

# Contrasting pressure evolution of $f$ -electron hybridized states in $\text{CeRhIn}_5$ and $\text{YbNi}_3\text{Ga}_9$ : An optical conductivity study

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Optical conductivity [ $\sigma(\omega)$ ] of  $\text{CeRhIn}_5$  and  $\text{YbNi}_3\text{Ga}_9$  have been measured at external pressures to 10 GPa and at low temperatures to 6 K. Regarding  $\text{CeRhIn}_5$ , at ambient pressure the main feature in  $\sigma(\omega)$  is a Drude peak due to free carriers. With increasing pressure, however, a characteristic midinfrared (mIR) peak rapidly develops in  $\sigma(\omega)$ , and its peak energy and width increase with pressure. These features are consistent with an increased conduction ( $c$ )- $f$  electron hybridization at high pressure and show that pressure has tuned the electronic state of  $\text{CeRhIn}_5$  from very weakly to strongly hybridized ones. As for  $\text{YbNi}_3\text{Ga}_9$ , in contrast, a marked mIR peak is observed already at ambient pressure, indicating a strong  $c$ - $f$  hybridization. At high pressures, however, the mIR peak shifts to lower energy and becomes diminished and seems to merge with the Drude component at 10 GPa. Namely,  $\text{CeRhIn}_5$  and  $\text{YbNi}_3\text{Ga}_9$  exhibit some opposite tendencies in the pressure evolution of  $\sigma(\omega)$  and electronic structure. These results are discussed in terms of the pressure evolution of  $c$ - $f$  hybridized electronic states in Ce and Yb compounds, in particular in terms of the electron-hole symmetry often considered between Ce and Yb.

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## I. INTRODUCTION

The physics of strongly correlated  $f$ -electron systems, most typically Ce-based and Yb-based intermetallic compounds, has attracted much interest for the last few decades [1]. Central to the problem is a duality between localized and delocalized characteristics exhibited by the  $f$  electrons. The  $f$  electrons intrinsically exhibit localized characteristics since the  $f$  orbitals are located closer to the nucleus than the conduction states. However, they may become partially delocalized by hybridizing with conduction ( $c$ ) electrons. This  $c$ - $f$  hybridization leads to various interesting phenomena such as the Kondo effect, heavy fermion (HF) formation, intermediate valence (IV), the Rudermann-Kittel-Kasuya-Yoshida interaction, and the associated magnetic ordering. It also plays an important role in the quantum critical phenomena (QCP) at the border of magnetic ordering.

In IV compounds, the  $c$ - $f$  hybridization is fairly strong, and the average Ce or Yb valence significantly deviates from 3 and takes an intermediate value well above and below 3 for Ce and Yb compounds, respectively [2,3]. Optical conductivity [ $\sigma(\omega)$ ] studies have provided much information about their microscopic electronic states [4]. A marked midinfrared (mIR) peak has been commonly observed in  $\sigma(\omega)$  of many Ce- and Yb-based IV metals, and its origin has been discussed in terms of the  $c$ - $f$  hybridized electronic states [5–17]. For example, a model of “renormalized  $c$ - $f$  hybridized bands” has been used to understand the mIR peak [6–10]. In this model, a flat  $f$  band renormalized by large  $f$  electron correlation ( $U$ ) hybridizes with a wide  $c$  band, forming a pair of hybridized bands near the Fermi level ( $E_F$ ) [18–20]. The mIR peak in this model results from optical excitations between the two bands [6]. Its peak energy is given as  $E_{\text{mIR}} \simeq 2\tilde{V}$ , where  $\tilde{V}$  is the  $c$ - $f$  hybridization renormalized by large  $U$  and expressed as

$$\tilde{V} \simeq \sqrt{T_K W}, \quad (1)$$

where  $T_K$  and  $W$  indicate the Kondo temperature and  $c$  bandwidth, respectively [18–20]. Measured  $E_{\text{mIR}}$  values of

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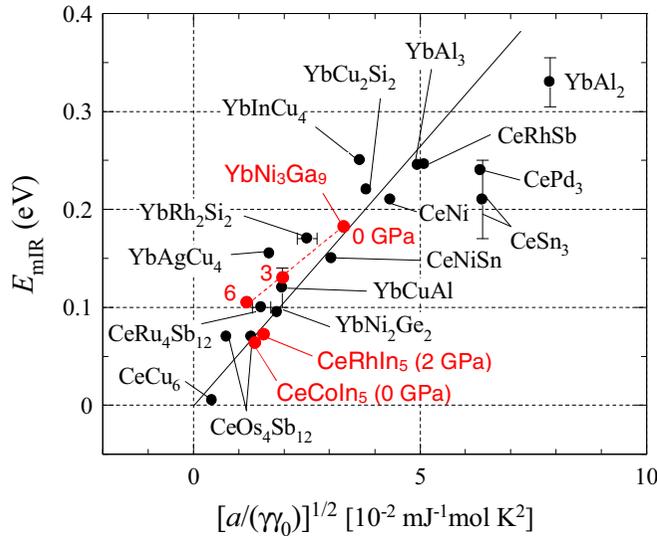


FIG. 1. A universal relation between the optical conductivity and the  $c$ - $f$  hybridization energy observed for Ce and Yb compounds (the red data points have been added here, while the others have been reproduced from Ref. [10]). Here, the mid-IR peak energy ( $E_{\text{mIR}}$ ) measured for these compounds are plotted as a function of their  $\sqrt{a/(\gamma\gamma_0)}$ , where  $\gamma$  and  $\gamma_0$  are the specific heat coefficients of the Ce (Yb) and La (Lu) compounds, respectively, and  $a$  is an  $f$ -degeneracy dependent constant [10]. Here,  $\sqrt{a/(\gamma\gamma_0)}$  is a measure of the  $c$ - $f$  hybridization energy  $\tilde{V}$ , through the relation  $2\tilde{V} \simeq \sqrt{T_K W} \propto \sqrt{(a/\gamma)(1/\gamma_0)}$ . The solid line is a guide to the eye.

