Anisotropic gap formation in $\text{Ce}M_2\text{Al}_{10}$ ($M = \text{Ru, Os}$)

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Abstract. We report an anomalous energy gap formation in $\text{Ce}M_2\text{Al}_{10}$ ($M = \text{Ru, Os}$) by using a polarized optical conductivity $\sigma(\omega)$ measurement. Near $28$ K ($= T_0$), both compounds undergo an antiferromagnetic transition order, the origin of which is not yet clear. We found that the $\sigma(\omega)$ spectra along the orthorhombic $b$-axis have characteristic peak structure at the energy gap edge, which implies the existence of the band nesting due to a charge instability. The peak structure does not originate from the conventional Kondo semiconducting property but can be explained by the charge-density wave (CDW) fluctuation based on the mean-field theory. This suggests that the opening of energy gap along the $b$-axis originates from the CDW or spin-density wave formation.

1. Introduction
Cerium-based compounds CeM$_2$Al$_{10}$ ($M = \text{Fe, Ru, Os}$) with the orthorhombic YbFe$_2$Al$_{10}$-type crystal structure [1] are classified into Kondo semiconductors/insulators (KI) because of the anisotropic electronic structure [2]. CeRu$_2$Al$_{10}$ and CeOs$_2$Al$_{10}$ show anomalous antiferromagnetic transition associated with insulator-to-metal and insulator-to-insulator transitions, respectively, at $T_0 = 28$ K [3, 4, 5], while CeFe$_2$Al$_{10}$ has no magnetic transition. Because $T_0$ is higher than the Neel temperature of Nd- and Gd-counterparts, the phase transition is considered to be driven by other mechanism than the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction, which gives rise to magnetic transitions in conventional rare-earth compounds [4, 6]. To clarify the origin of the anomalous magnetic transition at $T_0$, we have investigated the electronic structure by using temperature-dependent polarized optical conductivity $\sigma(\omega)$ spectra of CeM$_2$Al$_{10}$ along the three principal axes. As a result, we observed the strong anisotropy in the electronic structure, in spite of the rather isotropic Kondo semiconducting energy gap due to the hybridization between the conduction band and the nearly localized Ce 4$f$ state, namely $c$-$f$ hybridization [2]. In CeRu$_2$Al$_{10}$ and CeOs$_2$Al$_{10}$, there is another gap structure with a peak at the gap edge below the hybridization gap energy along the $b$-axis [7]. The gap structure and its temperature dependence are inconsistent with those of conventional Kondo semiconductors previously reported but can be explained by the charge-density wave (CDW) fluctuation based on the mean-field theory by Lee, Rice, and Anderson [8].
In this paper, we show the evidences that the antiferromagnetic ordering is induced by the charge instability with charge or spin density wave formation along the $b$-axis.

2. Experimental
Near-normal incident polarized optical reflectivity [$R(\omega)$] spectra were acquired in a very wide photon-energy region of 2 meV–30 eV to ensure an accurate Kramers-Kronig analysis (KKA). We used two FTIRs in the far-infrared to near-infrared range ($\hbar\omega = 2$ meV–1.5 eV) and a grating monochromator in the visible to vacuum-ultraviolet range (1.5–30 eV) with synchrotron radiation. In order to obtain $\sigma(\omega)$ via KKA of $R(\omega)$, the spectra were extrapolated below 2 meV with a Hagen-Rubens function, and above 30 eV with a free-electron approximation $R(\omega)/\omega^4$ [9]. Single-crystalline samples of Ce$_M$$_2$Al$_{10}$ ($M$ = Fe, Ru, Os) were synthesized by the Al-flux method [5] and was well-polished using 0.3 µm grain-size Al$_2$O$_3$ lapping film sheets for the $R(\omega)$ measurements.

3. Result and discussion
Obtained polarized $\sigma(\omega)$ spectra of the three compounds along all principal axes at the temperature of 10 K are shown in Fig. 1. Sharp peaks in the photon energy range of 15–50 meV originate from optical absorptions by TO-phonons, which were discussed elsewhere [10]. Besides these peaks, energy gaps (cross-hatchings) are observed at around 40-55 meV in all materials. In CeFe$_2$Al$_{10}$, the identical gap sizes of about 55 meV along all principal axes suggest the isotropic $c$-$f$ hybridization energy, which have already been reported [2]. In CeRu$_2$Al$_{10}$ and CeOs$_2$Al$_{10}$, there are shoulder and peak structures at about 40 and 45 meV, respectively, along all axes suggesting the isotropic $c$-$f$ hybridization gaps.

