

On Magnetism in Insulators*

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Slater's split-band theory of insulating anti-ferromagnetism is discussed in some detail by means of a simplified nondegenerate one-band model. A comparison with Anderson's theory of super-exchange is also made.

A teoria de Slater do antiferromagnetismo em isolantes é discutida em algum detalhe por meio de um modelo simplificado. É também feita uma comparação com a teoria de super-troca de Anderson.

1. Introduction

Magnetic interaction in insulators creates, with a few exceptions, antiferromagnetic order. This is generally the case of substances, like transition metals and rare earths oxides and salts, where the magnetic atoms are separated by relatively large diamagnetic radicals. One can expect the Heisenberg model to be applicable since the metallic ions are far apart. However, the direct exchange interaction is positive and therefore forces ferromagnetic ordering. Kramers¹ in 1934 introduced the concept of superexchange; the intermediate radicals cause spin dependent perturbations on the magnetic atoms wave functions transmitting an antiferromagnetic exchange effect at large distances. The reason why such an antiferromagnetic state might come about was clarified in a more intuitive way by Mott². The major contribution to the theory is due to Anderson^{3,4,5}. Anderson's idea is to assume that the band structure of these compounds has been already solved, thus incorporating the effect of the radicals. Next, one solves the many-electron problem. Using Wannier representation, it is possible to show that, up to second order in perturbation theory, a large contribution to the spin dependent interactions between d-electrons comes from virtual occupation of ionized states. Since electron transfer can occur, because of Pauli principle, only between electrons in neigh-

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bouring ions with antiparallel spins, the interaction is necessarily antiferromagnetic. The two parameters that enter the theory are the hopping integral b between nearest neighbours and the Coulomb interaction U . Since U is much larger than b , the real occupation of ionized states is energetically forbidden thus making these compounds insulators.

The effective magnetic interaction Hamiltonian in Anderson's theory can be written in the form of a Heisenberg-type interaction^{3,4,5}

$$H_{eff.} = - \sum_{ij} A_{ij} \mathbf{S}_i \cdot \mathbf{S}_j,$$

where

$$A_{ij} = \frac{2b_{ij}}{U} - J_{ij}.$$

The first term is the so called kinetic exchange whereas the second is the usual direct exchange. Antiferromagnetic order will result if kinetic exchange exceeds the potential exchange. This seems to be generally the case, explaining why a large number of transition metal compounds are antiferromagnetic. However, it should be remarked that the direct evaluation of both parameters is a formidable task, to our knowledge not yet accomplished.

Back in 1951, it was proposed a band model of antiferromagnetism in insulators⁶. According to this model, the electronic bands are assumed to be splitted due to a Hartree-Fock-type exchange potential which renders the material antiferromagnetic. If originally the band was half filled, now, under conditions that can assure non-overlapping sub-bands, the lower sub-band will be occupied whereas the upper sub-band is empty and the material becomes an insulator in the conventional Wilson sense. One difficulty associated with this molecular field model is that it predicts a metallic behaviour above the Néel temperature, when the sub-bands collapse, which contradicts the experimental observation that most of antiferromagnets remain insulators in the non-magnetic phase.

Several authors proposed a model of this kind to describe the cases of some transition metal oxides and sulfides which show a magnetic-non-metal to non-magnetic-metallic transition⁷. These results were shown to be incorrect: correlation effects, decisive near the transition point, should more properly be taken into account⁸. However, it is possible that a band model can give a good description of the ground state and the statistics of insulating antiferromagnets at low temperature⁸.

In a previous paper¹⁰, Slater's-split-band model and Anderson's theory of kinetic exchange were compared and a discussion was given on the common characteristics in the limit when the band width is small compared to the intra-atomic electron-electron interaction. Some advantages and disadvantages of both models were also pointed out. The present paper should be considered complementary to the paper of Ref. 10, since it considers in detail the characteristics of the ground state wave function, a discussion on the determination of the magnetic ordering being presented.

A simple example is given in the Appendix in order to further clarify the subject.

In the next Section, some results of Ref. 10 are summarized. Section 3 is devoted to the discussion of stability of different magnetic structures, and Section 4 deals with the characteristics of the ground state wave function.

