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Muon-spin-rotation measurements in the 'infinite-chain' Ca_2CuO_3

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Abstract

We performed zero-field muon-spin-rotation measurements of the 'infinite-chain' compound Ca_2CuO_3. A spontaneous magnetic field is observed below 13 K. The exchange interaction inferred from susceptibility measurements and two-magnon Raman scattering is approximately 10^3 K, which implies a remarkable suppression of the ordering temperature ($k_BT_N/J_{1d} \sim 0.01$). We discuss the relevance of these measurements to the problem of one-dimensional spin $\frac{1}{2}$ antiferromagnets.

The subject of one-dimensional (1D) spin $\frac{1}{2}$ chains has drawn considerable theoretical and experimental attention in the past few years [1]. However, this research is restricted by the difficulty of finding systems in which the magnetic interaction is approximately 1D. Therefore, there is considerable interest in new materials which could better serve this study. Because of their crystal structure, the 'infinite chain' (IC) compounds R_2CuO_3 (R is Sr or Ca) are possibly such materials. In the orthorhombic lattice of the IC compounds, the CuO chains lie along the $\hat{b}$ axis, while along the $\hat{a}$ axis there are no oxygen atoms to carry the superexchange interaction. The distance between Cu atoms along the $\hat{c}$ axis is roughly three times as large as this distance along the $\hat{a}$ or $\hat{b}$ axis [2,3]. This structure leads us to anticipate 1D behavior in the IC systems. Indeed, we previously found evidence [3] that Sr_2CuO_3 has a very small ratio of Néel temperature (T_N) to exchange interaction (J), an important criterion for a good 1D system. We confirm the low dimensionality of the Ca_2CuO_3 by measuring the Néel temperature with the μSR technique and by comparing it to the exchange interaction deduced from two-magnon Raman scattering and susceptibility experiments. In Ca_2CuO_3 we find that $T_N/J_{1d} = 0.01$. This ratio is smaller than any other prototypical 2D [4] or 1D [5] spin system known to us. The μSR experiment is also used to determine the magnetic properties of the IC in the ordered state.

The dc-susceptibility measurements were made using a commercial SQUID magnetometer between 5 and 300 K. The susceptibility data appearing in the inset of Fig. 1 is obtained after subtracting core diamagnetism ($\chi_{dia} = -78 \times 10^{-6}$ emu/G-mole) from the measured values [6]. In this figure we also plot the susceptibility after subtracting a magnetic impurity contribution, which was assumed to follow a Curie law. The estimated magnetic impurity density, assuming Cu^{2+} impurities, is 0.13%. In addition, in the inset of Fig. 1, we compare the susceptibility measurement with a model calculation for the 1d Heisenberg spin chain, consisting of $S=\frac{1}{2}$ moments coupled

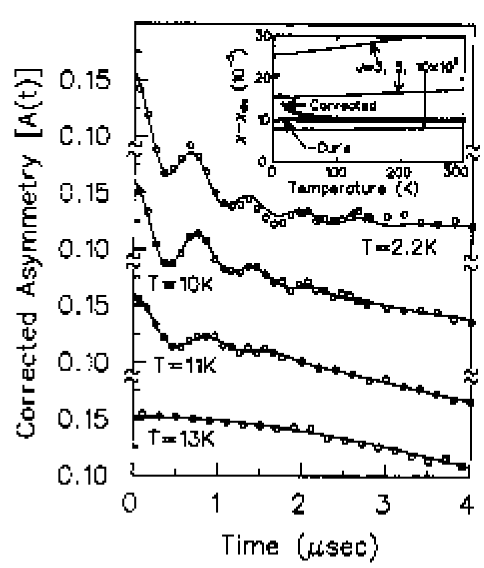

Fig. 1. Zero field μSR data in Ca_2CuO_3. The inset shows both the corrected susceptibility $\chi - \chi_{dia}$ (in units of emu/G-mole), and the corrected susceptibility with a Curie term subtracted as described in the text. The inset also shows the expected susceptibility in a one-dimensional model [7].

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antiferromagnetically, as given by Bonner and Fisher [7]. The temperature dependence of the theoretical dc-susceptibility is shown for several values of the exchange interaction J_{1d}. According to this theory, J_{1d} in Ca_2CuO_3 is of the order 10^3 K.

