Structural and dynamic properties of the magnetic order in the 90-K superconductor $HoBa_2Cu_3O_7$

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Zero-field μ^+ spin-rotation measurements in a ceramic sample of $HoBa_2Cu_3O_7$ reveal three-dimensional antiferromagnetic order below ≈ 5 K and some incoherent, perhaps two-dimensional, order up to 50 K in a diminishing fraction of the sample volume. With increasing temperature, at about 100 mK, the Ho moments change their orientation from parallel to the c axis to parallel to the a axis. The effective Ho moment is of the order of $2.6\mu_B$. Fluctuating field components perpendicular to the static field components at the μ^+ positions cause dynamical relaxation coexisting with static dipolar line broadening. Thermally activated slow "paramagnetic" fluctuations are observed up to 50 K.

I. INTRODUCTION

Shortly after the discovery of superconductivity in $YBa_2Cu_3O_7$ it was realized that the substitution of Y in the layered orthorhombic perovskite-type structure by a magnetic rare-earth (R) ion, such as Gd, Dy, Ho, and Er,[1-3] does not affect the superconducting properties of this system, not even at low temperature, where the $4f$ moments enter into a magnetically ordered state. This confirms the view that superconductivity must be closely confined to the CuO_2 planes, considering that these planes are the nearest-neighbor planes to the $Y(R)$ planes.

The Y or R planes themselves are separated from each other by approximately 12 Å, which leads one to suspect that the magnetic order among the R ions is mainly two dimensional (2D) in nature and that the exchange coupling between adjacent magnetic planes is rather weak. Therefore, it is very interesting to probe the R-ion-related magnetism in more detail because such studies could reveal possible interaction mechanisms which may be relevant with respect to the superconductivity in these oxide compounds.

Neutron-diffraction experiments revealed in detail the magnetic structure in $GdBa_2Cu_3O_7$,[4] $DyBa_2Cu_3O_7$,[5] and $ErBa_2Cu_3O_7$.[6] In the first two systems the structure was found to be of simple 3D antiferromagnetic order with the rare-earth moments aligned along the crystallographic c axis and a doubling of the unit cell in all three directions. In the third system a two-dimensional ordering was found with chains of ferromagnetically coupled spins aligned along the b axis and antiparallel orientation between adjacent chains. Some ordering below 140 mK was also detected in $HoBa_2Cu_3O_7$, but an unambiguous identification of the magnetic structure was not possible.[5] Assuming a 3D antiferromagnetic structure similar to the one in $DyBa_2Cu_3O_7$, but with a doubling of the unit cell only along the b and c directions, an effective moment of $\sim 2\mu_B$ was extracted.[5]

The slowing down of spin fluctuations in $HoBa_2Cu_3O_7$, starting at temperatures as high as 100 K, and an effective spin freezing below ~ 3 K were observed in a number of zero-field muon spin-rotation (μSR) experiments.[7-9] Since no precession was observed in the frozen state down to 2 K, the order appeared to be random or spin-glass-like, or of extremely short-range nature. Therefore, μSR and neutron-scattering results together seem to suggest that a change in the magnetic behavior must occur somewhere between ~ 2 K and ~ 140 mK. The present zero-field μSR study was intended to further investigate this phenomenon and to extend the temperature range distinctly down to lower temperatures.[10] The lowest temperature attained in this experiment was 39 mK.

Inelastic neutron scattering on $HoBa_2Cu_3O_7$ allowed to deduce the crystal-field splitting of the Ho^{3+} Hund's rule ground-state multiplet $^5I_8$.[11] The lowest state is a singlet state with zero magnetic moment. The actual appearance of a non-zero-ordered moment of the order of $2\mu_B$ per Ho site implies a strong admixture of excited states into the singlet ground state. Four excited singlet states are found within 50 K above the ground state.[11] Two of them have a nonzero matrix element $\langle J_z \rangle$ with the ground state. Previously, the low-temperature ordering was explained to occur out of a singlet ground state by a nuclear moment induced polarization effect.[12] Since, as demonstrated by our results discussed below, coherent antiferromagnetic order appears above 1 K, this is a rather unlikely explanation. We suggest that the observed ordering is due to overcritical exchange interactions. Such interactions destabilize the singlet ground state similar to what is observed in induced moment magnets such as Pr_3Tl, for example.[13] The crucial question is then, of course, of what origin these overcritical exchange interactions are.

The paper is organized as follows: In Sec. II we briefly provide some experimental details, we discuss our results

in Secs. III and IV, and Sec. V contains a summary of the present μSR study.

II. EXPERIMENTAL DETAILS

The zero-field μSR measurements were performed at the Paul Scherrer Institute (PSI, formerly SIN) using low-energy (surface) muons from the $\pi M3$ beam line. Two series of runs were performed with the PSI low-temperature μSR facility (LTF), which allowed to cover the temperature range from 39 mK to 2.8 K. A third series of runs, using a conventional μSR spectrometer, covered the range from 2.4 to 50 K.

