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Magnetic, specific heat and ^{151}Eu Mössbauer studies on $Ba_2EuRu_{1-x}Cu_xO_6$ ($0 \leqslant x \leqslant 0.2$) compounds

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Abstract

Magnetic, ^{151}Eu Mössbauer and specific heat measurements have been carried out on $Ba_2EuRu_{1-x}Cu_xO_6$ ($0 \leqslant x \leqslant 0.2$) compounds. From the magnetization data, all the compounds are found to be antiferromagnetically ordered near 40 K, independent of Cu content. A sharp peak at 40 K in the specific heat data also confirms the magnetic ordering. The Eu ions in all the compounds are in the 3+ state as inferred from the ^{151}Eu Mössbauer isomer shift values. However, a well split, six line pattern is observed in the ^{151}Eu Mössbauer spectra of these compounds below 40 K. This is rather rare for Eu^{3+} ($4f^6, J = 0$) which is a nonmagnetic ion. The magnetic hyperfine field at the Eu site is found to be ~28 T at 4.2 K and is presumed to arise due to the strong Ru–O–Eu exchange interaction.

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The discovery of superconductivity in Cu substituted $Sr_2YRu_{1-x}Cu_xO_6$ compounds [1] has initiated extensive studies in A_2RRuO_6 (A = alkaline-earth metal, R = rare-earth metal) compounds. The studies are directed to understand how superconductivity can occur in these compounds which lack Cu–O planes. We have prepared and studied several related compounds and here we report the magnetic properties of $Ba_2EuRu_{1-x}Cu_xO_6$ ($0 \leqslant x \leqslant 0.2$) compounds through magnetization, ^{151}Eu Mössbauer and specific heat measurements.

All the samples were prepared using the standard solid state reaction method. The final sintering of the pellets was carried out at 1100 °C for 24 h. Magnetization was measured using a SQUID magnetometer (Quantum Design, USA) and specific heat was measured by the relaxation method (PPMS, Quantum Design, USA). The ^{151}Eu Mössbauer absorption spectra were recorded at different temperatures between 4.2 and 300 K.

The Rietveld analysis of the X-ray diffraction data confirmed that the compounds form in a single phase cubic structure (space group—Fm$\bar{3}$m, No. 225). The lattice parameter for the parent compound is 8.405 Å with no appreciable change as Cu is substituted for Ru.

The magnetic ordering temperature (T_M) for the parent compound is 40 K. The magnetization is found to vary linearly with the magnetic field below 40 K [2], which indicates the antiferromagnetic nature of the ordering. No appreciable change in the magnetic ordering temperature is observed on substituting Cu for Ru in the parent compound [2]. The magnetic susceptibility for the $x = 0.1$ sample is shown in Fig. 1 as a function of temperature. The difference between the zero-field-cooled (ZFC) and the field-cooled (FC) magnetization for all the samples were found to be negligible. This type of behaviour is typical of an ideal antiferromagnet. The inverse susceptibility of all the compounds shows a deviation from the Curie–Weiss

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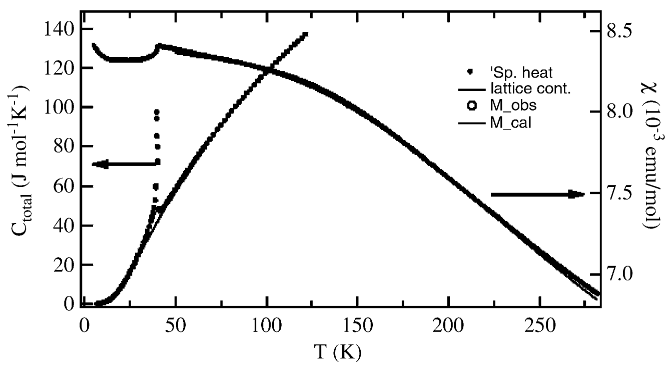

Fig. 1. Specific heat and magnetization vs. temperature for $Ba_2EuRu_{0.9}Cu_{0.1}O_6$.

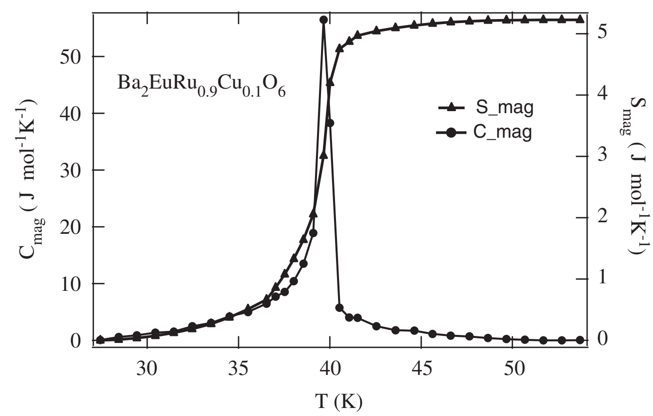

Fig. 2. Magnetic specific heat and magnetic entropy against temperature for $Ba_2EuRu_{0.9}Cu_{0.1}O_6$.

