RAPID COMMUNICATION

Observation of mode-like features in tunneling spectra of iron-based superconductors*

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We report scanning tunneling microscopy/spectroscopy (STM/STS) studies on iron-based superconductors of $Ba_{1-x}K_xFe_2As_2$ and nearly optimally doped Fe(Te,Se). Mode-like features were observed universally outside the superconducting gaps in the tunneling spectra, which are similar to our previous observations in other samples and can be ascribed to the interaction between electrons and spin excitations. Furthermore, an almost linear relationship between the superconducting gaps and the superconducting transition temperatures was noted and should also be taken into account in understanding the mechanism of iron-based superconductors.

Keywords: superconductor, scanning tunneling microscopy, paring mechanism, spin excitations

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1. Introduction

The discovery of iron-based superconductors has attracted tremendous attention in the past few years.[1–7] It provides an opportunity to take a fresh look on some fundamental problems of high-T_c superconductors. In the conventional BCS superconductors, superconductivity is achieved through the formation of cooper pairs mediated by electron–phonon interaction called "paring glue". However for cuprates, the paring mechanism is still under debate partially due to their abnormal behaviors in normal states. Unlike cuprates, the recently discovered iron-based superconductors usually have a normal metallic state, though the multiband effect[8–10] makes the understanding of paring mechanism not so easy as what expected in the beginning. In addition, the electron correlation seems not to be negligible[11,12] and may play a role in the regimes of both magnetism and superconductivity.[13,14] Thus, whether Fermi topology/"pairing glue"[15,16] or local correlation[11–14] is dominant in the origin of superconductivity is a pivotal problem.[17]

In the strong coupling regime based on BCS scenario, an electron–phonon interaction function can be derived from the tunneling spectrum of a normal metal–superconductor junction,[18,19] which self-consistently identifies phonon as the "pairing glue" in the conventional superconductors.[20] In this case, the phonon modes are reflected by some hump-dip features (called as "mode-like features" in this paper) at the corresponding mode energies in the tunneling spectrum (dI/dV vs. V). These mode-like features appear as some dips at positive voltage or some peaks at negative voltage in the second derivative of the tunneling current (d^2I/dV^2 vs. V). For cuprates and iron-based superconductors, neutron scattering experiments have observed spin resonance modes below T_c.[21,22] The presence of spin resonance suggests magnetic origin (such as spin fluctuations) of the pairing mechanism. Theoretically, this issue has been investigated in cuprates[23–25] and in iron-based superconductors.[26] Interestingly, mode-like features were observed in some iron-based superconductors,[27–31] which were seemingly related to the coupling between electrons and some kinds of bosonic modes. Furthermore, the mode-like features were studied in momentum space for LiFeAs through quasiparticle interference (QPI) by using STM/STS.[32] The uncovered electronic self-energy presumptively induced by electron–boson coupling is strongly momentum-dependent, which is inconsistent with the regime of electron–phonon coupling while can be explained by the coupling between electrons and spin excitations (in here could be antiferromagnetic spin fluctuations). More inspiringly, the reported mode energies from STM/STS are in good agreement with those of the spin resonance modes observed in inelastic neutron scattering experiments which have a linear relation-

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ship with superconducting temperatures.[33–43] However, before a final conclusion can be drawn, one question needs to be answered: to what extent can we accept that the mode-like features are universal in iron-based superconductors?

In this paper, we report our scanning tunneling microscopy/spectrocopy (STM/STS) studies on iron-based superconductors $Ba_{1-x}K_xFe_2As_2$ with different doping levels and nearly optimally doped Fe(Te, Se). Superconducting gaps and mode-like features were observed simultaneously in all samples. Based on the statistics of these data and the previously reported results, we present a more complicated relationship between the mode-like features and the spin excitations and an explicit linear relationship between the superconducting gaps and the superconducting transition temperatures.

