RAPID COMMUNICATION

Epitaxial LaFeAsO$_{1-x}$F$_x$ thin films grown by pulsed laser deposition

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Abstract

Superconducting and epitaxially grown LaFeAsO$_{1-x}$F$_x$ thin films were successfully prepared on (001)-oriented LaAlO$_3$ substrates using pulsed laser deposition. The prepared thin films show exclusively a single in-plane orientation with the epitaxial relation (001)[100] $\parallel$ (001)[100] and a full width at half-maximum value of 1$^\circ$. Furthermore, resistive measurement of the superconducting transition temperature revealed a $T_c$,90% of 25 K with a high residual resistive ratio of 6.8. The preparation technique applied, standard thin film pulsed laser deposition at room temperature in combination with a subsequent post-annealing process, is suitable for fabrication of high quality LaFeAsO$_{1-x}$F$_x$ thin films. A high upper critical field of 76.2 T was evaluated for magnetic fields applied perpendicular to the c-axis and the anisotropy was calculated to be 3.3 assuming single band superconductivity.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The discovery of high temperature superconductivity in RFeAsO$_{1-x}$F$_x$ (R = rare earth) iron pnictides [1, 2] triggered the search for new superconductors and phenomena. Recently, further iron-based superconducting compounds were discovered [3, 4]. Concerning quaternary iron pnictides (‘1111’-phase) most of the experimental investigation performed up to now used polycrystals or the available single crystals of NdFeAsO$_{1-x}$F$_x$ and SmFeAsO$_{1-x}$F$_x$ [5, 6]. Besides the challenge of single crystal growth also thin film fabrication of the fluorine doped oxypnictides has been addressed recently due to the importance of thin films for fundamental studies as well as for applications. Thin film fabrication of superconducting quaternary ‘1111’-compounds emerged to be very challenging [7, 8]. The first superconducting thin film has been grown by our group on LaAlO$_3$ substrate using pulsed laser deposition (PLD) [9, 10]. Difficulties and detailed investigation on the thin film growth of the La ‘1111’-phase is reported in [11]. To the best of our knowledge, there are no other reports on superconducting epitaxial thin films of the ‘1111’-phase. The fabrication of highly qualitative epitaxial grown thin films is mandatory for fundamental studies, e.g. on Josephson junctions, as well as for the development of multilayers or electronic devices. Furthermore, the deposition of textured thin films is so far the best alternative to the difficult single crystal growth. Due to their geometry and dimensionality, thin films open the beneficial way to a variety of experiments, such as transport current measurements, spectroscopy, multilayers, and possible new structures. Even though there are reports on single crystals of the LaFeAsO$_{1-x}$F$_x$ phase [12], no detailed measurements of physical properties have been published so far.

In this rapid communication we present the successful fabrication of epitaxial thin films of these new superconductors by pulsed laser deposition. Research using epitaxial grown LaFeAsO$_{1-x}$F$_x$ thin films offers great opportunities to study the intrinsic properties of the new iron-based superconductors. Structural as well as superconducting properties are discussed.

2. Experimental details

Thin films were deposited using a standard on-axis PLD setup with a KrF laser (Lambda Physik) with a wavelength $\lambda$ = 248 nm, a pulse duration $\tau$ = 30 ns, and an energy density $\varepsilon$ $\approx$ 4 J cm$^{-2}$ on the target surface. The total deposition time
of 2000 s with a laser pulse repetition rate of 10 Hz leads to a film thickness of app. 200 nm. Vacuum conditions in the deposition chamber were \( p = 10^{-6} \) mbar. From our early results on thin film deposition in this system we expected a high fluorine loss during deposition. Film deposition under \( \text{F}_2 \) or HF gas atmosphere might be an appropriate way to compensate for the fluorine loss, but was not considered in our experiments due to process safety concerns. Hence, a target with a rather high fluorine content was used. The nominal target composition was LaFeAsO_{1-x}F\(_x\) as calculated from the initial weight used for the target preparation.

The film presented in this publication was prepared using \textit{ex situ} phase formation. After thin film deposition using standard PLD at room temperature a 7 h heat treatment at 960 °C was applied. The post-annealing was performed in a sealed evacuated quartz tube. To avoid further fluorine or arsenide loss before phase formation a small piece of bulk material with the target composition was added. Following film deposition and \textit{ex situ} phase formation structural properties were investigated via standard x-ray diffraction methods. The superconducting transition temperature \( T_c \) was measured in a commercial physical property measurement system (PPMS Quantum Design) resistively in a four-point geometry up to 9 T.

