Optical conductivity of CeMIn$_5$ (M=Co, Rh, Ir)

F. P. Mena

Material Science Center, University of Groningen, 9747 AG Groningen, The Netherlands

D. van der Marel

Département de Physique de la Matière Condensée, Université de Genève, quai Ernest-Ansermet CH - 1211 Genève, Switzerland

J. L. Sarrao

Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

The optical properties of the heavy fermion family CeMIn$_5$ (M=Co, Rh, Ir) have been determined in the photon energy range from 2 meV to 4.5 eV using the combination of near normal incidence reflectivity (2 meV–0.8 eV) and ellipsometry (0.8–4.5 eV). In all compounds signatures of the formation of a coherent state are found. However, as a result of the competition with antiferromagnetism, the energy region where the coherent state occurs is much lower in the compound that orders magnetically.

PACS number(s): 78.20.−e, 71.27.+a, 78.30.−j

I. INTRODUCTION

Cerium based compounds are well known to exhibit strongly correlated electronic phenomena. Among them, the materials of the CeMIn$_5$ (M=Co, Rh, Ir) group have recently attracted considerable attention. The reason is their unusual properties, especially their unconventional superconductivity similar to the high-T_c superconductors[1,2] and, presumably, proximity to a quantum critical point (with pressure[2,3] and magnetic field[4] as tuning parameters). The analogy with the high-T_c superconductors even starts with their tetragonal crystal structure which can be seen as layers of CeIn$_3$ separated by MIn$_2$ layers. These heavy fermion compounds exhibit an ample gamut of properties that reveal the subtle interplay between magnetism and superconductivity (SC) as shown by their phase diagram[5] (which has been reproduced here in Fig. 1 for the sake of clarity). CeCoIn$_5$ is the heavy fermion compound with the highest superconducting transition temperature T_c at ambient pressure (2.3 K).[5,6] If Co is replaced by either Rh or Ir (all of them being isoelectric), T_c decreases monotonically. In the Rh direction, T_c becomes zero before reaching CeRhIn$_5$. On the other hand, in the Ir direction, it remains finite all the way to CeIrIn$_5$ where it is 0.4 K. From there, if Ir is replaced by Rh, T_c increases having a maximum at around a 50% concentration, then, it decreases to zero again before reaching CeRhIn$_5$. Although CeRhIn$_5$ does not show superconductivity at ambient pressure, it shows antiferromagnetism (AF) below T_N=3.8 K.[7] When Rh is replaced by either Co or Ir, T_N decreases monotonically and, in both cases, reaches zero before complete substitution is achieved. Moreover, in both cases, there are regions where AF and SC coexist.

The physics of a single magnetic impurity in a nonmagnetic metallic matrix is characterized only by the Kondo temperature T_K. Below T_K, the local moment of the impurity is screened by the conduction electrons and the system can be described as a Fermi liquid. Moreover, all thermodynamic quantities scale with the reduced temperature T/T_K. An important question in the Kondo lattice (a lattice of magnetic

centers) is whether the temperature below which the moments are locally screened (T_K) coincides with the temperature below which the Fermi liquid is formed ($T^\star$) and how these are related to the Kondo temperature of the single impurity.[8] Experimentally, $T^\star$ is usually associated with T_{coh} which is the temperature where the resistivity shows a maximum. If intersite interactions are not taken into account, calculations in the dynamical mean-field theory[8,9] indicate that there are two completely different energy scales. The first one, T_K, coincides with that of the single impurity case while the second one depends only on the ratio between the conduction and f-electron densities, (n_c/n_f). It turns out that when $n_c/n_f \sim 1$, $T^\star$ is slightly enhanced over T_K but rapidly decreases as the exhaustion regime is reached ($n_c \ll n_f$). If, on the other hand, intersite interactions are considered,[10] the single impurity and the lattice are characterized by two different Kondo temperatures.

