High-Energy Anomaly in the Band Dispersion of the Ruthenate Superconductor

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We reveal a "high-energy anomaly" (HEA) in the band dispersion of the unconventional ruthenate superconductor Sr_2RuO_4, by means of high-resolution angle-resolved photoemission spectroscopy (ARPES) with tunable energy and polarization of incident photons. This observation provides another class of correlated materials exhibiting this anomaly beyond high-T_c cuprates. We demonstrate that two distinct types of band renormalization associated with and without the HEA occur as a natural consequence of the energetics in the bandwidth and the energy scale of the HEA. Our results are well reproduced by a simple analytical form of the self-energy based on the Fermi-liquid theory, indicating that the HEA exists at a characteristic energy scale of the multielectron excitations. We propose that the HEA universally emerges if the systems have such a characteristic energy scale inside of the bandwidth.

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Understanding electron correlation is being actively pursued in modern condensed-matter physics. Landau's Fermi-liquid (FL) theory is a fundamental and a very successful paradigm describing normal metallic states of interacting electrons in many systems [1]. Within the framework of the FL theory, the interacting electrons can be described as "quasiparticles"—independent particles but with renormalized mass (m^*). In general, the effective mass enhancement is caused by two primal many-body interactions [2]: electron-electron interaction (EEI) and electron-boson interaction (EBI). EEI is responsible for band renormalization over a large energy scale (eV order) accompanying a "band narrowing," whereas EBI is responsible for a low energy-scale one near the Fermi level (E_F), which causes a "kink" in the energy-band dispersion at the bosonic-mode energy (sub-eV order).

Besides these well-established band renormalization effects, a "high-energy anomaly" (HEA) in the band dispersion is indicative of a strong renormalization effect, which has been widely observed using angle-resolved photoemission spectroscopy (ARPES) in various high-T_c cuprate families. Tremendous experimental [3–11] and theoretical [12–23] effort has been devoted to identifying the physics behind the HEA. Experimentally, the HEA has been characterized by two spectral features in the energy range of ~300–600 meV: a giant kink ("high-energy kink") and a nearly vertical dispersion ("waterfall"), though photoemission matrix-element effects also play an important role in the spectral appearance especially around the waterfall region [8,9]. After an intense debate on the origin of the HEA, in a long list of proposals given so far, the self-energy approach [20–23] has been received as the mainstream consensus. However, even though the HEA seemingly originates in self-energy effects, ambiguity remains in a key ingredient of excitations, whether these arise from strong correlation [7,14,15,19,23], or couplings to high-energy bosonic modes such as phonons [5], antiferromagnetic spin fluctuations or paramagnons [6,17,18,20], and plasmons [13]. One reason for this might be that, so far, the energy scale of the HEA has not been quantitatively determined experimentally.

In this Letter, we report the first observation of the HEA in the band dispersion of single layered ruthenate Sr_2RuO_4, using high-resolution ARPES. Further, we demonstrate two distinct types of band renormalization associated with and without the HEA in Sr_2RuO_4. One type, observed in the d_{xy} band, yields both positive and negative energy shifts, i.e., an effective mass enhancement near E_F as well as an increase in the bandwidth. Accordingly, the HEA emerges at around their crossover region identified by a characteristic energy (ω_c), at which the real and imaginary parts of the self-energy becomes zero and shows a local minimum, respectively. In contrast, the other type, observed in the d_{zx} band, yields only band narrowing because the bandwidth is smaller than ω_c. These distinct types of band renormalization can be reasonably explained by a simple self-energy analysis based on the FL theory, indicating that the multielectron excitations are responsible for the HEA. The generality of the HEA will also be detailed.

High-quality single crystals of Sr_2RuO_4 were grown by the floating-zone method using the self-flux technique [24]. The present data were collected at BL-1 of the Hiroshima Synchrotron Radiation Center using a VG-SCIENTA R4000 electron analyzer. All measurements were performed after cleaving the samples *in situ* under ultrahigh-vacuum conditions $\sim 2 \times 10^{-11}$ Torr at $T \sim 10$ K. To reduce complexity due to the surface-derived states [25], we aged the sample surface and then collected the ARPES data with the energy and angular resolution of 30 meV and 0.3°, respectively.

