Video Article

Angle-resolved Photoemission Spectroscopy At Ultra-low Temperatures

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Abstract

The physical properties of a material are defined by its electronic structure. Electrons in solids are characterized by energy (ω) and momentum (***k***) and the probability to find them in a particular state with given ω and ***k*** is described by the spectral function A(***k***, ω). This function can be directly measured in an experiment based on the well-known photoelectric effect, for the explanation of which Albert Einstein received the Nobel Prize back in 1921. In the photoelectric effect the light shone on a surface ejects electrons from the material. According to Einstein, energy conservation allows one to determine the energy of an electron inside the sample, provided the energy of the light photon and kinetic energy of the outgoing photoelectron are known. Momentum conservation makes it also possible to estimate ***k*** relating it to the momentum of the photoelectron by measuring the angle at which the photoelectron left the surface. The modern version of this technique is called Angle-Resolved Photoemission Spectroscopy (ARPES) and exploits both conservation laws in order to determine the electronic structure, *i.e.* energy and momentum of electrons inside the solid. In order to resolve the details crucial for understanding the topical problems of condensed matter physics, three quantities need to be minimized: uncertainty* in photon energy, uncertainty in kinetic energy of photoelectrons and temperature of the sample.

In our approach we combine three recent achievements in the field of synchrotron radiation, surface science and cryogenics. We use synchrotron radiation with tunable photon energy contributing an uncertainty of the order of 1 meV, an electron energy analyzer which detects the kinetic energies with a precision of the order of 1 meV and a He^3 cryostat which allows us to keep the temperature of the sample below 1 K. We discuss the exemplary results obtained on single crystals of Sr_2RuO_4 and some other materials. The electronic structure of this material can be determined with an unprecedented clarity.

Video Link

The video component of this article can be found at http://www.jove.com/video/50129/

Introduction

Nowadays ARPES is widely used to determine the electronic structure of solids. Usually, different variations of this method are defined by the source of the radiation needed to excite the electrons. We use synchrotron radiation since it offers a unique opportunity to tune the polarization and the excitation photon energy in a wide energy range and is characterized by high intensity, small bandwidth (uncertainty in energy hn) and it can be focused to a narrow beam to collect photoelectrons from a spot of a few tens of microns. Synchrotron radiation is generated in electron storage rings forcing electrons circulating in the ring with an energy of the order of 2 GeV ** to pass through periodic arrangements of strong magnets (undulators). The magnetic field deflects the electrons and when such fast electrons change their direction they emit radiation. Exactly this radiation is then directed into the so called beamline where it is further monochromatized by a diffraction grating and focused on the surface of the sample by several mirrors. There are many such facilities around the world. Our end-station is located at one of the beamlines of the BESSY storage ring which belongs to the Helmholtz-Zentrum Berlin.

The heart of this ARPES facility is the electron-energy analyzer (**Figure 1**). Since we are interested in both the kinetic energy and angle at which electrons leave the surface, it is very convenient to detect them in one measurement. A very simple principle makes this approach a reality. As in a basic experiment with an optical lens, which focuses a plane wave into the point in the back focal plane thus performing spatial Fourier transformation, the electron optical lens projects electrons which left the surface at particular angle to a point in the focal plane (**Figure 1**). In

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such a way we gain access to the reciprocal, *i.e.* momentum, space. The distance from the forward direction in the focal plane corresponds to the angle and thus to the momentum of the photoelectron. Now the electrons have to be analyzed in terms of energy. For this purpose the entrance slit of the hemispherical analyzer is placed exactly in the focal plane of the electron optical lens. Voltages on two hemispheres are chosen such that only electrons with particular kinetic energy (pass energy) will be guided exactly in the middle of two hemispheres and land on the central line of the two dimensional detector. Those which are faster will hit the detector closer to the outer hemisphere; those which are slower will be deflected towards the inner hemisphere. In such a way we can get the photoemission intensity distribution as a function of angle and kinetic energy simultaneously.

