Electronic properties across the first-order phase transition in Fe$_{1.11}$Te

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Abstract. We present electrical resistivity as well as scanning tunneling microscopy and spectroscopy (STM/S) studies on Fe$_{1.11}$Te single crystals. For this composition, a first-order phase transition is observed around 57 K. This transition is associated with a structural change from the tetragonal $P4/nmm$ to the monoclinic $P2_1/m$ space group. Also at this temperature, the compound becomes antiferromagnetic ($T_N = 57$ K) and the temperature dependence of the resistivity changes from $\log(-T)$ to $T^2$. This observation suggests that the material becomes a Fermi-liquid metal at low temperatures. Metallic behavior is also confirmed in the $I-V$ characteristics of the scanning tunneling spectroscopy measurements taken at $T < T_N$ on an atomically resolved surface.

1. Introduction

The observation of superconductivity in LaFeAsO$_{1-x}$F$_x$ with $T_c = 26$ K [1] led to the discovery of several new superconductors based on iron-pnictides and -chalcogenides. Among them, FeSe with $T_c = 8$ K has a comparatively simple PbO-type tetragonal structure [2]. Upon Te substitution, this temperature increases to $T_c = 15$ K [3] and with the application of pressure, $T_c$ can be as high as 37 K [4]. The parent compound, Fe$_{1+y}$Te is non-superconducting at ambient conditions, but exhibits a spin-density-wave (SDW) transition around $T_N = 67$ K. Simultaneously, a structural transition takes place from the tetragonal $P4/nmm$ to the monoclinic $P2_1/m$ space group [5]. With increasing amount of interstitial Fe, the SDW order becomes incommensurate with respect to the lattice, and the low-temperature structure is described within the orthorhombic $Pmmn$ space group [6]. The propagation vector of the SDW order is $(\pi, 0)$ in the Brillouin zone defined by the chemical unit cell [6] which is rotated by 45° with respect to the ordering in the FeAs-families. This suggests that the nesting properties of the Fermi surface does not play a role for the SDW order in Fe$_{1+y}$Te. Furthermore, unlike in iron-pnictides where the magnetic moment per Fe-atom is rather small, the size of the magnetic moment in Fe$_{1+y}$Te is about 2 $\mu_B$/Fe [6]. The magnetic susceptibility above $T_N$ follows a Curie-Weiss law [7] indicating that a local-moment model is more appropriate to describe the magnetic behavior of Fe$_{1+y}$Te.

One important aspect of the Fe-based superconductor is the nature of their electronic ground state of the parent compounds: Is the ground state a metallic one or is the system closer to a...
Figure 1. (a) Resistivity as a function of temperature in both cooling and heating cycles for Fe$_{1.11}$Te displaying a hysteretic first-order transition at $T_N = 57$ K. (b) For $T > T_N$, the temperature dependence of resistivity exhibits a nearly linear behavior on a semi-logarithmic plot. (c) Resistivity plotted as a function of $T^2$ showing a roughly linear dependence below $T_N$.

Mott-insulating state? [8]. In the case of Fe$_{1+y}$Te, above $T_N$, the electrical resistivity shows a non-metallic behavior. Nonetheless, the optical conductivity does not display any indication for a gap [9]. At the same time, a Drude peak typical of metallic conductivity was observed in optical conductivity only below $T_N$. Thus, optical conductivity studies describe the change of the electronic state across the phase transition as an incoherent-coherent metal transition. Here we analyze the temperature dependence of dc-resistivity above and below the SDW-transition in Fe$_{1.11}$Te. In order to investigate the local conductivity on an atomic scale, we conducted scanning tunneling spectroscopy in a scanning tunneling microscope (STM). Our results reveal that the system shows a gap of about 0.2 eV above $T_N$, but a Fermi-liquid-like metallic behavior below the transition temperature.

