

Problem 1. Exchange Consider single-particle wavefunctions on two neighboring identical atoms ψ_A, ψ_B , which may be assumed real. These are to be used as the basis for a two-electron state. Show that the charge density in a singlet (triplet) state made out of the two orbitals is given by

$$\rho(r) = |\psi_A(r)|^2 + |\psi_B(r)|^2 \pm 2 \langle \psi_A | \psi_B \rangle \psi_A(r) \psi_B(r).$$

Explain why the singlet state will usually be lower in energy.

Solution. The charge density $\rho(r)$ is equivalent to the electron density, which can be found by

$$\rho(r) = \langle \psi | \hat{\rho}(r) | \psi \rangle,$$

where

$$\hat{\rho}(r) = \sum_{i=1}^N \delta(r - r_i)$$

is the density operator [1]. For a two-electron state, the wavefunction is given by (6.9) of the lecture notes,

$$\psi(r_1, r_2) = \frac{\psi_A(r_1)\psi_B(r_2) \pm \psi_A(r_2)\psi_B(r_1)}{\sqrt{2}},$$

where the $+$ ($-$) is for the spin singlet (triplet) state. Since ψ_A and ψ_B are real, $\psi^* = \psi$. Then the density is

$$\begin{aligned} \rho(r) &= \langle \psi | [\delta(r - r_1) + \delta(r - r_2)] | \psi \rangle \\ &= \iint dr_1 dr_2 \psi(r_1, r_2) [\delta(r - r_1) + \delta(r - r_2)] \psi(r_1, r_2) \\ &= \int dr_2 \psi(r, r_2) \psi(r, r_2) + \int dr_1 \psi(r_1, r) \psi(r_1, r) \\ &= \frac{1}{2} \int dr_2 [\psi_A(r) \psi_B(r_2) \pm \psi_A(r_2) \psi_B(r)]^2 + \frac{1}{2} \int dr_1 [\psi_A(r_1) \psi_B(r) \pm \psi_A(r) \psi_B(r_1)]^2 \\ &= \frac{1}{2} \int dr_2 \left[|\psi_A(r)|^2 |\psi_B(r_2)|^2 \pm 2\psi_A(r) \psi_B(r) \psi_A(r_2) \psi_B(r_2) + |\psi_A(r_2)|^2 |\psi_B(r)|^2 \right] \\ &\quad + \frac{1}{2} \int dr_1 \left[|\psi_A(r_1)|^2 |\psi_B(r)|^2 \pm \psi_A(r) \psi_B(r) \psi_A(r_1) \psi_B(r_1) + |\psi_A(r)|^2 |\psi_B(r_1)|^2 \right] \\ &= \frac{1}{2} \left(|\psi_A(r)|^2 \int dr_2 |\psi_B(r_2)|^2 \pm 2\psi_A(r) \psi_B(r) \int dr_2 \psi_A(r_2) \psi_B(r_2) + |\psi_B(r)|^2 \int dr_2 |\psi_A(r_2)|^2 \right. \\ &\quad \left. + |\psi_B(r)|^2 \int dr_1 |\psi_A(r_1)|^2 \pm 2\psi_A(r) \psi_B(r) \int dr_1 \psi_A(r_1) \psi_B(r_1) + |\psi_A(r)|^2 \int dr_1 |\psi_B(r_1)|^2 \right) \\ &= \frac{1}{2} \left(|\psi_A(r)|^2 \pm 2\psi_A(r) \psi_B(r) \langle \psi_A | \psi_B \rangle + |\psi_B(r)|^2 + |\psi_B(r)|^2 \pm 2\psi_A(r) \psi_B(r) \langle \psi_A | \psi_B \rangle + |\psi_A(r)|^2 \right) \\ &= |\psi_A(r)|^2 + |\psi_B(r)|^2 \pm 2 \langle \psi_A | \psi_B \rangle \psi_A(r) \psi_B(r), \end{aligned}$$

where we have assumed both ψ_A and ψ_B are properly normalized. □

The singlet state will usually be lower in energy because ψ_A and ψ_B are not orthogonal, as mentioned on p. 110 of the lecture notes. The singlet state has a higher charge density near $r = 0$, as shown in Fig. 1. Since the electrons are therefore more likely to be located between the two atoms, they tend to be less excited, and so less energetic.

