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## A NEUTRON SCATTERING STUDY OF THE HEAVY FERMION SYSTEM $\text{UBe}_{13}$

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Neutron scattering measurements were performed on polycrystalline samples of  $\text{UBe}_{13}$  in order to establish more accurately the structure and probe the dynamics of the spin fluctuations. The structure was confirmed to be cubic with space group  $\text{O}_h^6$  (Fm3c) with BeII positional parameters  $y = 0.1763(1)$  and  $z = 0.1150(1)$ . The inelastic spectrum at  $T = 10$  K exhibits a broad response with half width at half maximum of 15 meV.

The recent discovery of heavy fermion systems (HFS) has created a great deal of excitement amongst physicists studying the properties of f electrons in solids [1]. At low temperatures, there is a large linear term in the specific heat which implies a large effective mass of the f electrons. The enhanced susceptibility at low temperatures with the absence of magnetic ordering suggests a narrow ( $\approx 1$  meV) density of states at the Fermi energy. These properties seem to be at the extreme limit of the mixed valence (MV) materials where localized f electrons lie near the Fermi energy [2]. In the HFS the characteristic energy scale associated with the width of the resonance is 1–2 meV whereas in the MV materials it is 10–20 meV. One unusual feature of the HFS's is that the same f electrons that pair to form a superconducting state also exhibit the large magnetic susceptibility. The nature of the superconducting state is not established but it is believed to be more complicated than a BCS singlet state.

$\text{UBe}_{13}$  is one of the first established HFS [3]. It does not exhibit any magnetic ordering, but becomes superconducting below 1.0 K. We have performed neutron measurements of the structure of  $\text{UBe}_{13}$  and inelastic measurements probing the energy dependence of the magnetic fluctuations in this system.

$\text{UBe}_{13}$  has the cubic  $\text{NaZn}_{13}$  type structure ( $\text{O}_h^6$ -Fm3c) with 8 molecular units per unit cell [4]. Surprisingly the structure has never been studied in detail and the general coordinates of the BeII sites have been assumed to be the same as the Zn II sites as measured in  $\text{NaZn}_{13}$  [5]. A neutron diffraction study was performed on a polycrystalline sample of  $\text{UBe}_{13}$  at several

temperatures between 10 and 250 K over the angular range  $20^\circ < 2\theta < 130^\circ$  encompassing 13 reflections [6]. The Rietveld method of profile analysis was used to determine the positions of the atoms, the lattice parameter and thermal factors. Good fits were obtained at all temperatures and the results are given in table 1. The values of  $y$  and  $z$  are slightly different from those measured in  $\text{NaZn}_{13}$  and are temperature independent within error limits. The lattice parameter decreases smoothly with temperature and exhibits a slightly larger thermal expansion than that of  $\text{ThBe}_{13}$  [6]. The thermal factor for the U atoms is substantially larger than that for the Th atoms in  $\text{ThBe}_{13}$  [6]. This is in qualitative agreement with predictions of Overhauser and Appel [7] although measurements should be extended to larger  $Q$  values and lower temperatures.

The inelastic measurements were performed on a large polycrystalline sample (25 g) of  $\text{UBe}_{13}$ . Both polarized and unpolarized neutrons were used. In the former case, the magnetic scattering can be uniquely separated from the ubiquitous nuclear "background" [8]. In the latter case, the nuclear "background" is subtracted by measurements on an identically shaped sample of the nonmagnetic compound  $\text{ThBe}_{13}$ . Fig. 1 shows our results at 10 K [9]. In the top portion, the unpolarized spectra  $\text{UBe}_{13}$  and  $\text{ThBe}_{13}$  are given separately. A relatively sharp feature at  $\Delta E = 13.0$  meV in the  $\text{ThBe}_{13}$  spectrum corresponds to a peak in the phonon density of states. The upswing at the higher energies is the start of a band of phonons associated with vibrations of the Be atoms. In the observed  $\text{UBe}_{13}$  spectrum, the peak near 13 meV is considerably

