INFLUENCE OF THE RING EXCHANGE ON THE MAGNETIC STRUCTURE OF MANGANITES

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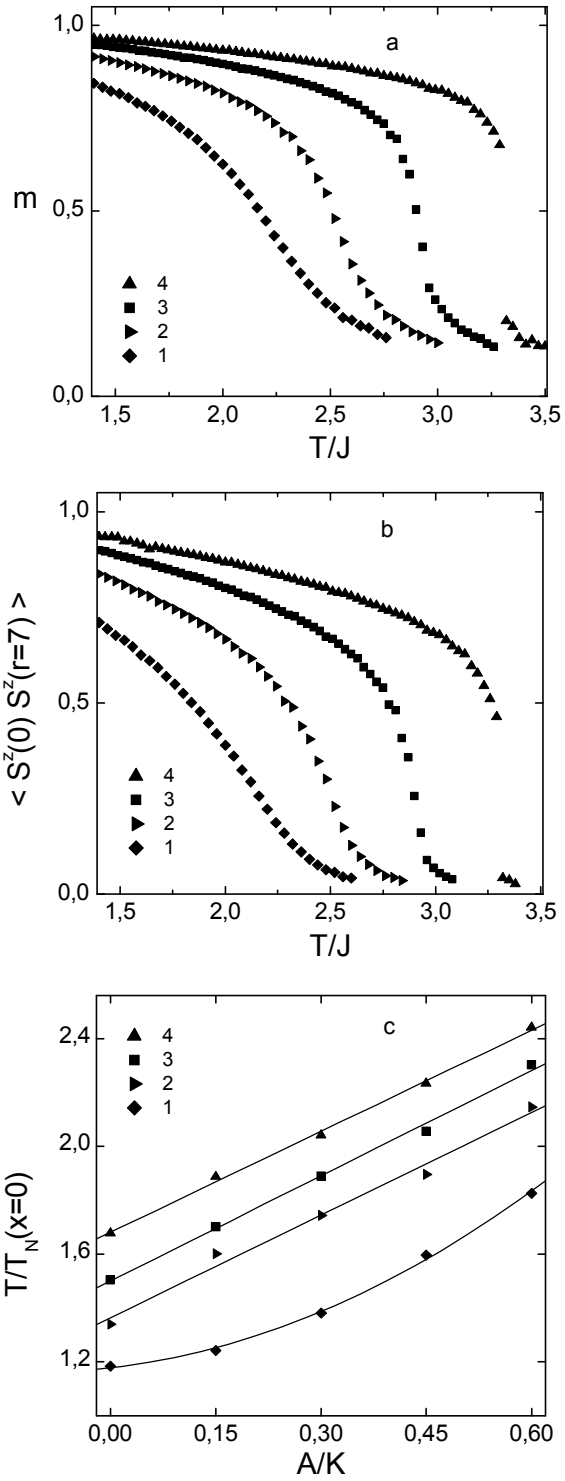
The manganites $Re_{1-x}A_xMnO_3$ (where Re=La, Pr, Nd etc. and A=Ca, Sr, Ba etc.) have been at the centre of attention in condensed matter for the last two decades because of the variety and novelty of electronic phenomena in them and the possibility of applications in spintronic . The initial interest was sparked by the discovery of colossal magnetoresistance . Subsequent work has been shown a variety of magnetic phases, phase transitions and phenomena depending on the doping x, temperature , and ionic species Re and A as well as external perturbations. The change of the kind of magnetic ordering from antiferromagnet to ferromagnet is observed at x=0.08 -0.09 in the $La_{1-x}A_xMnO_3$ (A=Ca, Sr)[1] . Ferromagnetic dielectric becomes a ferromagnetic metal with colossal magnetoresistance near T_c for $x > 0.2$. The early simulations explored the competition between (double exchange induced) ferromagnetism and anti- ferromagnetic (super) exchange .