2. Experiment

The $Ba_{1-x}K_xFe_2As_2$ ($x = 0.078, 0.09, 0.40$) single crystals and the Fe(Te,Se) single crystal studied were grown with the self-flux method.[44,45] The transition temperatures were determined by dc susceptibility measured by a superconducting quantum interference device (SQUID). The superconducting transition temperatures (T_c) of the $Ba_{1-x}K_xFe_2As_2$ samples are around 37 K, 22 K, and 16 K for $x = 0.40$, 0.09, and 0.078, respectively. The T_c of Fe(Te, Se) is about 14 K. All the spatially resolved tunneling experiments were carried out on our home-made low-temperature STM. The studied single crystals were cold-cleaved *in situ* and immediately inserted into the STM head, which had already been set at the desired temperatures. Tunneling spectra (dI/dV vs. V) were taken by using the lock-in technique.

3. Result and discussion

Figure 1(a) shows tunneling spectra taken at different positions on the cleaved surface of the optimally doped $Ba_{0.6}K_{0.4}Fe_2As_2$ single crystal. The spectra have been normalized to their normal-state backgrounds constructed from the spectra measured above T_c as presented in Fig. 1(b).[28] In the spectra shown in Fig. 1(a), a larger superconducting gap can be easily determined by a pair of coherence peaks and a smaller gap can be identified from two symmetric kinks inside the larger gap. The energies of those peaks and kinks are indicated by the gray vertical lines, which are well consistent with our previously-reported data obtained on the same sample[10] and in good agreement with the gap values from different Fermi surfaces detected by ARPES.[46] It was found that the surface configuration could affect the spectral shape by inducing finite low-bias conductance in some cases (and thus the low-bias U-shape of a fully-gapped spectrum is not so clear) while it does not change the gap values significantly.[10] In addition to the superconducting gaps, we also observe hump-dip features (indicated by the black arrows) outside the gaps. Such features correspond to a dip at positive bias voltage or a peak at negative bias voltage in the curve of d^2I/dV^2 vs. V, as shown in Fig. 1(c).[28] In analogy to the conventional superconductors with strong electron–phonon coupling, we have ascribed these abnormal features to the coupling between electrons and spin excitations, because the specific mode energy (Ω) determined from the positions of dips/peaks ($E_{dip,peak}$) and superconducting gaps (Δ) according to the relation $\Omega = E_{dip,peak} - \Delta$ is very close to the energy of the spin resonance mode detected by neutron scattering.[28] In order to find further evidence for this argument, we have carried out similar measurements on two underdoped samples of $Ba_{1-x}K_xFe_2As_2$ with $T_c = 22$ K ($x = 0.09$) and $T_c = 16$ K ($x = 0.078$).

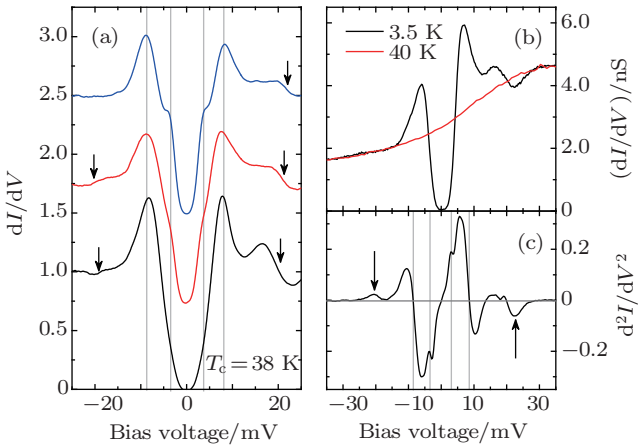


Fig. 1. (color online) (a) Normalized tunneling spectra of nearly optimally doped $Ba_{1-x}K_xFe_2As_2$ single crystals ($x = 0.4$) measured at 3.5 K. The grey vertical lines indicate the features of superconducting gaps, while the black arrows indicate the hump-dip feature mentioned in the paper. (b) Comparison between two spectra measured at low temperature and above T_c. (c) Derivative of the low-temperature spectrum shown in panel (b). The grey vertical lines indicate the features of superconducting gaps, while the black arrows indicate the hump-dip feature. The data in panels (b) and (c) are the same as what we have reported previously.[28]

