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ON A CRITERION FOR ANTIFERROMAGNETISM, CONNECTED WITH THE METAL-INSULATOR PHASE TRANSITION

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I. Introduction

It is well-known from experiments[1], that some of transition-metal oxides are metals at high temperatures, and insulators in the ground-state. Between them, a phase transition exists at a critical temperature. In several oxides, this transition point is connected with the Neel-point T_N , and the materials show antiferromagnetism (af) in the insulating phase. But also the possibility of non-magnetic insulators exists in the ground-state. Hereby, the transition is connected with a lattice distortion (ℓd). Both cases, af and ℓd are connected with a gap Δ which divides half-filled metallic bands in a completely filled subband and an empty subband. So the material becomes an insulator[2]. Now, the problem of this paper is to find a quantitative criterion for that, which oxides show af and which ℓd in the ground-state.

2. Model

Mattis and Langer[3] have discussed the role of the electron-photon interaction in the metal-insulator phase transition. Basing on their work, a study of the electron-spin interaction is possible. The model Hamiltonian has the form

$$H=\sum_{ks}(\epsilon_k-\mu)\,c^+_{ks}c_{ks}+\hbar\omega\sum_q a^+_q a_q+\frac{g}{\sqrt{N}}\sum_{kqs}c^+_{k+qs}c_{ks}(a_q+a^+_{-q})- \qquad (1)$$

$$-\frac{J}{2N}\sum_{nkq}e^{-iqR_n}\,[S^z_n(c^+_{k+q\uparrow}c_{k\uparrow}-c^+_{k+q\downarrow}c_{k\downarrow})+S^+_n c^+_{k+q\downarrow}c_{k\uparrow}+S^-_n c^+_{k+q\uparrow}c_{k\downarrow}]\,.$$

c^+_{ks} , a^+_q and S are the operators of electrons, phonons, and spins, resp. The first three terms are the Hamiltonian of Mattis and Langer, the last terms describe the electron-spin interaction in a usual form.

For crystals with cubic symmetry (sc and bcc), realized in several transition-metal oxides, the band energy $\epsilon(k)$ has the property $\epsilon(k+Q)=-\epsilon(k)$ if Q/π is a reciprocal lattice vector and

$$\delta_n \equiv \exp(iQR_n)=\pm 1 \qquad (2)$$

for R_n in a (fictive) sublattice A or B . For half-filled bands ($\epsilon_F=0$), Q transforms one point of the Fermi surface into another one. Therefore in the sum over q the transition impulse Q is the most important term, and all other's are only small perturbations. This gives the unperturbed Hamiltonian H^0 , in which the term Q replaces the sum over q .

The phonon part (with $J=0$ in (1)) is solved by Mattis and Langer, assuming macroscopic expectation values for a^+_Q and setting $a_Q = a^+_Q = -x\sqrt{N}$ with the variational parameter x . Analogous to this, here the contribution of the spin term of (1) is considered, with $x=0$ and $J \neq 0$. So, the unpertubed Hamiltonian has the form

$$H^0 = \sum_{ks} (\epsilon_k - \mu) c^+_{ks} c_{ks} - \frac{J}{2N} \sum_{nk} \delta_n [S^z_n (c^+_{k+Q\uparrow} c_{k\uparrow} - c^+_{k+Q\downarrow} c_{k\downarrow}) + \tag{3}$$

$$+ S^+_n c^+_{k+Q\downarrow} c_{k\uparrow} + S_n c^+_{k+Q\uparrow} c_{k\downarrow}] .$$

3. Free Energy

We use the molecular-field approximation[4], in which H^0 is divided into an electron part H_{el} and a spin part H_S . In H_{el} S is substituted by

$$< S^z_n > = \delta_n |< S^z_n >| \equiv \delta_n SM, \quad < S^+_n > = < S^-_n > = 0 , \tag{4}$$

with the variational parameter M . It follows

$$H_{el} = \sum_{ks} (\epsilon_k - \mu) c^+_{ks} c_{ks} - \frac{1}{2} JMS \, \Sigma_k (c^+_{k+Q\uparrow} c_{k\uparrow} - c^+_{k+Q\downarrow} c_{k\downarrow}), \tag{5}$$

In H_s the spin-polarization of electrons is substituted by

$$< c^+_{k+Q\uparrow} c_{k\uparrow} - c^+_{k+Q\downarrow} c_{k\downarrow} > \equiv \delta_n \sigma, \quad < c^+_{k+Q\downarrow} c_{k\uparrow} > = < c^+_{k+Q\uparrow} c_{k\downarrow} > = 0, \tag{6}$$

yielding

$$H_S = -\frac{1}{2} J \sigma \Sigma_n \delta_n S_n^z . \tag{7}$$

σ is a second variational parameter.