2. Slater's Antiferromagnet

Let us consider a material with one outer electron per ion, moving in a nondegenerate band, with $\phi_{\mathbf{k}}$ and $\varepsilon_{\mathbf{k}}$ being the single-electron Bloch functions and energies. These functions are assumed to contain all the effects of the diamagnetic radicals, but not the interaction with the other electrons. A magnetic state can be described by determinantal functions formed with the one-electron wavefunctions:

$$\psi_{\mathbf{k}}^{(-)}(\mathbf{r}) = \cos \theta_{\mathbf{k}} \phi_{\mathbf{k}}(\mathbf{r})|\uparrow\rangle + \sin \theta_{\mathbf{k}} \phi_{\mathbf{k}+\mathbf{Q}}(\mathbf{r})|\downarrow\rangle, \quad (2-1a)$$

$$\psi_{\mathbf{k}}^{(+)}(\mathbf{r}) = -\sin \theta_{\mathbf{k}} \phi_{\mathbf{k}}(\mathbf{r})|\uparrow\rangle + \cos \theta_{\mathbf{k}} \phi_{\mathbf{k}+\mathbf{Q}}(\mathbf{r})|\downarrow\rangle. \quad (2-1b)$$

A determinant Φ_0 formed with the functions (2-1a), in second quantization notation,

$$|\Phi_0\rangle = \prod_{\mathbf{k}} (\cos \theta_{\mathbf{k}} C_{\mathbf{k}\uparrow}^\dagger + \sin \theta_{\mathbf{k}} C_{\mathbf{k}+\mathbf{Q}\downarrow}^\dagger) |0\rangle \quad (2-2)$$

is an eigenfunction of the reduced Hamiltonian

$$\begin{aligned} H_{\text{red}} = & \sum_{\mathbf{k}\alpha} e_{\mathbf{k}} C_{\mathbf{k}\alpha}^\dagger C_{\mathbf{k}\alpha} + \frac{1}{2} \sum_{\mathbf{k}\sigma} \sum_{\mathbf{k}'\alpha'} [\langle \mathbf{k}, \mathbf{k}' | V | \mathbf{k}', \mathbf{k} \rangle \\ & - \langle \mathbf{k}, \mathbf{k}' | V | \mathbf{k}, \mathbf{k}' \rangle \delta_{\sigma\sigma'}] C_{\mathbf{k}\sigma}^\dagger C_{\mathbf{k}\sigma} C_{\mathbf{k}'\sigma'}^\dagger C_{\mathbf{k}'\sigma'} \\ & - \frac{1}{2} \sum_{\mathbf{k}\mathbf{k}'} \langle \mathbf{k} + \mathbf{Q}, \mathbf{k}' | V | \mathbf{k}, \mathbf{k}' + \mathbf{Q} \rangle C_{\mathbf{k}\uparrow}^\dagger C_{\mathbf{k}+\mathbf{Q}\downarrow} C_{\mathbf{k}'+\mathbf{Q}\downarrow}^\dagger C_{\mathbf{k}'\uparrow}, \end{aligned} \quad (2-3)$$

where V is the interparticle potential. Clearly, the first term in (2-3) is the kinetic or band energy, the second is the usual direct and exchange interaction energies and the last is the equivalent of the Weiss molecular field interaction; \mathbf{k} runs over the entire Brillouin zone. The ground state energy $E_0 = \langle \Phi_0 | \mathbf{H} | \Phi_0 \rangle$ follows immediately:

$$E_0 = \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} \cos^2 \theta_{\mathbf{k}} + \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}+\mathbf{Q}} \sin^2 \theta_{\mathbf{k}} + NV_0 - \sum_{\mathbf{k}\mathbf{k}'} V(\mathbf{k}-\mathbf{k}') \cos^2(\theta_{\mathbf{k}} - \theta_{\mathbf{k}'}). \quad (2-4)$$

We have assumed that the exchange matrix elements of the interaction potential only depends on the difference of wavenumbers \mathbf{k} of the initial and final Bloch states, i.e., $V(\mathbf{k}-\mathbf{k}') = \langle \mathbf{k}\mathbf{k}' | V | \mathbf{k}\mathbf{k}' \rangle = \langle \mathbf{k}+\mathbf{Q}, \mathbf{k}' | V | \mathbf{k}, \mathbf{k}'+\mathbf{Q} \rangle$, and $V_0/N = \langle \mathbf{k}\mathbf{k}' | V | \mathbf{k}'\mathbf{k} \rangle$.

We observe that the Bloch functions appearing in (2-1) must, in principle be determined through a Hartree-Fock calculation. The resulting Bloch functions would now contain the molecular field effect. This is a very difficult procedure, which we have replaced by a simpler one in which the ϕ 's are the usual Hartree-Fock-Bloch solutions and two variational parameters $\theta_{\mathbf{k}}$ and \mathbf{Q} have been introduced into the theory.