Figure 2 shows the spatially dependent tunneling spectra of the $Ba_{1-x}K_xFe_2As_2$ samples. Similar to the results of the optimally doped sample with $T_c = 37$ K, two superconducting gaps (indicated by the gray vertical lines) and mode-like features outside the gaps (indicated by the arrows) can be identified. $Ba_{1-x}K_xFe_2As_2$ with $T_c \approx 22$ K has also been studied by ARPES[47] and the derived gaps of 4.4 meV and 7.6 meV are close to our results of 3.8 meV and 6 meV within the experimental errors. Figures 3(a) and 3(b) show the temperature dependencies of the tunneling spectra averaged over a 10 nm×10 nm region for the sample with $T_c = 22$ K and a 8.3 nm×8.3 nm region for the sample with $T_c = 16$ K. Similar to the case of the optimally doped sample,[28] both gap features and mode-like features fade out with increasing temperature towards T_c. Due to great similarity in the spectra between these two underdoped $Ba_{1-x}K_xFe_2As_2$ samples and the

optimally doped $Ba_{0.6}K_{0.4}Fe_2As_2$ sample, we speculate that the hump-dip features/mode-like features observed in all these samples should have a common origin and be very likely related to the coupling between electrons and spin excitations. Correspondingly, the mode energies of the spin excitations can be determined by the positions of the dips or peaks in the d^2I/dV^2 vs. V curves (indicated by arrows) as shown in Figs. 3(c) and 3(d), that is, $\Omega = E_{\rm dip,peak} - \Delta$.

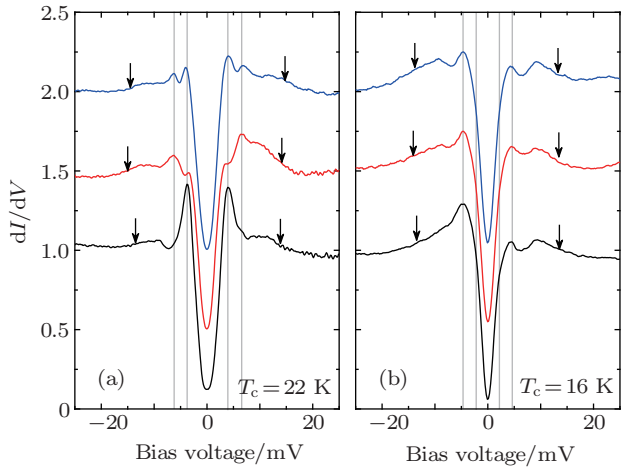


Fig. 2. (color online) Normalized tunneling spectra of underdoped $Ba_{1-x}K_xFe_2As_2$ single crystals with (a) $T_c = 22$ K and (b) $T_c = 16$ K. All the spectra were taken at 2.2 K. The grey vertical lines indicate the features of superconducting gaps, while the black arrows indicate the hump-dip feature.

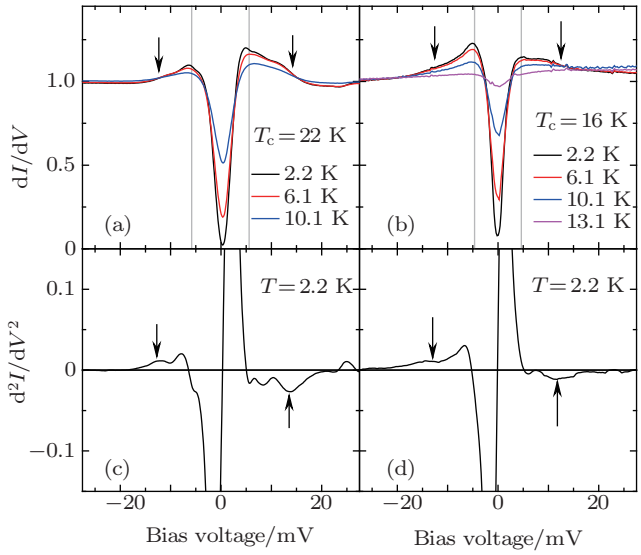


Fig. 3. (color online) (a) The averaged dI/dV spectra (over a 10 nm×10 nm region) of $Ba_{1-x}K_xFe_2As_2$ with $T_c = 22$ K. (b) The averaged dI/dV spectra (over a 8.3 nm×8.3 nm region) of $Ba_{1-x}K_xFe_2As_2$ with $T_c = 16$ K. Panels (c) and (d) present the derivatives of the 2.2 K spectra shown in panels (a) and (b), respectively.

