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Rare-earth magnetic ordering in the R_2CuO_4 cuprates (R = Tb, Dy, Ho, Er and Tm)

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Low-temperature AC magnetic susceptibility measurements on the heavy rare-earth antiferromagnetic cuprates R_2CuO_4 which have a T' Nd_2CuO_4-like crystal structure are reported. A single susceptibility maximum is observed in all the compounds at different temperatures ranging from 1.5 K to 10 K, which is interpreted as indicative of antiferromagnetic long-range ordering of the rare-earth ions. No double peak structure is observed in any of these weak ferromagnetic compounds, at variance with the weak ferromagnetic compound Gd_2CuO_4 for which a second sudden transition, of uncertain origin, has been previously observed at $T_L \approx 20$ K. The overall dependence of the antiferromagnetic ordering temperature on the single ion rare-earth magnetic moments do not follow a simple scaling with the De Gennes factor but rather a scaling with the full rare-earth magnetic moments, suggesting a pseudo-dipolar origin for the leading magnetic interaction coupling the rare-earth ions. Some important deviations are observed, however, which may be indicative of strong crystal field effects. It is suggested that the Suhl–Nakamura mechanism may be a relevant mechanism to understand the rare-earth magnetic ordering in the R_2CuO_4 series (R = Nd– Tm).

The remarkable effects associated with rare-earth magnetic ordering in superconductors have been widely investigated in the past and produce a rich variety of physical phenomena, including magnetic pair breaking, magnetic-field induced superconductivity and the coexistence of magnetic and superconducting order [1].

The discovery of high-temperature superconductivity in the layered cuprates offers a new exciting opportunity to investigate the subtle relationship between magnetism, electronic structure and superconductivity.

Rare-earth magnetic ordering in superconducting or antiferromagnetic cuprates has already been observed in several systems including $RBa_2Cu_3O_{7-\delta}$ (R = Nd–Er) [2], R_2CuO_4 (R = Nd,Sm,Gd) [3], $R_2Cu_2O_5$ (R = Tb–Tm) [4] and R_2BaCuO_5 (R = Sm–Yb) [5].

A detailed study of the magnetic interactions involved in these oxide materials may greatly contrib-

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ute to the elucidation of the microscopic mechanism in high-temperature superconductivity. It has been suggested that the apparent independence of both phenomena in the $RBa_2Cu_3O_7$ series is caused by a small exchange interaction between 4f and CuO_2 electrons. However, some evidence for a stronger interaction seems to exist in $Y_{1-x}Pr_xBa_2Cu_3O_7$ [6] and maybe also in $Sm_{1-x}Ce_xCuO_4$ [7]. In the Y–Pr systems, for instance, magnetic pair-breaking effects have been identified as a relevant mechanism responsible of the decrease of the superconducting transition temperature, in addition to the well stablished hole-filling mechanism.

The existence of long-range rare-earth magnetic ordering in R_2CuO_4 (R=Nd,Sm,Gd) was demonstrated by means of specific heat [8], magnetic susceptibility [3,9] and neutron diffraction [10] measurements. In all these systems the lanthanide magnetic ordering occurs at temperatures much lower than the Néel temperature of the copper sublattice. In Gd_2CuO_4 and other mixed oxides containing heavier rare earths [3,9,11], a complex magnetic behavior has been observed and is associated with the appearance of a weak ferromagnetic component in the copper sublattice which interacts with the rare-earth ions.

Recently, new T′ cuprates with heavy rare-earths (R=Tb, Dy, Ho, Er, Tm) and Y have been synthesized under high pressures [12] and their magnetic properties have been widely investigated by DC magnetization, AC magnetic susceptibility and microwave magnetoabsorption [13]. It was shown, for instance, that the copper sublattice orders antiferromagnetically at a temperature only slightly dependent on the lanthanide size while a weak ferromagnetic behavior exists in all these oxides. Some differences in the dynamic behavior of their magnetic moments were found evidence for, as compared to Gd_2CuO_4. In the heavier rare-earth compounds, such as Tb_2CuO_4, a complete freezing of the copper non-collinear spin components occurs, with all the signatures of spin glass behavior [13]. In Gd_2CuO_4, on the other hand, the weak ferromagnetic component is still relaxing, as indicated by AC magnetic susceptibility, down to $T_L \approx 20$ K, where a sudden magnetic transition occurs, implying the disappearance of an internal magnetic field on the Gd sublattice.

