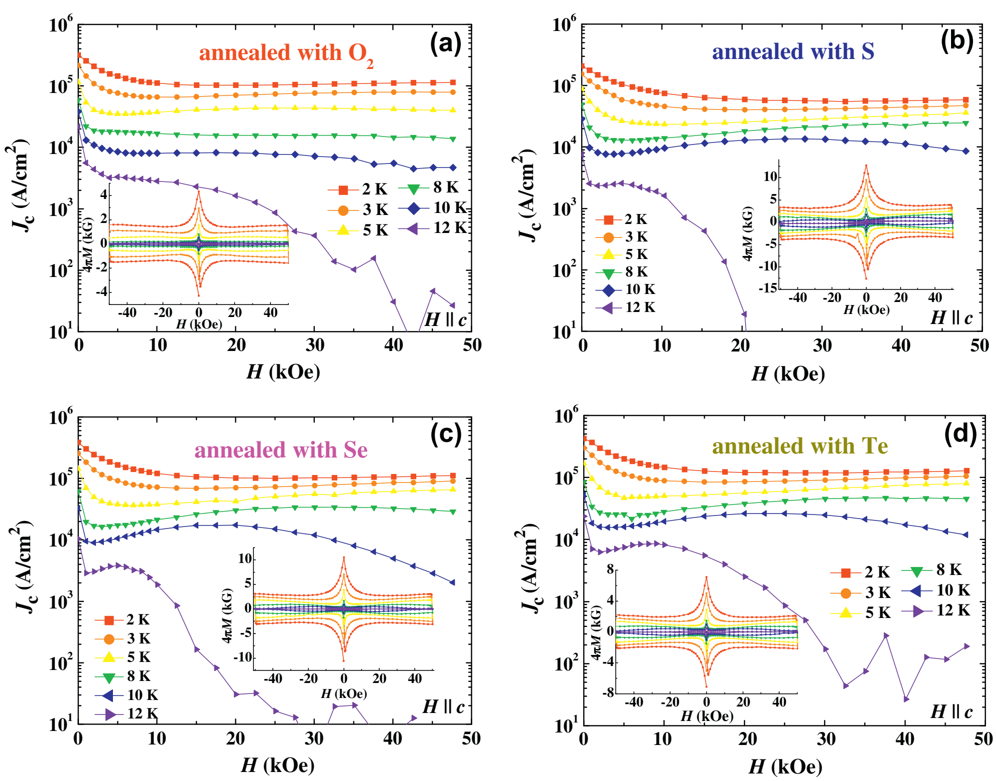

Fig. 2. Magnetic field dependence of critical current densities of (a) O_2, (b) S, (c) Se, and (d) Te annealed $Fe_{1+y}Te_{0.6}Se_{0.4}$ for $H \parallel c$ at temperatures ranging from 2 to 12 K. Insets are the magnetic hysteresis loops for the annealed samples.