The 4f-Hybridization Strength in CeₘMₙIn₃ₘ₊₂ₙ Heavy-Fermion Compounds Studied by Angle-Resolved Photoemission Spectroscopy

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We systematically investigate the nature of Ce 4f electrons in structurally layered heavy-fermion compounds CeₘMₙIn₃ₘ₊₂ₙ (with M = Co, Rh, Ir, and Pt, m = 1, 2, n = 0–2), at low temperature using on-resonance angle-resolved photoemission spectroscopy. Three heavy quasiparticle bands f¹, f₂/₃ and f₁/₂, are observed in all compounds, whereas their intensities and energy locations vary greatly with materials. The strong f⁰ intensity ratio is more suitable to reflect the 4f-state hybridization strength. DOI: 10.1088/0256-307X/38/10/107402

The heavy-fermion (HF) system is an excellent platform for studying the behavior of correlated electrons in unconventional superconductors, quantum criticality and non-Fermi liquids, antiferromagnetism. Previous studies on HF superconductors reveal that both dimensionality and hybridization are closely related to superconductivity. Tuning dimensionality and hybridization is the key to revealing the microscopic mechanism of unconventional superconductivity in heavy fermions.

It is found that the lattice parameters ratio c/a of CeₘMₙIn₅ (M = Co, Rh, Ir) has a roughly linear relationship with its superconducting transition temperature Tc.[⁸] Pu-based 115 materials Tc is also linear in c/a.[⁹] Uniaxial pressure measurement on CeIrIn₃ revealed that Tc changed linearly with both a-axis and c-axis pressure.[¹⁰] The c/a ratio may be the control parameter for the scaling of the superconducting dome width and Tc max. The value of Tc in magnetically mediated superconductors is believed to be dependent on dimensionality.[¹¹–¹⁶] Superconducting properties can be enhanced by lowering dimensionality. It is suggested that the difference in hybridization sets the overall temperature scale for each family, but dimensionality governs the behavior within each family.

To explore the character of f electrons is essential to understanding the complex physics in HF compounds. Notably, f-electrons participate in the formation of Fermi surface through hybridization with conduction electrons. The localized or itinerant properties of f-electrons, i.e., hybridization between conduction and f electrons (c–f), will significantly affect the band structure near the Fermi energy EF. After c–f electron hybridization, the complex itinerant heavy electron state is formed, which leads to the delocalization of f electron. Therefore, f electron posses...
the dual nature of local and itinerant even at very low temperatures, which makes the system complex and difficult to understand.\cite{11,12}

HF compounds $\text{Ce}_nM_n\text{In}_{3m+2n}$ ($M = \text{Co, Rh, Ir, and Pt}$) are very suitable for investigating dimensionality and hybridization, and a lot of studies have been carried out on such a system. Theoretical calculations have only one nearest and four next-nearest neighbors hybridization strength of Ce.\cite{29}

$\text{Pm}$ This family and crystallizes in the infinite-layer (cubic)\cite{29} phase, which is the distance between Ce and its nearest neighbors $M$. The table also gives the value of $c/a$, which is often used to describe the structural dimension.

$\text{CeIn}_3$ is a parent and fundamental unit of this family and crystallizes in the infinite-layer (cubic, 3D) structure ($\text{Pm}\overline{3}m$).\cite{20} $\text{CeIn}_3$ is an antiferromagnet at ambient pressure with predominantly localized moments, and Sommerfeld coefficient $\gamma$ is 120 mJ mol$^{-1}$ K$^{-2}$.\cite{20} It undergoes a superconducting transition at $T_c = 0.2$ K under pressure of 2.6 GPa.\cite{31}

In previous ARPES experiments, a weak heavy quasiparticle band with an energy dispersion of 4 meV was observed, indicating a weak $c-f$ hybridization.\cite{17}