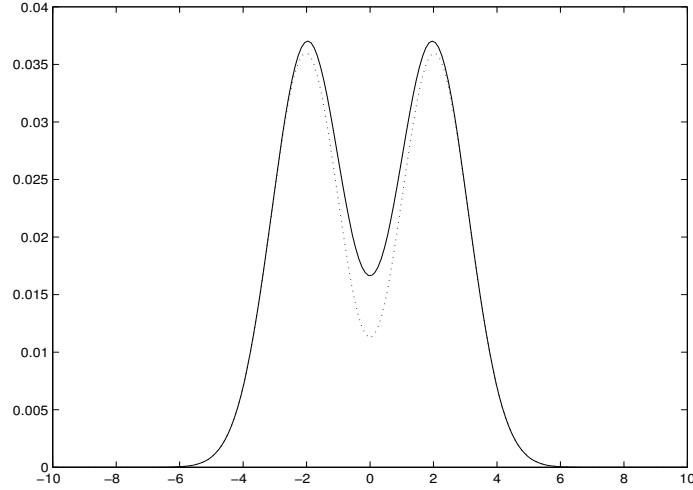


Figure 1: [Figure 6.2 of the lecture notes] Charge density for the wavefunctions in a singlet state (solid line) and a triplet state (dotted line).

Problem 2. One-dimensional spin waves Assume a one-dimensional chain of spins, precessing according to Eq. (6.30). By considering two neighbors of the n th spin, as in Fig. 6.8, each at relative angles θ , show that the rate of precession according to Eq. (6.30) is

$$\omega = \frac{4JS}{\hbar} [1 - \cos(\theta)].$$

Hence show that for a spin wave of wavevector q , the dispersion is

$$\hbar\omega = 4JS[1 - \cos(qa)].$$

Solution. Equation (6.30) in the lecture notes is

$$\delta\dot{\mathbf{S}}_n = \frac{2JS}{\hbar} \sum_{j=\text{n.n. of } n} (\delta\mathbf{S}_n - \delta\mathbf{S}_j) \times \hat{\mathbf{z}},$$

where $\hat{\mathbf{S}}_n$ is a vector in the xy plane and $\hat{\mathbf{S}}_n = S\hat{\mathbf{z}} + \delta\mathbf{S}_n$ by (6.29). For a one-dimensional chain, the only nearest neighbors are $j \in \{n-1, n+1\}$. Thus

$$\delta\dot{\mathbf{S}}_n = \frac{2JS}{\hbar} (2\delta\mathbf{S}_n - \delta\mathbf{S}_{n-1} - \delta\mathbf{S}_{n+1}) \times \hat{\mathbf{z}}.$$

Splitting into one equation for each component, we have

$$\delta\dot{S}_n^x = -\frac{2JS}{\hbar} (2\delta S_n^y - \delta S_{n-1}^y - \delta S_{n+1}^y), \quad \delta\dot{S}_n^y = \frac{2JS}{\hbar} (2\delta S_n^x - \delta S_{n-1}^x - \delta S_{n+1}^x), \quad \delta\dot{S}_n^z = 0, \quad (1)$$

since $\hat{\mathbf{x}} \times \hat{\mathbf{z}} = -\hat{\mathbf{y}}$ and $\hat{\mathbf{y}} \times \hat{\mathbf{z}} = \hat{\mathbf{x}}$. By analogy with phonon waves, we make the ansatz that the solutions have the form [2, p. 332]

$$\delta S_n^x = ue^{i(j\theta - \omega t)}, \quad \delta S_n^y = ve^{i(j\theta - \omega t)}, \quad \delta S_n^z = 0, \quad (2)$$

where u and v are constants and j is an integer. Making this substitution, Eq. (1) becomes

