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Enhanced superconducting properties in epitaxial FeSe thin films with self-assembled Fe_3O_4 nanoparticles

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ABSTRACT

In this paper we report epitaxial tetragonal iron selenide thin films grown on single crystal $SrTiO_3$ (STO) (0 0 1) and MgO (0 0 1) substrates by a pulsed laser deposition (PLD) technique. Deposition temperature and annealing process are found to be critical for achieving the tetragonal phase and the optimum superconducting properties of the films. The critical transition temperature of the thin films ranges from 2 K to 11.5 K depending on the deposition temperature and annealing condition. The samples with higher critical transition temperatures show better film crystallinity along with self-assembled Fe_3O_4 nanoparticles (~15 nm in average particle size) in the films according to both X-ray diffraction (XRD) and transmission electron microscopy (TEM) analysis. Besides the better crystallinity achieved in the films, the formation of Fe_3O_4 nanoparticles could assist the formation of the tetragonal FeSe phase and thus lead to the enhanced superconducting properties.

1. Introduction

Ever since the discovery of the iron-based superconductor of $LaFeAsO_{1-x}F_x$ [1] with the similar layered structure as cuprates, the new group of superconductors has attracted much research attention in exploring its superconducting mechanism, which could guide the exploration of the room temperature superconductor with comparison of the most studied cuprates since 1987 [2]. The discovery of the simplest iron-based superconductor iron selenide with a transition temperature T_c around 8 K [3] arouses much research interest. The superconducting FeSe has the tetragonal PbO structure containing Fe–Se planar sub-lattice with an interval of 5.518 Å. This layered structure is equivalent to the layered FeAs structure in previously found iron pnictide superconductors. FeSe provides a simple system to study the iron-based superconductors in comparison with cuprates [4]. The material contains identical Fe–Se layers similar to the layered structure in cuprates which is believed to be responsible for the superconductivity [5]. Since then, the effects of stoichiometry [6–8], structure variation [9–11], strain and stress [12], pressure [9,13,14] and doping [15,16] on its superconducting properties have been explored for FeSe.

Besides the bulk FeSe that has been explored extensively [3,10,14,15,17], the epitaxial FeSe thin films have recently attracted great research interests [12,18,19]. Most of the FeSe films reported were deposited by pulsed laser deposition (PLD). Compared with its bulk counterpart, FeSe thin film has a great potential in developing the ordered quasi-2D structure and is suitable for coating technology which has already been applied in $YBa_2Cu_3O_{7-x}$ coated conductors [20]. In the previous work on FeSe thin films, it was reported that the film thickness and orientation [18] as well as film stoichiometry and deposition temperature are critical factors for the superconducting properties [8,19,21] of FeSe thin film. Wang et al. reported the optimum deposition temperature for FeSe films on MgO is 500 °C. The T_c^{onset} and T_c^{zero} are 10 K and 4 K respectively with the film thickness of 140 nm. However the film is not in the *c*-axis orientation [18]. Nie et al. reported that the optimum deposition temperature of FeSe films with *c*-axis orientation on different substrates is 380 °C. The T_c^{onset} of the films is lower than 10 K on $SrTiO_3$ (STO) (0 0 1) and MgO (0 0 1). The upper critical field H_{c2} of the film was estimated to be 35 T with linear extrapolation method. It was also reported that the tensile strain in *a*–*b* plane suppresses the superconductivity [12]. Han et al. reported single-phased, *c*-axis oriented epitaxial $FeSe_{0.88}$ films on STO (0 0 1), (La, Sr) (Al, Ta)O_3 (0 0 1) and $LaAlO_3$ (0 0 1) with the T_c^{onset} of 11.8 K and the T_c^{zero} of 3.4 K [19]. Based on the previous studies, it is evident that the optimum growth temperatures, the film compositions and the resulted superconducting properties are largely varied. There is not yet a solid explanation for the variation of the superconducting properties of FeSe films based on the deposition conditions. The properties of FeSe films also need to be further optimized for the study on dopants incorporation.