In contrast with this point of view, from the study of the effect of substitution of the Ce for La in CeMIn$_5$, Nakatsuji and co-workers[11,12] have proposed that the single impurity energy scale "survives" when going to the lattice with $T^\star$ being the result of intersite correlations. At high temperatures, the Kondo lattice behaves as a set of noninteracting magnetic impurities (Kondo gas) which condense below $T^\star$ (Kondo liquid). However, they suggest, the condensation is not complete when the material is close to a quantum critical point, and at low temperatures the material can be described as a two-component system. Since there is no evidence for spatial phase separation, what occurs below $T^\star$ is a transfer of spectral weight from the high-frequency part of the spectrum (corresponding to the localized Kondo moments) to the low-frequency part (corresponding to the coherent state). In the present work we present optical measurements in the CeMIn$_5$ family to elucidate these spectral weight shifts. We experimentally see how antiferromagnetism (AF) competes with the formation of the coherent state. This competition manifests itself in lower values for the characteristic energies of CeRhIn$_5$ in comparison to the compounds that do not
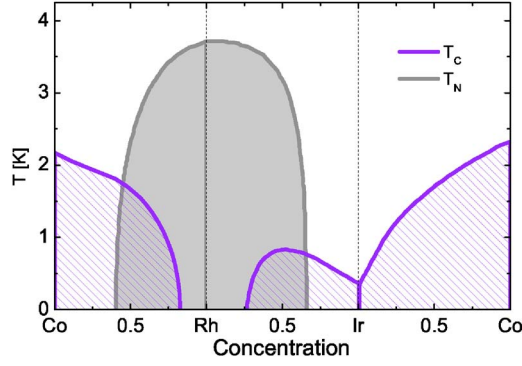
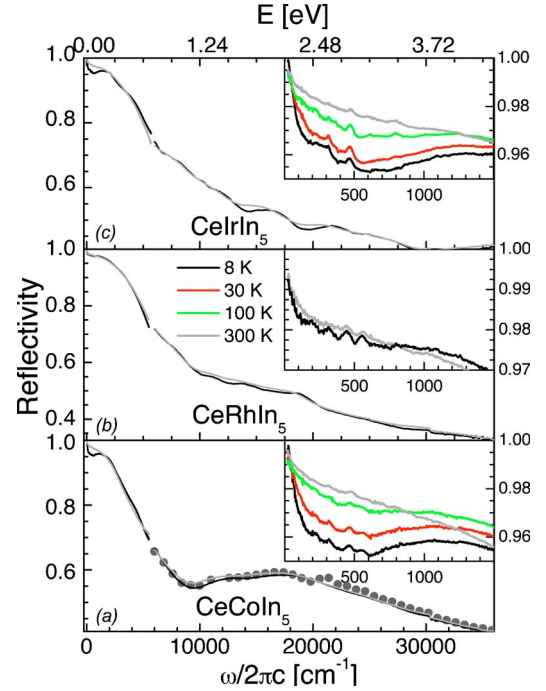FIG. 1. (Color online) Phase diagram of CeMIn$_5$ (adapted from Ref. 5).

FIG. 2. (Color online) Reflectivity of the different samples at several temperatures. Below 6000 cm^{-1}: measured. Above 6000 cm^{-1}: calculated from the dielectric function. The gray dots show the measured reflectivity of the *ab* plane of CeCoIn$_5$. Insets: Reflectivity below 2000 cm^{-1}.

order magnetically. We will see that the loss of spectral weight associated with the formation of the coherent state occurs in the same energy range for all three members of the CeMIn$_5$ group. Furthermore, the energy where this recovery occurs is much larger than the region where the mass enhancement occurs which seems to be a common feature in Kondo systems.

