Nuclear magnetic resonance in the heavy-fermion superconductor UBe_{13}

N. E. Alekseevskiĭ and E. G. Nikolaev

Institute for Problems in Physics, and Institute for High-Pressure Physics, USSR Academy of Sciences
(Submitted 24 April 1986)
Zh. Eksp. Teor. Fiz. **91**, 1820–1831 (November 1986)

We present the results of NMR investigations of ^{9}Be nuclei in the compound UBe_{13} when this compound is in its normal (i.e., nonsuperconducting) state. From the spectra of polycrystalline and single-crystal UBe_{13} samples we determine the isotropic NMR shifts for inequivalent positions of the beryllium atoms, and also the components of the anisotropic shift and electric field gradient tensors for the BeII position. We show that the values of the isotropic shift have different signs for the BeI and BeII positions; this fact can be explained by the presence of spatial oscillations in the spin polarization of conduction electrons around the magnetic uranium atoms. Measurements under hydrostatic compression show that the shift at the beryllium atoms in UBe_{13} decreases with pressure, which apparently indicates a decreased magnetic susceptibility for this system. We discuss how our results are connected with the possibility of *sf*-hybridization and formation of a heavy-carrier band in the compounds under investigation.

INTRODUCTION

Recently much attention has been directed toward investigating systems of heavy fermions.[1,2] The characteristic signature of a heavy-fermion compound is an anomalously large value of the electronic specific heat coefficient γ at low temperatures (~ 1 J/mole K^2); this fact allows us to postulate that a very narrow band ($T_F \sim 10$ K) is present in these systems near the Fermi level with a large density of states and effective carrier masses on the order of 200 m_e. Particular interest attaches to those heavy-fermion compounds in which superconductivity is observed. The value of the critical temperatures in this case is less than 1 K, and for the compounds $CeCu_2Si_2$ and UBe_{13} the derivative of the upper critical field with respect to temperature near T_c exceeds 200 kOe/K.[1] We should also note the large value of the specific heat discontinuity at the superconducting transition point, which agrees with the anomalous value of γ in these systems. These unusual properties, along with such facts as, e.g., the nonexponential character of the temperature dependence of the specific heat below T_c, support the hypothesis that electron pairing in the superconducting state of heavy-fermion systems has a triplet character.[2] Other explanations have also been advanced to explain the behavior of the specific heat in the superconducting state and the large value of $(\partial H_{c2}/\partial T)T_c$ (Ref. 4). The magnetic properties of heavy-fermion compounds are to a significant degree determined by the presence in these compounds of atoms with *f*-electrons. The temperature dependence of the magnetic susceptibility at high temperatures for the majority of these systems follows the Curie law, which is a characteristic of systems with localized moments.[1] In the region of very low temperatures (below 4.2 K), in many cases a temperature-independent susceptibility is observed,[5] which can be considered a consequence of the formation of a Fermi liquid of heavy electrons.[6] In some publications[7] this last circumstance has been associated with the formation of a Kondo lattice.

There is also an indubitable interest in investigating the local magnetic characteristics of these systems by the method of magnetic resonance, in order to ascertain the causes of the anomalous properties of heavy-fermion compounds. In this paper we present the results of NMR experiments on ^{9}Be nuclei in the heavy-fermion superconductor UBe_{13}. This compound has the $NaZn_{13}$ type of cubic structure (see Fig. 1) formed by atoms of uranium and icosahedral Be_{13} clusters. In each cluster there are two kinds of crystallographically inequivalent sites for beryllium atoms—one atom (BeI) in the center, and twelve atoms (BeII) at the vertices of the icosahedra. The positions of atoms in a unit cell are distributed in the following fashion: 8 U atoms at the positions $\pm(1/4,1/4,1/4)$; 8 BeI at (0,0,0) and (1/2,1/2,1/2); 96 BeII at $\pm(0,y,z)$, $\pm(0,y,\bar{z})$, $\pm(1/2,z,y)$, $\pm(1/2,\bar{z},y)$ plus all sites related to these by a threefold rotation, along with the face-centered positions equivalent to them. Here $y = 0.1763$, $z = 0.1150$ (Refs. 8,9). As we will show below,

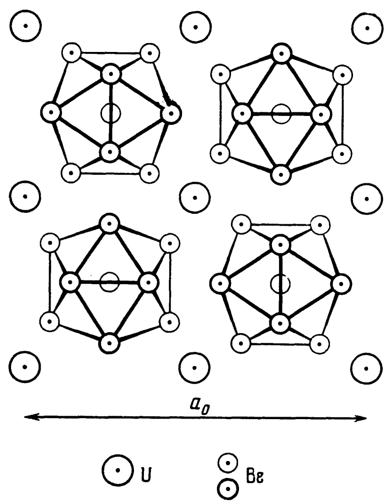

FIG. 1. Structure of the compound UBe_{13}.

 0038-5646/86/111078-07$04.00

such a structure gives rise to NMR spectra of ^{9}Be in UBe_{13} which are extremely complex. For this reason, our investigation was conducted both on polycrystalline and single-crystal samples.