Fig. 1 shows Zero Field (ZF) μSR time spectra measured in Ca_2CuO_3. A description of the ZF μSR technique can be found in Ref. [8]. At 13 K we find only muon spin relaxation while at 11 K we find muon spin oscillations indicating a static local field. Therefore, the Néel temperature lies in the range $11 < T_N < 13$ K. The critical region of less than 2 K is rather small and is consistent with a first order phase transition. In addition, no critical fluctuations were found by longitudinal field μSR measurements (not shown) around T_N. After several attempts we find that the best fit to the μSR data is achieved with the form

$$A(t) = A_d\, e^{-t/T_L} + e^{-(t/T_G)^2}\left(A_1 \cos(2\pi\nu_1 t) + A_2 \cos(2\pi\nu_2 t)\right). \quad (1)$$

The three amplitudes are fixed, and the contribution to the relaxation from nuclear moments and background is deduced from the high temperature data. It should be noted that the values we obtain for the frequencies depend only very weakly on the fitting scheme. The presence of two frequencies could be a result of two muon sites, or a unique crystallographic site, which is not symmetric with respect to the magnetic lattice. That the observed asymmetries are different, $A_1 = 0.008$ and $A_2 = 0.0175$ (with $A_d = 0.11$), supports the first possibility. Interestingly, the muon relaxation function in Ca_2CuO_3 is very similar to the one observed in the metallic spin density wave system $(TMTSF)_2$–X, even though the IC is insulating [9].

The temperature dependence of the two fitted frequencies is shown in Fig. 2. We see a fast reduction in the frequencies with increasing temperature, expected for the sub-lattice magnetization of a low dimensional spin system. The same behavior is observed in the sister compound Sr_2CuO_3 [3]. The similar temperature dependence of the two frequencies supports the assumption that they result from two different muon sites. In the inset of this figure we show the parameters $1/T_L$ and $1/T_G$ as a function of temperature. The absence of rapid dynamical fluctuations at low temperatures in an LF-μSR measurement (not shown) suggests that the T_L relaxation originates from a random component of the local field which is static or at most slowly fluctuating (on a time scale of 1 MHz). The increase in $1/T_L$ as $T \to 0$ is ascribed to an increase in the RMS of this random field. The relaxation of the oscillating part, $1/T_G$, could also result from static field inhomogeneity.

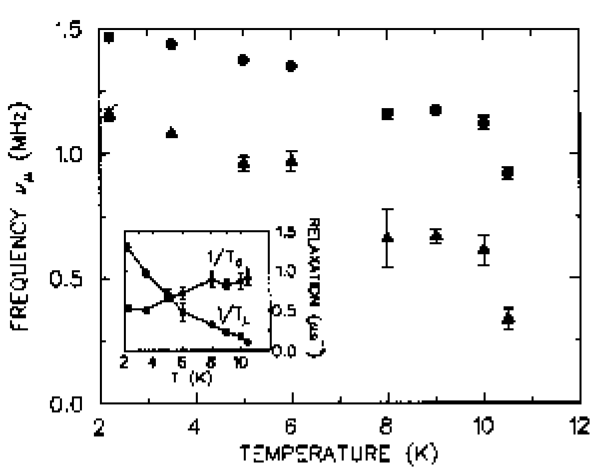

Fig. 2. The temperature dependence of the two frequencies. The inset shows the temperature dependence of the relaxation rates given in Eq. (1).

The μSR determination of T_N is completely unambiguous. On the other hand, there is a certain systematic ambiguity in the estimate of J from uniform susceptibility, due to the subtraction of an impurity Curie and diamagnetic terms. Therefore, it is important to estimate J from more direct measurements. For that purpose, Watanabe and Tajima performed a two-magnon Raman scattering experiment in SRL Tokyo [10]. They found a very broad peak in the frequency shift at $T = 7$ K centered at $\Delta E = 2200$ cm^{-1}, indicating an exchange constant J in the order of 10^3 K [11].

The combination of the μSR results with the susceptibility and two-magnon Raman scattering demonstrates that Ca_2CuO_3 is a very good quasi one-dimensional compound with a ratio $T_N/J_{1d} \leq 0.01$. This ratio indicates that the exchange interaction between the CuO chains is extremely small, though finite. For comparison, in the 'infinite-layer' system $CaCuO_2$ in which oxygen atoms are placed along both the $\hat{a}$ and $\hat{b}$ axis (CuO_2 planes) and the interaction is roughly 3D with $J \sim 10^3$ K, the Néel temperature is 540 K [3]. It will be interesting to study charge-doped CuO chains as a realization of the 1D Hubbard model when such specimens become available.

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