Pressure-induced ferromagnetism in antiferromagnetic Fe_{1.03}Te

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The magnetic properties of Fe_{1.03}Te under hydrostatic pressure up to $p \simeq 5.7$ GPa were investigated by means of muon spin rotation, dc magnetization, and neutron depolarization measurements. All of these methods show consistently that the commensurate antiferromagnetic order is weakened with increasing pressure before the system becomes a low temperature bulk ferromagnet above $p \simeq 2$ GPa.

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By applying hydrostatic pressure the magnetic and superconducting properties of iron-based superconductors can be directly controlled, since the carrier concentration and the exchange interaction through the compressed lattice are changed.^{1–4} Within this new class of superconductors, the iron chalcogenides have attracted considerable interest, because this system shows a large effect on the superconducting and magnetic properties upon both chemical and hydrostatic pressure. Substitution of Se by Te in superconducting FeSe leads to almost a doubling of the value of the superconducting transition temperature T_c until the system enters a state of coexistence of superconductivity and magnetism, ending at antiferromagnetic FeTe.⁵ On the other hand, application of hydrostatic pressure leads to a nonlinear increase of T_c with a maximum value of $\simeq 36$ K.⁶ Furthermore, magnetic order appears and superconductivity and magnetism coexist on a microscopic level in the whole sample.^{7,8} In antiferromagnetic FeTe a T_c even higher than that of FeSe was predicted,^{9–11} but not observed until now.¹² Instead, the parent compound FeTe exhibits peculiar magnetic properties. At the Néel temperature $T_N \simeq 70$ K a drastic drop in the magnetic susceptibility is seen.^{13,14} This is related to a first-order phase transition to a monoclinic crystal structure at T_S that coincides with the appearance of commensurate antiferromagnetism at T_N .¹⁵ However, at ambient pressure the magnetic properties of FeTe depend strongly on the amount of excess Fe.^{16,17} For nearly stoichiometric Fe_{1+x}Te a distorted monoclinic structure and a commensurate antiferromagnetic order in the ab plane was observed at low temperatures. In contrast, at a high amount of excess Fe the value of T_N decreases, and the magnetic order changes to an incommensurate antiferromagnetic one.^{16,17}

Pressure studies of the electrical resistivity of FeTe_{0.92} up to 19 GPa revealed that the anomaly in the resistivity at T_S shifts toward lower temperatures with increasing pressure.¹² Additionally, at high pressures a new anomaly in resistivity emerges, strongly indicating that the electronic properties of Fe_{1+x}Te are closely correlated with its crystal structure.

In this Rapid Communication, a systematic investigation of the magnetic properties of Fe_{1.03}Te under hydrostatic pressure using neutron depolarization, magnetization, and muon spin rotation (μ SR) experiments is reported. All three experimental

techniques reveal consistently a transition from a low pressure antiferromagnetic phase to a high pressure bulk ferromagnetic phase in Fe_{1.03}Te. Note that only a few compounds show similar behavior under pressure.^{18–21} The single crystals were grown by the Bridgman method similar to the ones prepared in Ref. 5. The samples were partially ground into powder for the experiments.

The neutron depolarization experiments on powderized crystals of Fe_{1.03}Te were performed at the Paul Scherrer Institute (PSI), Switzerland, using the TASP three-axis spectrometer equipped with a MuPAD polarimeter.^{22,23} Figure 1(a) presents data of the polarized neutron experiments taken on Fe_{1.03}Te in a piston cylinder cell with a neutron wavelength of 3.2 Å. In these measurements the polarization of a monochromatic neutron beam is measured after transmission through the sample. Because of the Larmor precession of the neutrons, the polarization of the beam will rotate around the randomly aligned ferromagnetic domains, resulting in a loss of initial polarization. The temperature dependence of the neutron polarization at ambient pressure shows no loss of polarization, whereas at $p \simeq 2$ GPa the neutrons lose $\sim 5\%$ of their polarization below the ferromagnetic ordering temperature T_C (2 GPa) $\simeq 60$ K [see Fig. 1(a)].