Minimizing $E_0(\theta_{\mathbf{k}}, \mathbf{Q})$ with respect to $\theta_{\mathbf{k}}$, we obtain the self-consistency condition

$$2\Delta(\mathbf{k}) = \sum_{\mathbf{k}'} V(\mathbf{k}-\mathbf{k}') \sin 2\theta_{\mathbf{k}'}, \quad (2-5)$$

where $\theta_{\mathbf{k}'}$ is given by

$$\tan 2\theta_{\mathbf{k}} = \frac{2\Delta(\mathbf{k})}{\hat{\varepsilon}_{\mathbf{k}+\mathbf{Q}} - \hat{\varepsilon}_{\mathbf{k}}}, \quad (2-6)$$

with

$$\hat{\varepsilon}_{\mathbf{k}} = \varepsilon_{\mathbf{k}} + V_0 - \sum_{\mathbf{k}'} V(\mathbf{k}-\mathbf{k}') \sin^2 \theta_{\mathbf{k}'}, \quad (2-7a)$$

and

$$\hat{\varepsilon}_{\mathbf{k}+\mathbf{Q}} = \varepsilon_{\mathbf{k}+\mathbf{Q}} + V_0 - \sum_{\mathbf{k}'} V(\mathbf{k}-\mathbf{k}') \cos^2 \theta_{\mathbf{k}'}. \quad (2-7b)$$

Let us observe that $\sin^2 \theta_{\mathbf{k}} = \langle \mathbf{O} | n_{\mathbf{k}+\mathbf{Q}} | \Phi_0 \rangle$ and $\cos^2 \theta_{\mathbf{k}} = \langle \Phi_0 | n_{\mathbf{k}} | \Phi_0 \rangle$, and therefore E are the normal Hartree-Fock one-electron energies.

The Hamiltonian (2-3) can be diagonalized by means of the Bogoliubov transformation,

$$\alpha_{\mathbf{k}(-)} = \cos \theta_{\mathbf{k}} C_{\mathbf{k}\uparrow} + \sin \theta_{\mathbf{k}} C_{\mathbf{k}+\mathbf{Q}\downarrow}, \quad (2-8a)$$

$$\alpha_{\mathbf{k}(+)} = -\sin \theta_{\mathbf{k}} C_{\mathbf{k}\uparrow} + \cos \theta_{\mathbf{k}} C_{\mathbf{k}+\mathbf{Q}\downarrow}, \quad (2-8b)$$

the one-electron eigen-energies being

$$\omega_{\mathbf{k}}^{(\pm)} = \frac{1}{2} (\tilde{\varepsilon}_{\mathbf{k}+\mathbf{Q}} + \tilde{\varepsilon}_{\mathbf{k}}) \pm \sqrt{\frac{1}{4} (\tilde{\varepsilon}_{\mathbf{k}+\mathbf{Q}} - \tilde{\varepsilon}_{\mathbf{k}})^2 + |\Delta_{\mathbf{k}}|^2} \quad (2-9)$$

The magnetic character of state (2-2) can be put into evidence by evaluating the average magnetization

$$M_+(\mathbf{r}) = \mu \sum_{\mathbf{k}} \phi_{\mathbf{k}}^*(\mathbf{r}) \phi_{\mathbf{k}+\mathbf{Q}}(\mathbf{r}) \sin 2\theta_{\mathbf{k}}, \quad (2-10)$$

which is different from zero whereas Eq. (2-5) admits a non-trivial, $A \neq 0$, solution. Q , which should be determined by minimizing $E_0(\theta_{\mathbf{k}}, Q)$, determines the magnetic structure and will be considered in the next section. It would be noted that Q was assumed to be commensurate with the lattice, i.e., $\mathbf{k} + 2Q$ represents the same state as \mathbf{k} . Otherwise, one would have to include higher harmonics in the present formalism in order to obtain a self-consistent average field solution. Let us remark that if we assume (2-2) as the ground state wavefunction, it is implicit that the bandwidth is small compared to A , in order to prevent an overlapping of the sub-bands. Hence, $|\Phi_0\rangle$ is the Hartree-Fock ground state of Hamiltonian (2-3) and represents a fully occupied lower sub-band. The system is clearly an insulator.

We now make the approximation of replacing the exchange matrix elements $V(\mathbf{k}-\mathbf{k}')$ by a constant U/N , which implies a contact-type interaction between electrons. This is the case of a highly screened interaction in an electron gas or the case of highly localized electrons. $\Delta_{\mathbf{k}}$ is then independent of \mathbf{k} as can be seen from Eq. (2-5).

One needs next to establish the stability of this antiferromagnetic state. This is not possible to accomplish in general. We shall consider here only the comparison with the saturated ferromagnetic and paramagnetic states. The difference in Hartree-Fock energies per particle are, respectively,

$$E_0 - E_{\text{FM}} = \frac{U}{4} - \frac{\Delta^2}{U} - \frac{1}{N} \sum_{\mathbf{k}} x_{\mathbf{k}}^2 E_{\mathbf{k}}^{-1} \quad (2-11)$$

and

$$E_0 - E_{\text{PM}} = -\frac{\Delta^2}{U} - \frac{1}{N} \sum_{\mathbf{k}} x_{\mathbf{k}}^2 E_{\mathbf{k}}^{-1} + \frac{1}{N} \sum_{\mathbf{k}} |\varepsilon_{\mathbf{k}} - \mu|, \quad (2-12)$$

