A0009/77

ABR,1977

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SPIN POLARONS IN NARROW, ALMOST HALF-FILLED s BAND : A VALID CONCEPT IN b. c. c. ^3He ?

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77/14
Phys. Sol.

ABSTRACT :

In a narrow enough, half-filled band of interacting fermions, a hole creates a ferromagnetic spin polarization spreading over a large volume around it. We suggest this model might apply to the case of a vacancy in b. c. c. ^{3}He. This would provide a possible explanation of the susceptibility anomaly near the Néel Point.

Polarons de spin dans une bande s étroite, presqu'à moitié remplie : un concept valable dans ^{3}He cubique centré ?

RESUME :

Dans une bande suffisamment étroite et à moitié remplie de fermions en interaction, un trou crée une polarisation de spin ferromagnétique s'étendant sur un grand volume autour de lui. Nous suggérons que ce modèle puisse s'appliquer au cas d'une lacune dans ^{3}He cubique centré. Ceci fournirait une explication possible de l'anomalie de susceptibilité près du point de Néel.

I - INTRODUCTION -

In the first part of this paper (section 2 and 3) we study a crystal with very nearly one fermion per site, with hard core repulsive interaction U large compared with the overlap integral t between nearest neighbour one fermion wave functions. We neglect orbital degeneracy. The relevant Hamiltonian is the Hubbard Hamiltonian. For large $U/_t$ and one vacancy in a b. c. c. crystal, the ground state seems to be a spin polaron (sect. 3) ; the spin polaron is stable in a temperature range which can be large compared with the Néel temperature (sect. 4).

In the second part of this paper, we discuss the validity of this concept for b. c. c. ^3He and suggest that spin polarons were observed in recent susceptibility measurements of the system (1) (sect. 5).

2 - THE INFINITE U/t LIMIT.

Let us first examine the case of a single vacancy, at T = 0, in the limit $U/t \to \infty$. Then, configurations with two fermions on the same site are forbidden. Thus we are left with the problem of a vacancy propagating through the lattice by exchanging its place with a neighbouring fermion. The motion of the vacancy depends on the magnetic order. We know that in a b. c. c. lattice, the ground state is the fully ferromagnetic state (2). The physical reason is that any departure from the complete alignment of spins suppresses walks for the vacancy propagation and thus increases the kinetic energy of the vacancy. This partial localization is also reflected in the single particle density of states of the vacancy (2). In the ferromagnetic configuration, the vacancy band is the full band for non interacting particles, extending from - zt to + zt, where z is the number of nearest neighbours. For an antiferromagnetic spin configuration, the vacancy band is narrowed, now stretching from $- \omega_o zt$ to $+ \omega_o zt$ $(\omega_o = \frac{2\sqrt{z-1}}{z})$.
However a band tail extends up to the ferromagnetic band edge ; this is because a completely spin aligned cluster of N fermions $(S = N/2, S_z = 0)$ has a small, and rapidly decreasing with N, overlap with the up and down "antiferromagnetic"state of these spins. In these large ferromagnetic clusters the kinetic energy of the vacancy is decreased, leading to the exponentially small band tail mentionned above (2).

The mobility of the vacancy (of a hole in a Mott insulator in ref. (2)) has been studied : whereas it is infinite in the ferromagnetic case, it is reduced to that of a Brownian particle for states above the pseudo band edge in the antiferromagnetic case (2). This is also the case in the paramagnetic phase.

3 - THE SPIN POLARON STATE.

We will consider the problem of one vacancy at $T = 0$, with a large but finite value of U/t. Virtual transitions to states with double occupancy of lattice sites introduce superexchange terms in the hamiltonian. This affects the states above the pseudo band edge only slightly (3). However, states in the tail are profoundly changed.

