Neutron scattering study of magnetic $Fe_{1+d}Te$ thin films grown under tensile stress

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Polarized neutron scattering reveals the presence of magnetic order in $Fe_{1+d}Te$/MgO ($0.04 \leq d \leq 0.1$) grown under tensile stress. Neutron diffraction data shows the magnetic ordering with propagation vector (-0.5 0 -0.5) and the transition temperature is found $T_N \approx 67$K from the temperature dependent measurement. In addition possible understanding of the strain effect on the lattice parameters of FeTe is discussed based on the present neutron data.

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I. INTRODUCTION

Neutron scattering has been the scattering technique of choice for the analysis of magnetic structures and their dynamics for many decades. The advent of magnetic thin film systems has posed new challenges since such samples have inherently small scattering volumes. More particularly, in the recently discovered Fe-based materials[1], the antiferromagnetic and superconducting states appear to be more intimately connected and, superconductivity and magnetic order coexist in the phase diagrams.[2] In one of different families of Fe-based superconductors, for the "11" Fe-chalcogenides (Ch= Se, Te), the tetragonal $Fe_{1+y}Se$ with T_c=8 K can be considered as a reference material due to its binary atomic pattern.[3] $Fe_{1+d}Te$ with an analogous crystal structure to the superconducting $Fe_{1+y}Se$, shows a complex interplay of magnetic and structural phase transitions depending on the excess amount of Fe.[4–6]

Successful fabrication of superconducting monolayer FeSe on $SrTiO_3$ substrate represented a real breakthrough in searching for FeCh superconducting material with higher T_c.[7] Experiments on FeSe thin films with different orientations, that affect the structural distortion, showed that superconductivity is strongly sensitive to the structural transition. The structural distortion lowers the symmetry from tetragonal to orthorhombic, as does the nematic transition in LaFeAsO[8] , but in FeSe it is not followed by magnetic ordering at lower temperatures, and the transition is not caused by the magnetic order. However, it is accompanied by a resistivity anomaly, but not as strong as that in pnictides.[9] This tetragonal to orthorhombic transition then seems to be different in nature from the transition in the pnictides and still not very well understood. The tetragonal to orthorhombic distortion in FeSe is also different from the distortion seen for FeTe, which is directly connected with and coincident with the double-stripe magnetic order[2,6]. In the non-superconducting FeTe bulk samples the first order magnetic and structural transition occurs around 70 K and is accompanied by anomaly in resistivity and susceptibility measurements.[10] On the other hand, $Fe_{1+d}Te$ was expected to have a strongest spin density wave (SDW) ordering and a highest transition temperature in the "11" family.[11] This argument was supported by the increase of T_c up to 37 K under high pressure in FeSe[12], and in that case the application of pressure is found to enhance spin fluctuations. That gave the motivation to make superconducting FeTe thin films using different methods. For exemple $Fe_{1.08}Te:O_x$ thin films epitaxially grown on $SrTiO_3$ substrates in a controlled oxygen atmosphere have been reported as superconducting.[13] Tensile strain was also used during the growth and is reported as being at the origin of superconductivity in FeTe thin films.[14] Han *et. al.* reported superconductivity at ∼13 K in $Fe_{1+d}Te$ thin films grown on different substrates (MgO, $SrTiO_3$, $LaAlO_3$), and attributed the emergence of superconductivity to the tensile stress. They have observed in-plane extension and out-of-plane contraction in all FeTe films, independent of the lattice mismatch. While for the non-superconducting FeTe bulk samples, a first order magnetic and structural phase transition occurs around 70 K, it is was not clearly reported whether SDW exists or not in FeTe thin films.[14]

In this work we present our results from neutron diffraction experiment performed on $Fe_{1+d}Te$ thin films. Polarized neutrons were used as a probe for magnetism and superconductivity in $Fe_{1+d}Te$ thin films grown on MgO substrate. We show various number of reflections coming from $Fe_{1+d}Te$ and/or from the MgO substrate. The magnetic Bragg peak from the film was observed at commensurate position (-0.5 0 -0.5) in contrast to what is reported previously in Ref. [4–6]. The magnetic ordering transition that occurs around ∼67 K clarified the origin of unclear resistivity anomaly observed previously in $Fe_{1+d}Te$ thin films.[14] Because of the very les amount

