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INDETERMINACY OF MULTIPLICITY (IM) AND PSEUDOMULTIPLICITY (IPM) OF CRYSTALS WITH MAGNETIC AND OTHER ORDER

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Résumé. — La réalisation d’un état antiferromagnétique colinéaire (afm) nécessite IM. On ne peut alors pas expliquer l’afm par un hamiltonien d’Heisenberg isotrope. Un hamiltonien anisotrope (commutant avec l’opérateur renversement du temps) ne permet pas d’obtenir un état afm dans un état non dégénéré de spin entier. Pour stabiliser l’afm, il est nécessaire d’introduire un hamiltonien impair par rapport au renversement du temps. IM est aussi nécessaire pour stabiliser des structures magnétiques non colinéaires. Pour les structures héliocoldales IM dépend de l’angle de l’hélice. La théorie quantique du magnétisme est généralisée aux systèmes électroniques non magnétiques en introduisant un pseudospin lié à la population des niveaux électroniques. IPM est important pour les états normaux et supraconducteurs. Le niveau de Fermi étant I dans le premier état IPM est égal à la racine carrée du produit du nombre d’électrons et de trous dans la bande d’énergie. IPM devient 0 dans le cas d’un diélectrique à bande pleine, pseudoferromagnétisme (pfm). On rencontre un IPM maximum dans les métaux alcalins ayant une bande à moitié occupée (deux domaines pfm). L’état supraconducteur correspond à une paroi de Bloch entre deux pseudodomains. La méthode du champ pseudomagnétique est équivalente à la théorie de Bardeen-Cooper-Schrieffer. Le rejet de l’approximation du champ pseudomoléculaire conduit à une généralisation de la théorie BCS.

Abstract. — The realization of colinear antiferromagnetism (afm) requires IM. Therefore afm cannot be explained by an isotropic Heisenberg Hamiltonian. With an anisotropic Hamiltonian (commuting with the time reversal operator) afm cannot be realized in a non degenerate state of integral spin. For stabilization of afm it is necessary to introduce a Hamiltonian that is odd with respect to time reversal. IM is also necessary for the stabilisation of non colinear magnetic structures. For spiral structures IM depends on the helical angle. The quantum theory of magnetism is generalized for non magnetic electron systems by introducing a pseudospin related to the population of electron levels, IPM is important for normal and superconducting states. The Fermi index being I in the former state, IPM is equal to the square root of the product of the number of electrons and holes in the energy band. IPM becomes 0 in the case of a dielectric with a full band, pseudoferromagnetism (pfm). Maximum IPM occurs in alkali metals with a half occupied band (two-domain pfm). The superconducting state corresponds to a Bloch wall between two pseudodomains. Therefore the pseudomagnetic field method is equivalent to the theory of Bardeen-Cooper-Schrieffer. Rejection of the pseudomolecular field approximation leads to generalization of the BCS theory.

1. In earlier papers we have considered the properties of the electronic system of a crystal associated with a change in the separate ion or conduction electron pair multiplicity. It was established that in many important cases (minimum in the electrical resistance of alloys containing impurities; the lowering of the critical temperature of the superconducting transition in La when impurities of rare earth elements are added; the paramagnetic susceptibilities of superconductors; the coexistence of superconductivity and fm or afm and so on) it is essential to use the generalized Hamiltonian for s = \delta(\mathcal{A}) direct and indirect exchange coupling and the interactions of conduction electrons, taking into account the change of ion or electron pair multiplicity [1]. In this report we have considered the properties of the electronic system of a crystal, that are connected with IM of the crystal as a whole.

2. The IM of a crystal as a whole is essential for realization of magnetic ordering different from colinear fm. First we consider the problem of stabilization of the afm. In the case of a crystal with two sublattices I and II the total spin-vector operator \( \vec{S} \) is determined by the sum

\[ \vec{S} = \vec{S}_I + \vec{S}_II, \]

where \( \vec{S}_I \) and \( \vec{S}_II \) are the spin-vector operators of the I and II sublattices. The afm state is characterized by simultaneous satisfaction of the following relations:

\[ < \vec{S}_z > = 0, \quad < S_{Iz} > \neq 0, \quad < S_{IIz} > \neq 0. \]  

If we introduce the antisymmetric operator \( \mathcal{A} = \vec{S}_I - \vec{S}_II \),

then it follows from (1) and (4):

\[ \vec{S}_I = \frac{1}{2} (\vec{S} + \mathcal{A}), \quad \vec{S}_II = \frac{1}{2} (\vec{S} - \mathcal{A}). \]

