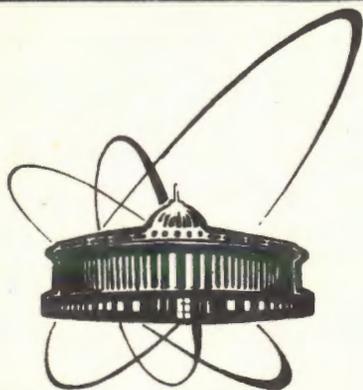


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V.G.Grebinnik, V.N.Duginov, V.A.Zhukov,
S.Kapusta, A.B.Lazarev, V.G.Olshevsky,
V.Yu.Pomjakushin, S.N.Shilov, I.I.Gurevich ¹,
B.F.Kirillov ¹, E.P.Krasnoperov ¹, B.A.Nikolsky ¹,
A.V.Pirogov ¹, A.N.Ponomarev ¹, V.A.Suetin ¹,
A.I.Morozov ²

THE μ SR STUDY OF THE RELAXATION OF Ho
AND Er MAGNETIC MOMENTS IN HIGH- T_c
1-2-3 COMPOUNDS

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¹I.V.Kurchatov Institute of Atomic Energy, Moscow,
USSR

²Moscow Institute of Radioengineering, Electronics
and Automation, Moscow, USSR

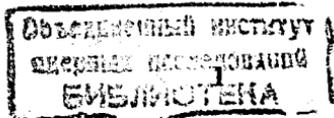
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History

Zero field (ZF) μ SR-measurements of the temperature dependence of the muon spin relaxation rate is a widely known method for studies of dynamic effects in magnetics [1]. In the present paper we discuss the results of our ZF μ SR-experiments with polycrystalline metal oxide superconductors of 1-2-3 type with rare earth (RE) elements holmium and erbium substituting for yttrium. It is a well established fact that in 1-2-3 compounds RE substitutes such as Er, Ho, Gd and Dy possess effective magnetic moments produced by the state of ions with valence 3^+ . These ions do not suppress superconductivity because the interaction of magnetic moments with conduction electrons is extremely weak. The temperature of magnetic ordering of RE ions such as Ho, Gd and Er doesn't exceed 2.5K. On the other hand, ZF μ SR-experiments revealed a considerable increase of the muon spin relaxation rate in $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{Ho}_{0.5}\text{Y}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ at temperatures 10 - 20K [2-5] well above the Neel temperature. The increase in the relaxation rate may be explained by assumption of the slowing down of local magnetic field fluctuations. This fact can hardly be considered as the consequence of the critical fluctuations because the temperatures corresponding to the noticeable increase in the muon relaxation rate exceed the Neel temperature by a factor of 10-50. Another experimental fact needed an explanation: in the Er substituted sample the relaxation rate does not show any change in the same temperature region in contrast to Ho substituted compounds, though the magnetic moments of Ho and Er ions with valence 3^+ are of the same order of magnitude. We will try to give a reasonable explanation to this difference taking into account the crystalline field effects.

Experiment

Our μ SR-experiments were carried out on the muon channel of the LNP JINR phasotron (Dubna). Three polycrystalline 1-2-3 samples were investigated: $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$,



$\text{Ho}_{0.5}\text{Y}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{ErBa}_2\text{Cu}_3\text{O}_{7-\delta}$. The samples with Ho were prepared by the cryochemical procedure [6] and the sample with Er by the selfpropagating synthesis procedure [7]. The measurements were performed in the temperature interval 4.2K - 270K, the samples were cooled in a continuous He-flow cryostat. By the fitting procedures the experimental time dependences of the muon polarisation $P(t)$ were described by the power-exponential function:

$$P(t) = \exp[-(\sigma t)^\alpha] \quad (1)$$

Figure 1 shows the experimental dependences: $\sigma(T)$ and $\alpha(T)$ for the samples with the substitution for yttrium by holmium. Figure 2 shows $\sigma(T)$ dependence for $\text{ErBa}_2\text{Cu}_3\text{O}_{7-\delta}$ obtained by fitting data to formula (1) with $\alpha=1$. Formula (1) is a satisfactory approximation when the

