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The bulk-representative low-energy spectrum of Sr_2RuO_4 can be directly measured by angle-resolved photoemission. We find that the quasiparticle spectral line shape of Sr_2RuO_4 is sensitive to both temperature and momentum. Along the $(0, 0)$-$(\pi, 0)$ direction, both γ and β bands develop a sharp quasiparticle peak near k_F at low temperatures, but as the temperature increases the spectra quickly lose coherent weight and become broad backgrounds above $\sim$130 K, which is the metal-nonmetal crossover temperature, T_M, in the c-axis resistivity. However, spectra along the $(0, 0)$-(π, π) direction evolve smoothly across T_M. A simple transport model can describe both in-plane and c-axis resistivity in terms of the quasiparticle line shape. Comparisons are also made to the cuprates, with implications for two dimensionality, magnetic fluctuations, and superconductivity.

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The ruthenate, Sr_2RuO_4, has generated new interest since the discovery of superconductivity with $T_c \sim 1.5$ K [1]. It is the only noncuprate perovskite superconductor that is isostructural to the high-T_c cuprate $La_{2-x}Sr_xCuO_4$, and it is thus a good material to compare to high-T_c cuprates. Despite the structural similarity, Sr_2RuO_4 has different electronic and magnetic properties. While most cuprates have a single Cu3$d_{x^2-y^2}$-O2p band near the Fermi energy, Sr_2RuO_4 has multiple orbitals with Ru4$d_{xy,yz,zx}$-O2p character. The superconducting state of Sr_2RuO_4 is believed to have p-wave symmetry with enhanced ferromagnetic correlations. In contrast, high-T_c cuprates have a d-wave order parameter with proximity to antiferromagnetic ordering. The normal state transport properties of Sr_2RuO_4 also differ from the cuprates. In Sr_2RuO_4, while the in-plane resistivity ρ_{ab} is always metallic, the c-axis resistivity ρ_c is nonmetallic ($d\rho/dT < 0$) above $T_M \sim 130$ K, and becomes metallic ($d\rho/dT > 0$) below T_M [2]. Below 25 K, both ρ_{ab} and ρ_c have Fermi-liquid-like T^2 behavior, although with a large anisotropy of $\sim$600 [2]. In comparison, most cuprates have non-Fermi-liquid transport. Moreover, ρ_c in cuprates remains nonmetallic or incoherent down to T_c in most cases, implying that the cuprates are two dimensional in terms of coherent single-particle transport [3]. The 2D-3D crossover in Sr_2RuO_4 may shed light on the influence of two dimensionality on superconductivity. It is worth noting that this crossover has an electronic origin, since no associated structural change is observed [1].

In conventional metals, charge transport is understood in terms of the quasiparticle (QP) scattering rate (inverse lifetime) through the Drude formalism. However, in the layered correlated systems, such as cuprates and ruthenates, the c-axis transport can be nontrivial due to weak interlayer hopping. Many experimental [4–7] and theoretical [3,8–13] studies have investigated the peculiar c-axis transport. But the issue is complicated in cuprates by the high anisotropy and disorder induced by dopants, which make it difficult to obtain accurate and intrinsic c-axis transport properties. In contrast, Sr_2RuO_4 is a stoichiometric material with much less disorder. Therefore Sr_2RuO_4 is ideally suited for investigating the intrinsic c-axis transport and its effects on other properties, including superconductivity.

In this Letter, we report an angle-resolved photoemission spectroscopy (ARPES) study of spectral line shape in Sr_2RuO_4. We find that at low temperatures, the spectra along all Fermi surfaces are sharp and QP-like. However, along the $(0, 0)$-$(\pi, 0)$ (Γ-M) direction, sharp QP peaks quickly lose coherent intensity as T approaches T_M. In contrast, the QP linewidth along the $(0, 0)$-(π, π) (Γ-X) direction shows a smooth and conventional T dependence across T_M. Our analysis indicates that both in-plane and c-axis transport can be understood from the linewidth and coherent weight of QPs. In particular, the c-axis transport is controlled by interlayer tunneling of single particles, similar to the cuprates.