However Millis [2] has been shown that the "double exchange" model disagreed with several experimental consequences by an order of magnitude or more. The discrepancy was resolved by including polaron effects due to a very strong electron- phonon coupling coming from a Jahn-Teller splitting of the Mn^{3+} ion. The measurements of specific heat indicates also an anomalous softening of the lattice observing from T^3 – term in the specific heat in a fairly wide x region ($0.1 < x < 0.3$) [3] , but is not apparently relevant to the crystal (rhombohedral- orthorhombic) transition. This anomalous is ascribed to the subsisting dynamic Jahn- Teller distortion down to low or zero temperature. The strong coupling with lattice may induce major ring exchange (A) the value of which is proportional to $AS^4 \sim \frac{1}{Ma^2}(\frac{\nabla J}{\theta_D})^2$, where θ_D –Debey temperature (θ_D=320 K- 450 K) [4], $M \sim 10^{-22}$ g, $a \sim 4 \cdot 10^{-10}$ m, S is changed from S=2 to 3/2 versus doping. Ring exchange resulted from the exchange by virtual phonons is A~ 0.1 meV for ($\nabla J / \theta_D$) ~0.03 . Relevant contribution to the ring exchange can be given by I_{s-d} interaction moving electron with localized one. Using small parameter W/E_g (W- bandwidth, E_g – gap) Heisenberg exchange is $J \sim I_{s-d}^2 W^2/E_g^3$, ring exchange is $A \sim I_{s-d}^4 W^4/E_g^7$ and ratio $J/A \sim I_{s-d}^2 W^2/E_g^4$. At doping the gap in the single electron excitation spectrum is decreased and ring exchange value may be compared with the value of bilinear exchange. So the $e_g(\sigma^*)$ band is about 1 eV wide, and the $t_{2g} – e_g$ separation is about 1.5 eV [5], I_{s-d} is (0.5 – 1) eV.

Another problem in manganites the isotherms H/M never intercept the M^2 axis, even at temperatures much lower than the temperature of the minimum in $\partial M/\partial T$. The extrapolation from low field, where the approximated is justified, neither cuts the M^2 axis. This makes it impossible to define the order parameter. Moreover, the isotherms never reach the origin, they intercept the H/M axis at a finite value, giving a fixed susceptibility [6]. Following this criterion the magnetic transition out of the range $0.275 < x < 0.43$ is not a true phase transition [6] and unlike in common continuous magnetic phase transition.

Magnetic properties we consider in the Heisenberg model with random exchange interactions .

$$H = -\sum_{i,j}^{L} J_{i,j} S_i S_j - \sum_{i,j}^{L} A_{i,j,k,l} (S_i S_j)(S_k S_l)$$



where J_{ij} – exchange between the nearest neighbors , $A_{i,j,k,l}$ – ring exchange, $\sigma_i = \pm 1$, L is size of cube. Manganites have perovskite –type crystal structure where the rare –earth ions are situated in the center of cube and manganese ions are in the cube vertexes . If at the left and at the right from the cube face the various valences ions are found then exchange interactions between the manganese ions are equal to $K > 0$ and $A > 0$. The magnetic structure of $LaMnO_3$ consists of the ferromagnetic ordering spins in plane ($J_{xy} > 0$) which ordered antiferromagnetically ($J_z < 0$). The ratio of the exchange parameters have been determined from the spin dynamics as a result of a fit of the dispersion using a Heisenberg model with four first in plane neighbor couplings (J) (ferromagnetic) to J_{AF} - antiferromagnetic along [001] $J_{AF}/J \approx -0.7$ and an effective single-ion anisotropy $D/J_1=0.2$ for $LaMnO_3$ [7] . Monte Carlo procedure is performed on cubic lattice with 14 x 14 x 14, 18 x 18 x 18 sites and periodical boundary conditions. We used 10 000 – 20 000 MC/spin. Substitution of ions are realized in the lattice by using random numbers.

Neel and Cure temperatures are determined from the calculation of sublattice magnetization (simple magnetization for FM) , spin –spin correlation function that are shown in Fig.1 .

