Kink during the formation of the Kondo resonance band in the heavy fermion system

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We have shown that the kink behavior in the spectral function of heavy fermion can appear during the formation of the Kondo resonance (KR) band and the hybridization gap (HG). We have investigated the heavy fermion compound CeCoGe2, using a combined approach of the density functional theory (DFT) and the dynamical mean field theory (DMFT). Low temperature (T) spectral functions show dispersive KR states, similar to the recent experimental observation. During the evolution from the nonf conduction band state at high T to the dispersive KR band state at low T, which have topologically different band shapes, we have found the existence of kinks in the nonf spectral function near EF. The observation of kink is clearly in correspondence with the multiple temperature scales of the formation of the KR band.

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The Hamiltonians for the heavy fermion compounds, such as the Kondo lattice model and the periodic Anderson lattice model, are represented in terms of the kinetic energies of conduction electrons, the correlation energies among localized electrons, and the hybridization between them. In Ce-based compounds, the local kinetic energies of conduction electrons, the correlation functions show dispersive KR states, similar to the recent experimental observation. During the evolution from the nonf conduction band state at high T to the dispersive KR band state at low T, which have topologically different band shapes, we have found the existence of kinks in the nonf spectral function near EF. The observation of kink is clearly in correspondence with the multiple temperature scales of the formation of the KR band.

Depending on the temperature, the localization of 4f electrons is determined by the competition between the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and Kondo effect. The RKKY interaction is the conduction electron-mediated exchange energy between 4f electrons. The Kondo effect induces the spin singlet state of the localized and delocalized electrons. The hybridization strength between 4f and nonf electrons strongly affects both the RKKY interaction and the Kondo effect. At the weak hybridization, the RKKY interaction drives the localized 4f electrons to be magnetic. The Kondo effect at the strong hybridization produces the Fermi liquid states with heavy mass.

The momentum-dependent spectral function (A(k, ω)) at low T was in good agreement with the measured spectrum and provided the information of the orbital-dependent hybridization strength.

In this study, we have theoretically described the formation of the dispersive KR band and the HG in the heavy fermion compound CeCoGe2. It has the orthorhombic base-centered structure, shown in Fig. 1. Though the nearest neighbours of Ce atoms are Ge atoms, the sp states in Ge ions are almost empty. The open-core calculation (4f states set to be inside the core), which is equivalent to the electronic structure at high T, demonstrated that Co 3d states have the most contribution to EF. So, Co 3d states can be regarded as main character of nonf states. The KR states are composed of dispersive bands arising from the hybridization of nonf and renormalized f bands, and show quantitative agreement with the recent ARPES measurement. We have shown that the formation of the KR band, which brings about a topological change of the band structures, should be accompanied by the kinks in the spectral function. We propose that the kink could appear in the nonf spectral function (A_nonf(k, ω)) prior to the formation of the KR band around T*.
The kink, the abrupt change in the band dispersion, is usually observed in the ARPES measurements of high \( T_C \) superconductors. \cite{24-26} The discontinuous quasiparticle band is induced by various collective excitations, such as phonon \cite{13-15} and spin-fluctuation. \cite{16} When the strong electron-phonon coupling disturbs the velocity and the scattering rate of electrons, ARPES spectrum near the phonon energy shows the abrupt change in the slope of the energy-momentum dispersion. The ARPES experiment on \( \text{USb}_2 \) also revealed the kink feature in the dispersion of \( f \) bands, which was explained by the combination of quasiparticle bands and many-body correction of the electron-spin-fluctuation coupling. \cite{21} On the other hand, it was reported that the pure electronic correlation in the Hubbard model can produce such a kink near \( E_F \), which suggests that the one-particle picture should be strongly renormalized below a certain energy \( \omega^* \) to have the kink feature in momentum space. This analogy was applied to the periodic Anderson model, showing the existence of the kink. \cite{22} Recently, the energy scale of the kink was proved to be smaller than the width of the central peak. \cite{22} Other subsequent studies, however, showed that the kink in the Hubbard model arises from the internal spin-fluctuation mode. \cite{23, 24} In the present study, instead of kinks in correlated bands, we show that kinks can be phenomenologically observed in noncorrelated bands due to the formation of correlated \( f \) states in the heavy fermion system.