where $x_{\mathbf{k}} = (1/2)(\tilde{\varepsilon}_{\mathbf{k} + \mathbf{Q}} - \tilde{\varepsilon}_{\mathbf{k}})$, $E_{\mathbf{k}} = (x_{\mathbf{k}}^2 + \Delta^2)^{1/2}$ and μ is the paramagnetic Fermi energy. As shown in Ref. 10, (2-11) is non-positive. This is not possible to show for (2-12) without a knowledge of the band structure. However, it is obvious that for Δ large enough compared to the band energies, the insulating state is doubtless more stable. This observation leads us to suspect that, in principle, an instability as compared to the paramagnetic state could occur for some critical value of U/Γ , where Γ is the non-magnetic band-width. But, in lowering the ratio U/Γ , a sub-band overlap is not excluded to happen previous to the non-magnetic instability. For an f.c.c. structure, and in the tight binding approximation, band overlap began when $\Delta/\Gamma \sim 0.15$. When the nesting condition, $\varepsilon_{\mathbf{k} + \mathbf{Q}} = -\varepsilon_{\mathbf{k}}$, is satisfied such overlap does not occur for all values of the ration U/Γ (See Appendix). The Statistical Mechanics of the model, based on a thermally averaged exchange field, leads to a vanishing thermal gap for a certain critical temperature T_c , resulting in a transition to the metallic paramagnetic state. This kind of behaviour was believed to apply to the thermal transitions in titanium and vanadium oxides⁷, but the method is invalid at intermediate and high temperatures⁸. Recently, it has been speculated that stability of the magnetic phase can be improved when an additional correlation is introduced with further combinations of band-Bloch functions¹². Another point that should be stressed is that we have been constantly considering U and Γ as independent of the state of the system. This is a crude oversimplification, which however is not important in the limit of large U/Γ ratio. Correlation effects will be discussed in a forthcoming paper.

We omit here the comparison with Anderson's theory of superexchange given in Ref. 10 and proceed in the next Section to discuss the parameter \mathbf{Q} .

3. The Magnetic Structure

The condition for energy (2-4) be extremal in relation to \mathbf{Q} is

$$\frac{\partial E_0(\theta_{\mathbf{k}}, \mathbf{Q})}{\partial \mathbf{Q}} = - \sum_{\mathbf{k}} \cos 2\theta_{\mathbf{k}} \frac{\partial \varepsilon_{\mathbf{k} + \mathbf{Q}}}{\partial \mathbf{Q}} = 0. \quad (3-1)$$

In deriving Eq. (3-1) it was taken into account the \mathbf{Q} dependence of the

angle θ_k . Using the relationship

$$\cos 2\theta_k = x_k/E_k$$

and assuming that the Brillouin zone has a center of symmetry, i.e., that $\varepsilon_{\mathbf{k}} = \varepsilon_{-\mathbf{k}}$, it is straightforward to show that a solution of Eq. (3-1) implies $\varepsilon_{-\mathbf{k}+\mathbf{Q}} = \varepsilon_{\mathbf{k}+\mathbf{Q}}$. This means that either $\mathbf{Q} = 0$ or \mathbf{Q} is a vector of the reciprocal space with end point on the Brillouin zone surface. Let us consider the limit of small Γ/Δ ratio. Expanding the Bloch functions in terms of Wannier functions and neglecting overlap between Wannier functions centered on different lattice sites \mathbf{R} , the magnetization (2-10) becomes

$$M(\mathbf{r}) = \mu_{eff} \sum_{\mathbf{R}} |w(\mathbf{r}-\mathbf{R})|^2 \exp(i\mathbf{Q} \cdot \mathbf{R}), \quad (3-2)$$

where

$$\mu_{eff} = \frac{\mu}{N} \sum_{\mathbf{k}} \sin^2 \theta_{\mathbf{k}}.$$

The magnetization of (3-2) shows that a fractional Bohr magneton μ_{eff} per electron is distributed around each lattice site in the "plus" or "minus" direction according to the phase factor $\exp(i\mathbf{Q} \cdot \mathbf{R})$ being plus or minus one, thus defining the magnetic structure. Obviously $\mathbf{Q} = 0$ produces ferromagnetic ordering.

In the limits of $\Gamma/\Delta \ll 1$ and $\Gamma/\Delta \gg 1$, it can easily be shown that E_c depends quadratically on the order parameter A . Under these conditions one can use the arguments advanced by Dzyaloshinskii¹³ to derive a "selection rule" for \mathbf{Q} stating that the values of \mathbf{Q} that make the energy an extremum are those whose group vector¹⁴ contain the inversion operator. Consequently, the group vector of \mathbf{Q} should belong to one of the groups D_2 , D_{2d} , D_{3h} , D_3 , D_{3h} , S_4 , D_4 , D_6 , T , T_d , and O .