The $HoBa_2Cu_3O_7$ ceramic sample used in this experiment is a roughly rectangular plate with dimensions of $13\times22\times2$ mm^3. It was oriented perpendicular to the μ^+ beam. The sample was prepared as outlined in Ref. 3. It was examined by neutron diffraction and found to be essentially single-phase material. The transition temperature T_c was determined by transverse-field μSR measurements to be $\sim$90 K. These measurements will be discussed elsewhere.

III. RESULTS

Figure 1 shows the time evolution of the μ^+ polarization observed at different temperatures. In particular, one notices a strongly damped oscillating signal at the lowest temperature. This oscillating component is still faintly visible in the 2.4-K spectrum. The oscillation can no longer be resolved at 5 K. However, at 7 K and even at 30 K two components continue to exist as is apparent from the change of slope in the data. The fastly damped component obviously decreases in amplitude as the temperature is increased. The solid lines in the figure are fits to the data of the function

$$P_\mu(t)=A_\parallel e^{-\lambda t}+A_\perp e^{-(1/2)\sigma^2 t^2}\cos\omega_\mu t\,. \tag{1}$$

$A_\parallel$ and $A_\perp$ are amplitudes reflecting the fractions of the muons contributing to the two components; ω_μ is the μ^+ Larmor frequency which is determined by the static average local field B_μ at the μ^+ sites

$$\omega_\mu=\gamma_\mu B_\mu \quad (\gamma_\mu/2\pi=13.55\ \text{kHz/G})\,. \tag{2}$$

Various χ^2 tests proved that for the oscillating component a Gaussian relaxation function fits the data best. This implies that the damping of the oscillating component is primarily caused by a quasistatic inhomogeneous field distribution across the μ^+ sites leading, in the language of NMR, to an inhomogeneous line broadening. The relaxation rate σ as defined in Eq. (1), is given by the square root of the second moment $\langle\Delta B_l^2\rangle$ of the distribution of B_μ,

$$\sigma=\gamma_\mu\sqrt{\langle\Delta B_l^2\rangle}\,. \tag{3}$$

The second nonoscillating component in Eq. (1), which had to be included in order to describe the data accurately, displays clearly an exponential damping.

