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Optical properties and electronic structures of $CeSn_3$ and $LaSn_3$

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The dielectric functions of $CeSn_3$ and $LaSn_3$ were remeasured to correct errors in previous measurements. The partial-sum rule for the optical conductivity shows that $CeSn_3$ has about 0.54 electrons per formula unit more than $LaSn_3$ contributing to the conductivity in the 1.5–5.4 eV region.

The optical properties of $CeSn_3$ and $LaSn_3$ (Ref. 1) were measured to investigate the role of the $4f$ electron using a rotating-polarizer-rotating-analyzer ellipsometer (RPRAE) (Ref. 2) with two quartz Rochon prisms for the polarizer and the analyzer. The optical conductivities of both materials were also calculated using the same potential as Ref. 3. There was a difference in both the measured and calculated conductivities of the compounds, and a $4f$-electron contribution to the optical conductivity for $CeSn_3$ was demonstrated.

First, the original calculated optical conductivities of both materials should be multiplied by a factor of 2, since in the final calculations the spin degeneracy was inadvertently omitted. Also, the directions of the transitions in Table I of Ref. 1 should be reversed. In addition, Kim *et al.* did not calculate the contributions from the exterior of the muffin-tin spheres. These contributions are 10% and 5% for $CeSn_3$ and $LaSn_3$, respectively.

Second we report measurements of the optical conductivities of $CeSn_3$ and $LaSn_3$. The samples and their preparation method are similar to those of Ref. 1 but the samples were placed in the UHV chamber more quickly. The chamber pressure was kept below 5×10^{-7} Torr for the first measurement at room temperature. The chamber was then baked out at 110 °C for one day. The complex dielectric functions were measured with a rotating-analyzer ellipsometer (Ref. 4) in the 1.5–5.4 eV range.

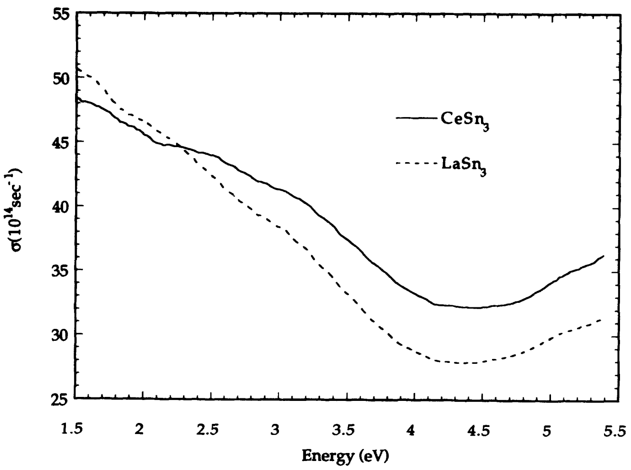

FIG. 1. Optical conductivities of $CeSn_3$ and $LaSn_3$. Note that the zero of the optical conductivity is suppressed.

The measured optical conductivity spectra decrease with time. The difference between measurements before and after baking is the largest, indicating heavy oxidation during baking due to the high temperature and poorer vacuum. After baking, the pressure was kept below 7×10^{-10} Torr, reducing considerably the oxide growth rate. After baking we made three more measurements at intervals of a day or two. If the oxide overlayer is not too thick we can estimate its effects and deduce the optical conductivity of the bulk without the oxide overlayer to first order of d/λ, where d is the thickness of the oxide overlayer and λ is the wavelength of the incident light. The corrected optical conductivities are shown in Fig. 1. The corrections are 11–14% for $CeSn_3$ and 13–17% for $LaSn_3$.

There are two reasons for the difference between measurements of Kim *et al.* and ours. First, the RPRAE with quartz Rochon prisms causes a difference[5] due to inadequate correction for the optical activity of the quartz. Second, the oxide overlayer is likely to reduce the magnitude of measured optical conductivities.

The conductivity partial-sum rule applied to the measured spectra gives 0.53 electrons per formula unit more for $CeSn_3$ than for $LaSn_3$.

In decomposing the interband transitions into contributions from states based on particular atomic transitions there are several approaches because the problem is not well defined—charge density cannot be unambiguously assigned to a particular subshell of a particular atom, especially in interstitial regions, and there are interference effects in the squared dipole matrix elements, making contributions of individual transitions only qualitatively interpretable. We have reanalyzed the calculated interband conductivity, and find the contribution to the partial-sum rule from $f \rightarrow d$ transitions to be small (0.087 for $CeSn_3$ and 0.031 for $LaSn_3$) because of small matrix elements in this energy range. While there are some differences in the contribution from $d \rightarrow f$ transitions, they are suspect because the standard local density approximation does not place the f^2 final state configuration at the appropriate energy.

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