II. EXPERIMENT AND RESULTS

High quality single crystals of CeMIn$_5$ (M=Co, Rh, Ir) were grown as described elsewhere.[1,6,7] After polishing, the optical properties of the *ab* plane were determined at different temperatures by combining reflectivity at near normal incidence (11°) and ellipsometry. Both experiments were performed at the same set of temperatures: every 5 K below 40, 50, 75, 100, and 200 K, and room temperature. The reflectivity was measured from 30 to 6000 cm^{-1} using a Fourier transform spectrometer. For measuring the temperature dependence we used a home-built cryostat and the intensities were calibrated against a gold reference film evaporated *in situ* on the sample. At room temperature we also measured the reflectivity from 6000 to 36 000 cm^{-1} using a monochromator.[13]

The ellipsometry experiment of the *ab*-plane surface was performed from 6000 to 36 000 cm^{-1} at an angle of incidence of 80°. The temperature dependence was performed using an ultrahigh vacuum cryostat. From this experiment we obtained the pseudodielectric function. We used the normal incidence reflectivity data in the same frequency range to calculate the correction for the admixture of the *c*-axis dielectric function in the pseudodielectric function, which turned out to be negligible in agreement with Aspnes.[14] To illustrate the results, we show the reflectivity (measured and

calculated from ellipsometry) in the whole experimental range in Fig. 2.

From the complete data set, the complex dielectric function, $\epsilon(\omega)=\epsilon_1(\omega)+i(4\pi/\omega)\sigma_1(\omega)$, was calculated in the infrared (30–6000 cm^{-1}) using Kramers-Kronig relations (KK) following the procedure described in Ref. 15, which includes a fit of the available data to a Drude-Lorentz (DL) model dielectric function:

$$\epsilon(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\Gamma)} + \sum_j \frac{\omega_{p,j}^2}{(\omega_{o,j}^2 - \omega^2) - i\omega\Gamma_j}. \quad (1)$$

The fit parameters obtained at the lowest temperature in the far infrared are presented in Table I while in Fig. 3, $\sigma_1(\omega)$ in the entire frequency range is shown. In the lower panel of the latter we also show the error bars corresponding to the absolute error and which is representative of all compounds. However, it has to be kept in mind that if we consider a specific compound, the error bars of the conductivity difference when comparing two different temperatures is smaller. The conclusions presented below are consistent with these

TABLE I. Low-frequency fit parameters, in cm^{-1}, at 8 K [see Eq. (1)].

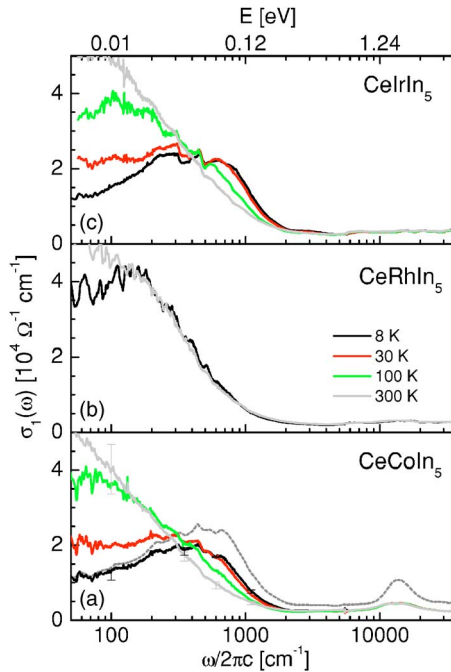| Sample	ω_p	Γ	$\omega_{o,1}$	$\omega_{p,1}$	Γ_1	$\omega_{o,2}$	$\omega_{p,2}$	Γ_2
CeCoIn$_5$	7454	8	181	21630	531	562	21555	694
CeRhIn$_5$	8810	14	122	31241	392			
CeIrIn$_5$	8864	14	248	28696	629	777	21668	683

FIG. 3. (Color online) Real part of the optical conductivity at several temperatures. The shown error bars correspond to an absolute error of 0.5% in reflectivity (the relative error between different temperatures is smaller). The lower panel also shows the obtained $\sigma_1(\omega)$ if the high-frequency reflectivity of Ref. 20 is used in the KK analysis (gray dotted line).

error bars. It is also necessary to mention that the method presented here decreases the error bars when compared with the one that uses only reflectivity.[15]