In order to understand the origin of this component one

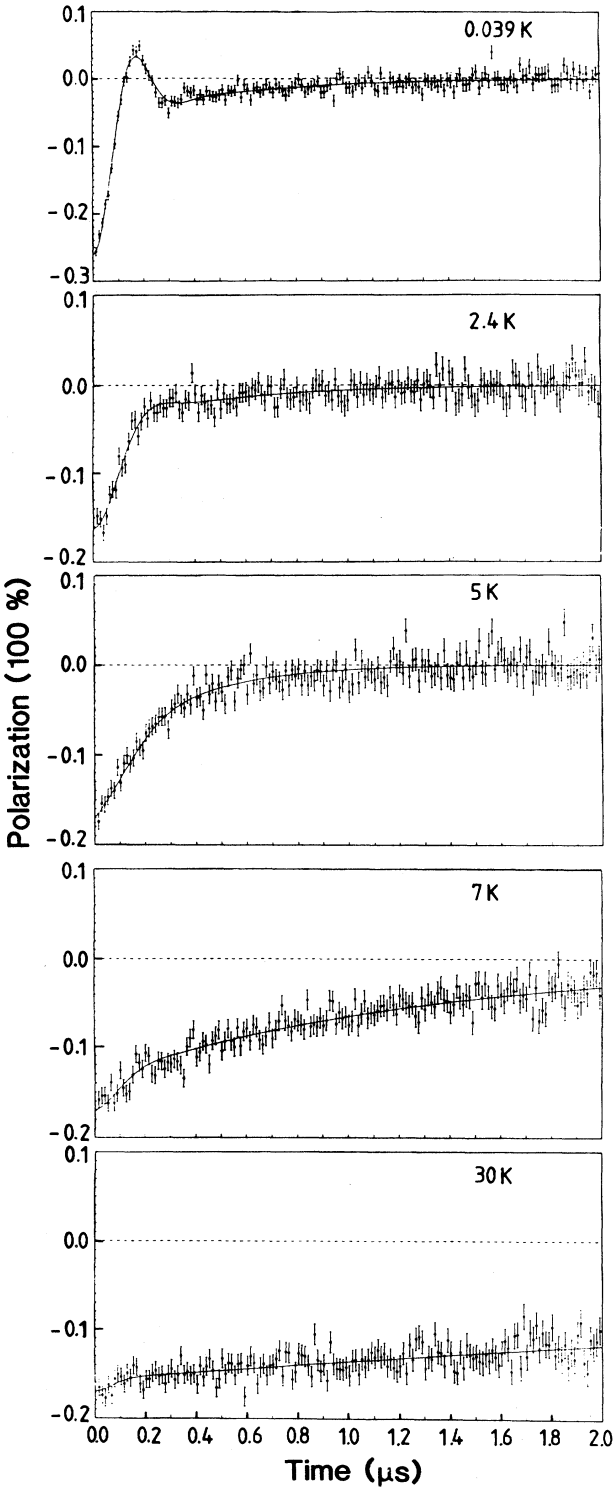

FIG. 1. Evolution of the μ^+ polarization $P_\mu(t)$ at various temperatures. The absolute values of $P_\mu(0)$ are not identical since the data were taken with different instruments and under different conditions. The solid lines are fits to Eq. (1). The original fit to the μSR signal made use of the expression $N_{e^+}=N_0\exp(-t/\tau_\mu)[1+P_\mu(t)]+P_{BG}$, where $\tau_\mu=2.2$ μs is the μ^+ lifetime and P_{BG} is a time-independent background (Ref. 14).

has to recall that for a completely random distribution of the directions of the internal field $\mathbf{B}_\mu$ one-third of all μ^+ will on the average experience an internal field along their initial spin direction. These muons will not precess and the muon-spin-rotation signal is reduced to $\frac{2}{3}$. Usually the nonprecessing $\frac{1}{3}\mu^+$ fraction will not be visible, unless it is time dependent, i.e., if it relaxes. Relaxation in this instance must be a dynamic or T_1 process caused by fluctuating field components perpendicular to the μ^+-spin, or to $\mathbf{B}_\mu$. The resulting relaxation function is exponential and the damping constant $\lambda = 1/T_1$ is proportional to the power of the fluctuation spectrum $F(\omega)$ at the μ^+ Zeeman frequency ω_μ.[15] If the fluctuations can be characterized by a correlation time τ_c, $1/T_1$ can be expressed as[15]

$$\lambda = \frac{1}{T_1} = \gamma_\mu^2 \langle B_l^2 \rangle \frac{\tau_c}{1+\omega_\mu^2 \tau_c^2} . \quad (4)$$

$\langle B_l^2 \rangle$ is the mean of the square of the transverse-field components.

Because the total asymmetry or amplitude $A_\parallel + A_\perp$ of the observed μSR signal accounts for all implanted μ^+ (no missing asymmetry) and because the investigated $HoBa_2Cu_3O_7$ sample is polycrystalline ($\mathbf{B}_\mu$ is randomly directed with respect to the initial μ^+ polarization) the nonoscillating, exponentially relaxing component must originate from those μ^+, whose spins are parallel to $\mathbf{B}_\mu$ and the observed damping reflects the presence of fluctuating field components.

However, there is a slight difficulty with this explanation. Figure 2 shows the fitted $A_\perp$ and $A_\parallel$ as a function of temperature. Considering only the low-temperature regime we find $A_\perp = 19\%$ and $A_\parallel = 6\%$ and $A_\parallel / A_\perp = 0.32$ instead of the expected $\frac{1}{3}/\frac{2}{3} = 0.5$. It implies that on the average less μ^+ are experiencing a field parallel to their spin than expected for a truly polycrystalline sample. It is not unreasonable, however, to assume that the sample preparation technique leads to a certain preference of crystal orientations inside the sample.

Figure 3 displays the fit results for $\nu_\mu = \omega_\mu / 2\pi$. Below 75 mK ν_μ amounts to about 2.5 MHz, corresponding to

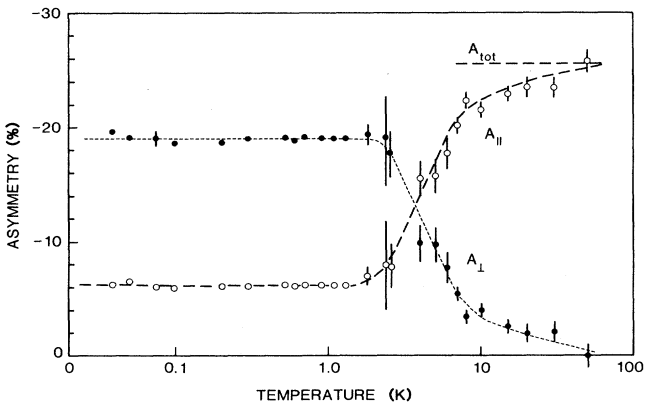

FIG. 2. Temperature dependence of the fitted amplitudes $A_\parallel$ and $A_\perp$. Data from different instruments were renormalized so that $A_{tot} = A_\parallel + A_\perp = 25.6\%$. The drawn lines are only intended to guide the eye.