The ferromagnetic behavior of Fe_{1.03}Te under pressure is further evidenced by field-dependent magnetization measurements taken with a commercial Quantum Design 7 T magnetic property measurement system (MPMS) XL superconducting quantum interference device (SQUID) magnetometer. The measurements up to $p \simeq 6$ GPa were performed on a single crystal with a mass of $\simeq 90$ μ g in a diamond anvil cell, especially designed for magnetization measurements.²⁴ The field dependencies of the magnetization after subtraction of the zero pressure measurements at 10 K are shown in Fig. 1(b). The typical hysteretic behavior of a ferromagnet is obvious above $p \simeq 1.7$ GPa. At higher pressure this behavior is more pronounced. However, the coercive field seems to be maximal with a value of 0.35 T at $p \approx 2$ GPa and reaches ~ 0.15 T with increasing pressure. Furthermore, the pressure dependence of the estimated saturated ferromagnetic moment in units of μ_B was extracted from the magnetization measurements. As can be seen in the inset to Fig. 1(b), it increases with increasing

FIG. 1. (Color online) (a) Temperature dependence of the neutron polarization of Fe_{1.03}Te at ambient pressure and at $p = 2$ GPa. A clear loss of polarization is observed at $p = 2$ GPa below $T \simeq 60$ K, indicating that the sample is ferromagnetic. (b) The measured hysteresis loops of Fe_{1.03}Te at $T = 10$ K after subtraction of the ambient pressure data. Above $p \simeq 1.7$ GPa, for a ferromagnet, the typical opening of the hysteresis loop is evident, indicating the appearance of ferromagnetism in the sample under pressure. The inset in (b) shows the estimated saturated magnetic moment in units of μ_B as a function of pressure at $T = 10$ K. The dashed line is a guide to the eyes. (c) Temperature dependence of the magnetic moment m of Fe_{1.03}Te at $\mu_0 H = 0.1$ T after subtraction of the zero pressure data for various pressures. The inset shows the determination of the onset temperature T_C of ferromagnetic ordering by linear extrapolation of m to zero.

pressure, reaching a value of $\sim 3\mu_B$ per Fe atom at the highest pressure investigated. This strongly suggests the occurrence of bulk ferromagnetism in FeTe under pressure.

The evaluation of the ferromagnetic state was further investigated by means of temperature-dependent magnetization measurements in a magnetic field of $\mu_0 H = 0.1$ T shown in Fig. 1(c). Similar to the field dependencies, the ambient pressure measurements were subtracted in order to extract the ferromagnetic signal of the sample. In addition, the

paramagnetic contribution above T_C of the 2.3 GPa data was fitted to the expression $m \propto aT + C/T$ and subtracted for all pressures. The onset of the ferromagnetic ordering T_C was determined from the intersection of the linear extrapolation of the magnetic moment close to T_C with the zero line [see the inset of Fig. 1(c)].

A microscopic view of the magnetic properties of Fe_{1.03}Te is obtained by μ SR measurements performed on the μ E1 beam line with the GPD instrument at PSI. In such experiments spin polarized muons are implanted into the sample. By monitoring the time evolution of the muon spin polarization, information on the local magnetic field at the muon stopping site $B_\mu^{\text{af/f}}$ and the magnetic volume fraction M_{tot} are obtained (af and f refer to the antiferromagnetic and ferromagnetic states, respectively).

The internal magnetic field distribution of Fe_{1.03}Te was investigated for different pressures by means of zero field (zf) μ SR at 10 K. The obtained time spectra presented in Fig. 2(a) show a spontaneous precession of the muon spins, indicating a long-range ordered magnetic state in the sample. The μ SR asymmetry $A(t)$ with the corresponding zero time values indicated in Fig. 2(a) was analyzed using the free software package MUSRFIT.²⁵ Here, $A(t)$ is described by a model consisting of a superposition of two typical functions describing magnetic signals:²⁶ one for the antiferromagnetic g_{af} and one for the ferromagnetic g_{f} oscillations. M^{af} and M^{f} describe the relative contributions of the ferromagnetic and the antiferromagnetic phases, respectively, to the total normalized magnetic volume $M_{\text{tot}} = M_{\text{af}} + M_{\text{f}} = A(t = 0, T)/A(t = 0, T = 0)$:

$$A(t) = A(0) [M_{\text{af}} g_{\text{af}}(t) + M_{\text{f}} g_{\text{f}}(t)], \quad (1)$$

where the muon polarization function $g_{\text{af/f}}(t)$ has the typical shape

$$g_{\text{af/f}}(t) = (1 - a)e^{-\Lambda_{\text{af/f}}^t} \cos(\omega_{\text{af/f}} t + \phi_{\text{af/f}}) + ae^{-\lambda_{\text{af/f}}^t}. \quad (2)$$

Here a is the fraction of the muon spins parallel and $(1 - a)$ the fraction perpendicular to the magnetic moments with a typical value of $a \simeq 1/3$. $\Lambda_{\text{af/f}}^t$ and $\lambda_{\text{af/f}}^t$ describe the transverse (t) and longitudinal (l) relaxations in the antiferromagnetic and ferromagnetic parts, respectively, with the corresponding precession frequencies $\omega_{\text{af/f}}$. $\phi_{\text{af/f}}$ is the initial phase of the muon ensemble.

