Pressure-induced ferromagnetism in antiferromagnetic Fe$_{1.03}$Te

M. Bendele, 1,2,* A. Maisuradze, 1,2 B. Roessler, 3 S. N. Gvasaliya, 4 E. Pomjakushina, 5 S. Weyeneth, 1 K. Conder, 6 H. Keller, 1 and R. Khasanov 2

1 Physik-Institut der Universität Zürich, Winterthurerstrasse 190, CH-8057 Zürich, Switzerland
2 Laboratory for Muon Spin Spectroscopy, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland
3 Laboratory for Neutron Scattering, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland
4 Neutron Scattering and Magnetism, Laboratorium für Festkörperphysik, ETH Zürich, CH-8093 Zürich, Switzerland
5 Laboratory for Developments and Methods, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland

(Received 3 September 2012; revised manuscript received 13 February 2013; published 25 February 2013)

The magnetic properties of Fe$_{1.03}$Te under hydrostatic pressure up to $\rho \simeq 5.7$ 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 $\rho \simeq 2$ GPa.

DOI: 10.1103/PhysRevB.87.060409

PACS number(s): 74.62.Fj, 74.70.Xa, 75.30.—m, 76.75.+i

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. Within this new class of superconductors, the iron chalcogenides have attracted considerable interest, because their magnetic properties upon both chemical and hydrostatic pressure. Substitution of Se by Te in superconducting FeSe leads to 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. On the other hand, application of hydrostatic pressure leads to a non-linear increase of $T_c$ with a maximum value of $\simeq 36$ K. Furthermore, magnetic order appears and superconductivity and magnetism coexist on a microscopic level in the whole sample. In antiferromagnetic FeTe a $T_c$ even higher than that of FeSe was predicted but not observed until now. Instead, the parent compound FeTe exhibits peculiar magnetic properties. At the Néel temperature $T_N \simeq 70$ K a dramatic drop in the magnetic susceptibility is seen. This is related to a first-order phase transition to a monoclinic crystal structure at $T_N$ that coincides with the appearance of commensurate antiferromagnetism at $T_N$. However, at ambient pressure the magnetic properties of FeTe depend strongly on the amount of excess Fe. 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.

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. 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 (μSR) 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. 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 powdered 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. 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 $\rho \simeq 2$ 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 T’ magnetic property measurement system (MPMS) XL superconducting quantum interference device (SQUID) magnetometer. The measurements up to $\rho \simeq 6$ GPa were performed on a single crystal with a mass of $\simeq 90 \mu g$ in a diamond anvil cell, especially designed for magnetization measurements. 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 $\rho \simeq 1.7$ 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 $\rho \approx 2$ GPa and reaches $\sim 0.15$ T with increasing pressure. Furthermore, the pressure dependence of the estimated saturated ferromagnetic moment in units of $\mu_B$ was extracted from the magnetization measurements. As can be seen in the inset to Fig. 1(b), it increases with increasing
A microscopic view of the magnetic properties of Fe$_{1.03}$Te is obtained by μSR measurements performed on the μE1 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 $B_{\mu}^{\text{af}}$ and the magnetic volume fraction $M_{\text{tot}}$ are obtained ($\text{af}$ 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) μSR 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 μSR asymmetry $A(t)$ with the corresponding zero time values indicated in Fig. 2(a) was analyzed using the free software package MUSRFIT. Here, $A(t)$ is described by a model consisting of a superposition of two typical functions describing magnetic signals: one for the antiferromagnetic $g_{\text{af}}$ and one for the ferromagnetic $g_{\text{f}}$ oscillations. $M_{\text{af}}$ and $M_{\text{f}}$ describe the relative contributions of the ferromagnetic and the antiferromagnetic phases, respectively, to the total normalized magnetic volume $M_{\text{tot}} = M_{\text{af}} + M_{\text{f}} = A(t = 0, T)/A(t = 0, T = 0)$:

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

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

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

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 $a \simeq 1/3$. $A_{\text{af/f}}$ and $\lambda_{\text{af/f}}$ 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 μSR 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), $B_{\mu}^{\text{af}}$ 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 $\lambda_{\text{af}}$ just before the ordering [Fig. 2(c)].

In addition, the muon phase $\phi_{\text{sf}}$ 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. 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 μSR time spectra which is allocated to the ferromagnetic moment and is substantially larger than $B_{\mu}^{\text{af}}$ [see Fig. 2(a)]. This clearly indicates a change in the microscopic structure. The ferromagnetic volume fraction $M_{\text{f}}$
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...obtained from the zf μSR 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 $M_{\text{tot}}$ in weak transverse field (wtf) μSR 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. 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 Fe$_{1.03}$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 $\lambda^{pc}$ [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 μSR measurements are summarized in a phase diagram exhibiting different magnetic states (see Fig. 3). At ambient pressure Fe$_{1.03}$Te exhibits commensurate antiferromagnetic ordering along the ($\pi$,0) direction. The magnetic transition at $T_N$ is accompanied by a first-order structural phase transition. Application of pressure first leads...
to a decrease of $T_N$ and a reduction of $B^{\mu}_p$, indicating a weakening of the antiferromagnetic state. At high pressure the commensurate antiferromagnetic order of Fe$_{1.03}$Te changes, with the muon spectra showing characteristics compatible with the incommensurate order reported in a recent neutron study.\cite{9-11} Surprisingly, above a relatively low pressure of $\sim$2 GPa the system becomes bulk ferromagnetic. Thus, the observed disorder before the occurrence of ferromagnetism could be due to nonoriented 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.\cite{28} At even higher pressures above 4 GPa a collapsed tetragonal structure was observed at room temperature, indicating a change in the crystal structure.\cite{29} Note that different pressure-induced magnetic phases were already suggested in an earlier work, however, they could not be identified.\cite{12}

Turning an antiferromagnet into a bulk ferromagnet by applying hydrostatic pressure is a rare feature\cite{18-21} and has not yet been observed in the Fe-based superconductors. In general, the opposite is observed.\cite{30,31} Magnetization measurements on Fe$_{1.03}$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, $\mu$SR experiments clearly indicate a drastic change in the microscopic magnetic structure, since a high frequency associated with the ferromagnetic moment occurs as soon as Fe$_{1.03}$Te is in the ferromagnetic state. In comparison, an antiferromagnet can be easily tuned into a ferromagnet by application of a high magnetic field.\cite{12,32} 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 $N(E_F)$,\cite{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.\cite{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.\cite{33} 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 Fe$_{1.03}$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 $\mu$SR 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.

Helpful discussions with R. Puzniak are gratefully acknowledged. This work was supported by the Swiss National Science Foundation and the NCCR program MaNEP. The experiments were partially performed at the Swiss Muon Source SINQ and at the Swiss spallation neutron source SINQ of the Paul Scherrer Institute, PSI Villigen, Switzerland.
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