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Magnetic order in $La_{1.65}Eu_{0.20}Sr_{0.15}CuO_4$ studied by ^{57}Fe Mössbauer spectroscopy

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Abstract. ^{57}Fe Mössbauer effect studies of $La_{1.65}Eu_{0.20}Sr_{0.15}CuO_4$ doped with 0.5 at% ^{57}Fe performed in the temperature region 300 K $> T >$ 4.2 K give an onset temperature for magnetic ordering of 35 $\pm$ 5 K. This temperature practically is the same as that found in Nd doped $La_{2-x}Sr_xCuO_4$. It indicates that the magnetic ordering temperature in the LTT phase of rare earth (RE) doped $La_{2-x}Sr_xCuO_4$ is independent of the RE moment. The direction of the ^{57}Fe magnetic moment in the magnetically ordered state is within the CuO_2 plane, while it has been found to be parallel to the c-axis in Nd doped $La_{2-x}Sr_xCuO_4$.

PACS. 76.80.+y Mössbauer effect; other γ-ray spectroscopy – 74.25.Ha Magnetic properties – 74.72.Dn La-based cuprates

Rare earth (RE) doped $La_{2-x}Sr_xCuO_4$ has attracted considerable interest in recent years due to the fact that the low-temperature tetragonal (LTT) phase of these compounds exhibits a competition between superconductivity and magnetic order [1–4]. The latter one is especially interesting since neutron diffraction experiments have manifested that the magnetic order occurs in spin stripes separated by quasi one-dimensional charge walls [5]. As it was shown in a previous paper [6] Mössbauer Effect (ME) spectroscopy can be quite useful for studying the magnetic order in the LTT phase of RE doped $La_{2-x}Sr_xCuO_4$. Indeed, it were ^{57}Fe as well as ^{119}Sn ME experiments which revealed magnetic order in $La_{1.25}Nd_{0.60}Sr_{0.15}CuO_4$ below $T_N \approx 32$ K [6] a long time before this was later established by neutron scattering [5] and by muon spin rotation (μSR) experiments [7]. Based on this fact we decided to investigate by means of ^{57}Fe ME spectroscopy if magnetic order also occurs in the LTT phase of Eu doped $La_{2-x}Sr_xCuO_4$. The question is particularly interesting due to the following reason: in contrast to Nd^{3+} the Eu^{3+} ($^7F_0$) ions do not carry a magnetic moment in $La_{2-x}Sr_xCuO_4$; the frustration effect of the spin stripes crossing at right angles between neighbouring CuO_2 planes may lead to a suppression of the long range magnetic order for the case that the RE ions sitting between the CuO_2 planes do not carry a magnetic moment.

The polycrystalline sample of $La_{1.65}Eu_{0.20}Sr_{0.15}CuO_4$ doped with 0.5 at% ^{57}Fe was prepared using a solid state reaction technique described elsewhere [8]. X-ray diffraction measurements show the LTT structural phase occurring below $T_{LT} \approx 130$ K. AC susceptibility measurements reveal a strongly reduced superconductivity (strong reduction of AC shielding signal as well as of T_c) [9]. ESR measurements on $La_{1.70}Eu_{0.15}Sr_{0.15}CuO_4$ samples doped with 1 at% Gd indicate a considerable slowing down of the frequency of the antiferromagnetic fluctuations below ≈ 50 K by almost three orders of magnitude [10]. ^{57}Fe experiments have been performed in a variable temperature He cryostat in the temperature region $4.2 \leq T \leq 300$ K. The ^{57}Co:Rh ME source always was kept at room temperature outside of the He cryostat.

Figure 1 shows the ^{57}Fe ME spectra of $La_{1.65}Eu_{0.20}Sr_{0.15}CuO_4$ taken at various temperatures between 300 K and 4.2 K. The pure quadrupole doublet appearing at 300 K broadens at low temperatures (see *e.g.* the 18 K-spectrum) and finally transforms into a magnetically split ME spectrum at 4.2 K. Such spectra can be attributed either to a slowing down of magnetic relaxation with decreasing temperature or to a magnetic ordering at low temperature with a very broad distribution of the ordering temperature T_N. In order to determine more exactly the temperature at which the broadening of the quadrupole doublet starts, we have measured in addition the ^{57}Fe ME spectra at a small velocity scale (see Fig. 2). The temperature dependence of the resonance absorption area A of the quadrupole doublet, *i.e.* of the nonmagnetic part of the ME spectrum, was determined from these spectra. The result, shown in Figure 3, indicates an increase of A with decreasing temperature in the region 300 K to 100 K, which is due to the increase in the Mössbauer f-factor, followed by a *decrease* in A below $T_{onset} = (35 \pm 5)$ K. This temperature T_{onset} gives the temperature below which the