III. DISCUSSION

A. Optical conductivity

The temperature dependence of the optical conductivity of the Co and Ir compounds at low frequencies follows closely what has been previously seen in other heavy fermion systems.[16,17] Above T_{coh}, the optical conductivity at low frequencies has a Drude-like behavior while below it, a minimum appears signaling the appearance of a narrower Drude mode. The origin of this behavior is the development of the so-called hybridization gap, Δ, as a result of the hybridization between the conduction electrons and the localized f moments.[18] The intraband transitions in the lower band produce the narrow Drude mode while the interband transitions between the two resulting bands produce the bump seen at higher frequencies.[18,19] In the Rh compound these characteristics are less strong but still a decrease in the optical conductivity can be seen at low frequencies. Since this compound eventually becomes AF at 3.8 K, this seems to be the result of the competition between the magnetic ordering and the coherent state. We will discuss more about this point below.

When comparing our results for CeCoIn$_5$ with those of a previous report,[20] we notice that both show a minimum in $\sigma_1(\omega)$ at the same position and a shoulder at 250 cm^{-1}. The
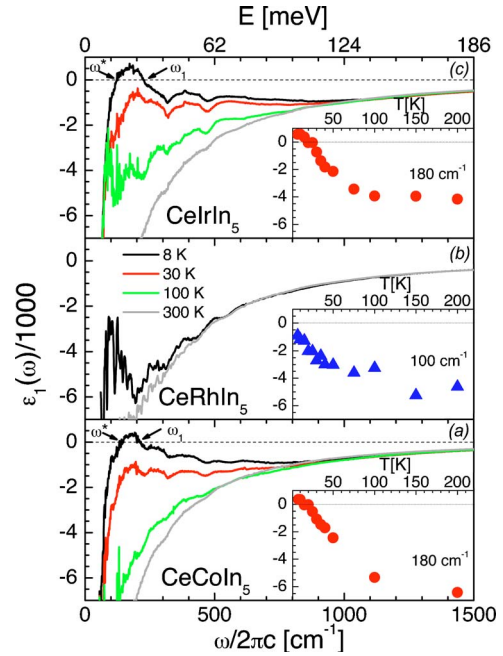

FIG. 4. (Color online) Real part of the dielectric function in the infrared. The arrows indicate the zero crossings of $\epsilon_1(\omega)$. Insets: Temperature dependence of $\epsilon_1(\omega)$ at selected frequencies.

later feature was interpreted as a Holstein band (an absorption corresponding to the bosons that couple the electrons in the SC state) inside of the gap which could be attributable to AF critical fluctuations.[11,20] The argument for such assignment is that, in contrast with what has been seen in other heavy fermion systems,[16] there is not a sudden decrease of the optical conductivity just below the peak due to the transitions across the hybridization gap. The interband transitions were related to a strong peak seen at 600 cm^{-1} which has less spectral weight in our results. Besides the understandable differences between different experimental sets, the origin of such a large spectral weight could be due to an overestimation of the reflectivity in the visible range (which we measured directly with ellipsometry). To check this supposition, we have performed KK using our own reflectivity data and the reflectivity from Ref. 20 above 6000 cm^{-1}. The result is presented as the gray dotted line in Fig. 3(a). Such a procedure gives more spectral weight near 600 cm^{-1}. Therefore the whole structure between $\approx$100 and 1000 cm^{-1} should correspond to transitions across the hybridization gap. This assignment is further supported below when we discuss the properties of $\epsilon_1(\omega)$.

B. Dielectric function

The development of the hybridization gap can also be seen in the real part of the dielectric function at low frequencies (Fig. 4). As the temperature is lowered, $\epsilon_1(\omega)$ changes from purely metallic (monotonically decreasing) to a region (below $\approx$300 cm^{-1}) with a maximum. The insets of Fig. 4 show the detailed temperature dependence of $\epsilon_1(\omega)$ at the frequency where the maxima occur at the lowest temperature. In those insets it is possible to see the gradual develop-

TABLE II. Characteristics parameters of CeMIn$_5$. T_{coh} is taken from the dc resistivities of Refs. 1, 6, and 7. $m^\star/m$ is calculated from spectral weight considerations.