In our previous investigation of R_2CuO_4 (R=Tb, Dy, Ho, Er, Tm, Y) cuprates down to 5 K [13], a single low-temperature maximum in the magnetic susceptibility was detected in Tb_2CuO_4 and Dy_2CuO_4, but there was insufficient experimental evidence to attribute this maximum to an antiferromagnetic transition or to a spin reorientation transition, similar to Gd_2CuO_4.

We report in this communication new AC magnetic susceptibility measurements at much lower temperatures (80 mK–16 K) which allow us to identify an antiferromagnetic transition not reported previously in the R_2CuO_4 compounds (R=Ho, Er, Tm) and to assess the absence of any additional susceptibility peak in the compounds, where a susceptibility maximum was previously detected (R=Tb, Dy). These measurements allow us to claim that the unique susceptibility maxima may be interpreted in terms of antiferromagnetic ordering of the rare earths, and we discuss why the behavior of these compounds is different from that of Gd_2CuO_4. From the dependence of the Néel temperature on the single ion rare-earth magnetic moment, we suggest that a pseudodipolar magnetic interaction mechanism is the most plausible mechanism for this long-range magnetic ordering.

The synthesis of R_2CuO_4 polycrystalline samples was carried out from stoichiometric mixtures of the single oxides under high pressure and temperature conditions. Typical experimental conditions were 8 GPa, 1000–1200°C, produced in a belt-type apparatus. Extensive details on the synthesis procedure and the structural characterization may be obtained in refs. [12] and [13]. It is important to stress that all these oxides display some lattice superstructure in both the X-ray and electron diffraction patterns, which indicate the development of a distorted crystal structure. Actually, the tetragonal T′ structure may be considered only as an average structure, similar to the case of Gd_2CuO_4, where an in-plane oxygen disorder has also been detected [14].

The lower symmetry associated with these distortions has been proposed as the origin of the observed in-plane weak ferromagnetic component because an antisymmetric exchange interaction would be allowed in this case [13]. It is also important to stress that the oxygen stoichiometry of these oxides may not be greatly modified [15] and so their magnetic

properties depend very little on the modifications of the oxygen content [16].

The magnetic properties of the R_2CuO_4 cuprates have been investigated through measurements of the complex AC susceptibility. The high-temperature measurements were performed in a Lake Shore susceptometer with $h=10$ Oe and $\nu=111$ Hz. The low-temperature investigation was carried out in a SHE dilution refrigerator ($0.08\ K<T<16\ K$) with an LR 201 impedance bridge operating at a frequency of 16 Hz and $h=0.2$ Oe. The polycrystalline samples were sealed inside a plastic bag and tightly fit inside a section of a first order gradiometer secondary coil. The thermal contact with the metal support of the coils was increased with Apiezon N grease.

From the high temperature in-phase component of χ_{AC}, the effective magnetic moments of the rare-earth ions were deduced and found to be essentially coincident with those corresponding to the free ions, as has been reported in ref. [13]. Deviations from a Curie–Weiss law were observed, however, at low temperatures due to crystal field effects.

The low-temperature in-phase χ' and out-of-phase χ'' components of the complex susceptibility of the heavier R_2CuO_4 (R=Tb, Dy, Ho, Er, Tm) cuprates are displayed in figs. 1(a–e). As may be observed, these compounds display a single susceptibility maximum in both χ' and χ'' within the 1.5 K–10 K temperature range. Although the maxima for Tb_2CuO_4 and Dy_4CuO_4 appear to be broader than in previous measurements [13] performed under a higher AC excitation field (10 Oe versus 0.2 Oe), the locations of their χ' maxima are essentially coincident. The observation of a single maximum in χ' at low temperatures in all these compounds allows us to interpret these maxima as a signature of long-range magnetic ordering of the rare-earth ions.

