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^{9}Be KNIGHT SHIFT IN THE NORMAL STATE OF UBe$_{13}$

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^{9}Be NMR spectra have been studied in the heavy-fermion compound UBe$_{13}$ as functions of spectrometer frequency, crystal orientation, and temperature. Be^{I}-site isotropic Knight shift and Be^{II}-site anisotropic shifts, corresponding hyperfine fields, and quadrupolar tensors have been determined. For $T \leq 10$ K a typical heavy-fermion Knight shift anomaly is observed.

1. Introduction

The intermetallic compound UBe$_{13}$ has attracted much attention since its discovery as a heavy-electron system [1], in large part because its superconducting transition at 0.84 K makes it one of the heavy-electron superconductors [2]. Previous nuclear magnetic resonance (NMR) studies of UBe$_{13}$ have concentrated on the ^{9}Be spin-lattice relaxation rate [3,4]. The noise power of local field fluctuations at ^{9}Be sites is three orders of magnitude larger than in the non-5f reference compound ThBe$_{13}$, and the relaxation rate in the superconducting state follows a power law in temperature instead of the conventional activated BCS temperature dependence. In addition, preliminary measurements of NMR spectra [3] indicated little change of frequency of line-width upon entereing the superconducting state.

In this paper we report measurements of ^{9}Be NMR absorption spectra in single-crystal UBe$_{13}$ in the normal state as a function of spectrometer frequency, crystal orientation, and temperature. Knight shifts, hyperfine fields, and quadrupolar couplings are derived from the data. The hyperfine fields are found to be no smaller than in isostructural 4fBe$_{13}$ compounds.

2. Experiment

UBe$_{13}$ crystallizes in the NaZn$_{13}$-type structure, with a fcc space lattice [5]. For our experiments we used a flux grown single crystal, dimensions $6 \times 4 \times 1$ mm^3, spark-cut with faces perpendicular to the cubic axes. A conventional NMR pulse method was used, with field calibration obtained from the ^{27}Al resonance of a reference sample next to the UBe$_{13}$ crystal.

The observed ^{9}Be ($I = 3/2$) field-swept NMR spectra consisted of ten lines due to the existence of two inequivalent Be sites, Be^{I} and Be^{II}. The Be^{I} site is of cubic point symmetry (m3), while the symmetry of the Be^{II} site is quite low (m), this leads to an anisotropic Knight shift and quadrupolar splitting [6] of the spectra from Be^{II} nuclei. After the individual lines were identified, the temperature dependence of the Knight shift was obtained for the inequivalent Be^{I} and Be^{II} sites. The relation between shift and bulk magnetic susceptibility was used to obtain the corresponding hyperfine fields. Approximate corrections for the demagnetization and Lorentz fields were

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made using the factors of an ellipsoid [7] Uncertainty in the extra field produced by the UBe$_{13}$ paramagnetic moment caused an error of ±0 01% in the absolute values of the Knight shifts

3. Results and discussion

3 1 Frequencydependence (T = 4 2 K)

The crystal was mounted with a (001) axis parallel to the external field, and data were taken for spectrometer frequencies $\omega_0/2\pi$ between 6 and 31 MHz (fields between 10 and 52 kOe) In fig 1 the relative line positions $\gamma H(i)/\omega_0$, corrected for demagnetizing and Lorentz fields, are plotted vs inverse frequency The data group into three triplets, each with slopes $(S, 0, -S)$ and the same intercept Conventional NMR theory yields $S = \omega_Q/[2(1+K)]$, where $\omega_Q/2\pi$ is the quadrupolar splitting frequency and K is the Knight shift, the intercept is $1/(1+K)$ These triplets are associated with Be^{II} sites In addition, the Be^{I} sites give a line of zero slope without satellites The signal amplitudes are consistent with these assignments

Knight shifts and quadrupolar splitting frequencies for nonidentical nuclei are obtained from figs 1–3, and are summarized in table 1 The three values of K^{II} and ω_Q^{II} values are the principle-axis values from the orientation dependence discussed in 3 2 below The isotropic (average) Be^{II} shift $(K_A^{II} + K_B^{II} + K_C^{II})/3 =$ 0 09(1)% at 4 2 K, which is enhanced by about

