New crystal field level scheme of CeB₆ deduced from Raman and neutron spectroscopy (abstract)

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An inelastic electronic excitation has been observed in CeB₆ near 47 meV (545 K) by means of neutron and Raman spectroscopy. This excitation has been identified as the $\Gamma_8 \rightarrow \Gamma_7$ crystal-field-electric-field (CEF) transition. From the anomalous energy shift of this excitation at low temperatures, detectable due to the high resolution of Raman spectroscopy ($\pm 5$ K), we deduce a $\Gamma_8$ ground-state split by about 30 K. With this new CEF level scheme the first consistent interpretation of so far seemingly unrelated thermal, elastic, and magnetic data is achieved. In spite of the large body of thermal, magnetic, and elastic data of CeB₆ accumulated over the past years, various diverging proposals for the CEF level scheme have been reported. In all schemes a $\Gamma_7$ ground state was assumed, but the CEF splittings ranged from 10 K to more than 400 K. The absence of CEF excitations in direct spectroscopic measurements up to 400 K has been puzzling, and pointed to the necessity of high-energy neutron experiments. We have performed inelastic neutron scattering experiments using high-energy incident neutrons up to 185 meV from the Intense Pulsed Neutron Source at Argonne National Laboratory. An inelastic peak at 46 meV (530 K) is clearly identified as magnetic scattering by its $Q$ dependence. The absolute intensity corresponds to the value of a $\Gamma_8 \rightarrow \Gamma_7$ transition. In addition, we have carried out Raman measurements on (100) faces of CeB₆. At room temperature we found an inelastic excitation at 372 cm⁻¹ (530 K). It is absent in the reference compound LaB₆. From the polarization analysis we found that the transition has $\Gamma_8^1$ and $\Gamma_7^3$ symmetry. Both facts characterize this excitation as the $\Gamma_8 \rightarrow \Gamma_7$ CEF transition, which is the first CEF excitation seen in a metal by means of Raman spectroscopy. The high resolution of Raman spectroscopy enabled us to detect a 10-cm⁻¹ shift of the $\Gamma_8 \rightarrow \Gamma_7$ transition energy to 382 cm⁻¹ for temperatures below 20 K. This can only be explained by assuming a $\Gamma_8$ ground state, which is split by 30 K. From the spectroscopic data we can establish a completely new CEF level scheme for CeB₆ with a $\Gamma_8$ ground state, split by about 30 K and a $\Gamma_7$ state 545 K above. This allows a straightforward interpretation of various other experimental data, such as magnetic entropy, static magnetic susceptibility, high-field magnetization, magnetic form factor, temperature, and magnetic-field-dependent elastic constants and the antiferroquadrupolar ordering below $T_Q = 3.3$ K. A recent theoretical investigation of magnetic ordering of a periodic Anderson Hamiltonian with orbital degeneracy has been applied to CeB₆. On the basis of entropy and magnetization data it was assumed that the quartet $\Gamma_8$ is the ground state. Hence it was predicted that the low-temperature phase II ($2.1 \text{K} < T < 3.3 \text{K}$) is due to an orbital antiferromagnetic ordered state, contrary to the conclusions of an antiferroquadrupolar ordering. This work was in part supported by Deutsche Forschungsgemeinschaft, SFB 125 and SFB 126.


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