



ELSEVIER

Physica B 320 (2002) 34–36

PHYSICA B

www.elsevier.com/locate/physb

Double exchange model for $\text{RuSr}_2(\text{Eu}, \text{Gd})\text{Cu}_2\text{O}_8$

H. Aliaga, A.A. Aligia*

Comisión Nacional de Energía Atómica, Centro Atómico Bariloche and Instituto Balseiro, 8400 S.C. de Bariloche, Argentina

Abstract

We propose a double exchange model to describe the RuO_2 planes of $\text{RuSr}_2(\text{Eu}, \text{Gd})\text{Cu}_2\text{O}_8$. The Ru^{+5} ions are described by localized spins, and additional electrons provided by the superconducting CuO_2 planes are coupled ferromagnetically to them by Hund rules coupling. We calculate the spin structure factor, magnetic susceptibility and magnetization as a function of magnetic field and temperature, using a Monte Carlo algorithm in which the Ru^{+5} spins are treated as classical. Several experiments which seemed in contradiction with one another are explained by the theory. © 2002 Elsevier Science B.V. All rights reserved.

PACS: 75.10.-b; 75.40.Mg; 74.72.-h

Keywords: Ruthenates; Double exchange; Magnetic susceptibility; Spin structure factor

$\text{RuSr}_2(\text{Eu}, \text{Gd})\text{Cu}_2\text{O}_8$ is a very interesting material because of the coexistence of magnetic order and superconductivity below $T_S \sim 45$ K [1,2]. The system orders magnetically at $T_M = 133$ K, but the type of magnetic order remains controversial. The first measurements in Gd samples indicated ferromagnetic (FM) ordering of the Ru moments [1], but neutron diffraction experiments found superlattice reflections consistent with an antiferromagnetic (AF) order with nearest-neighbor spins antiparallel in all three directions [3]. The Ru contribution to the magnetic susceptibility at temperature $T > T_M$ can be very well described by $\chi = C/(T - \Theta)$, with $\Theta = 100 \pm 3$ K [4]. The fact that $\Theta > 0$ seems difficult to reconcile with AF order at low temperatures.

From previous intensive research on the similar system $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ [5], it seems clear that each

of the superconducting CuO_2 planes has doping slightly < 0.1 holes per Cu ion in order to lead to the observed superconducting critical temperature T_S . Since Eu or Gd are expected to be in the oxidation state +3, assuming as usual that the inactive SrO layers are neutral, charge balance imposes that the RuO_2 planes have an electron doping $n \lesssim 0.2$ per Ru ion [5]. Neglecting covalency with O atoms, this means that there are $1 - n$ Ru^{+5} and n Ru^{+4} ions per unit cell. The superconducting CuO_2 planes and the magnetic RuO_2 planes can be regarded as separate entities as a first approximation. This is supported by symmetry considerations and band-structure calculations [6]. We propose that the electronic structure of the RuO_2 planes can be described by an effective double exchange model, with one localized spin at each site representing the Ru^{+5} ions, and additional n itinerant electrons per Ru coupled ferromagnetically with the localized spins.

*Corresponding author. Fax: +54-2944-445299.

E-mail address: aligia@cab.cnea.gov.ar (A.A. Aligia).

The model is the two-dimensional (2D) version of one widely used in manganites:

$$H = - \sum_{\langle ij \rangle \sigma} t(c_{i\sigma}^\dagger c_{j\sigma} + \text{h.c.}) - J_H \sum_i \mathbf{s}_i \cdot \mathbf{S}_i + K \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j. \quad (1)$$

Here \mathbf{S}_i is the localized spin operator representing the Ru^{+5} ion at site i , $c_{i\sigma}^\dagger$ is the operator creating an itinerant electron of spin σ at this site, and $\mathbf{s}_i = \sum_{\alpha\beta} c_{i\alpha}^\dagger \boldsymbol{\sigma}_{\alpha\beta} c_{i\beta}$ gives the spin of this electron. For simplicity we take the Hund coupling, $J_H \rightarrow +\infty$.