The main advantage of our approach over existing methods is the use of the He^3 cryomanipulator. There are at least two reasons to carry out the measurements at low temperatures. The higher the temperature of the material, the more smeared out the electronic states become in energy and momentum. To determine the electronic structure with high precision this temperature broadening has to be avoided. Also, many physical properties are temperature dependent, some ordering phenomena set in at low temperatures and the knowledge of the electronic structure in the ground state of the system, *i.e.* at T=0, is of fundamental importance. One of the most effective ways to cool down the sample down to tenths of a Kelvin is to liquefy He^3 gas. In many experiments reaching sub-Kelvin temperatures is not a problem, since thermal radiation, the main enemy of ultra-low temperatures, can be effectively shielded. Unfortunately, this is not the case in photoemission experiments. We need to provide free access for the incoming light and outgoing electrons. This is realized by specially designed slits in three radiation shields, having different temperatures. In order to compensate for the heat load caused by the photon beam and room-temperature radiation, the cooling power of the cryostat should be very high. This is achieved by the very large pumping speed of two pumps which reduce the vapor pressure above the liquid He^3, thus cooling the cold finger and sample. The design specifications of our He^3 system make it the most powerful worldwide. It is perhaps the only place on the planet where one can see a 1 K cold surface through a room temperature window, the "coldest visible".

The sketch of the modern photoemission experiment is shown in **Figure 1**. The synchrotron beam (dashed green line) illuminates the 1 K cold surface of the sample and excites photoelectrons. Electrons are projected to the entrance slit of the hemispherical analyzer, sorted in terms of angle (yellow, magenta and cyan traces correspond to different tilt angles) and then are analyzed in terms of kinetic energy. **Figure 2** shows the typical intensity distribution as a function of tilt angle and kinetic energy. Such an intensity distribution is indeed expected as the comparison with band structure calculations of this material shows (right panel). This is our window into reciprocal space.

By scanning voltages on the lens and hemispheres and rotating the sample around the vertical axis (polar angle) we can explore the broad binding energy range as well as wide regions of the reciprocal space with unprecedented detail. In particular, plotting intensity at the Fermi level as a function of both components of the in-plane momentum, calculated from tilt and polar angles, we have direct access to the Fermi surface (FS).

* Under "uncertainty" we understand the experimenter's best *estimate* of how far an experimental quantity might be from the "true value."

** The low-energy ring can have an energy of ~0.8 GeV, the high-energy one - up to 8 GeV.

Protocol

1. Mounting the Sample

1. This experiment uses synchrotron radiation produced by the BESSY storage ring of Helmholtz-Zentrum Berlin. The photons travel a beamline to our end-station where a sample is mounted.
2. Begin with a single crystal of the material to be investigated, here strontium ruthenate. Use silver-based epoxy to glue the sample to the sample holder. The silver-based epoxy ensures good thermal and electrical contact.
3. Glue an aluminum top-post to the surface of the single-crystal. The top-post will be used to cleave the sample in ultra-high vacuum to expose an atomically clean surface.
4. Mount the sample holder in the load lock.

2. Achieving Ultra-high Vacuum and Thermal Isolation

1. Begin evacuating the load lock to minimize contamination of the ultra-high vacuum chamber.
2. Monitor the pressure. Once a pressure of about 10^{-8} mbar has been achieved, transfer the assembly to the preparation chamber, and subsequently to the main chamber.The cold finger and sample holder have been specially designed to guarantee the best possible thermal contact with the helium pot.
3. These demonstration versions show how this is achieved by using conical surfaces to increase the area of contact. The conical surfaces are pressed against one another and the sample holder and cold finger are firmly fixed in place using a titanium nut and bolt.

3. Positioning and Cooling the Sample

1. The next step is to orient the sample within the cold finger along the azimuth using the transfer arm. Fix the position of the sample by tightening the nut while applying a counterforce with the supporting arm attached to the opposite side of the chamber.
2. Cleave the sample by moving the manipulator upwards so that the top-post is removed by interaction with the supporting arm.
3. With the beam shutter closed, move the sample into position in the beamline using the manipulator. Once the sample is in place, make sure the cryoshields are closed properly.
4. Start pumping on the 1-K pot and circulate the helium-3 gas in order to cool the sample to the base temperature. The temperature is measured close to the sample and will not change during the experiment.

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5. Open the beam shutter of the beamline. Use the micrometer screws on the apparatus to adjust the position of the sample so that it is at the focus of the analyzer lens. This adjustment is crucial.