In Table I, we give the corresponding values of \mathbf{Q} that reproduce different magnetic structures in sc, bcc and fcc materials shown in Fig. 1¹⁵. In order to determine which is the more stable magnetic structure, it is necessary to make a direct comparison of the ground state energies (See Appendix).

It would be instructive to study the ground state wave function of the Slater model, mainly in the limit of small Γ/U , i.e., the region of equivalence with Anderson's theory. This is done in the next Section.

Crystal Structure	Magnetic Structure	Q (units of π/a)	Brillouin Zone point	Symmetry Group
<i>sc</i>	First class	(1, 1, 1)	<i>R</i>	O_h (m3m)
	Second class	(1, 1, 0)	<i>M</i>	D_{4h} (4/mmm)
	Third class	(1, 0, 0)	<i>X</i>	D_{4h} (4/mmm)
<i>bcc</i>	First class	(2, 0, 0)	<i>H</i>	O_h (m3m)
	Second class(a)	(1, 1, 0)	<i>N</i>	D_2 ($\overline{222}$)
	Second class(b)	(1, 1, 1)	<i>P</i>	T_d (43m)
<i>fcc</i>	First class	(2, 0, 0)	<i>X</i>	D_{4h} (4/mmm)
	Second class	(1, 1, 1)	<i>L</i>	D_3 (32)
	Fourth class (two magnetization axes)	(1, 0, 2)	<i>W</i>	D_{2d} ($\overline{42m}$)

Table 1.

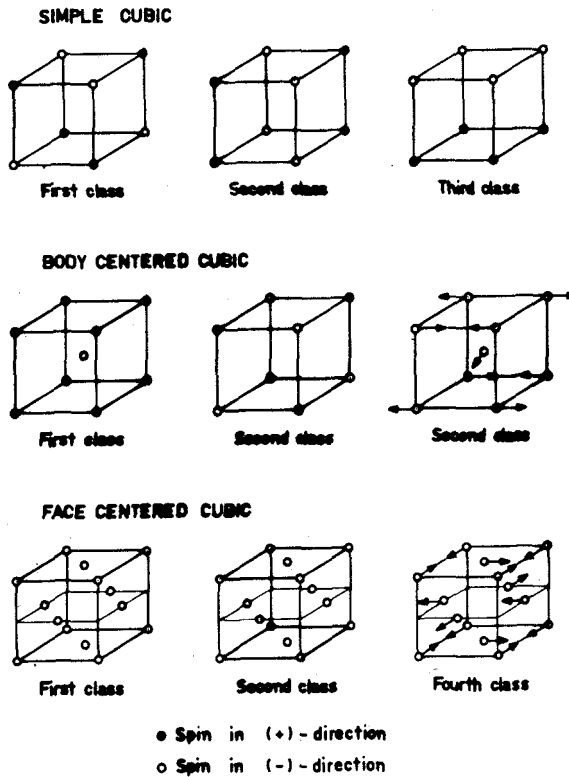


Fig. 1 - Different classes of magnetic ordering.

4. Ground State Wave Function

The ground state wavefunction of the insulating antiferromagnet (2-2) can alternatively be written

$$|\Phi\rangle = \prod_k (\cos \theta_k + \sin \theta_k b_k^\dagger) |\text{FM}\rangle \quad (4-1)$$

where

$$b_k^\dagger = C_{k+Q\downarrow}^\dagger C_{k\uparrow}$$

and

$$|\text{FM}\rangle = \prod_k C_{k\uparrow}^\dagger |0\rangle \quad (4-2)$$

is the wavefunction of the insulating ferromagnetic state.

In the limit of small Δ , θ_k becomes small and

$$|\Phi_0\rangle \sim |\text{FM}\rangle + \sum_k \theta_k b_k^\dagger |\text{FM}\rangle, \quad (4-3)$$

which corresponds to the creation of a small amplitude static spin density wave in the ferromagnetic state¹⁶. The wavefunction (4-1) shows the character of the AFM state: a static spin density wave of large amplitude and period $2\pi/Q = 2R$ superimposed on the ferromagnetic state. When $\Delta = 0$ both states are degenerate in energy. In fact, there are 2^N degenerate states that correspond to all possible orientations of the N electrons spins one per atom.