different IV metals have been compared with their  $\sqrt{T_K W}$  (or related quantities) estimated by other experiments, and a universal relation between  $E_{\text{mIR}}$  and  $\sqrt{T_K W}$  has been found over a variety of Ce and Yb compounds [8–11]. An example of such a universal relation [10] is shown in Fig. 1. This universal relation may be regarded as an optical analog to the well known Kadowaki-Woods relation [21,22]. More detailed analyses including effects of  $f$  level degeneracy and/or the band structure have also been reported [12–16]. These studies suggest that the  $c$ - $f$  hybridized band model is an oversimplification for the actual IV metals. For example, it has been suggested that, for Ce compounds,  $E_{\text{mIR}}$  should correspond to the energy separation from the  $c$ - $f$  hybridized band below  $E_F$  to the bare  $f$  states above  $E_F$  [16]. In fact, the observed  $E_{\text{mIR}}$  values of some IV compounds seem too large to result between the  $c$ - $f$  hybridized bands, and such a model may offer a useful alternative to the  $c$ - $f$  hybridized band model. Nevertheless, it is still true that the mIR peak energy roughly scales with  $\sqrt{T_K W}$  over many IV metals [8–11]. Clearly, the characteristics of the mIR peak involve the Kondo physics and are not due to accidental band structures. Furthermore, effects of momentum-dependent  $c$ - $f$  hybridization have been considered in analyzing  $\sigma(\omega)$  of Ce compounds [17].

Note that both Ce- and Yb-based IV metals seem to follow the same universal relation [10,11], as seen in Fig. 1. For  $\text{Ce}^{3+}$  and  $\text{Yb}^{3+}$  ions, their respective  $f^1$  and  $f^{13}$  configurations have an electron-hole ( $e$ - $h$ ) symmetry, since  $f^{13}$  is equivalent to  $h^1$ . It has been an important question as to what degree this  $e$ - $h$  symmetry is reflected on the properties of Ce and Yb compounds. An example of common property between them,

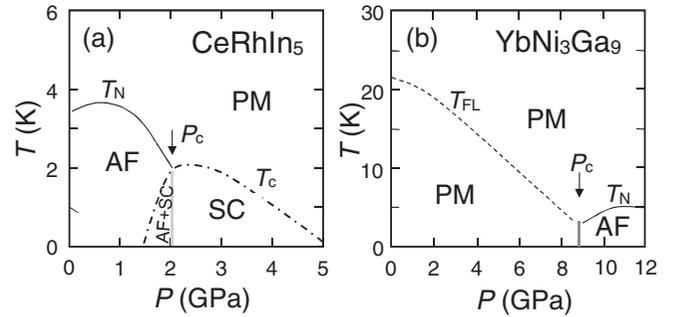


FIG. 2. Schematic phase diagrams of (a)  $\text{CeRhIn}_5$  (after Ref. [32]) and (b)  $\text{YbNi}_3\text{Ga}_9$  (after Ref. [36]) as functions of temperature ( $T$ ) and external pressure ( $P$ ). AF: antiferromagnetic, PM: paramagnetic, SC: superconducting,  $T_N$ : Neel temperature,  $T_c$ : superconducting transition temperature,  $T_{FL}$ : the temperature below which Fermi liquid characteristics are observed,  $P_c$ : critical pressure where AF ordering appears or disappears.

which is consistent with the  $e$ - $h$  symmetry, is the formation of HF state with large effective mass. However, Ce and Yb compounds also exhibit noted differences [23,24]. A useful experimental technique to examine the  $e$ - $h$  symmetry is the application of an external pressure ( $P$ ) [23–26]. Since  $\text{Ce}^{4+}$  ( $f^0$ ) and  $\text{Yb}^{3+}$  ( $f^{13}$ ) ions have smaller ionic radii than  $\text{Ce}^{3+}$  ( $f^1$ ) and  $\text{Yb}^{2+}$  ( $f^{14}$ ) ions, respectively, an applied  $P$  generally increases the average valence ( $v$ ) of Ce toward 4 and that of Yb toward 3. For both Ce and Yb cases,  $P$  should also increase the bare (unrenormalized)  $c$ - $f$  hybridization, since a reduced interatomic distance should increase the overlap between the  $c$  and  $f$  wave functions. For Ce compounds, an increase of  $c$ - $f$  hybridization with  $P$  has been observed, for example, by an increase in  $T_K$  [23]. Then, in Fig. 1, upon applying  $P$ , a Ce compound should shift to the upper right. For Yb compounds, in addition to  $v$  increases, effective mass increases and magnetic order have been found at high  $P$  [23,24,26]. Namely, Yb compounds seem to exhibit more localized  $f$  electron states at high  $P$ . This suggests a reduced  $T_K$ , and hence a reduced  $\tilde{V}$  from Eq. (1), although the bare hybridization in a Yb compound should increase with  $P$  as explained above. Therefore, it is intriguing how a Yb compound should shift with  $P$  in Fig. 1.

In this work, we have addressed the above questions by studying the  $\sigma(\omega)$  of  $\text{CeRhIn}_5$  and  $\text{YbNi}_3\text{Ga}_9$  at  $P$  to 10 GPa and at temperatures ( $T$ ) to 6 K. These compounds have attracted much attention for their remarkable properties at high  $P$ , as summarized in Fig. 2.  $\text{CeRhIn}_5$  exhibits an antiferromagnetism (AF) at  $P = 0$  with a Neel temperature ( $T_N$ ) of 3.8 K and an electronic specific heat coefficient of  $\gamma = 420 \text{ mJ/K}^2 \text{ mol}$  above  $T_N$  [27]. With increasing  $P$ , the AF is gradually suppressed, and near a critical pressure ( $P_c$ ) of  $\sim 2$  GPa, a superconductivity with a transition temperature ( $T_c$ ) of 2.6 K is observed [27,28]. Around  $P_c$ , various anomalous properties related with QCP have been observed [29–32].  $\sigma(\omega)$  of  $\text{CeRhIn}_5$  at ambient  $P$  has already been measured and analyzed in detail [17,33], but  $\sigma(\omega)$  at high  $P$  has not been explored yet.  $\text{YbNi}_3\text{Ga}_9$ , in contrast, is a paramagnetic IV compound at  $P = 0$  with  $\gamma = 30 \text{ mJ/K}^2 \text{ mol}$ , indicating a strong  $c$ - $f$  hybridization [34,35]. With increasing  $P$ , the

