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Direct observation of superconducting gaps in iron-based superconductors by laser ARPES

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Abstract. We have developed a new apparatus for carrying out ultra-high energy resolution laser-excited angle-resolved photoemission spectroscopy (laser ARPES). The achieved energy resolution is $\sim$ 70 μeV at a sample temperature $\sim$ 1.5 K. As a test case, we have measured the superconducting gap of elemental metal Sn (T_c = 3.7 K). We have used this laser-ARPES apparatus, to directly observe the superconducting gap of various iron-based superconductors. The spectra of $FeTe_{0.6}Se_{0.4}$ below T_c show a very clear temperature dependent superconducting coherence peak. Preliminary momentum-dependent results indicate an anisotropic superconducting gap in $FeTe_{0.6}Se_{0.4}$.

1. Introduction

Since the discovery of the iron-based superconductors (FeSCs) [1], the pairing mechanism of their superconductivity has been extensively studied. One of the most intriguing aspects of FeSCs is their multi-band and multi-orbital nature and how they relate with various properties. Most of the FeSCs have characteristic electronic structures composed of hole-like Fermi surfaces (FSs) around Brillouin-zone (BZ) center and electron-like FSs at the BZ corner. The interband scattering between these hole-like and electron-like FSs is generally believed to be important for the pairing interaction based on the experimental observations that many FeSCs show superconductivity only upon carrier doping in the antiferromagnetic parent compounds [2]. Theoretically, spin-fluctuation mediated $s_{\pm}$ superconductivity, where the hole-like and electron-like FSs have opposite signs of the superconducting order parameter, have been proposed[3, 4]. However, there exist exceptional examples, such as: (i) Fe(Te,Se), whose parent compound $Fe_{1+y}Te$ is antiferromagnetic [5], but Fe(Te,Se) shows superconductivity upon isovalent substitution i.e. without carrier doping [6], (ii) KFe_2As_2, which is an extremely hole-doped compound of the series $(Ba,K)Fe_2As_2$ and has only hole FSs [7] but still shows superconductivity [8], and (iii) LiFeAs, which can be regarded as a parent compound, but also shows superconductivity without substitution [9]. To reveal the details of the superconducting pairing interactions, it should be crucial to investigate their superconducting (SC) order parameters or SC gaps including anisotropy in the momentum space and node structures.

Angle-resolved photoemission spectroscopy (ARPES) is the most powerful tool to investigate the magnitude of SC gaps. We can directly observe them utilizing this technique, although their phase cannot be detected. The direct observation of SC-gap anisotropy and nodes would provide us indispensable informations for understanding the superconducting pairing mechanism. Recently, we have developed a new laser-based ARPES (laser ARPES) apparatus,

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where a vacuum ultra violet (VUV) laser is adopted for an excitation light source. We have achieved an unprecedented energy resolution of 70 μeV at a low temperature of 1.5 K. Utilizing this laser-ARPES apparatus, we can directly observe the SC gaps of various FeSCs including above mentioned interesting materials, $FeTe_{0.6}Se_{0.4}$ (T_c = 14.5 K) [10], KFe_2As_2 (T_c = 3.4 K) [11], and LiFeAs (T_c = 18 K) [12].

2. Performance of new laser ARPES apparatus

Figure 1(a) shows a schematic description of our laser-ARPES apparatus. This apparatus mainly consists of a VG-Scienta HR8000 hemispherical electron analyzer, an upright helium-impounding cryostat, and a $Nd:YVO_4$ laser system with a Fabry-Perot etalon and a non-linear optical crystal $KBe_2BO_3F_2$ [13]. Because the hemisphere radius of the electron analyzer R = 200 mm and the smallest width of the installed slit is w = 50 μm, the highest energy resolution that can be achieved with this spectrometer, evaluated as $2R/w$, can be as high as 8000. Because the lowest pass energy of this spectrometer is 0.5 eV, we can expect the best energy resolution of this electron analyzer ΔE_a = 0.5 eV / 8000 = 62.5 μeV. The linewidth of the incident laser photons can be narrowed with the Fabry-Perot etalon down to ΔE_l = 25 μeV. Thus, we can expect the best total energy resolution of this laser-ARPES apparatus to

