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Direct observation of a quasiparticle band in $\mathrm{CeIrIn_5}$: An angle-resolved photoemission spectroscopy study

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We have performed a high-resolution angle-resolved Ce 4*d*-4*f* resonant photoemission experiment on the heavy fermion superconductor $\mathrm{CeIrIn_5}$. We have observed a quasiparticle band, which has an energy dispersion of ~30 meV in the Ce 4*f* on-resonance spectra. The result suggests that although the 4*f* spectra are dominated by the localized and/or correlated character, the small itinerant component is responsible for the superconductivity in this compound.

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I. INTRODUCTION

The discovery of superconductivity in heavy fermion (HF) *f*-electron compounds near a magnetic quantum critical point (QCP) has stimulated the studies of superconductivity in this class of materials.[1,2] In these compounds, both magnetism and superconductivity are originated from the *f* electrons, which are considered to have dual character, i.e., both localized and delocalized character.[3] The recently discovered $\mathrm{Ce}T\mathrm{In_5}$ ($T=\mathrm{Rh,Ir,Co}$) compounds have attracted much attention because they are antiferromagnets or superconductors depending on the chemical composition and pressure, which suggests that the system is located near a QCP. Therefore, they are good target materials to study how magnetism and superconductivity are related to each other. At ambient pressure, $\mathrm{CeRhIn_5}$ (Ref. 4) becomes antiferromagnetic below $T_N=3.8$ K, while $\mathrm{CeIrIn_5}$ (Ref. 5) and $\mathrm{CeCoIn_5}$ (Ref. 6) become superconducting below $T_C=0.4$ and 2.3 K, respectively. For the $\mathrm{CeRh}_x\mathrm{Ir}_{1-x}\mathrm{In_5}$ alloy compounds, it was reported that the antiferromagnetic (AF) long-range order and the superconductivity coexist for $0.3<x<0.6$.[7] These differences in the ground-state properties of the different $\mathrm{Ce}T\mathrm{In_5}$ compounds are thought to be originated from the differences in the lattice constants of the quasi-two-dimensional (2D) $\mathrm{CeIn_3}$ layers and, hence, the difference in the *f*-*p* hybridization strength. Therefore, this system provides an opportunity to study the relationship between magnetism and superconductivity in *f*-electron systems.

There are several experimental studies on the electronic structure of *f* states in these compounds. For antiferromagnetic $\mathrm{CeRhIn_5}$, Shishido *et al.*[8] have performed de Haas van Alphen (dHvA) experiments and found that the obtained branches are essentially the same as those of $\mathrm{LaRhIn_5}$, suggesting that the Ce 4*f* electrons in $\mathrm{CeRhIn_5}$ are localized. The localized nature of Ce 4*f* electrons in this compound is also evident from the dHvA experiments on $\mathrm{Ce}_x\mathrm{La}_{1-x}\mathrm{RhIn_5}$ by Los Alamos group[9] and the optical conductivity measurements.[10] For the superconducting $\mathrm{CeIrIn_5}$, on the other hand, there has been a controversy between the localized and delocalized pictures for the Ce 4*f* electrons. A nuclear quadrupole resonance (NQR) study has suggested that the Ce 4*f* electrons in this compound are much more itinerant than known other Ce HF based compounds.[11] dHvA measurements on this compound have also suggested that the observed branches are well explained by the itinerant 4*f*-electron model.[8,12] In addition, Harrison *et al.*[9] have compared the dHvA branches of $\mathrm{CeIrIn_5}$ and $\mathrm{LaIrIn_5}$, and found that the expansion of Fermi surface (FS) volume on going from $\mathrm{LaIrIn_5}$ to $\mathrm{CeIrIn_5}$. This implies the formation of a quasiparticle band in $\mathrm{CeIrIn_5}$. All these results suggest that 4*f* electrons in $\mathrm{CeIrIn_5}$ have itinerant character. On the other hand, photoemission studies have been made for this compound, and the localized character of *f*-electrons has been observed. In a previous paper, we have studied $\mathrm{Ce}T\mathrm{In_5}$ ($T=\mathrm{Rh,Ir}$) by angle-resolved photoemission spectroscopy (ARPES) using low energy photon ($h\nu=21.2$ eV) and Ce 3*d*-4*f* resonant photoemission spectroscopy (RPES).[13] The RPES spectra of both compounds were understood within the framework of the single impurity Anderson model (SIAM), suggesting that the nearly localized nature of Ce 4*f*-states in these compounds. Since we could not observe delocalized spectral features of the Ce 4*f* states in $\mathrm{CeIrIn_5}$, we concluded that the Ce 4*f* electrons in $\mathrm{CeRhIn_5}$ and $\mathrm{CeIrIn_5}$ were nearly localized, but they were slightly delocalized in $\mathrm{CeIrIn_5}$ compared to $\mathrm{CeRhIn_5}$. Because the spectral difference between $\mathrm{CeIrIn_5}$ and $\mathrm{CeRhIn_5}$ was very small, the origin of the large difference in their ground-state properties was not well understood from the photoemission spectra.