The model is solved using a classical Monte Carlo (MC) procedure for the localized spins, in conjunction with exact diagonalization of the conduction electron system [7]. The localized spins are taken to be classical and of magnitude one. The conduction electrons are assumed to occupy a single orbital, and from the condition $J_H \rightarrow +\infty$ only one spin projection is possible at each site. Then, for each configuration of localized spins, the effective hopping t_{ij} of itinerant electrons between two sites i and j is affected by the factor $\langle \uparrow_i | \uparrow_j \rangle$, where $|\uparrow_j\rangle$ is the state of a spin $\frac{1}{2}$ pointing in the direction of the localized spin j . Then,

$$t_{ij} = t \left(\cos \frac{\theta_i}{2} \cos \frac{\theta_j}{2} + e^{-i(\phi_i - \phi_j)} \sin \frac{\theta_i}{2} \sin \frac{\theta_j}{2} \right), \quad (2)$$

where θ_i and ϕ_i are the polar angles of spin \mathbf{S}_i . The resulting electronic energy levels are then filled by the available number of electrons in the canonical ensemble. The MC simulation proceeds from the partition function with classical spins. The general features of this model as a function of K/t and doping have been studied before in the context of the manganites [8,9]. The calculations reported here are in a square of $N = 64$ atoms with periodic boundary conditions and six mobile electrons ($n = \frac{6}{64} \cong 0.1$). We have taken $K/t = 0.06$. We expect that the order of magnitude of t is near 0.25 eV, and then $K \sim 180$ K, near the observed T_M .

We have calculated the spin structure factor defined as $S(\mathbf{q}) = \sum_{i,j} \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle e^{i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)} / N^2$ for all nonequivalent values of \mathbf{q} in our 8×8 cell. In Fig. 1 we represent the two superlattice peaks of highest intensity as a function of temperature. Clearly, the peak at $\mathbf{q} = (\pi, \pi)$ is the dominant one, in agreement with neutron experiments [3]. An

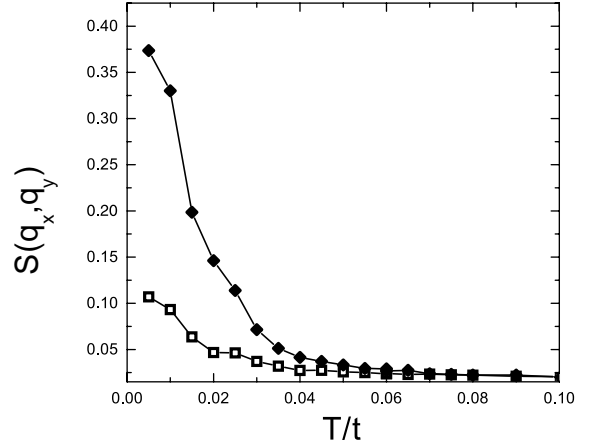


Fig. 1. Spin structure factor as a function of temperature for two wave vectors \mathbf{q} : $\mathbf{q} = (\pi, \pi)$ (diamonds), $\mathbf{q} = (0, 0)$ (squares).

important fact is that at $T = 0$ the amplitude of $S(\mathbf{q})$ is smaller than one. This is consistent with experiments because the magnetic moment deduced from magnetic measurements [4] is larger than that deduced from the amplitude of the superlattice reflections [3]. This reduction in our simulations is due to the fact that, in general, there are small ferromagnetic islands (magnetic polarons) around the mobile electrons. Since these islands are not entirely compensated in our small systems and that their kinetics is slow at low temperatures, they give rise to a small contribution to $S(0, 0)$ at small temperatures, as displayed in Fig. 1. Since we are dealing with a strictly 2D system in our simulations (instead of 3D in the real system), and a continuous symmetry cannot be broken in 2D, Fig. 1 does not show a phase transition to the paramagnetic phase.

The mobility of the ferromagnetic polarons makes it easy to orient them under small applied magnetic fields. Then, as observed experimentally [4] the magnetization increases abruptly with small magnetic fields (~ 5 T) compared with the Neel temperature $T_M \cong 133$ K. This fact is unusual for an ordinary AF system. The magnetization curve of the localized spins in our system is shown in Fig. 2. In spite of the difficulties with the sluggish statistics of the ferromagnetic polarons mentioned above, one can see an abrupt increase at low fields, due to the alignment of these polarons, followed by a linear increase with a smaller slope, in agreement with experiment [4].

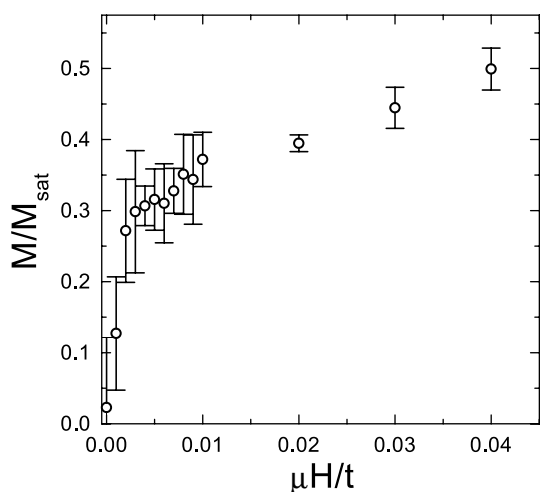


Fig. 2. Magnetization as a function of applied magnetic field at temperature $T = 0.02$ K. M_{sat} is the saturation magnetization.