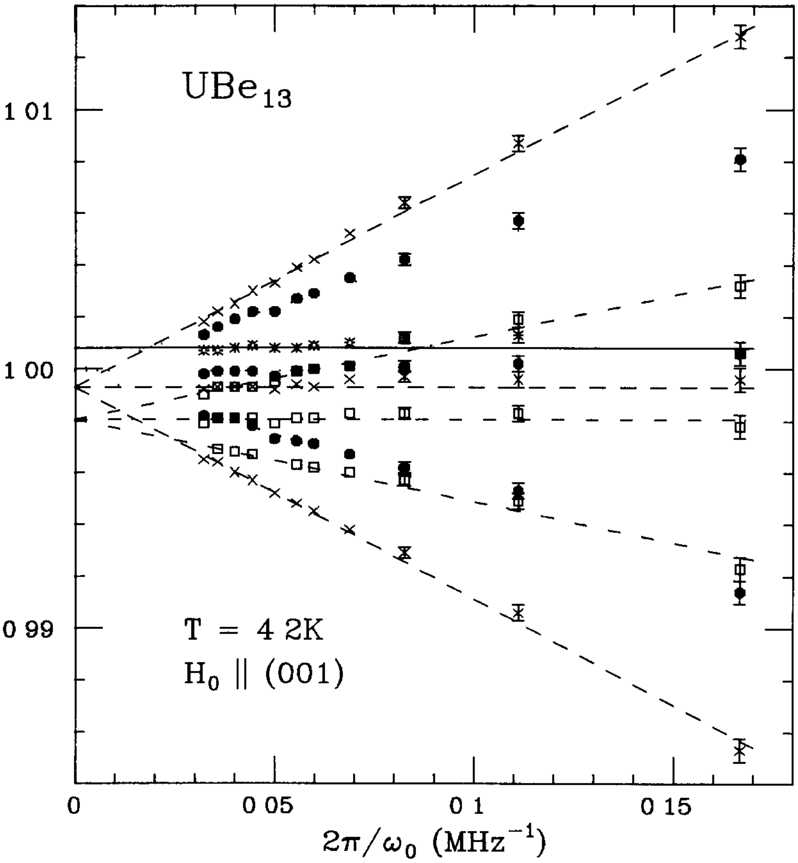

Fig 1 Relative ^{9}Be NMR line positions in single-crystal UBe$_{13}$ as functions of inverse spectrometer frequency $2\pi/\omega_0$ The lines are grouped into three triplets (×, ●, and □, dashed, dotted, and dash-dot lines) and a singlet (*, solid line) γ is the ^{9}Be gyromagnetic ratio, and $H(i)$ is the resonance field for line i The straight lines give linear least-squares fits to the data

Table 1
^{9}Be Knight shift at the Be^I site. ^{9}Be Knight shift and quadrupolar tensor principal-axis values at the three Be^{II} sites and hyperfine fields H_{hf} in UBe$_{13}$ ($T = 4.2$ K)

Site	K_i (%)	$\omega_{Qi}/2\pi$ (kHz)	$(H_{hf})_i$ (Oe/μ_B)
Be^I	−0.08(1)	-	−118
(Be^{II})$_A$	+0.07(1)	±164(2)^a	414
(Be^{II})$_B$	+0.01(1)	∓97(2)	147
(Be^{II})$_C$	+0.19(1)	∓64(2)	874
(Be^{II})$_{iso}$	+0.09(1)	±1(1)	478

[a] The relative signs of ω_{Qi} are obtained from the vanishing trace of the quadrupolar tensor ($\sum \omega_{Qi} = 0$).

25 compared to Be metal [8]. The shift $K^I = -0.08(1)\%$ is of the same order but of opposite sign. These values for the isotropic part of K indicate a large enhancement of the transferred hyperfine interaction in UBe$_{13}$. The data also yield values of the quadrupolar coupling constant $e^2qQ/h = 328(4)$ kHz and the quadrupolar asymmetry parameter $\eta = 0.20(2)$ at 4.2 K.

3.2. *Orientation dependence* (T = 4.2 K)

Fig. 2 gives the relative fields for resonance as functions of rotation angle about a (001) axis perpendicular to the external field. The observed extrema of fields for Be^{II} resonance lie along the (100) and (010) directions (0° and 90°, respectively), which indicates that both shift and quadrupolar tensor principal axes are oriented in these directions. The curves in fig. 2 were calculated with this assumption, using the values obtained as described in 3.1 above as principal values. There is good agreement, which confirms our assumption.