SAMPLE PREPARATION AND EXPERIMENTAL METHODS

Polycrystalline samples of the compound UBe_{13} were obtained by fusing the original constituents in an induction furnace. The single crystals were also grown in an induction furnace by slow cooling of the melt in a beryllium-oxide crucible. An X-ray diffraction analysis showed that all the samples were practically single-phase, and had lattice constants close to the value 10.254 Å. The single-crystal sample used for the NMR experiments had a lattice constant of 10.257 Å. We should point out that the value of the critical temperature in this case, which was measured to be 0.55 K,[1] differed significantly from T_c for the majority of samples (0.9 K). This fact can be related to the presence of finite quantities of impurities or defects in the sample in question. As shown in Refs. 11 and 12, a small quantity of impurities and defects strongly lowers the critical temperature, although such impurities turn out not to affect the magnitude of the specific heat and magnetic properties of the compound UBe_{13} significantly while it is in the normal state. In our case, we also observed very good agreement with regard to a number of NMR spectral parameters of ^{9}Be between polycrystalline samples with high values of T_c and single-crystal samples with relatively low values of T_c.

For the NMR experiments, the polycrystalline samples were ground into powder in an agate mortar and mixed with paraffin. The single-crystal sample used for NMR was a stack of height 5.4 mm made of 13 plates, each with dimensions $0.35\times3.9\times4.9$ mm^3, pasted together with paper. The orientation of the plates was such that in the plane of the films a fourfold axis, a threefold axis and a twofold axis were all present; a further twofold axis was directed perpendicular to the films.

NMR measurements were carried out on a stationary autodyne NMR spectrometer with a superconducting magnet[13] in the temperature range 1.8 to 100 K and in magnetic fields up to 32 kOe. For experiments under pressure the RF circuit with the sample was placed in a beryllium-bronze cylinder with a channel diameter of 8.5 mm, making it possible to apply pressures up to 10 kbar. A special sample holder was used in studies of the single crystal, which allowed the sample to be oriented relative to the magnetic field. Measurements of the susceptibility were carried out on a magnetic balance, using the Faraday method.

EXPERIMENTAL RESULTS

In Fig. 2 we show a ^{9}Be NMR spectrum for a polycrystalline sample of UBe_{13}; the spectrum was obtained at a frequency of 13 MHz and a temperature of 4.2 K. As is clear from the figure, the spectrum consists of two central components and several satellites. Measurements at various frequencies show that the form of the spectrum is due to a complex combination of magnetic and electric hyperfine interactions. The presence of two central components is apparently related to the fact that in the UBe_{13} structure there are two inequivalent beryllium positions, which correspond to different values of the isotropic shift. The central component has an asymmetric shape which is characteristic of an

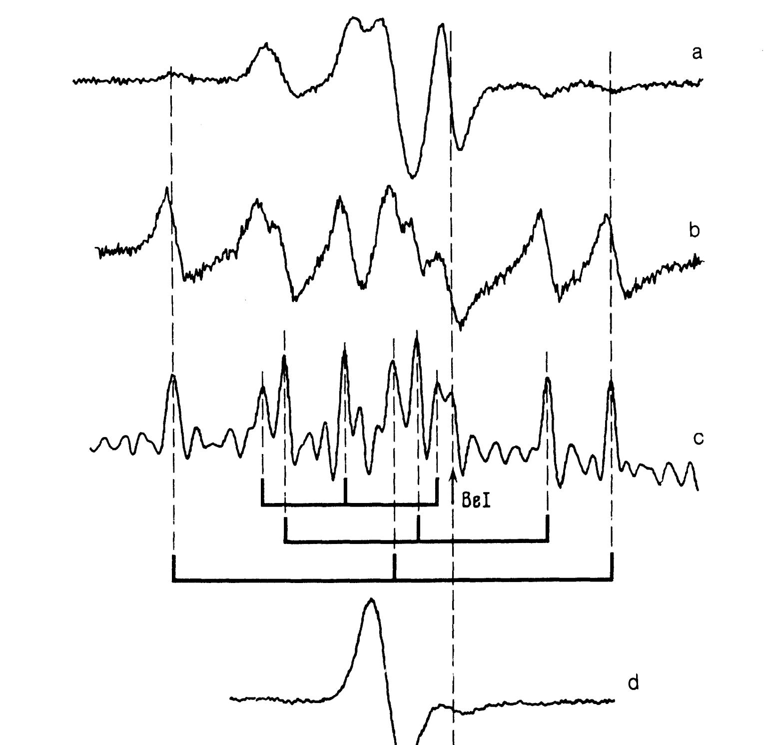

FIG. 2. Spectra of ^{9}Be in the compound UBe_{13}, obtained at a frequency of 13100 kHz for $T=4.2$ K; a—spectrum of a polycrystalline sample, b—spectrum of a single crystal oriented so that [100]‖**H**, c—results of processing the single-crystal spectrum for the [100]‖**H** orientation. The dotted lines show the separate NMR lines (see text). Below it we show individual groups of lines for the inequivalent BeII positions relative to the direction of the magnetic field. The arrow shows the position of the BeI line. d—single-crystal spectrum for the orientation [111]‖**H**. The experimental spectra a, b, and d are derivatives of the NMR signal; spectrum *c* is an absorption spectrum with line narrowing.

anisotropic shift; in addition, the quadrupole satellites must also correspond to the low-symmetry BeII positions. The second central component is probably related to the BeI position, for which the anisotropic shift and quadrupole interactions must be absent as a consequence of the high symmetry. Analysis of the ratio of the intensities agrees with this interpretation of the spectrum for polycrystalline samples of UBe_{13}.

