RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 32, NUMBER 3 1 AUGUST 1985

Low-frequency Einstein mode in the heavy-fermion compound UBe_{13}

B. Renker, F. Gompf, W. Reichardt, H. Rietschel, and J. B. Suck
Kernforschungszentrum Karlsruhe, Institut für Nukleare Festörperphysik, Postfach 3640,
D-7500 Karlsruhe, Federal Republic of Germany

J. Beuers
Max-Planck-Institut für Metallforschung, Institut für Werkstoffwissenschaften,
Heisenbergstrasse 5, D-7000 Stuttgart, Federal Republic of Germany

(Received 12 March 1985)

We present results of inelastic neutron scattering on polycrystalline samples of the heavy-fermion compound UBe_{13}. We analyze these data on the basis of a Born–von-Kármán model. Our results give clear evidence for the existence of an Einstein-mode-like oscillator at 13 meV, independently of temperature. From our deduced phonon density of states, we calculate the lattice contribution to the low-temperature heat capacity. It is found to be small compared to the electronic part.

By use of inelastic neutron scattering, we determined the phonon density of states (PDOS) in the heavy-fermion compound UBe_{13}. Our investigations were guided by the following object: Because of the very high mass ratio of its constituents, and because of the large interatomic distances for the heavy U atoms, UBe_{13} is expected to show characteristic low-lying phonon branches. For brevity we will designate the whole complex of these branches as "resonant mode," although this term was originally coined for the case of heavy impurities in a light matrix.

Representing an interesting problem already in itself, the existence of such a resonant mode will acquire additional importance if it is linked with the peculiarities of heavy-fermion physics.[1] Since the resonant mode is caused by vibrations of the U atoms, i.e., by those atoms which carry the f electrons, it could possibly couple to the heavy quasiparticles. If so, the strong temperature dependence of the electronic properties for T around $T_K \sim 8$ K should at least be partially reflected in the phonons as well.

Another important point concerns the evaluation of experimental heat-capacity data. There, the decisive electronic part can only be extracted if the lattice part is known. For UBe_{13}, the situation is presently controversal. Ott and coworkers[2,3] argue that superconductivity in UBe_{13} is caused by triplet pairing, a mechanism so far known to be realized only in liquid ^3He. Their assumption is mainly based on the low-temperature results for the specific heat $C(T)$: Plotted over appropriate scales, the data strongly resemble those for liquid ^3He. In their analysis, the lattice contribution to $C(T)$ is estimated to be virtually negligible in the temperature range under consideration ($T \lesssim 8$ K). But this estimate is in conflict with theoretical work by Overhauser and Appel (OA). In a recent publication,[4] these authors proposed an s-f hybridization model to explain most of the experimental results for UBe_{13}. In its main essence, their model represents a one-particle description of the electrons based on conventional singlet pairing in order to describe the superconducting state. In order to obtain agreement between their theory and the experimental specific heat, OA had to assume a considerable lattice contribution $C_{\text{latt}}(T)$ which they fitted within a Debye model through $C_{\text{latt}}(T) = \alpha T^3$ with $\alpha = 9\times10^{-4}$ J/K^{4}mole for 1 K $\lesssim T \leqslant 12$ K. This value for α implies a Debye temperature $\Theta_D \sim 310$ K which is much lower than what could be expected from scaling down the Debye temperature for Be via the square root of the ratio of the mean atomic masses. Also, from measurements of the low-temperature heat capacity, Bucher *et al.*[5] found the much higher value of 618 K in the related compound $ThBe_{13}$. As a source for their unexpectedly low θ_D in UBe_{13}, OA invoked the above mentioned resonant mode which, in order to give the required $C_{\text{latt}}(T)$, would have to lie around $\hbar\omega \sim 5$ meV.