4. Collecting Data

1. Once the setup is ready, switch to the angle-resolved mode of the analyzer and record the spectrum in swept mode. This will generate data for two-dimensional energy-angle plots.
2. Construct a Fermi surface map using the data. Select polar angles that correspond to Fermi level crossings for study of the superconducting gap of strontium ruthenate.
3. Record high-resolution spectra at the chosen polar angles above and below the superconducting transition temperature of strontium ruthenate to investigate the behavior of the superconducting gap.

Representative Results

The ultra-low temperatures of our setup together with the high resolution of the beamline and analyzer allow us to record spectra with very high overall resolution. This is illustrated in **Figure 3**. The usual test of the energy resolution is to measure the width of the Fermi edge of a metal. In this case it is a freshly evaporated indium film. The full width at half maximum (FWHM) of the Gaussian, which when convoluted with the step-function precisely describes the edge, is of the order of 2 meV. Of more importance for detailed studies of the low-energy electronic structure of solids is the angle-resolved spectrum of the dispersing feature. Such an example is shown in the middle panel of **Figure 3**. A very sharp superconducting peak is observed in the iron-based superconductor LiFeAs [1] representing one of the sharpest features ever detected by ARPES. The same holds for momentum resolution. The FWHM of 0.23 ° is the record value for the wide angular mode of electron energy analyzers. The system has been designed to combine three achievements, 1 meV bandwidth of the beamline, 1 meV resolution of the analyzer and 1 K temperature of the sample. This aim gave the name to our system "1-cubed ARPES". If all the three components were minimized, one would expect the FWHM~ 1.4 meV. Our current measurements show that overall resolution of the order of 2 meV can be achieved.

Another representative result is our investigation of the electronic structure of the superconductor Sr_2RuO_4 with critical temperature 1.35 K. This material is a well-known oxide possessing a wide spectrum of interesting physical properties. It was the first discovered superconducting oxide after the cuprates [2]. Its superconducting state is unusual: electrons coupled in pairs have their spins oriented in the same direction. This is the so called triplet superconductivity. It still remains not well understood and the main problem is to define the symmetry of the superconducting order parameter. In order to do that, the superconducting energy gap should be determined as a function of momentum - the exact task for ARPES. Because such low temperatures needed for superconductivity were not accessible in photoemission experiments before, it was not possible to address this problem. Here we make an attempt to do this. First of all, one needs to determine the Fermi surface. For this purpose we have recorded many cuts at different polar angles. Some of them are shown for illustration in **Figure 4**. Now, if we consider intensity only in the vicinity of the Fermi level, and plot it as a function of both angles, we will obtain the locus of Fermi momenta, *i.e.* the Fermi surface map. Such a Fermi surface map is shown in **Figure 5** together with the calculated Fermi surface [3]. The agreement is very good, but the experimental data show many more features. Some of them are unexpected and unusual [4]. Now we can try to measure the superconducting gap. For this we have to record the spectra above and below the critical temperature, *i.e.* 1.35 K. In **Figure 6** we show such pairs of spectra. We indeed observe some changes, compatible with the opening of the energy gap, but even the present energy resolution (or perhaps also the temperature, which can be higher right at the surface of the sample) does not allow us to draw a definite conclusion as for the energy gap in Sr_2RuO_4.