We may now ask why the anomalous low temperature transition observed in the weak ferromagnetic compound Gd_2CuO_4 does not exist in the present new weak ferromagnetic compounds. As we have mentioned above, an essential difference has been observed in the high-temperature AC susceptibility of Tb_2CuO_4 and Gd_2CuO_4 [13]. While in the former a field-independent Curie–Weiss behavior is observed, indicating a freezing of the copper weak ferromagnetic components (also evidence can be found in ZFZ–FC hysteresis), in Gd_2CuO_4 a pure Curie–Weiss law is never observed and the AC susceptibility is field dependent down to the anomalous transition observed at $T_L\approx 20$ K.

A different dynamical magnetic behavior in both compounds may be understood as arising from quantitative changes in the thermally activated two-level relaxation of the weak ferromagnetic components. The energy barrier separating two equilibrium spin directions should depend on the degree of in-plane oxygen disorder in the T′ structure. Actually, the symmetry reduction associated with this oxygen displacement allows the appearance of an antisymmetric Dzyaloshinski–Moriya (D–M) interaction and hence of the weak ferromagnetic component. As shown by Bordet et al. [12] and Galez et al. [14], the displacement of in-plane oxygens from their high symmetry position in the CuO_2 planes seems to increase with the increase of the rare-earth atomic number. This is simply an effect of the chemical bond mismatch across the interface of the CuO_2 plane and the R_2O_2 fluorite layer [17], and thus we might expect an enhanced energy barrier in Tb_2CuO_4, as compared to Gd_2CuO_4. In this way, an essentially frozen weak ferromagnetic component may be obtained below $T\approx 200$ K in Tb_2CuO_4, while in Gd_2CuO_4 the relaxation extends to much lower temperatures.

On approaching the rare-earth magnetic ordering temperatures, further magnetic interactions (such as the R–Cu and R–R interactions) become involved in the determination of the magnetic ground state and hence a competition between different terms of the magnetic hamiltonian might be anticipated.

In the heavy rare-earth R_2CuO_4 cuprates, the D–M interaction should be much larger than the R–R interaction and hence the weak ferromagnetic component is not altered by the rare-earth long-range order. In Gd_2CuO_4, instead, a competition among different terms (D–M, R–R and R–Cu) is established and, finally, a collinear antiferromagnetic order of the Cu sublattice is stabilized by the Gd sublattice which has a collinear antiferromagnetic order (as demonstrated by Chattopadhyay et al. by means of neutron diffraction [10]).

The knowledge of the rare-earth lattice transition temperatures for the whole R_2CuO_4 series allows us to analyze which is the leading magnetic interaction driving this magnetic transition. In the $RBa_2Cu_3O_7$