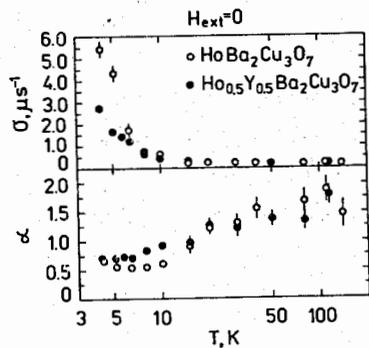


Fig.1. The relaxation rate σ and the parameter α in the Ho substituted compounds for $P(t) = \exp[-(\sigma t)^\alpha]$.

values of the frequencies of the local field fluctuations on the muon $\nu \geq \langle B_\mu^2 \rangle^{1/2} / \gamma_\mu$ where $\langle B_\mu^2 \rangle^{1/2}$ is the r.m.s. value of the magnetic field distribution at the interstitial sites of the muon localization and γ_μ is the gyromagnetic ratio for

muons. This power-exponential law was proved in [2] by numerical calculations of $P(t)$. The decrease of α in the region where $\alpha < 1$ is a consequence of the decrease of the fluctuation frequencies of local fields. For the case of the static local fields the Kubo - Toyabe function [8] is often used which corresponds to the Gaussian type of the field distribution with $\langle B_\mu \rangle = 0$. In the region where the fluctuation frequencies are very high and the visible effect of fluctuations on μSR spectra is averaged to zero, the $P(t)$ dependence reflects the static distribution of local fields produced by nuclear dipole moments. In this case we can see only the relaxing part of the Kubo-Toyabe dependence because of low r.m.s. value and the limited time interval of the observation $5\tau_\mu$, where τ_μ is the muon life time ($\approx 2.2\mu\text{s}$). So the time-dependent part of $P(t)$ may be described approximately by the Gaussian law (α values are close to 2) as well. The decrease of α down to 1 may be considered as an indication of the increasing influence of the fluctuating local field on the $P(t)$ dependence due to slowing down of the fluctuations.

The muon spin relaxation rate values in $\text{Y}_{0.5}\text{Ho}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ show the behaviour similar to that in the pure $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ sample. In contrast to Ho compounds in $\text{ErBa}_2\text{Cu}_3\text{O}_{7-\delta}$ we have not seen any change in the relaxation rate down to the temperature 4.2K.

Discussion

As was mentioned, from the phenomenological point of view Ho and Er substituted 1-2-3 compounds are similar. The results of the measurements of the magnetic susceptibility in the 1-2-3 samples with Ho and Er [9] proved that the magnetic moments of Ho and Er ions don't noticeably differ. To explain the difference between the temperature dependences of the relaxation rate in Ho and Er substituted samples the crystalline electric field effects can be taken into account.

In contrast to $\text{ErBa}_2\text{Cu}_3\text{O}_{7-\delta}$ where the ground state of

Er ions in the crystalline field is the doublet, $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ is the singlet antiferromagnetic compound.

The transition from the singlet ground state of the Ho ion to the first excited state requires the energy $\Delta=0.55\text{meV}$ [10]. The long range magnetic order in these systems takes place only if the exchange (or dipole) interaction between the magnetic moments exceeds the critical value.

According to paper [11], in the framework of the two-level model of singlet magnetism the following inequality is to be valid for observation of the long-range order:

$$J(Q_0) \geq \Delta/2, \quad (2)$$

where Q_0 is the wave vector corresponding to the maximum Fourier-component of the exchange interaction $J(r)$. (Below we shall omit the argument Q_0 , so $J(Q_0) \equiv J$).

In the case of validity of inequality (2) the magnetic phase transition temperature T_N can be determined from the equation

$$\chi_{V-V}^{J=1}, \quad (3)$$

where χ_{V-V} is the Van-Vleck magnetic susceptibility of the ion. After the substitution of χ_{V-V} in Eq.(3) one can obtain [11]

$$\frac{2J}{\Delta} \text{th}(\Delta/2T_N) = 1 \quad (4)$$

In the limit $1 \gg \delta = 2J/\Delta - 1 > 0$ the value of T_N is

$$T_N \approx \Delta / (|\ln(\delta/2)|) \quad (5)$$

If we take for granted that $T_N = 140\text{mK}$, as it follows from the neutron diffraction experiment [12], then we must admit that in this model δ is unreally small.