$$\begin{aligned} -i\omega ue^{i(n\theta - \omega t)} &= -\frac{2JS}{\hbar} \left(2e^{i(n\theta - \omega t)} - e^{i[(n-1)\theta - \omega t]} - e^{i[(n+1)\theta - \omega t]} \right) v, \\ -i\omega ve^{i(n\theta - \omega t)} &= \frac{2JS}{\hbar} \left(2e^{i(n\theta - \omega t)} - e^{i[(n-1)\theta - \omega t]} - e^{i[(n+1)\theta - \omega t]} \right) u, \end{aligned}$$

or

$$\begin{aligned} -i\omega u &= -\frac{2JS}{\hbar} \left[2 - (e^{-i\theta} - e^{i\theta}) \right] v = -\frac{4JS}{\hbar} [1 - \cos(\theta)] v, \\ -i\omega v &= \frac{2JS}{\hbar} \left[2 - (e^{-i\theta} - e^{i\theta}) \right] u = \frac{4JS}{\hbar} [1 - \cos(\theta)] u. \end{aligned}$$

This system can be solved using the matrix equation

$$0 = \begin{vmatrix} -i\omega & 4JS[1 - \cos(\theta)]/\hbar \\ -4JS[1 - \cos(\theta)]/\hbar & -i\omega \end{vmatrix} = -\omega^2 + \frac{16J^2S^2}{\hbar^2} [1 - \cos(\theta)]^2,$$

which has the solutions

$$\omega = \frac{4JS}{\hbar} [1 - \cos(\theta)]$$

as we wanted to show [2, p. 332]. □

For a spin wave of wavevector q , the phonon waves have the form $e^{i\mathbf{q}\cdot\mathbf{r}} e^{i\omega t}$ by (5.5) of the lecture notes. Assuming the distance between adjacent spins is a , then $\mathbf{q} \cdot \mathbf{r} \rightarrow qa$ in one dimension and $\theta = qa$ by inspection of Eq. (2). So the dispersion is

$$\hbar\omega = 4JS[1 - \cos(qa)]$$

as desired.

Problem 3. Colossal magnetoresistance In a crystal like that shown in Fig. 6.10 the magnetism arises from a mechanism called double exchange, which is a version of itinerant exchange but involving two types of d bands. The prototype compound is $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$, where the valence of La is 3+ and Sr is 2+. This is a cubic (perovskite) crystal structure where the Mn ions are nominally equidistant from six oxygen neighbors in three Cartesian directions.

3(a) Explain why the valence of Mn in the compound $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ is expected to be between 3+ and 4+ and that the occupancy of the d levels is expected to be $4 - x$ electrons per Mn ion.

Solution. The relevant electron configurations are [3, 4, 5, 6]

$$\text{La} : 5d^1 6s^2, \quad \text{Sr} : 5s^2, \quad \text{O} : 2s^2 2p^4, \quad \text{Mn} : 3d^5 4s^2.$$

The atoms (excluding Mn) have valence

$$\text{La} : 3+, \quad \text{Sr} : 2+, \quad \text{O} : 2-.$$

This is because La and Sr have few valence electrons and will tend to donate them, while O will fill its $2p$ shell upon receiving two more electrons.

The two extremes are LaMnO_3 ($x = 0$) and SrMnO_3 ($x = 1$). For LaMnO_3 , O_3 has valence 6− and La 3+, so Mn must have valence 3+ to form a neutral compound. For LaMnO_3 , O_3 again has valence 6− but Sr has valence 2+, so in this case Mn must have valence 4+. Thus for any $x \in [0, 1]$, the valence of Mn must be between 3+ and 4+. The expected valence is $(3 + x)+$.

When Mn ionizes, it loses its two $4s$ electrons first [7]. For LaMnO_3 ($x = 0$), it also loses one $3d$ electron, leaving it with $3d^4$, or $4 = 4 - x$ electrons in the d levels. For SrMnO_3 ($x = 1$), Mn loses two $3d$ electrons, leaving it with $3d^3$, or $3 = 4 - x$ electrons in the d levels. Therefore Mn loses an expected $1 + x$ electrons from its $3d$ levels, leaving it with $5 - (1 + x) = 4 - x$ remaining.