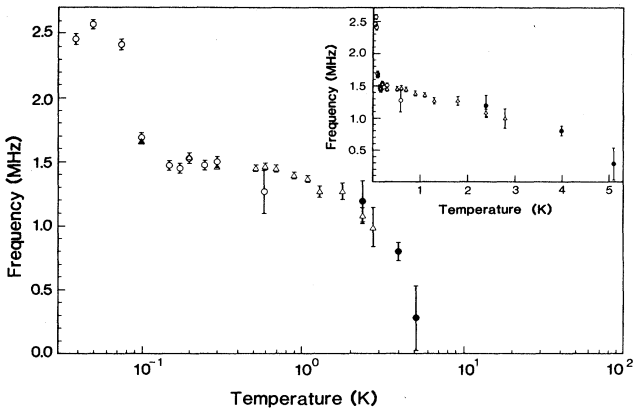

FIG. 3. Temperature dependence of the μ^+ Larmor frequency $\omega_\mu/2\pi$. The temperature scale is logarithmic and linear in the inset. Different symbols refer to different series of measurements.

$B_\mu = 185$ G. Between 75 and 150 mK ν_μ drops relatively sharply to 1.5 MHz ($B_\mu = 111$ G). Upon increasing the temperature further, ν_μ remains constant up to about 600 mK and then starts to drop smoothly until it becomes zero somewhere between 5 and 6 K (Fig. 3). Turning back to Fig. 2 one notices that the amplitude of the oscillating signal starts to decrease above ~ 1.5 K, while the component of the exponentially damped signal rises by the same proportion. Although ω_μ approaches zero somewhere around 5–6 K, it does not mean that this component disappears altogether. Rather this component continues to exist as a Gaussian-damped, nonoscillating signal until its amplitude vanishes around 50 K. The exponentially damped component becomes the dominating one above 4 K and seems to be the only one describing the data at 50 K. Consequently, one-component Gaussian fits to the 50-K data showed visible disagreement.

The fitted relaxation rates σ and λ are displayed in Figs. 4 and 5. σ shows essentially no temperature dependence up to 15 K and amounts to $\sim 7\ \mu s^{-1}$ or $(\langle \Delta B_l^2 \rangle)^{1/2} = 82$ G. Above 15 K the fitted σ value increases and its error starts to increase drastically due to the decreasing $A_\perp$. In contrast λ shows a pronounced

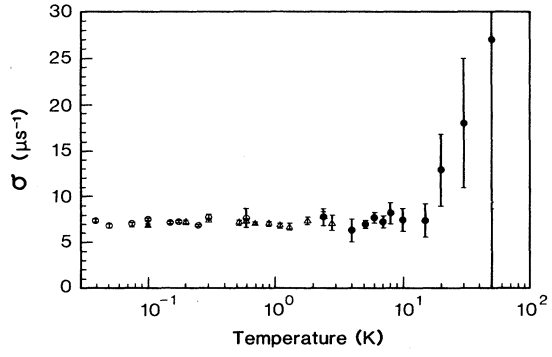

FIG. 4. Temperature dependence of the Gaussian relaxation rate σ.

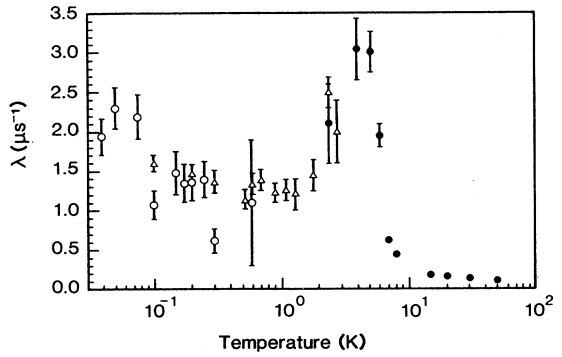

FIG. 5. Temperature dependence of the exponential relaxation rate λ.

temperature dependence over the full temperature range exhibiting in particular a clear peak at 4.5 K. The change in frequency around 100 mK is also accompanied by a change in λ, while, surprisingly, σ does not vary.

IV. DISCUSSION

The average field B_μ by which the muon precesses depends on the magnitude and order of the magnetic moments as well as on the muon site in the $HoBa_2Cu_3O_7$ lattice. The available information which can be used in assigning a site to the muon are the μ^+ relaxation rate above $T_c = 90$ K and the local field seen by the μ^+ in the magnetically ordered state of $GdBa_2Cu_3O_7$.[16,17] Furthermore a number of possible sites have been suggested by calculations of potential energy surfaces for μ^+ in $YBa_2Cu_3O_7$.[18]