Our inelastic neutron scattering (INS) experiments were performed at the time-of-flight spectrometer TOF 1 of the Melusine reactor in Grenoble. For these measurements which were carried out at 300 K and at 8 K, 312 detectors were distributed over scattering angles between 40° and 120° and the energy E_0 of the incident neutrons was chosen to be 30 meV. Owing to the finite instrumental resolution, only energy transfers down to 3.5 meV could be registered without interfering with the elastic line. At lower temperatures (22 and 7 K), additional measurements with $E_0 = 11.6$ meV and smaller scattering angles (10–90) were made to look for possible low-lying excitations. Under such conditions, the spectra could be analyzed down to 1 meV.

The samples were prepared from 3N6 Be and 3N7 U materials by repeated arc melting under argon, starting with a 1 at.% Be surplus to compensate for losses due to evaporation. X-ray analysis revealed homogeneous and single-phased UBe_{13}. Measurements of the specific heat showed a bulk transition into the superconducting state with a midpoint $T_c \sim 850$ mK.

INS on polycrystalline compounds provides a generalized phonon density of states (PDOS) $G(\hbar\omega)$ which differs from the true PDOS $F(\hbar\omega)$ by weighting the vibrations of an atom i with the ratio σ_i/M_i (σ_i: bound scattering cross section; M_i: atomic mass).[6] In Fig. 1. we show our experimental results for $G(\hbar\omega)$ in UBe_{13} at 300 K. It consists of two distributions separated from each other by a deep valley. The energetically lower part has a pronounced peak around 13 meV while the upper part represents a broad spectrum composed of optical phonons and centers around 60 meV with a cutoff energy of about 90 meV, very similar to the overall shape of the PDOS in pure Be. For a further analysis of these data, we employed a simple Born–von-Kármán model to describe the lattice dynamics in UBe_{13} and

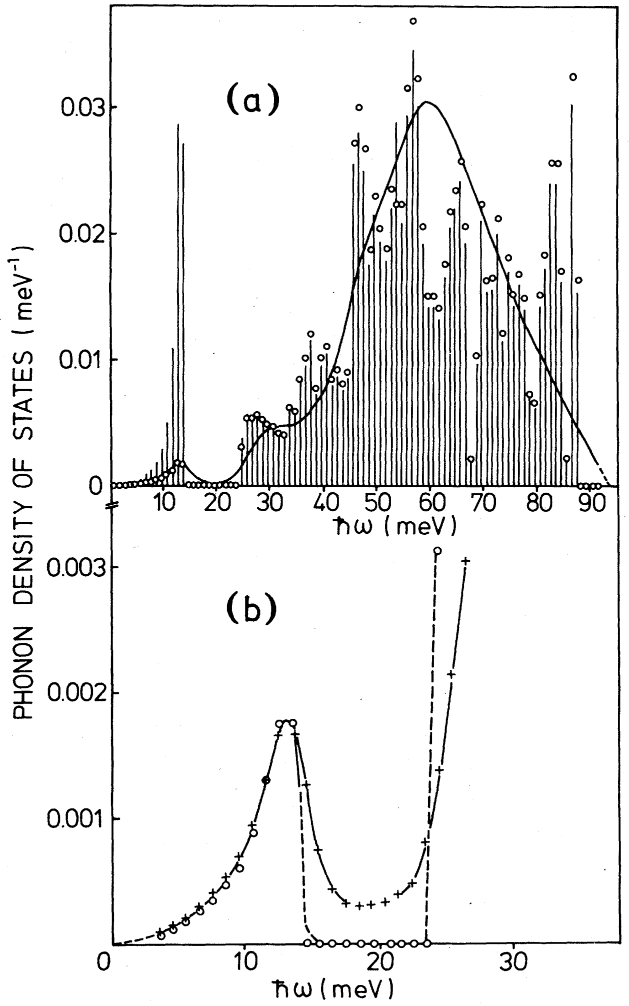

FIG. 1. (a) Generalized PDOS $G(\hbar\omega)$ and PDOS $F(\hbar\omega)$ for UBe_{13}; solid line: $G(\hbar\omega)$ as deduced from experiment; circles: $G(\hbar\omega)$ as calcualted from the Born–von-Kármán model; histogram: $F(\hbar\omega)$ as calculated from the same model. (b) Calculated (circles) and experimental (crosses and solid line) $G(\hbar\omega)$ on a larger energy scale.

to calculate both $G(\hbar\omega)$ and $F(\hbar\omega)$. In this model, only three different force constants were employed: one of longitudinal character for Be-Be interactions and two of longitudinal and transversal character, respectively, for the U-Be interactions.