The tunability of the excitation energy ($h\nu$) and polarization [Fig. 1(a)] of incident photons are indispensable in the present ARPES study. In Fig. 1(c), two bands derived from the d_{zx} and d_{xy} orbitals cross E_F along the Γ–M or Z–M high-symmetry line [Fig. 1(b)], and these can be selectively observed using p- and s-polarized light as shown in Figs. 1(d) and 1(e) following the dipole selection rule [26,27]. Note that those experimental conditions were carefully determined to examine wide energy-band dispersions by surveying $h\nu$'s (22–100-eV) and momentum spaces (1st and 2nd Brillouin zones). For instance, the significant matrix-element effects become apparent under certain conditions, as seen in Figs. 1(f) and 1(g), where spectral weights near the zone center (Γ or Z) are strongly suppressed as similarly observed in cuprates [8,9,20].

The experimental energy-band dispersions are accurately determined over an extended energy region by standard line-shape analysis (see caption to Fig. 2) as seen in Figs. 2(a) and 2(b), both, indicating good agreement between experimental dispersions and contour ridges of the ARPES images. By comparing the ARPES and local-density approximation (LDA) dispersions [Figs. 2(c) and 2(d)], the large energy-scale renormalization effects seem at first glance to be highly band dependent. The group velocity of the ARPES dispersion for the d_{zx} band is entirely slower than that of the LDA dispersion. By contrast, for the d_{xy} band, the ARPES dispersion shows slower and faster group velocities compared with the LDA dispersion at the respective energies above and below the intersection of their dispersions. Furthermore, the d_{xy} band undergoes vertical dispersion near the intersection of the ARPES and LDA dispersions. This waterfall-like dispersion pattern is an unambiguous signature of the HEA, as reported for cuprates [3–11].

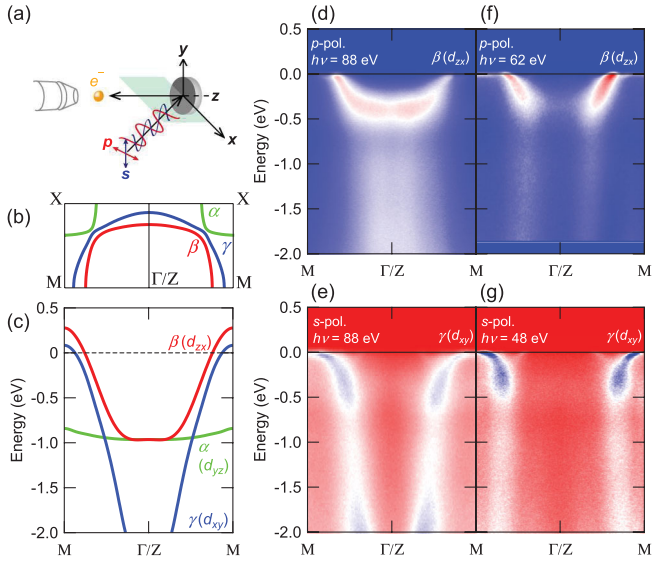

FIG. 1 (color). (a) Schematic polarization geometry used in the ARPES experiment, where the p-polarized light and s-polarized light are incident on the sample parallel and perpendicular with respect to the detection plane (xz plane), respectively. (b),(c) Respective Fermi surfaces and electronic bands along the Γ–M or Z–M line of Sr_2RuO_4 as determined from LDA calculations without hybridization between Ru $4d$ and O $2p$ states [26]. (d)–(g) ARPES images recorded along the Γ–M or Z–M line with different polarizations and incident photons.