We now look for the ground state of the system which would be the antiferromagnetic state, stabilized by the Anderson superexchange in the absence of the vacancy. In the case of one vacancy and finite U/t, Nagaoka's theorem (2) states that it is not the ferromagnetic state, and we show hereafter that it is not either the antiferromagnetic state.

Following Mott (4), we assume that the vacancy, in order to decrease its kinetic energy creates a ferromagnetic polarization of the spins in a sphere of radius R around it. R would be infinite for $t/U = 0$. Now, the ferromagnetic polarization costs an exchange energy proportionnal to the volume of the sphere. The vacancy energy is written as :

$$E(R) \cong - zt + \pi^2 \, t \left(\frac{a}{R}\right)^2 + \frac{4\pi}{3} \left(\frac{R}{a}\right)^3 \frac{zt^2}{U} \qquad (1)$$

where a is the lattice spacing.

In (1), the first term is the lowest vacancy energy in a ferromagnetic medium ; the second one is the energy necessary to localize the vacancy inside the ferromagnetic sphere, and the third one is the exchange energy cost of the polarization. Minimizing E(R) with respect to R, we obtain the minimum energy of the vacancy :

$$E_o \cong - zt \left(1 - \frac{5\pi^2}{3z} \left(2 \frac{zt}{\pi U}\right)^{2/5} \right) \qquad (2)$$

corresponding to a number of aligned spins

$$N = \frac{4\pi}{3} \left(\frac{\pi U}{2zt} \right)^{3/5} = \frac{4\pi}{3} \left(\frac{R_o}{a}\right)^3 \qquad (3)$$

E_o is a variational approximation for the ground state energy. In this state, the vacancy is dressed with a ferromagnetic polarization over a large volume, forming a spin polaron. We call it a "strong" spin polaron, by contrast with "weak" spin polarons, where the magnetization per site within the spin polaron is small compared with the nuclear magneton (5).

A polaron of radius R contributes to the vacancy density of states a term proportional to $\frac{dR}{dE}$. Thus, this density of states exhibits an infinite peak at E_o. For smaller values of $\frac{U}{t}$, it may happen that E_o exceeds the antiferromagnetic band edge energy. The critical value $(\frac{U}{t})_c$ is given by :

$$E_o\left((\tfrac{U}{t})_c\right) = -2\sqrt{z-1}\,t$$

Then the vacancy does not stabilize a strong magnetic polaron, which would decay by Brownian motion of the vacancy. We have $(\frac{U}{t})_c = 460$ for $z = 8$ and $(\frac{U}{t})_c = 1460$ for $z = 6$. A large number of nearest neighbours is favourable to the spin polaron formation.

Remarks

a) We only considered spherical polarons. Non spherical geometries should lead to slightly different kinetic energy. This should probably give a polaron peak with finite width and finite amplitude.

b) We have assumed that the spin polarization was uniform inside the polaron, whereas it seems more plausible that $<S_z>$ decreases away from the polaron center. However, we think that $<S_z> = \frac{1}{2}$ over most of the polaron volume in strong polarons and decreases only near the surface ensuring a sharp frontier between the polaron and the antiferromagnetic medium, at least at $T = 0$ (3). Thus, this effect is not expected to change very much the radius and the energy of the polaron.

c) We have assumed the vacancy to be completely localized inside the ferromagnetic volume. In fact, for $\frac{U}{t} > (\frac{U}{t})_c$, the vacancy wave function decreases exponentially outside the polaron. This effect decreases the energy. $(\frac{U}{t})_c$ is lowered to about 160 (5).

d) If a lattice relaxation is allowed, the polaron energy is reduced again. We estimate that this decreases also $(\frac{U}{t})_c$ by about 20 % (5).

e) We expect the lifetime of the spin polaron to be large below $k_B T << \Delta$, where $\Delta(\frac{U}{t})$ is the energy difference between E_o and the pseudo band edge in the antiferromagnetic configuration.

f) The spin polaron discussed in this letter is qualitatively different from the spin polaron formed by an excess electron in an empty conduction band when it interacts through sf,sd or df exchange interaction with a set of localized ionic spins (4).The difference is analogous to that between itinerant electron magnetism in 3d metals and Rare Earth magnetism.