$\text{Ce}_4\text{IrIn}_8$ is a tetragonal compound. The observation of a spin-glass state,\cite{41} a relatively high Sommerfeld coefficient ($\sim 700$ mJ mol$^{-1}$ K$^{-2}$),\cite{30} and the absence of long-range magnetic order indicate that the Ce 4f electrons in $\text{Ce}_4\text{IrIn}_8$ have itinerant character. ARPES is observed with a flat band near the $E_F$ around the $\Gamma$ point, which shows no large difference in Fermi momentum.\cite{18}

$\text{Ce}_4\text{RhIn}_8$ has a tetragonal crystal structure is an antiferromagnetic superconductor with two magnetic transitions at $T_N = 2.8$ K and $T_m = 1.65$ K.\cite{32,42} ARPES measurements suggested that the Ce 4f electrons are essentially localized.\cite{26,43,44}

$\text{CeCoIn}_5$ is a well-studied compound. Its low energy behavior is similar to that of the underdoped cuprates, with a relatively high $T_c \approx 2.3$ K at ambient pressure. In the beginning, $f$ electrons were considered to be itinerant at low temperature because the experimental results of optical conductivity\cite{45} de Haas-van Alphen (dHvA)\cite{46,47} and scanning tunneling microscope (STM)\cite{48,49} were in good agreement with the theoretical calculation based on fully itinerant $f$ electrons.\cite{7,46} Earlier ARPES experiments suggested that $f$ electrons were itinerant up to 105 K,\cite{22} while subsequently there appeared evidence to support the dominance of localized $f$ electrons at low temperature,\cite{50,51} which showed an itinerant nature due to partial hybridizations.\cite{52} Recent ARPES experiments suggested that the localized-to-itinerant transition occurs at surprisingly high temperatures, and $f$ electrons are largely localized even at the lowest temperature.\cite{10} $\text{CeIrIn}_5$ is a superconductor with $T_c = 0.4$ K at

$\text{f}_{3/2}$.\cite{13,23} The $f_{3/2}^1$ peak observed near $E_F$ is actually the tail of the Kondo resonance peak, and $f_{3/2}^1$ is the satellite peak. According to the single impurity Anderson model (SIAM),\cite{24} generally, the more localized the Ce 4f is, the stronger the $f^0$ peak is, and the weaker the $c-f$ hybridization is.\cite{25,26}

In this study, we carried out high-resolution on-resonance ARPES experiments on HF compounds $\text{Ce}_nM_n\text{In}_{3m+2n}$, with $n = 1, 2$, and $\text{Pt}$, and discuss the localization or itinerancy of Ce 4f in these compounds and the relationship between the dimensionality and the $c-f$ hybridization strength. It is found that compared with the $f^{1-3}$ intensity ratio, the $f_{3/2}^1$ intensity ratio can better reflect $c-f$ hybridization strength.

### Table 1. Physical properties of cerium compounds studied in the present study. $T_c$: Néel temperature.

| Compounds          | Structure | $\gamma$ [mJ/(mol·Ce·K$^2$)] | Order | $T_c$ (K) | $T_N$ (K) | $a$ (Å) | $c$ (Å) | $c/a$ | $d_{Ce-M}$ (Å) |
|--------------------|-----------|-------------------------------|-------|-----------|-----------|---------|---------|------|----------------|
| $\text{CeIn}_3$   | Cubic     | 120\cite{27}                  | AF    | 0.2       | 10.3\cite{31} | 4.6876  | 4.6876  | 1     |               |
| $\text{Ce}_4\text{IrIn}_8$ | Tetragonal | 700\cite{30}                 | AF    | 2.0\cite{32} | 12.195\cite{31} | 2.600   | 3.73    |       |               |
| $\text{Ce}_4\text{RhIn}_8$ | Tetragonal | 400\cite{30}                |       |           |           | 4.667   | 12.247  | 2.624 | 3.74          |
| $\text{CeCoIn}_5$ | Tetragonal | 290\cite{33}                 | AF    | 2.2\cite{33} | 7.540\cite{34}  | 1.639   | 3.77    |       |               |
| $\text{CeIrIn}_5$ | Tetragonal | 720\cite{35}                 | AF    |           |           | 4.668   | 7.515   | 1.610 | 3.76          |
| $\text{CeRhIn}_5$ | Tetragonal | $\sim 400$\cite{30}         | AF    | 0.1\cite{36} | 3.8\cite{37}  | 4.652   | 7.542   | 1.621 | 3.77          |
| $\text{CePt}_2\text{In}_7$ | Tetragonal | 328\cite{38}                | AF    | 2.1\cite{39} | 21.625\cite{40} | 4.692   | 4.98    |       |               |

