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

Authors
Zirngiebl, E
Hillebrands, B
Blumenröder, S
et al.

Peer reviewed

New crystal field level scheme of CeB$_6$ deduced from Raman and neutron spectroscopy (abstract)

E. Zirngiebl, B. Hillebrands, S. Blumenröder, and G. Güntherodt

II. Physikalisches Institut, Universität Köln, 5000 Köln 41, Federal Republic of Germany

M. Loewenhaupt

Institut für Festkörperforschung, KFA Jülich, 5170 Jülich 1, Federal Republic of Germany

J. M. Carpenter

IPNS, Argonne National Laboratory, Argonne, Illinois 60439

K. Winzer

I. Physikalisches Institut, Universität Göttingen, 3400 Göttingen, Federal Republic of Germany

Z. Fisk

Los Alamos National Laboratory, Los Alamos, New Mexico 87545

An inelastic electronic excitation has been observed in CeB$_6$ near 47 meV (545 K) by means of neutron and Raman spectroscopy. This excitation has been identified as the $\Gamma_8 \rightarrow \Gamma_7$ crystalline-electric-field (CEF) transition. From the anomalous energy shift of this excitation at low temperatures, detectable due to the high resolution of Raman spectroscopy (± 5 K), we deduce a Γ_8 ground-state split by about 30 K.[1] 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 CeB$_6$ accumulated over the past years,[2-5] various diverging proposals for the CEF level scheme have been reported. In all schemes a Γ_7 ground state was assumed, but the CEF splittings ranged from 10 K[6] to more than 400 K.[7] The absence of CEF excitations in direct spectroscopic measurements up to 400 K[7] has been puzzling, and pointed to the necessity of high-energy neutron experiments. We have performed inelastic magnetic 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 CeB$_6$. At room temperature we found an inelastic excitation at 372 cm^{-1} (530 K). It is absent in the reference compound LaB$_6$. From the polarization analysis we found that the transition has Γ_3^+ and Γ_5^+ 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 Γ_8 ground state, which is split by 30 K.[1,8] From the spectroscopic data we can establish a completely new CEF level scheme for CeB$_6$ with a Γ_8 ground state, split by about 30 K and a Γ_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,[8] temperature, and magnetic-field-dependent elastic constants and the antiferroquadrupolar ordering below $T_Q = 3.3$ K.[9] A recent theoretical investigation of magnetic ordering of a periodic Anderson Hamiltonian with orbital degeneracy has been applied to CeB$_6$.[10] On the basis of entropy and magnetization data it was assumed that the quartet Γ_8 is the ground state. Hence it was predicted that the low-temperature phase II (2.1 K $< T <$ 3.3 K) is due to an orbital antiferromagnetic ordered state, contrary to the conclusions of an antiferroquadrupolar ordering.[9,11,12]

[1] E. Zirngiebl, B. Hillebrands, S. Blumenröder, G. Güntherodt, M. Loewenhaupt, J. M. Carpenter, K. Winzer, and Z. Fisk, Phys. Rev. B **30**, 4052 (1984).

[2] T. Kasuya, K. Takegahara, Y. Aoki, K. Hanzawa, M. Kasaya, S. Kunii, T. Fujita, N. Sato, H. Kimura, T. Komatsubara, T. Furuno, and J. Rossat-Mignod, in *Valence Fluctuations in Solids*, edited by L. M. Falicov, W. Hanke, and M. B. Maple (North-Holland, Amsterdam, 1981), p. 215.

[3] T. Fujita, M. Suzuki, T. Komatsubara, S. Kunii, T. Kasuya, and T. Oktsaka, Solid State Commun. **35**, 569 (1980).

[4] M. Kawakami, S. Kunii, T. Komatsubara, and T. Kasuya, Solid State Commun. **36**, 435 (1980).

[5] P. Burlet, J. X. Boucherle, J. Rossat-Mignod, J. W. Cable, W. C. Koehler, S. Kunii, and T. Kasuya, J. Phys. (Paris) Colloq. **43**, C7-273 (1982).

[6] T. Goto, A. Tamaki, S. Kunii, T. Nakajima, T. Fujimura, T. Kasuya, T. Komatsubara, and S. B. Woods, J. Magn. Magn. Mater. **31–34**, 419 (1983).

[7] S. Horn, F. Steglich, M. Loewenhaupt, H. Scheuer, W. Felsch, and K. Winzer, Z. Phys. B **42**, 125 (1981).

[8] E. Zirngiebl, S. Blumenröder, and G. Güntherodt (to be published).

[9] B. Lüthi, S. Blumenröder, B. Hillebrands, E. Zirngiebl, G. Güntherodt, and K. Winzer, Z. Phys. B (in press).

[10] F. J. Ohkawa, J. Phys. Soc. Jpn. **52**, 3897 (1983).

[11] K. Hanzawa and T. Kasuya, J. Phys. Soc. Jpn. **53**, 1809 (1984).

[12] J. M. Effantin, J. Rossat-Mignod, P. Burlet, H. Bartholin, S. Kunii, and T. Kasuya, Proceedings of the International Conference on Valence Fluctuations, Cologne, 1984 (to be published).