We have used the DFT+DMFT method implemented in the linearized Muffin-Tin orbital band method. \cite{25} We have considered the experimental crystal structure and the Brillouin zone shown in Fig. 1. The DMFT calculation considers only the local self-energy of \( 4f \) orbital, and other orbitals are considered in the DFT part. The local correlation effect is contained in the self-energy \( \Sigma(\omega) \), which can be calculated from the corresponding impurity problem. To solve the impurity problem, we used a vertex corrected one-crossing approximation (OCA) which is a self-consistent diagrammatic method perturbed in the atomic limit. \cite{26} We used the same interaction parameters \( U=5.0 \) and \( J=0.8 \) eV, as in previous works on \( \text{CeIrIn}_5 \). \cite{27, 28} It has been checked that the OCA describes well the \( T \)-dependent spectral function of heavy fermion compound. \cite{27, 28} We neglected the crystalline electric field (CEF) effect on the local correlation energy because \( \text{CeCoGe}_2 \) has been confirmed as \( j=5/2 \) heavy fermion. \cite{29}

Figures 2(a) and 2(b) show \( A^{nonf}(k, \omega) \)'s along \( S-R-T-Y \) at \( T=1200 \) and \( 10 \) K, which demonstrates the formation of the \( \text{KR} \) bands near \( E_F \). The dispersive \( spd \) bands at high \( T \) (1200 K) look almost vertical due to the narrow energy range. At low \( T \), as a result of the hybridization, new coherent quasiparticle bands are formed to give the different band geometry near \( E_F \). For example, there are two separate bands crossing \( E_F \) between \( S \) and \( R \) at high \( T \), while, at low \( T \), additional \( j=5/2 \) bands with the bandwidth of \( \sim 10 \) meV are introduced to produce degenerated three composite quasiparticle bands. As a result, the lower part of two bands observed at high \( T \) are warped due to the hybridization, as indicated by green circle in Fig. 2(b). One band closer to \( S \) is pushed down below \( E_F \), but another band still intersects \( E_F \). It is interesting that the formation of the parabolic-like hybridized band originated from high \( T \) separated bands below \( E_F \) around \( k=D \) at low \( T \). All these features result in the change of geometry in the FS, as will be shown in Figs. 3(e) and 3(f).

\( T \)-dependent formation of the HG has been examined at the chosen \( k \)-points in Figs. 2(c)-(f). At high \( T \), \( A^{nonf}(k, \omega) \) shows a general quasiparticle spectral feature with a single Gaussian function at each \( k \)-point. With lowering \( T \), the spectral weights of \( A^{nonf}(k, \omega) \) near \( E_F \) begin to be transferred to upper and lower peaks separated by \( \Delta_{HG} \) to form a gap structure. At low \( T \), the clear gap can be observed at each chosen \( k \)-point. The size of gap \( \Delta_{HG} \) in \( A^{nonf}(k, \omega) \) can be measured by the optical conductivity. Because \( \Delta_{HG} \) has a variation of 70 to 100 meV depending on \( k \)-points, the measured \( \Delta_{HG} \) should show multiplet structures or widespread shape in the optical conductivity measurements. The small peaks near \( E_F \) at 20 K are induced due to the formation of quasiparticle bands of \( j=5/2 \) states within the HG.

Figure 3 shows both \( A^{nonf}(k, \omega) \) and the \( f \) spectral function \( (A^f(k, \omega)) \) along \( R-T \), and the integrated density of states (DOS) around \( E_F \) at \( T=1200, 300, \) and 10 K. The \( T \)-dependent development of the \( \text{KR} \) states is clearly confirmed in the DOSs of Figs. 3(c), (f), (j). At high \( T \), the upper and lower Hubbard bands are located near 2 to 3 eV above \( E_F \) and 2 eV below \( E_F \), respectively (not shown here). At the elevated \( T \), the profile of the DOS near \( E_F \) comes mostly from non\( f \) states, although there is a weak background spectrum of \( 4f \) states. With lowering \( T \), the weights of the upper and lower Hubbard bands are reduced and transferred to the \( \text{KR} \) states near \( E_F \). The \( 4f \) states give the main contribution to the DOS near \( E_F \) below \( T^* \).