### Summary of Studied Compounds.

The HF compounds $\text{Ce}_nM_n\text{In}_{3m+2n}$ possess a relatively higher $T_c$ than other HF materials. Its crystal structure is composed of $m$-layers of $\text{CeIn}_3$ separated by $n$-layers of $M\text{In}_2$ along the $c$ axis of the tetragonal unit cell. This family has rich phase diagrams and is a good platform to investigate the effect of dimensionality, hybridization, magnetism, superconductivity, etc. The basic physical properties of HF compounds investigated in the present study are listed in Table 1. From top to bottom, $\text{CeIn}_3$, $\text{Ce}_4\text{MIn}_8$, $\text{Ce}_4\text{IrIn}_3$, and $\text{Ce}_4\text{PtIn}_7$, the electronic structure becomes more two-dimensional, and the superconducting transition temperature $T_c$ is enhanced. The last column $d$ is the $Ce-M$ bond distance, which is the distance between Ce and its nearest neighbors $M$ atoms. It is worth noting that $\text{Ce}_4\text{MIn}_3$ has only one nearest and four next-nearest $M$ atoms, while $\text{Ce}_4\text{IrIn}_3$ has two nearest and eight next-nearest neighbor $M$ atoms. In other words, the hybridization strength of $\text{Ce}_4\text{MIn}_3$ will be significantly stronger than that of $\text{Ce}_4\text{IrIn}_3$. The table also gives the value of $c/a$, which is often used to describe the structural dimension.

$\text{CeIn}_3$ is the parent and fundamental unit of this family and crystallizes in the infinite-layer (cubic, 3D) structure ($\text{Pm}\overline{3}m$).\cite{20} $\text{CeIn}_3$ is an antiferromagnet at ambient pressure with predominantly localized moments, and Sommerfeld coefficient $\gamma$ is 120 mJ mol$^{-1}$ K$^{-2}$.\cite{20} In previous ARPES experiments, a weak heavy quasiparticle band with an energy dispersion of 4 meV was observed, indicating a weak $c-f$ hybridization.\cite{17} In previous ARPES experiments, a weak heavy quasiparticle band with an energy dispersion of 4 meV was observed, indicating a weak $c-f$ hybridization.\cite{17}
ambient pressure.[35] Early dHvA experiments suggested that 4f electron delocalizes and involves forming Fermi surface at low temperature, which means that 4f electrons have itinerant character.[46,47,52] Earlier ARPES experiments suggested that 4f electrons were nearly localized.[25] Later, on-resonance ARPES experiments have observed heavy quasiparticle peaks, suggesting that although 4f electrons are mainly localized, a small itinerant component is responsible for superconductivity.[53] Recently, ARPES measurements found that the localized 4f electrons evolve into the HF state starting from a temperature much higher than the coherence temperature.[20]

CeRhIn₅, unlike CeCoIn₅ and CeIrIn₅, is an antiferromagnetic compound with Néel temperature $T_N = 3.8$ K at ambient pressure.[37] In contrast to CeCoIn₅ and CeIrIn₅, dHvA experiments suggested that the Ce 4f electrons are localized in CeRhIn₅.[16,47,52] The theoretical calculation also gives the picture of localized f states.[54] Different ARPES research groups found seemingly conflicting results. Some believed that 4f electrons were nearly localized in the paramagnetic state,[25] while others suggested that the 4f electrons participate in band formation.[55] Recently, ARPES experiments found band-dependent $c$–$f$ hybridization.[21]

CePt₂Ir₇ with body-centered tetragonal crystal structure is an antiferromagnetic superconductor with $T_c = 2.1$ K near 3.5 GPa.[59] Quantum oscillation[56] and ARPES[13,23,57] measurements suggested that partially 4f-electrons contribute to FS formation.