High-quality Sr_2RuO_4 single crystals were prepared by the floating zone method and subsequently annealed, resulting in a sharp superconducting transition at $T_c \sim 1.36$ K. ARPES experiments were performed at the Synchrotron Radiation Center, Wisconsin, and the National Synchrotron Light Source, New York. Several undulator beam lines were used at various photons energies (10–34 eV). We used Scienta analyzers capable of multiangle detection. Samples were cleaved *in situ* in a vacuum better than 8×10^{-11} Torr and yielded a flat (001) surface. The energy resolution is $\sim$10–20 meV, and the momentum resolution $\sim$0.02 Å^{-1}.

Earlier ARPES results [14,15] on the issue of the Fermi surface (FS) in Sr_2RuO_4 have some inconsistencies with

de Haas–van Alphen (dHvA) results and band calculations [16–18]. Recent ARPES studies [19–21] have since clarified this issue, showing that this is mainly due to the addition of a surface state near M $(\pi, 0)$, induced by a mild surface reconstruction, as observed by LEED and STM [22]. However, it has been observed that the photoemission matrix element has a different photon energy dependence for bulk and surface states in this material [19]. We find that at a 32 eV photon energy, the ARPES intensity from the surface state is greatly suppressed, as shown in Fig. 1 where we compare spectra along Γ-M for photon energies of 21.2 and 32 eV. While strong intensity from the surface state near M is seen for 21.2 eV photons, there is a substantial reduction of intensity from this surface contribution at 32 eV. Furthermore, the surface state in the vicinity of M can be almost eliminated by aging the sample surface *in situ,* as shown in Fig. 1(c). Note that the intensity of bulk peaks is also reduced by surface disorder. However, the aged coherent peak dispersion and line shape (without background) are found to be almost identical to fresh ones, indicating that the two peaks are bulk representative. In addition, we find they are perfectly consistent with dHvA measurements and local-density approximation band predictions [16–18].

We have extensive and reproducible data at many momenta and temperatures. Spectra of three representative k points, labeled A, B, and C, are displayed in Fig. 2. A and B are Fermi surface crossing points (FSCPs) along the $(0, 0)$-$(\pi, 0)$ direction, while C is a FSCP along $(0, 0)$-(π, π). The temperature dependence of the spectra at A and B are almost identical, as shown in Fig. 2(a). At low temperatures, the spectra at both points show sharp QP-like peaks. As T increases, the QP peak quickly loses its intensity and leaves a broad background-like feature at high temperature; see $T = 180$ K. Note that the temperature range in which the coherent peak vanishes is close to the metal-nonmetal crossover temperature of c-axis transport. In contrast, the spectral behavior at C is quite different, as seen in Fig. 2(b). As T increases, the spectral linewidth at C broadens smoothly from 20 to 180 K, and there is no rapid loss of the coherent peak. This rather conventional behavior is consistent with the metallic behavior of the in-plane transport.

It is known that transport of Sr_2RuO_4 is anisotropic; the temperature dependence of in-plane and c-axis resistivity are quite different. Since the transport is intrinsically related to quasiparticle lifetime, which is inversely proportional to the QP linewidth, the different QP behavior in k space may cause the anisotropic transport properties. In the following we develop a more quantitative analysis to compare ARPES and transport results.

For the in-plane transport, since optical reflectivity experiments observe a Drude peak [23], it is reasonable to use the Drude formula $\sigma_{ab} = (ne^2\hbar/m^*\Gamma)$. To obtain the QP linewidth, Γ, we use a simple fit as demonstrated in Fig. 3, where a Lorentzian is used to fit the QP peak, and a linear term to fit the incoherent background. We also include the effects of the Fermi function and energy resolution. Excellent fitting results were achieved for every spectrum in Fig. 2, with some examples shown in Fig. 3(a).

