Local density of states and superconducting gap in the iron chalcogenide superconductor Fe$_{1+y}$Se$_{1-x}$Te$_x$ observed by scanning tunneling spectroscopy

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We report on the investigation of the quasiparticle local density of states and superconducting gap in the iron chalcogenide superconductor Fe$_{1+y}$Se$_{1-x}$Te$_x$ ($T_c \sim 14$ K). The surface of a cleaved crystal revealed an atomic square lattice, superimposed on the inhomogeneous background, with a lattice constant of $\sim 3.8$ Å without any reconstruction. Tunneling spectra measured at 4.2 K exhibit the superconducting gap, which completely disappears at 18 K, with a magnitude of $\sim 2.3$ meV, corresponding to $2\Delta/k_B T_c = 3.8$. In stark contrast to the cuprate superconductors, the value of the observed superconducting gap is relatively homogeneous, following a sharp distribution with a small standard deviation of 0.23 meV. Conversely, the normal-state local density of states observed above $T_c$ shows spatial variation over a wide energy range of more than 1 eV, probably due to the excess iron present in the crystal.

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Since the discovery of superconductivity in LaFeAsO$_{1-x}$F$_x$ (the so-called 1111 system) at 26 K, several other iron-based high-$T_c$ superconductors with similar layered structures have been found, such as $Ae$Fe$_2$As$_2$ (the 122 system, where $Ae$ is an alkaline-earth metal) and FeCh (the 11 system, where Ch is a chalcogenide). Among these materials, the iron chalcogenide superconductor FeSe ($T_c \sim 8$ K) has attracted much attention as the simplest model system. By partially substituting Te for Se, $T_c$ can be enhanced up to 14 K. Moreover, the application of pressure increases the $T_c$ of FeSe to 37 K. The superconducting gap in the simplest systems of the iron-based compounds is crucial in order to understand the mechanisms of superconductivity. In this letter, we report on the observation of the LDOS and superconducting gap in the iron chalcogenide superconductor Fe$_{1+y}$Se$_{1-x}$Te$_x$.

Single crystals of Fe$_{1+y}$Se$_{1-x}$Te$_x$ were grown using a self-flux method. A mixture of Fe, Se, and Te powders with a starting composition of Fe$_{0.25}$Se$_{0.75}$Te$_{0.75}$ was placed in an alumina crucible, which was then sealed in an evacuated quartz tube. The tube was heated at 950°C for 10 h, cooled to 700°C at a rate of $-5^\circ$C/h, and then annealed at 400°C for 100 h. The composition of the crystal obtained was determined as Fe$_{1.05}$Se$_{0.15}$Te$_{0.85}$ using the energy dispersive x-ray spectroscopy. The onset temperature of the superconducting transition in the in-plane resistivity was approximately 14 K. A laboratory-built low-temperature scanning tunneling microscope (STM) was used for the STS measurements. The sample mounted on the STM head was cooled down to 4.2 K in pure helium gas and then cold-cleaved in situ to expose a clean ab surface. The measurements were made in a helium gas environment. Surface topographic images were ob-
FIG. 1: (Color online) (a) Large-scale surface topography of the cleaved surface at 4.2 K (72 × 72 nm$^2$). (b) Magnified topography of the region outlined by the box in panel (a), revealing a square atomic lattice with $a_0 \sim 3.8 \AA$ ($V = 1.0$ V, $I = 40$ pA). [(c) and (d)] Tunneling conductance dependence of the STM image in the same region (6.8 × 6.8 nm$^2$ obtained at 18 K) for (c) low conductance (500 mV, 0.5 nA) and (d) high conductance (50 mV, 0.5 nA).

FIG. 2: (Color online) (a) Spatially averaged tunneling conductance spectrum in the high-energy range, measured at 4.2 K. (b) Low-energy part of the spatially averaged spectrum measured below (4.2 K) and above (18 K) $T_c$, indicating that the gap feature at low temperature corresponds to the superconducting gap. The dotted line on the 4.2 K spectrum indicates the calculated V-shaped background conductance (see text). The 18 K spectrum has been shifted vertically for clarity.