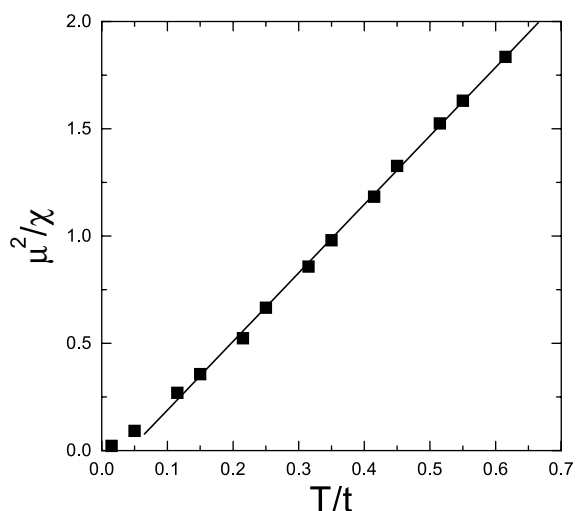


Fig. 3. Inverse magnetic susceptibility as a function of temperature. The straight line is a fit using $\chi^{-1} = (T - \Theta)/C$.

Another unexpected result for an AF system is the fact that the inverse magnetic susceptibility χ^{-1} at high temperatures extrapolates to a positive temperature for $\chi^{-1} = 0$. As shown in Fig. 3, our model also reproduces this result. The constant Θ obtained fitting $\chi = C/(T - \Theta)$ is $\Theta = (0.041 \pm 0.004)t$. This is a subtle issue which is related with the way in which the ferromagnetic polarons are reduced as the temperature is increased, and their effect on the magnetic correlations on their

neighborhood. A mean field picture in which the kinetic energy of the carriers is neglected seems unable to explain this result even qualitatively. The resulting value of the slope gives $C = (0.314 \pm 0.006)\mu^2$, where μ is the magnetic moment of the localized spins. This value is near to the classical value $\mu^2/3$ for noninteracting spins. The value of C in the real system depends on the spin of the configurations $4d^5$ and $4d^6$ of Ru^{+5} and Ru^{+4} [4].

To conclude, we have proposed a double exchange model for $\text{RuSr}_2(\text{Eu}, \text{Gd})\text{Cu}_2\text{O}_8$ which is able to explain the main unusual features of these systems. Previous studies of the model [8,9] indicate that for small densities there is phase separation in the model. However, in the real system, particularly due to the small number of carriers [5], the effect of long-range Coulomb repulsions, not included in the model should be present and avoid macroscopic phase separation [10], while phase separation in a microscopic scale does not affect our conclusions.

We thank B. Alascio and B. Normand for useful discussions. We are partially supported by CONICET. This work was sponsored by PICT 03-06343 of ANPCyT and PIP 4952/96 of CONICET.

References

- [1] C. Bernhard, et al., Phys. Rev. B 59 (1999) 14099.
- [2] A. Fainstein, et al., Phys. Rev. B 60 (1999) R12597.
- [3] J.W. Lynn, et al., Phys. Rev. B 61 (2000) R14964.
- [4] A. Butera, et al., Phys. Rev. B 63 (2001) 054442.
- [5] A.A. Aligia, J. Garcés, Phys. Rev. B 49 (1994) 524, and references therein; J.J. Neumeier, H.A. Zimmermann, Phys. Rev. B 47 (1993) 8385; A.A. Aligia, E.R. Gagliano, P. Vairus, Phys. Rev. B 52 (1995) 13 601.
- [6] W.E. Pickett, R. Weht, A.B. Shick, Phys. Rev. Lett. 83 (1999) 3713.
- [7] S. Yunoki, A. Moreo, Phys. Rev. B 58 (1998) 6403.
- [8] H. Aliaga, et al., Phys. Rev. B 64 (2001) 024422.
- [9] S. Yunoki, A. Moreo, E. Dagotto, Phys. Rev. Lett. 81 (1998) 5612; J.L. Alonso, et al., Phys. Rev. B 63 (2001) 054411; J.L. Alonso, et al., Nucl. Phys. B 596 (2001) 587.
- [10] A.A. Aligia, J. Garcés, J.P. Abriata, Physica C 221 (1994) 109; J. Lorenzana, C. Castellani, C. Di Castro, cond-mat/0106608.