The interpretation described above receive further confirmation when the single crystal was investigated. In Fig. 2b we show the ^{9}Be NMR spectrum of a sample for which the fourfold axis was oriented along the constant magnetic field ([100]||**H**); this spectrum was obtained under the same conditions as the polycrystalline spectrum presented above. It is clear from the figure that some lines overlap to a considerable degree, making analysis and processing of the spectrum difficult. In order to resolve these lines, the spectrum was processed in the following way: using the spectral lines farthest to the left and right (the "boundary" lines), which have insignificant overlap with the remaining components, we reconstructed the shape of an isolated line of this spectrum. Here we should note that in this case the width and shape of the lines are apparently identical for all the spectral components, since they are primarily determined by the distribution of magnetization in the sample (due to the large value of the susceptibility and the sample's nonellipsoidal shape) and by a dispersive admixture to the signal due to the fact that the plate thicknesses in the sample are on the order of the skin depth.(2) Then the single-crystal spectrum can be written in the following form:

$$G(H)=\int_{-\infty}^{+\infty} g(H')A(H-H')dH', \qquad (1)$$

where $G(H)$ is the single-crystal spectrum including line broadening and $A(H)$ is the shape of an isolated line; $g(H)$ is the single-crystal spectrum without including line broadening. We can determine the positions of the individual lines once we have evaluated $g(H)$. The function $g(H)$ was found by Fourier transforming (with preliminary optimized filtering using the FILTER program) both the isolated line shape $A(H)$ and the total spectral shape $g(H)$, using the method described in the paper by Kosarev and Pantos.[14] Calculations were performed on the HP-1000 computer at the Institute for Problems in Physics, USSR Academy of Sciences. The results are shown in Fig. 2c. As is clear from the figure, all the lines are well resolved, and their positions and centers can be identified with some confidence; however, some oscillations did appear due to the absence of higher harmonics in the Fourier transform. Subsequently we evaluated the number of lines in the spectrum more precisely, and also their positions, by using the method of least squares. Ten lines were resolved in all, whose positions are shown in Fig. 2 by the dashed lines.

We can write the following expression[15] for the positions of the NMR lines in the single-crystal spectrum, taking into account the anisotropic shift and quadrupole interaction to first order in perturbation theory(3)

$$H_{m\to m-1}=H_0(1-K_{iso})-\frac{K_{an}^{ax}}{2}H_0(3\cos^2\theta_1-1)+\frac{K_{an}^{as}}{2}H_0\sin^2\theta_1\cos 2\Phi_1-\frac{2\pi}{\gamma_n}\frac{\nu_Q}{2}\left(m-\frac{1}{2}\right)(3\cos^2\theta_2-1)+\frac{2\pi}{\gamma_n}\frac{\nu_Q\eta}{2}\left(m-\frac{1}{2}\right)\sin^2\theta_2\cos 2\Phi_2, \qquad (2)$$

where $H_0=\nu_0/(\gamma_n/2\pi)$, γ_n is the nuclear gyromagnetic ratio, ν_0 is the frequency of the RF field, K_{iso} is the isotropic shift, and m is the nuclear spin projection along the direction of the magnetic field; for the case of a ^{9}Be nucleus, m takes on the values 3/2, 1/2, and $-1/2$ in formula (2), while

$$K_{an}^{ax}=K_{an}^{z}, \qquad K_{an}^{as}=K_{an}^{y}-K_{an}^{x},$$

K^x_{an}, K^y_{an}, K^z_{an} are the components of the anistropic shift tensor in principal axis coordinates;

$$|K_{an}^{y}|\leqslant|K_{an}^{x}|\leqslant|K_{an}^{z}|,$$
$$\nu_Q=3e^2qQ/2I(I-1)h$$

where ν_Q is the quadrupole frequency, eQ the nuclear quadrupole moment, I the nuclear spin, $eq=V_{zz}$ [in the case of ^{9}Be, $I=3/2$ and $(\nu_Q=V_{zz}eQ/2h)$]; $\eta=(V_{yy}-V_{xx})/V_{zz})$ is the asymmetry parameter, where V_{xx}, V_{yy} and V_{zz} are the tensor components of the electric field gradient along principal axes;

$$|V_{yy}|\leqslant|V_{xx}|\leqslant|V_{zz}|,$$

θ_1,Φ_1 and θ_2,Φ_2 are the polar and azimuthal angles for the magnetic field direction relative to the principal axes of the anisotropy-shift tensor and the corresponding axes for the electric field gradient. Expression (2) is valid up to the terms of order $(K_{iso}+K_{an})^2H_0$ and $(K_{iso}+K_{an})\nu_Q(2\pi/\gamma_n)$.