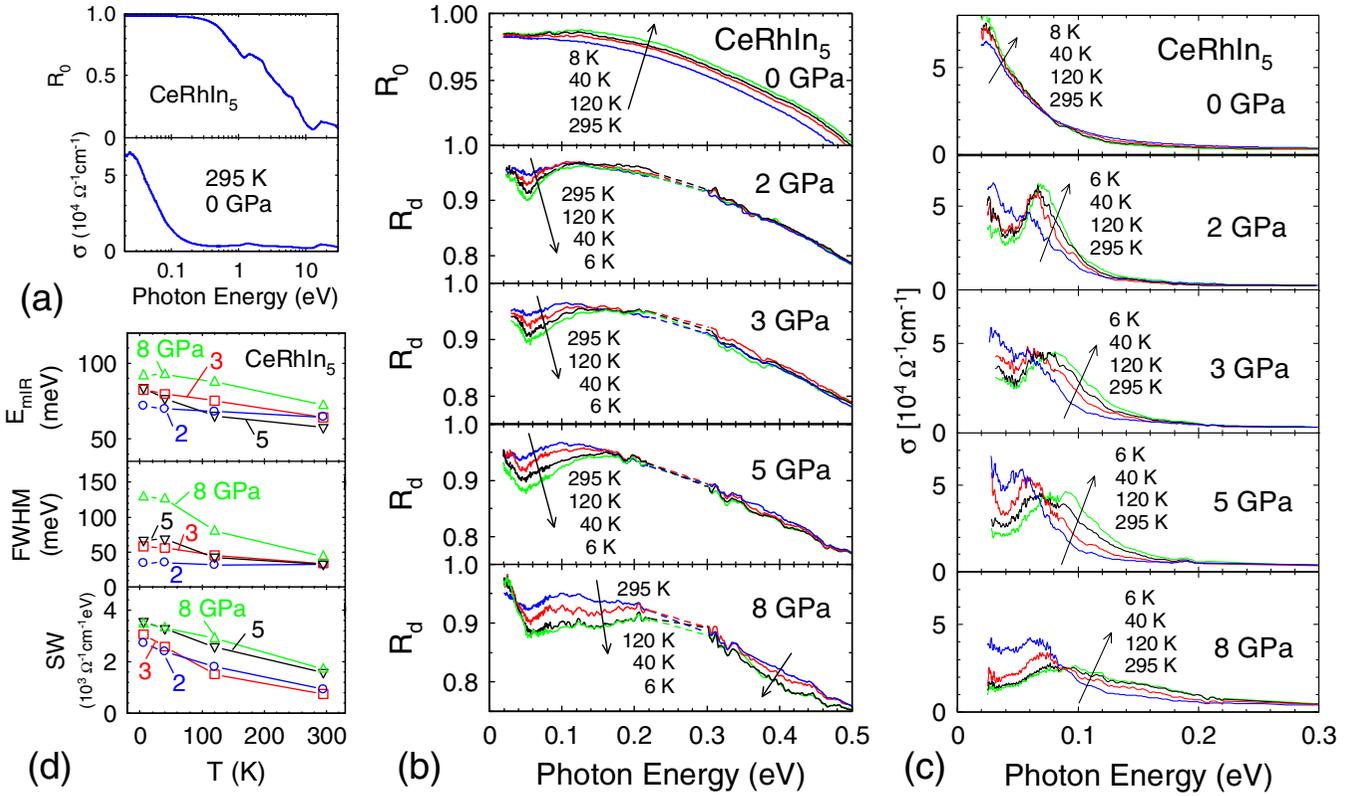


FIG. 3. (a)  $R(\omega)$  and  $\sigma(\omega)$  spectra of CeRhIn<sub>5</sub> measured at  $T = 295$  K and  $P = 0$ . (b)  $R(\omega)$  at  $P = 0, 2, 3, 5,$  and  $8$  GPa and at  $T = 295$  K (blue curves),  $120$  K (red),  $40$  K (black), and  $8$  K ( $0$  GPa) or  $6$  K ( $2$ – $8$  GPa) (green).  $R_0$  and  $R_d$  denote  $R(\omega)$  measured at sample/vacuum and sample/diamond interfaces, respectively. The broken-curve portions of the spectra indicate interpolations, which were needed because of strong absorption by the diamond [49]. (c)  $\sigma(\omega)$  at the same values of  $P$  and  $T$  as in (b). (d) Peak position ( $E_{\text{mIR}}$ ), the full width at half maximum (FWHM), and the spectral weight (SW) of the mIR peak in  $\sigma(\omega)$ , given by the spectral fitting.

measured  $v$  increases from  $2.6$  at  $P = 0$  to  $2.88$  at  $P = 16$  GPa, and an AF state appears above  $P_c \simeq 9$  GPa [36]. In addition,  $\gamma$  increases significantly with  $P$ , reaching  $\gamma = 1 \text{ J/K}^2 \text{ mol}$  at  $9$  GPa [37]. Namely, with increasing  $P$ , CeRhIn<sub>5</sub> shows a crossover from localized to delocalized electronic states, while YbNi<sub>3</sub>Ga<sub>9</sub> shows that from delocalized to localized ones. Although the lowest  $T$ 's in our study,  $6$  K for CeRhIn<sub>5</sub> and  $8$  K for YbNi<sub>3</sub>Ga<sub>9</sub>, are above  $T_c$  and  $T_N$ , our study should still provide important information about the  $P$  tuning of the underlying  $c$ - $f$  hybridized state behind the QCP-related properties below  $T_c$  and  $T_N$ . The mIR peaks of CeRhIn<sub>5</sub> and YbNi<sub>3</sub>Ga<sub>9</sub> have indeed shown quite contrasting  $P$  evolution, which is discussed in terms of the  $c$ - $f$  hybridized electronic states at high  $P$  and in terms of the  $e$ - $h$  symmetry.

## II. EXPERIMENTAL

The samples of CeRhIn<sub>5</sub> and YbNi<sub>3</sub>Ga<sub>9</sub> used were single crystals grown with the self-flux method. The reflectance spectrum [ $R(\omega)$ ] of a sample was measured on an as-grown surface without polishing.  $\sigma(\omega)$  was derived from  $R(\omega)$  using the Kramers-Kronig (KK) analysis [38].  $R(\omega)$  at  $P = 0$  was measured at photon energies between  $15$  meV and  $30$  eV covered by several light sources [39], including the vacuum uv synchrotron radiation at the beamline BL7B of the UVSOR Facility [40].  $R(\omega)$  spectra at high  $P$  were measured using a diamond anvil cell (DAC) [41]. Type IIa diamond anvils

with  $0.8$  mm culet diameter and a stainless steel gasket were used to seal the sample with glycerin as the pressure transmitting medium [42–44]. A flat surface of a sample was closely attached on the culet surface of the diamond anvil, and  $R(\omega)$  at the sample/diamond interface was measured. Small ruby pieces were also sealed to monitor the pressure via its fluorescence. A gold film was placed between the gasket and anvil as a reference of  $R(\omega)$ . In the KK analysis of  $R(\omega)$  measured with DAC, the refractive index of diamond ( $n_d = 2.4$ ) was taken into account as previously discussed [45].  $R(\omega)$  at high  $P$  and low  $T$  were measured at photon energies from  $25$  meV (CeRhIn<sub>5</sub>) or  $20$  meV (YbNi<sub>3</sub>Ga<sub>9</sub>) to  $1.1$  eV, using synchrotron radiation as a bright IR source [46] at the beamline BL43IR of SPring-8 [47,48]. Below the measured energy range,  $R(\omega)$  was extrapolated with the Hagen-Rubens function [38]. More details of the high pressure IR experiments can be found elsewhere [41].