FIG. 3: (Color online) Spatially averaged spectrum at 4.2 K normalized to the background conductance represented by the dotted line in Fig. 2(b). The curve indicates a fit of the calculated DOS for an $s$-wave superconductor to the data.
on the V-shaped background, indicating the existence of an energy gap. The disappearance of this feature above $T_c$ is evidence that this energy gap is responsible for the superconductivity.

In order to determine the value of the superconducting gap $\Delta$, the contribution of the V-shaped background conductance was removed using the following procedure. First, the background on both sides of the region, displaying the gap feature ($|V| > 5$ meV), was extracted from the average spectrum measured at 4.2 K. The backgrounds for the positive and negative bias regions were separately fitted by linear functions, which were then extended to the Fermi energy. The resulting V-shaped background was convoluted with the Fermi function at 4.2 K to simulate thermal broadening and is indicated by the dotted line in Fig. 2(b). Finally, the measured spectrum was normalized to the simulated V-shaped background and is shown by the circles in Fig. 3. The normalized spectrum was fitted using the $s$-wave BCS gap function with a Dynes broadening factor $\Gamma$, as shown by the curve in Fig. 3. The resulting value of the gap is $\Delta = 2.3$ meV, which gives the ratio $2\Delta/k_BT_c \sim 3.8$, slightly larger than that for a conventional superconductor. Because of the relatively high temperature of the measurements with respect to $T_c$ and the arbitrary nature of the background estimation, we were unable to determine the symmetry of the gap from the fitting procedure. We note that the $d$-wave gap function with $\Delta = 3$ meV and $\Gamma = 1$ meV gives almost the same curve as the fitted $s$-wave function.

The shape of the gap feature observed in the normalized spectra measured at different points on the sample surface seems to be rather uniform. Figure 4 displays a series of spectra taken along a 10-nm-long linecut across the surface; each spectrum has been normalized to the background conductance given by the dotted line in Fig. 2(b). The magnitude of the gap does not show significant spatial variation, in marked contrast to the cuprate superconductors, which exhibit gross gap inhomogeneity. The zero-bias conductance and the gap edge peaks vary slightly with location, and the values of $\Delta$ obtained by fitting each normalized spectrum to the $s$-wave function follow a sharp distribution with a small standard deviation of $\sigma_\Delta = 0.23$ meV.

While the superconducting gap opens homogeneously below $T_c$, the normal-state electronic states in the high-energy range show spatial variation. Figure 5 shows a representation of the spatial evolution of the high-energy LDOS measured above $T_c$. The differential conductance map measured at 50 mV shown in Fig. 5(b) correlates well with the high-conductance STM image shown in Fig. 5(a). Thus, the LDOS spatially varies in accord with the surface topography. In order to visualize the spectral changes occurring with location, Fig. 5(c) shows a series of tunneling spectra measured along a linecut denoted by the arrows in Figs. 5(a) and 5(b). Seven typical spectra taken at the labeled positions are shown in Fig. 5(d). In the darkest region of the STM image (spectrum 2), the LDOS exhibits a simple V-shape with strong bias asymmetry, and the low-energy spectral weight is strongly de-
pressed. In contrast, at the positions of bright spots in the STM image (spectra 4 and 6), the LDOS around 100 meV is enhanced, and the asymmetry and V-shape are less pronounced, with substantial spectral weight remaining at low energy. This change in the nature of the LDOS remains up to 1 eV, as can be seen in the high-bias STM image in Fig. 1[b]. As mentioned earlier, the bright spots are most likely related to excess iron present between the layers. Therefore, the spatial variation in the normal-state LDOS seems to be caused by the excess iron, although the mechanism by which this occurs is unclear. The weak spatial variation in the normalized low-energy LDOS in the superconducting state probably originates from this spatially inhomogeneous background electronic structure.

In conclusion, scanning tunneling microscopy and spectroscopy have been performed on the iron chalcogenide superconductor Fe$_{1+y}$Se$_{1-x}$Te$_x$ both above and below $T_c$. Atomically resolved topographic STM images of the cleaved surface were acquired, revealing a square lattice without any reconstruction. The tunneling spectra reveal a superconducting gap with an average value of approximately 2.3 meV. The normal-state background electronic structure shows strong spatial variation, probably due to the excess iron in the crystal. Further investigation of the LDOS and superconducting gap in the simplest iron-based compounds will ensure progress in understanding the mechanism involved in the iron-based superconductors.

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