Actually, even if $\delta < 0$, the phase transition to the singlet antiferromagnetic state occurs owing to the hyperfine interaction with the nuclear spins or to the

exchange interaction with the paramagnetic impurities having a nonsinglet ground state in the crystalline field [13].

In this case the long-range order is caused by the additional contribution of these subsystems to the inverse magnetic susceptibility, which is inversly proportional to the temperature T .

The expression for T_N with allowance for hyperfine interaction has been obtained in paper [14]. But if the strength of this interaction isn't extremely large, it cannot induce magnetic moments required for the creation of the observed values of σ .

The elementary excitations of the singlet antiferromagnetic display the characteristic features of the soft mode, so we can use the phenomenological theory interpreting the influence of the orientable defects on the structural phase transition (see for example [14]) for the description of the effect of paramagnetic impurities. According to this theory, the presence of the orientable defects produces the following correction to the inverse susceptibility

$$\chi^{-1} - \chi_0^{-1} = -\gamma x J'^2 / T \quad (6)$$

here γ is the constant of the order of unit, x is the concentration of impurities, J' is the value of the exchange interaction between the impurity spin and the spin of the nearest ion of the matrix, and

$$\chi_0^{-1} = \frac{\Delta}{2} \text{th}^{-1}(\Delta/2T) - J. \quad (7)$$

From Eq.(6) with allowance for Eq.(7) for the case $\delta < 0$, $|\delta| \ll 1$ one has with the accuracy to $\exp(-\Delta/T_N)$

$$T_N = \gamma' x J'^2 / (\Delta - 2J) \quad (8)$$

γ' is the constant of the order of the unit.

So, the small (with respect to Δ) value of T_N in the compound $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ can be interpreted as the phase transition induced by the low concentration of paramagnetic impurities in the compound with $\delta < 0$.

In the paramagnetic phase this impurity polarises the

matrix in its vicinity. The size of this polarised region is of the order of the correlation radius of the magnetic moments in the pure compound r_{CO} . The value of r_{CO} in the singlet antiferromagnetic with $|\delta| < 1$ considerably exceeds the interatomic distance d .

For $T \leq \Delta$ the order of magnitude r_{CO} is

$$r_{CO} = d/|\delta|^{1/2} \quad (9)$$

and in the case $T \gg \Delta$

$$r_{CO} = d/(T/J-1)^{1/2}. \quad (10)$$

Hence with the temperature decreasing to $T \sim \Delta$ the size of the polarised region near the paramagnetic impurity strongly increases.

The appearance of this region is analogous to the polaronic effect and leads to the same consequences. The probability of coherent transition to the state with the opposite value of the effective spin projection sharply diminishes. Noncoherent transition (with destruction of the spin-polaron state) obeys the activation law. As a result, the reorientation time of the impurity spin and σ increase when the temperature decreases.

The estimation of the authors of paper[4] shows that one can get the observable values of σ if the characteristic time of changing of the magnetic field from the magnetic moments of ions acting on the muon is $\tau_0 \approx 10^{-8}$ s. This fact confirms the presence of the narrow central peak in the order parameter fluctuation spectrum, which is induced by the coupling between the order parameter and the slowly relaxed variable [14]. In our case the spin of the paramagnetic impurities plays the role of this variable.

If the distance between the muon site and the paramagnetic impurity greatly exceeds r_{CO} , the muon spin depolarisation is due to the nuclear magnetic moments, and

its rate isn't very large. This is the reason for the slow relaxation component of the muon spin.

In the compound $ErBa_2Cu_3O_{7-\delta}$ the spin polaron states are not formed and the value of τ_0 has the order of magnitude

$$\tau_0 = h/|T-T_N| \approx 10^{-11} s/|\beta|, \quad (11)$$

where $\beta = (T-T_N)/T_N$. Far from T_N ($\beta \gg 1$) the small value of τ_0 leads to the small contribution of electron spins to the muon spin relaxation rate.