For the mean value of the z-projection according to (2) we have

\[ < \vec{S}_{Iz} > = \frac{1}{2} < \mathcal{A}_z >, \quad < \vec{S}_{IIz} > = - \frac{1}{2} < \mathcal{A}_z >. \]

From (6) and (3) it follows that the necessary condition for the realization of the afm state is [2]:

\[ < \mathcal{A}_z > \neq 0. \]

In the states where \( \vec{S} \) and \( \vec{S}_I \) have definite values the following equality holds:

\[ < S, S_z | \mathcal{A}_z | S, S_z > = A_z^s S_z, \]

where \( A_z^s \) is some constant. We see from (2) and (8) that \( < \mathcal{A}_z > = 0 \), thus violating condition (7). Therefore, it is impossible to realize afm in a state with definite multiplicity and definite value of the z-projection of the total crystal spin \( S_z \). Hence an exchange isotropic Heisenberg Hamiltonian possessing (at positive exchange integral) a singlet ground state, with the definite values \( S = 0 \) and \( S_z = 0 \), cannot describe the afm. The necessary condition (7) is violated also at definite values of \( \vec{S} \) and indefinite values of \( \vec{S}_I \) for a system of any even number of sites with identical spins. In this case the expansion of the state \( | S > \)
in the states $|S, S_z\rangle$ gives us the equality:

$$<S | \hat{A}_x | S\rangle = 0.$$  \hspace{1cm} (9)

From a comparison of (9) with (7) it follows that in a system made up of even number of sites with identical spins afm is possible only in states having an IM.

We have also proved a more general theorem [2]:

if the Hamiltonian of the system commutes with the time-reversal operator $\hat{\Theta}$ then $<A_x> = 0$ in any degenerate state with integral spin. Hence, for stabilization of a non degenerate afm state, it is necessary to have the Hamiltonian odd with respect to time reversal (\textsuperscript{*)}). This theorem must be taken into account in introducing the anisotropic Hamiltonian for stabilizing afm. For example, the Hamiltonian for the magnetic anisotropy energy with axial symmetry [3]:

$$\hat{H}_{1A} = K \sum_{j,k} (\hat{S}_{kj}^2 + \hat{S}_{kj}^2) + \sum_{k} (\hat{S}_{k}^2 + \hat{S}_{k}^2),$$  \hspace{1cm} (10)

(where $j, k$ are the site numbers of sublattices I and II and $K$ is the anisotropy constant) commutes with the time reversal operator $\hat{\Theta}$, and therefore cannot stabilize the afm. The opposite conclusion obtained in paper [3] is due to the main assumption:

$$\hat{S}_{kj} \approx S_z - (2 S_z)^{-1} (\hat{S}_{kj}^2 + \hat{S}_{kj}^2),$$

$$\hat{S}_{kj} \approx S_z + (2 S_z)^{-1} (\hat{S}_{kj}^2 + \hat{S}_{kj}^2),$$

which changes the character of the symmetry of $\hat{H}_{1A}$ with respect to time reversal. On the other hand the Hamiltonian was used by Ziman [4]:

$$\hat{H}_{1A} = \sum_{k} g_{\mu_B} \hat{S}_{k} H_A \exp(iwR_k) = pH_x,$$  \hspace{1cm} (12)

(where $p = |g\mu_B H_A \exp(iwR_k)|$ is odd with respect to $\hat{\Theta}$ and therefore can stabilize the afm). If $\hat{H}_{1A}$ is the total Hamiltonian the ground state corresponds to two Néel sublattices and IM is equal to the maximum possible value of the crystal spin.

3. IM is also necessary for noncolinear magnetic structures. In the state of weak fm described by the Hamiltonian:

$$\hat{H}_{1lm} = I(\hat{S}_I \hat{S}_M) + D[\hat{S}_I \hat{S}_M] + pH_x,$$  \hspace{1cm} (13)

we have for mean values of the spin projections [5]:

$$<\hat{S}_x> = \frac{pD_x}{F}, <\hat{S}_y> = -\frac{pD_y}{F}, <\hat{S}_z> = 0,$$  \hspace{1cm} (14)

where

$$F = \frac{1}{2} (I^2 + D^2) + 2 p^2 + I \left(\frac{I^2 + D^2}{4} + p^2\right) \frac{1}{2}.$$

From (14) it is clear that the nonzero spin projection occurs only in case, the parameters $p$ and $D$ both have nonzero values. Therefore for realization of weak fm besides the Dzyaloshinsky Hamiltonian ($D \neq 0$) the term $pH_x$, odd with respect to time reversal $\hat{\Theta}$ is also necessary. The IM is determined by the relation

$$\Delta S^2 = \left(1 + \frac{I^2}{D^2 + 4 p^2}\right)^{-1}.$$  \hspace{1cm} (15)

\textsuperscript{(*)} As is evident from paper [7] the presence in the Hamiltonian of terms odd with respect to the operator $\hat{H}$ (Zeeman terms) is of vital importance also for the stabilization of fm.