The muon precession frequencies extracted from the μ SR spectra are directly proportional to the local magnetic field $\omega_{\text{af/f}} \propto B_\mu^{\text{af/f}}$ at the muon stopping site. As seen in Fig. 2(b), B_μ^{af} at 10 K decreases with increasing pressure. This indicates that the antiferromagnet moment decreases, and thus the ordering is weakened. For higher pressures close to the ferromagnetic state ($p \sim 1.7$ GPa) the magnetic order changes to a more complex one. This is seen in the steep increase of the longitudinal relaxation λ_{af}^l just before the ordering [Fig. 2(c)]. In addition, the muon phase ϕ_{af} changes at this pressure from zero to $\simeq 60^\circ$ [see Fig. 2(c)]. This appears to be consistent with the occurrence of incommensurate order that is supposedly indicated by a recent neutron study.²⁷ In the ferromagnetic state ($p \gtrsim 1.9$ GPa) an additional high field of $B_\mu^{\text{f}} \gtrsim 1.7$ T is observed in the μ SR time spectra which is allocated to the ferromagnetic moment and is substantially larger than B_μ^{af} [see Fig. 2(a)]. This clearly indicates a change in the microscopic structure. The ferromagnetic volume fraction M_{f}

FIG. 2. (Color online) (a) Selected zF μ SR time spectra $A(t)$ of $\text{Fe}_{1.03}\text{Te}$ for various pressures at $T = 10$ K. In the ferromagnetic state at high pressures $p \geq 1.9$ GPa, an additional high local magnetic field $B_{\mu}^f \gtrsim 1.7$ T appears in the spectra. The lines are fits to the data. The tick labels on the asymmetry axis refer to the zero time value of each spectrum. (b) Pressure dependence of the internal magnetic fields B_{μ}^{af} (left axis) and B_{μ}^f (right axis). (c) Pressure dependence of the longitudinal relaxation rate λ'_{af} at 10 K (left axis). The right axis shows the pressure dependence of the initial muon phase ϕ_{af} . (d) Pressure dependence of the volume fraction M_f of the ferromagnetic phase. (e) Temperature dependence of the magnetic fraction M_{tot} for selected pressures. (f) Temperature dependence of the normalized additional relaxation $\lambda^{pc}/\lambda_0^{pc}$ of the pressure cell due to the ferromagnetic state of the sample. The lines in (b), (c), (e), and (f) are guides to the eyes.

obtained from the zF μ SR measurements is shown in Fig. 2(d). Obviously, M_f is increasing continuously until at $p = 2.4$ GPa the long-range ferromagnetic order is fully established.

The magnetic transition temperatures were obtained by monitoring the temperature dependence of the magnetic volume fraction M_{tot} in weak transverse field (wtf) μ SR measurements [see Fig. 2(e)]. In such measurements, a wtf is applied perpendicular to the initial muon spin polarization. The muons stopping in a magnetic region lose their polarization very fast (within 100 ns), leading to a reduction of the muon asymmetry in the magnetic parts of the sample. In contrast, the muons stopping in the nonmagnetic parts of the sample give rise to long living oscillations, representing the coherent precession of the muon spins around the external magnetic field. The ordering temperatures are taken from the midpoint of the transition. It is rather sharp in the antiferromagnetic state, since it is first-order-like.⁵ In contrast, the ferromagnetic transition is significantly broader. The ferromagnetic ordering temperature T_C can also be determined from these measurements with the help of the pressure cell signal (especially in the pressure range where T_C is smaller than T_N). In the ferromagnetic state $\text{Fe}_{1.03}\text{Te}$ is magnetized in the weak field applied. Thus, the stray field leads to an increase of the field inhomogeneities in the pressure cell, and consequently the signal of the pressure cell shows an additional relaxation λ^{pc} [see Fig. 2(f)]. Note that the relaxation of the pressure cell itself is pressure independent.