In the mixed compound $Ho_{0.5}Y_{0.5}Ba_2Cu_3O_{7-\delta}$ the spin polaron state has smaller density because the susceptibility of Y ions is very small. The value of $|\delta|$ increases as the average distance between Ho ions increases and J decreases. Then the characteristic size of the polarized region r_{CO} in $Ho_{0.5}Y_{0.5}Ba_2Cu_3O_{7-\delta}$ will be smaller than in $HoBa_2Cu_3O_{7-\delta}$. The diminishing of the polaron effect leads to the decrease of τ_0 and σ values.

If our explanation is valid, the fast increase of σ with decreasing temperature is to be observed in other 1-2-3 compounds with singlet magnetic ions: $TbBa_2Cu_3O_{7-\delta}$, $TmBa_2Cu_3O_{7-\delta}$, etc.

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References

- [1] A.Schenck. Muon spin rotation spectroscopy (AdamHilger Ltd., Bristol and Boston, 1985)
- [2] N.Nishida, H.Miyatake, S.Okuma, Y.Kuno, Y.Watanabe, T.Yamazaki, S.Hikami, M.Ishikawa, T.Takabatake, Y.Nakazawa, S.R.Kreitzmann, J.H.Brewer and C-Y.Huang Jpn.J.Appl.Phys. 27 (1988), L94
- [3] V.N.Duginov, I.E.Graboi, V.G.Grebinnik, I.I.Gurevich, A.R.Kaul, B.F.Kirillov, E.P.Krasnoperov, A.B.Lazarev, B.A.Nikolsky, V.G.Olshevsky, A.V.Pirogov, V.Yu.Pomjakushin, A.N.Ponomarev, S.N.Shilov, V.A.Suetin, V.A.Zhukov JINR Rapid Commun. 4(30) (1988), 63
- [4] P.Birrer, F.N.Gygax, B.Hitti, E.Lippelt, A.Schenck, M.Weber Phys.Rev.B 39 (1989), 11449

- [5] V.G.Grebinnik, V.N.Duginov, V.A.Zhukov, S.Kapusta, A.B.Lazarev, V.G.Olshevsky, V.Yu.Pomjakushin, S.N.Shilov, D.T.Bezhitadze, I.I.Gurevich, B.F.Kirillov, E.P.Krasnoperov, B.A.Nikolsky, A.V.Pirogov, A.N.Ponomarev, V.A.Suetin, G.F.Tavadze Hyperfine Interactions (1990) in press. Proceedings of the 18 International Conference on Hyperfine Interactions (Prague, 14-19 August, 1989)
- [6] A.R.Kaul, I.A.Graboy, Yu.D.Tretyakov Superconductivity 1, 8 ed. by V.I.Ozhogin, Moscow 1987
- [7] G.F.Tavadze et al. Metallurgy 131 (1988), 97
- [8] R.Kubo and T.Toyabe, in: Magnetic Resonance and Relaxation, ed. by R.Blinc (1967)
- [9] Gang Xiao et al. Sol.St.Comm. 63 (1987), 817
- [10] A.Furrer, P.Briesch, P.Unternahrer Phys. Rev.B, 38,N7,1988, 4616-23
- [11] A.K.Zvezdin, V.M.Matveev, A.A.Muhin, A.I.Popov, Rare-earth ions in magnetically ordered crystals, (Nauka, Moscow, 1985)
- [12] P.Fischer, K.Kakurai, M.Steiner, K.N.Clausen, B.Lebeck, F.Hulliger, H.R.Ott, P.Bruesch, P.Unternahrer, Physica C152, 1988, 145
- [13] H.B.Moller, J.Z.Jensen, M.Wulff, A.R.Mackintosh, O.D.McMasters, K.A.Gschneidner Jr., Phys.Rev.Lett.,49, N7, 1982, 482
- [14] A.P.Levanyuk, A.S.Sigov. Defects and Structural Phase Transitions (Gordon and Breach Science Publishers, NY, 1988)

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