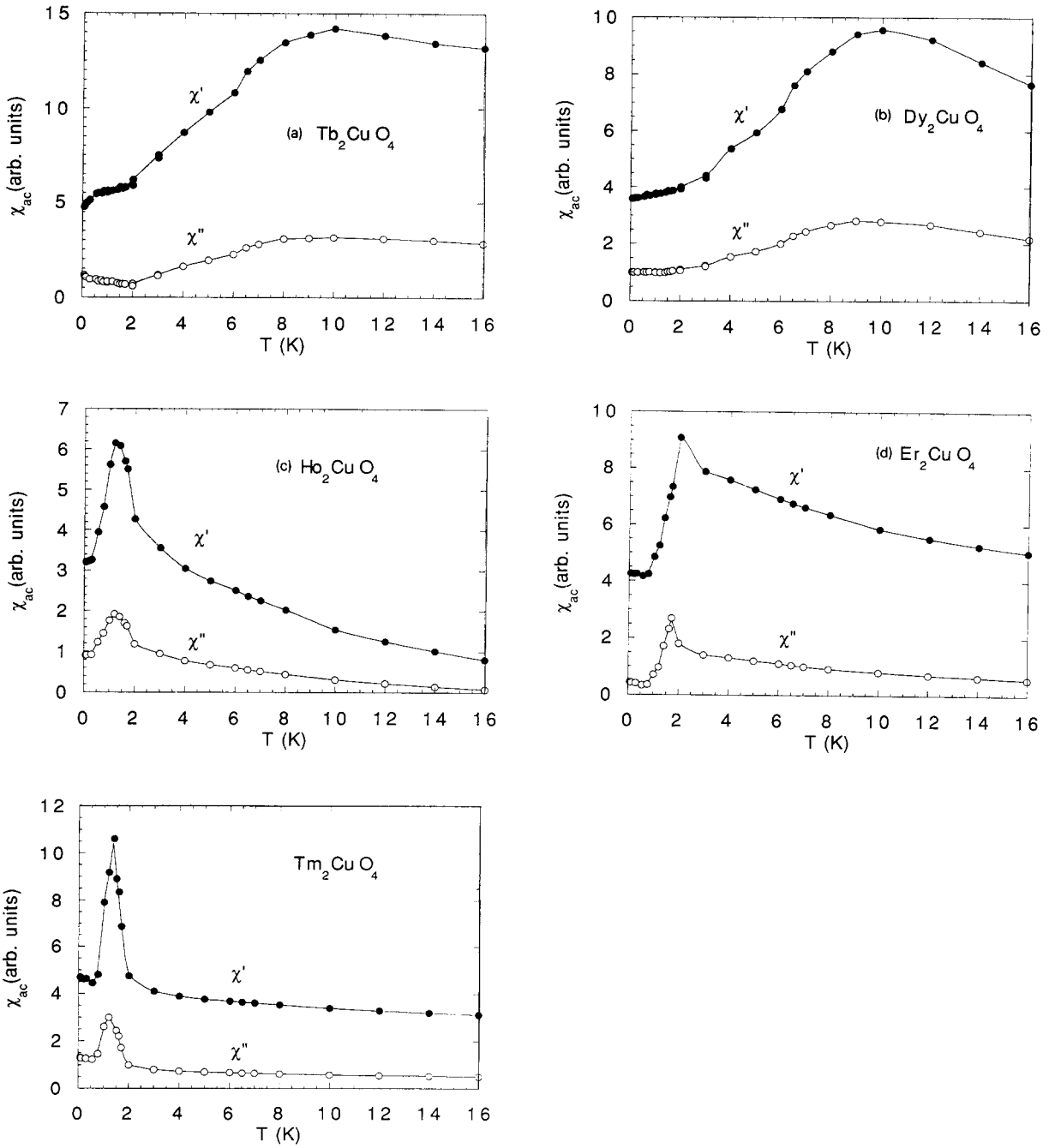

Fig. 1. Real and imaginary parts of the AC susceptibility, $\chi'(T)$ and $\chi''(T)$, measured at $\nu = 16$ Hz and $h = 0.2$ Oe, of R_2CuO_4: (a) Tb_2CuO_4, (b) Dy_2CuO_4, (c) Ho_2CuO_4, (d) Er_2CuO_4, (e) Tm_2CuO_4.

cuprates, for instance, it has been suggested [2] that the scaling of T_N with the de Gennes factor favors the superexchange or RKKY mechanism. However, evidence against the second possibility is obtained, for instance, when considering for $GdBa_2Cu_3O_{7-\delta}$ the independence of T_N on x, the parameter that

controls the change from the metallic ($x \leqslant 0.55$) to the insulating state ($x \geqslant 0.55$).

In fig. 2 we display the experimental values of T_N for the R_2CuO_4 oxides and compare them to theoretical curves, obtained by assuming either that T_N is proportional to $(g_J J)^2$ or to $(g_J - 1)^2 J(J+1)$, where g_J is the Landé g factor and J is the total angular momentum for the free rare-earth ions.

When comparing both dependences with the experimental points, a better agreement is observed for the line corresponding to a dipolar interaction between the rare-earth ions. This conclusion may be immediately drawn from the observation of an enhanced T_N in Dy_2CuO_4 and Tb_2CuO_4 with respect to Gd_2CuO_4. The main exceptions to this general rule are Sm_2CuO_4 and Ho_2CuO_4, where the true magnetic moments may be strongly modified by crystal field effects, so a complete analysis taking into account crystal field effects should be necessary to determinate the true magnetic moments. Our assessment indicates in any case that the classical dipolar interaction or a pseudodipolar term is the leading contribution in the establishment of antiferromagnetic order of the rare-earth ions in the T' R_2CuO_4 series.