Experimental Details. High-resolution ARPES experiments were performed on SIS X09LA beamline at the Swiss Light Source, using a VG-SCIENTA R4000 photoelectron spectrometer. The characteristics of Ce 4f electrons were obtained by on-resonance ARPES at a low temperature of 10 K, which is a practical approach to explore the 4f electron states. All compounds were measured with 122 eV photons, except CeIrIn₅ and CePt₂Ir₇ compounds measured with 123 eV photons. The energy and momentum resolution was set to ~25–35 meV and 0.2°, respectively. All the samples were single crystals, and cleaved in situ, and measured in an ultra-high vacuum. All measurements were carried out under very similar conditions. Since the thermocouple does not directly contact the sample, the actual temperature of the sample is slightly higher than the measured temperature. Thus, all samples, including CeIn₃, are in the paramagnetic phase.

Results and Discussion. Figures 1(a1)–1(a7) show the spectra taken from freshly cleaved HF compounds $\text{Ce}_m\text{M}_n\text{In}_{m+n+2n}$. Because ARPES spectra is a surface sensitive technique with 122/123 eV photon energies, this results in a large Ce 4f spectra weight distribution at deeper binding energies. Figures 1(b1)–1(b7) display the dispersion reproduced by second-derivative with respect to energy corresponding to (a1)–(a7), respectively.

[Fig. 1. Photoemission images for HF compounds $\text{Ce}_m\text{M}_n\text{In}_{m+n+2n}$. (a1)–(a7) On-resonance ARPES spectra for CeIn₃, Ce₂IrIn₈, Ce₂RhIn₈, CeCoIn₅, CeIrIn₅, CeRhIn₅, and CePt₂Ir₇, respectively. (b1)–(b7) The second-derivative images with respect to energy corresponding to (a1)–(a7), respectively.]
corresponding to $f^1_{3/2}$ and $f^1_{1/2}$ final states.\textsuperscript{[13]–[21]} The intensities of $f^0_{3/2}$ and $f^0_{1/2}$ final states also vary significantly with materials. The $f^0_{3/2}$ final state, the tails of Kondo resonant peaks, can be easily distinguished from the raw images of CeIn$_3$ and CeMnIn$_5$, while those of Ce$_2$IrIn$_5$, Ce$_2$RhIn$_5$ and CePt$_2$In$_7$ show only limited spectral weight. The $f^1_{1/2}$ final state, i.e., the spin-orbit coupling sideband of the $f^1_{3/2}$ state, located at around $-300$ meV, can be observed in the raw data of all samples. As seen in Fig. 1(a4), the $f^1$ state of CeCoIn$_5$ is heavily overlapped with the Co 3$d$ state, giving the fact that the photoionization cross-section of the Co 3$d$ level is of one order higher than those of the Rh 4$d$ and Ir 5$d$ at the Ce 4$d$–4$f$ excitation threshold.