4- PROPERTIES OF THE SPIN POLARON

Whereas the bare vacancy is strongly scattered by the system of spins in the antiferromagnetic medium,it is free to propagate inside the polaron with an infinite mobility.The spin polaron state in the antiferromagnetic phase is also an eigenstate of the wave vector,in contradistinction to the bare vacancy state,but the polaron has a high effective mass m^* which may be estimated by calculating the matrix element between states where the center of the polaron is at site i and at site i + δ, nearest neighbour of i :

$$\frac{\hbar^2}{2m^* a^2} = t_{eff} = < i \mid H \mid i + \delta > = \xi^2 \ t$$

where $\xi = (\frac{1}{2})^{N_s/4} \sim e^{-0,2\, N_s}$ is the overlap of the ferromagnetic state of $\frac{N_s}{2}$ spins with the up-down "antiferromagnetic state" of these spins, N_s being the number of spins at the surface of the polaron. This gives a very low value for $\frac{m}{m^*}$. The result is a very low mobility (5) in the presence of a finite mean free path (phonons, impurities, spin fluctuations).

Let us now investigate the stability of the polaron at temperature $T > T_N$ (Néel point). The radius of the polaron R(T) and the magnetization of the spins inside the polaron $m(T) = 2 < S_z >$ are temperature dependent quantities. First consider low enough temperatures, so that $m(T) = 1 - \varepsilon(T)$ with $\varepsilon << 1$. The free energy of the polaron may be written, in the mean field approximation (3) :

$$F_p = -zt(1-\alpha\varepsilon) + \pi^2 \ t\left(\frac{a}{R}\right)^2 + \frac{4\pi}{3}\left(\frac{R}{a}\right)^3 m^2 \frac{zt^2}{4u} + \frac{4\pi}{3}\left(\frac{R}{a}\right)^3 kT\left(\mathrm{Log}\ 2 + \frac{\varepsilon}{2}\mathrm{Log}\frac{\varepsilon}{2} - \frac{\varepsilon}{2}\right) \quad (4)$$

Here we have used the fact that the band edge energy corresponding to a magnetization $1 - \varepsilon$ is $- zt(1 - \alpha\varepsilon)$ ($\alpha = 0,35$ for a s. c. lattice (3)). The last term in (4) is the entropy term. Minimizing with respect to R and ε , we get :

$$\mathrm{Log}\frac{\varepsilon}{2} \simeq - \frac{\alpha z t}{kT} \times \frac{3}{2\pi}\left(\frac{kT_N + 2kT\ \mathrm{Log}\ 2}{\pi t}\right)^{3/5}$$

$$\frac{R}{a} \simeq \left(\frac{\pi t}{kT_N + 2kT\ \mathrm{Log}\ 2}\right)^{1/5}$$

The physical meaning is clear: the spin polarization results from an effective magnetic field $g\mu_B h = 2\,\alpha zt/N$. Through the variation of R, h increases with T, so that the polarization remains large up to temperatures of the order of $kT_o = at$ ($a \simeq 1$ in a b.c.c. lattice).

We have to investigate the stability of the polaron with respect to the decay of the vacancy into the band of diffusive motion (with density of states $\rho(\omega)$). The condition of stability is

$$F_p < -kT \, \mathrm{Log} \int_{-\omega_o zt}^{\omega_o zt} \exp(-\omega/kT)\,\rho(\omega)\,d\omega \qquad (5)$$

The fulfilment of this condition is very much affected by the approximation made in the kinetic energy term. In a better treatment, taking into account the exponential decay of the vacancy wave function outside the polaron, this term may be reduced by a factor $b^2(T)$ ($1 < b(T) < 2$) (5). When this correction is taken into account, the condition (5) is always satisfied when $b < 2$.