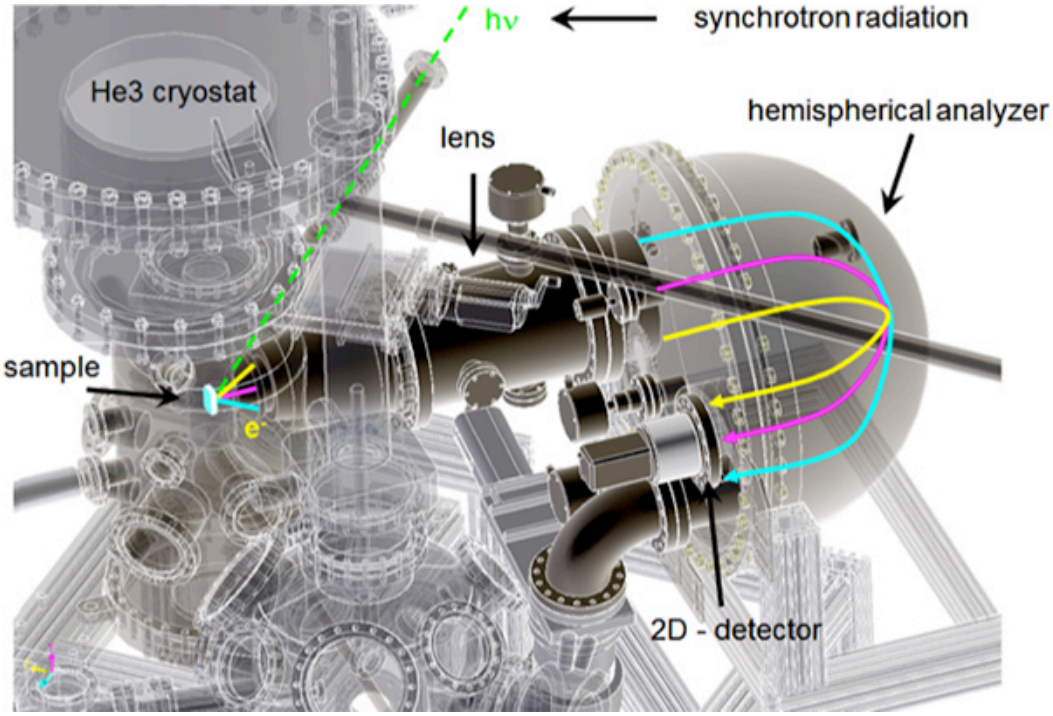


Figure 1. Schematic of the experimental setup.

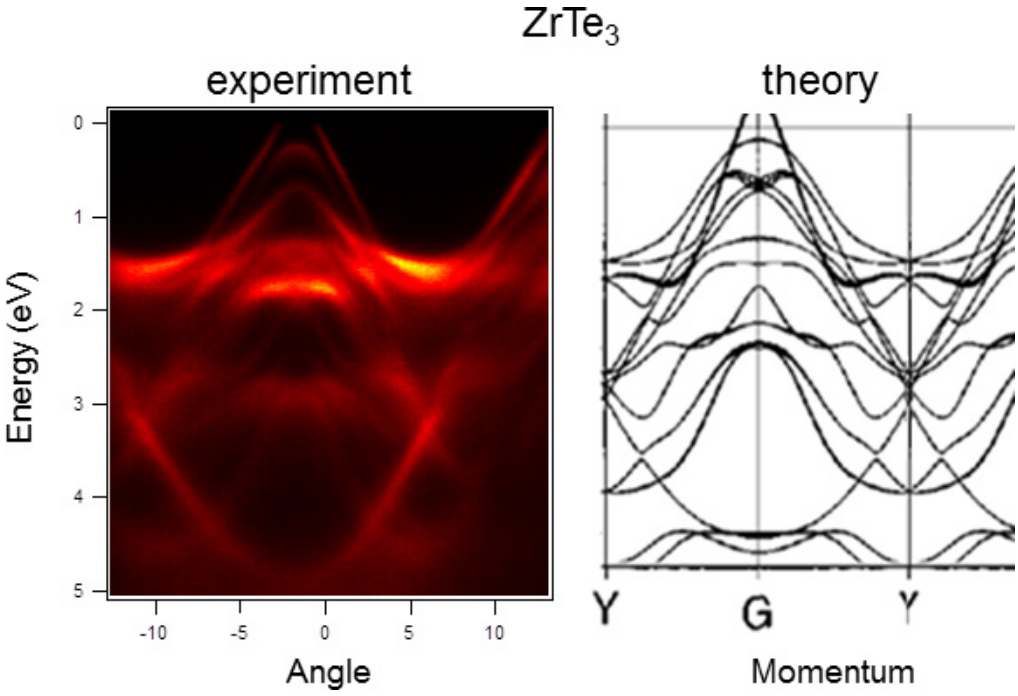
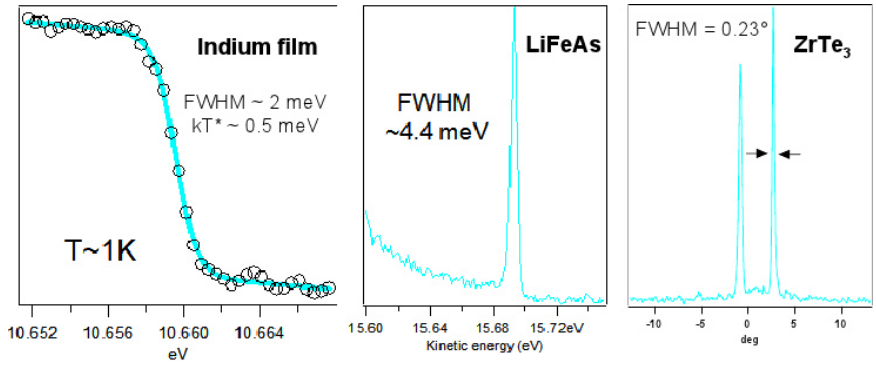