Through fitting, we extract two quantities, namely, the coherent linewidth, Γ, and the coherence ratio (Fermi liquid quasiparticle residue), Z, estimated as the ratio of the coherent to total spectral weight [24]. Both quantities are shown together in Fig. 3(b). For the three k points (A, B, and C), Γ increases with T. The behavior of Z at A

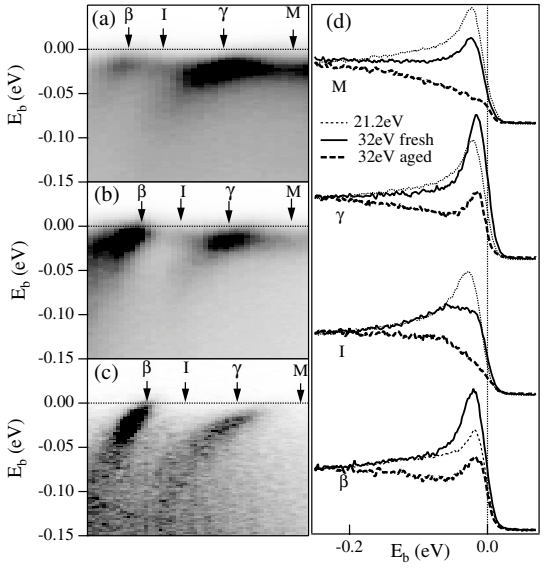

FIG. 1. Photon energy dependence of ARPES spectra along Γ-M. Intensity plots for (a) 21.2 eV photons, (b) 32 eV photons on a fresh surface, and (c) 32 eV photons on an aged surface. (d) Comparison of spectra at 32 eV fresh (solid lines), 32 eV aged (thick dashed lines), and 21.2 eV (thin dashed lines) at four locations marked in the intensity plots.

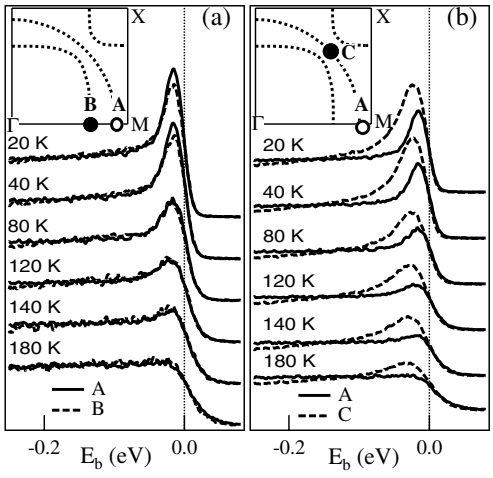

FIG. 2. Temperature dependence of spectra at the three FSCPs: A, B, and C (see the insets). (a) Comparison of spectra between A and B. (b) Comparison of spectra between A and C. The insets show measurement locations in the Brillouin zone.

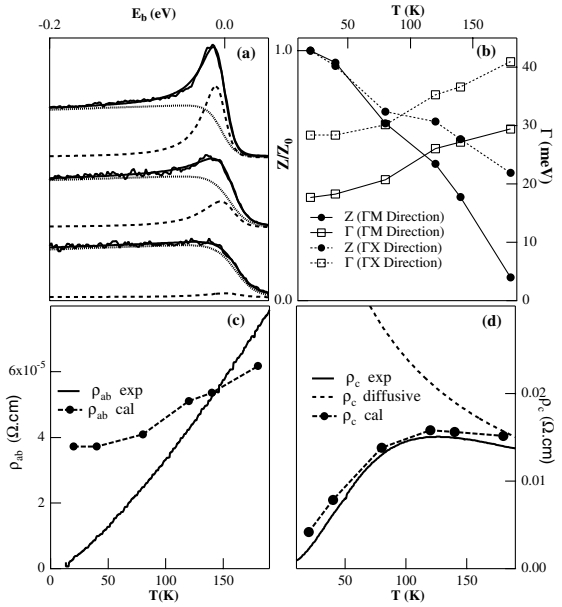

FIG. 3. (a) Example fits extracting Lorentzian coherent peaks from spectra at point *A*. (b) QP linewidth Γ and coherence ratio Z/Z_0 for points *A* and *C* (results from point *B* are almost identical to *A*). Calculated and experimental resistivity is plotted for (c) the in-plane and (d) the *c* axis. The effect of energy resolution is removed from the linewidth in the calculation.

and *B*, which are close to $(\pi, 0)$, is rather unusual—Z decreases rapidly as T increases, approaching zero after passing through T_M. In contrast, the reduction of Z is much slower at *C*. Even at 180 K, it retains about 60% of its low-T value.