Interestingly, in a previous study of "111"-type iron-based superconductor $Na(Fe_{1-x}Co_x)As$ by our collaborators, similar mode-like features were observed with a value very close to neutron data.[30] In order to further examine such universality, we have studied another iron-based superconductor Fe(Te,Se) with the most simple "11"-type structure. Fe(Te,Se) only consists of Fe-based blocks repeated along the c axis without intercalations between the blocks. As shown in Figs. 4(a) and 4(b), the exposed surface after a natural cleavage is the nonpolar Te(Se) atomic layer, which is much more cleaner than that of the "122"-type samples. Thus an atomic-level flat surface can be seen in the STM topographic image shown in Fig. 4(b), in which the brighter spots are Te atoms and the less bright spots are Se atoms, meanwhile the most bright big spot in the center of the image is an interstitial iron impurity.[45] The measured tunneling spectra are spatially homogeneous on the cleaved surface of Fe(Te, Se), while a sharp zero-bias peak shows up in the spectrum taken around the iron impurity.[45] Figures 4(c) and 4(d) show the temperature dependences of the tunneling spectra and the normalized ones. The background adopted here for normalization is the spectrum taken at 10.1 K, the highest temperature measured as shown in Fig. 4(c). In both figures, strong depression of density of states at low energies limited by two coherence peaks can be clearly seen in the spectra of low temperature, indicating a finite superconducting gap about 2 meV which corresponds to that opened in the hole bands.[48] Outside the coherence peaks, there exists a hump-dip/mode-like feature (indicated by the vertical dotted line). With increasing temperature, this feature fades out in the wake of the suppression of superconductivity. Therefore, the mode-like feature observed here in "11"-type iron-based superconductors is very similar to that in the above-mentioned "122"-type and "111"-type samples in all respects. From the dip position at the positive voltage of the d^2I/dV^2 vs. V curve (not shown here), the "mode" energy is determined to be 5.4 meV, which is close to the characteristic energy of a spin resonance peak observed by inelastic neutron scattering.[37]

In Fig. 5, we plot the relationship between the mode energies and T_c (solid symbols) obtained from all our tunneling experiments on different types of iron-based superconductors. The relation between the spin resonance modes and T_c (open symbols) obtained from neutron scattering measurements is also presented for comparison.[33,35–38,40,41] A good agreement between the data from STS and neutron scattering indicates that the hump-dip structure/mode-like feature observed in the tunneling spectra should be closely related to electron–spin excitations coupling. In addition, since we reported the consistency between STS experiment and neutron scattering measurements,[28,30] there have been two other STS experiments on LiFeAs ("111"-type)[29] and FeSe ("11"-type)[31] to study the relationship between the mode-like features in tunneling spectra and the neutron scattering data.[41,42] All these works support the picture of electron–spin excitations coupling in iron-based superconductors and the obtained mode energies fall into the same line as shown in Fig. 5.[22]