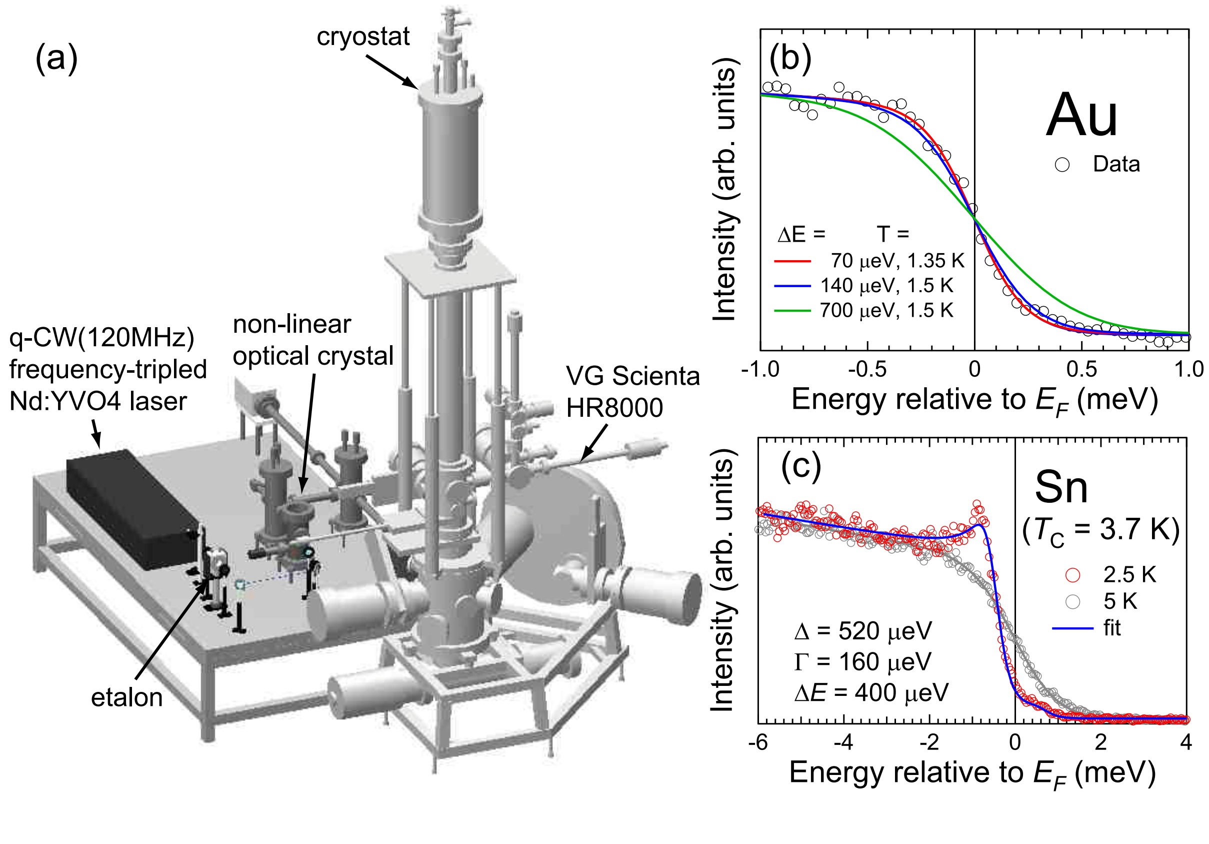

Figure 1. Performance of our recently developed laser-ARPES apparatus. (a) Schematic description of our laser-ARPES apparatus. (b) Photoemission spectrum of an evaporated gold film measured with the laser-ARPES apparatus, compared with the several fitting results. (c) Photoemission spectra of Sn (T_c = 3.7 K) below and above T_c. The superconducting coherence peak is clearly observed for the spectrum at 2.5 K.

be $\Delta E_{total} = \sqrt{\Delta E_a^2 + \Delta E_l^2} = \sqrt{62.5^2 + 25^2} \sim 67.5$ μeV. Figure 1(b) shows the spectrum of an evaporated gold film for the best performance of our new laser-ARPES apparatus. We have carried out a least-squares fitting of the gold spectrum to the Fermi-Dirac function convoluted with a Gaussian of FWHM ΔE to obtain the total energy resolution. If we fixed ΔE to be 70 μeV, the value of temperature T was obtained to be $T = 1.35$ K by the least-squares fitting. Accordingly, the quality of the fit in terms of the residual error was the best when we used $\Delta E = 70$ μeV and $T = 1.35$ K, while it became worse for $\Delta E = 70$ μeV and $T = 1.5$ K. For comparison, we show fits using $\Delta E = 70$ μeV, $T = 1.35$ K, $\Delta E = 140$ μeV, $T = 1.5$ K and $\Delta E = 700$ μeV, $T = 1.5$ K. Figure 1(c) shows the spectrum of a fractured polycrystalline tin(Sn) sample ($T_c = 3.7$ K) measured at 2.5 K ($< T_c$) and 5 K ($> T_c$). The leading-edge shift below T_c clearly indicates the SC-gap opening and the superconducting coherence peak is also observed.

