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The search for the lost entropy of CeB₆

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Among the many puzzles of CeB₆, there is the question of its magnetic entropy. In the paramagnetic range, the entropy curves derived from specific heat measurements [1,2] are inconsistent with the fourfold degenerate, well isolated, crystal field ground state [3]. This question is solved thanks to new specific heat measurements together with a better description, in the cage context, of the phonons and crystal field contributions.

Accurate analysis of the antiferromagnetic transition shows that it is actually first-order, resulting in an entropy step at T_N. Starting from the characteristic dispersion curves of a cage system, an improved modelling of the phonons contribution is proposed. This gives a physical ground to the derivation of CeB₆ phonons specific heat from the LaB₆ reference data.

The obtained temperature variation for CeB₆ magnetic entropy is consistent with the fourfold crystal field ground state. However, starting from an entropy value of only $8 \approx R \ln(2.6)$ J/K.mol at the ordering temperature T_Q, it takes as much as 30 K to reach the quadruplet entropy ($R \ln(4)$ J/K.mol). This temperature scale fits with the simultaneous shift in the energy separation of the crystal field levels [3]. These phenomena are readily explained if one considers that the rare-earth movement splits the crystal field ground state. Accounting for this cage distributed crystal field yields a quantitative description of the temperature dependence of the paramagnetic entropy.

The example of CeB₆ shows that a specific treatment of the crystal field is required in rare-earth cage compounds.

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