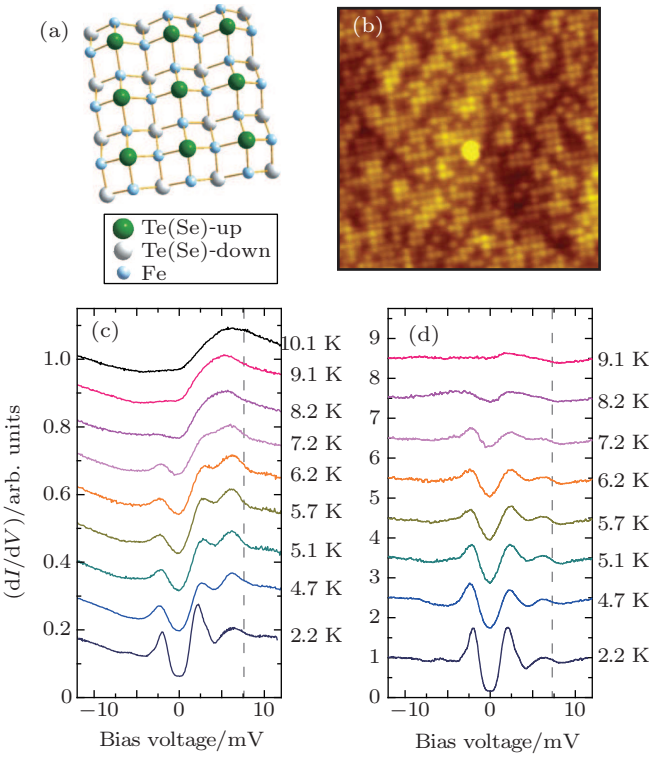


Fig. 4. (color online) Atomic topography and tunneling spectra of Fe(Te,Se) single crystal. (a) Schematic crystal structure of a single Fe(Te,Se) block. (b) Topographic image of Fe(Te, Se) (17 nm×17 nm). The exposed surface is the Se/Te layer, wherein the bright dots are Te atoms and the less bright dots are Se atoms. It was taken at 2.4 K with a bias voltage of 20 mV and a tunneling current of 250 pA. (c) Tunneling spectra taken on the surface of Fe(Te, Se) at different temperatures. (d) The spectra in panel (c) normalized to the spectrum taken at 10.1 K. All the spectra in panels (c) and (d) are shifted for clarity. Dotted lines indicate the location where the bosonic mode lies.

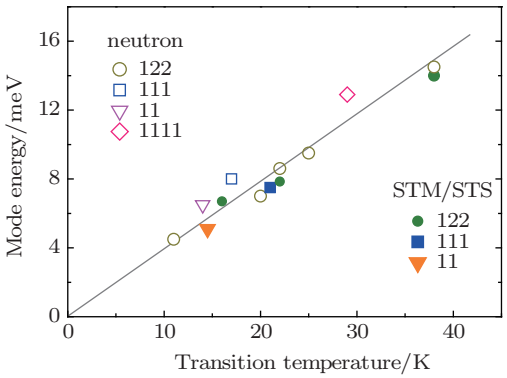


Fig. 5. (color online) Relationship between the characteristic energies of bosonic modes and superconducting transition temperatures. Open symbols indicate the data obtained from neutron scattering,[33–38,40,41] solid symbols indicate the data from our new STS experiments and previous reports.[28,30] The grey line is for guiding eyes.

Before a conclusion can be drawn, the discrepancy should be noted between the analysis of STS data in those reports and that in our works. As shown in Fig. 6(a), in all our previous and present works, the mode energies were derived from the downslope (the right arrow in blue) in the $\mathrm{d}I/\mathrm{d}V$ vs. V curve, i.e., the dip in the $\mathrm{d}^2I/\mathrm{d}V^2$ vs. V curve. However, the mode energies were determined by the dip in the $\mathrm{d}I/\mathrm{d}V$ vs. V curve (the left arrow in black) in Refs. [27] and [29], and by the upslope (the middle arrow in red), i.e., the peak in the positive-voltage part of the $\mathrm{d}^2I/\mathrm{d}V^2$ vs. V curve, in Ref. [31]. This means that the universal relation between Ω and T_c was obtained by using different ways to determine the mode energies.[27–31] On the other hand, if the same way was taken to determine Ω in all experiments, the obtained Ω could be discrete and no universal law can be obtained. This is just an experimental fact as shown in Fig. 6(b), where a summary of the present works and all the reported STS experiments we known is presented.[27–31,49,50] No obvious rule can be derived for the three different ways to determine Ω as indicated in Fig. (a). Such discrepancy can be understood by taking into account the following factors. First, the mode energy is derived by $\Omega = E_{\mathrm{dip,peak}} - \Delta$, while there are multiple gaps in various iron-based superconductors. Thus, determination of Ω depends on which gap is detected by the STS measurement (or is selected). Second, the interaction between electrons and spin excitations is more complicated than what has been expected and thus the relation between Ω and the mode-like feature is not so explicit. Third, the anisotropy of the superconducting gap can also change the spectral shape of the mode-like feature. Finally, the spin resonance mode observed by neutron scattering can not reflect all spin excitations coupled with electrons.