At high \( T \) (1200 K), \( A^f(k, \omega) \) shows weak intensity near \( E_F \), but has clear dispersive band feature similar to \( A^{nonf}(k, \omega) \). This means that small hybridization still
FIG. 2: (color online) Theoretical demonstration of the HG feature from $T$-dependent $A^{nonf}(k,\omega)$. $A^{nonf}(k,\omega)$'s are provided along S-R-T-Y (a) at $T = 1200$ K, and (b) at $T = 10$ K. S, R, T, and Y are $k = (\pi/2, \pi/2, 0)$, $(\pi/2, \pi/2, \pi/2)$, $(0, \pi/2, \pi/2)$, and $(0, \pi/2, 0)$, respectively. The green circle in (b) is for the emphasis of the $T$-dependent feature from $nonf$ states. As $T$ decreases, the momentum dependence of dispersive KR peaks are correctly captured in the calculation. The weak dispersion-less feature, which is the precursor of $nonf$ states, shows the feature of kinks near $E_F$ to the total LDOS and the total DOS, as shown in Figs. 3(c), (f), (i) provide the $T$-dependent enhancement of the spin-orbit multiplet around $-0.3$ eV. It is noteworthy that the multiplet shows almost flat feature because the incoherent feature (broadening of bands) is much bigger than the dispersion of the KR states.

Because the spectra of high and low $T$ show clearly different quasiparticle band structures near $E_F$, the $T$-dependent evolution should show some feature of phase transition or crossover. Figures 3(d) and (e) show the spectra in the intermediate $T$. $A^f(k,\omega)$ around $E_F$ shows effectively dispersion-less feature, which is the precursor of the formation of the KR states. Below and above the KR state, the $nonf$ bands are warped in different directions. At the energy of the KR state, the $nonf$ bands are not well defined due to the incoherent contribution of $A^f(k,\omega)$ to the $nonf$ states. As a result, $A^{nonf}(k,\omega)$ shows the feature of kinks near $E_F$. Distinctly from the
kinks observed in other correlated systems, such as high $T_C$ superconductor, the kinks in heavy fermion system should appear in the noncorrelated bands during the formation of the KR bands and the HG.

Figures 3(a)-(d) show the schematic picture of emergence of kink during the formation of the KR bands At high $T$ in Fig. 3(a), there are only non-$f$ conduction bands that can be usually well described by the open-core band calculation, in which the occupied Ce 4$f$ state is treated as a core level. With lowering $T$ in Fig. 3(b), the incoherent KR states of 4$f$ electrons start to contribute to $E_F$, whereby the kink feature starts to emerge in $A_{nonf}(k, \omega)$. Here the kink structure is far from the "water-fall" shape, rather close to a "bell-profile" shape, indicated by the arrow in Fig. 3(d), since the dispersion changes happen rather close to a "bell-profile" shape, indicated by the arrow in Fig. 3(d), since the dispersion changes happen at two spin-orbit multiplets ($j = 5/2, 7/2$). As decreasing $T$ further, the $f$ electrons start to be coherent slowly, and the non-$f$ bands are still being deformed. This procedure corresponds to Fig. 1(c), where the coherent character of bands becomes enhanced around $E_F$. In this case, $A_{nonf}(k, \omega)$ has the kink structure of the "water-fall" shape due to the separation of the upper and lower hybridized bands. At lower $T$ in Fig. 3(d), most $f$ electrons near $E_F$ become coherent to make the fully coherent bands near $E_F$. Accordingly, the region of the kink feature is changed into that of the HG feature. Interestingly, the electron FS area gradually enlarges during this procedure.