## III. RESULTS AND DISCUSSIONS

### A. $R(\omega)$ and $\sigma(\omega)$ of CeRhIn<sub>5</sub> at high pressures

Figure 3(a) shows  $R(\omega)$  and  $\sigma(\omega)$  of CeRhIn<sub>5</sub> at  $P = 0$  and  $T = 295$  K over the entire measured spectral range.  $R(\omega)$  below  $0.3$  eV is very high, which indicates highly metallic characteristics of CeRhIn<sub>5</sub>.  $\sigma(\omega)$  has a Drude component rising toward zero energy, which is due to free carrier dynamics. Figures 3(b) and 3(c) show  $R(\omega)$  and  $\sigma(\omega)$  measured

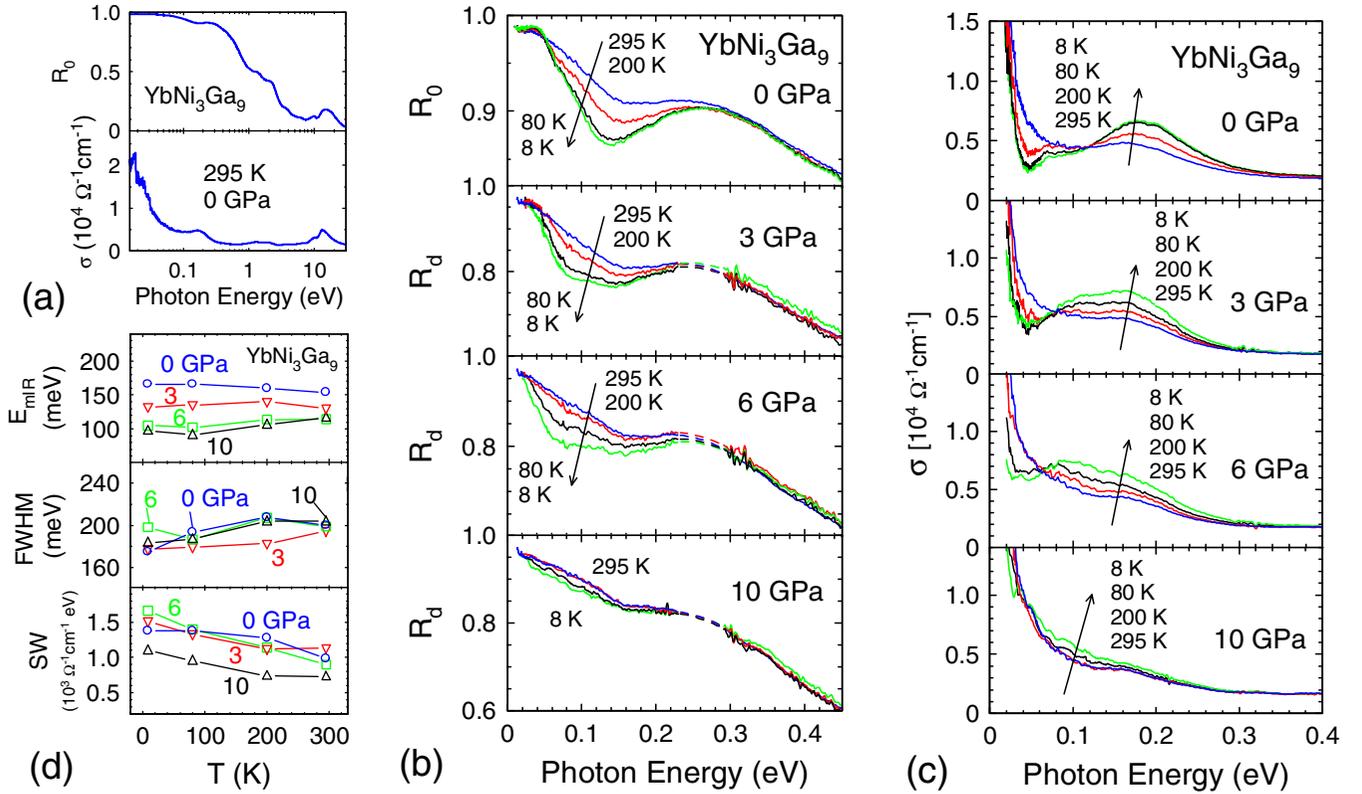


FIG. 4. (a)  $R(\omega)$  and  $\sigma(\omega)$  spectra of  $\text{YbNi}_3\text{Ga}_9$  measured at  $T = 295$  K and  $P = 0$ . (b)  $R(\omega)$  spectra at  $P = 0, 3, 6,$  and  $10$  GPa and at  $T = 295$  K (blue curves),  $200$  K (red),  $80$  K (black), and  $8$  K (green). The broken curve portions indicate smooth interpolations, as already mentioned in the caption of Fig. 3(b) [49]. (c)  $\sigma(\omega)$  spectra at the same values of  $P$  and  $T$  as those in (b). (d) Results of the spectral fitting, with the same notations as those in Fig. 3(d).