From (15) it follows that only in the limiting case $I^2 > D^2 + 4 p^2$ does the IM approaches zero. In the general case $\Delta S^2 \neq 0$, when $D^2 + 4 p^2 > I^2$ the IM approaches the maximum possible value of one in the system of two spins ($S_z = S_M = 1/2$).

4. In the case of spiral magnetic structures the IM depends on the helical angle $\varphi$. For example, in the case of two lattice sites when $\varphi$ increases from $0$ to $\pi$, IM monotonically increases from a zero value typical of single domain fm to a maximum value of unity, typical of afm ordering of two spins. In the case of many lattice sites ($N > 2$) oscillations of the nonzero IM take place due to the possibility of representing some spiral structures as the aggregate of afm sublattices turned against each other. Therefore, the IM oscillates about the value $N/2$ typical of Néel afm. The oscillations in the interval $0 < \varphi < 2 \pi$ have $N = 2$ maxima and minima, and a gigantic peak of the IM takes place near the fm state ($\varphi \sim 0$) with a value of the order of $0.09 N^{3/2}$.

Therefore IM is essential for realization of noncolinear and colinear magnetic structures different from colinear fm.

5. The quantum theory of magnetism is generalized for crystal conduction electrons by introduction of pseudospins $S_k^i$ associated with the population $n_{ek}$ of electron energy levels $k, \sigma$ by the relation

$$n_{ek} = \frac{1}{2} - S_k^i.$$  \hspace{1cm} (16)

The IPM is important for the normal and superconducting states. In the normal state with the Fermi index equal to unity the IPM of electrons in the energy band is determined by the relation

$$\Delta S^2 = (N_e N_h) \frac{1}{2},$$  \hspace{1cm} (17)

where $N_e$ and $N_h$ represent the number of electrons and holes respectively in the energy band. The IPM becomes equal to zero according to (17) in the case of a dielectric with a full band ($N_h = 0$). Taking into account that in this case, according to (16), all $S_k^i$ have values equal to $-1/2$, the dielectric may be considered as the pseudo fm. The maximum of IPM, which equal to $(N_e + N_h)/2$ occurs in the case of alkali metals with half occupied bands. If we take into account that in this latter case $N_e = N_h = (N_e + N_h)/2$ and $S^e = \sum_k S_k^e = 0$ (because for all levels under the Fermi surface $S_k^e = -1/2$ and for all levels above is $S_k^e = 1/2$) we can consider it as two pseudo-fm domains with zero projection of total pseudospin. The superconducting state corresponds to a Bloch wall between these domains. In such case the method of pseudomolecular field [6] is equivalent to the theory of BCS. The rejection of the pseudomolecular field approximation leads to the following expression for the BCS Hamiltonian

$$\hat{H} = \frac{1}{2} \sum_k V + 2 \sum_{k>k'} e_k - \sum_k e_k S_k^e - VS^2,$$  \hspace{1cm} (18)

where $V$ is the parameter of mutual electron interaction, $k$ is the wave vector, $e_k$ is the conduction electron kinetic energy, and $S$ is the total pseudospin of the electron system in the range of effectiveness of the mutual electron interaction. The analysis of the pro-
properties of the Hamiltonian in equation (18) leads to the following generalizations for the BCS theory [8]:

1) In analogy with the quantum theory of magnetism, where spin ordering occurs both with positive and negative exchange integrals, the ordering in a system of conduction electrons associated with appearance of an energy gap in the energy spectrum of elementary excitations may take place both in the case of mutual electron attraction and repulsion.

2) In the case of mutual electron attraction, in the superposition of states with different values of the total pseudospin the role of states with greater values of total pseudospin is increased in comparison with the normal state. In the case of electron repulsion, the role of states with smaller values of the total pseudospin is increased. Therefore, in the repulsion case, the starting state for constructing the wave function of the system in the range of effectiveness of mutual electron interaction is the pseudo afm state, i.e. the state with $S = 0$ and $S' = 0$.

6. Thus, the theory developed above shows that the IM of the electron system of a crystal as a whole is quite essential for magnetic ordering phenomena. Generalization of the results of the quantum theory of magnetism to conduction electron systems in crystals through introduction of pseudospins is a fruitful method for study of properties in both normal and superconducting states.

References