2. Experimental

The single crystals of Fe$_{1.11}$Te were grown using a horizontal Bridgman setup. The details of the growth have been presented elsewhere [10]. The single crystals were characterized by Laue diffraction and energy-dispersive x-ray spectroscopy (EDX). The electrical properties were measured using a four-point contact setup in a physical property measurement system (PPMS, Quantum Design). The scanning tunneling microscopy and spectroscopy were performed in the temperature range of 30 - 300 K using a commercial variable-temperature STM (Omicron Nanotechnology). A sample with freshly cleaved surface ($ab$-plane) was introduced into the ultra-high vacuum chamber of the STM. The topographic scans were obtained in the constant current mode ($I=0.6$ nA) by applying a bias voltage of $+0.8$ V (i.e. the empty sample states above the Fermi-level were visualized). The spectroscopic measurements were carried out while sweeping the bias voltage from -1 to +1 V by keeping the feed-back loop open.

3. Results and discussion

The temperature dependence of resistivity $\rho(T)$ of Fe$_{1.11}$Te is presented in figure 1(a). A sharp drop in the resistivity is observed at $T_N = 57$ K. This drop is associated with a simultaneous antiferromagnetic as well as a structural transition from a tetragonal to a monoclinic structure. The suppression of $T_N$ compared to the reported value of 67 K [5] may result from an increased amount of excess Fe. In our sample investigated here the resistivity displays a clear thermal hysteresis revealing the first-order nature of the phase transition. Above $T_N$, the resistivity follows a $\rho \propto \log(1/T)$ behavior, as can be seen in figure 1(b). A similar $\log(1/T)$-divergence was also reported by Liu et al. for Fe$_{1.11}$Te$_{0.64}$Se$_{0.36}$ below 50 K [11] and in Fe$_{1.09}$Te$_{0.55}$Se$_{0.45}$
Figure 2. (a) An STM topographic image of Fe$_{1.11}$Te obtained at room temperature on an area of 2.02 × 2.02 nm$^2$ displaying a square lattice of Te atoms. (b) Line-scan along the white line in (a) showing an average inter-atomic distance of ≈ 3.7 Å. (c) $I-V$ characteristics measured on an atomically resolved surface of Fe$_{1.11}$Te at $T = 298$ K and 44 K. (d) Derivative $dI/dV$ as a function of the bias voltage $V$ for the curves shown in (c).

below 130 K [10]. This behavior is attributed to a disorder-induced electronic localization in the ternary iron-chalcogenides. However, in Fe$_{1.11}$Te, the nature of the temperature dependence drastically changes to a nearly $T^2$ behavior below $T_N$, figure 1(c), suggesting that the system becomes a Fermi-liquid-like metal. This result is in consistence with the optical conductivity where a Drude-peak develops at low frequency that reflects a reduction in the carrier scattering rate in the magnetically ordered state [9].

In order to explore the electronic properties at the atomic scale, we performed STM measurements at temperatures well above and below the SDW ordering temperature. STM is a powerful tool to study the density of states (DOS) of the materials on an atomic scale. The DOS is directly proportional to the derivative $dI/dV$, where $I$ is the tunneling current and $V$ is the bias voltage. The technique has been efficiently used to determine the superconducting gap [12] as well as the symmetry of the order-parameter [13] in the case of Fe-chalcogenides. In those cases, a large surface area could be atomically resolved and was found to be superimposed on an inhomogeneous electronic back ground. In the case of Fe$_{1.11}$Te, however, so far we could obtain only small patches of the surface with atomic resolution in our topographic scans, see figure 2(a). Scans on larger areas showed an inhomogeneous distribution of bright and dark regions, with the bright areas occasionally revealing atomic resolution. In those regions, the average inter-atomic distances were found to be ≈ 3.7(0.2) Å, figure 2(b), which is in good agreement with the Te-Te (3.82 Å) distance in the lattice. In figure 2(c), $I-V$ characteristics measured at $T = 298$ K and 44 K are plotted. The $I-V$ curve at 298 K clearly shows a gap-like feature, with a gap of 0.2 eV. This is even more apparent in figure 2(d), where $dI/dV$ as a function of $V$ is plotted. At 298 K, the zero-bias conductance $(dI/dV)_{V=0}^{T=298 K} ∼ 0$. This behavior is in contrast with the results of the optical conductivity [9], where a semiconducting gap has not been observed. At $T < T_N$, the material becomes more and more metallic, with the $I-V$ curve displaying a finite slope at the bias voltage $V = 0$. The zero-bias conductance
at 44 K is found to be \((dI/dV)_{V=0}^{T=44 K} = 0.06 \text{ nS}\), figure 2(d), which is rather small suggesting that Fe_{1.11}Te is a bad metal at low temperatures. However, the finite DOS at the Fermi level as inferred from the finite conductance at zero-bias is in consistency with the metallic behavior observed in the optical conductivity \cite{9} at low temperatures in Fe_{1.05}Te.

