AC susceptibility in weak ferromagnetic $R_2CuO_4$ cuprates

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We present ac susceptibility measurements for $R_2CuO_4$ with $R =$ Gd, Tb, Dy, Ho, Er and Tm, in the temperature range from 4 to 300 K. The frequency dependence of the in-phase $\chi'(\omega, T)$ and out-of-phase $\chi''(\omega, T)$ is analyzed as a function of temperature.

Antiferromagnetic order [1] is a common characteristic of the rare-earth cuprates, $R_2CuO_4$. For the light rare earths ($R =$ Pr, Nd, Sm and Eu) electron doping through Ce substitution leads to high-$T_c$ superconductivity [2]. For heavier rare earths, instead, a weak ferromagnetic (WF) component is present [3-5] and, up to now, it has not been possible to induce superconductivity in these materials. The dc magnetization [3] reflects a history-dependent magnetization of the Cu lattice and an associated internal field polarizing the paramagnetic rare-earth ions. The onset of WF in $Wb_2CuO_4$ has been found to be accompanied with spin-glass-like characteristics [5], such as differences between the field-cooled (FC) and zero-field-cooled (ZFC) magnetization and a logarithmic time decay of the remanent magnetization. We present here an analysis of the frequency dependence of the real (in-phase) and imaginary (out-of-phase) components of the ac susceptibility, which further characterizes the spin-glass-like features associated with the weak ferromagnetism of these systems.

Ceramic samples of $R_2CuO_4$, with $R =$ Tb, Dy, Ho, Er, and Tm, were prepared from the oxides under high pressure (8-9 GPa) in a belt type apparatus at temperatures ranging from 800 to 1200 °C. X-ray diffraction showed in all cases the basic $T'$ structure [6] but showed many extra weak peaks that could be indexed as superstructure reflections. Details of the synthesis and structural studies will be published separately [7]. Single crystals of $Gd_2CuO_4$ were grown from a CuOP-PbO flux.

We have measured the real, $\chi'(\omega, T)$, and imaginary, $\chi''(\omega, T)$, parts of the ac susceptibility, in the temperature range 4.2 K $< T <$ 320 K. X-ray diffraction showed the peaks in $\chi'(\omega, T)$ have associated anomalies in $\chi''(\omega, T)$, as shown in figs. 1 and 2 for $Tb_2CuO_4$ and $Gd_2CuO_4$. In the case of $R =$ Dy, Ho, Er and Tm, the anomalies were less intense and we were not able to detect the out-of-phase component.

For $T \gg T_{max}$ the ac and dc susceptibility measurements [3-5] are in agreement, following a Curie-Weiss law with effective magnetic moments close to the free-ion values for $R^{3+}$.

For $R =$ Tb and heavier rare earths, $\chi'(\omega, T)$ decreases below $T_{max}$, reaching for $T \approx T_{max}$ the same Curie-Weiss dependence found at high temperatures. For $Gd_2CuO_4$, $\chi'(\omega, T)$ also presents a maximum but it remains significantly higher than $\chi_{dc}(T)$ at lower temperatures.

As shown in fig. 1, the frequency dependence of $\chi'(\omega, T)$ is almost negligible for $T > T_{max}$, increases at lower temperatures, and becomes again independent.
of \( \omega \). The out-of-phase component \( \chi''(\omega, T) \) presents a broad maximum as a function of temperature whose shape and position are strongly frequency dependent. \( \chi''(\omega, T) \) drops to zero at temperatures close to those where \( \chi'(\omega, T) \) becomes frequency independent. At high temperatures, \( \chi''(\omega, T) \) is an increasing function of \( \omega \), and a decreasing one at low temperatures.

This behavior may be described in terms of relaxation processes characterized by a spectral distribution of relaxation times \( P(\tau) \). For a single relaxation time, \( P(\tau) = \delta(\tau - \tau_0(T)) \), the ac susceptibility is given by

\[
\chi'(\omega, T) = \chi_0(T) + \frac{\chi_0(T) - \chi'(T)}{1 + \omega^2 \tau_0^2(T)},
\]

and

\[
\chi''(\omega, T) = \frac{\chi_0(T) - \chi'(T)}{1 + \omega^2 \tau_0^2(T)},
\]

where \( \chi_0(T) \) and \( \chi'(T) \) are the low and high frequency limits of \( \chi'(\omega, T) \), respectively. The imaginary part \( \chi''(\omega, T) \) tends to zero in both limits and presents a maximum for \( \omega = \tau_0^{-1}(T) \). For a distribution of relaxation times, \( \chi''(\omega, T) \) becomes a flattened function of frequency, but its maximum value still corresponds to the average or dominant relaxation rates in the system \( \tau(T) \).

The available excitation frequencies in our ac measurements correspond to a time window \( 10^{-3} \text{ s} < \tau < 10^{-1} \text{ s} \). From our experimental results we conclude that \( \tau(T) < 10^{-3} \text{ s} \) for \( T > T_{\text{max}} \) and coincides with our time window at \( T = 250 \text{ K} \) for Tb$_2$CuO$_4$ and at \( T = 200 \text{ K} \) for Gd$_2$CuO$_4$. At lower temperatures, the dominant relaxation times rapidly increase and then \( \chi'(\omega, T) \) becomes again frequency independent, while \( \chi''(\omega, T) \) goes to zero. This observation is in agreement with the logarithmic decay of the remanent magnetization observed [5] for Tb$_2$CuO$_4$ at \( T = 100 \text{ K} \), i.e. below the temperature range where strong relaxation is observed in our ac susceptibility measurements.

The adiabatic susceptibility \( \chi_a(T) \), measured for \( T \leq 150 \text{ K} \), follows a Curie–Weiss law for \( R = \text{Tb} \) and the heavier rare earths. This behavior and the observation of hysteresis loops with a slowly varying remanent magnetization [5] indicates the freezing of the Cu moments. Then, only the rare-earth moments are able to respond to the excitation of the oscillating magnetic field.

For Gd$_2$CuO$_4$, the low-temperature limit of \( \chi_a(T) \) resembles also a Curie–Weiss law, but it corresponds to an effective magnetic field much larger than the applied ac excitation field \( H_{\text{exc}} \).

As mentioned above, at a second characteristic temperature, \( T_L \), some of these materials show a spin reorientation transition [3,8] that suppresses the WF. We have found that \( T_L \) varies from \( \approx 20 \text{ K} \) for \( R = \text{Gd} \) to \( \approx 10 \text{ K} \) in the cases of \( \text{Tb} \) and \( \text{Dy} \). For \( R = \text{Ho}, \text{Er} \) and \( \text{Tm} \), we have not found indications of this transition down to \( 4.5 \text{ K} \).

In conclusion, we have identified a high-temperature region where the ac susceptibility presents characteristics of spin-glass materials. The observed behavior may be described in terms of a distribution of relaxation times whose average value varies as a function of temperature. Above \( T_{\text{max}} \) the relaxation is fast and the whole system behaves paramagnetically. In an intermediate temperature range, \( \tau(T) \) varies in the time domain of our ac measurements \( (10^{-1} - 10^{-3} \text{ s}) \) and both \( \chi'(\omega, T) \) and \( \chi''(\omega, T) \) present a strong frequency dependence. Finally, the Cu moments become frozen at lower temperatures \( T < 150 \text{ K} \) for \( R = \text{Tb} \) and heavier rare earths. For \( R = \text{Gd} \), instead, the freezing is not complete. The analysis of the interaction of the WF component of the Cu sublattice with the rare-earth magnetic moments at low temperatures and its effects on the observed transitions at \( T_L \) is presently under way and will be published separately.

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References