at different values of  $P$  and  $T$ . At  $P = 0$ ,  $R(\omega)$  and  $\sigma(\omega)$  have only minor  $T$  dependences, which is consistent with the previous report [33]. With increasing  $P$ , as shown in Figs. 3(b) and 3(c), a dip appears and develops in  $R(\omega)$ , and a pronounced mIR peak develops in  $\sigma(\omega)$ . At 2 GPa, the mIR peak is barely visible at 295 K but becomes very pronounced with cooling to 6 K. As discussed in the Introduction, such a mIR peak is a hallmark of the  $c$ - $f$  hybridized state in Ce compounds. Namely,  $\text{CeRhIn}_5$  at 2 GPa has much stronger  $c$ - $f$  hybridization than at 0 GPa. Note that the  $T$  evolution of the mIR peak at  $P = 2$  GPa is strikingly similar to that of  $\text{CeCoIn}_5$  at  $P = 0$ , with very close  $E_{\text{mIR}}$  [33,50,51].  $\text{CeCoIn}_5$  at  $P = 0$  is also a superconductor with almost the same  $T_c$  as that of  $\text{CeRhIn}_5$  at 2 GPa [50]. These similarities between  $\text{CeCoIn}_5$  at 0 GPa and  $\text{CeRhIn}_5$  at 2 GPa indicate that their  $c$ - $f$  hybridized electronic states are also similar. In Fig. 1,  $E_{\text{mIR}}$  of  $\text{CeCoIn}_5$  at 0 GPa and  $\text{CeRhIn}_5$  at 2 GPa have been added using their  $\gamma$  and  $\gamma_0$  data [52]. Clearly, they follow the universal relation well, and their plots are indeed close to each other reflecting their similarity. At  $P = 3$  GPa, the mIR peak of  $\text{CeRhIn}_5$  is broader than that at 2 GPa. This broadening of the mIR peak should basically indicate a broadening of the  $f$  band and hence a stronger  $c$ - $f$  hybridization. From the evolution of  $\sigma(\omega)$  from  $P = 0$  to 3 GPa, it is clear that the electronic structure of  $\text{CeRhIn}_5$  in the normal state at 6 K changes significantly from very weakly to moderately hybridized ones, which should be an important basis for the QCP observed below 2 K. From 3 to 8 GPa, the mIR peak

becomes apparently much broader, and its spectral weight (SW) shifts toward higher energy. In addition, at 5 and 8 GPa the mIR peak is clearly observed even at room  $T$ , which is a feature often observed for IV Ce compounds. Namely,  $\text{CeRhIn}_5$  above 5 GPa is likely a strongly hybridized IV compound. Unfortunately,  $\gamma$  data of  $\text{CeRhIn}_5$  above 2 GPa are unavailable, so  $E_{\text{mIR}}$  above 2 GPa cannot be plotted in Fig. 1. However, it is almost certain that the plot for  $\text{CeRhIn}_5$  shifts to the upper right with  $P$ , since the resistivity data [28] strongly suggest that the hybridization is much stronger at 8 GPa.

To analyze the evolution of mIR peak more quantitatively, we have performed spectral fitting on the measured  $\sigma(\omega)$  using the Drude-Lorentz oscillator model [38]. Details of the fitting procedures and examples of the fitted spectra are given in the Appendix. In Fig. 3(d), the  $E_{\text{mIR}}$ , full width at half maximum (FWHM), and the SW of the mIR peak given by the fitting are summarized. The fitting results in Fig. 3(d) confirm the features discussed above, namely the  $P$ -induced increases in  $E_{\text{mIR}}$ , SW, and the width. However, they also reveal that the increases in  $E_{\text{mIR}}$  and SW are at most about 30%. In contrast, the increase in the width at 6 K is particularly large from 5 to 8 GPa, suggesting that the  $f$  electron bandwidth should rapidly increase at this  $P$  range.

### B. $R(\omega)$ and $\sigma(\omega)$ of $\text{YbNi}_3\text{Ga}_9$ at high pressures

Figure 4(a) shows measured  $R(\omega)$  and  $\sigma(\omega)$  of  $\text{YbNi}_3\text{Ga}_9$  over a wide energy range at  $P = 0$  and  $T = 295$  K, and

Figures 4(b) and 4(c) show those below 0.4 eV at different values of  $P$  and  $T$ . At  $P = 0$ , a dip is observed in  $R(\omega)$  at 0.1–0.2 eV range and a mIR peak in  $\sigma(\omega)$  at 0.1–0.3 eV range, both of which are strongly  $T$  dependent. A Drude component is also observed in  $\sigma(\omega)$  below 0.1 eV, which rises steeply toward zero energy. With cooling, the dip becomes deeper, and the mIR peak becomes more pronounced and slightly shifts to higher energy. In addition, a shoulder appears in  $\sigma(\omega)$  near 70 meV at low  $T$ . Furthermore, the onset of Drude component becomes extremely sharp at low  $T$ . This is because  $\sigma(0)$  at low  $T$  is very large, exceeding  $1 \times 10^6 \Omega^{-1} \text{cm}^{-1}$  at 8 K [34,36]. This extremely narrow Drude component is due to the Drude response of heavy quasiparticles formed at low  $T$  [5]. These features are qualitatively very similar to those previously observed for other Yb-based IV metals such as  $\text{YbAl}_3$  [55]. Using the observed  $E_{\text{mIR}} = 0.18 \text{ eV}$  at 8 K with  $\gamma = 30 \text{ mJ/K}^2 \text{mol}$  and  $\gamma_0 = 6.3 \text{ mJ/K}^2 \text{mol}$  [34], a plot for  $\text{YbNi}_3\text{Ga}_9$  at  $P = 0$  has been added in Fig. 1. Clearly,  $\text{YbNi}_3\text{Ga}_9$  well follows the universal relation.

The  $\sigma(\omega)$  spectra at various values of  $T$  and  $P$  have been analyzed by spectral fitting, similarly to those of  $\text{CeRhIn}_5$ . The results are summarized in Fig. 4(d), and examples of the fitting are given in the Appendix. At 3 GPa,  $R(\omega)$  and  $\sigma(\omega)$  spectra are still strongly  $T$  dependent. Note that, at low  $T$ , the mIR peak seems to consist of two peaks, located at  $\sim 0.1$  and  $\sim 0.17 \text{ eV}$ . A similar two-peak feature is also observed at 6 GPa. The origins for the two peaks are unclear, hence we define  $E_{\text{mIR}}$  as the center-of-mass position of the Lorentz oscillators used to fit the mIR peak. As indicated in Fig. 4(d), the obtained  $E_{\text{mIR}}$  decreases with  $P$  from 0 to 3 GPa and also from 3 to 6 GPa. The  $E_{\text{mIR}}$ 's at  $P = 3$  and 6 GPa and  $T = 8 \text{ K}$  given by the spectral fitting have been added in Fig. 1, using the  $\gamma$  data measured at high  $P$  [37,56]. The plot for  $\text{YbNi}_3\text{Ga}_9$  shifts to the lower left with  $P$ , namely, it actually shifts in an opposite manner to that of Ce compounds. At 10 GPa, the mIR peak seems much weaker than at 6 GPa and almost merged with the Drude component. The remaining mIR component has been evaluated by the fitting as in Fig. 4(d), which indicates that  $E_{\text{mIR}}$  further decreases compared with that at 6 GPa. From Figs. 4(b) and 4(c), the  $T$  variations of  $R(\omega)$  and  $\sigma(\omega)$  at 10 GPa are much smaller than those at 6 GPa. This strongly suggests that the  $T$ -dependent hybridization has become much weaker with  $P$  from 6 to 10 GPa. This is reasonable since the  $f$  electron state should be more localized and less hybridized above  $P_c \sim 9 \text{ GPa}$ , where an AF state appears below  $T_N = 5 \text{ K}$ . The fitting results in Fig. 4(d) also indicate that  $E_{\text{mIR}}$  does not change much with  $T$ , although it clearly decreases with  $P$ . In addition, the peak width seems to exhibit no systematic changes with  $T$  and  $P$ .