For the case of **H** parallel to the fourfold axis, there are three types of inequivalent positions of the BeII relative to the magnetic field directions (these are labeled with the letters *a*, *b* and *c* in Fig. 3); each of these corresponds to a set of angles $\theta_1,\Phi_1,\theta_2,\Phi_2$. In agreement with Eq. (2), each of these

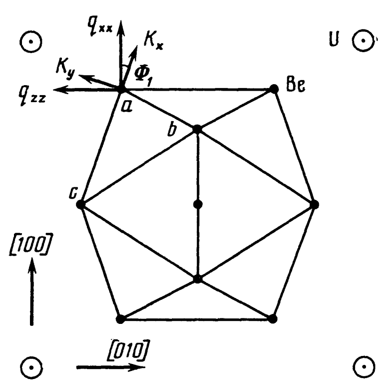

FIG. 3. Directions of the principal axes of the tensor K_{an} and the electric field gradient tensor for the BeII position. The letters *a*, *b*, and *c* denote the three inequivalent BeII positions relative to the magnetic field [100]||**H**. The plane of the drawing coincides with the (001) plane, which for position *a* is a reflection plane. Only the axis lying in the plane of the drawing is shown. The directions of the *Z* axis for K_{an} and the *Y* axis for the electric field gradient at position *a* are perpendicular to the plane of the figure. The angle Φ_1 is obtained by calculating the tensor K_{an}, assuming direct dipole interactions (see text), and equals 19.3°.

TABLE I.

Sample	K_{iso}^{I}, %	K_{iso}^{II}, %	K_{an}^{ax}, %	K_{an}^{as}, %	ν_Q, kHz	η
Polycrystalline	−0.085±0.02	~0.08	~0.10	–	83.0±1.0	–
Single-crystal л	−0.083±0.02	0.107±0.02	0.108±0.005	0.077+0.005*	83.0±0.2	0.20

*For the angle Φ_1 (Fig. 3), which equals 19.3°.

positions gives rise to a group of three equally-spaced lines in the spectrum (Fig. 2c). The spacing between the lines in each group is determined by the anisotropic shift for a given BeII position. Consequently, for a given orientation the spectrum contains nine lines originating from the BeII positions. The tenth line, shown by the arrow in Fig. 2c, must belong to the BeI position.

For the BeII positions in the UBe_{13} structure, there is one symmetry element—a reflection plane perpendicular to the fourfold axis of the crystal which connects the nearest uranium atom to the beryllium atom in question. It is obvious that one of the principal axes of the anisotropic shift tensor and one of the principal axes of the electric field gradient tensor must be perpendicular to this plane, and consequently must coincide with the fourfold axis. The two other principal axes of the tensors K_{an} and the electric field gradient must lie in the reflection plane.

To determine the directions of the principal axes of the anisotropic shift tensor, we performed a calculation of K_{an} using formulae from Ref. 16, assuming direct dipole interaction between the beryllium nuclei and the magnetic moments of the uranium atoms. In the calculation, contributions from all the uranium atoms in a sphere of 50 Å radius around a given beryllium atom were summed. The results showed that the Z axis of the tensor K_{an} had to be perpendicular to the reflection plane. In Fig. 3 we show the directions of the X and Y axes of the tensor K_{an}, which lie in the reflection plane. To determine the directions of the principal axes of the electric field gradient tensor, we took into account the fact that the positions of the outermost quadrupolar satellite lines in the polycrystalline spectrum (Fig. 2a) coincided closely with the position of the outermost lines in the single crystal spectrum for the orientation [100]||**H** (Fig. 2c). It is also essential that these lines belong to one and the same group, corresponding to the position *a,b* or *c* having the largest quadrupole splitting (Fig. 2c). According to Eq. (2), this implies that for the position in question the condition $\theta_2 = 0$ holds, and consequently that the Z axis of the electric field gradient tensor coincides with the direction of the magnetic field and with one of the fourfold axes. If we assume that the directions of the principal axes of the tensor K_{an} are close to our calculated directions, then analysis of the spectrum for the [100]||**H** orientation shows that all the principal axes of the electric field gradient tensor must coincide with the fourfold axes, as shown in Fig. 3. In this case, the angles for which the quadrupole splitting vanishes by virtue of the conditions

$$3\cos^2\theta_2 - 1 = 0, \qquad \cos 2\Phi_2 = 0,$$

must correspond to an orientation in which the third-order axis is parallel to the direction of the magnetic field ([111]||**H**). In this case, the axial component of the anisotropic shift must also go to zero, since $3\cos^2\theta_1 - 1 = 0$. In Fig. 2d we show the NMR spectrum of ^9Be in single-crystal UBe_{13} for [111]||**H**. As is clear from the figure, the spectrum in this case contains both lines, one of which (the right) coincides in position with the BeI line in the previous spectrum. The significantly more intense line on the left corresponds to the BeII position. The width of this line is quite a bit larger than that of the BeI line and those of the individual lines in Fig. 2b. One reason for this is apparently that for the orientation in question $\cos 2\Phi_1 \neq 0$; consequently there is a small unresolved splitting connected with the asymmetric components of the anisotropy shift. Another reason could be nonideal orientation of the sample relative to the magnetic field direction, which gives rise to a small quadrupole splitting.