Sample	ω^* cm^{-1}	ω_1 cm^{-1}	T_{coh} cm^{-1} (K)	N_{eff} (2000 cm^{-1}) carriers/formula unit	$m^\star/m$	T^* [Eq. (3)] cm^{-1} (K)	T^* (Refs. 11 and 12) cm^{-1} (K)
CeCoIn$_5$	140	200	31 (45)	1.5	15	44 (64)	31 (45)
CeRhIn$_5$	<100	<100	28 (41)	1.8	13	<32 (<48)	10 (15)
CeIrIn$_5$	120	220	36 (52)	2.0	16	40 (58)	14 (20)

ment of such maxima which seems to start above T_{coh}. In the Co and Ir compounds, $\epsilon_1(\omega)$ even has two extra zeros whose values are given in Table II. This behavior shows the definitive establishment of the coherent state.[18] The lowest zero, ω^*, may be thought of as the screened heavy fermion plasma mode[18] while the second one, ω_1, comes from the contribution of the interband transitions to $\epsilon_1(\omega)$. Since these transitions have a threshold energy Δ, ω_1 gives an estimate of the value of the hybridization gap. The shoulders seen at ~ 250 cm^{-1} in the Co and Ir compounds are above the gap and therefore most likely correspond to transitions across the gap. This interpretation is corroborated by the DL fit where the first Lorentz oscillator is located at a frequency $\omega_{o,1} \sim \omega_1$. The dielectric function of CeRhIn$_5$, though it shows a maximum at 8 K, has no zero crossings indicating that, in contrast to the other compounds, the coherent state is not completely formed. We interpret this phenomenon as the well known competition between magnetic ordering and the formation of the correlated state. Those systems that form a magnetic ordered state usually present a smaller heavy fermion character.[21]

C. Generalized Drude formulation

To get more insight about the competition between magnetism and the correlated state, we analyze our results with the generalized Drude formulation:

$$\sigma(\omega) = \frac{\omega_p^2}{4\pi} \frac{1}{1/\tau(\omega) - i\omega m^\star(\omega)/m}, \quad (2)$$

where we used ω_p corresponding to N_{eff} (see below) at 2000 cm^{-1} and given in Table II. The obtained frequency-dependent masses are plotted in Fig. 5. The increase of the effective mass is evident at low frequencies as soon as the temperature is lowered below T_{coh}. At the lowest measured frequency, the mass enhancement depends on the sample. However, from the DL fit, we estimate similar mass enhancements at $\omega \rightarrow 0$. Analogous results are obtained if the effective mass is calculated using spectral weight arguments (Table II).[22] The difference between them is the energy range where the mass enhancement occurs, being much smaller for CeRhIn$_5$. In contrast, specific-heat measurements[1,6,7] (which is also an indication of the mass enhancement) indicate, in the normal state, $\gamma = 290$, 400, and 750 mJ/(mol K^2) for Co, Rh, and Ir, respectively. At least for CeRhIn$_5$, as indicated by measurements of the de Haas–van Alphen experiment[23] and of the specific heat in magnetic field,[24] this value of γ seems to be overestimated as the specific heat contains large con-

tributions from magnetic interactions and not only from correlation effects. For completeness, we present the frequency dependent scattering rates in the insets of Fig. 5. The same characteristics are seen here. Both the Co and Ir compound show the typical characteristics of a completely formed coherent state while this is not the case in CeRhIn$_5$. Notice also the frequency dependence of the scattering rate at the lowest temperatures which is similar to the temperature dependence of the dc resistivity. This characteristic demonstrates the presence of only one characteristic energy in the formation of a heavy fermion system.