In conclusion, we have analyzed the temperature dependence of electrical resistivity above and below the SDW ordering temperature in Fe_{1.11}Te. The resistivity behavior drastically changes from a \(\rho(T) \sim \log (1/T)\) to \(\rho(T) \sim T^2\) below \(T_N\). The STM topographic images display atomic resolution only on small areas. Within these areas the lattice constant corresponds to the inter-atomic Te-Te distance. The \(I-V\) spectroscopy taken on the atomically resolved surface display a gap-like feature at room temperature, but metallic behavior below \(T_N\). In order to confirm a gapped high-temperature phase more detailed spectroscopic imaging scanning tunneling microscopy (SI-STM) should be performed. Although the electronic ground state of the high-temperature phase could not be determined unambiguously, the metallic low-temperature phase has been confirmed by our scanning tunneling spectroscopy.

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References

[1] Kamihara Y, Watanabe T, Hirano M and Hosono H 2008 J. Am. Chem. Soc. 130 3296
[2] Hsu F C, Luo J Y, Yeh K W, Chen T K, Huang T W, Wu P M, Lee Y C, Huang Y L, Chu Y Y, Yan D C and Wu M K 2008 Proc. Natl. Acad. Sci. USA 105 14262
[3] Fang M H, Pham H M, Qian B, Liu T J, Vehstedt E K, Liu Y, Spinnu L and Mao Z Q 2008 Phys. Rev. B 78 224503
[4] Margadonna S, Takabayashi Y, Ohishi Y, Mizuguchi Y, Takano Y, Kagayama T, Nakagawa T, Takata M and Prassides K 2009 Phys. Rev. B 80 144506
[5] Li S, de la Cruz C, Huang Q, Chen Y, Lynn J W, Hu J, Huang Y L, Hsu F C, Yeh K W, Wu M K and Dai P 2009 Phys. Rev. B 79 064503
[6] Bao W, Qiu Y, Huang Q, Green M A, Zajdel P, Fitzsimmons M R, Zhernenkov M, Chang S, Fang M, Qian B, Vehstedt E K, Yang J, Pham H M, Spinnu L and Mao Z Q 2009 Phys. Rev. Lett. 102 247001
[7] Yang J, Matsui M, Kawa M, Ohta H, Miichioka C, Dong C, Wang H, Yuan H, Fang M and Yoshimura K 2009 arXiv:0911.4758
[8] Si Q and Abrahams E 2008 Phys. Rev. Lett. 101 076401
[9] Chen G F, Chen Z G, Dong J, Hu W Z, Li G, D Z X, Zheng P, Luo J L and Wang N L 2009 Phys. Rev. B 79 140509
[10] Rößler S, Cherian D, Harikrishnan S, Bhat H L, Elizabeth S, Mydosh J A, Tjeng L H, Steglich F and Wirth S 2010 Phys. Rev. B 82 144523
[11] Liu T, Ke X, Qian B, Hu J, Fobes D, Vehstedt E, Pham H, Yang J, Fang M, Spinnu L et al. 2009 Phys. Rev. B 80 174509
[12] Kato T, Mizuguchi Y, Nakamura H, Machida T, Sakata H and Takano Y 2009 Phys. Rev. B 80 180507
[13] Hanaguri T, Niitaka S, Kuroki K and Takagi H 2010 Science 328 474