The table presents values of the components of the K_{an} and electric field gradient tensors for positions BeII, and also the isotropic shifts for the inequivalent positions BeI and BeII, as determined from the spectra of a polycrystalline sample and a single crystal at 4.2 K. It should be noted that the values of the components of the K_{an} tensor given in the table differ from the corresponding quantities obtained from calculations. Thus, the calculated value of K_{an}^{ax} amounts to 0.189%, which exceeds the experimental value given in the table. Possible reasons for this discrepancy will be discussed in the following sections; however, the existence of such a discrepancy between experiment and calculation suggests that the actual directions of the principal axis of the K_{an} and electric field gradient tensors for the BeII positions may differ from those shown in Fig. 3. Nevertheless, starting with the experimental results and the considerations stated above, we can confirm that the directions of the principal axes of the electric field gradient tensor coincide fairly closely with the fourfold axes. The direction of the Z principal axis of the K_{an} tensor also, apparently, coincides with one of the fourfold axes. Additional confirmation of this comes from the fact that, as is clear from the table, the estimate of K_{an}^{ax} from the polycrystalline sample is less than the corresponding value for the single crystal. The angle between the X axis for K_{an} and the X axis for the electric field gradient tensor (Φ_1 in Fig. 3) is less than 45°. This follows from an investigation based on Eq. (2) of the relation between the quadrupole splitting and the relative position of each of the groups of lines in Fig. 2c.

Measurements carried out on a polycrystalline sample at various temperatures show that the temperature depen-

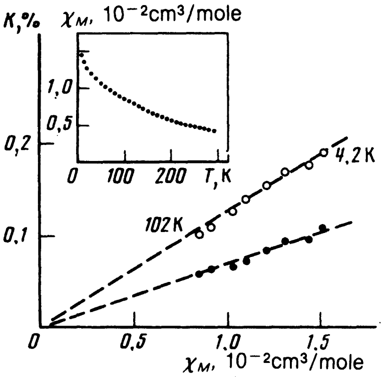

FIG. 4. Dependence of the quantity ($K^{\rm II}_{\rm iso} - K^{\rm I}_{\rm iso}$) (O) and $K^{\rm ax}_{\rm an}$ (●) on the magnetic susceptibility of the compound UBe_{13}. The inset shows the temperature dependence of the magnetic susceptibility of UBe_{13}.

dence of the shifts correlate with the temperature dependence of the magnetic susceptibility of the compound UBe_{13}. In Fig. 4 we show the dependence on magnetic susceptibility of the quantity ($K^{\rm II}_{\rm iso} - K^{\rm I}_{\rm iso}$), which characterizes the relative splitting between the BeI and BeII positions, and that of the quantity $K^{\rm ax}_{\rm an}$. The values of ($K^{\rm II}_{\rm iso} - K^{\rm I}_{\rm iso}$) and $K^{\rm ax}_{\rm an}$ are correlated once we take into account the results for the single crystal. Analogous dependences are observed also for each of the quantities $K^{\rm I}_{\rm iso}$ and $K^{\rm II}_{\rm iso}$. Here we also present a plot of the temperature dependence of the susceptibility in UBe_{13}. In the polycrystalline samples we also obtained NMR spectra of ^{9}Be under conditions of isotropic compression for 4.2 K. We established that the splitting between the BeI and BeII positions decreases under pressure, as does the anisotropic shift at the position of the BeII. The derivatives

$$\partial \ln(K^{\rm II}_{iso}-K^{\rm I}_{iso})/\partial p \text{ and } \partial \ln K_{an}{}^{ax}/\partial p$$

amount to $(-9\pm 2)\times 10^{-3}$ kbar^{-1}.

DISCUSSION OF RESULTS

As is clear from the table, the isotropic shifts for the inequivalent positions BeI and BeII differ significantly; furthermore, they have opposite signs. This circumstance, and also the strong temperature dependence of the shifts which is correlated with the temperature dependence of the susceptibility (Fig. 4), is connected in a fundamental way with the presence in the system of uranium atoms which contain $5f$ electrons. In Fig. 5 we show the central part of the ^{9}Be NMR spectrum for a polycrystalline sample of the isostructural compound $ThBe_{13}$. Thorium, as is well-known, has no $5f$ electrons; as is clear from the figure, in this case we observe only a single central component of the isotropic shift and magnetic susceptibility are considerably smaller here than in UBe_{13}, and depend only weakly on temperature.