With increasing T, the decrease of R(T) causes an increase of b(T). When b reaches the value 2, the potential well with depth $zt-\omega_o zt$ and radius R does not anymore bound a state. The polaron disappears. This happens at temperature $kT_I \simeq 0.6t$ (5).

5 - APPLICATION TO b. c. c. ^{3}He.

The Hubbard model has been used extensively to account for the properties of Mott insulators (2, 3, 4). It has also been proposed that b. c. c. ^{3}He might be properly described by the Hubbard model with one fermion per site (6). ^{3}He atoms interact through a short range potential U, the strenght of which is large compared with the hopping integral t accounting for the tunneling due to the large zero point motion of ^{3}He atoms (7). This model has been used by Widom and Sokoloff (12) to predict a ferromagnetic transition near the melting curve (with a vacancy concentration $\simeq 5\ 10^{-2}$). In this simple model, there is a complete analogy between b. c. c. ^{3}He and the electronic system of a b. c. c. Mott insulator. At T = 0, ^{3}He spins, localized on lattice sites should be antiferromagnetically ordered by the Anderson superexchange, of order of $\frac{t^2}{U}$ (7). A vacancy in this lattice is equivalent to an excess hole in an antiferromagnetic insulator. Differences between the two systems (a hole in a magnetic insulator and a vacancy in ^{3}He) may only come from different values of $\frac{t}{U}$. In the well known Mott insulators, such as NiO, a typical value of 3.10^{-2} is commonly admitted (4). In b. c. c. ^{3}He , there is no direct and precise measurements of t and U. Furthermore X rays measurements, spin diffusion coefficient values, NMR experiments, isochoric pressure measurements (8), magnetic ordering temperature values (7) do not yield the same estimate for t/U for the same molar volume. An indirect determination of t has been obtained from N.M.R. experiments. The value $t \simeq 50$ mK has been proposed (7). From X Rays measurements, which give the creation energy of Schottky vacancy, we obtain a lower limit for U (i.e. the creation energy of an interstial-vacancy pair) $U_I \simeq 6$ K. With $t \simeq 50$ mK we get $U/t > U_I/t = 120$. From $J = 2t^2/U = 0.75$ mK ($V = 24.5$ cm^3/mole) we get $U_2/t = 133$, and from $U_3/t = 5.52t/kT_N$ (9), $U_3/t = 240$.

These discrepancies, according to us, probably mean that the Hubbard model is too simple to describe quantitatively all the magnetic properties of b. c. c. ^3He Presumably, other physical quantities should be introduced, such as next nearest neighbour tunneling to have a better description. This situation is reminiscent of the difficulties encountered when one tries to describe the magnetic propertie of ^3He with a nearest neighbour exchange Heisenberg Hamiltonian (7, 8).

However, it is clear that the effects discussed in this letter are not restricted to the simplest Hubbard model, but still persist when other exchange terms are included, provided that t remains small compared with the vacancy creation energy and large compared with the exchange energy. Then, we only have to replace U/t by $q = zt/\Delta E$ in the results of sect.3 and 4. ΔE is the energy cost per spin of the ferromagnetic polarization. With only nearest neighbour exchange, $q = t/J = 133$. When next nearest neighbour exchange and four spins exchange are included, as in the Hetherington's model (13), $q = 144$ in the "SCAF" phase and 178 in the "NAF" phase, according to the numerical values of ref. (13). These values are above the variationnal critical value $q = 130$ of sect.3, remark d). Then, the results of sections 3 and 4 should apply. We expect thermal vacancies to be self trapped below $T_I \simeq 30$ mK. Those self trapped strong spin polarons should be independent at low enough concentration x, such that $x(R/a)^3 \ll 1$.