To make a rough estimate for ρ_{ab}, we use the linewidth (Γ) at point *C*, and n, m^* from dHvA results [16], in the Drude formula. The calculated results are plotted along with the experimental ρ_{ab} in Fig. 3(c), showing good agreement in terms of the order of magnitude. The discrepancy between the calculated and experimental values at low T is mainly due to the oversimplification based on a single Fermi surface point, and the possible overestimate of ARPES line shape due to overlapping of two bands at this point [21].

The same Drude approach, however, clearly does not work for the *c*-axis transport since ρ_c is hundreds of times larger than ρ_{ab}. We consider the hopping process along the *c* axis and follow the analysis by Mackenzie [16],

$$\sigma_c^{\text{direct}} = \frac{4e^2 c}{\pi\hbar^4}\langle t_\perp^2 m^* \tau\rangle, \tag{1}$$

where c is the *c*-axis unit cell length (12.7 Å), $t_\perp$ is the hopping integral along the *c* axis, m^* is the in-plane effective mass, and τ is the in-plane lifetime. $\langle \ldots \rangle$ denotes the sum over all three FSs. Equation (1), however, requires that the QP mean free path along the *c* axis be longer than the interlayer distance, $l_c > c/2$, which implies direct *c*-axis interlayer tunneling. A shorter mean free path leads to diffusive tunneling, during which in-plane momentum is not conserved. Since $l_c \sim l_{ab} c \Delta k_F$, where Δk_F is the *c*-axis bandwidth, we find that only the quasiparticle component of the in-plane spectral function with sufficient lifetime can contribute to direct tunneling. Using the data from Ref. [16], we estimate the corresponding maximum linewidth $\Gamma \sim 30$ meV to satisfy the condition $l_c > c/2$. This implies that only the sharp QP peaks contribute to the direct hopping. Therefore, one needs to *add* the coherence ratio, Z^2, as a prefactor to the above expression for σ_c. The reason for using Z^2 instead of Z is that $t_\perp$ is proportional to Z. The same conclusion is implied in Ref. [11]. Furthermore, we note that the main contribution to Eq. (1) comes from the FSs in the vicinity of $(\pi, 0)$. This is mainly because the enhanced density of states (DOS) around $(\pi, 0)$, due to a saddle point in the band dispersion, would make the $(\pi, 0)$ contribution stronger, by increasing the effective mass, m^*. We then obtain

$$\sigma_c^{\text{direct}} \simeq \frac{4e^2 c}{\pi\hbar^3}\sum_{\beta,\gamma}\frac{Z^2 (t_\perp^0)^2 m^*}{\Gamma_{(\pi,0)}}. \tag{2}$$

In Fig. 3(d), we plot the calculated ρ_c along with the experimental ρ_c. In our calculation, ρ_c has both direct and diffusive hopping contributions, $1/\rho_c = 1/\rho_c^{\text{direct}} + 1/\rho_c^{\text{diffusive}}$. As first proposed by Kumar *et al.*, the diffusive term should have a form of A/τ, where $\tau \propto k_B T + \tau_0$ for a finite range of T [8]. The resulting resistivity depends only on two parameters, $t_\perp^0$ and τ_0. As can be seen in Fig. 3(d), our simple model gives good agreement with the experimental ρ_c. The $t_\perp^0$ used in the calculation is $\sim$2.2 meV (25 K), in good agreement with the value ($\sim$ several 10 K) obtained from both band calculations [17,18] and dHvA measurement [16]. The good description of ρ_c and the correct estimate of $t_\perp^0$ supports this simple model of the *c*-axis transport.

From the analysis above, we see a one-to-one correspondence between the emergence of a sharp QP and the onset of the *c*-axis coherent transport. Because of the anisotropy of the effective mass and the interlayer hopping integral, such a correspondence may be limited only to some certain momenta along the FS. We believe that the association between the in-plane QP and the coherent *c*-axis transport may in general be valid for many quasi-2D materials. A similar observation has been reported for other layered materials, such as the cobaltates [25].