To probe the localized and/or delocalized nature of f-states, the ARPES experiment is the most powerful experimental method since the observation of dispersive f-bands is the direct evidence for the itinerant character of f states. There have been some attempts to detect the energy dispersions of the Ce $4f$ band by ARPES. Arko *et al.*[14] have reported an ARPES study of $CeSb_2$, and observed the intensity modulation of the Ce $4f$ peak near the Fermi level (E_F) as a function of momentum. However, they measured the spectra at $h\nu=60$ eV, where contributions not only from Ce f, but also Ce d and Sb s,p states were substantial, and therefore, it was difficult to extract the Ce $4f$ contribution unambiguously.[15] To distinguish the Ce $4f$ contribution from the other states, the RPES technique has been utilized. Denlinger *et al.*[16] measured angle-resoled RPES (AR-RPES) spectra of $CeRu_2Si_2$, and found the amplitude variation of the Ce $4f$ derived component near E_F. However, they could not observed the experimental energy dispersion of the Ce $4f$ derived component, probably due to the weakness of the dispersion compared to the energy resolution ($\Delta E \sim 60$ meV).

In the present paper, we report on a high-resolution (~ 30 meV) AR-RPES study of $CeIrIn_5$ in the $4d$-$4f$ excitation region to study the dispersion of the $4f$ band, thereby clarifying the f-electron states in $CeIrIn_5$. The improved energy resolution has enabled us to detect the energy dispersion of the Ce $4f$ states in $CeIrIn_5$ as we shall discuss below.

II. EXPERIMENT

The experiment was performed at the linear undulator beamline (BL-1) of a 700 MeV electron-storage ring (HiSOR) in Hiroshima Synchrotron Radiation Center (HSRC), Hiroshima University. The photon energies of $h\nu \sim 122$ and 112 eV were used for $4d$-$4f$ on- and off-resonance, respectively. The energy resolution of 18 and 30 meV were used for the angle-integrated and angle-resolved photoemission experiments, respectively. Single crystals of $CeRhIn_5$ and $CeIrIn_5$ were grown by the self-flux method described in Ref. 4. Clean surfaces were obtained by *in situ* cleaving the samples parallel to the a-b plane. The sample temperature was kept at 10 K during the course of the measurements. The position of E_F was carefully determined using evaporated gold film.

III. RESULTS AND DISCUSSION

Figure 1(a) shows angle-integrated $4d$-$4f$ RPES spectra of $CeRhIn_5$ and $CeIrIn_5$. The spectra show an intense peak at around 2.5 eV below E_F corresponding to the f^0 final state, and weak structures near E_F corresponding to the f^1 final states. The $4f^1$ final-state feature shows peak at ~ 280 meV arising from the spin-orbit interaction. These spectral features have been observed in other Ce-based compounds and understood within the framework of the SIAM as in the case of $3d$-$4f$ RPES study for these compounds.[13] The intensity of the f^1 final-state signal near E_F is much weaker than that of the f^0 final state peak located at ~ 2.5 eV. It has been pointed out that the Ce $4f$ electrons in the surface layers are more localized than in the bulk electronic structure.[17] Sekiyama *et al.*[18] have demonstrated that the surface-sensitive $4d$-$4f$ RPES spectra of HF Ce-based compounds are significantly different from the bulk-sensitive $3d$-$4f$ RPES spectra. In fact, the f^0 peak intensity is much stronger in the present $4d$-$4f$ RPES spectra than in the previous $3d$-$4f$ RPES spectra,[13] indicating that Ce $4f$ electrons in the surface layers are more localized than in the bulk.