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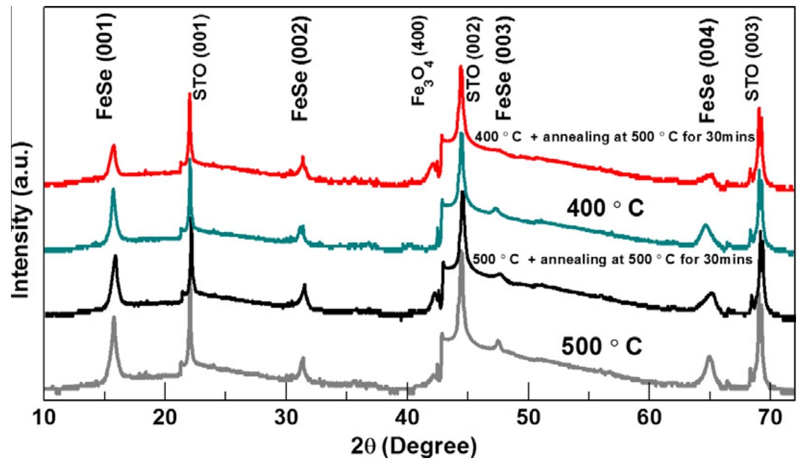

Fig. 1. XRD patterns of the FeSe thin films on STO deposited at 400 °C and 500 °C compared with the patterns for the samples after annealing at 500 °C.

In this report, we focus on the optimization of pure FeSe thin films with different growth conditions using PLD and post-annealing procedures. The microstructure properties of the films including the epitaxial quality, interface structure and secondary phase have been studied and correlated with the superconducting properties.

2. Experimental

The FeSe thin films were deposited on single crystal MgO (0 0 1) and STO (0 0 1) substrates in a PLD system with a KrF excimer laser (Lambda Physik Compex Pro 205, $\lambda = 248$ nm). The laser power density was varied from 3 J/cm^2 to 5 J/cm^2 by adjusting the laser

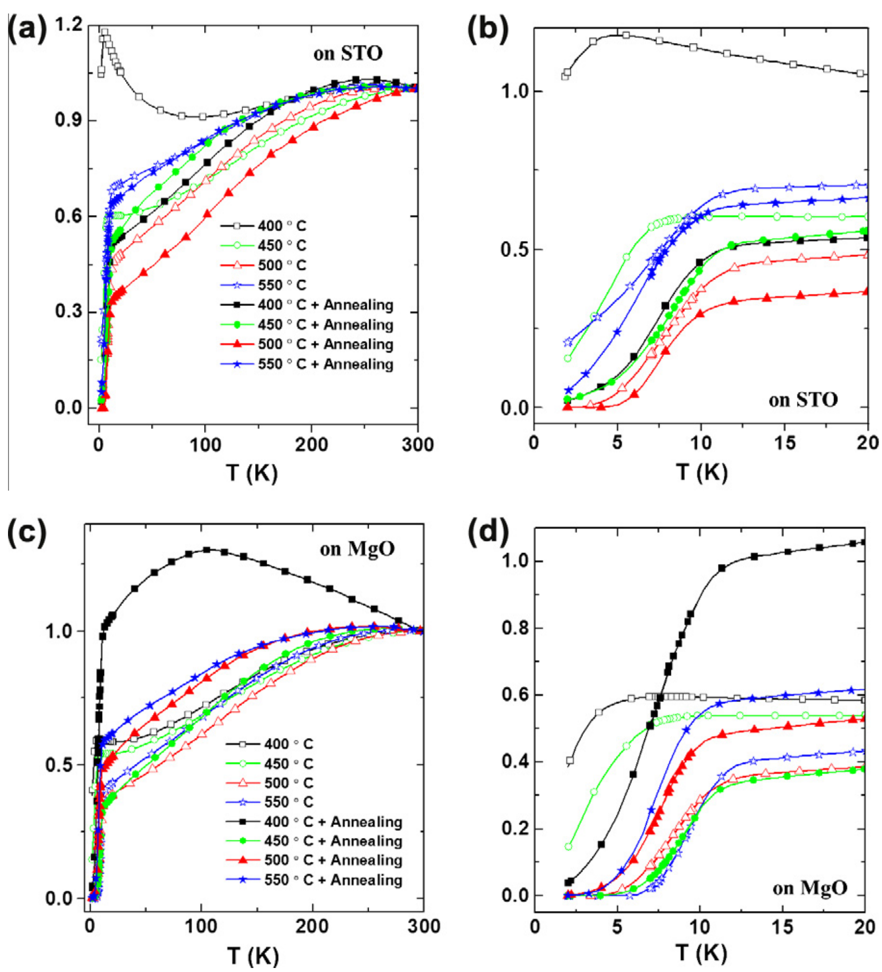

Fig. 2. Normalized *R–T* plots of FeSe thin film samples on (a) STO and (c) MgO substrates with different deposition temperatures and annealing conditions. (b) and (d) show the details from 2 K to 20 K for (a) and (b) respectively.