If Q tends to zero, one has

$$\theta_k \sim \frac{\pi}{4} - \frac{\mathbf{Q} \cdot \nabla_k \epsilon_k}{4\Delta}$$

and

$$|\Phi_0\rangle \sim |\Phi_x\rangle - |\Phi'\rangle, \quad (4-4)$$

where

$$|\Phi_x\rangle = \frac{1}{\sqrt{2}} \prod_k (C_{k\uparrow}^\dagger + C_{k+Q\downarrow}^\dagger) |0\rangle \quad (4-5)$$

and

$$|\Phi'\rangle = \sum_{\mathbf{k}} \frac{\mathbf{Q} \cdot \nabla \varepsilon_{\mathbf{k}}}{4\Delta} (C_{\mathbf{k}\uparrow}^\dagger - C_{\mathbf{k}+\mathbf{Q}\downarrow}^\dagger)(C_{\mathbf{k}\uparrow} + C_{\mathbf{k}+\mathbf{Q}\downarrow})|0\rangle. \quad (4-6)$$

The state can be described as an FM state with domains of width $\sim 2\pi/\mathbf{Q}^{17}$.

In the limit of small Γ/U and \mathbf{Q} describing antiferromagnetic ordering of the kind considered in Section 3, one finds

$$\theta_{\mathbf{k}} \sim \frac{\pi}{4} + \frac{x_{\mathbf{k}}}{2\Delta}. \quad (4-7)$$

Hence

$$|\Phi_0\rangle \sim |\Phi_x\rangle + \sum_{\mathbf{k}} \frac{x_{\mathbf{k}}}{\Delta} L_{\mathbf{k}}^\dagger R_{\mathbf{k}} |\Phi_x\rangle, \quad (4-8)$$

where we have defined

$$L_{\mathbf{k}}^\dagger = \frac{1}{\sqrt{2}} (C_{\mathbf{k}\uparrow}^\dagger - C_{\mathbf{k}+\mathbf{Q}\downarrow}^\dagger), \quad (4-9a)$$

$$R_{\mathbf{k}} = \frac{1}{\sqrt{2}} (C_{\mathbf{k}\uparrow} + C_{\mathbf{k}+\mathbf{Q}\downarrow}). \quad (4-9b)$$

Introducing the Wannier transformation, we find

$$|\Phi_0\rangle \sim |\Phi_x\rangle + \sum_{\mathbf{R}\tau} \frac{b_\tau}{U} (\exp\{i\mathbf{Q} \cdot \tau\} - 1) L_{\mathbf{R}+\tau}^\dagger R_{\mathbf{R}} |\Phi_x\rangle, \quad (4-10)$$

where b_τ is the transfer or hopping integral, \mathbf{R} and τ are lattice vectors and

$$L_{\mathbf{R}}^\dagger = \frac{1}{\sqrt{2}} [C_{\mathbf{R}\uparrow}^\dagger - \exp(i\mathbf{Q} \cdot \mathbf{R}) C_{\mathbf{R}\downarrow}^\dagger],$$

$$R_{\mathbf{R}} = \frac{1}{\sqrt{2}} [C_{\mathbf{R}\uparrow} + \exp(i\mathbf{Q} \cdot \mathbf{R}) C_{\mathbf{R}\downarrow}].$$

This is quite general. To obtain some insight into the characteristics of function (4-10), we shall consider antiferromagnetic ordering of the first class in a *bcc* material. In this case the antiferromagnet can be described by two interpenetrated magnetic *sc* lattices, one with spins in $(-x)$ -direction with their positions indicated by \mathbf{L} in the original *bcc* lattice, and the other with spins in $(+x)$ -direction with their positions indicated by \mathbf{R} in the original *bcc* lattice. Evidently, $\mathbf{L} = \mathbf{R} + \boldsymbol{\tau}$, where $\boldsymbol{\tau}$ is the vector displacement from a point \mathbf{L} or \mathbf{R} to a nearest neighbour *bcc* lattice point.

Let us choose the origin of coordinates in an R-type point. Then $\exp(i\mathbf{Q} \cdot \mathbf{R}) = 1$ and $\exp(i\mathbf{Q} \cdot \mathbf{L}) = -1$, since $\exp(i\mathbf{Q} \cdot \boldsymbol{\tau}) = -1$. Using these results, we obtain, when considering only nearest neighbours,

$$\begin{aligned} |\Phi_0\rangle \sim |\Phi_x\rangle - \frac{2b}{U} \sum_{\tau} \sum_{\mathbf{R}}^{n.n.} R_{\mathbf{R}+\tau}^{\dagger} R_{\mathbf{R}} |\Phi_x\rangle \\ - \frac{2b}{U} \sum_{\tau} \sum_{\mathbf{L}} L_{\mathbf{L}+\tau}^{\dagger} L_{\mathbf{L}} |\Phi_x\rangle \end{aligned} \quad (4-11)$$

where

$$\begin{aligned} R_{\mathbf{R}} &= \frac{1}{\sqrt{2}} (C_{\mathbf{R}\uparrow}^{\dagger} + C_{\mathbf{R}\downarrow}), \\ L_{\mathbf{L}} &= \frac{1}{\sqrt{2}} (C_{\mathbf{L}\uparrow}^{\dagger} - C_{\mathbf{L}\downarrow}), \end{aligned}$$

and $b_{\tau} = b$ is a constant independent of τ . Clearly, $R^{\dagger} \mathbf{R}$, for instance, transfers an electron with (+) spin from a lattice point of R-type to a neighbouring L-type lattice point, without spin flip.

