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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 Tb_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 $4.2\ K < T < 320\ K$, using a Lake Shore Susceptometer operating at different excitation frequencies of the magnetic field, $\nu = \omega/2\pi = 10$, 111 and 1000 Hz.

A maximum of $\chi'(\omega, T)$ has been found for all the ceramic samples at temperatures varying from $T_{max} \simeq 285$ K for R = Tb to $T_{max} \simeq 260$ K for R = Tm. Subsequent maxima were observed at lower temperatures indicating either spin reorientation transitions or rare-earth magnetic order. The overall magnetic behavior is similar to that found [3,8] for Gd_2CuO_4, where weak ferromagnetism was first reported for these T′ cuprates.

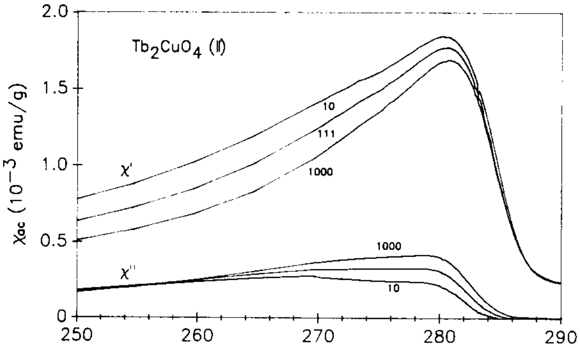

Fig. 1. Real and imaginary parts of the ac susceptibility, measured at two different frequencies for a ceramic sample of Tb_2CuO_4 with $H_{exc} = 10$ G.

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 $T \ll T_{max}$ 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

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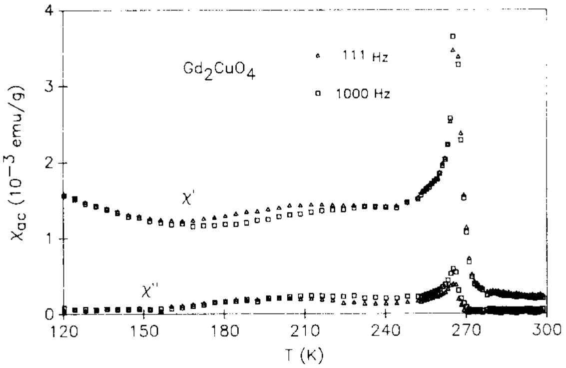

Fig. 2. Real and imaginary parts of the ac susceptibility measured for a Gd_2CuO_4 single crystals, with $H_{exc} = 1$ G, as a function of temperature for two different frequencies.

of ω. 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 ω, 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 $\chi'(\omega, T) = \chi_\infty(T) + (\chi_0(T) - \chi_\infty(T))/(1 + \omega^2\tau_0^2(T))$ and $\chi''(\omega, T) = \omega\tau_0(T)(\chi_0(T) - \chi_\infty(T))/(1 + \omega^2\tau_0^2(T))$, where $\chi_0(T)$ and $\chi_\infty(T)$ 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 $\bar{\tau}(T)$.

The available excitation frequencies in our ac measurements correspond to a time window 10^{-3} s $< \tau < 10^{-1}$ s. From our experimental results we conclude that $\bar{\tau}(T) < 10^{-3}$ s for $T > T_{max}$ and coincides with our time window at $T \approx 250$ K for Tb_2CuO_4 and at $T \approx 200$ K for Gd_2CuO_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_2CuO_4 at $T = 100$ K, i.e. below the temperature range where strong relaxation is observed in our ac susceptibility measurements.

The adiabatic susceptibility $\chi_\infty(T)$, measured for $T \lesssim 150$ K, follows a Curie–Weiss law for R = 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_2CuO_4, the low-temperature limit of $\chi_\infty(T)$ resembles also a Curie–Weiss law, but it corresponds to an effective magnetic field much larger than the applied ac excitation field H_{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 ≈ 20 K for R = Gd to ≈ 10 K in the cases of Tb and Dy. For R = Ho, Er and Tm, we have not found indications of this transition down to 4.5 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_{max} the relaxation is fast and the whole system behaves paramagnetically. In an intermediate temperature range, $\bar{\tau}(T)$ varies in the time domain of our ac measurements (10^{-1}–10^{-3} s) and both $\chi'(\omega, T)$ and $\chi''(\omega, T)$ present a strong frequency dependence. Finally, the Cu moments become frozen at lower temperatures ($T < 150$ K) for R = Tb and heavier rare earths. For R = 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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