Figure 2 displays the energy distribution curves (EDCs) of Ce$_m$M$_n$In$_{3m+2n}$, which is integrated over the momentum range shown in Fig. 1. It is found that our ARPES results are consistent with the previous results.\textsuperscript{[13,17–21,26]} The $f^0$, $f^1_{3/2}$ and $f^1_{1/2}$ final states mentioned above were observed in EDCs. These spectral features were widely observed in Ce-based HFs. Several notable features can be seen from the figures. The relative intensities of the three final states change significantly with the material. Only in CeIn$_3$ and CeIrIn$_5$ can sharp peaks of $f^1_{3/2}$ and $f^1_{1/2}$ be observed. For others, the intensity of $f^1_{3/2}$ is considerably weaker than that of $f^1_{1/2}$. The $f^0$ final state intensity is very strong for the whole family members, except CeCoIn$_5$, suggesting that localized electrons dominate the Ce 4$f$ states. The $f^0$ intensities of CePt$_2$In$_7$ and CeCoIn$_5$ were the strongest and the weakest in the family, respectively. According to the SIAM, they may correspond to the most localized (weakest hybridized) and the most itinerant (most hybridized), respectively. The deduction for CePt$_2$In$_7$ seems to be consistent with the fact that it has the largest $d$ value in this family.\textsuperscript{[30]} However, the inference for CeCoIn$_5$ is inconsistent with previous experimental and calculation results, which suggested that the $c$–$f$ hybridization weakness in CeRhIn$_5$, CeCoIn$_5$, and CeIrIn$_5$.\textsuperscript{[7,21]} It seems thus not a good idea to use the $f^0$ strength alone to reflect the hybridization strength and itinerancy of $f$ electrons. A more suitable parameter is needed to quantitatively reflect the hybridization strength.

The $4d$–$4f$ on-resonance spectra in Ce-based HF compounds can be well understood by the SIAM\textsuperscript{[24–26]} which considers that the ground state is a linear combination of the $f^0$ and $f^1_{3/2}$ states. Previous experiments suggested that the stronger the $c$–$f$ hybridization, the stronger the $f^1$ peaks.\textsuperscript{[25,26]} To compare hybridization strength quantitatively, the $f^1$–$f^0$ intensity ratios $[I(f^1_{1/2})+I(f^1_{3/2})]/I(f^0)$ were calculated, which has been considered to reflect the $c$–$f$ hybridization strength.\textsuperscript{[25]} The angle-integrated PES spectra were well fitted with multiple Lorentzian peaks.\textsuperscript{[16]} Figures 3(a) and 3(b) display the typical fitting results of CeIn$_3$ and CeCoIn$_5$ spectra. The filled shadow curves represent the heavy $f$ response. As evidenced by red, yellow, and cyan dot-dashed lines, the spectra include a large contribution of conduction

![Figure 2](image_url)

**Fig. 2.** Comparison of angle-integrated PES spectra of CeIn$_3$, Ce$_2$IrIn$_5$, Ce$_2$RhIn$_5$, CeCoIn$_5$, CeIrIn$_5$, CeRhIn$_5$, and CePt$_2$In$_7$.

![Figure 3](image_url)

**Fig. 3.** (a) and (b) Angle-integrated PES spectra (open circles) and the corresponding fitting results (solid lines and dot-dashed lines). Filled green, purple, and blue profiles represent the fitting results of $f^0$, $f^1_{3/2}$, and $f^1_{1/2}$ final states, respectively. Other dot-dashed lines originate from conduction bands and background. (c) The $f^1$–$f^0$ and $f^1_{3/2}$–$f^0$ intensity ratios of $\frac{[I(f^1_{1/2})+I(f^1_{3/2})]}{I(f^0)}$.
peak position (eV)

CePt2In7CeRhIn5CeIrIn5CeCoIn5Ce2RhIn8Ce2IrIn8CeIn3
-0.24
-0.28
-0.32
-2.0
-2.2
-2.4
-2.6

bands.