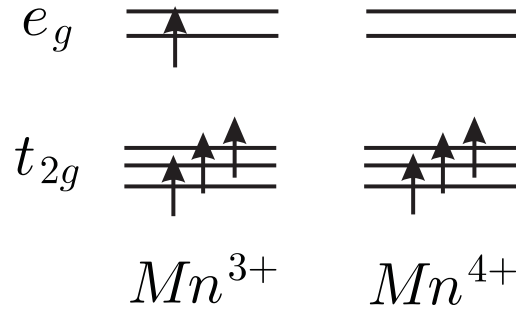


Figure 2: [Figure 6.12 of the lecture notes]

3(b) The degeneracy of the 5 d levels in the free ion is split by the cubic environment into a low energy threefold degenerate subset (whose notation is t_{2g}) and a higher energy doubly degenerate orbital set (e_g). Explain why the spin configurations of these levels for the Mn^{3+} and Mn^{4+} ions are expected to be as shown in Fig. 6.12.

Solution. Figure 6.12 in the lecture notes is reproduced in Fig. 2. The lower-energy t_{2g} levels will be filled first according to the aufbau principle. By Hund's rule as mentioned on p. 107, all 5 d levels will receive a single electron before any d level receives a second electron. This allows all of the electrons to have the same spin, which maximizes the total spin of the system. Since this is a symmetric spin state, the electrons stay further apart due to Coulomb repulsion and therefore have lower energy than they would if they were not all aligned [2, pp. 307–308]. This explains why Mn^{4+} has one electron in each of the three t_{2g} levels, all with the same spin, and why the fourth electron of Mn^{3+} is added to the e_g level, again with all electrons having the same spin.

3(c) The lowest three electron states can be regarded as forming a classical spin $S = 3/2$ which has negligible hopping from site to site, whereas the highest state is potentially itinerant. Now consider two neighboring sites i, j in the solid, each having the same “core” spin S , and sharing a single itinerant e_g electron, that has a tight-binding matrix element

$$t = \langle \phi_{e_g}(\mathbf{r} - \mathbf{R}_i) | H | \phi_{e_g}(\mathbf{r} - \mathbf{R}_j) \rangle$$

for hopping from site to site.

In second-quantized notation, the full Hamiltonian can be written as

$$H = t \sum_{i,j=\text{n.n.}} \sum_{\sigma} \hat{c}_{i,\sigma}^{\dagger} \hat{c}_{j,\sigma} + U \sum_i \hat{n}_{i,\sigma} \hat{n}_{i,-\sigma} - J \sum_i \hat{\mathbf{s}}_i \cdot \mathbf{S}_i + J_x \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j.$$

Explain the origin of the terms

$$H_{\text{int}} = J \sum_i \hat{\mathbf{s}}_i \cdot \mathbf{S}_i + J_x \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j$$

in the total Hamiltonian ($\hat{\mathbf{s}}_i$ is the spin of the e_g electron) and suggest relative magnitudes of U , J , and J_x .

Solution. The term with coefficient J represents the alignment of the itinerant electron's spin with the core's spin at the same site. That is, the larger J is, the more compelled the itinerant electron is to align its spin with the core. The term with coefficient J_x represents the alignment of the spins of the cores of neighboring Mn sites.

Similarly, U is the “coupling” for antialignment of electrons within one d level. This is strongly disfavored by Hund's rule. Further, it is more favorable for the itinerant electron to align its spin with its core than for

adjacent Mn ions to have the same spin, since the cores are much further apart from one another than the e_g electron is to the core of its site. Thus the relative magnitudes are

$$J > J_x > U.$$

3(d) Consider two neighboring core spins $\mathbf{S}_i, \mathbf{S}_j$ that are at a relative angle θ_{ij} . By considering that the spin wavefunction of the itinerant electron must, for $J \gg t$, be always aligned with the local core spin \mathbf{S} , explain why the Schrödinger equation for the itinerant electron can be simplified to one in which the tight-binding hopping matrix element from site i to site j is replaced by

$$t_{\text{eff}} = t \cos\left(\frac{\theta_{ij}}{2}\right).$$

To do this, you may wish to note that under a rotation by an angle θ , the spin wavefunction transforms as

$$\begin{pmatrix} |\uparrow'\rangle \\ |\downarrow'\rangle \end{pmatrix} = \begin{pmatrix} \cos(\theta/2) & \sin(\theta/2) \\ -\sin(\theta/2) & \cos(\theta/2) \end{pmatrix} \begin{pmatrix} |\uparrow\rangle \\ |\downarrow\rangle \end{pmatrix}.$$