law below 190 K which may be attributed to the crystalline electric field effects originating from the low lying multiplets of Eu^{3+}. The total susceptibility can be written as

$$\chi = \chi(Eu^{3+}) + \chi(Ru) + \chi_0 \quad (1)$$

where $\chi(Ru) = C/(T - \theta_p)$ is the Curie–Weiss contribution to the susceptibility from the Ru ions and χ_0 is the temperature independent susceptibility. The molar susceptibility of Eu^{3+} is given by [3]

$$\chi(Eu^{3+}) = \frac{N_A \mu_B^2}{3\gamma T k_B} \times \frac{24 + X + Y + Z}{1 + 3e^{-\gamma} + 5e^{-3\gamma} + 7e^{-6\gamma}} \quad (2)$$

Here $X = (13.5\gamma - 1.5)e^{-\gamma}$, $Y = (67.5\gamma - 2.5)e^{-3\gamma}$, $Z = ((189\gamma - 3.5)e^{-6\gamma}$ and $\gamma = \lambda/k_B T$ in which λ is the spin–orbit coupling constant. The observed susceptibility above 50 K can be nicely fitted to Eq. (1) (Fig. 1) and yields the following parameters: $\chi_0 = 0.0014\,emu\,mol^{-1}$, $C = 0.8725\,emu\,K\,mol^{-1}$, $\theta_p = -624\,K$ and $\lambda = 365\,cm^{-1}$. The value of C corresponds to an effective magnetic moment of $2.94\,\mu_B$ per Ru ion, which may be compared with the value of $3.1\,\mu_B$ [2] for Ru^{5+} ion.

The specific heat for all the samples shows a sharp peak at 40 K (Fig. 1) which is again consistent with the magnetic ordering temperature. The magnetic contribution to the specific heat (C_{mag}) for the $x = 0.1$ sample was calculated by subtracting the background (lattice contribution) and is shown in Fig. 2 along with the magnetic entropy (S_{mag}). The entropy corresponding to the magnetic contribution was calculated by integrating C_{mag}/T vs. T. S_{mag} is found to be $5.22\,J\,mol^{-1}\,K^{-1}$, which is close to R ln$(2S + 1)$ with $S = \frac{1}{2}$ for Ru.

Fig. 3 shows the ^{151}Eu Mössbauer spectra for the $x = 0.1$ sample at 40 K and 4.2 K below the magnetic ordering temperature. The Isomer shift values correspond to Eu^{3+} which is a nonmagnetic state. However, the spectra show a well-split six line pattern below 40 K (Fig. 3), which is unusual. This may be due to the strong magnetic hyperfine field acting at the Eu site through the Ru–O–Eu exchange interaction [4]. Magnetic hyperfine field at the Eu site is found to be ~28 T at 4.2 K which decreases with the increase in temperature and disappears above 40 K, as evident from a single line Mössbauer spectra.

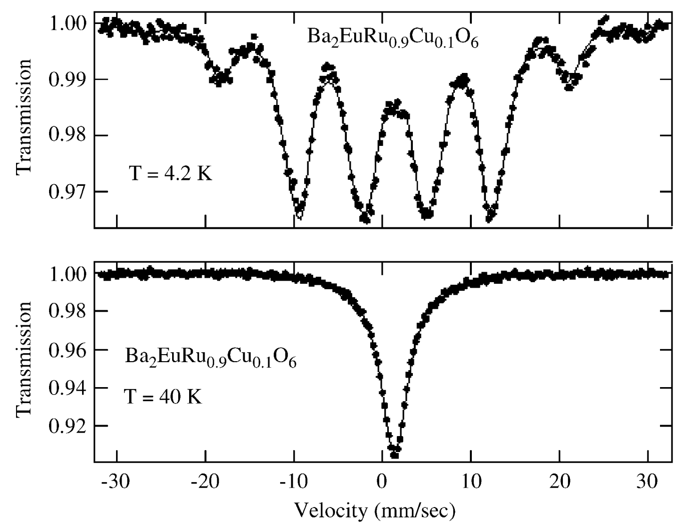

Fig. 3. ^{151}Eu Mössbauer spectra at 4.2 K and 40 K for $Ba_2EuRu_{0.9}Cu_{0.1}O_6$.

In conclusion, we have shown that the substitution of Cu for Ru in $Ba_2EuRuCuO_6$ does not affect the magnetic properties of the compound. A large hyperfine field exists at the Eu site, as inferred from the Mössbauer data due to the strong exchange interaction between the ordered Ru moments.

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