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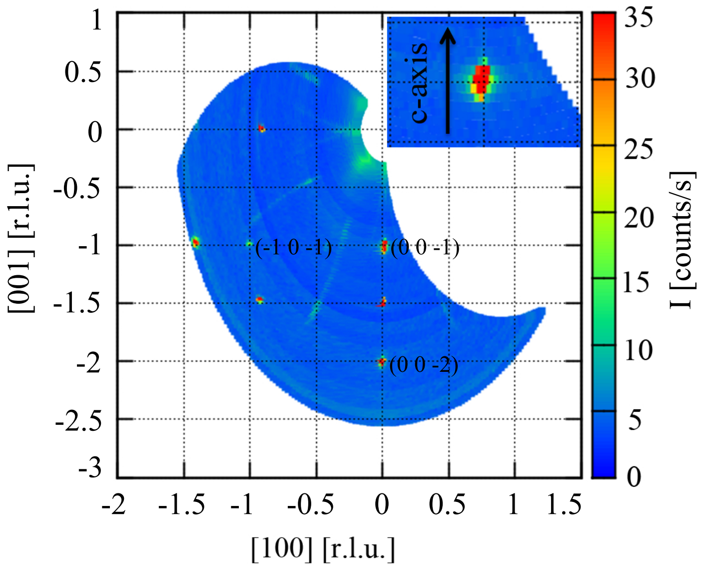

FIG. 1: (Color online) Color-coded map for the polarized neutron diffraction intensities from 21 pieces of 90 nm $Fe_{1+d}Te$ thin films for non spin-flip channel reflections under x-field at 5 K. Structural peaks for FeTe thin film are labelled on the map as (0 0 -1), (0 0 -2) and (-1 0 -1). Reflections appearing at incommensurate position close to following points (-1 0 0) and (0 0 -1.5) are structural peaks from the substrate both coming from $\lambda/2$ contribution. Reflections close to (-1 0 -1.5) and (-1.5 0 -1) points were also expected coming from $\lambda/2$ contributions. The inset represent a zoom on the structural Bragg peak (0 0 -1). This map is obtained using the $Fe_{1+d}Te$ thin film lattice constants presented in Table I. Here the axes are defined as [100] [r.l.u.]$= 2\pi/a$ and [001] [r.l.u.]$= 2\pi/c$ respectively.

of scattering material (total mass of FeTe $\sim$ 1.4 mg) from the thin film, and the experimental resolution limit, we didn not observe a clear indication of superconducting signal.

II. EXPERIMENTAL DETAILS

Polarized neutron scattering experiment has been performed on superconducting $Fe_{1+d}Te$ thin films pulsed laser deposited from targets with nominal composition of $FeTe_{1.4}$ on MgO substrate under based pressure of 4×10^{-5} Pa and at $\sim$ 540°C.[14] Thin films are grown with different thickness of 60, 90, 120 and 150 nm, only those with thickness of 90 nm with maximum T_c have been used in this work. Chemical analysis using Energy-dispersive X-ray spectroscopy (EDAX) has been performed and Fe:Te ratio is found to be from 1.04 to 1.10 over 1. No

Compound	a (Å)	c (Å)
$Fe_{1+d}Te$ Film	3.86	6.31
$Fe_{1+d}Te$ Bulk[19]	3.834	6.249
MgO Substrate	4.21	4.21

TABLE I: Lattice parameters of strained $Fe_{1+d}Te$ and MgO substrate.

