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Journal of Physics and Chemistry of Solids 66 (2005) 1412–1416
JOURNAL OF PHYSICS AND CHEMISTRY OF SOLIDS
www.elsevier.com/locate/jpcs

Ring exchange and correlated fermions

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Abstract

The concept of exchange in strongly-correlated fermions is reviewed with emphasis on the generalization of the Heisenberg pair exchange to higher order *n*-particle permutations. The 'frustration' resulting from competing ferromagnetic three-spin exchange and antiferromagnetic two- and four-spin exchanges is illustrated on a two-dimensional model system: solid ^{3}He films. Recent experimental results proving the presence of significant four-spin exchange interactions in the CuO_2 plaquettes of high T_c cuprates are reported.

Keywords: Quantum spin systems; ^{3}He; Cuprates.

1. Historical introduction

The concept of exchange interactions in almost-localized correlated fermions first appeared in the pioneering papers of Heisenberg [1] and was formulated in a more general mathematical way by Dirac [2,3]. Although the early papers by Dirac already contain the general expression of the Hamiltonian in terms of n-particle permutations, no attention was paid, up to the sixties, to higher orders than the 'pair-exchange' Heisenberg term. Thouless [4] was the first to point out that higher order exchanges as three- and four-spin cyclic permutations might be important in quantum solids like ^{3}He. But only ten years later, striking experimental results on nuclear magnetism in the bcc phase of solid ^{3}He, in the millikelvin range [5–7], were interpreted by Hetherington, Delrieu and Roger through a ring-exchange model with two-three-and four-particle interactions of comparable magnitude [8–10]. From general considerations put forward by Thouless, permutations of even parity (like three-particle cycles) induce ferromagnetism while odd permutations (pair and four-particle exchanges) favor antiferromagnetism, and the striking phase diagram of bcc solid ^{3}He corresponds to a highly frustrated quantum-spin system with competing three- and four-spin permutations. During the last two decades a lot of progress has been accomplished in the investigation of solid ^{3}He films adsorbed on graphite, a simple model system exhibiting even more frustration since the frustrated nature of ring-exchange interactions is enhanced by the frustrated geometry of the triangular lattice. The conceptual beauty of solid ^{3}He lies in the fact that the system is simple enough (the pair interactions between ^{3}He atoms are mainly of hard core nature) to allow the calculation of exchange frequencies from first principles [11] and a quantitative comparison with experimental results.

The relevance of the multi-spin exchange concept is not restricted to the physics of nuclear ^{3}He spins. Delrieu [12] suggested that three-spin exchange might be dominant in the two-dimensional electron Wigner solid near the quasi-classical limit, and this has been corroborated through WKB calculations [13]. More recent Monte-Carlo simulations have proved that competing three and four-spin exchange interaction should occur near melting [14].

The first fourth-order t/U expansion of the one-band Hubbard Hamiltonian in terms of four-spin interactions has been published in 1977 by Takahashi [15]. Soon after the discovery of high-T_c superconductors, Roger and Delrieu [16] suggested, on the basis of an expansion of the three-band Hubbard model, that four-spin exchange might be significant in the CuO_2 planes of cuprates. During the last ten years many experimental results have revealed the presence of four-spin exchange interactions in the Cu–O plaquettes of cuprates [17–19] and copper-based spin-ladder materials [20–23].

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doi:10.1016/j.jpcs.2005.05.065

2. Dirac formalism with illustration on the one-band Hubbard model

Dirac formalism is introduced in the framework of degenerate perturbation theory. The Hamiltonian is written $H=H_0+V$, where H_0 describes independent particles and V is a perturbation. The 'unperturbed' degenerate ground-states for N distinguishable particles can be written as products of independent particle states:

$$|\psi\rangle = |\alpha_1\rangle^{(1)}|\alpha_2\rangle^{(2)}|\alpha_3\rangle^{(3)}\cdots|\alpha_N\rangle^{(N)} \tag{1}$$

which means that the particle numbered (i) is in a state $|\alpha_i\rangle$. Each of these states is itself a product of two kets corresponding to the orbital and spin variables respectively: $|\alpha_i\rangle=|R_i\rangle\sigma_i\rangle$

$$|\psi\rangle = |R_1\rangle^{(1)}|\sigma_1\rangle^{(1)}\cdots|R_n\rangle^{(N)}|\sigma_N\rangle^{(N)} \tag{2}$$

Hence $|\psi\rangle=|\phi^R\rangle|\xi^\sigma\rangle$ appears as a product of an orbital wavefunction:

$$|\phi^R\rangle = |R_1\rangle^{(1)}|R_2\rangle^{(2)}\cdots|R_N\rangle^{(N)} \tag{3}$$

and a spin wavefunction:

$$|\xi^\sigma\rangle = |\sigma_1\rangle^{(1)}|\sigma_2\rangle^{(2)}\cdots|\sigma_N\rangle^{(N)} \tag{4}$$