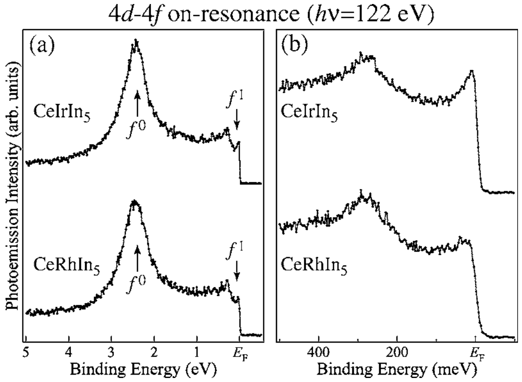

FIG. 1. $4d$-$4f$ resonant photoemission spectra of $CeIrIn_5$ and $CeRhIn_5$ ($h\nu=122$ eV): (a) wide range and (b) near E_F part.

The weakness of the $4f^1$ signal compared to other itinerant f-electron compounds in the present spectra are consistent with the nearly localized character of $4f$ electrons in the bulk. To address the question of whether the present $4d$-$4f$ RPES can be used for probing the bulk electronic structure of these compounds, we note that because the Ce $4f$ electrons in these compounds are nearly localized, the Ce $4f$ electrons in the surface layer must be almost completely localized. This means that the surface component of the Ce $4f$ states should have very small spectral weight in the delocalized (f^1) part of the spectra. Therefore, we assume that the f^1 peak in these spectra can be mostly considered largely the bulk origin. Figure 1(b) shows the near E_F part of the RPES spectra. The spectra consist of the $f^1_{5/2}$ final state peak located at E_F (in fact, the tail of the Kondo peak located above E_F) and the $f^1_{7/2}$ peak located ~ 280 meV below E_F. The clearest difference between the spectra of $CeIrIn_5$ and $CeRhIn_5$ is that the spectral intensity at E_F, the Ce $4f_{5/2}$ peak or the tail of the Kondo peak, is higher in $CeIrIn_5$ than in $CeRhIn_5$. Because its intensity is higher for more strongly hybridized compounds, the absence of the sharp peak structure in $CeRhIn_5$ suggests that Ce $4f$ electrons in this compound is almost completely localized in $CeRhIn_5$ while those in $CeIrIn_5$ have some itinerant character. Therefore, there should be only minor contributions from Ce $4f$ state at the vicinity of E_F in the ARPES spectra of $CeRhIn_5$. These results are consistent with the results of optical conductivity measurements where the hybridization gap is observed in $CeIrIn_5$ but not in $CeRhIn_5$.[10]

We have further performed AR-RPES experiments on $CeIrIn_5$ to reveal its momentum dependence of the Ce $4f$ spectrum. Figure 2 shows the angle-resolved $4d$-$4f$ RPES spectra of $CeIrIn_5$. The left panel shows the spectra in a wide energy range, and the right panel does details of the spectra near E_F. The emission angle relative to the surface normal is also presented. It can be seen that the spectra show a clear

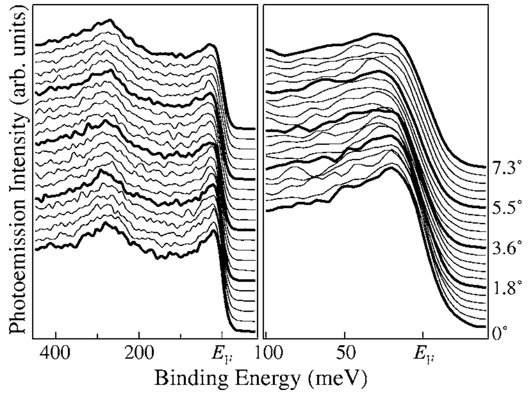

FIG. 2. On-resonance ARPES spectra near E_F, taken at $h\nu=122$ eV.

angular dependence. In particular, the intensity and shape of $4f_{5/2}$ peak located just below E_F show a clear momentum dependence. Its intensity is largest around $\theta=0°$ and becomes weaker when the emission angle is increased. Moreover, the peak shows a small but distinct energy dispersion. The peak moves toward the deeper binding energy side as it approaches $\theta\sim 4°$, and it again moves toward E_F as the emission angle is further increased. These changes are observed only in the $4f_{5/2}$ final state peak and are not seen in the $4f_{7/2}$ final state peak. This suggests that the changes are not caused by matrix-element effects but by band dispersion.