3. Results and discussion

Detailed investigation of film growth and phase formation of the LaFeAsO\(_{1-x}\)F\(_x\) system was performed using different preparation methods [9, 11]. As result, a successful preparation technique was developed to fabricate epitaxial films grown on \((001)\)-oriented LaAlO\(_3\) single crystals as substrates. The PLD process at room temperature was applied for all kinds of substrates like SrTiO\(_3\), MgO, NdGaO\(_3\), LSAT and LaAlO\(_3\). However, only LSAT and LaAlO\(_3\) turned out to be suitable for the post-annealing process at high temperatures necessary for the LaFeAsO\(_{1-x}\)F\(_x\) phase formation; and superconducting thin films could only be prepared on LaAlO\(_3\) substrates. The x-ray diffraction pattern (figure 1) revealed a \( c \)-axis textured LaFeAsO\(_{1-x}\)F\(_x\) thin film growth. Only \((00l)\)-peaks of the LaFeAsO\(_{1-x}\)F\(_x\) phase are visible besides a small peak of LaOF, indicating nearly phase-pure \( c \)-axis textured growth. As proven by TEM investigation on our polycrystalline thin films LaOF formation is a typical feature of this fabrication process. LaOF is formed at the interface between the substrate and the film and at the film surface [11], which results in a film roughness of app. 10 nm rms as measured by AFM. Oxygen impurities like LaOF also appeared during the single crystal growth as reported in [12]. Optimizing deposition condition and phase formation heat treatment, especially by reducing the oxygen partial pressures during film preparation and annealing, we succeeded in suppressing strongly the oxygen impurity phases resulting in an epitaxial film growth.

To check the epitaxial growth of the \( c \)-axis textured LaFeAsO\(_{1-x}\)F\(_x\) thin film (figure 1), the \((112)\) pole figure (figure 2(a)) was measured. In addition the \((114)\), \((102)\) and \((314)\) pole figures were measured and considering the standard x-ray diffraction pattern (Bragg–Brentano) a \( P4/\text{mmm} \) crystal structure with lattice parameters of \( c = 0.8665 \pm 0.0005 \) nm and \( a = b = 0.407 \pm 0.003 \) nm was determined. This is a reasonable finding in fluorine doped LaFeAsO\(_{1-x}\)F\(_x\) (\( x > 0.1 \)) whereas the undoped compound has a lattice constant of \( c = 0.874 \) nm and \( a = b = 0.403 \) nm [1]. As reported by Chen et al [13], the fluorine doping reduces the \( c \)-axis lattice constant. The pole figures reveal a sharply textured in-plane aligned film. The in-plane orientation distribution of about 1° was determined by an extra phi-scan of the \((112)\) peaks depicted in figure 2(b). Furthermore, the phi-scan indicates,
that only one single epitaxial component is present with the epitaxial relationship (001)[100] \parallel (001)[100].

Resistive measurement results of the superconducting transition are shown in figure 3. \( T_{c,90\%} \) of the epitaxial LaFeAsO\(_{1-x}\)F\(_x\) film is 25 K whereas \( T_{c}(R = 0) \) is at 17 K. The quality of the textured thin film emerges in the high residual resistive ratio (RRR = 6.8). Figure 3(b), (c) show the resistive behaviour of the thin film in an applied magnetic field up to 9 T in both major directions: figure 3(b) shows out-of-plane (parallel to \( c \)-axis) and figure 3(c) in-plane (perpendicular to \( c \)-axis) measurements.

Based on \( R(T) \) measurements, part of the magnetic phase diagram has been plotted (figure 4). The upper critical field, \( \mu_0 \mu H_\parallel(0) \), evaluated by the \( T_{c,90\%} \) criterion, i.e. 90% of \( R_{\text{normal}} \), is approximated by the Werthamer–Helfand–Hohenberg (WHH) model for a single band \( (\mu_0 \mu H_\parallel(0) = 0.693 T_c \times \frac{d \ln \mu H_\parallel}{d T} |_{T_c}) \). The irreversibility line, evaluated by the \( T_{c,10\%} \) criterion, is fitted using a power law with exponent 1.5. The upper critical field for magnetic fields applied parallel to the \( c \)-axis was determined to be \( \mu_0 \mu H_\parallel(0) = 23.2 \) T with a slope of \( \frac{d \ln H_\parallel}{dT} |_{T_c} = -1.34 \) T K\(^{-1}\). In the case of an applied field perpendicular to the \( c \)-axis, the upper critical field has a value of \( \mu_0 \mu H_\perp(0) = 76.2 \) T with a slope of \( \frac{d \ln H_\perp}{dT} |_{T_c} = -4.4 \) T K\(^{-1}\) which is very similar to bulk data [14]. Consequently, the anisotropy factor was determined to be \( \gamma = 3.3 \) based on single band calculations.

4. Summary

In this study we report the first successful epitaxial growth of superconducting LaFeAsO\(_{1-x}\)F\(_x\) thin films with a \( T_{c,90\%} \) of 25 K and a single in-plane orientation with FWHM of 1°. The epitaxial relationship to the LaAlO\(_3\) substrate is (001)[100] \parallel (001)[100]. We determined the anisotropy of the upper critical field to be 3.3 with a high \( \mu_0 \mu H_\perp(0) = 76.2 \) T perpendicular to the \( c \)-axis. The anisotropy was calculated with the assumption of single band superconductivity. Measurements on polycrystalline bulk LaFeAsO\(_{1-x}\)F\(_x\) (\( x = 0.11 \)), however, indicate a two band behaviour [15]. To investigate the full magnetic phase diagram and clarify one or multiband behaviour high field measurements on this epitaxially grown thin films are required.

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