Dependences of the shifts on the susceptibility similar to those shown in Fig. 4 are observed in a whole series of intermetallic rare-earth compounds.[17] In these systems it is postulated that fields at the nonmagnetic atoms are caused by the polarization of conduction electrons by the magnetic moments of the rare-earth atoms. The shift at the nucleus of the nonmagnetic atoms in these systems is described by the following expression

$$K=K_0\left(1+\frac{g-1}{g}\frac{\Gamma_{sf}}{N_A\mu_B}\chi_f\right), \tag{3}$$

where K_0 is the shift in the isomorphic nonmagnetic compound, g is the g-factor of the rare-earth atom, N_A is Avogadro's number, μ_B is the Bohr magnetron, χ_f the molar susceptibility, and Γ_{sf} is a parameter which characterizes the sf interaction. Different values of the shifts for inequivalent positions can be considered as consequences of spatial oscillations in the spin polarization around the magnetic atoms. A qualitative description of this phenomena can be obtained within, e.g., the Ruderman-Kittel model,[18] which involves the usual exchange interaction between conduction electrons and localized point moments. This model predicts

$$\Gamma_{sf}=-6\pi Z J_{sf}\sum_i F(2k_F R_i), \tag{4}$$

where $F(x) = (x\cos x - \sin x)/x^4$ is the Ruderman-Kittel function, Z is the number of conductions per atom, k_F is the Fermi momentum, R_i is the distance from a given nucleus to the ith rare-earth atom, and J_{sf} is the exchange integral for the sf interactions. The oscillatory character of the function $F(x)$ can lead to a situation where the values of the sum $\Sigma_i F(2k_F R_i)$ for inequivalent positions will differ significantly, and consequently the shifts will differ for inequivalent positions. This model can apparently be used for the case of sufficiently well-localized $4f$ states located considerably below the Fermi level.

Oscillations in the spin density around magnetic atoms are also observed in dilute alloys of copper in $3d$-elements.[19] We should note that in these systems the $3d$ states have an appreciably more delocalized character and are located relatively close to the Fermi level. To describe such systems, Friedel and Anderson developed an approach based on a model of virtual bound states.[20,21] The interaction of conduction electrons with magnetic atoms in this case is determined primarily by hybridization, and the effective sd exchange integral has a negative sign. The asymptotic form of the spin density oscillations around a magnetic atom in their model coincides with the Ruderman-Kittel oscillations;

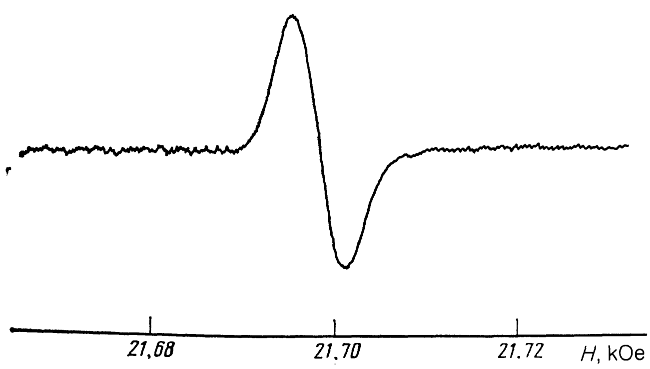

FIG. 5. Central component of the ^{9}Be spectrum for the compound $ThBe_{13}$ (derivative of the NMR signal), taken at a frequency of 12980 kHz for $T = 4.2$ K.

however, the short-distance behavior of these oscillations is markedly different.[19]

Apparently this approach is more correct for a description of the interaction of conduction electrons with the 5f states of uranium. These states also are significantly more delocalized than the 4f electrons of rare-earth atoms. The data presented in this paper on the shifts in UBe_{13} can be compared with the results of NMR studies of the isostructural compound $NdBe_{13}$.[22] It is well-known that trivalent neodymium is a 4f analogue of trivalent uranium; the authors of Ref. 22 established that the isotropic shift at the BeII position was negative and amounted to -0.2% at 77 K. The quantity K_{iso} at the BeI position was not given in this paper. For UBe_{13}, the isotropic shift at the BeII position has a positive value for the entire range of temperatures investigated, from 1.8 K to 100 K. This difference can be considered to be a consequence of differences in the spatial distribution of the spin-polarized conduction electrons. In the case of $NdBe_{13}$ this distribution is apparently closer to the Ruderman-Kittel model, whereas for UBe_{13} the distribution of spin-polarized conduction electrons has a different character due to the important role of sf hybridization.

As we mentioned in the previous section, the experimental value of the axial component of the anisotropic shift tensor in UBe_{13} at the BeII position is almost twice as small as the value calculated by assuming direct dipole interactions between the beryllium nuclei and the magnetic moments of the uranium atoms. This discrepancy can be related to the fact that in our calculations we did not take into account the finite magnetic moments of the uranium atoms. Actually the radius of the 5f orbitals is rather larger (~ 0.8 Å) and their spatial configuration in this case are not spherically symmetric. This latter circumstance, as was noted in Ref. 22, can be important when crystal field effects are present. In addition, this additional contribution to the anisotropic shift for positions whose symmetry is lower than cubic can lead to anisotropic components in the magnetization of the conduction electrons, which is also caused by their interaction with the localized moments.[19] Comparing the calculated and experimental values of K_{an}^{ax}, we can conclude that in the case of UBe_{13} this contribution is apparently less than half the dipole contribution and must have a negative sign.