Solution. Intuitively, the expression for t_{eff} makes sense: $t_{\text{eff}} = t$ when θ_{ij} and the core spins are aligned; this makes hopping possible even when $J \gg t$. When $\theta_{ij} = \pi$, the core spins are completely antiparallel, and $t_{\text{eff}} = 0$, rendering hopping impossible as we would expect.

This term may come about by explicitly including the spin part of the wave function in the matrix element. That is, we would have something like

$$t_{\text{eff}} = \langle \phi_{e_g}(\mathbf{r} - \mathbf{R}_i), \chi_i | H | \phi_{e_g}(\mathbf{r} - \mathbf{R}_j), \chi_j \rangle,$$

where $|\chi_i\rangle$ is the spin of the i th site. The possibilities are

$$|\chi_i\rangle = \begin{cases} |\uparrow_i\rangle, \\ \frac{|\uparrow_i\rangle + |\downarrow_i\rangle}{\sqrt{2}}, \\ \frac{|\uparrow_i\rangle - |\downarrow_i\rangle}{\sqrt{2}}, \\ |\downarrow_i\rangle. \end{cases}$$

Any χ_j may be rotated through an angle θ_{ij} by

$$\begin{pmatrix} |\uparrow_j\rangle \\ |\downarrow_j\rangle \end{pmatrix} = \begin{pmatrix} \cos(\theta_{ij}/2) & \sin(\theta_{ij}/2) \\ -\sin(\theta_{ij}/2) & \cos(\theta_{ij}/2) \end{pmatrix} \begin{pmatrix} |\uparrow_i\rangle \\ |\downarrow_i\rangle \end{pmatrix} = \begin{pmatrix} \cos(\theta_{ij}/2) |\uparrow_i\rangle + \sin(\theta_{ij}/2) |\downarrow_i\rangle \\ -\sin(\theta_{ij}/2) |\uparrow_i\rangle + \cos(\theta_{ij}/2) |\downarrow_i\rangle \end{pmatrix}.$$

Noting that

$$\begin{aligned} \langle \uparrow_i | \uparrow_j \rangle &= \langle \uparrow_i | \left[\cos\left(\frac{\theta_{ij}}{2}\right) |\uparrow_i\rangle + \sin\left(\frac{\theta_{ij}}{2}\right) |\downarrow_i\rangle \right] = \cos\left(\frac{\theta_{ij}}{2}\right), \\ \langle \uparrow_i | \downarrow_j \rangle &= \langle \uparrow_i | \left[-\sin\left(\frac{\theta_{ij}}{2}\right) |\uparrow_i\rangle + \cos\left(\frac{\theta_{ij}}{2}\right) |\downarrow_i\rangle \right] = -\sin\left(\frac{\theta_{ij}}{2}\right) \langle \uparrow_i | \uparrow_i \rangle, \\ \langle \downarrow_i | \downarrow_j \rangle &= \langle \downarrow_i | \left[-\sin\left(\frac{\theta_{ij}}{2}\right) |\uparrow_i\rangle + \cos\left(\frac{\theta_{ij}}{2}\right) |\downarrow_i\rangle \right] = \cos\left(\frac{\theta_{ij}}{2}\right), \\ \langle \downarrow_i | \uparrow_j \rangle &= \langle \downarrow_i | \left[\cos\left(\frac{\theta_{ij}}{2}\right) |\uparrow_i\rangle + \sin\left(\frac{\theta_{ij}}{2}\right) |\downarrow_i\rangle \right] = \sin\left(\frac{\theta_{ij}}{2}\right) \langle \downarrow_i | \downarrow_i \rangle, \end{aligned}$$