To visualize the energy renormalization, we derived the real part of the self-energy $\Sigma'(\omega)$ [28] of the d_{zx} and d_{xy} bands, displayed respectively in Figs. 2(e) and 2(f) together with previous theoretical calculations [29]. The global features of $\Sigma'(\omega)$ show the close resemblance between experiment and theory, implying the validity of parameter values used in Ref. [29] ($U = 1.2$ eV, $J = 0.2$ eV). A clear deviation can be recognized in the low-energy region for the d_{xy} band, though not for the d_{zx} band, where the contributions due to the EBIs as well as the spin-orbit interaction [26,27] can be expected. A further quantitative evaluation of low-energy renormalization effects is in progress, but is beyond the scope of this work.

Next, the $\Sigma'(\omega)$ values were fitted by an analytical formula, $\Sigma_{\mathrm{model}}(\omega) = g\omega/(\omega + i\gamma)^2$, which satisfies causality and the FL theory [28]. As evidenced by the good agreement between experimental and model $\Sigma'(\omega)$ and $\Sigma''(\omega)$ shown in Figs. 2(e) and 2(f), the HEA should be related to the electronic excitations. This is also supported by the fact that the modeled spectral functions well reproduce the global spectral shapes and intensities as seen in Figs. 2(g) and 2(h).

Our results place strong constraint as to what is responsible for the HEA. Notably, there exists a characteristic energy (ω_c) of $\sim$1.2 eV satisfying $\Sigma'(\omega_c) = 0$ and a local minimum $\Sigma''(\omega_c)$, thereby yielding a maximum linewidth $2|\Sigma''(\omega_c)|$. The energy scale of the HEA should correspond to the ω_c, at which the sign of the $\Sigma'(\omega)$ reverses as well as the spectral intensity becomes suppressed because of the enhanced linewidth broadening. Thus, one cannot rigorously define a group velocity in the sense of the one-electron picture around the ω_c or the waterfall-type spectral shape where the coherent peak is not well defined. If the coupling parameter for the HEA (g/γ^2 in our model [28]) is strong enough, the energy band should be split into high- and low-energy branches. This situation is similar to that expected from the "coherent-incoherent" picture recently discussed for cuprates [11,21]. However, we stress here that the observed d_{xy} band is not independent of $\boldsymbol{k}$ even for $|\omega| > |\omega_c|$ but is indeed well dispersed, obviously different than "incoherent states." This observation thus favors the "quasiparticle" scenario [5,7] in which the Zhang-Rice singlet band [30] is described by a single quasiparticle band with a large bandwidth.

The observed characteristics of the HEA—that is, the zero point of the $\Sigma'(\omega)$ as well as the local minimum of the $\Sigma''(\omega)$—cannot be explained by the EBI. This is because any bosonic modes, irrespective of bosonic type such as