Our best fit to the experimental $G(\hbar\omega)$ is also plotted in Fig. 1(a) together with the PDOS $F(\hbar\omega)$ which is represented by the histogram. As for the optical spectrum, this fit is far from being perfect, sketching only the overall shape. But since, for the very subject of this work, the details of the optical spectrum are of no concern, we forwent further improvement through more sophisticated models but left this together with a more detailed description of the lattice dynamics to a forthcoming paper. The lower part of the spectrum (from 14 meV on downwards), however, is well reproduced [see Fig. 1(b)]. By inspection of the corresponding polarization vectors, we could clearly identify the peak at 13 meV as being mainly produced by vibrating U atoms. This becomes also evident by direct comparison of $G(\hbar\omega)$ and $F(\hbar\omega)$. Finally, from our calculations we find that the valley between the two parts of the spectrum is likely to be a gap which in our measurements is at least partially smeared out due to the finite resolution of the spectrometer.

Since, at low temperatures, INS is effective in energy loss only, our results for 8 K are restricted to energy transfers $\hbar\omega < E_0$ which allowed the data to be analyzed up to about 17 meV. Within this range and within our experimental resolution, our low-temperature data (not shown as figure) are identical to those taken at 300 K. In particular, no shift of the resonant mode upon cooling was observed. Further, after lowering E_0 from 30 to 11.6 meV, no additional structure could be found. As to possible magnetic contributions to our INS results, from the good agreement between the calculated and the measured $G(\hbar\omega)$ we conclude that such contributions, if present at all, should be relatively small and structureless.

From our fitted Born–von-Kármán model, we generated the PDOS $F(\hbar\omega)$ [see Fig. 1(a)]. The ratio of the spectral weights between the lower and higher part is 1/14. Since UBe_{13} has two formula units per unit cell, the lower part of the spectrum comprises three acoustic *and* three optical branches, while the upper part consists of 78 optical branches. This $F(\hbar\omega)$ was then used to calculate the lattice contribution $C_{\mathrm{latt}}(T)$ to the specific heat via standard thermodynamics. It is shown in Fig. 2. Having its energy centered at 13 meV, the resonant-mode-like oscillator in UBe_{13} lies much higher than assumed by OA.[4] If treated exactly as an Einstein oscillator, its contribution to the low-temperature specific heat would be completely negligible, below, say, $T \sim 20$ K. However, our results indicate addi-

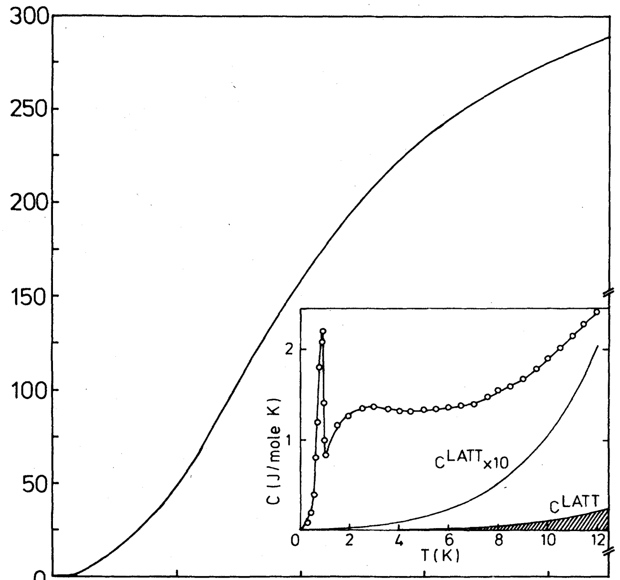

FIG. 2. Lattice contribution to the heat capacity as calculated from the histogram shown in Fig. 1; the inset compares this contribution with the experimental specific heat as measured by Ott *et al.* (Ref. 3) in UBe_{13} (circles).