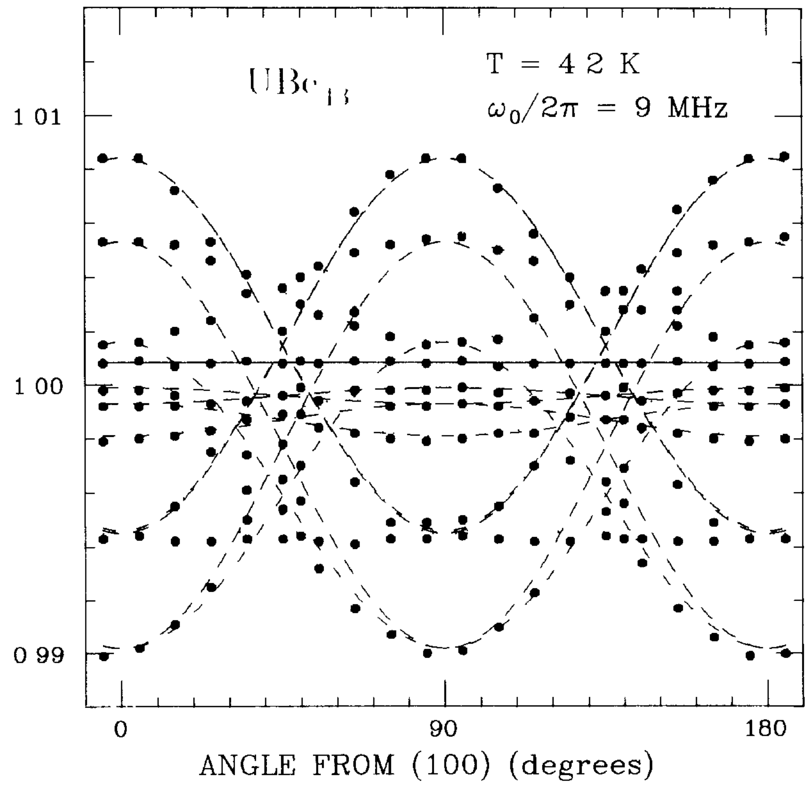

Fig. 2. Angular dependence of relative ^{9}Be NMR line positions. Curve designations as in fig. 1.

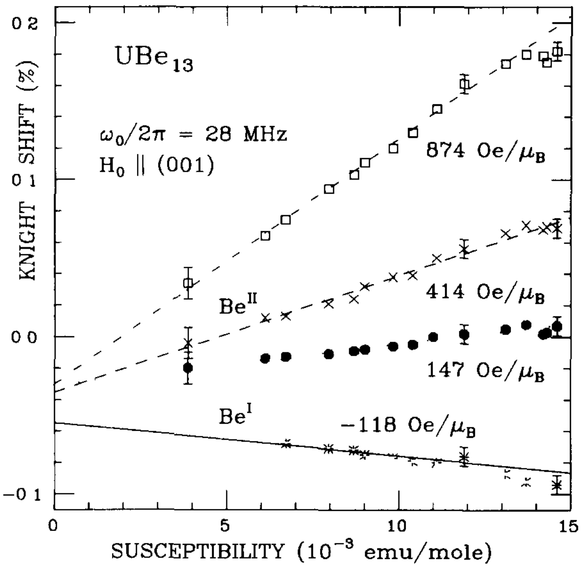

Fig. 3. Be^I Knight shift and Be^{II} principal-axis Knight shift values as functions of the molar susceptibility, with temperature an implicit parameter. Symbols and line designations as in fig. 1. The straight lines are linear least-squares fits to data for $\chi_m < 13 \times 10^{-3}$ emu mol^{-1} ($T \geq 15$ K).

3.3 Temperature dependence

The Be^I shift and the three princpal values of the Be^{II} shift tensor are plotted against the bulk molar magnetic susceptibility χ_m in fig. 3, with the temperature as an implicit variable. The data follow linear relations except for the deviations when $\chi_m \geq 13.5$ emu/mol ($T \leq 10$ K), where the shifts depend less on temperature than χ_m. Hyperfine fields H_{hf} were obtained from fits of the high-temperature linear dependences to the general relation $K(T) = (H_{hf}/N\mu_B)\chi_m(T)$, where N is Avogadro's number and μ_B is the Bohr magneton [9]. Values of H_{hf} are given in table 1 for all four resonances.

Although no comparable single-crystal NMR study of other 4f or 5f beryllides has been made, a Knight shift of less than 0.005% at 4.2 K has been reported for CeBe$_{13}$ [9]. This is an intermediate-valent compound with a susceptibility an order of magnitude smaller than that of UBe$_{13}$, so that an upper bound on H_{hf}(CeBe$_{13}$) is of the order of the measured value of H_{hf} in UBe$_{13}$. The latter is therefore not anomalously small.

A departure of $K(\chi)$ from linearity at low temperatures has been observed in several mixed-valent and Kondo-lattice compounds [10], which is associated with entry into a new physical regime. There is some evidence that this crossover is due to the onset of coherence in the heavy-electron system [10].

4. Conclusions

We have identified lines in the ^{9}Be nuclear resonance spectrum of single-crystal UBe$_{13}$ by measurements as a function of spectrometer frequency and external field orientation. The isotropic Knight shifts at 4.2 K for both Be^I and Be^{II} sites are enhanced by a factor of ≈25 with respect to Be metal, due to the high density of heavy-electron states at the Fermi surface. The temperature dependence exhibits a departure from a linear $K(\chi)$ relation below ≈10 K, which identifies a crossover at this temperature to a low-temperature coherent Fermi-liquid state.

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