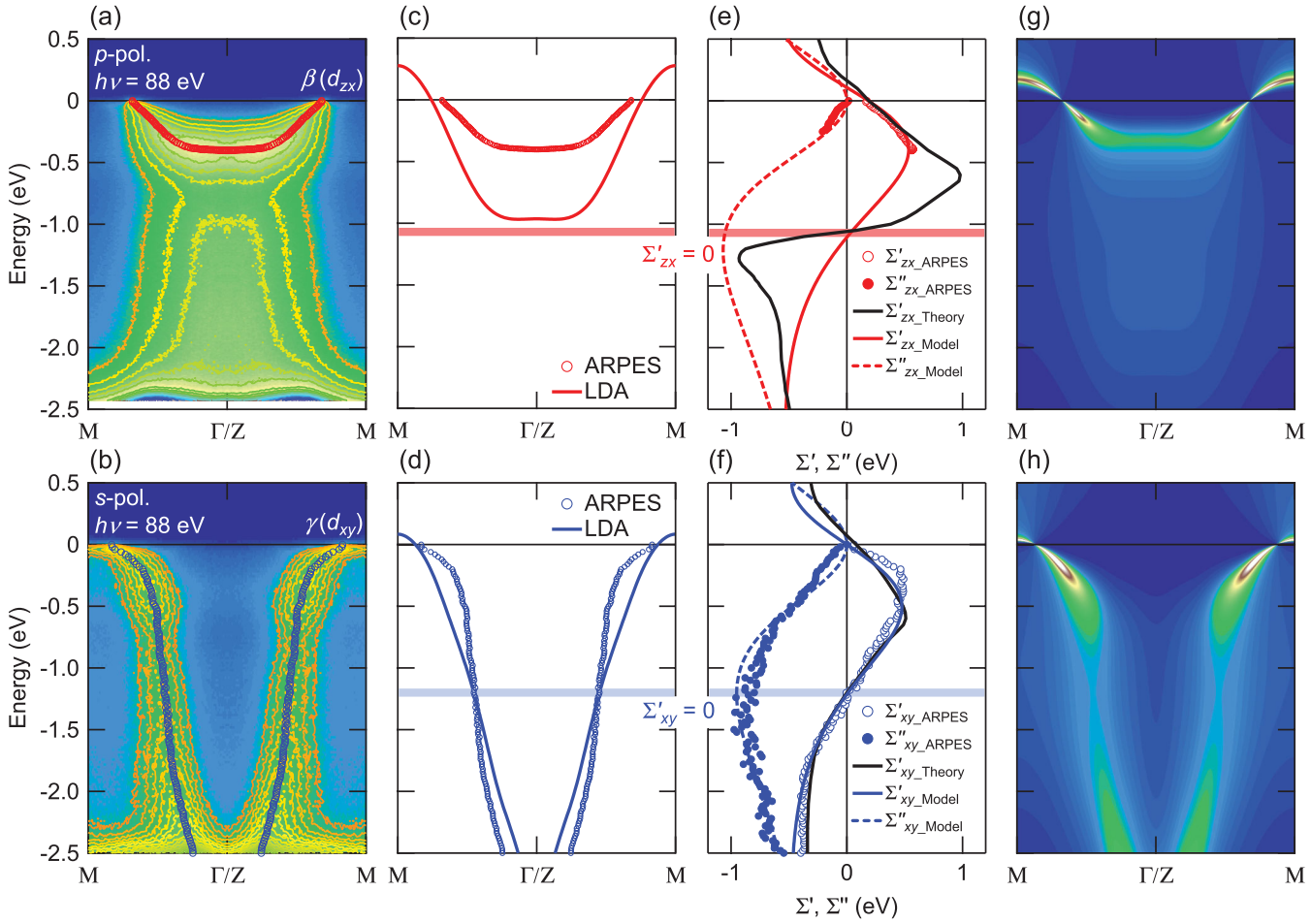

FIG. 2 (color). (a),(b) ARPES images of Sr_2RuO_4 along the Γ–M or Z–M line and their contour plots together with the energy-band dispersions (circles) recorded with 88-eV photons with p and s polarization, respectively; ARPES intensities are symmetrized with respect to the zone center (Γ or Z). (c),(d) Comparison of the energy-band dispersion between ARPES results (circles) and LDA calculations (line) for d_{zx} and d_{xy} bands, respectively. The experimental dispersions were determined by fitting the momentum distribution curves, except for the flat dispersion around the bottom of the d_{zx} band, which was determined by fitting the energy distribution curves. (e),(f) Real and imaginary parts of the self-energy $\Sigma'(\omega)$ (open circles) and $\Sigma''(\omega)$ (closed circles) for d_{zx} and d_{xy} bands, respectively. These were obtained by $\Sigma'(\omega) = \omega - \varepsilon_k^{\mathrm{LDA}}$ and $\Sigma''(\omega) = -\frac{1}{2} v_0^{\mathrm{LDA}} \Delta k$, where $\varepsilon_k^{\mathrm{LDA}}$, v_0^{LDA}, and Δk represent the LDA dispersion, and the velocity of the LDA dispersion, and the momentum distribution curve full width, respectively. For comparison, the $\Sigma'(\omega)$ (black line) from previous theoretical calculations [29] is also shown. Solid and dashed lines in blue (red) represent the real and imaginary parts of the modeled self-energy [$\Sigma'_{\mathrm{model}}(\omega)$ and $\Sigma''_{\mathrm{model}}(\omega)$] obtained by fitting $\Sigma'(\omega)$. (g), (h) Calculated spectral functions for d_{zx} and d_{xy} bands, respectively, where LDA dispersion is used as the bare dispersion and blue and red lines in (e) and (f) are used as the input self-energies.