Here, we consider the momentum position of the present ARPES cut in k space. The momentum of photoelectron parallel and perpendicular to the surface are given by

$$k_{\parallel} = \sqrt{\frac{2m}{\hbar^2}E_{\mathrm{kin}}}\sin\theta, \tag{1}$$

$$k_{\perp} = \sqrt{\frac{2m}{\hbar^2}(E_{\mathrm{kin}}\cos^2\theta + V_0)}, \tag{2}$$

where E_{kin} is the kinetic energy of the photoelectron, V_0 is the inner potential, and θ is the emission angle of the photoelectron relative to the surface normal. To determine the $k_{\perp}$ value, we need to determine the inner potential V_0. In the previous ARPES experiments on CeIrIn$_5$,[19] the V_0 is estimated to be 15 eV. However, this value is somewhat larger than those of other HF compounds, such as CeRu$_2$Si$_2$ (12 eV)[16] and CeNi$_2$Ge$_2$ (13.8 eV).[20] We assume that the inner potential is 12 eV, which can be regarded as a typical value for HF materials. Figure 3(b) shows the momentum position of the APRES cut as a function of $k_{\perp}$ and $k_{\parallel}$ for on-resonance ARPES spectra. Because the escape depth of

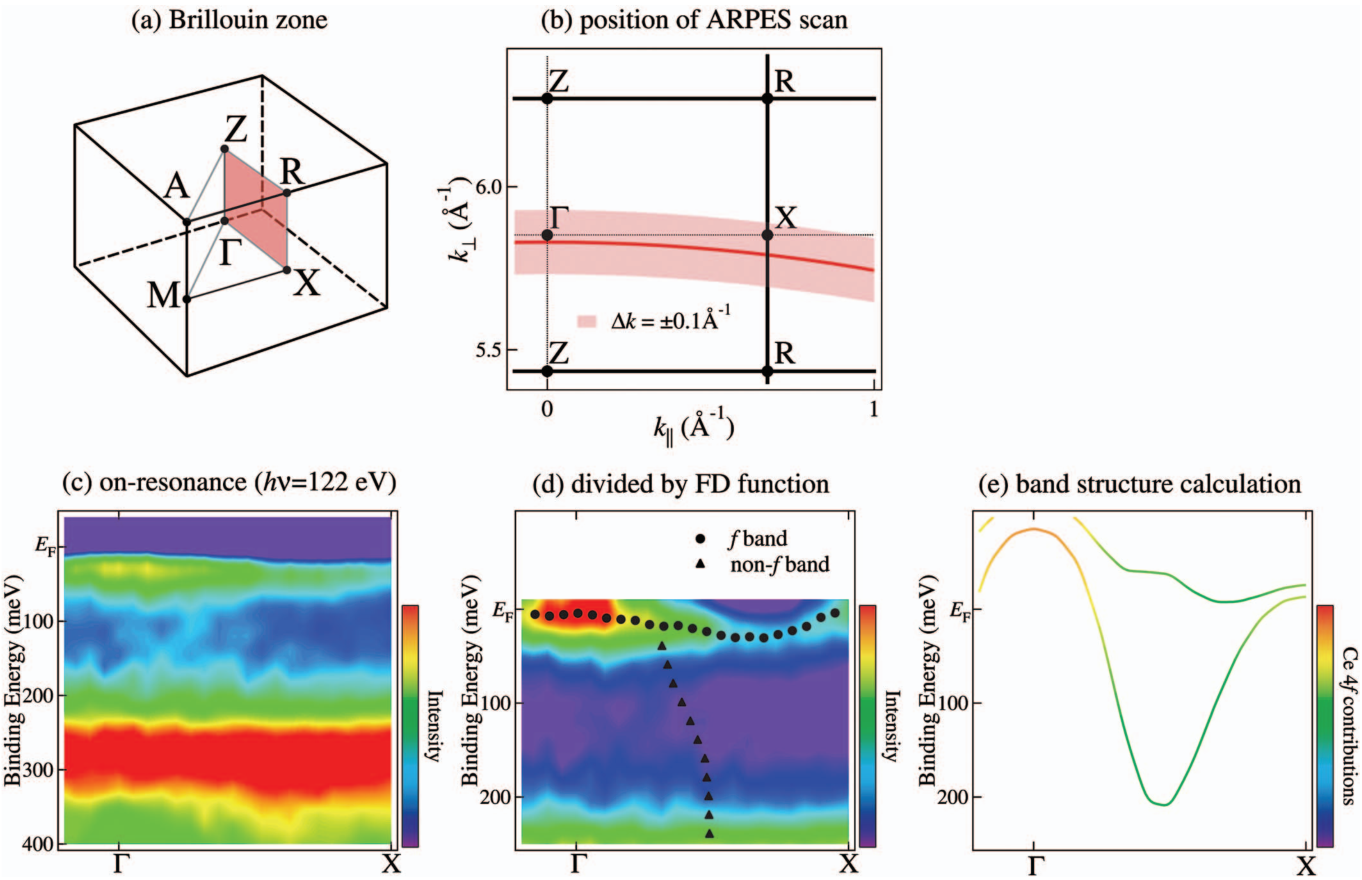

FIG. 3. (Color) ARPES spectra of CeIrIn$_5$: (a) Brillouin zone, (b) momentum position of ARPES cuts in k space. (c) on-resonance ARPES spectra, (d) on-resonance divided by the Fermi-Dirac function, and (e)Result of the band structure calculation along the Γ- X direction, together with the positions of peaks in the on-resonance spectra and the He I spectra.

the photoelectron in the present experiment is about 5–10 Å, the momentum broadening for the $k_\perp$ direction should be $\Delta k_\perp \sim 0.1-0.2$ Å^{-1}. This broadening width ($\Delta k_\perp = \pm 0.1$ Å^{-1}) is also shown as shaded area in Fig. 3(b). Since the contributions from the high symmetry lines are dominant in ARPES spectra, we assume that the present AR-RPES spectra probe the high-symmetry Γ-X line of the eighth Brillouin zone. The spectrum measured at $\theta=0°$ corresponds to the Γ point, and that measured at $\theta=7°$ does the X point.