Before finalizing this section, we would like to make some remarks about the applicability of Eq. (2). The generalized Drude analysis is valid in the frequency range where there are no interband transitions, and the optical conductivity is characterized by a single frequency-dependent mass renormalization function. In the previous paragraphs, we have attributed ω_1 to an interband transition. A meaningful interpretation of the frequency-dependent mass and scattering rate is therefore limited to frequencies $\omega < \omega_1$. Indeed above ω_1 the function $m^*(\omega)$ reaches negative values, which
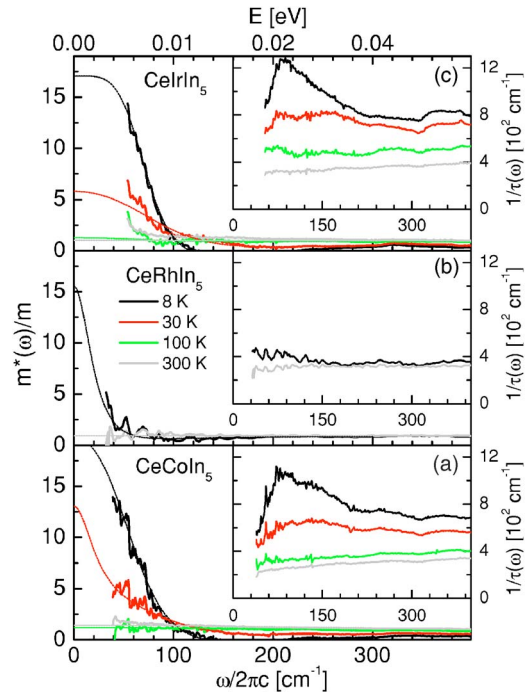

FIG. 5. (Color online) Measured frequency-dependent mass (solid lines) and obtained from a fit to the experimental data (dotted lines). Insets: Frequency-dependent scattering rate.

thus appears to be an artifact caused by applying the generalized Drude analysis to experimental data which for those frequencies are a superposition of intraband and interband transitions.

D. Kondo-Lattice temperature

Another way to see the smaller heavy fermion character of CeRhIn₅ is by studying a simple relation obtained from the calculation of the dielectric function for the Anderson lattice:[18]

$$\omega^{*2} = 6(1 + n_f/n_c)T^{*2}, \qquad (3)$$

where n_c and n_f are the density of conduction and f electrons. To estimate values of T^*, we have used this equation with $n_f = 1$ and $n_c = N_{eff}$ at 2000 cm^{-1} (see next section). These estimated values give again a smaller value of the characteristic temperature for the formation of the Fermi liquid in CeRhIn₅. From the study of the specific heat and the magnetic susceptibility,[11,12] different values of T^* were obtained (Table II) contradicting our observation of a completely settled coherent state in CeIrIn₅ at 8 K. We believe that the idea (see Sec. I) is not complete and more sophisticated calculations are needed to understand the various energy scales involved and how they impact the spectral weight transfer.

E. Spectral weight

Important information about the relevant energy scale of the electron-electron correlations is gleaned from the spectral weight function, which is defined as

$$N_{eff}(\omega) = \frac{m\Omega}{2\pi e^2} \int_0^\omega \sigma_1(\omega')d\omega', \qquad (4)$$