phonons, magnons, and plasmons, are expected to couple most strongly to the electrons at their own specific mode energies, always driving the effective mass enhancement associated with the gradient but not the zero point of the $\Sigma'(\omega)$ [31]. In addition, the energy scale of the HEA is completely different from that of bosonic couplings already reported for Sr_2RuO_4: the phonon distribution is just up to ~80 meV [26,27,32–34], and the ferromagnetic or antiferromagnetic fluctuations are only related with the low-lying excitations below ~8 meV [35–37]. Judging from the eV-order energy scale of the ω_c, we thus conclude that electron correlation due to the EEI is the remaining candidate for the origin of the HEA. In the present case, the HEA energy-scale can be attributed not to the exchange interaction $J \sim 0.2$–0.7 eV [29,38], but to the on-site Coulomb interaction $U = 1.2$–1.5 eV [29,39].

Our results highlight two distinct types of band renormalization depending on the energetic relationship between the ω_c and bare band-bottom energies ($\omega_0^{xy,zx} \sim \omega_{\mathrm{LDA}}^{xy,zx}$). One, observed in the d_{zx} band, yields only band narrowing associated with the increased effective mass, because $|\omega_{\mathrm{LDA}}^{zx}| < |\omega_c|$, that can be well described by the canonical coherent-incoherent picture as predicted by the infinite-dimensional Hubbard model calculations [40]. The other,

observed in the d_{xy} band, yields that the effective mass of the d_{xy} electrons becomes heavier for $|\omega| < |\omega_c|$ but the band-bottom energy is reduced leading to a larger W, resulting in the HEA because $|\omega^{xy}_{\mathrm{LDA}}| > |\omega_c|$. The indication is that the effective mass enhancement is not always simply measured by U/W [41]. Indeed, the large energy-scale mass enhancement, $1 - \partial\Sigma'_{\mathrm{model}}/\partial\omega|_{\omega=0}$, was estimated to be about 3 times heavier compared with the LDA band mass irrespective of band characteristics (d_{xy}, d_{zx}), but consistent with the dynamical mean-field theories [29,38].

In most $3d$ transition metal oxides, the ω_c might be related to the exchange interaction J because $U \gg W$. Indeed, in the case of cuprates, the reported energy scales of the HEA ($\sim$ 300–600 meV) are indeed similar to $3J \sim 0.35$ eV [42]. However, it has been also argued that the doping dependence of the HEA cannot be simply described by the J scale [7]. To pin down the energy scale and the origin of the HEA in cuprates, an analysis similar to that presented here on various doping levels and families in a cuprate system is highly desired.

One intriguing question is: How generic is the HEA-type renormalization for other systems? We clearly demonstrated that at the heart of this type of renormalization is the existence of the characteristic energy (ω_c), specifically giving the zero and local minimum points in the $\Sigma'(\omega)$ and $\Sigma''(\omega)$, respectively. In the present model, these characteristic structures in the self-energy stems from two conditions for the $\Sigma''(\omega)$ [28]: (1) ω^2 dependence near E_{F} to satisfy the FL theory, and (2) the zero-convergence at infinite energy, i.e., $\lim_{\omega\to\infty}\Sigma''(\omega) = 0$, to satisfy the causality. From the second condition, the existence of the ω_c should be generally expected for the electron self-energy, as long as the $\Sigma''(\omega)$ has some energy dependence in the low-energy region irrespective of its functional form. Hence, the HEA and band narrowing are a ubiquitous electronic feature in the systems with $|\omega_c| < |\omega_0|$ and $|\omega_c| > |\omega_0|$, respectively. These new and generic aspects of the band renormalization provide a platform for a comprehensive understanding of electron correlation.

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