As we already noted in the Introduction, the large value of the electronic specific heat coefficient in heavy-fermion systems at low temperatures suggests the presence in these systems of a very narrow band of carriers with large effective mass. In Ref. 4 it was shown that several other properties of UBe_{13}, e.g., the anomalous Hall effect at low temperatures, could be explained assuming that there exists two bands of carriers—a heavy-carrier band and the usual conduction band made up of light carriers. If we make this assumption, then the heavy-carrier band must apparently be related to the 5f states of the uranium; however, the formation of a band due to the direct overlap of the f-orbitals in the present case is highly improbable, since the spacing between uranium atoms in this system exceeds 5 Å. Apparently, the hybridization of the 5f states of uranium with the light-carrier band, i.e., the conduction band, plays an important role in the formation of the heavy-fermion band. We must point out that in this case the localized character of the 5f states of uranium must be preserved to a considerable degree.

The nature of the heavy-fermion band is not clear at this time. In the phenomenological model of Overhauser and Appell[23] proposed to explain the properties of UBe_{13}, the 5f state is located exactly at the Fermi level, and the width of the band is directly determined by hybridization. A similar situation is encountered in compounds with intermediate valence[24]; however, characteristic values of the width of the f-band for these systems are at least an order of magnitude larger. In the Kondo-lattice model, used to explain the properties of heavy-fermion compounds with cerium,[7] the f states are located somewhat below the Fermi level, and the heavy-fermion band forms because of collective effects in the system of ordered Kondo centers. It is unclear to what extent this model is applicable to uranium compounds. However, as was noted in Ref. 2, the unstable character of the 5f state, which can be related to the closeness of these states to the Fermi level, as well as hybridization with conduction electrons, plays an important role in the formation of the heavy-fermion band in UBe_{13}.

The nearness of the 5f states to the Fermi level is manifested in the significant influence of pressure on the NMR shift at the beryllium nuclei in UBe_{13}. The isotropic shift at the nucleus of a nonmagnetic atom, as is clear from Eq. (3), is proportional to the susceptibility of the localized moments of the f-atoms and to the sf interaction parameter. Therefore, these quantities can be significantly changed by pressure in the system under discussion here. As we pointed out above, the primary contribution to the anisotropic shift in UBe_{13} is given by the direct dipole interaction, and consequently its change under pressure must be connected with the change in susceptibility. As we pointed out in the previous section, the isotropic and anisotropic shifts decrease under pressure, and the logarithmic derivatives of these quantities with pressure are equal to one another. Consequently, we can assume that the decrease in these shifts with pressure is apparently due to the decrease of the magnetic susceptibility with pressure, and that the quantity $(\partial \ln \chi/\partial p)_{4.2\,K}$ is of the same order of magnitude as the corresponding quantity for the shifts, i.e., -10^{-2} kbar^{-1}. If we choose the error in determining the change of the shifts with pressure as an upper limit on the variation of the sf interaction, then $|\partial \ln J_{sf}/\partial p| \sim 2\cdot 10^{-3}$ kbar^{-1}. To confirm this assumption it is necessary to measure directly the change in magnetic susceptibility with pressure. The decrease in the susceptibility with pressure for UBe_{13} can be regarded as a consequence of a decrease in the degree of filling of the f states, because under pressure the f level shifts upward relative to the Fermi level.

It should also be noted that in our preliminary data on UBe_{13} we observe a very weak effect of pressure on the critical temperature for the superconducting transition ($\partial T_c/\partial p \sim 5\times 10^{-3}$ K/kbar). For normal superconductors this quantity is -10^{-2} K/kbar, while for certain Chervil phases the quantity $\partial T_c/\partial p$ is considerably larger; in the case of Mo_6S_8Sn it reaches a value of -0.15 K/kbar. In contrast to UBe_{13}, no significant effect of pressure on the susceptibility is observed in these systems with iron impurities, while the magnitude of the exchange interaction of conduction electrons with the localized moments is significantly increased by pressure.[25] (In Ref. 25, $\partial \ln J/\partial p \sim 2\times 10^{-2}$ kbar^{-1}).

CONCLUSION

In this paper we have investigated nuclear magnetic resonance of ^{9}Be nuclei in the heavy-fermion superconductor UBe_{13}. NMR spectra were obtained both for polycrystalline samples and single crystals, for the orientation [100]||**H** and [111]||**H**. We determined the isotropic shifts for the inequivalent positions of the beryllium atoms, and also the tensor components of the anisotropic shift and electric field gradient for the low-symmetry BeII position. We established that the tensor K_{an} and the electric field gradient tensor at these positions both have a significant nonaxial character, while the directions of their principal axes do not coincide with each other. The isotropic shifts for the BeI and BeII positions have opposite signs; this fact may be explained by spatial oscillations in the spin density around the magnetic moments of the uranium atoms, which arise from the interaction of conduction band electrons with the 5*f* states of uranium. This interaction is apparently determined to a significant degree by *sf* hybridization. The important role of hybridization in the *sf* interaction is connected with the fact that the 5*f* states of uranium in UBe_{13} are located close to the Fermi level. This circumstance leads to a significant decrease in the shifts with pressure in these systems.