Se contaminations were reported in the $Fe_{1+d}Te$ thin films. Polarized neutron diffraction experiments were performed on 21 $Fe_{1+d}Te$ thin film samples with thickness of 90 nm stacked together in a thin Al sample holder. The c-axis of thin films were oriented parallel to the incident neutron beam, and the experiment was performed in a temperature range varying from 3.5 K to 95 K. The thin film assembly was oriented by taking into account the scattering plane [001] identified before the neutron scattering experiments using X-ray Laue diffraction based on the MgO substrates simple cubic structure. The magnetic structure of superconducting $Fe_{1+d}Te$ thin films was studied by polarized neutron diffraction using the DNS (Diffuse Neutron Scattering) instrument of FRM II reactor in Garching (`http://www.mlz-garching.de/dns`). Neutron diffraction studies of thin films are quite challenging due to the fact that the signal coming from the substrate is usually higher then the one coming from the sample itself, e.g. in our case only 0.01% of the total volume is constituted by the $Fe_{1+d}Te$, and its total mass is less then $\sim$1.4 mg. Therefore neutron experiments were performed on a packed sample assembly in order to enhance the signal coming from the sample. Samples were mounted together with the [100] direction of $Fe_{1+d}Te$ thin films vertically in order to access the (h0l) type reflections in the horizontal plane. Data is corrected for detector efficiency and for the flipping ration using the vanadium and

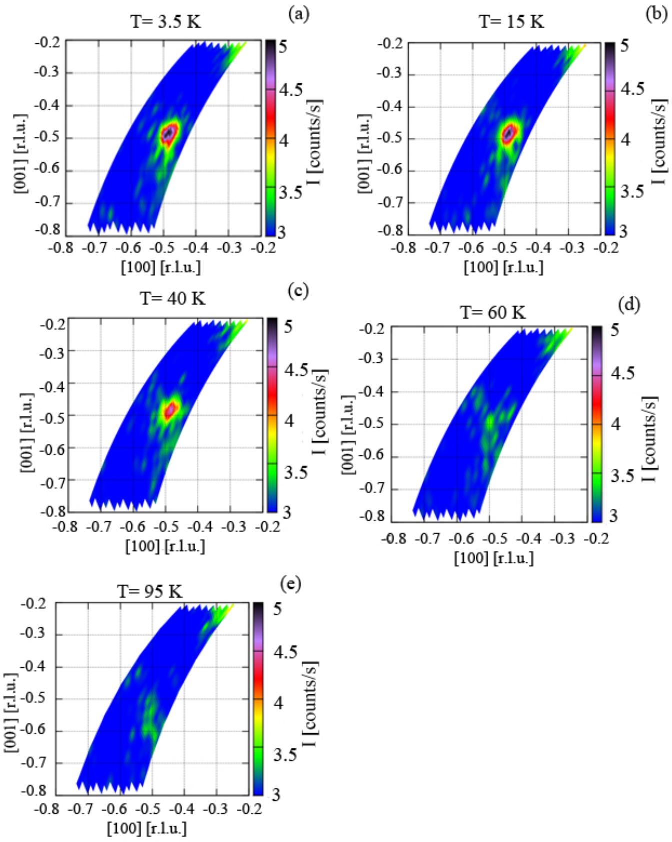

FIG. 2: (Color online) (a-e)Temperature dependence of the magnetic reflection (-0.5 0 -0.5) from the $Fe_{1+d}Te$ thin films measured with polarized neutron diffraction in a temperature range varying from 3.5 K to 95 K.