we also have

$$\frac{\langle \uparrow_i | + \langle \downarrow_i |}{\sqrt{2}} \frac{|\uparrow_j\rangle + |\downarrow_j\rangle}{\sqrt{2}} = \frac{\langle \uparrow_i | \uparrow_j \rangle + \langle \uparrow_i | \downarrow_j \rangle + \langle \downarrow_i | \uparrow_j \rangle + \langle \downarrow_i | \downarrow_j \rangle}{2} = \cos\left(\frac{\theta_{ij}}{2}\right),$$

$$\frac{\langle \downarrow_i | + \langle \uparrow_i |}{\sqrt{2}} \frac{|\downarrow_j\rangle + |\uparrow_j\rangle}{\sqrt{2}} = \frac{\langle \downarrow_i | \downarrow_j \rangle + \langle \downarrow_i | \uparrow_j \rangle + \langle \uparrow_i | \downarrow_j \rangle + \langle \uparrow_i | \uparrow_j \rangle}{2} = \cos\left(\frac{\theta_{ij}}{2}\right).$$

So all of the pairs where $|\chi_i\rangle$ and $|\chi_j\rangle$ have the same form give us

$$t_{\text{eff}} = \langle \phi_{e_g}(\mathbf{r} - \mathbf{R}_i), \chi_i | H | \phi_{e_g}(\mathbf{r} - \mathbf{R}_j), \chi_j \rangle \cos\left(\frac{\theta_{ij}}{2}\right) = t \cos\left(\frac{\theta_{ij}}{2}\right)$$

as desired.

3(e) Sketch the density of states of the itinerant electrons for different alignments of the core spins \mathbf{S} :

- (i) *ferromagnetic* (all core spins aligned),
- (ii) *antiferromagnetic* (all neighboring core spins anti-aligned),
- (iii) *paramagnetic* (core spins randomly aligned).

Discuss how the total free energies of these states differ, and suggest what is the magnetic ground state when $x = 0$, and when $tx > J_x$; give rough estimates of the transition temperatures of the ordered magnetic states toward high temperature paramagnetism.

Solution. The density of states $g(E)$ for each case is shown in Fig. 3. These illustrations are based on Fig. 4.2 in the lecture notes. For the ferromagnetic case in Fig. 3 (left), the core spins are all aligned which facilitates hopping. This makes the same number of states available for a relatively wide range of energies, since hopping does not change the number of available states but does allow for the electrons to decrease their kinetic energy. For the antiferromagnetic case in Fig. 3 (middle), hopping is disallowed, which means the electrons must remain localized in the same number of available states. They cannot change their energy by hopping, so these states are only available at one value of the energy. The paramagnetic case in Fig. 3 (right) lies somewhere in between these two extremes.

The free energy $F = U - TS$, where U is internal energy, T is temperature, and S is entropy [8, p. 48]. Assuming the second term does not differ between the three states, we need only concern ourselves with U . From (2.14) of the lecture notes,

$$U = \int dE E g(E) f(E).$$

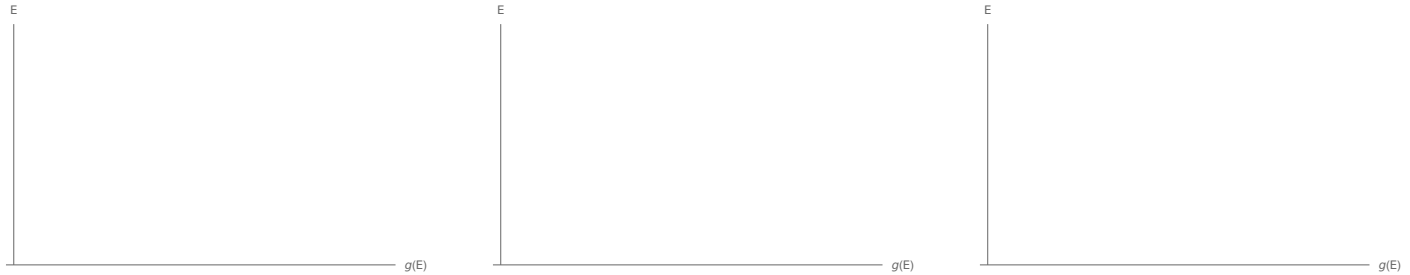


Figure 3: Density of states $g(E)$ for the ferromagnetic case (left), antiferromagnetic case (middle), and paramagnetic case (right).