It is very unlikely, however, that dipolar magnetic interactions may lead to transition temperatures as high as 10 K for the rare-earth sublattice, the most frequent range being an order of magnitude lower [18]. Similar rare-earth transition temperatures have also been observed in rare-earth nickelates such as Nd_2NiO_4 ($T_N = 11$ K) [19] where the disappearance of the Ni sublattice antiferromagnetism strongly decreases the Néel temperature of the rare-earth sublattice [20]. Unusually high transition temperatures have also been observed in non-superconducting $PrBa_2Cu_3O_7$ ($T = 17$ K) [21] where the Pr ions have a mixed valence behavior. Some authors have suggested that an unusual Suhl–Nakamura mechanism [22,23], involving the antiferromagnetic copper sublattice could explain this enhanced T_N. Within the context of this model an effective R–R interaction develops as a consequence of the R–Cu and Cu–Cu exchange interactions and hence the rare-earth Néel temperature might be determined mainly by J_{R-Cu}. This exchange interaction has been evaluated in the scope of the mean-field approximation in Gd_2CuO_4 and Tb_2CuO_4, and it turns out to be of the same order of magnitude as kT_N ($J_{R-Cu} \approx 1$ meV and $J_{R-Cu} \approx 0.2$ meV, respectively) [13,24]. We may then suggest that the copper sublattice antiferromagnetic order plays an essential role in the unusually high antiferromagnetic ordering temperatures of the rare-earth ions in the R_2CuO_4 series.

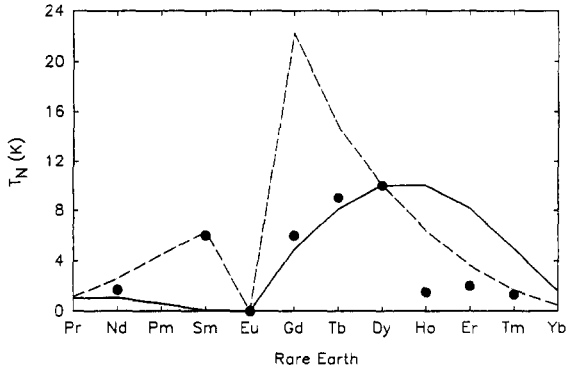

Fig. 2. Rare-earth antiferromagnetic transition temperatures vs. R for R_2CuO_4 oxides. The dashed line is the de Gennes scaling while the solid line is the full magnetic moment, dipolar-like, scaling. In both cases the scaling curves have been normalized to the transition temperature of Dy_2CuO_4.

It is straightforward to note that such a mechanism could be a natural explanation of the strong decrease of the rare-earth Néel temperature in $Nd_2NiO_{4+\delta}$ [20] and R_2BaCuO_5 [5] when the antiferromagnetic order in the Ni or Cu sublattice is either destroyed by hole doping or eliminated through substitution with non-magnetic Zn. Similarly, the validity of the mechanism implies that short range antiferromagnetic correlations persisting in the superconducting state [25] are still active in the effective R–R interaction in compounds such as $Sm_{2-x}Ce_xCuO_4$ or $Nd_{2-x}Ce_xCuO_4$, where T_N is decreased by 25% for $x = 0.15$ (7.5% doping). This decrease rate is higher than that predicted by simple dilution models based on superexchange interactions [26] and thus an alternative explanation involving the spin correlated state, observed in the superconducting state, might be invoked.

Further analysis of the Suhl–Nakamura mechanism appears necessary, however, to understand how this mechanism can lead to an effective R–R interaction having a pseudodipolar character.

In conclusion, we have shown that the rare-earth magnetic ordering in the R_2CuO_4 series does not fol-

low a simple superexchange mechanism, which would lead to a scaling of T_N with the de Gennes factor. A scaling of T_N with the full rare-earth magnetic moment seems more appropriate, thus suggesting a pseudodipolar coupling mechanism. It is very unlikely, however, that a simple classical dipolar interaction could account for the large values of the observed Néel temperatures. We suggest that other mechanisms, such as the Suhl–Nakamura interaction, should be considered to explain the puzzling behavior of the rare-earth sublattice in this series of cuprates.

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