tional spectral weight below 13 MeV decreasing roughly like $\sim \omega^2$ (of course, for sufficiently low ω it has to follow an ω^2 law). This low-frequency wing has its origin in the three acoustic branches which are considerably lowered in comparison to their counterparts in a pure Be lattice. These low-lying modes are mainly responsible for $C_{\mathrm{latt}}(T)$ below 20 K. Nevertheless, as can be seen from the inset in Fig. 2, the total $C_{\mathrm{latt}}(T)$ is smaller by a factor $\gtrsim 5$ than assumed by OA. At 12 K (the highest temperature considered by OA), $C_{\mathrm{latt}}(T)$ amounts to about 10% of the total specific heat as measured by Ott and co-workers.[2,3] So we conclude that, in UBe_{13}, the specific heat below 12 K is essentially electronic in origin, in agreement with the assumption of Ref. 3.

A last interesting point is the apparent insensitivity of the lower part of the spectrum to cooling. But although this observation may be considered as an indication of only weak coupling between the resonant mode and the heavy electrons, it must not be taken as a proof. Since the resonance-like increase of the quasiparticle density of states at low temperatures takes place only within ~ 10 K around the Fermi energy, it is as well conceivable that the coupling is still strong but that only phonons with $h\omega/k_B \lesssim 10$ K are affected. We have also performed similar investigations at 300 and 8 K for the related compound ThBe_{13} which has no $5f$ electrons and which is not a heavy-fermion system. We find an almost identical spectrum of lattice vibrations. In particular, the resonance mode appears at 13.5 meV independently of temperature at an energy which is only 0.5 meV higher than in UBe_{13}.[7] This small difference may well be understood by the larger ionic radius of Th. Therefore, from the present work we can exclude that a low-lying Einstein mode of U atoms contributes essentially to the specific heat in UBe_{13} which is a crucial point in Ref. 4. Furthermore, by comparison with the related compound ThBe_{13} it becomes clear that this particular mode does not couple to the heavy electrons to any larger extent.

Note added. H. R. Ott has informed us that he has determined the lattice contribution to the specific heat of UBe_{13} from his data and he finds a value of 250 mJ/mole K at 13 K which is in excellent agreement with our results.

We gratefully acknowledge many stimulating discussions with P. Fulde during the course of these investigations.

---

[1] For a recent review on heavy-fermion systems see, e.g., G. R. Stewart, Rev. Mod. Phys. **56**, 755 (1984).

[2] H. R. Ott, H. Rudigier, Z. Fisk, and J. L. Smith, Phys. Rev. Lett. **50**, 1595 (1983).

[3] H. R. Ott, H. Rudigier, T. M. Rice, K. Ueda, Z. Fisk, and J. L. Smith, Phys. Rev. Lett. **52**, 1915 (1984).

[4] A. W. Overhauser and J. Appel, Phys. Rev. B **31**, 193 (1985).

[5] E. Bucher, J. P. Maita, G. W. Hull, R. C. Fulton, and A. S. Cooper, Phys. Rev. B **11**, 440 (1975).

[6] P. Schweiss, B. Renker, E. Schneider, and W. Reichardt, *Proceedings of the Second Rochester Conference on Superconductivity in d- and f-Band Metals,* edited by D. H. Douglass (Plenum, New York and London, 1976), p. 189.

[7] F. Gompf, B. Renker, H. Rietschel, and N. Nücker, Proceedings of the International Conference on Materials and Mechanisms of Superconductivity, Ames, Iowa, May 1985 (unpublished).