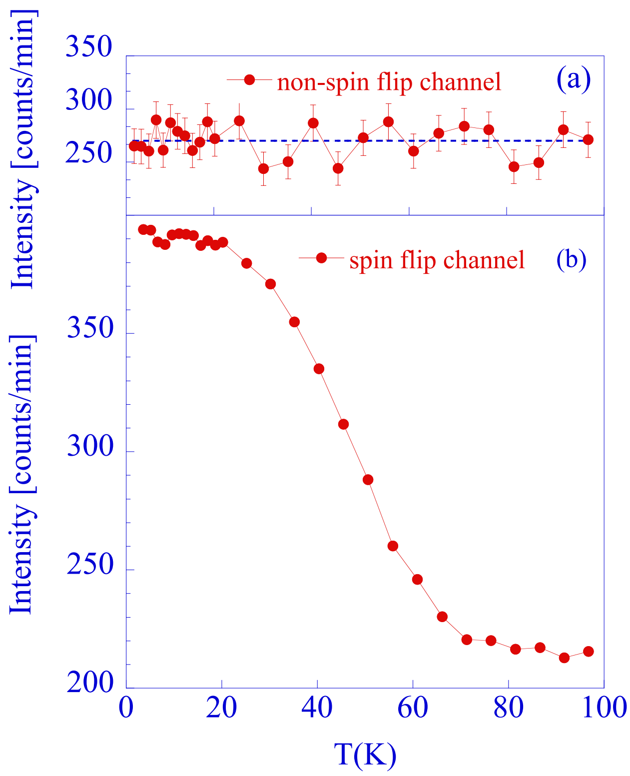

FIG. 3: (Color online) (a)Intensities of the magnetic peak of $Fe_{1+d}Te$ thin films for non-spin flip channel. No discontinuity is found due to any coexistence of magnetism and superconductivity. (b) Temperature dependence of the spin-flip channel counts of the magnetic peak intensity for $Fe_{1+d}Te$ thin films. The transition temperature is extracted as 67 K. Here the errors bars are within the marker size.

NiCr standard samples respectively. While the reflections coming from $\lambda/2$ contamination were still detectable.

III. RESULTS AND DISCUSSION

A. Magnetism in $Fe_{1+d}Te$ thin films

Figure 1 (a) show the color coded map of the diffracted intensity in the non spin-flip (nsf) channel measured at 5 K. In this channel only reflections from structural peaks will be available. The reflections labelled as (0 0 -1), (0 0 -2) and (-1 0 -1) are the structural Bragg peaks of $Fe_{1+d}Te$ thin film. Reflections seen at incommensurate position close to the points (-1 0 0) and (0 0 -1.5) were identified as structural peaks coming from $\lambda/2$ of the MgO substrate. Here the latter peak has both contribution from (0 0 3) FeTe and (0 0 2) MgO structural peaks. Reflections close to (-1 0 -1.5), and (-1.5 0 -1) are expected to be from higher order scattering. From the simple principle that FeTe thin films are grown under tensile strain, one may assume that FeTe adopt the lattice parameters of MgO substrate during the growing process. In Figure 2 (a-e) we present the temperature evolution of the magnetic Bragg peak (-0.5 0 -0.5) of $Fe_{1+d}Te$ using color-coded maps of the reciprocal spaces covering the peak vicinity. Maps were recorded at different temperatures starting from the base temperature 3.5 K, that allowed us to see the magnetic transition around ∼67 K where the peak intensity disappears. The magnetic transition temperature can be accurately measured as shown in Figure 3 (b) where the temperature dependence of neutron diffraction intensity is presented. The obtained value for the transition temperature is $T_N \approx 67$ K is in agreement with values determined for the bulk system.[4–6,15]

Low temperature magnetic structure of $Fe_{1+d}Te$ bulk system has been reported to be highly sensitive to the stoichiometry[4,5], and it has been argued that the amount of interstitial Fe ions is a parameter that tunes the magnetic ordering peak from commensurate to an incommensurate position. In addition $Fe_{1+d}Te$ undergoes different structural phase transitions driven by the Fe-content.[16] Recent neutron diffraction measurements indicated the same commensurate ordering wave vector (0.5 0 0.5) for $Fe_{1.075}Te$[4] and $Fe_{1.068}Te$[6] where the low temperature crystal structure is found to be monoclinic. While for $Fe_{1+d}Te$ polycrystalline samples the low temperature magnetic order is reported[16]to be commensurate within the $0.06 < d < 0.11$ doping range.