Note that for the half-filled Hubbard model, $|R_i\rangle$ simply represents the site occupied by the particle (i). The permutation P of two particles can be expressed as a product of two operators:

$$P = P^R P^\sigma \tag{5}$$

P^R acting on orbitals and P^σ acting on spin variables. If the Hamiltonian does not depend explicitly on the spin, we can as a first step concentrate on the orbital part of the wave function and solve the eigenvalue problem:

$$H|\phi^R\rangle = E|\phi^R\rangle \tag{6}$$

for the orbital wavefunction $|\phi^R\rangle$ describing N *distinguishable* particles. The ground-state of the unperturbed part H_0 of the Hamiltonian is N! fold degenerated and the corresponding subspace Ω_0 is spanned by the N! states:

$$P^R|\phi^R\rangle = |R_{\nu_1}\rangle^{(1)}|R_{\nu_2}\rangle^{(2)}\cdots|R_{\nu_N}\rangle^{(N)} \tag{7}$$

where $\{\nu_1, \nu_2,\ldots, \nu_N\}$ represents a permutation P of the N integers $\{1, 2,\ldots,N\}$. We now apply degenerate perturbation theory [25,26] to the perturbed Hamiltonian $H=H_0+V$. At first order, the splitting of the $N!$ degenerated energy levels is given by the eigenvalues of the Hamiltonian $V^{(1)}$ defined by its matrix elements:

$$V^{(1)}_{a,b} = \langle\phi^R|P^R_a V P^R_b|\phi^R\rangle \tag{8}$$

where P^R_a and P^R_b are two permutations of the symmetric group S_N. Since V is invariant with respect to any permutation, we can write:

$$V^{(1)}_{a,b} = \langle\phi^R|VP^R_aP^R_b|\phi^R\rangle = \langle\phi^R|VP^R|\phi^R\rangle = V^{(1)}_P$$

where $P=P^R_aP^R_b$, and the eigenvalue problem, restricted to the subspace Ω_0 can be formally represented by the Hamiltonian:

$$H^{(1)} = -\sum_{P^R\in S_N} V^{(1)}_P P^R \tag{9}$$

where the summation runs over permutations P^R of the symmetric group S_N. This result extends straightforwardly to higher order degenerate perturbation theory: the higher orders are expressed in terms powers of V and projection operator P_0 on Ω_0[25,26], and these operators commute with permutation operators. Hence, at arbitrary order in degenerate perturbation theory, we can write:

$$H \approx -\sum_{P^R\in S_N} V_P P^R \tag{10}$$

We now have to introduce the spin degrees of freedom an express that the global wavefunction is completely antisymmetric. As a general result from Group theory a completely antisymmetric wave function can be expressed by the following bilinear expression [27]:

$$|\psi\rangle = \sum_{\lambda,\mu} c_{\lambda,\mu}|\phi^R_\lambda\rangle|\xi^\sigma_\mu\rangle \tag{11}$$

where $|\phi^R_\lambda\rangle$ represents a linear combination of different permutations $P^R_a|\phi^R\rangle$ corresponding to a given irreducible representation of the symmetry group schematized by a Young diagram, while $|\xi^\sigma_\mu\rangle$ represents a linear combination of permutation $P^\sigma_a|\xi^\sigma\rangle$ corresponding to the representation associated with the 'complementary' Young diagram, obtained by exchanging the lines and the columns. For spin-1/2, a complete antisymmetrisation of the spins cannot be realised over more than 2 variables, hence the corresponding Young diagrams have at most two lines and each diagram corresponds to a given value of the total spin S. It is then possible to establish a correspondence between the expression (10) of the Hamiltonian acting only on the orbital variables with an equivalent Hamiltonian acting only on the spin variables. Expressing the antisymmetry of the wave function:

$$P|\psi\rangle = (-1)^p|\psi\rangle \tag{12}$$

where p is the parity of the permutation, we can write:

$$P^RP^\sigma|\psi\rangle = (-1)^p|\psi\rangle \tag{13}$$

and multiplying to the left by $(P^R)^{-1}$:

$$P^\sigma|\psi\rangle = (-1)^p(P^R)^{-1}|\psi\rangle \tag{14}$$

Taking into account that in Eq. (10) P^R and the inverse permutation $(P^R)^{-1}$ appear with the same weight V_P, the Hamiltonian is written equivalently in spin space:

$$H \approx -\sum_{P^\sigma\in S_N} (-1)^p V_P P^\sigma \tag{15}$$