In the case of cuprates, as mentioned above, the normal state *c*-axis transport is incoherent in the optimally and underdoped region, and ARPES observes a broad, ill-defined peak in the vicinity of $(\pi, 0)$ above T_c. This can be seen from Fig. 4, where we compare the T evolution of Sr_2RuO_4 and Bi2212 at $(\pi, 0)$ and along $(0, 0)$-(π, π). Similar to Sr_2RuO_4, the loss of the QP-like peak in Bi2212 is dramatic at $(\pi, 0)$, where the spectrum becomes a smeared background-like feature above a certain T. In Sr_2RuO_4, this scale is T_M, while in Bi2212 it is T_c. The behavior along the $(0, 0)$-(π, π) (nodal) direction is quite different; the QP-like peak seen at low T broadens

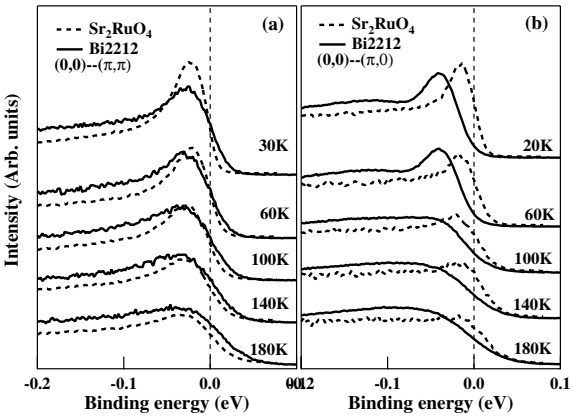

FIG. 4. Comparison of spectra at different temperatures for slightly underdoped Bi2212 and Sr_2RuO_4 at FS crossing points (a) along the $(0, 0)$-(π, π) direction and (b) near $(\pi, 0)$.

smoothly through T_c, remaining a relatively well-defined peak even at elevated T.

Applying the same analysis as in the ruthenate, we conclude that in cuprates ρ_c is controlled by the $(\pi, 0)$ spectrum, and ρ_{ab} by the nodal QP. A similar claim has been suggested by other groups [10,12]. Despite many similarities in spectra, there is one major difference between the two materials in the coherent QP peak at $(\pi, 0)$. Unlike the ruthenate whose QP peak is related to the coherent single-particle interlayer tunneling, the cuprate develops a $(\pi, 0)$ coherent peak in the superconducting state where it is suggested that at T_c the single-particle c-axis tunneling may still be incoherent [3]. Note that the superconducting coherent peak is not the consequence of the opening of an energy gap, as seen in Fig. 4(b) where there is no sharp peak in the 100 K curve which has a clear pseudogap, consistent with previous observations [26,27]. Therefore the origin of this $(\pi, 0)$ coherent peak developing below T_c in Bi2212 is an important issue currently under debate. We also note that ARPES has observed that the $(\pi, 0)$ coherent peak in Bi2212 has a similar temperature and doping dependence as the superfluid density [28,29], suggesting that this single-particle coherent peak may be closely related to pair coherence in cuprates.

With the establishment of links between quasiparticle coherence and coherent interlayer tunneling, one remaining question is what kind of scattering process causes the rapid loss of QP coherence. In the cuprates, it is argued that the (π, π) antiferromagnetic fluctuations, which connect two FS points near $(\pi, 0)$, enhance QP scattering in the vicinity of $(\pi, 0)$. However, this issue is far from clear in Sr_2RuO_4. It is generally believed that ferromagnetic correlations are important in this material. A possible explanation for the rapid loss of QP coherence near $(\pi, 0)$ is due to a small Q-vector scattering off the high DOS caused by Van Hove singularity (VHS) at $(\pi, 0)$. At high T, electrons at E_F encounter strong scattering from the the highly enhanced DOS due to the VHS, since the VHS energy is very close to E_F [17,18], within a few times of k_BT. At low T when the thermal excitation is smaller than the energy of the VHS, these scattering channels are cut off, a true QP then emerges in the spectral function. Therefore the position of the VHS determined the crossover temperature, T_m.

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