In $YBa_2Cu_3O_7$ the μ^+ relaxation rate above T_c, $\sigma(T > T_c)$, is entirely determined by the Cu nuclear dipole fields at the μ^+ positions. Even in the polycrystalline average this relaxation is quite site dependent. In $YBa_2Cu_3O_7$, $\sigma(T > T_c)$ is known from previous μSR studies[19,20] to amount to 0.127(3) μs^{-1} and to be field independent between 100 G and 7.47 kG. We have calculated $\sigma(T > T_c)$ for a number of lattice positions for three different situations: (i) in the Van Vleck limit, in which the Zeeman interaction of the Cu nuclear spins dominates over possible quadrupole interactions (σ_{vv}); (ii) in the quadrupole interaction limit with the electric-field gradients (EFG's) at the Cu nuclei oriented along the μ^+-Cu-nucleus distance vectors (σ_Q^{rad}); (iii) again in the quadrupole interaction limit but with the EFG's at the Cu nuclei oriented along the crystallographic c axis (σ_Q^{ax}).[14] The latter configuration is the one expected for the Cu(2) site on the basis of nuclear quadrupole resonance (NQR) results (see e.g., Ref. 21). The NQR results also suggest that within the range of magnetic fields applied in the μSR experiments the quadrupole interaction is dominant.

For the same lattice positions we have also calculated the dipolar fields B_{dip}^R originating from the Gd moments ($\mu_{Gd} \simeq 7.5\mu_B$) using the well-determined antiferromagnetic structure of $GdBa_2Cu_3O_7$.[4] μSR measurements in $GdBa_2Cu_3O_7$ yield a local average field of ~340 G.[16,17] The results of all these calculations are compiled in Table I. Sites 6, 7, and 8 are interstitial positions of high symmetry. None of these are compatible with the μSR data. Site 4 is an oxygen vacancy in the Cu-O chains and site 5 corresponds to the midposition between two oxygen atoms in the Ba plane. Sites 12 and 13 were suggested in Ref. 17. They all can be ruled out. Sites 1, 2, 3, and 9 are the

TABLE I. Compilation of calculated relaxation rates σ and net dipole fields B_{dip} at various possible μ^+ sites (for details see Sec. IV). Sites (1), (2), (3), and (9) were suggested in Ref. 18 and sites (12) and (13) in Ref. 17; site (4) is an oxygen vacancy. Lattice constant used (Ref. 5) $a = 3.814$ Å, $b = 3.882$ Å, and $c = 11.63$ Å. The known experimental values for σ in $YBa_2Cu_3O_7$ above T_c and for the local fields in $GdBa_2Cu_3O_7$ (R moments) and in the magnetically ordered tetragonal $YBa_2Cu_3O_6$ (Cu moments) are also listed.

				(MHz)			(G)	
μ^+ site	x/a	y/b	z/c	σ_{vv}	σ_Q^{rad}	σ_Q^{ax}	B_{dip}^R [a]	B_{dip}^{Cu} [b]
(1)	0.026	0.400	0.083	0.119	0.168	0.121	450	
(2)	0.0	0.265	0.108	0.159	0.173	0.226	393∥a	
(3)	0.245	0.395	0.0	0.125	0.178	0.141	190	
(4)	0	0.5	0	0.131	0.185	0.147	−257∥a	
(5)	0	0.5	0.159	0.0635	0.0899	0.0662	1010	297
(6)	0.5	0.0	0.0	0.138	0.195	0.154	−253∥b	
(7)	0.0	0.0	0.5	0.198	0.281	0.279	0	
(8)	0.5	0.5	0	0.0687	0.0972	0.0769	0	
(9)	0.015	0.555	0.087	0.100	0.142	0.103	413∥a	
(10)	0.015	0.555	0.065	0.117	0.165	0.122	332∥a	
(11)	0.171	0.5	0.065	0.0669	0.134	0.100	346∥a	100
(12)	0.0	0.5	0.171	0.0622	0.0881	0.0650	−1176∥a	
(13)	0.5	0	0.171	0.0636	0.0898	0.0666	−1141∥b	
	Experiment				0.127 [c]		340 [d]	~300 [e]

[a] $\mu_{Gd} = 7.5\mu_B$, AF arrangement according to Ref. 4.
[b] $\mu_{Cu} = 0.5\mu_B$, AF arrangement according to Ref. 23, $\mu_{Ho} \| \langle 110 \rangle$.
[c] References 19 and 20.
[d] References 16 and 17.
[e] References 22 and 23.

suggested μ^+ positions in Ref. 18. According to Ref. 18 site 1 is associated with the deepest potential well followed closely by the one at site 9. All of these positions do not lead to predictions which are consistent with the data. In calculating $B^R_{\rm dip}$ for other positions around site 9 it was noticed that a very good agreement with B_μ could be achieved by lowering the z coordinate to 0.065 (site 10). Keeping this new position and changing the y and z coordinates had little effect on $B^R_{\rm dip}$. Fixing the y coordinate at 0.5 and keeping the z component at 0.065 the x coordinate was determined by requiring the distance between μ^+ and the nearest-neighbor atom [this oxygen atom is at the position (0,0.5,0)] to be 1 Å. This is a typical length for the O−H bond.[18] The thus obtained site 11 leads to a $B^R_{\rm dip}$ in almost complete agreement with B_μ. Also the calculated $\sigma^{\rm rad}_Q$ is very close to the experimental value. The fact that $\sigma^{\rm rad}_Q$ and not $\sigma^{\rm ax}_Q$ is what the muon measures is consistent with the observation that the EFG at the nearest-neighbor Cu atom, which belongs to the Cu-O chains, is highly anisotropic[21] and therefore perhaps better represented by the situation (ii).