### C. Comparison of pressure evolution between $\text{CeRhIn}_5$ and $\text{YbNi}_3\text{Ga}_9$

To compare more clearly the observed  $P$  evolution of  $\sigma(\omega)$  between  $\text{CeRhIn}_5$  and  $\text{YbNi}_3\text{Ga}_9$ , the  $\sigma(\omega)$  spectra at the lowest measured  $T$  are displayed in Fig. 5. Clearly, the  $P$  evolution of  $\text{CeRhIn}_5$  and  $\text{YbNi}_3\text{Ga}_9$  has some contrasting and opposite tendencies: With increasing  $P$ , the mIR peak of  $\text{CeRhIn}_5$  appears and grows and shifts to higher energy.

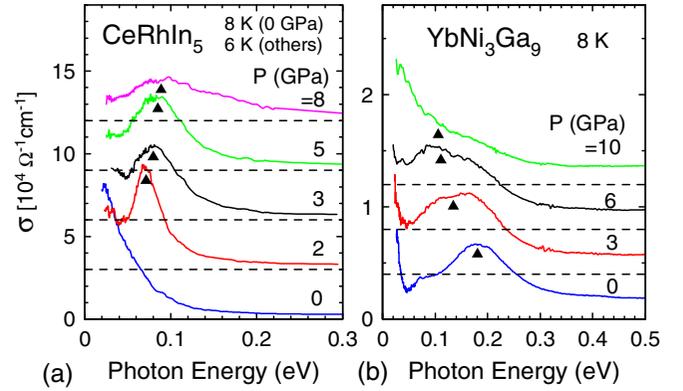


FIG. 5. Comparison of  $P$  evolution of  $\sigma(\omega)$  at low  $T$  between (a)  $\text{CeRhIn}_5$  and (b)  $\text{YbNi}_3\text{Ga}_9$ . The spectra are vertically offset for clarity, and the triangles indicate the  $E_{\text{mIR}}$  values.

In contrast, the mIR peak of  $\text{YbNi}_3\text{Ga}_9$  is well developed already at  $P = 0$  and shifts to lower energy with  $P$  and becomes diminished at 10 GPa. On the other hand, not all the  $P$  evolution of the mIR peak exhibits opposite tendencies between them. For example, the mIR peak width of  $\text{CeRhIn}_5$  significantly increases with  $P$ , but that of  $\text{YbNi}_3\text{Ga}_9$  does not exhibit a narrowing or any other systematic change, as indicated in Fig. 6(b). In addition, the  $P$ -induced shift of  $E_{\text{mIR}}$  seems much larger for  $\text{YbNi}_3\text{Ga}_9$  than for  $\text{CeRhIn}_5$ . Below, we consider these results in terms of the  $P$  evolution of IV states in Ce and Yb compounds.

In a Ce compound with  $v = 3$  and completely localized  $f^1$  state,  $\sigma(\omega)$  should consist only of a Drude component due to  $c$  electrons, since the system is a metal where the localized  $f$  electrons do not contribute to the Drude response. This applies well to  $\text{CeRhIn}_5$  at 0 GPa, since its  $\sigma(\omega)$  has only a Drude component in Fig. 3(c) and its  $v$  should be very close to 3. As stated in the introduction, an external  $P$  on a Ce compound should increase  $v$  from 3. In this IV state,  $\sigma(\omega)$  would exhibit a mIR peak due to the hybridized state, as actually observed in  $\sigma(\omega)$  of  $\text{CeRhIn}_5$  at 2 and 3 GPa. With further increasing  $P$ , both the energy and width of the mIR peak should increase, since the hybridization and  $f$  bandwidth increase with  $P$ . This is again consistent with the observed  $\sigma(\omega)$  at 3–8 GPa. Namely, the observed  $P$  evolution of  $\sigma(\omega)$  for  $\text{CeRhIn}_5$  seems quite consistent with that expected for a Ce compound.  $P$ -induced higher-energy shifts and development of an IR peak in  $\sigma(\omega)$  have also been observed for  $\text{CeRu}_4\text{Sb}_{12}$  (Ref. [57]) and  $\text{CeIn}_3$  [58].

As for Yb compounds, as stated in the introduction, an ionic radius consideration suggests that  $v$  of an IV Yb compound should increase toward 3 with  $P$ . In the limit of exactly  $v = 3$  state with the completely localized  $f^{13}$  state,  $\sigma(\omega)$  would consist only of a broad Drude component due to  $c$  electrons, similarly to the  $f^1$  ( $\text{Ce}^{3+}$ ) case. Namely, the main feature in  $\sigma(\omega)$  of an IV Yb compound should evolve from a mIR peak at  $P = 0$  into a broad Drude component in the limit of very high  $P$ . Clearly, such a  $P$  evolution is qualitatively consistent with that observed for  $\text{YbNi}_3\text{Ga}_9$  in Fig. 5(b):  $\sigma(\omega)$  has a well developed mIR peak at  $P = 0$ , which shifts to lower energy with  $P$  and becomes much weaker at 10 GPa.  $\sigma(\omega)$  at 10 GPa actually looks like a broad Drude component, and

the residual mIR peak SW would become even weaker if  $P$  is further increased since  $v$  still increases from 2.84 at 10 GPa to 2.88 at 16 GPa [36].