In coordinate representation, the wavefunction (4-10) is the Slater determinantal function formed with the one-electron functions

$$\left[w(\mathbf{r} - \mathbf{R}_j) - \frac{2b}{U} \sum_{\tau}^{n.n.} w(\mathbf{r} - \mathbf{R}_j - \boldsymbol{\tau}) \right] \eta(\mathbf{R}_j), \quad (4-12)$$

where

$$\eta(\mathbf{R}_j) = \frac{1}{\sqrt{2}} (|\uparrow\rangle + |\downarrow\rangle)$$

if \mathbf{R}_j belongs to sub-lattice of type R and

$$\eta(\mathbf{R}_j) = \frac{1}{\sqrt{2}} (|\uparrow\rangle - |\downarrow\rangle)$$

if \mathbf{R}_j belongs to sub-lattice type L. The order is antiferromagnetic and the orbital part is an admixture of Wannier functions, centered on each lattice site, with Wannier functions centered on nearest neighbouring lattice sites. This is the type of one-electron wave-functions extensively discussed by Anderson^{3,4}. As he stated, kinetic exchange occurs because the best Hartree-Fock orbitals must be orthogonal but, when they are antiparallel, the orbitals may overlap each other. An energy of the order $-2b^2/U$ per orbit pair can be gained.

It is proper to remark, however, that Slater's treatment is more general than Anderson's because it is not restricted to small b/U values since it implies that one starts with the insulating ferromagnetic state (4-2) modifying it with the creation of a giant static spin density wave of period $2\pi/Q$. In Anderson's model, one starts with 2^N degenerate states of N -localized electrons with intra-atomic interaction and then using kinetic (or band) energy as a perturbation, one finds the AFM state to have lower energy. In Section 2, we did not compare the stability of the AFM state against other possible states, except the insulating FM and normal paramagnetic states. A more general comparison would require methods similar to those of Penn¹⁸. In fact, Penn finds a ferromagnetic state as a more stable state but with the hypothesis, not justified, of neglecting the (positive) electrostatic energy involved in the formation of a static charge density wave which is also present. To discuss this kind of structures, it is necessary to consider lattice deformation¹⁹.

Finally, let us observe that the magnetic moment per electron is fractionary. In the limit of small b/U , one obtains

$$\mu_{eff} \sim \mu [1 - (16b^2/U^2)].$$

For systems satisfying the "nesting condition" $\epsilon_{\mathbf{k}+\mathbf{Q}} = -\epsilon_{\mathbf{k}}$, when the insulating antiferromagnet is stable for all values of b/U , μ_{eff}/μ can have any value between 0 and 1. It is also interesting to note that even if there exists superposition of sub-bands for a critical value of b/U in fcc structures, it is possible to stabilize new insulating states by successive phase transformations in the way described by Kohn¹². This can be done with admixtures of states of wavevector $(\pi/a)(1, 0, 2)$ and $(\pi/2a)(1, 1, 1)$. Such combination produces complex magnetic structures.

In conclusion, we may say that the discussion of a simplified Slater's band model presented here, even if not quite realistic, attempts to put in evidence some characteristics of the magnetic insulating state.

Appendix. Magnetic Ordering in SC Structure

In the tight binding approximation, the band structure of a simple cubic crystal is

$$\epsilon_{\mathbf{k}} = b_0 - b(\cos ak_x + \cos ak_y + \cos ak_z); \quad (\text{A-1})$$

when $A \gg b$, we have

$$\begin{aligned} \frac{\partial E_0}{\partial Q_i} &\simeq \sum_{\mathbf{k}} \frac{x_{\mathbf{k}}}{\Delta} \frac{\partial \epsilon_{\mathbf{k}+\mathbf{Q}}}{\partial Q_i} = \frac{b^2}{\Delta} a \int_{-\pi/a}^{\pi/a} dk_x dk_y dk_z \sum_{j=i}^3 \left\{ \cos(k_j + Q_j)a - \cos k_j a \right\} \cdot \sin(k_i + Q_i)a \\ &\sim -\sin aQ_i \end{aligned} \quad (\text{A-2})$$

Therefore, $(\partial E_0 / \partial Q_i) = 0$ implies $Q_i = 0$ or (π/a) . Consequently, the \mathbf{Q} values that make the AFM energy an extremum are the surface Brillouin Zone points R, X and M as predicted in Table 1. Furthermore,

$$\frac{\partial^2 E_0}{\partial Q_i \partial Q_j} = 0 \quad \text{if } i \neq j,$$

$$\frac{\partial^2 E_0}{\partial Q_i^2} \sim -\cos aQ_i = \begin{cases} 1 & \text{if } Q_i = \pi/a, \\ -1 & \text{if } Q_i = 0. \end{cases}$$

