5–7 How Heavy Electrons are Formed in Eu-Based Compounds

—Direct Observation of the Band Structure of Eu 4*f* Orbitals, Which Have Magnetic and Localized Characteristics, by Synchrotron Radiation Experiments—

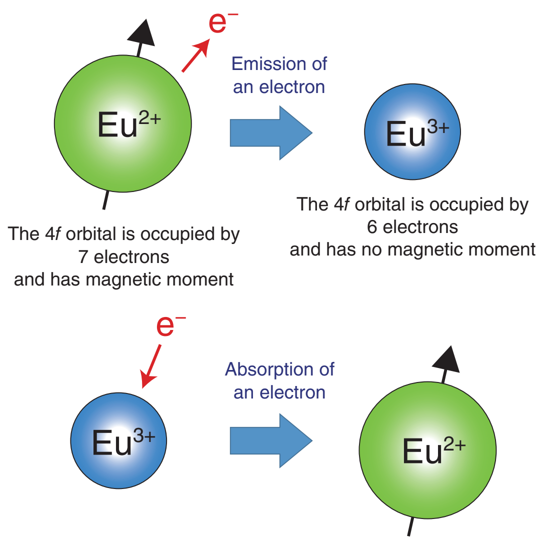

Fig.5-14 Valence instability of Eu-based compounds
Because of the small energy difference between Eu^{2+} and Eu^{3+} states, the valence state of Eu atoms fluctuates as a function of time and spatial coordinates.

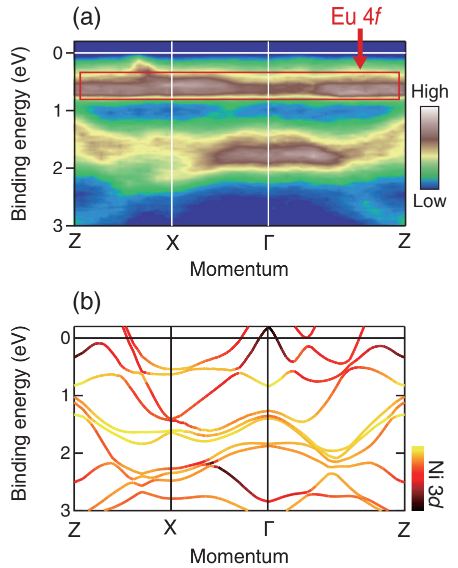

Fig.5-15 Experimental and calculated band structures of $EuNi_2P_2$
(a) The band structure of $EuNi_2P_2$ determined by angle-resolved photoemission experiments. The abscissa represents the position in the momentum space, and the higher-intensity part corresponds to the position of the energy bands. (b) The corresponding calculated band structure of $EuNi_2P_2$. The color scale shows the contribution of Ni 3*d* state.

Rare-earth–based compounds possess various intriguing properties such as superconductivity and thus have garnered much attention in the field of strongly correlated electron physics. Moreover, these compounds have been considered as potential candidates for magnetic devices as the rare earth atoms have large magnetic moments. The physical properties of the rare-earth–based compound are accounted for by the 4*f* electrons of the rare earth atoms. The 4*f* electrons acquire itinerant properties through hybridization with other electron orbitals and form energy bands and fermi surfaces. This hybridization is the origin of the various magnetic orderings and superconductivities of rare-earth–based compounds. However, a relatively small 4*f* binding energy is required to hybridize with other electron orbitals, and therefore, observations of superconductivity have been reported only for Ce- and Yb-based compounds so far. On the other hand, in recent years, it has been recognized that for some Eu-based compounds, the hybridization effect on the 4*f* electrons becomes important and leads to various unusual properties.

It is well known that most rare earth atoms are nearly trivalent states in intermetallic compounds. However, some Eu-based compounds show a valence instability between the trivalent Eu^{3+} and divalent Eu^{2+} states because of the small energy difference between these states. As shown in Fig.5-14, this valence fluctuation corresponds to an instability of the number of 4*f* electrons. Because of this instability, the 4*f* electrons acquire a mobility that enables them to move throughout the crystal and govern the electronic and physical properties of Eu-based compounds. Although no Eu-based superconductor has been discovered until now, it has been reported that some Eu-based compounds exhibit heavy electron behavior in which the effective electron mass is strongly enhanced because of the electron correlation effect.

$EuNi_2P_2$ is known as the first heavy-electron system among the Eu-based compounds. In this study, we have performed angle-resolved photoemission experiments on $EuNi_2P_2$ at the JAEA beamline BL23SU at SPring-8. In angle-resolved photoemission, the energy and angular distributions of the photoelectrons were measured to directly observe the band structure of the materials.

Fig.5-15(a) shows the experimental band structure determined by angle-resolved photoemission. We observed a flat band structure originating from the Eu 4*f* orbitals. The flatness of the bands reflects a large effective electron mass, and thus, this band shape seems to be the origin of the heavy electron behavior in $EuNi_2P_2$. A comparison with the calculated band structure (Fig.5-15(b)) shows that Eu 4*f* electrons hybridize with other orbitals such as Ni 3*d*. This result confirms that Eu 4*f* electrons interact with other electron orbitals and contribute to the formation of the band structure because of valence instability. Moreover, we confirmed the presence of Eu 4*f* at the Fermi level. This means that the electronic and thermodynamic properties of this compound are markedly affected by the Eu 4*f*–derived heavy electrons as these properties are usually governed by electron excitations at the Fermi level.

The present result provides an important clue for understanding the mechanism of formation of heavy electrons in Eu-based compounds. We hope that Eu-based superconductors will be discovered in future experimental studies. Such a discovery will further expand the research field of Eu-based compounds.

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Reference

Kawasaki, I. et al., Electronic Structure of the Intermediate-Valence Compound $EuNi_2P_2$ Studied by Soft X-Ray Photoemission Spectroscopy, Physical Review B, vol.104, issue 16, 2021, 165124, 8p.