ON THE FEASIBILITY OF ORBITAL MAGNETISM

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Submitted 5 January 1970
ZhETF Pis. Red. 11, No. 3, 206 - 209 (5 February 1970)

The interaction of localized atomic electrons is usually considered with the s-shell electrons as an example. The Hamiltonian of the exchange interaction is expressed in this case in terms of the electron spin operators in the well-known manner:

$$\mathcal{H}_{H-L} = I\mathbf{S}_1\mathbf{S}_2. \tag{1}$$

Formula (1) is the basis of the Heitler-London theory of the chemical bond, and leads on the other hand to the spin-magnetism theory associated with Heisenberg.

There are, however, possible situations in which the exchange interaction couples electrons having nonzero orbital angular momenta.

It is usually assumed that the orbital angular momenta are quenched in a crystal. However, if the crystal fields acting on the electrons are weak compared with the exchange energy, then the angular momentum of the electron is weakly coupled to the lattice. The spin-orbit interaction is also assumed to be weak compared with the exchange interaction.

To describe the latter we use Hubbard's model Hamiltonian [1]

$$\mathcal{H}_H = v\sum_{\mathbf{r}\mathbf{a}m\sigma} a^+_{\mathbf{r}m\sigma}\, a_{\mathbf{r}+\mathbf{a}m\sigma} + V\sum_{\mathbf{r}}\frac{1}{2}\Big(\sum_{m\sigma} n_{\mathbf{r}m\sigma}\Big)\Big(\sum_{m\sigma} n_{\mathbf{r}m\sigma} - 1\Big) \tag{2}$$

$a^+_{\mathbf{r}m\sigma}$ ($a_{\mathbf{r}m\sigma}$) are the operators of creation (annihilation) of an electron at the site $\vec{\mathbf{r}}$ with momentum projection m and spin projection σ (for concreteness, we consider p-electrons). The second term in the Hamiltonian (2) describes the Coulomb repulsion of the electrons per atom. We assume that $v \ll U$ and regard the first term of (2) as a perturbation. We assume that the number of electrons coincides with the number of atoms. The ground state of the unperturbed Hamiltonian is strongly degenerate. It contains, for each atom, one electron with arbitrary projections of the angular momentum and spin. The perturbation lifts the degeneracy, so that in second order of perturbation theory we can obtain an effective Hamiltonian in the form

$$\mathcal{H}_{\text{eff}} = I\sum_{\mathbf{r}\mathbf{a}}\{[(\mathbf{L}_\mathbf{r}\mathbf{L}_{\mathbf{r}+\mathbf{a}})^2 + (\mathbf{L}_\mathbf{r}\mathbf{L}_{\mathbf{r}+\mathbf{a}}) - 2] +$$

$$+ (2\mathbf{S}_\mathbf{r}\mathbf{S}_{\mathbf{r}+\mathbf{a}} - \frac{1}{2})[(\mathbf{L}_\mathbf{r}\mathbf{L}_{\mathbf{r}+\mathbf{a}})^2 + (\mathbf{L}_\mathbf{r}\mathbf{L}_{\mathbf{r}+\mathbf{a}}) - 1]\}, \tag{3}$$

$$\left(I = \frac{v^2}{V}\right),$$

which depends on the spin and orbital-momentum operators of the electrons at each site. A similar calculation for the s-electrons leads to the usual Heisenberg Hamiltonian (cf., e.g., [2, 3].

The total orbital angular momentum, the spin, and their projections are conserved in the system described by the Hamiltonian $\mathcal{H}_{\text{eff}}$. A rigorous construction of the ground state is just

as difficult as in the Heisenberg model of an antiferromagnet. We shall therefore consider the simplest possibilities of configurations of systems of orbital angular momenta and spins. Let, for example, the spin system be ferromagnetic. Then it follows from (3) that the orbital angular momenta form an antiferromagnetic system. Putting $\langle \vec{L}_0 \vec{L}_a \rangle \simeq -1$, we obtain for the energy of the state per atom $E_1 \simeq 2Iz$ (z is the number of nearest neighbors). Let us consider another state in which $\vec{L}$ form an antiferromagnetic system. Then the spin system is antiferromagnetic. The energy of such a state per atom is $E_2 \simeq -Iz$. Thus, within the framework of the model under consideration, the former state is energetically favored. The described system is similar in its magnetic properties to a ferromagnet (it has a large spontaneous moment). The neutron-diffraction pattern, however, is very similar to that obtained in an antiferromagnet. Namely, in the case of sufficiently strong magnetic scattering, the neutron reflections will be twice as frequent as the x-ray reflections.

There are no grounds for assuming that the spin and orbital orders are destroyed at the same temperature. Rough estimates show that the spin order is more likely to be destroyed at a lower temperature than the orbital order. This means that the system experiences a transition from the state described by us into an antiferromagnetic state, where the orbital angular momenta are ordered, and then into a paramagnetic state.

The actual picture may turn out to be more complicated, since our estimates are only preliminary. We wish to emphasize that a model using p-electrons is not realistic, and the phenomenon described by us can be realized on d- or f-shells. The calculations for these cases are much more complicated, but the qualitative picture remains the same.

Experiments aimed at detecting orbital magnetism should be performed either with poor metals having a narrow conduction band, or in ferrodielectric compounds with large transition temperatures.

We are grateful to I. E. Dzyaloshinskii and A. P. Kazantsev for a useful discussion.

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