In CeFe$_2$Al$_{10}$, there is no peak structure below the energy of the hybridization gap except for sharp peaks of TO-phonons. In both CeRu$_2$Al$_{10}$ and CeOs$_2$Al$_{10}$, however, there is a broad peak at around 20 meV (namely “20-meV peak” hereafter) only along the $b$-axis. Such a peak

![Figure 1](image-url)
has never been observed within the hybridization gap of conventional KIs.

To investigate the evolution of the 20-meV peak along the -axis, we measured the temperature dependence by a small temperature step. The obtained \( \sigma(\omega) \) spectra of CeRu\(_2\)Al\(_{10}\) and CeOs\(_2\)Al\(_{10}\) are shown in Fig 2. Some sharp peaks at around 25–30 meV originate from TO-phonons described above. A shoulder at 40 meV in CeRu\(_2\)Al\(_{10}\) and a small peak at 44 meV in CeOs\(_2\)Al\(_{10}\) originate from the optical transition across the \( c-f \) hybridization gap. The \( c-f \) hybridization gaps in both materials still appear at 34 K in CeRu\(_2\)Al\(_{10}\) and 40 K in CeOs\(_2\)Al\(_{10}\). In the figures, the tails of the \( c-f \) hybridization gap, are drawn by dashed lines. The 20-meV peaks become visible below the temperatures (namely \( T^* \)) of 31 ± 1 K in CeRu\(_2\)Al\(_{10}\) and 36 ± 2 K in CeOs\(_2\)Al\(_{10}\). This suggests that the 20-meV peak develops after the \( c-f \) hybridization gap opened. The shape of the 20-meV peak is akin to that due to the band nesting by a CDW or spin-density wave (SDW) formation in (TMTSF)\(_2\)PF\(_6\) and so on [11].

These spectral features and the temperature dependence are consistent with the fluctuation of the CDW formation. The CDW fluctuation has been discussed to explain the decrease in the magnetic susceptibility data of other quasi one-dimensional CDW compounds [12, 13] using the mean-field theory [8]. Thereby, the CDW fluctuation occurs at the temperature \( T_{CF}^M \) higher than the CDW transition temperature \( T_C \). At \( T_{CF}^M \), the magnetic spin susceptibility starts

![Figure 2](image-url)

**Figure 2.** Temperature dependence of optical conductivity \([\sigma(\omega)]\) spectra of CeRu\(_2\)Al\(_{10}\) and CeOs\(_2\)Al\(_{10}\) along the -axis in the photon energy range below 70 meV. The backgrounds due to the \( c-f \) hybridization gaps below 40 meV in CeRu\(_2\)Al\(_{10}\) and 45 meV in CeOs\(_2\)Al\(_{10}\) are depicted by dashed lines. Sharp peaks at around 25–30 meV are due to TO-phonons. Each of the lines is offset by \( 2 \times 10^3 \ \Omega^{-1} \text{cm}^{-1} \) for clarity.
to decrease from a constant Pauli spin susceptibility at higher temperature, and the density of states at the Fermi level also decreases indicating the energy gap opening. The relation between $T_{MF}^C$ and $T_C$ can be attributed to a SDW case. If $T^*$ and $T_0$ are regarded as $T_{MF}^C$ and $T_C$, respectively, the temperature dependences of both $\sigma(\omega)$ and magnetic susceptibility of CeRu$_2$Al$_{10}$ and CeOs$_2$Al$_{10}$ can be explained by the CDW fluctuation theory. This relationship strongly suggests that the energy gaps along the $b$-axis of CeRu$_2$Al$_{10}$ and CeOs$_2$Al$_{10}$ originate from the CDW/SDW formation.

4. Conclusion
To summarize, the mechanism of the energy gap formation of CeRu$_2$Al$_{10}$ and CeOs$_2$Al$_{10}$ along the $b$-axis was studied by using the optical conductivity $[\sigma(\omega)]$ spectra. The temperature dependence of the gap formation in $\sigma(\omega)$ spectra and the magnetic susceptibility data are consistent with the behavior of the CDW fluctuation based on the mean-field theory. This result strongly suggests that the energy gap along the $b$-axis originates from the CDW/SDW formation.

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