Figure 2 shows the magnetic Bragg peak of $Fe_{1+d}Te$ thin films at commensurate position (-0.5 0 -0.5). The earliest characterization of $Fe_{1+d}Te$ gave a rather large ordered magnetic moment of 2.07 μB with components of magnetic moment along all crystallographic axes.[15] Recent studies suggested that the moment direction is in the b-axis direction including also projections of the moment along the a and c axes with 0.72(2) μ_B and 0.7(1) μB respectively. Finite moments along the c-axis were attributed to the finite moments of the excess Fe ions.[6] The average calculated magnetic moment was reported such as 1.78 μ_B/Fe for powder samples and similar values 1.9 μ_B were reported for $Fe_{1.1}Te$ polycrystalline samples.[17] From the present neutron diffraction data we have estimated the magnetic moment as $\sim 2\ \mu_B$ using the integrated intensities of the magnetic and nuclear Bragg peaks.[17,18] Here the contribution from interstitial Fe atoms to the estimated magnetic moment has been nearly neglected since the system under study is a thin film grown under tensile stress. Nevertheless, the estimated moment is in agreement with those reported for bulk $Fe_{1+d}Te$.

B. Strain effects on iron-chalcogenide thin films

Figure 1 shows the color-coded map of the diffracted intensity in the non spin-flip (nsf) channel obtained using the using the unstrained FeTe lattice parameters by slightly arranging them in order to fit the reciprocal space grid notations and this by referring to the highest intensity nuclear peaks. This allowed us to extract experimentally a and c lattice constant for tensile-strained $Fe_{1+d}Te$ thin film shown in Table I. Here lattice constants value for the bulk material are taken from ref. [19] from crystals having the same Fe content. Neutron data shows that thin film under tensile stress is not completely adopting the substrates lattice parameters but only accommodates to them. Both a and c axes lattice parameters for the tensile-strained film are slightly different then from the bulk material. One can interpret this difference as following; in $Fe_{1+d}Te$ thin film only a few layers at immediate vicinity of the MgO substrate are mostly affected by the tensile strain and are forced to adopt the MgO lattice constants. While when one goes to upper layers of $Fe_{1+d}Te$ a relaxation occurs in the lattice parameters and one ends up with unstrained $Fe_{1+d}Te$ lattice. Indeed the effect of the relaxation can be seen in the neutron data presented in the inset of Figure 1 where the structural peak (0 0 -1) of $Fe_{1+d}Te$ thin film is broadened (stretched) along the c axis. Recent DFT study on the effect of in-plane strain in bulk and monolayer iron-chalcogenides revealed that low dimensionality induce superconductivity in FeTe[20], and for both bulk and monolayer (ML) form, magnetic phase transition is reported upon increasing the tensile strain. Magnetization measurements performed on $Fe_{1+d}Te$/MgO thin films showed that 22% of the total volume is superconducting.[14] While present neutron scattering experiment performed on a total scattering mass less then $\sim$1.5 mg shows has no clear indication (deep) for the emergence of superconductivity at low temperature (see in Fig. 3 (b)). Some confusing discontinuity is still visible in this regime. We assume that the expected superconducting signal is beyond the detectable limit of polarized neutrons. An additional approach might be that the superconductivity is only present in a few interface layer of FeTe in the vicinity of the substrate. For example, it was argued that the spin density wave (SDW) is suppressed, in the FeSe monolayer deposited on $SrTiO_3$[21], by the charges transferred from the oxygen-vacancy-induced states of the substrate.

IV. CONCLUSIONS

We have successfully observed magnetic and structural Bragg peaks using polarized neutron diffraction method in $Fe_{1+d}Te$ thin films, with a total mass less the $\sim$1.4 mg, grown substrate under tensile stress on MgO . In $Fe_{1+d}Te$ thin films magnetic phase transition was observed around 67 K, and magnetic peak with propagation vector $\mathbf{k}$=(0.5 0 0.5) is found at commensurate position with an estimated moment of $\sim 2\ \mu_B$. The effect of tensile strain visible on structural peaks, was proposed to be effective only at the interface layers of FeTe while both in plane and out-of-plane lattice parameters are relaxed giving rise to very broad structural Bragg peaks along the [001] axis.

Acknowledgements

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