The values of T_C and T_N extracted from the neutron, magnetization, and μ SR measurements are summarized in a phase diagram exhibiting different magnetic states (see Fig. 3). At ambient pressure Fe_{1+x}Te exhibits commensurate antiferromagnetic ordering along the $(\pi, 0)$ direction.¹⁴ The magnetic transition at T_N is accompanied by a first-order structural phase transition. Application of pressure first leads

FIG. 3. (Color online) Pressure dependence of the magnetic ordering temperatures T_N and T_C of $\text{Fe}_{1.03}\text{Te}$. AFM denotes the antiferromagnetic, FM the ferromagnetic, and PM the paramagnetic state. At $p \simeq 1.6$ GPa the AFM changes to an incommensurate magnetic state. Above $p \sim 2$ GPa, $\text{Fe}_{1.03}\text{Te}$ becomes a bulk ferromagnet. In the crossover region from the AFM to the FM the antiferromagnetic order is distorted due to a possible reorientation of the spin clusters, also seen in the PM before the ferromagnetic order is fully established. The dashed lines indicating $T_N(p)$ and $T_C(p)$ are guides to the eyes.

to a decrease of T_N and a reduction of B_μ^{af} , indicating a weakening of the antiferromagnetic state. At high pressure the commensurate antiferromagnetic order of $\text{Fe}_{1.03}\text{Te}$ changes, with the muon spectra showing characteristics compatible with the incommensurate order reported in a recent neutron study.²⁷ Surprisingly, above a relatively low pressure of ~ 2 GPa the system becomes bulk ferromagnetic. Thus, the observed disorder before the occurrence of ferromagnetism could be due to nonorientated spin clusters in general seen in crossover regions from an antiferromagnet to a ferromagnet. Recently, a symmetry conserving structural transition was observed at low temperatures in the pressure range of the transition.²⁸ At even higher pressures above 4 GPa a collapsed tetragonal structure was observed at room temperature, indicating a change in the crystal structure.²⁹ Note that different pressure-induced magnetic phases were already suggested in an earlier work, however, they could not be identified.¹²

Turning an antiferromagnet into a bulk ferromagnet by applying hydrostatic pressure is a rare feature^{18–21} and has not yet been observed in the Fe-based superconductors. In general, the opposite is observed.^{30,31} Magnetization measurements on $\text{Fe}_{1.03}\text{Te}$ revealed that the magnetic moment in the ferromagnetic state increases with increasing pressure and reaches a value of $\sim 3\mu_B$ per Fe atom at 5.7 GPa and at $T = 10$ K. In addition, μSR experiments clearly indicate a drastic change in the microscopic magnetic structure, since a high frequency associated with the ferromagnetic moment occurs as soon as $\text{Fe}_{1.03}\text{Te}$ is in the ferromagnetic state. In comparison, an antiferromagnet can be easily tuned into a ferromagnet by application of a high magnetic field.^{32,33} However, in this case not an itinerant antiferromagnetic spin density wave, but localized moments order ferromagnetically

in these compounds. On the other hand, Fe_{1+x}Te with a high density of states, close to the Fermi level $N(E_F)$,⁹ is an itinerant antiferromagnet without any rare earth atoms. A high $N(E_F)$ indicates that either ferromagnetism or superconductivity may be present, whereas the latter one was suggested earlier with a T_c even higher than that in FeSe .^{9–11} However, here it is shown that a ferromagnetic state occurs. Such a scenario was proposed earlier in a theoretical model describing the magnetic order in Fe_{1+x}Te .³⁴ Within the framework of this model it was discussed that commensurate antiferromagnetic Fe_{1+x}Te could be in the vicinity of an incommensurate antiferromagnetic and a ferromagnetic phase.

In conclusion, neutron depolarization experiments show the appearance of a ferromagnetic phase at high pressure in the antiferromagnet $\text{Fe}_{1.03}\text{Te}$. Furthermore, field-dependent magnetization measurements revealed hysteretic behavior typical of a ferromagnet, whereas at the maximum pressure investigated the magnetic moment reaches $\sim 3\mu_B$ per Fe atom. In addition a drastic change in the microscopic magnetic structure is observed by means of μSR measurements. All techniques used demonstrate a transition from a low pressure antiferromagnetic to a high pressure and low temperature bulk ferromagnetic phase above $p \sim 2$ GPa. This peculiar observation may help to unravel the complex magnetic and superconducting properties in Fe-based systems.

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