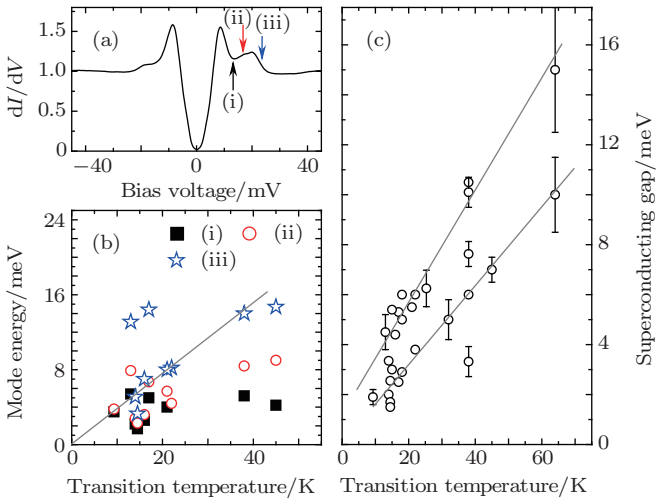


Fig. 6. (color online) (a) Three different ways to determine the mode energy (indicated by the arrows numbered (i)–(iii)) as depicted in the text. (b) Relationship between superconducting transition temperatures and the mode energies derived with the ways shown in panel (a). (c) Relationship between transition temperatures and the superconducting gaps measured by STS. The tunneling data used in panels (b) and (c) come from the current study and references.[27–31,45,49–58]

According to the above discussion, the spin resonance mode is a consequence of superconductivity, which indicates magnetic origin (such as antiferromagnetic spin fluctuations) of paring mechanism in iron-based superconductors. Then what is the dominant factor determining T_c in iron-based superconductors? To get further insight into this issue, a summary of superconducting gaps measured by STS experiments

is presented in Fig. 6(c).[27–31,45,49–58] Two groups of gaps can be seen, which are separated from each other, and both gaps have an approximate linear relationship with T_c. The gap ratio of $2\Delta/k_BT_c$ (for the larger gap) varies between 5 and 8, locating in the strong coupling regime as compared with the ratio of 3.5 in the BCS weak coupling regime, which is consistent with the reported ARPES data.[47,48,59–69] According to a local strong-coupling pairing model,[70] the local superconducting gap can be expressed as $\Delta(r) = \Omega(r)\exp(-1/N_0 g_{\rm eff}(r))$, where N_0 is the density of states at the Fermi level, $g_{\rm eff}(r)$ is the effective electron–boson coupling constant, and $\Omega(r)$ indicates spin excitations. Since $g_{\rm eff}(r)$ also depends on $\Omega(r)$, $\Delta(r)$ can be a nonmonotonic function of $\Omega(r)$. According to all these observations and considerations, it can be concluded that electron–spin excitations coupling is closely related to the origin of superconductivity in iron-based superconductors, while pairing strength Δ is still the dominant factor for T_c and depends on the spectrum of spin excitations in a nonmonotonic way.

4. Summary

Scanning tunneling spectroscopy experiments were performed on some iron-based superconductors with "122"-type structure (underdoped $Ba_{1-x}K_xFe_2As_2$) and "11"-type structure (Fe(Te,Se)). Mode-like features (hump-dip structure) outside the superconducting gaps were observed in tunneling spectra for each sample. By combining current data and the previous reports on STS and neutron scattering, we ascribe these features to the coupling between electrons and spin excitations. Furthermore, T_c is found to be dominated by the superconducting gap, which may have a nonmonotonic dependence on the characteristic energies of spin excitations.

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