$\mathbf{Q} = (\pi/a) (1, 1, 1)$, first class magnetic structure, corresponds to a minimum energy. This result is in agreement with that derived by ter Haar and Lines²⁰ using Anderson's model. This value of \mathbf{Q} satisfy the "nesting condition" $\varepsilon_{\mathbf{k}+\mathbf{Q}} = -\varepsilon_{\mathbf{k}}$ and the one-electron energies become

$$\omega_{\mathbf{k}}^{\pm} = \pm \sqrt{\varepsilon_{\mathbf{k}}^2 + \Delta^2}. \quad (\text{A-3})$$

The stability of the AFM solution when compared to the PM state can easily be verified. Eq. (2-12) becomes

$$E_0 - E_{\text{PM}} = -N \frac{\Delta^2}{U} - 2 \int_0^{3b} d\varepsilon g(\varepsilon) \left[-\frac{\varepsilon^2}{\sqrt{\varepsilon^2 + \Delta^2}} - \varepsilon \right], \quad (\text{A-4})$$

where $g(\varepsilon)$ is the band density of states. This result can also be obtained by application of the Pauli-Feynman theorem

$$E_0 - E_{\text{PM}} = \int_0^U \frac{dV}{U} \langle H \text{ int} \rangle, \quad (\text{A-5})$$

where $\langle H \text{ int} \rangle = -\Delta^2/U$, and the gap equation is

$$(N/U) = \sum_{\mathbf{k}} (\Delta^2 + \varepsilon_{\mathbf{k}}^2)^{-1/2}.$$

Fig. 2 shows the qualitative behaviour of $E_0 - E_{\text{PM}}$ vs Δ/Γ . The energy gap is given in Fig. 3.

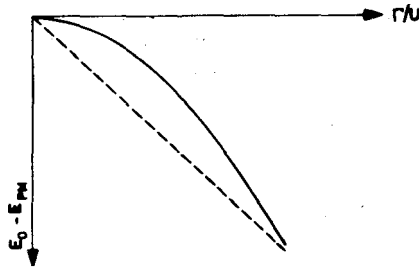


Fig. 2 - Comparison of antiferromagnetic and paramagnetic energies as a function of the band width to intra-atomic interaction ratio.

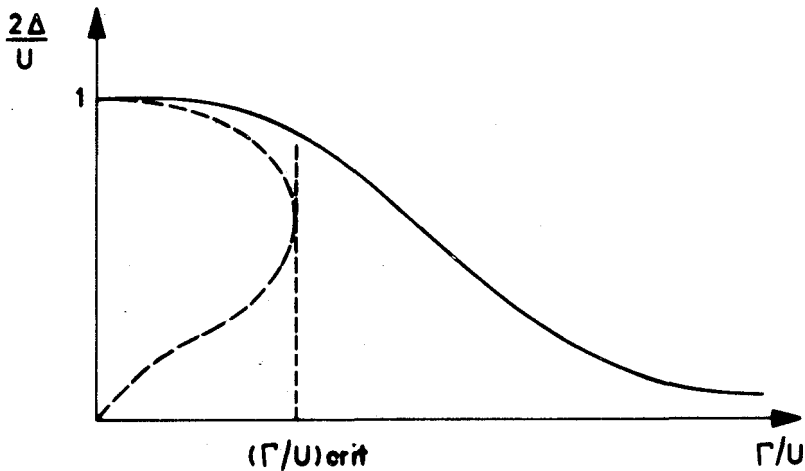


Fig. 3 - The energy gap as a function of the band width to intra-atomic interaction ratio.

For small Γ/U values ($\Gamma = 6b$), the gap equation is approximately given by

$$\left(\frac{2\Delta}{U}\right)^3 - \left(\frac{2\Delta}{U}\right)^2 + 2c\left(\frac{b}{U}\right)^2 = 0, \quad (\text{A-6})$$

where

$$cb^2 = \frac{1}{N} \sum_k x_k^2.$$

The solution of cubic equation (A-6) is indicated by the dashed line in Fig. 3. It can be obtained in second order perturbation theory in b/Δ^5 . Based on this result, Anderson was lead to conclude that, above a critical Γ/Δ value, the magnetic state can no longer be maintained, becoming unstable with respect to a redistribution of the electrons making the metallic-paramagnetic state more stable. As a result of the present discussion, one can see that this is not the case. The perturbation series has a weak convergence and their exact sum is the self-consistency or gap equation. The insulating AFM state is stable for all values of Γ/U . For large values of Γ/U , the gap decreases as $\exp[\text{const}(\Gamma/U)]$ and so do the magnetization and effective magnetic moment.

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