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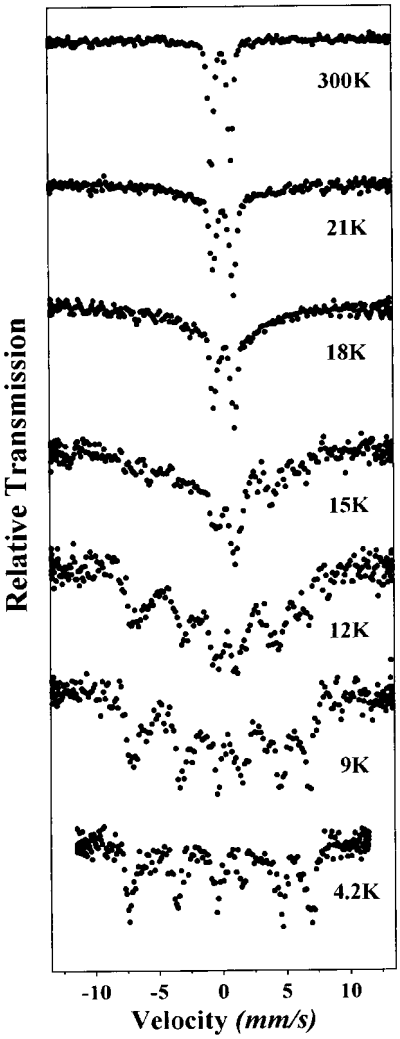

Fig. 1. ^{57}Fe ME spectra of $La_{1.65}Eu_{0.20}Sr_{0.15}CuO_4$ doped with 0.5 at% ^{57}Fe at various temperatures ($300 \geq T \geq 4.2$ K) measured with high drive velocities ($v_{max} \approx 12$ mm/s).

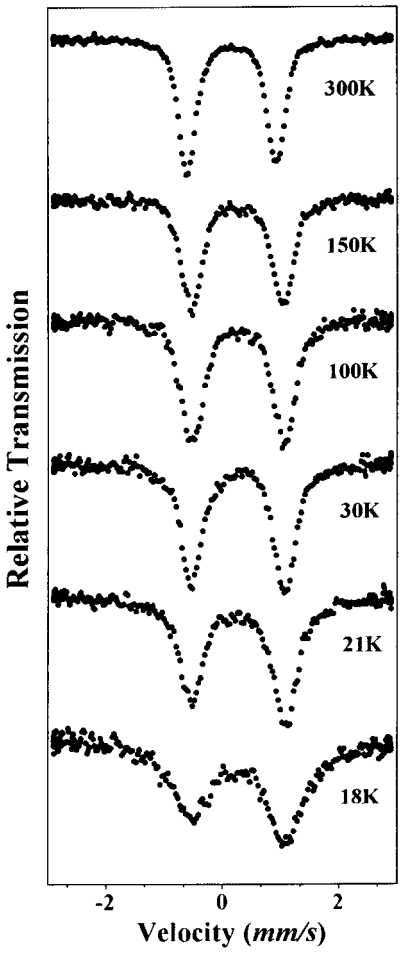

Fig. 2. ^{57}Fe ME spectra of $La_{1.65}Eu_{0.20}Sr_{0.15}CuO_4$ doped with 0.5 at% ^{57}Fe at various temperatures ($300 \geq T \geq 18$ K) measured with low drive velocities ($v_{max} \approx 3$ mm/s).

magnetic relaxation time τ starts to become $> 10^{-10}$ s. In this respect, T_{onset} gives an upper limit for the magnetic ordering temperature T_N. If one looks at the high-velocity ME spectra (see Fig. 1) one immediately recognizes that even at $T = 18$ K (corresponding to $T = 0.5T_{onset}$) a large part of the ^{57}Fe nuclei still show fast paramagnetic relaxation. This indicates that the magnetic ordering temperature experienced by most of the ^{57}Fe nuclei is quite below 35 K. Recent μSR studies on the same compound (however *not* doped with ^{57}Fe) reveal a begin of magnetic ordering at $T_N \approx 27$ K which is seen as the starting of a rotation signal [11]. If one considers the different time scales τ of the two measuring techniques for detecting the onset of slow magnetic relaxation (^{57}Fe ME spectroscopy: $\tau > 10^{-10}$ s, μSR: $\tau > 10^{-6}$ s) one expects a higher T_{onset} in the case of ^{57}Fe ME spectroscopy compared to that found in μSR. Thus, both microscopic techniques working with different local probes (^{57}Fe and muon, respectively) essentially are in agreement concerning the onset temperature of magnetic ordering in $La_{1.65}Eu_{0.20}Sr_{0.15}CuO_4$. This temperature practically is the same as that found for $La_{1.25}Nd_{0.60}Sr_{0.15}CuO_4$ [6]. Therefore, the first conclusion which can be drawn is the following: the magnetic ordering temperature T_N in the LTT phase of RE doped $La_{2-x}Sr_xCuO_4$ is independent of the RE magnetic moment. However, as we will see in the following, this is not the case for the *direction* of the ^{57}Fe magnetic moments in the magnetically ordered state.