In conclusion, Eq. (3) of the previous section can provide an estimate of the parameter Γ_{sf} for UBe_{13}. The absolute value of the isotropic shift of ^{9}Be in the compounds $LaBe_{13}$[22] and $ThBe_{13}$ (in the present work) is less than 0.01%. If we use this value as K_0, take K and χ_f from the experimental data at 4.2 K and assume that the g-factor in UBe_{13} is close to the corresponding value for trivalent neodymium (0.727), then the absolute value of Γ_{sf} is found to be of order 0.1 eV for the BeI and BeII positions. Thus, the effective exchange interaction in UBe_{13} is rather large, which is one of the interesting properties of this superconducting compound; further investigations are needed to explain this fact.

The authors are grateful to V. I. Nizhankovski and E. P. Khlybov for help with the experiment, E. L. Kosarev for help in processing the results on the computer, and also to co-worker I. Warchulski at the International Laboratory for Strong Magnetic Fields and Low Temperatures (Wroclaw, Poland) for help in carrying out the measurements of the magnetic susceptibility.

(1) Measurements of the critical temperatures for the superconducting transition in UBe_{13} samples were carried out in an erbium-aluminum-garnet adiabatic demagnetization apparatus.[10]

(2) We took into account the correction to the line center position due to admixture of dispersion in the signal when we determined the isotropic shift. The correction to the isotropic shifts due to the demagnetizing field was less than the errors given in the table below.

(3) The second-order quadrupole splitting was less than 0.6 Oe at a frequency of 13 MHz, and hence was not included.

1. G. R. Stewart, Rev. Mod. Phys. **56**, 755 (1984).
2. N. E. Alekseevskiĭ and D. I. Khomskiĭ, Usp. Fiz. Nauk **147**, 767 (1985) [Sov. Phys. Usp. **28**, 1136 (1985)].
3. H. R. Ott, H. Rudigier, T. M. Rice, K. Ueda, Z. Fisk, and J. L. Smith, Phys. Rev. Lett. **52**, 1915 (1984).
4. N. E. Alekseevskiĭ, V. N. Narozhniĭ, V. I. Nizhanovskiĭ, E. G. Nikolaev, and E. P. Khlybov, Pis'ma Zh. Eksp. Teor. Fiz. **40**, 421 (1984) [JETP Lett. **40**, 1241 (1984)].
5. B. Batlogg, J. P. Remeika, A. S. Cooper, and Z. Fisk, J. Appl. Phys. **55**, 2001 (1984).
6. T. M. Rice, K. Ueda, H. R. Ott, and H. Rudigier, Phys. Rev. **B31**, 594 (1985).
7. N. B. Brandt and V. V. Moshchalkov, Adv. Phys. **33**, 373 (1984).
8. E. Bucher, J. P. Maita, G. W. Hull, R. C. Fulton, and A. S. Cooper, Phys. Rev. **B11**, 440 (1975).
9. A. I. Goldman, S. M. Shapiro, D. E. Cox, J. L. Smith, and Z. Fisk, Phys. Rev. **B32**, 6042 (1985).
10. N. E. Alekseevskiĭ, A. P. Dodokin, C. Bazan, Kh. S. Bagdasarov, E. A. Fedorov, and L. M. Belyaev, Cryogenics **21**, 598 (1981).
11. G. R. Steward and A. L. Giorgi, J. Low Temp. Phys. **56**, 379 (1984).
12. N. E. Alekseevskiĭ, A. B. Mitin, V. I. Nizhanovskiĭ, V. I. Firsov, and E. P. Khlybov, Pis'ma Zh. Eksp. Teor. Fiz. **41**, 335 (1985) [JETP Lett. **41**, 410 (1985)].
13. N. E. Alekseevskiĭ and E. P. Krasnoperov, Dokl. Akad. Nauk SSSR **190**, 1325 (1970) [Sov. Phys. Dokl. **15**, 171 (1970)].
14. E. L. Kosarev and E. Pantos, Prib. Tekh. Eksp. **3**, 92 (1985).
15. G. C. Carter, L. H. Bennet, and D. J. Kahan, Prog. Mater. Sci. **20**, part I, 62 (1977).
16. S. K. Arif, D. St. P. Bunbury, G. J. Bowden, and R. K. Day, J. Phys. **F5**, 1048 (1975).
17. E. D. Jones, Phys. Rev. **180**, 455 (1969).
18. M. A. Ruderman and C. Kittel, Phys. Rev. **96**, 99 (1954).
19. J. D. Cohen and C. P. Slichter, Phys. Rev. **B22**, 45 (1980).
20. J. Friedel, Nuovo Cimento (Suppl.) **2**, 287 (1958).
21. P. W. Anderson, Phys. Rev. **124**, 41 (1961).
22. F. Borsa and G. Olcese, Phys. Status Solidi **A17**, 631 (1973).
23. A. W. Overhauser, and J. Appel, Phys. Rev. **B31**, 193 (1985).
24. D. I. Khomskiĭ, Ups. Fiz. Nauk **129**, 443 (1979) [Sov. Phys. Usp. **22**, 879 (1979)].
25. N. E. Alekseevskiĭ, V. N. Narozhniĭ, and E. P. Khlybov, Zh. Eksp. Teor. Fiz. **84**, 1538 (1983) [JETP Lett. **57**, 896 (1983)].

Translated by F. J. Crowne