where m and e are the electron mass and charge, respectively, and Ω is the volume of one formula unit. This quantity is depicted in Fig. 6 and the temperature dependencies at 400 and 2000 cm^{-1} are shown in Fig. 7. When the hybridization gap develops, spectral weight is removed from the low-frequency part as evinced, for example, by the behavior of N_{eff} at 400 cm^{-1}. The spectral weight does not start recovering immediately above the gap but eventually does at an energy range much larger than the gap itself. In the compounds that do not order magnetically (CeCoIn₅ and CeIrIn₅) this energy is ~2000 cm^{-1} (0.25 eV), i.e., at least ten times the hybridization gap. Other Kondo systems show similar behavior. In particular, an interesting case is FeSi, where the spectral weight lost during the formation of the Kondo insulator state is transferred to an energy range on the order of several eV.[15,25] The difference between the energies where the recovery of spectral weight takes place seems to be related with the nature of the local moments, d vs f electrons. While the f electrons form extremely narrow bands, the d electrons form rather broad bands.
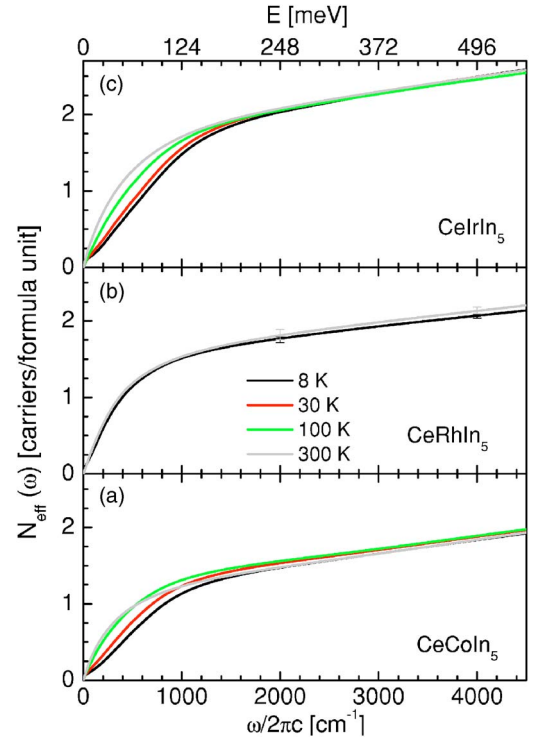

FIG. 6. (Color online) Spectral weight of the optical conductivity below 4500 cm^{-1}.

IV. CONCLUSIONS

In conclusion, we have presented optical measurements on one of the recently most studied families of heavy fermion systems, CeMIn₅ (M=Co, Rh, Ir). We have shown that
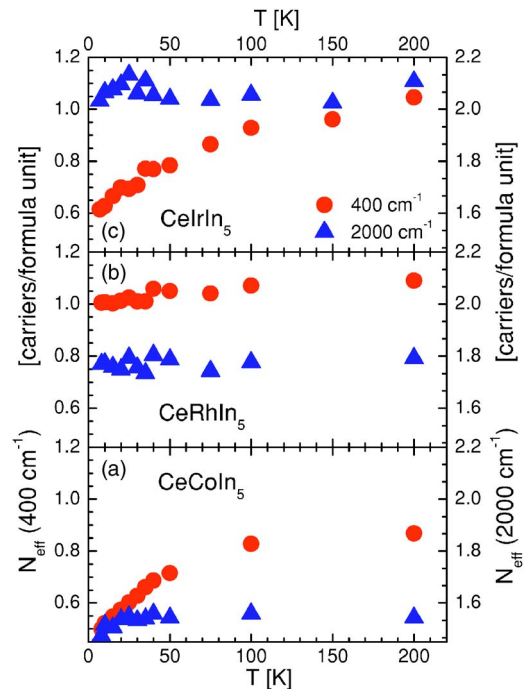

FIG. 7. (Color online) Temperature dependencies of N_{eff} at 400 (circles) and 2000 (triangles) cm^{-1}.

in CeCoIn$_5$ and CeIrIn$_5$, which do not order magnetically, the spectral weight is recovered in a frequency range that is considerably larger than the hybridization gap. We have also observed directly in the optical spectra the quenching of the coherent state by a competing antiferromagnetic phase.

[1] C. Petrovic *et al.*, Europhys. Lett. **53**, 354 (2001).

[2] V. A. Sidorov, M. Nicklas, P. G. Pagliuso, J. L. Sarrao, Y. Bang, A. V. Balatsky, and J. D. Thompson, Phys. Rev. Lett. **89**, 157004 (2002).

[3] T. Mito, S. Kawasaki, Y. Kawasaki, G.-q. Zheng, Y. Kitaoka, D. Aoki, Y. Haga, and Y. Onuki, Phys. Rev. Lett. **90**, 077004 (2003).