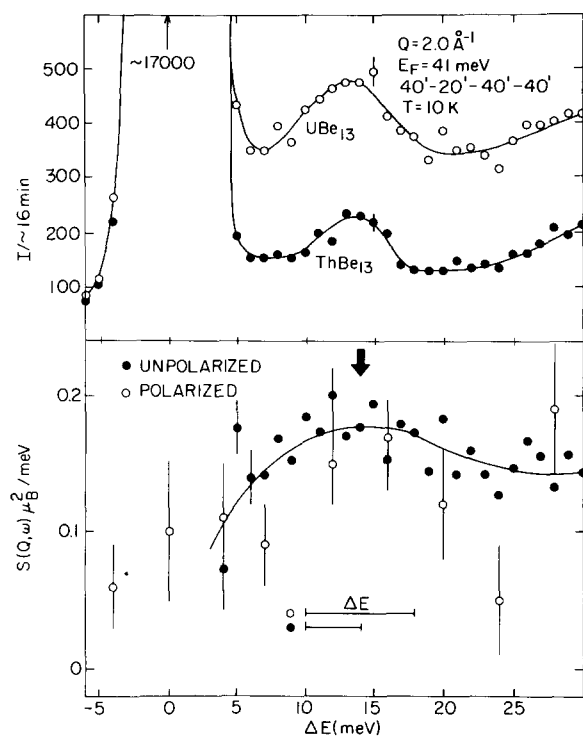


Fig. 1. Top: The inelastic spectra of  $UBe_{13}$  and  $ThBe_{13}$ . The latter has been scaled by 1.1 to take into account the differences in sample volume and absorption. Bottom: The difference spectrum  $I(UBe_{13}) - 1.1I(ThBe_{13})$ . Also shown is the spectrum determined from polarized neutron measurements. The horizontal bar denotes the resolution. (After Goldman, et al. [9].)

broadened and corresponds to a mixture of magnetic scattering and phonons.

In the bottom portion of fig. 1, the subtracted spectrum is shown along with the results of the polarized

beam measurements. The measurements have been put into absolute units by normalizing the intensity to a powder peak with its known structure factor [8,9]. The  $Q$  dependence of the magnetic scattering was measured by performing a  $Q$  scan with the spectrometer set for a constant energy transfer. For both the polarized and unpolarized neutron measurements, the intensity decreased with increasing  $Q$  and was consistent with the form factor of the uranium 5f electrons.

The magnetic scattering spectrum of fig. 1 is broad with a maximum at  $\Delta E \approx 15$  meV. This is consistent with a Lorentzian lineshape of the imaginary part of the susceptibility,  $\chi''(\omega)/\omega \sim \Gamma(\omega^2 + \Gamma^2)^{-1}$  with  $\Gamma = 15.0$  meV. There are two important features of these results which should be emphasized:

- (i) The data in fig. 1 can be properly summed and compared with the bulk susceptibility  $\chi_B$  [8–10]. At  $T = 10$  K, we obtain  $\chi_N/\chi_B = (0.7 \pm 0.2)$  using  $\chi_B = 12 \times 10^{-3}$  emu/mol [1] and  $\chi_N$  is the susceptibility determined by neutrons. One might expect  $\chi_N$  to be somewhat less than  $\chi_B$  since we integrated our spectrum only out to 28 meV and, as seen in fig. 1, there is some intensity extending out to higher energies. However, in the integration this intensity is reduced by  $\omega^{-1}$ . Thus, within error, the magnetic susceptibility is nearly exhausted by the broad scattering in fig. 1. Any additional response must have a very small spectral weight.
- (ii) No sharp quasielastic response is observed around  $\Delta E = 0$  as would be expected on the basis of the small energy width associated with the narrow f-level resonance ( $\sim 1$  meV). [1]. Measurements were performed with better resolution and no additional signal was observed around  $\Delta E = 0$ . It thus remains a puzzle why the neutron measurements, which directly couple to the f electron response, are not exhibiting the low energy response expected from specific heat and susceptibility measurements.

Table 1

Results of Rietveld refinement of  $UBe_{13}$ . The space group is  $O_h^6$  with U in 8(a) sites at (000); Be(I) in 8(b) sites at 0, 0, 0 and 1/2, 1/2, 1/2; Be(II) in 96(i) sites at  $\pm 0, y, z$ ; etc. The  $B$ 's are individual isotropic temperature factors. The numbers in the parenthesis are estimated standard deviations referred to the least significant digit

$T$ (K)	$a_0$ (Å)	$y$	$z$	$B_U$ <sup>a)</sup>	$B_{BeI}$ <sup>b)</sup>	$B_{BeII}$ <sup>c)</sup>
10	10.2489 (1)	0.1763 (1)	0.1150 (1)	0.7 (1)	0.1 (1)	1.1 (1)
50	10.2499 (1)	0.1763 (1)	0.1148 (1)	0.3 (1)	0.2 (2)	0.8 (1)
100	10.2504 (1)	0.1761 (1)	0.1150 (1)	0.7 (1)	0.2 (1)	1.1 (1)
175	10.2540 (1)	0.1765 (1)	0.1152 (1)	0.9 (1)	0.8 (1)	1.4 (1)
250	10.2602 (1)	0.1763 (1)	0.1150 (1)	0.9 (1)	0.4 (1)	1.3 (1)

<sup>a)</sup> Thermal factor for the 8(a) uranium sites.

<sup>b)</sup> Thermal factor the 8(b)  $Be_I$  sites.

<sup>c)</sup> Thermal factor for the 96(i)  $Be_{II}$  sites.

The meaning of the broad response is not totally clear, but the spectra of fig. 1 bear a striking resemblance to the mixed valent materials. In fact, measurements on the isostructural compound  $CeBe_{13}$  [11] also exhibit a broad magnetic response with a width  $\Gamma \approx 25$  meV. Additional measurements probing the temperature dependence are now in progress.

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