3. Direct observation of the superconducting gap in $FeTe_{0.6}Se_{0.4}$

Single crystals of $FeTe_{0.6}Se_{0.4}$ were prepared by a melt-growth technique. Chemical composition of the grown crystals was determined by electron probe microanalysis (EPMA) and inductively coupled plasma (ICP) atomic emission spectrometry. Details have been described in Ref. [10]. ARPES data were collected using the above-mentioned laser-ARPES apparatus. The overall energy resolution was set to $\sim$ 1.2 meV for the SC-gap measurements and the angular resolution was set to 0.1° to obtain a reasonable count rate for the angle-resolved measurements. The Fermi edge of an evaporated gold film was measured to calibrate the positions of Fermi level (E_F).

Figure 2(a) shows the FS map of $FeTe_{0.6}Se_{0.4}$ measured at 25 K with circularly polarized light. The image was made by integrating spectral intensity within ± 5 meV from E_F and symmetrized assuming the tetragonal crystal structure. The symbols indicate the positions of Fermi momenta (k_F) determined from the momentum distributions curves (MDCs) at E_F. The solid line corresponds to the shape of FS determined by fitting the positions of k_F to the model function having a fourfold symmetry. We have then measured a detailed temperature dependence of energy distribution curves (EDCs) at k_F with FS angle φ of 0°. The results are shown in Fig. 2(b). While the spectra above T_c have a cutoff of the Fermi-Dirac function at E_F, the spectrum at the lowest temperature (2.5 K) shows a very clear superconducting coherence

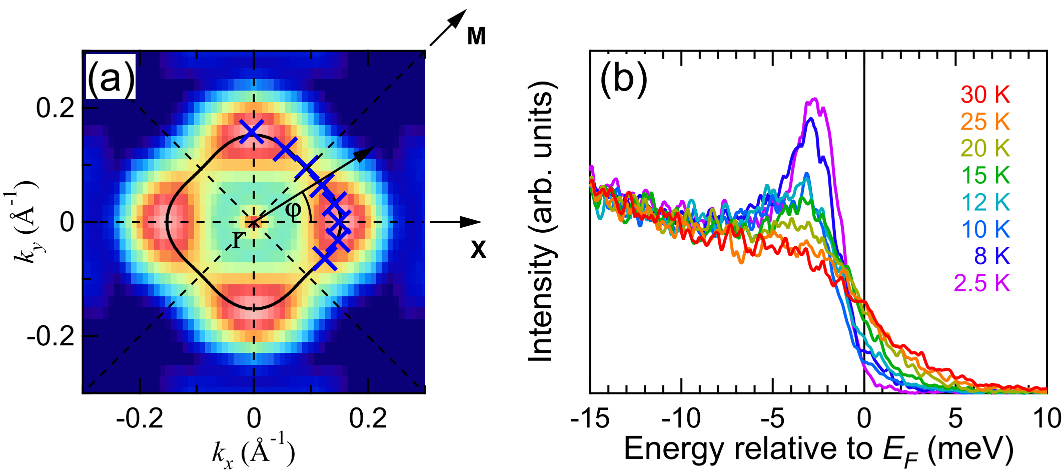

Figure 2. (a) FS intensity map of $FeTe_{0.6}Se_{0.4}$ measured at 25 K with circularly polarized light. k_F positions are indicated by the symbols. (b) Temperature-dependent EDCs at k_F with FS angle φ of 0°. The evolution of the superconducting gap with temperature is recognized very clearly.

peak. The gap at the lowest temperature can be nicely fitted to a BCS spectral function and provides a gap value of $\Delta_0 \sim 2$ meV. Preliminary results of the momentum dependence of the gap indicate an anisotropic s-wave behavior for the Γ-centered hole FS. This result is in contrast to earlier ARPES studies [14, 15] but consistent with thermodynamic results [16]. The origin of the anisotropy has been theoretically discussed recently [17].

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