We have also tried to explain the field at the μ^+ site in the magnetically ordered phase of tetragonal $YBa_2Cu_3O_6$ which is not superconducting. This field amounts to roughly 300 G.[22,23] Since in this compound oxygen has disappeared from the Cu-O chains the nearest-neighbor oxygen atom for site 11 is missing. Therefore, it is very likely that the μ^+ will assume a new position. Placing the μ^+ at the nearby (0,0.5,0.159) position (site 5) and assuming an antiferromagnetic arrangement of the Cu $3d$ moments ($=0.5\mu_B$) in the CuO_2 planes as found in Ref. 24 we calculate a dipolar field of $B^{\rm Cu}_{\rm dip}=297$ G which is in excellent agreement with the μSR result. In keeping with the concept of O−H bond formation we can move the μ^+ closer to one of the two nearest-neighbor oxygen atoms which, as long as the z position is not changed, has little effect on the calculated $B^{\rm Cu}_{\rm dip}$.

In effect it seems that the position (0.171,0.5,0.065) (see Fig. 6) is a reasonable choice for the μ^+ location in the fully oxygenated compound. Having thus obtained at least a rough identification of the μ^+ location one can attempt to interpret the B_μ found in the $HoBa_2Cu_3O_7$ crystal, assuming that the μ^+ site does not depend on the type of Y substitute.

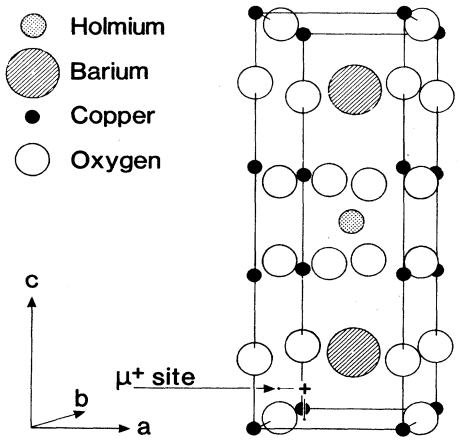

FIG. 6. Structure of the unit cell of $HoBa_2Cu_3O_7$ with the proposed μ^+ site indicated (site 11 of Table I).

Assuming the antiferromagnetic structure suggested in Ref. 5 and mentioned in the Introduction, we obtain a dipolar field of -72 G/μ_B. Comparing this number with $B_\mu=185$ G for $T<100$ mK we calculate an effective Ho moment of $2.56\mu_B$. This is in good agreement with the estimate presented in Ref. 5. The fact that a frequency is seen at all in the μSR data is also proof for a 3D antiferromagnetic arrangement. Since the μ^+ location is almost halfway between the Ho layers a 2D arrangement of magnetic order would probably have prevented the appearance of an oscillating component.

The drop of B_μ around 100 mK may have different origins. One possibility is a rotation of the Ho moment into the (a,b) plane. This is suggested by the crystal-field splitting of the Ho ground-state multiplet which predicts an easy axis of magnetization along the a direction.[11] Assuming the Ho moments to be aligned along the a axis and a magnetic unit cell which is doubled in all three direction we calculate a dipolar field at site 11 of $B^{\rm Ho}_{\rm dip}=42$ G/μ_B. By comparison with the measured field of $B_\mu=111$ G above 100 mK we find an effective moment of $2.65\mu_B$ which is essentially unchanged from the one for $T<100$ mK. This nice consistency renders this explanation as the most likely one. In view of the crystal-field level scheme[11] it is the ordering of the Ho moments along the c axis below 100 mK which is in fact unexpected. A reason for the assumed alignment might be found in competing influences of isotropic and anisotropic two-ion exchange interactions.

Another possibility is an unchanged structure but a drop in the value of the ordered moment. However, there is no obvious reason why something like that should occur. In fact, the temperature independence of σ is strong evidence against such a mechanism since in this instance one would expect a scaling of σ with $\mu_{\rm Ho}$.

A third very interesting, although remote possibility would be an onset of magnetic order among the Cu $3d$ moments (e.g., as found in Ref. 24) below ~100 mK while the Ho moments would retain the order already achieved above this temperature. The resulting field at the μ^+ would be of the order of 200 G, in reasonable agreement with the measured 185 G.