Figure 2. Left panel. Example of photoemission intensity distribution as a function of kinetic energy and angle. The band structure of the solid is directly seen. **Right panel.** Results of the band structure calculations for the same material. Theoretical data are taken from [5].



$$\sqrt{(1\ \mathrm{meV})^2 + (1\ \mathrm{meV})^2 + (1\ \mathrm{K})^2} \Rightarrow$$

FWHM ~ 1.4 meV
kT* ~ 0.4 meV

Figure 3. Performance of the experimental end-station. **Left panel.** Fermi edge of freshly evaporated indium film. **Middle panel.** Energy distribution curve (EDC) in a superconducting sample of LiFeAs. Data are taken from [1]. **Right panel.** Momentum distribution curve (MDC) at the Fermi level of $ZrTe_3$. Anticipated overall energy resolution of the system is expressed by the formula (1K= 0.0862 meV). The actual performance of the system is very close to the one we initially aimed for. Click here to view larger figure.

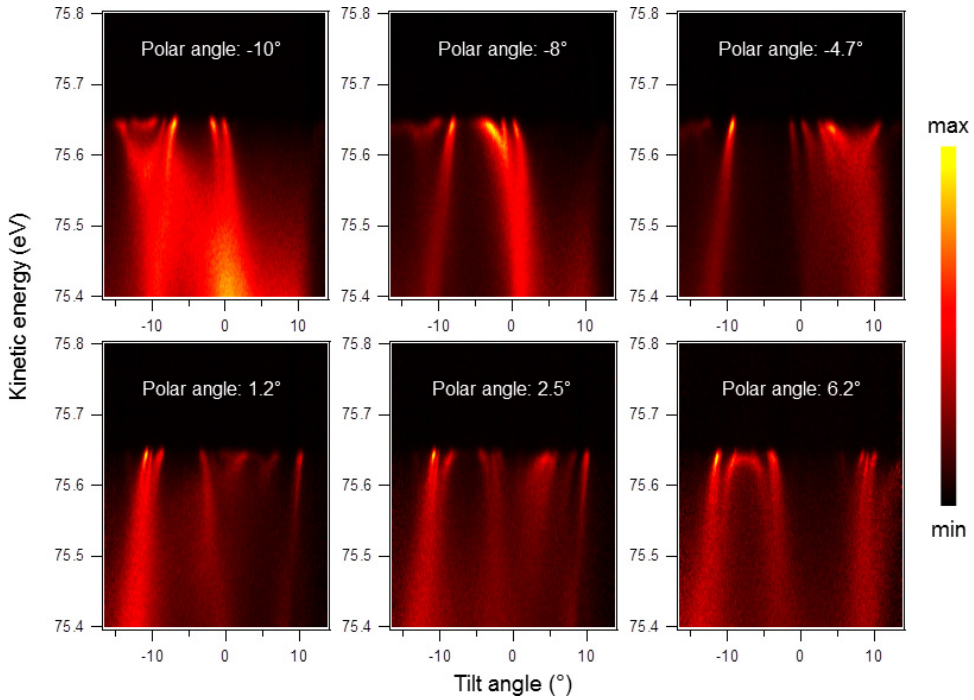


Figure 4. Energy-momentum cuts recorded at different polar angles (step 10) for Sr_2RuO_4. Click here to view larger figure.