[4] A. Bianchi, R. Movshovich, I. Vekhter, P. G. Pagliuso, and J. L. Sarrao, Phys. Rev. Lett. **91**, 257001 (2003); J. Paglione *et al.*, *ibid.* **91**, 246405 (2003).

[5] P. G. Pagliuso *et al.*, Physica B **312-313**, 274 (2002).

[6] C. Petrovic *et al.*, J. Phys.: Condens. Matter **13**, 337 (2001).

[7] H. Hegger, C. Petrovic, E. G. Moshopoulou, M. F. Hundley, J. L. Sarrao, Z. Fisk, and J. D. Thompson, Phys. Rev. Lett. **84**, 4986 (2000).

[8] S. Burdin, A. Georges, and D. R. Grempel, Phys. Rev. Lett. **85**, 1048 (2000), and references therein.

[9] A. N. Tahvildar-Zadeh, M. Jarrell, and J. K. Freericks, Phys. Rev. B **55**, R3332 (1997).

[10] B. Coqblin, C. Lacroix, M. S. Gusmao, and J. R. Iglesias, Phys. Rev. B **67**, 064417 (2003).

[11] S. Nakatsuji, S. Yeo, L. Balicas, Z. Fisk, P. Schlottmann, P. G. Pagliuso, N. O. Moreno, J. L. Sarrao, and J. D. Thompson, Phys. Rev. Lett. **89**, 106402 (2002).

[12] S. Nakatsuji *et al.*, cond-mat/0304587.

[13] For the reflectivity measurement in the visible region we started with the sample with gold evaporated on its surface. The dielectric function of this film was determined first via ellipsometry. Later, in near normal incidence configuration, the intensity of the gold film was measured. Then the film was removed and the intensity of the sample was measured. The ratio of these two quantities, corrected by the gold reflectivity (obtained from the ellipsometric measurement), gives the high-frequency reflectivity.

[14] D. E. Aspnes, J. Opt. Soc. Am. **70**, 1275 (1980).

[15] F. P. Mena, Ph.D. thesis, University of Groningen, 2004.

[16] S. V. Dordevic, D. N. Basov, N. R. Dilley, E. D. Bauer, and M. B. Maple, Phys. Rev. Lett. **86**, 684 (2001).

[17] F. Marabelli and P. Wachter, Phys. Rev. B **42**, 3307 (1990).

[18] A. J. Millis and P. A. Lee, Phys. Rev. B **35**, 3394 (1987); A. J. Millis, M. Lavagna, and P. Lee, *ibid.* **36**, 864R (1987).

[19] P. Coleman, Phys. Rev. Lett. **59**, 1026 (1987); M. Jarrell, Phys. Rev. B **51**, 7429 (1995).

[20] E. J. Singley, D. N. Basor, E. D. Bauer, and M. B. Maple, Phys. Rev. B **65**, 161101(R) (2002).

[21] J. R. Iglesias, C. Lacroix, and B. Coqblin, Phys. Rev. B **56**, 11820 (1997).

[22] $m^\star/m$ is the ratio between the contributions to the spectral weight coming from the broad Drude mode at room temperature and the narrow mode at low temperatures. For the former the integration limit was 2000 cm^{-1}. At the lowest temperature the limits were 55, 35, and 60 cm^{-1} for the Co, Rh, and Ir compounds, respectively.

[23] U. Alver, R. G. Goodrich, N. Harrison, D. W. Hall, E. C. Palm, T. P. Murphy, S. W. Tozer, P. G. Pagliuso, N. O. Moreno, J. L. Sarrao, and Z. Fisk, Phys. Rev. B **64**, 180402(R) (2001).

[24] J. S. Kim, J. Alwood, D. Mixson, P. Watts, and G. R. Stewart, Phys. Rev. B **66**, 134418 (2002).

[25] Z. Schlesinger, Z. Fisk, H. T. Zhang, M. B. Maple, J. F. Di Tusa, and G. Aeppli, Phys. Rev. Lett. **71**, 1748 (1993).