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(Cu0.97Fe0.03)3O7-x [R = Y, Pr, Er]
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MÖSSBAUER HYPERFINE FIELDS IN $\text{RBa}_2(\text{Cu}_{0.97}\text{Fe}_{0.03})_3\text{O}_{7-x}$ [$\text{R} = \text{Y, Pr, Er}$]

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Abstract. - Room temperature $^{57}\text{Fe}$ Mössbauer spectra of $\text{RBa}_2(\text{Cu}_{0.97}\text{Fe}_{0.03})_3\text{O}_{7-x}$ [$\text{R} = \text{Y, Pr, Er}$] were obtained from samples with varying $x$. A magnetically-split hyperfine field spectrum was observed for the most oxygen-deficient $\text{Y}$ sample, for all the $\text{Pr}$ samples, and for none of the $\text{Er}$ samples.

A large number of investigators [1] have studied the Mössbauer spectra of $^{57}\text{Fe}$-substituted $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ samples. Though different in the details, all of the results have certain similarities. There is an “outer” doublet (isomer shift, I.S. $\approx 0$ and quadrupole splitting, $\Delta \approx 1.9 \text{ mm/s}$), and an “inner” doublet (I.S. $\approx 0$ and $\Delta \approx 1.0 \text{ mm/s}$). In addition, there is a resolved single line (I.S. $\approx 0.1$), which has been most often interpreted as part of a doublet whose other line is unresolved. Additional unresolved doublets have also been invoked, in part to attempt to account for asymmetric lines. Complex magnetic hyperfine spectra reminiscent of spin glasses, with a distribution of hyperfine fields and a maximum $H_{\text{eff}} \approx 60 \text{ kG}$, have been observed at low temperatures. There have been few reports [2] of simple 6-line magnetic hyperfine splitting with $H_{\text{eff}} \approx 300 \text{ kG}$ at room temperature, which is the subject of the present paper.

Suitable portions of $\text{Y}_2\text{O}_3$, $\text{Pr}_2\text{O}_3$, or $\text{Er}_2\text{O}_3$ were mixed with $\text{BaCO}_3$, $\text{CuO}$, and $^{57}\text{Fe}_2\text{O}_3$ to produce $\text{RBa}_2(\text{Cu}_{0.97}\text{Fe}_{0.03})_3\text{O}_{7-x}$, where $\text{R} = \text{Y, Pr, and Er}$. These were fired under oxygen for 30 hours at $930 \degree \text{C}$, slowly cooled, reground and pressed in pellets. Each pellet was placed in a small alumina crucible and heated to $950 \degree \text{C}$ under flowing oxygen for four hours. The samples were then quenched from varying temperatures into liquid nitrogen in order to vary the oxygen content. X-ray diffraction showed that all these samples are tetragonal or pseudo-tetragonal. Most of the $\text{R} = \text{Y}$ and $\text{R} = \text{Er}$ samples were superconducting, whereas none of the $\text{R} = \text{Pr}$ were.

The room-temperature $^{57}\text{Fe}$ Mössbauer spectra were obtained for these quenched samples. A magnetically-split six-line hyperfine field with $H_{\text{eff}} = 295 \text{ kG}$, accounting for $\approx 25 \pm 5 \%$ of the spectral area, was found at $300 \text{ K}$ for the $\text{Y}$ sample quenched from $950 \degree \text{C}$ ($x \approx 0.9$). For the $\text{Pr}$ samples quenched from temperatures varying from $920 \degree \text{C}$ to $30 \degree \text{C}$ ($x \approx 0$ to $0.9$), hyperfine fields with $H_{\text{eff}} \approx 270 \text{ kG}$ were found, accounting for $\approx 20 \pm 5 \%$ of the spectral area (see Figs. 1 and 2). None of the $\text{Er}$ samples quenched from any temperature up to $900 \degree \text{C}$ showed a magnetic hyperfine field at $300 \text{ K}$.
Neutron diffraction experiments have shown [3, 4] that tetragonal $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ($\delta \approx 1$) is antiferromagnetic, with $T_N \approx 500$ K. The magnetism arises from antiferromagnetic coupling of the planar Cu(2) atoms. We believe that those iron atoms which substitute for the planar Cu(2) atoms experience this antiferromagnetism, resulting in the room-temperature hyperfine field. It can be expected (and preliminary neutron diffraction experiments appear to confirm [5]) that the same type of antiferromagnetism arises when $R = \text{Pr}$. Pr is probably at least partially tetravalent in this oxide, and may mimic the effect of oxygen depletion by adding electrons to the system. Antiferromagnetism is reflected in the room-temperature magnetic hyperfine fields, which are evident for all quench temperatures, implying an above-room temperature antiferromagnetism for a large range of oxygen compositions.

In conclusion, the antiferromagnetism displayed by the Cu-O planes is apparently not related to the magnetic moments of the rare-earth atoms. Thus, the Er superconductors do not show any room-temperature magnetic hyperfine fields even though $\text{Er}^{3+}$ has a larger magnetic moment ($\approx 9 \mu_B$) than either $\text{Pr}^{3+/4+}$ ($\approx 3 \mu_B$ or less) or $\text{Y}^{3+}$ (0 $\mu_B$). Temperature studies of these samples are underway. None of the samples showing magnetic hyperfine splitting at room temperature are superconducting, at least down to 10 K. This contrasts with the coexistence of magnetism and superconductivity observed [6] when a magnetic hyperfine pattern appears at low temperatures.

We further conclude that, based on the observed relative spectral areas, the percentage of Fe atoms substituting for the planar Cu(2) in these samples where 3 % of the Cu atoms have been replaced with Fe can be estimated. When the oxygen deficiency causes the occurrence of antiferromagnetism, only those Fe atoms residing on the magnetically aligned planes probe the magnetic order and produce the 6-line pattern. Therefore, approximately 20 % of the Fe atoms reside on the Cu(2) sites, for both $R = \text{Y}$ and $R = \text{Pr}$. When the magnetic pattern is present, the intensity of the outer doublet is unchanged, whereas the inner doublet and singlet appear somewhat reduced in intensity. This is consistent with the previous conclusion [7], based on quadrupole doublet asymmetry arguments, that the majority of atoms in the outer doublet are associated with Fe in the Cu(1) chain sites.