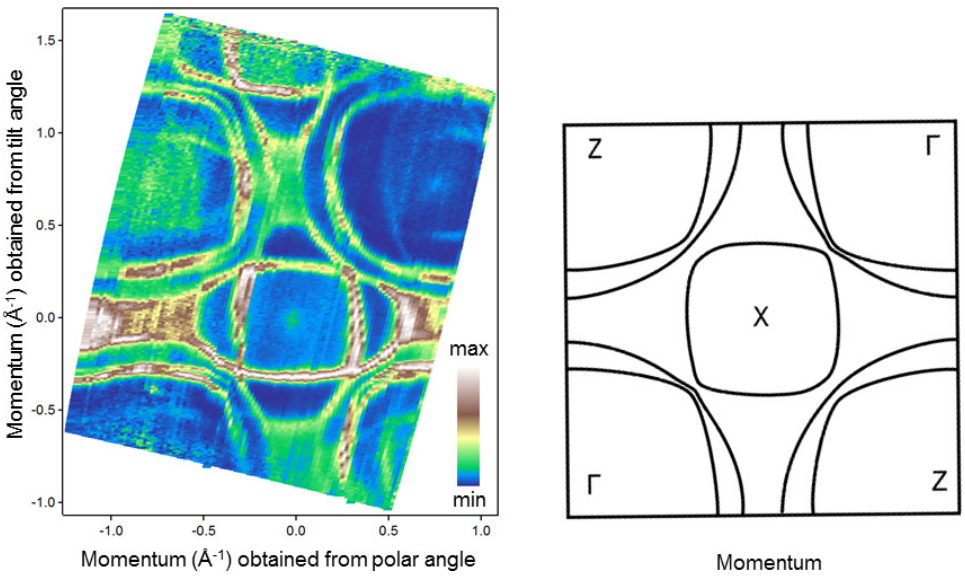


Figure 5. Fermi surface map of Sr_2RuO_4 taken using 80 eV linearly polarized light at ~1K. Click here to view larger figure.