Fig.1 Magnetization (m) (a) , spin-spin correlation function $<S^z(0)S^z(r=7)>$ (b) versus temperature for x=0.3, A/K=0.15 (1), 0.3 (2), 0.45 (3), 0.6 (4). Cure temperature normalized to Neel temperature of A-type AFM versus the ring exchange value for x=0.1 (1), 0.2(2), 0.3(3), 0.5(4) (c) .

At increasing ring exchange value the type of magnetic phase transition varies from continuous to discontinuous . The critical value A_c dependents on concentration and agree with analytical results for uniform system $A_{c1} \approx 0.5$ [8] at x=0.5 . T_c (A) dependence fits well by linear $(T_c(A) - T_c(x,A=0)) / T_N(x=0) = 0.6$ A/K at $x \geq 0.2$.

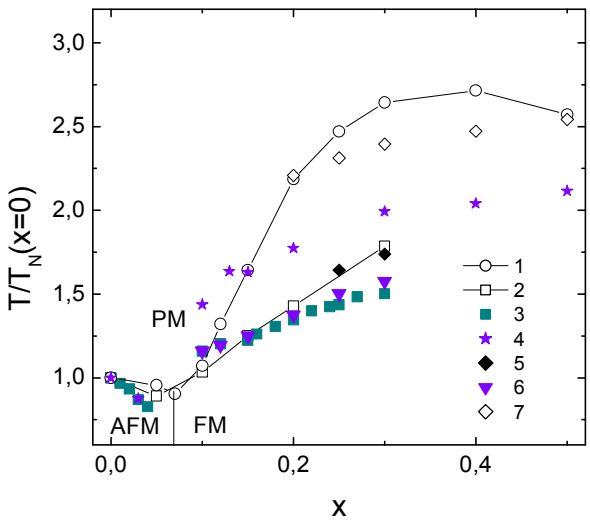


Fig.2 Phase diagram of antiferromagnetic (AFM) , ferromagnetic (FM) and paramagnetic (PM) states in plane $T_{c(N)}$ /$T_{c(N)}$(x=0) – A=Sr, Ca ions concentration (x). Experimental data A=Sr (1)[1], Ca (2)[7], MC results A=0 K/J=2, (3), K/J=3 (4), K/J=2 , A=0.15 (5), A=0.05 (6), K/J=3, A=0.3 (7).

The critical concentrations and ferromagnetic value exchanges (K) corresponding to state transition from AFM to FM are equal to x_c =0.08 ± 0.01, K ≈ 2 J for La_{1-x} Ca_xMnO_3 and x_c =0.06 ± 0.01, K ≈ 3 J for La_{1-x} Sr_xMnO_3. Neel and Cure temperatures are plotted in Fig.2 for K/J=2 , 3. Satisfactory agreement with experimental data [1] are observed at x < 0.2 for La_{1-x} Sr_xMnO_3 and x < 0.25 for La_{1-x} Ca_xMnO_3 without using ring exchange . For larger doping concentrations the best agreement with experimental data is achieved by allowing for ring exchange. Neel and Cure temperatures La_{1-x} A_xMnO_3 (A=Ca, Sr) are satisfactory described by model of modification of exchange on the boundary of surface La – Ca (Sr) with ring exhange A/K=0.15 for Ca and A/K=0.3 for Sr.

Change of conductivity type from semiconductor to metallic is observed at x_{c2}^{ex} =0.22 [7] for doping La by Ca and at x_{c2}^{ex} =0.17 [1] for doping La by Sr . For x > x_{c2} the main contribution to ring exchange value is attributed to electron hopping. Our ring exchange values is essentially less as compared to second critical value A_{c2} ~ (3- 4) S^{-2} ≈1.2 at which is formed a quantum quadruple state [8].

Summing up, the substitution of the rare- earth ion by the bivalent ion in the La_{1-x} A_xMnO_3 (A=Ca, Sr) causes formation of ferromagnetic exchange between manganese ions having value K/J ≈ 2, and ring exchange A/K=0.15 for doping La by Ca and K/J=3, A/K=0.3 for doping La by Sr. Formation of the ring exchange arises from metal – dielectric transition .

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