Now, the Hamiltonian is decoupled, and also the free energy. But by calculation of F we must consider, that the electron-spin interaction appears twofoldly, in H_{el} and in H_S. So a correction term is necessary and we have

$$F = F_{el} + F_S + \frac{1}{2} N J S M \sigma . \tag{8}$$

Using a canonical transformation, H_{el} can be diagonalized and it follows

$$F_{el} = \frac{N}{\beta} \int d\epsilon \, \rho(\epsilon) \, \ln [f(\mu - \sqrt{\epsilon^2 + \Delta^2}) f(\mu + \sqrt{\epsilon^2 + \Delta^2})] \tag{9}$$

with the energy gap $\Delta \equiv \frac{1}{2} JMS$, dividing the band $\epsilon(\mathbf{k})$ in the mentioned two subbands, and $f(x) \equiv (1 + \exp \beta x)^{-1}$. For F_S follows in a simple manner

$$F_S = -\frac{N}{\beta} \ln [sh(\frac{2S+1}{4} \beta J \sigma) / sh (\frac{1}{4} \beta J \sigma)] . \tag{10}$$

4. Antiferromagnetism

We have in F three parameters: μ , σ and Δ (or M). μ is given by the electron density $\bar{n}$ with $\bar{n} = 1$ for half-filled bands. σ and Δ are given by minimalization of F to

$$\Delta = \frac{1}{2} JSB_s(\frac{1}{2}\beta JS\sigma) \tag{11}$$

(with Brillouin's function $B_S(x)$) and

$$\sigma = \Delta \int \frac{d\epsilon\ \rho(\epsilon)}{\sqrt{\epsilon^2+\Delta^2}} [f(-\sqrt{\epsilon^2+\Delta^2}-\mu)-f(\sqrt{\epsilon^2+\Delta^2}-\mu)]. \tag{12}$$

In a simple approximation we use (see e.g.[3])

$$\rho(\epsilon) = 1/2w \text{ for } -w<\epsilon<w, \tag{13}$$

with the band width $2w$ and $\rho = 0$ outside the band. For $\bar{n} = 1$ we can eliminate μ and the electron polarization σ in eqs. (11) and (12) and obtain the transzendent equation

$$\Delta = \frac{1}{2} JSB_s[\frac{\beta JS\Delta}{2w} \int_0^w \frac{d\epsilon}{\sqrt{\epsilon^2+\Delta^2}} \text{th}(\frac{\beta}{2}\sqrt{\epsilon^2+\Delta^2})] \tag{14}$$

for Δ only. (14) has the trivial solution $\Delta = 0$ for all T , corresponding to the paramagnetic case. In the low-temperature region a nontrivial solution exists with $\Delta(T) \neq 0$, depending on T . For

$T = 0$ follows $\Delta(0) = \frac{1}{2} JS$. This second solution corresponds to af and is the thermodynamic stable solution. The solution $\Delta \neq 0$ disappears at a second-order phase transition point T_N, following from

$$\frac{1}{2}\beta_N w \int_0^{\frac{1}{2}\beta_N w} dx \frac{\text{th}\, x}{x} = \frac{6 w^2}{S(S+1)J^2}, \quad \beta_N^{-1} = k_B T_N \approx \sqrt{\frac{6S}{S+1}}\, \Delta(0) . \qquad (15)$$

The energy difference at $T=0$ between af and the paramagnetic state is

$$\delta E_{af} = -\frac{N}{2}\left[\sqrt{w^2 + \Delta^2} - w + \frac{\Delta^2}{w} \text{Ar sh}\left(\frac{w}{\Delta}\right)\right] < 0 , \qquad (16)$$

so the ground-state is antiferromagnetic. For $T > T_N$, af disappears, and at the same temperature T_N we have the insulator-metal transition $(\Delta \to 0)$.

5. Comparison

Now we can compare with the result, given by Mattis and Langer for ℓd. We consider

$$a \equiv |\delta E_{\ell d}| / |\delta E_{af}| . \qquad (17)$$

$\delta E_{\ell d}$ is the analogic energy difference between distorted and nondistorted lattice /3/. For $a < 1$ af is the stable ground-state, for $a > 1$ the ground-state is the paramagnetic case with ℓd. Therefore, the ground-state is antiferromagnetic, if

$$\frac{g^2}{\hbar\omega} < \frac{w}{2} \{ \operatorname{Arcth} [\sqrt{1+(\frac{JS}{2w})^2} + (\frac{JS}{2w})^2 \operatorname{Arsh} (\frac{2w}{JS})] \}^{-1} \approx \frac{JS}{2} , \quad (18)$$

and nonmagnetic in the other case. On the left-hand side of (18) we have the phonon parameters g and $\hbar\omega$, on the right-hand side the spin parameters J and S and the band width $2w$. With (18) a theoretical classification of the transition-metal oxides is possible in two groups:

i) oxides with a distorted nonmagnetic insulating ground-state, and

(ii) oxides with a nondistorted antiferromagnetic ground-state. A comparison of this theoretical argument with experiments is not possible at present, since not all of the parameters, necessary in (18), are known exactly.

By the way, the above considered model possesses no ground-state with both af and ℓd, in agreement with the experiments. Minimalizing the corresponding free energy one obtains no solution with simultaneous nonvanishing variational parameters M, σ and x.

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