Since the largest area under the $g(E)$ curve is largest in the ferromagnetic case, smallest in the antiferromagnetic case, and somewhere in between in the paramagnetic case, so too is U . Thus,

$$F_{\text{ferro}} > F_{\text{para}} > F_{\text{antiferro}}.$$

When $x = 0$, we have LaMnO_3 which contains only Mn^{3+} . In this case the magnetic ground state is **ferromagnetic**. The ferromagnetic configuration facilitates hopping of the itinerant electrons, and this hopping decreases the system's kinetic energy. When all of the Mn atoms have an itinerant electron, these electrons will be perpetually hopping between ions (and being replaced by an itinerant electron from another ion), and the total energy of the system will be as small as possible.

When $tx > J_x$, hopping will occur regardless of whether neighboring core spins are aligned. J_x being small means that there is no incentive. Thus the ground state is **antiferromagnetic** in this case.

The transition temperatures are in the **few hundred Kelvin** range. The transition temperature increases with x because the frequency of hopping decreases as x increases.

3(f) Figure 6.13 shows the resistivity as a function of temperature of several samples of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ with different concentrations x , as well as the magnetic field dependence of the resistivity (which gives rise to the label “colossal” magnetoresistance). Discuss this data in light of the results above.

Solution. Figure 6.13 in the lecture notes is reproduced here as Fig. 4. Resistivity is inversely proportional to conductivity [9, p. 296]. The more freely the itinerant electrons can travel, the more conductive and the less resistive the crystal will be.

When $x \leq 0.15$, the ground state is ferromagnetic. Increasing the temperature gives the itinerant electrons more kinetic energy and allows them to hop more freely among the Mn ions, thereby decreasing the resistivity. When a ferromagnetic transition occurs such as for $x = 0.15$ in Fig. 4 (left), the spin antialignment of some neighboring cores in the paramagnetic phase hinders hopping, increasing resistivity. But when the temperature is sufficiently increased past this point, resistivity decreases once more.

When $x > 0.15$ in Fig. 4 (left), the ground state is antiferromagnetic. Resistivity increases with temperature until T_c , after which resistivity decreases slowly or plateaus. This is because the paramagnetic phase allows the itinerant electrons to travel more freely than the antiferromagnetic phase.

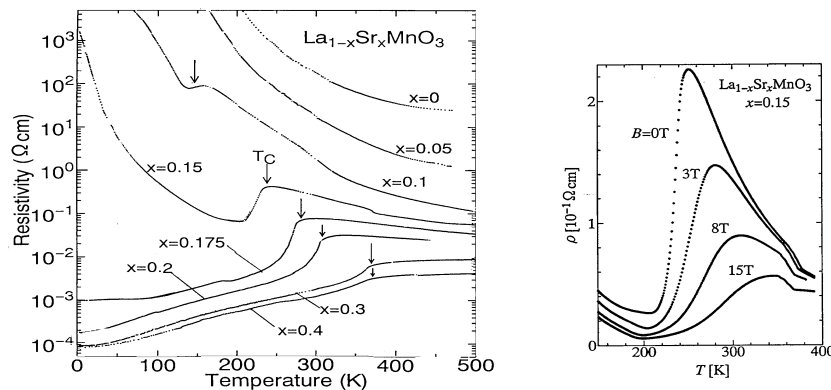


Figure 4: [Figure 6.13 of the lecture notes] Resistivity as a function of temperature and magnetic field for a series of samples of doped manganese oxides with different compositions. The ferromagnetic transition temperatures T_c are marked by the arrows.

The resistivity decreases with increasing magnetic field intensity as shown in in Fig. 4 (right). This is because the application of a magnetic field to $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ tends to align the electron spins (both the itinerant electrons and the cores), which eases hopping of the itinerant electrons and increases conductivity. The stronger the magnetic field, the better the alignment among the spins, and the more conductive the crystal becomes.

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