In any case the ordering must be rather short ranged, since no magnetic diffraction peaks were found in the neutron work above 140 mK.[5]

The temperature independence of σ up to ~15 K is a puzzling phenomenon. It implies that the associated quasistatic field spread ΔB_l [see Eq. (3)] does not change with rising temperature despite the fact that the average field as reflected in the behavior of ω_μ eventually becomes zero and that the fraction of μ^+ (or the associated volume fraction) contributing to the Gaussian-damped signal shows a steady decline above ~1.5 K (see Fig. 2). The actual origin of the field spread ΔB_l is not clear. The magnitude of the field spread is of similar order as B_μ and must, therefore, be associated with the magnetic moments in the vicinity of the muon. This can explain its indepen-

dence from the actual extension of the correlated regions. Perhaps there is a more complex magnetic structure than the simple antiferromagnetic one discussed above leading to magnetically inequivalent μ^+ sites.

The temperature dependence of ω_μ above 1 K most likely reflects to what extent the magnetic order in the respective volume is losing its 3D antiferromagnetic structure, perhaps by adopting a more 2D character above 5 K. The increase of σ above 15 K may even point to an arrangement of randomly frozen Ho moments, as in a spin glass, which will give rise to a field spread of the order of $\Delta B \simeq 340$ G or $\sigma \simeq 29\ \mu s^{-1}$ as estimated below.

The data show that magnetic order does not evolve by means of a cooperative phase transitions. Rather, this evolution starts on a microscopic scale around 50 K and is essentially complete at around 1.5 K, involving the whole sample volume (see Fig. 2).

Another puzzling fact is that σ does not reveal any influence from fluctuating field components along B_μ, while the exponentially damped component in the μSR signal clearly implies the presence of fluctuating field components perpendicular to B_μ. As we will discuss further below, fluctuation rates must be of the order of $10^8\ s^{-1}$ which, if present also along B_μ, should have left their mark on σ as well. So we conclude that there are no fluctuating field components along B_μ. This may be understood, if one assumes that the Ho moments are actually precessing in an uncorrelated fashion around the axis of alignment, thus producing a static component μ_l along this direction and a time-dependent component $\mu_t(t)$ perpendicular to it. This model could also provide a natural explanation for the static field spread ΔB_l, if the static component μ_l is quantized according to

$$\mu_l = m_J g_J \mu_B$$

and, therefore, displays a distribution in values restricted by the requirement that the overall antiferromagnetic arrangement is conserved. Assuming that $\langle \mu_l^2 \rangle = \langle \mu_z^2 \rangle = \frac{1}{3}\langle \mu_{\mathrm{Ho}}^2 \rangle$, the total moment μ_{Ho} is estimated to be $3\mu_B \langle \mu_l \rangle \simeq 4.5 \mu_B$. The moment arrangement resulting from this model may be viewed as a novel type of spin glass.

Finally we turn to the discussion of the exponentially damped component in the μSR signal. The relaxation rate data displayed in Fig. 5 are characterized by three different temperature ranges. In the range that extends from the lowest temperature up to ~ 1.5 K λ scales with ω_μ, between ~ 1.5 K and ~ 5 K λ increases sharply, and above 5 K it initially decreases drastically, then more slowly up to the highest temperature measured.

Since ω_μ is rather small, we expect $\omega_\mu \tau_c \ll 1$ and therefore

$$\lambda = \frac{1}{T_1} = \gamma_\mu^2 \langle B_l^2 \rangle \tau_c \,. \tag{5}$$

In the first range, assuming that $B_l \approx B_\mu$, we estimate the correlation time τ_c to be of the order of 10^{-8} s, independent of temperature. The temperature independence is consistent with our assertion that the fluctuating perpendicular field components arise from the uncorrelated precession of the Ho moments around the c axis below ~ 100 mK and around the a axis above ~ 100 mK. The rate

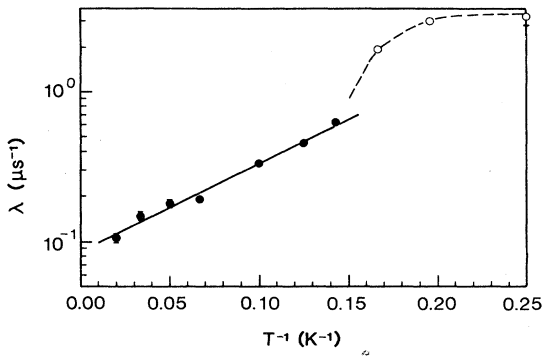

FIG. 7. Logarithm of λ vs inverse temperature (Arrhenius plot) for temperatures above 4 K.

$1/\tau_c$ is a measure of the loss of phase coherence among neighboring Ho moments and, therefore, also a measure of the average precession frequency of the Ho moments.