Figure 3(c) shows the density plot of ARPES spectra. The momentum dependences of the ARPES spectra are summarized as follows. The $4f_{5/2}$ peak located at around E_F show small energy dispersions. On the other hand, for the $4f_{7/2}$ peak, while their positions are almost unchanged, their widths have some momentum dependences. Therefore, they have very different behaviors, suggesting again that these changes are not due to the matrix element effects. To see the angular dependent behavior of the peak near E_F more clearly, we have divided the AR-RPES spectra by the Fermi-Dirac function convoluted with a Gaussian function representing the instrumental resolution (30 meV). This method has been utilized to remove the effect of the Fermi cutoff and to reveal spectral feature around E_F. Figure 3(d) shows the density plot of the ARPES spectra in the E-k plane divided by the Fermi-Dirac function. In this plot, we have marked the positions of the peak maximum as solid circles as a function of binding energy and momentum. In the near E_F part of the spectra, a dispersive feature is clearly recognized. The size of this energy dispersion is estimated to be about 30 meV. Mena *et al.*[10] have measured the optical conductivity of Ce*M*In$_5$ (M=Rh, Ir, and Co) and observed a hybridization gap of $\Delta=50-70$ meV for CeIrIn$_5$. This energy scale is of the same order as the energy dispersion observed in the ARPES spectra and consistent with our observation of the narrow hybridized band. This behavior of narrow *f*-band can be explained within the framework of the renormalized *f*-band model,[21] according to which the renormalized *f* band is mixed with the strongly dispersive non-*f* band, and the weakly dispersive hybridized bands are formed near E_F. In Fig. 3(d), we have also plotted the peak positions of ARPES spectra measured at He I ($h\nu=21.2$ eV)[13] as filled triangles, which can be regarded as contributions mostly from In 4*p* bands. Although these spectra do not correspond to the same momentum position of the ARPES scan measured by $h\nu=122$ eV for $k_\perp$ direction, it is known that ARPES spectra with He I represent the contributions mostly from the high symmetry lines.[22] Therefore, we take these ARPES spectra as representing the contributions from the non-*f* bands in high symmetry Γ-X line. The high intensity part in the quasiparticle bands is distributed inside of Fermi surface crossing points of the non-*f* bands, suggesting that the features can be understood within the framework of renormalized band theory. Therefore, in the present case, the narrow 4*f* quasiparticle band is originated from the hybridization between the renormalized Ce 4*f* states and the In 4*p* states.