The discussions above indicate that the opposite tendencies in the  $P$  evolution of  $\sigma(\omega)$  between CeRhIn<sub>5</sub> and YbNi<sub>3</sub>Ga<sub>9</sub>, including the opposite  $P$ -induced shifts in Fig. 1, are consistent with the consideration of ionic radius and  $e$ - $h$  symmetry under high  $P$ . The expression for the renormalized hybridization  $\tilde{V}$  in Eq. (1) has been derived for an  $f^1$  (Ce<sup>3+</sup>) system [18–20] but is also valid for an  $h^1$  ( $f^{13}$ , Yb<sup>3+</sup>) system under the  $e$ - $h$  symmetry. Hence, for both Ce and Yb compounds,  $\tilde{V}$  and  $E_{\text{mIR}}$  should become smaller with increasing localization and decreasing  $T_K$ . This has been actually demonstrated by the Ce and Yb compounds plotted in Fig. 1. Then, according to Eq. (1), the  $P$ -induced decrease of  $E_{\text{mIR}}$  for YbNi<sub>3</sub>Ga<sub>9</sub> indicates that  $\tilde{V}$  decreases with  $P$ , although the bare (unrenormalized) hybridization should increase with  $P$  as already discussed. This peculiar property of a Yb compound has been discussed [59] in terms of the  $c$ - $f$  exchange energy  $J_{cf}$ , expressed as [60]

$$J_{cf} \simeq \frac{|V|^2}{|E_F - \epsilon_f|}, \quad (2)$$

where  $V$  is the bare  $c$ - $f$  hybridization averaged over  $k$  space and  $\epsilon_f$  is the one-electron (unrenormalized)  $f$  level. Equation (2) has been derived for an  $f^1$  system with sufficiently large  $U$  but is also valid for the  $h^1$  system under the  $e$ - $h$  symmetry.  $J_{cf}$  is related with  $T_K$  as  $T_K \simeq W \exp(-W/J_{cf})$ . Note that  $|V|$  in Eq. (2) increases with  $P$  for both Ce and Yb cases, as discussed earlier. In addition, note that  $\epsilon_f$  should increase with  $P$  relative to  $E_F$  [59,61]. For the Ce case,  $\epsilon_f$  is located below  $E_F$  and approaches  $E_F$  with increasing  $P$ . Hence  $|E_F - \epsilon_f|$  decreases in Eq. (2), so that  $J_{cf}$  increases with  $P$ . For the Yb case, in contrast,  $\epsilon_f$  is the  $f$  hole level located above  $E_F$  and shifts away from  $E_F$  with  $P$ . Hence  $|E_F - \epsilon_f|$  increases with  $P$  in Eq. (2), so that  $J_{cf}$  may either increase or decrease depending on which of  $|E_F - \epsilon_f|$  and  $|V|^2$  increases more. Therefore, the  $P$ -induced decrease of  $E_{\text{mIR}}$  for YbNi<sub>3</sub>Ga<sub>9</sub> suggests that  $|E_F - \epsilon_f|$  increases with  $P$  more than  $|V|^2$  does.

As already mentioned, some of the observed  $P$  evolution of  $\sigma(\omega)$  are not opposite or symmetrical between CeRhIn<sub>5</sub> and YbNi<sub>3</sub>Ga<sub>9</sub>. For example, the mIR peak of CeRhIn<sub>5</sub> shows progressive and significant broadenings with  $P$ , while that of YbNi<sub>3</sub>Ga<sub>9</sub> does not show a narrowing or any systematic change with  $P$ . In addition,  $P$ -induced shift of  $E_{\text{mIR}}$  seems much larger for YbNi<sub>3</sub>Ga<sub>9</sub> (0.18 to 0.1 eV on going from 0 to 6 GPa) than for CeRhIn<sub>5</sub> (70 to 90 meV on going from 2 to 8 GPa). Microscopic mechanisms for these results are unclear, but they should involve microscopic differences between Ce<sup>3+</sup> and Yb<sup>3+</sup> not considered in the simple  $e$ - $h$  symmetry argument. For example, the  $4f$  orbital of Yb<sup>3+</sup> is much more localized than that of Ce<sup>3+</sup>, leading to a much smaller  $f$  bandwidth and  $|V|$  for Yb<sup>3+</sup> [23,24]. In addition, the spin-orbit splitting of Yb<sup>3+</sup> ( $\sim 1.3$  eV) is much larger than that of Ce<sup>3+</sup> ( $\sim 0.3$  eV). It has been pointed out [23,24] that, due to these differences, the  $P$ -induced variation of  $v$  from 3.0 in a Ce compound should be at most to  $\sim 3.16$ , while that in a Yb compound can be changed more widely between 2 and 3. Experimentally,  $v$  of CeRhIn<sub>5</sub> at high  $P$  has not been reported but that of CeCoIn<sub>5</sub> has been reported

to vary from 3.00 at  $P = 0$  to 3.05 at 8 GPa [62]. On the other hand,  $v$  of YbNi<sub>3</sub>Ga<sub>9</sub> varies from 2.60 at  $P = 0$  to 2.84 at  $P = 10$  GPa [36]. These different ranges of variation in  $v$  may be related to the much larger  $P$ -induced shifts of  $E_{\text{mIR}}$  for YbNi<sub>3</sub>Ga<sub>9</sub>, since  $v$  is closely related with  $T_K$  and  $\tilde{V}$ . To further understand  $P$  evolution of  $\sigma(\omega)$  and electronic structures for Yb compounds, more studies on other Yb-based IV compounds are clearly needed. For example, YbCu<sub>2</sub>Ge<sub>2</sub> (Ref. [63]) and YbAl<sub>2</sub> (Ref. [64]) are other examples that exhibit large  $P$  dependences in their physical properties.  $\sigma(\omega)$  studies of these compounds at high  $P$  are in progress [65].

#### IV. SUMMARY

Optical conductivity studies of CeRhIn<sub>5</sub> and YbNi<sub>3</sub>Ga<sub>9</sub> at high  $P$  have been performed to probe the  $P$  evolutions of their  $c$ - $f$  hybridized electronic structures. The main feature in the measured  $\sigma(\omega)$  is a mIR peak, which has exhibited many opposite or symmetrical  $P$  evolution between CeRhIn<sub>5</sub> and YbNi<sub>3</sub>Ga<sub>9</sub>: With increasing  $P$ , the mIR peak develops and shifts to higher energy for CeRhIn<sub>5</sub>, while it shifts to lower energy and becomes diminished at high  $P$  for YbNi<sub>3</sub>Ga<sub>9</sub>. These results are qualitatively consistent with the  $e$ - $h$  symmetry and  $P$ -induced variations in the Ce and Yb ionic radii. However, YbNi<sub>3</sub>Ga<sub>9</sub> has also exhibited  $P$  evolution of mIR peak not opposite to those of CeRhIn<sub>5</sub>, which are likely due to microscopic differences between Ce and Yb not included in the simple  $e$ - $h$  symmetry arguments.