Above ~ 1.5 K $A_\parallel$ starts to rise which, as discussed before, signals the formation of paramagnetic regions within the sample. Fluctuating magnetic fields arise now from correlated or uncorrelated fluctuations of the Ho moments which, if slow enough, constitute an efficient source for μ^+ depolarization. The rise in λ is probably the result of such a mechanism whereby one has to be aware that the exponentially damped component in reality now consists of two contributions: (i) a contribution from the magnetically correlated regions which decrease in volume as the temperature is increased, and (ii) a contribution from the paramagnetic regions which increase in volume as the temperature increases. The fitted λ is, therefore, to be considered as an average effective damping rate only.

The paramagnetic phase dominates in the third temperature range and the fitted λ will be mainly determined by the induced relaxation rate in this phase. The temperature dependence of λ above 6 K is in fact well described by the expression

$$\lambda = \lambda_0 \exp\left(\frac{U}{kT}\right) \tag{6}$$

with $\lambda_0 \simeq 0.085$ MHz and $U/k \simeq 15$ K (see Fig. 7). λ_0 can be converted to a correlation time τ_0 with the help of Eq. (5) by noting that $\langle B_l^2 \rangle$ has now to be replaced by the second moment of the field distribution arising from a random orientation of the Ho moment. Using for this moment the estimated value of $4.5\mu_B$ we calculate for site 11 $\gamma_\mu^2 \langle \Delta B^2 \rangle \simeq 828\ \mu s^{-2}$ or $\Delta B \simeq 340$ G in the Van Vleck limit. τ_0 is then estimated to be of the order of 10^{-10} s.

It thus appears that the Ho moment fluctuations are thermally activated with an activation energy of about 15 K. The fluctuation rates are relatively small even in the infinite temperature limit. Below 7 K some enhanced slowing down is indicated. However, a cooperative phase transition is clearly not manifested in these data.

V. SUMMARY

From μSR spectroscopy the evolution of magnetic correlations and magnetic order among the Ho $4f$ mo-

ments in the high-T_c superconductor $HoBa_2Cu_3O_7$ is found to be rather unusual. The main conclusions can be summarized as follows.

(1) Magnetic order of incoherent (probably 2D) and coherent 3D structure evolves below ~ 50 and 5 K, respectively, without any discernible cooperative phase transition as far as the μSR data can tell. The order develops first in a vanishingly small fraction of the total sample volume which grows with decreasing temperature and reaches 100% at ~ 1.5 K. Recently, a very similar behavior has been observed in the heavy electron compound $CeAl_3$.[25]

(2) The μSR data below 100 mK are consistent with an antiferromagnetic structure of the Ho moments in which the moments are aligned along the c axis and the magnetic unit cell is doubled along the b and c directions in comparison with the crystallographic unit cell. The ordered Ho moment is determined to be $\sim 2.6\mu_B$. The structure and the moment value are both in agreement with neutron diffraction data.[5] At about 100 mK the Ho moments realign antiferromagnetically along the a axis with a doubling of the unit cell in all three directions. Since the a axis is expected to be the easy axis of magnetization on ground of the crystal-field splitting of the ground-state multiplet[11] this change of the moment arrangement may be caused by anisotropic exchange interactions, as mentioned in Sec. IV.

(3) Dynamic spin relaxation of those μ^+ whose spin is oriented parallel to the internal field B_μ implies the presence of fluctuating field components perpendicular to B_μ while no fluctuating component along B_μ was observed. This can be understood, if the Ho moments perform an uncorrelated precession around the axis of alignment, leading to the appearance of perpendicular time-dependent components of the Ho moments. The precession rates are the order of 10^8 s^{-1}. Above 1.5 K a paramagnetic phase starts to develop, displaying relatively slow moment fluctuations of a thermally activated nature above 6 K with an activation eneryg of the order of 15 K.

(4) The internal field B_μ shows a temperature-independent distribution width of $\Delta B_l \simeq 82$ G up to 15 K. This value may be compared with the width arising in case of a random distribution of the Ho moments which amounts to ~ 340 G. This width seems to be approached at the higher temperatures. ΔB_l appears to be also independent of the change in magnetic structure at around 100 mK. This feature is not understood at present. We conjecture that ΔB_l is a result of the quantized nature of the static moment component μ_l along the axis of alignment or the axis of quantization. Possible values of μ_l are restricted by the requirement that the overall arrangement remains antiferromagnetic. The average value of μ_l determines the average B_μ and is given by the value quoted above ($\simeq 2.6\mu_B$).

The onset of magnetic order in a singlet ground-state system is unusual by itself and requires, as is known from previous work in metallic rare-earth systems,[13] an over-critical exchange interaction. *This exchange interaction is facilitated by conduction electrons in the normal metallic state but is clearly less easy to account for in a superconducting state well below T_c.* We thus regard the occurrence of three-dimensional magnetic order in the singlet ground-state system $HoBa_2Cu_3O_7$ below 2 K as an unsolved puzzle.

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