Figure 3(e) shows energy band dispersions calculated using local density approximation (LDA) for the Γ-X line. The degree of the Ce 4*f* contribution is also indicated by the color of each line. At first glance, the calculated overall band dispersion seems very different from the observed dispersions of quasiparticle bands. However, the shape of the Ce 4*f* dispersion seems to correspond to the band around E_F but being extremely narrowed. Although the Fermi surface crossing points are different, this Ce 4*f* dispersion can be interpreted as the band around E_F being extremely narrowed. The calculated energy dispersion of this band is about 300 meV, which is about ten times larger than the experimentally observed Ce 4*f* dispersion, and the size of Fermi surface is smaller in the experiment than in the calculation. The calculated band mass is estimated to be $m_b=0.64m_0$. In the dHvA study of CeIrIn$_5$, a small spherical FS having $m^* = 6.3m_0$ has observed, and it was suggested that this branch is originated from this calculated band.[12] This electron mass is ten times heavier than that of the calculation, consistent with our observation of the narrowing of the Ce 4*f* band dispersion. In the dHvA experiments, the size of the observed Fermi surface is about 40% of the calculation. In the present experiment, although it is difficult to estimate the exact size of the Fermi surface, its size seems smaller than the calculation, and the tendency is consistent with the dHvA data. According to the calculation, this Fermi surface has very large contributions from Ce 4*f* states, and they are strongly renormalized. Therefore, the LDA cannot describe the behavior of this band well.

Accordingly, we have found that although the global features of the angle-integrated spectra of CeIrIn$_5$ can be understood within the framework of the impurity model,[13] the contribution from the small itinerant component is observed as the dispersive Ce 4*f* band near E_F and explains the observation of the heavy quasiparticle bands in CeIrIn$_5$ in the dHvA experiments.[12] This dual nature of *f* states observed in CeIrIn$_5$ is originated from the very small but substantial hybridization to produce the quasiparticle band. When the hybridization is smaller than CeIrIn$_5$, the quasiparticle band will be vanished as is observed in CeRhIn$_5$. On the other hand, if the hybridization is larger than CeIrIn$_5$, the localized nature will disappear. In fact, the RPES spectra of CeCoIn$_5$,[23] which have the largest hybridization strength among the series of compounds, is very different from those of CeIrIn$_5$ as well as from CeRhIn$_5$, suggesting that *f* states are not no longer "nearly localized" in more strongly hybridized compounds. Therefore, the strength of the hybridization in CeIrIn$_5$ is very close to the critical value for the formation of the quasiparticle band and is the origin of the dual nature of *f* states. Pagliuso *et al.*[7] have inferred that the "band like" nature of the *f* states is important for the coexistence of magnetism and superconductivity for Ce-based compounds. However, we have found the nearly localized but slightly delocalized nature of 4*f* states even for CeIrIn$_5$, which is a nonmagnetic superconductor. Therefore, the nearly localized nature with a small itinerant component may be essential for such coexistence of magnetism and superconductivity in Ce-based compounds located near QCP.

IV. CONCLUSION

In conclusion, we have found that although the Ce 4*f* electrons in CeIrIn$_5$ are nearly localized, there exists the small itinerant contribution observed as the quasiparticle band near E_F. This quasiparticle band has an energy dispersion of ~30 meV, consistent with the results of dHvA and optical conductivity measurements, and is responsible for the superconductivity. This dual nature of f states is originated from the very small but substantial hybridization to produce the quasiparticle band in $CeIrIn_5$.

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