The area of the electron FS around $R$, which is identified as $\alpha$ branch in Figs. 1(e) and (f), increases continuously with lowering $T$. In our recent DMFT study, on the FS of heavy fermion CeIrIn$_5$, two temperature scales are proposed: one ($T_f$) for the $T$-dependent evolution of the FS size, and the other ($T_m$) for the $T$-dependent evolution of the cyclotron effective mass ($m^*$). $T_f$ should be related to the contribution of local $f$ electron to conduction electron. On the other hand, $T_m$ is a characteristic of the formation of coherent 4$f$ bands in the lattice since the $m^*$ reflects the renormalization of the carriers. Although $T_m$ is defined by the change of the effective mass of the FS, it should be similar to $T^*$ where the formation of the coherent KR states begins. Similar to CeIrIn$_5$, the same scaling behavior is also shown in CeCoGe$_2$. The FS branch $\alpha$, which is the well-defined FS branch at all $T$, as shown in Figs. 1(e) and (f), is chosen for this study. By analyzing $T$-dependent scaling behaviors in Figs. 1(g) and (h), we found $T_f \sim 200$ K and $T_m \sim 90$ K, respectively, for $\alpha$ branch.

The kink can be observed around $T_f$, where the incoherent $f$ state contributes to $E_F$. (see the Supplementary Movie.) The kink phenomenon is changed into the gap feature gradually between $T_f$ and $T_m$, where the contribution of incoherent 4$f$ electron states disturb the band dispersion near $E_F$. Well below $T_m$, the HG and KR states are well defined. So, the formations of the kink around $T_f$ (200 K) will be the precursor of the HG below $T_m$ (90 K). Note that the kink features are also observed around 0.3 eV above $E_F$, as shown in Fig 3(d), due to the incoherent contribution of spin-orbit multiplet. The multiplet around $-0.3$ eV does not give the kink because the contribution of $f$ state is too weak to distort the non-$f$ bands.

In summary, we have analyzed the $T$-dependent evolutions of $A(k, \omega)$ in the heavy fermion compound CeCoGe$_2$. It is shown that the DFT+DMFT calculations are consistent with the experimental measurements. We propose that the kink of $A_{nonf}(k, \omega)$ around $E_F$ can be identified during the evolution from the dispersive non-$f$ state at high $T$ to the HG and KR states at low $T$. Phenomenologically, the kinks observed in this work will show the similar shape to those in other experiments, even though the conventional kinks appear in the correlated bands via the interaction with other excitations, such as phonon and spin-fluctuation. The kinks in current work should be distinguished also from the one only with the electronic correlation in previous studies. As indicated in Fig. 4, the kink can be observed between $T_m$ and $T_f$, while all other kinks in previous studies should be shown well below $T^*$ ($\sim T_m$). The kink induced by the correlation between the incoherent 4$f$ and dispersive non-$f$ electrons above $T^*$ can be investigated in the state-of-the-art $T$-dependent ARPES experiments. We suggest that the detailed analysis on the abrupt change of electron velocity and the scattering rate near the Fermi level will provide crucial information for the heavy fermion system.

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(a) High T  
(b) Kink happens  
(c) Enhanced Hybridization  
(d) After Hybridization

FIG. 4: (color online) The formation of the HG can be divided into four processes in (a)-(d), using $T_f$ and $T_m$ that are obtained by the scaling analysis in (g) and (h). (a) The high $T$ quasiparticle band is displayed. The blue color represents $A^{nonf}(k, \omega)$. (b) For $T \sim T_f$, the quasiparticle band is interrupted by the incoherent $4f$ electrons. The red dotted-line is provided as a guide for the high $T$ band. (c) For $T_f > T > T_m$, the hybridization between non$f$ and $4f$ electrons is strengthened. (d) For $T < T_m$, the HG and $k$-dependent (dispersive) KR state can be well defined. The orange color represents $A^f(k, \omega)$. With lowering $T$, $E_R$ increases gradually due to the participation of the new carrier from the incoherent $4f$ electrons. The FS on $k = Y$ plane at high and low $T$ are provided in (e) and (f), respectively. Color represents the different band indices. The scaling behaviors of $T$-dependent renormalized de Haas van Alphen frequency ($\Delta_{HG}$) and cyclotron effective mass ($m^*$) for the FS branch $\alpha$ shown in (e) and (f) are analyzed in (g) and (h), respectively.

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