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#### APPENDIX: SPECTRAL FITTINGS ON $\sigma(\omega)$

The spectral fittings were performed using the Drude-Lorentz model [38]. In this model, the complex dielectric function is expressed as a sum of Drude and Lorentz oscillators, which represent free and bound electrons, respectively, as

$$\hat{\epsilon}(\omega) = \epsilon_\infty + \sum_j \frac{\omega_{p,j}^2}{\omega_{0,j}^2 - \omega^2 - i\omega\gamma_j}. \quad (A1)$$

Here,  $\omega_p$ ,  $\omega_0$ , and  $\gamma$  are the plasma frequency, natural frequency, and damping, respectively.  $j$  denotes the  $j$ th oscillator, and  $\omega_0 = 0$  for a Drude oscillator.  $\epsilon_\infty$  represents contribution from higher-lying interband transitions. In the fitting, these parameters are adjusted so as to reproduce a measured  $\sigma(\omega)$  through the relation  $\sigma(\omega) = (\omega/4\pi)\text{Im}(\hat{\epsilon})$ .

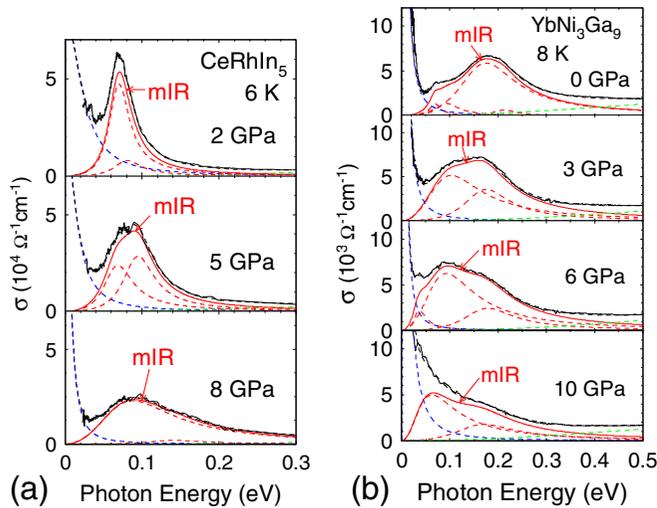


FIG. 6. (a) Examples of Drude-Lorentz fitting on  $\sigma(\omega)$  of CeRhIn<sub>5</sub> measured at  $T = 6$  K and at  $P = 2, 5,$  and  $8$  GPa, and (b) those of YbNi<sub>3</sub>Ga<sub>9</sub> measured at  $8$  K at  $P = 0, 3, 6,$  and  $10$  GPa, as discussed in the Appendix. Shown in each graph are the measured data (black solid curves), total fit (black broken), Drude component (blue broken), two Lorentz oscillators (red broken), mIR peak (red solid) which is the sum of the Lorentz oscillators, and the background (green broken).

Figure 6(a) shows examples of fitting for  $\sigma(\omega)$  of CeRhIn<sub>5</sub> at  $6$  K. The measured  $\sigma(\omega)$  spectra can be reproduced well by using two Lorentz oscillators for the mIR peak, in addition to a Drude oscillator and a broad Lorentz oscillator peaked at  $0.45$  eV serving as a background.  $\epsilon_\infty = 5$  is used for all the fitting, and the fitted mIR peak is the sum of the two Lorentz oscillators. Although not shown,  $\sigma(\omega)$  at other values of  $P$  and  $T$  can be fitted similarly. Note that two Lorentz oscillators are used simply because a single Lorentz oscillator is not sufficient to fit the mIR peak and that the two Lorentz

oscillators are not assigned to any specific origins. In addition, due to the use of DAC, the measured spectral range does not cover low-enough energies for fitting the Drude component. However, since our main focus here is the evolution of the mIR peak with  $P$  and  $T$ , the uncertainty regarding the Drude peak fitting is not a serious problem. To reduce the uncertainty in fitting the Drude component,  $\sigma(0)$  value given by the fitting was kept in the range  $8\text{--}20 \times 10^4 \Omega^{-1} \text{cm}^{-1}$ , and  $\sigma(0)$  was increased with decreasing  $T$ . These constraints on the Drude component are implied from the  $\sigma(0)$  at  $P = 0$  [Fig. 3(c)] and also from the measured dc conductivities [27].

Figure 6(b) shows examples of fitting for YbNi<sub>3</sub>Ga<sub>9</sub>. In some cases more than two Lorentz oscillators are needed to fit the mIR peak reasonably well. A background Lorentz oscillator at  $0.75$  eV and  $\epsilon_\infty = 5$  are used. Here, the fitting parameters are chosen so that  $\sigma(0)$  at each data matched the measured dc conductivity at the same  $T$  and  $P$  [34,36,66]. This procedure greatly reduced the uncertainty in fitting the Drude component. At  $0\text{--}6$  GPa, as mentioned in Sec. III B, the Drude component is extremely narrow due to the large values of  $\sigma(0)$ . At  $10$  GPa, the mIR peak is not well resolved from the Drude component any more, but the fitting was nevertheless performed to evaluate the remaining mIR component, as shown in Fig. 6(b). Again, measured  $\sigma(0)$  [36], for example  $4.5 \times 10^4 \Omega^{-1} \text{cm}^{-1}$  at  $P = 10$  GPa and  $T = 8$  K, were used to reduce the uncertainty. The fitting results suggest that the SW of the mIR peak at  $10$  GPa is still sizable but is indeed much smaller than that at  $6$  GPa, as discussed in the main text.

The  $E_{\text{mIR}}$ , FWHM, and SW of the mIR peak obtained from the fitting are displayed in Figs. 3(d) and 4(d) for CeRhIn<sub>5</sub> and YbNi<sub>3</sub>Ga<sub>9</sub>, respectively. Here,  $E_{\text{mIR}}$  is defined as the center of mass of the Lorentz peaks, namely the mean of  $\omega_0$ 's of the Lorentz oscillators weighted by their respective SW's. The SW of a Lorentz oscillator is defined as its area in  $\sigma(\omega)$ , and the SW of the mIR peak is the area of the fitted total mIR peak, namely the red solid curves in Fig. 6.

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