Information on the direction of the ^{57}Fe magnetic moments is obtained from the ^{57}Fe ME spectrum of the magnetically ordered state at 4.2 K (see ME spectrum at the bottom of Fig. 1). Just by comparing this spectrum with that obtained for $La_{1.25}Nd_{0.60}Sr_{0.15}CuO_4$ [6] one immediately sees that these spectra have a reversed asymmetry, *i.e.* the angle between the magnetic hyperfine field B_{hf} and the main component V_{zz} of the electric field gradient (EFG) tensor for $La_{1.65}Eu_{0.20}Sr_{0.15}CuO_4$ is quite different from that in $La_{1.25}Nd_{0.60}Sr_{0.15}CuO_4$. The 4.2 K-spectrum has been fitted taking the complete Hamiltonian (magnetic and quadrupole interaction) and assuming that the quadrupole interaction ΔE_Q is given by the

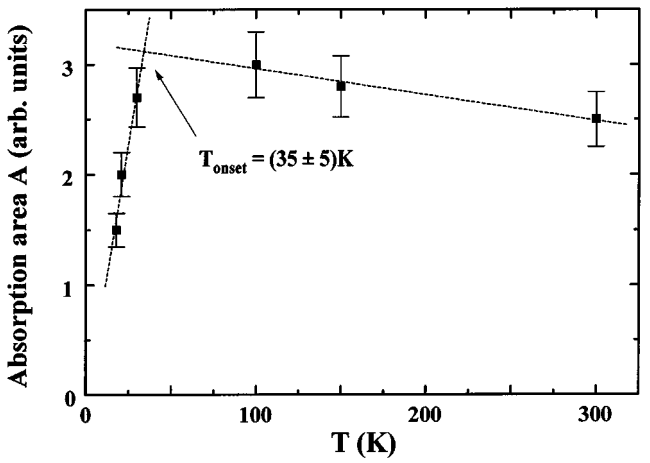

Fig. 3. Resonance absorption area A of quadrupole doublet as a function of temperature. The dashed straight lines through the data points above and below T_{onset} are least squares fits.

ΔE_Q values measured at 100 K and 30 K and extrapolated to $T \to 0$ ($\Delta E_Q(0) = 1.4$ mm/s). We obtain a value for the magnetic hyperfine field $B_{hf} = 44$ T. This value just is between that found for undoped $La_{1.85}Sr_{0.15}CuO_4$ ($B_{hf} = 42$ T [12]) and that for $La_{1.25}Nd_{0.60}Sr_{0.15}CuO_4$ ($B_{hf} = 46$ T [6]). The angle θ between the directions of B_{hf} and V_{zz} is $\theta = 90°$, *i.e.* since V_{zz} is parallel to the c-axis the ^{57}Fe magnetic moments are lying in the CuO_2 plane. The same was found for undoped $La_{2-x}Sr_xCuO_4$ with $x \leq 0.15$ [12]. In the case of Nd doped $La_{2-x}Sr_xCuO_4$, however, the ^{57}Fe magnetic moment direction has been measured to be parallel to the c-axis, *i.e.* to be out of the CuO_2 plane [6].

It is tempting to conclude from this observed difference in the ^{57}Fe magnetic moment direction between Eu doped and Nd doped $La_{2-x}Sr_xCuO_4$ that also the direction of the Cu magnetic moments in the magnetically ordered state depends on the RE moment. This would mean that the coupling between the Cu magnetic moments in two adjacent CuO_2 planes *via* the RE magnetic moments, sitting between these two planes, results in a change of the Cu magnetic moment direction from in-plane to out-of plane in the case of Nd. However, while this definitely is true for the ^{57}Fe magnetic moments, it may be not valid for the much smaller Cu magnetic moments ($\mu_{Cu} \approx 0.3\mu_B$, $\mu_{Fe} \approx 3\mu_B$) which may couple much weaker to the RE magnetic moments. Nevertheless, our finding with respect to the ^{57}Fe magnetic moment direction certainly gives a hint for interesting further experiments regarding the Cu magnetic moment direction in the LTT phase of RE doped single crystals of $La_{2-x}Sr_xCuO_4$. Cu NMR experiments on single crystals of RE doped $La_{2-x}Sr_xCuO_4$ certainly would be most suitable for such studies.

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