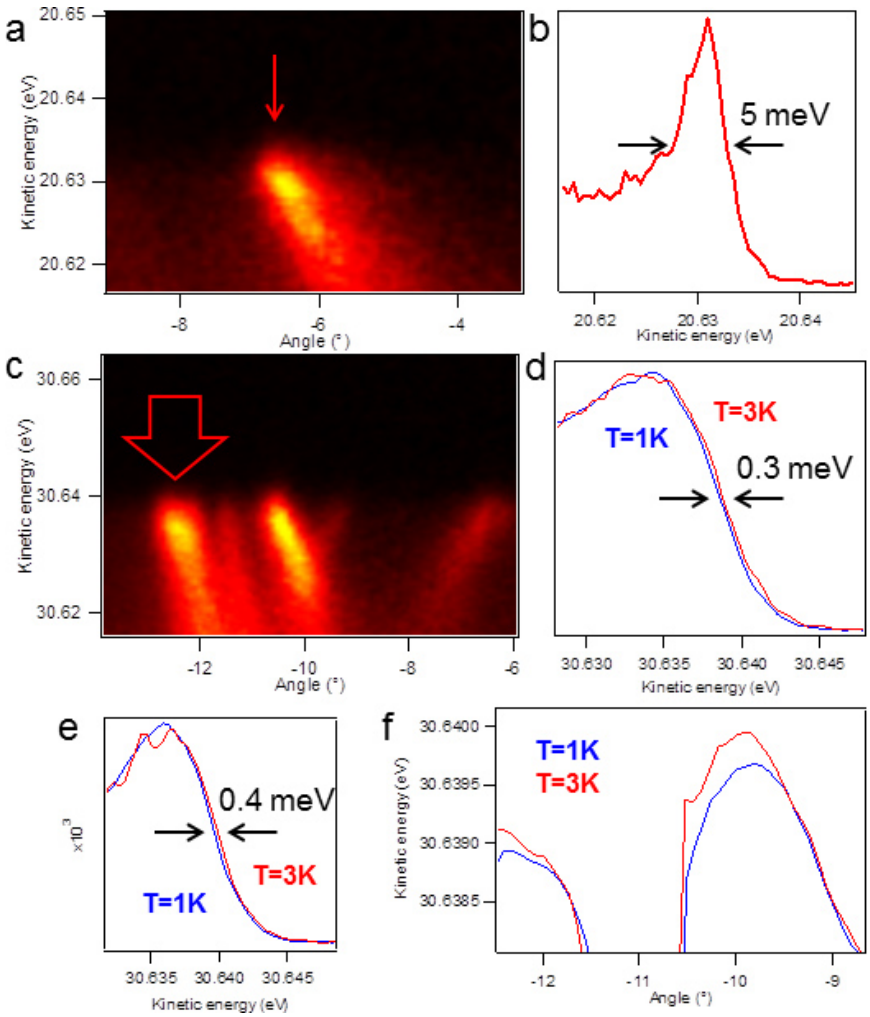


Figure 6. a, b Typical spectra taken to study the superconducting gap of Sr_2RuO_4. Red arrow indicates the momentum corresponding to a single energy distribution curve (EDC). T= 970 mK. **c, d** Shift of the leading edge of the integrated EDC. Momentum window is represented by the width of the red arrow. The gap corresponds to a FS point on the band near the BZ diagonal. **e** Shift of the kF EDC as a function of temperature from another point on the FS. **f** Typical temperature behavior of the binding energy of the leading edge in the vicinity of the crossing of two FSs.

Discussion

As is shown above, the implemented method is very efficient in studying the low-energy electronic structure of single crystals. Recent instrumental improvements have turned ARPES from a mere characterization and band-mapping tool into a sophisticated many-body spectroscopy. A modern experiment delivers information about the electronic structure of a solid or a nano-object with a new level of precision. Access to the Fermi surface in the case of a metal, energy gaps of semiconductors and insulators, their surface states, band structures and momentum dependent Fermi velocities allows one to characterize the electronic structure on a general level. Comparison with ab-initio calculations yields bandwidth- and Fermi velocity renormalizations and thus determines the complexity of the material in terms of the strength of correlations. Fine structures near the Fermi level often provide the possibility to detect the fingerprints of interaction between electrons and other degrees of freedom, like phonons, plasmons, spin-fluctuations etc. Systematic momentum dependent studies can identify the dominant interaction and single out, for example, a pairing mediator in superconductors or density wave systems. More thorough investigations involve the determination of the symmetry of the order parameter, thus providing critical tests for existing theories or stimulating new approaches on a fundamental level.

As in every experimental method, there are certain drawbacks. It is known that photoelectrons are strongly scattered inside the crystal, thus having a relatively short inelastic mean free path. As a result, the escape depth can be very small, down to several lattice constants. This defines the sensitivity of the method to the surface and in some cases the electronic structure of the surface is indeed different from the bulk [6]. However, ARPES offers many tools to monitor this situation. One of them is the use of various excitation photon energies. As has been mentioned previously, for a given photon energy it is possible to also estimate the component of momentum perpendicular to the surface. Observed periodic structures in ARPES spectra allow one to determine absolute $\boldsymbol{k}_z$ and the corresponding momentum resolution gives uncertainty of this quantity. In such a way the experimental value of the escape depth can be estimated from the uncertainty principle $\delta\boldsymbol{k}_z * \delta_z \sim 1$. The next tool to control the surface sensitivity of the method is variable polarization of the light. It was demonstrated earlier that using circularly polarized light it is

possible to distinguish between surface and bulk contributions to the photoemission signal [7]. Another advantage of using various polarizations and photon energies delivered by the synchrotron light source is the possibility to disentangle matrix element effects from genuine features of the spectral function. The matrix element is a transition probability which can suppress photoemission signal in particular regions of momentum space and result in misinterpretation of the ARPES spectra [8-10].

Obviously, the method is not very suitable for strongly 3D materials, which are difficult to cleave *in situ* and obtain atomically clean and flat surfaces. Finally, ARPES on insulators is much more complicated since it is necessary to compensate charging occurring because the flux of outgoing electrons cannot be compensated by electrical contact with the sample holder [11].

Disclosures

No conflicts of interest declared.

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