Recently Bernier and Delrieu (1) have measured the magnetic susceptibility of solid ^3He ($V = 24$ cm^3/mole) down to 1 m°K. Below about 5 m°K they found a departure from the Curie Weiss law obeyed at higher temperatures : when T decreases, the susceptibility increases faster and at $T = T_N$, it is about twice as large as expected from the Curie Weiss law. This anomaly is difficult to understand in terms of pretransitionnal effects near T_N, in a temperature range as large as $T_N < T < 5\,T_N$. We propose an explanation for this effect. If there exists a concentration x of vacancies they form polarons. Their contribution to the susceptibility obeys a Curie Law, since they are independent at low concentration. The effective moment per polaron is $\mu_{eff} = N\mu$ where N is the number of spins belonging to one polaron and μ is the magnetic moment of one ^3He atom. The total susceptibility is then

$$\chi = \frac{C(1-x)}{T+\theta} + x \frac{(\mu_{eff})^2}{3k_B T}$$

$$= \frac{C}{T+\theta} + x \frac{C}{T} \left(\frac{4\pi}{3}\right)^2 \left(\frac{\pi t}{b\,k\,(T_N + 2\,T\,\mathrm{Log}\,2)}\right)^{6/5}$$

(Below T_N, we expect polarons to gradually freeze magnetically in the AF lattice their contribution to X should decrease to a constant at 0°K (5)).

Above T_N, we obtain the right order of magnitude with a concentration of vacancie $x \simeq 2.5\ 10^{-4}$ (using here $kT_N = 4\ J = 3$ mK) .In this experiment, the solid was grown in a Pomeranchuk cell, and the susceptibility measured along the melting curve. Under such conditions, this concentration is not unreasonable. (II)

Clearly, the susceptibility should depend, through x on the conditions of the solidification. If the solid had been grown at high temperature and then cooled down to $T \simeq T_N$, we should expect x to have the equilibrium value at $T \simeq \frac{\Delta}{k_B}$ since they are trapped at that temperature. This value is extremely small for thermal vacancies (6, 7). An anomalous increase of the susceptibility in these conditions could only come, in our model, from zero point vacancies (10). This would provide a measurement of their concentration, if we accept our interpretation of the anomaly. At a given concentration, the effect of vacancies may be increased by applying a pressure at low temperature ($k_B T < \Delta$). x remains constant since the vacancies are trapped, but $J = \frac{t^2}{U}$ rapidly decreases (8). Thus the extra susceptibility is expected to increase.But,the simplest check of our interpretation would be to observe the rapid saturation of the polaron magnetization,which should occur with applied fields as low as $2kT/\mu N$(I4).

The magnetic transition at I mK is a first order transition with an unusually low critical temperature ($T_N \simeq 0.4|\theta|$).We insist on the fact that we have no explanation for these anomalies.. Would the polarons have such effects (which has to be proved),we think that given the orders of magnitude in the problem, the vacancy concentration is much too small to explain the experimental data in this way.

We believe that the formation of strong spin polarons in b. c. c. ^{3}He is plausible. If the concentration of vacancies is large enough, these polarons may have detectable effects on the properties of the solid, such as magnetic susceptibility, but also the spin diffusion, the ultrasonic absorption, the phonon modes, etc... (5). Further experiments below 5 m°K would be of interest.

In usual transition metal oxyde magnetic semiconductors, the U/t values seem to barely reach the critical value for polaron formation at O K; we are not aware of experimental data allowing to discuss this in more details but when U/t is large enough, strong polarons of large size must be trapped around excess holes at low temperatures in magnetic semiconductors.

Acknowledgements :

We thank G. Toulouse for a remark which stimulated our work. We had numerous discussions with A. Landesman, J. M. Delrieu, M. Chapellier, M. T. Béal-Monod and M. Papoular. J. Friedel helped us clarify some points. We gratefully acknowledge a critical discussion with I. Kagan. One of us (P.L) would like to thank the CBPF and the CNP_q for their hospitality during the completion of this work.

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