Figure 3(c) display the calculated $f^1-f^0$ intensity ratio, together with $f_{f/2}^1-f^0$ intensity ratio $I(f_{f/2}^1)/I(f^0)$. It can be seen that both intensity ratios $f^1-f^0$ and $f_{f/2}^1-f^0$, vary greatly with the materials. From CeIn3 to CePt2In7, the ratios do not change monotonically with c/a or d. Interestingly, CeIn3, which has a 3D structure, and CePt2In7, which is towards the two-dimension limit in the Ce$_m$M$_n$In$_{3m+2n}$ family, both have very low intensity ratio and possess the weakest hybridization/itinerant. The ratios of four antiferromagnets (CeIn3, Ce$_2$RhIn5, CeRhIn5, and CePt2In7) are very small. The strong localized nature of f electron may be responsible for the antiferromagnetic transition at low temperatures. The $f^1-f^0$ value of CeMIn$_5$ is the largest of this family, suggesting that CeMIn$_5$ may have the strongest c–f hybridization strength in the Ce$_m$M$_n$In$_{3m+2n}$ family.

![Fig. 4. Energy positions of $f^0$ and $f_{f/2}^1$ final states.](image)

However, in the subfamily CeMIn$_5$, the $f^1-f^0$ intensity ratio decreases in the order of CeCoIn5, CeRhIn5, and CeRhIn5. According to the SIAM, the c–f hybridization weakens in the same order, which is inconsistent with the previous experimental and theoretical results. Noticeably, the $f_{f/2}^1-f^0$ intensity ratio is in line with previous results, which seems to reflect the hybridization strength of CeMIn$_5$ better. Both intensity ratios could thus be employed. Significantly, our results suggest that the $f_{f/2}^1-f^0$ intensity ratio is more suitable than the $f^1-f^0$ to quantify the hybridization strength. In fact, this is an intuitive result. Previous studies have shown that compared with the $f^1_{f/2}$ state, the $f_{f/2}^1$ state is not sensitive to temperature. This means that the $f^1_{f/2}$ state is not a good parameter for determining Kondo temperature $T_K$, and the $f_{f/2}^1$ state is a more suitable parameter. With the development of ARPES, we may even further consider using the low-lying crystal electric field excitation of $f_{f/2}^1$ state to quantitatively determine $T_K$.

We note that the change of atomic distance $d$ in subfamily CeMIn$_5$ is very small, while the hybridization strength changes significantly. Similarly, the change of c/a in CeMIn$_5$ is not consistent with the change of hybridization strength. However, the c/a ratio can roughly distinguish the hybridization strength among different subfamilies. If the number of adjacent M atoms is taken into account, c can also be used to distinguish the hybridization strength among different subfamilies.

Finally, Fig. 4 summarizes the energy location of $f_{f/2}^1$ and $f^0$ final state of Ce$_m$M$_n$In$_{3m+2n}$. Since the $f_{f/2}^1$ state close to $E_F$ is the tail of Kondo resonant peak, it is greatly influenced by the resolution-convoluted Fermi–Dirac distribution. The $f_{f/2}^1$ will not be discussed here. The energy locations of $f_{f/2}^1$ and $f^0$ final states vary significantly with the material. They have similar behaviors with the change of materials, but not at the same magnitude. The variation of $f^0$ position is nearly an order of magnitude larger than that of $f_{f/2}^1$. For example, the positions of $f_{f/2}^1$ and $f^0$ peaks are $-0.26$ and $-2.08$ eV at CeIn3 and $-0.32$ and $-2.53$ eV at CePt2In7. The $f_{f/2}^1$ and $f^0$ states are shifted down by 0.06 and 0.45 eV, respectively.

In summary, we have studied the electronic structures of HF compounds Ce$_m$M$_n$In$_{3m+2n}$ by ARPES and observed the heavy quasiparticle bands $f_{f/2}^1$ for $f_{f/2}^1$ and $f^0$ in all compounds. It is found that the strength of all three peaks varies greatly with compositions of materials. The hybridization strength of the subfamily CeMIn$_5$ is the strongest in the Ce$_m$M$_n$In$_{3m+2n}$ family. Compared with the $f^1-f^0$ intensity ratio, the $f_{f/2}^1-f^0$ intensity ratio is a better indicator of hybridization strength. The c/a ratio and Ce–M band distance d, considering the number of adjacent M atoms, set the overall hybridization strength within each subfamily.

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