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Antiferromagnetic indirect exchange in ^3He adsorbed layers

M. Héritier

Laboratoire de Physique des Solides (*), Université de Paris-Sud, Centre d'Orsay, 91405 Orsay, France

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Résumé. — Nous proposons un nouveau mécanisme d'échange indirect dans les couches adsorbées très denses d' ^3He . Ce couplage antiferromagnétique provient d'échanges virtuels de particules entre la couche solide et le liquide. Il doit dominer de plus d'un ordre de grandeur les différents mécanismes ferromagnétiques proposés dans la littérature. Il rend compte du signe et de l'ordre de grandeur de la température de Curie-Weiss récemment observée.

Abstract. — We propose a new mechanism for indirect exchange in highly dense ^3He adsorbed layers. This antiferromagnetic coupling arises from virtual exchange of particles between the solid layer and the liquid. It should predominate by more than one order of magnitude over the various ferromagnetic mechanisms proposed in the literature. It accounts for the sign and the order of magnitude of the Curie-Weiss temperature recently observed.

Liquid ^3He in contact with various substrates forms highly dense adsorbed solid layers. In these layers, because of the strong Van der Waals attraction with the surface, the interatomic distance d is much smaller than in bulk b.c.c. ^3He along the melting curve. In the bulk solid, such small values of d (i.e. under about 400 atm) would correspond to very low values of the nearest neighbour direct exchange constant J , of the order of a few μK 's. Such a decrease of J is due to the strong hard core repulsion between ^3He atoms. Indeed, in the first monolayer adsorbed on Grafoil, neutron diffraction data indicate a compact triangular lattice with $d \simeq 3.32 \text{ \AA}$ [1]. N.M.R. studies on the same system gives $J \simeq 3 \mu\text{K}$ [2] (in the bulk solid, at the melting pressure, $d \simeq 3.72 \text{ \AA}$ and $J \simeq 0.75 \text{ mK}$ [3]). Various exchange mechanisms have been proposed as predominating over this very weak antiferromagnetic direct exchange : a ferromagnetic coupling induced by vacancies [4] or interstitials [5], a large cyclic three-spin exchange [6], and an indirect RKKY-like exchange, *via* the liquid [7]. All these ferromagnetic mechanisms might provide an alternative explanation to the tendency to ferromagnetism observed in ^3He in confined geometries [8]. The first proposed theory attributes this ferromagnetism to a Stoner instability *in the liquid*, favoured by an increased density of states near the surface [9]. On the contrary, the other couplings proposed would

induce a surface ferromagnetism *in the solid* layer. In this letter we propose a new mechanism for indirect exchange in the solid layer, which is antiferromagnetic, and thus opposed to the above ferromagnetic mechanisms.

Qualitatively, this mechanism is as follows : an atom of the solid layer can be emitted into the liquid, leaving a vacant site. In this 2 D quantum crystal, the vacancy can tunnel to a neighbouring site, which can be filled again by an atom of the liquid. These virtual transitions are only possible when the spins on the concerned lattice sites are parallel, and as shown below, they increase the energy because intermediate states with bonding vacancy wave function, i.e. positive vacancy kinetic energy are more easily excited. Thus, the coupling is antiferromagnetic. This third-order process might be non negligible because the effect of the fermion repulsion is much weaker in the liquid than in the solid layer.

More precisely, the solid layer is a quantum crystal with a tunnelling frequency t very much larger than the exchange frequency J , which we find convenient to describe by a simple Hubbard model with exactly one atom per lattice site (vacancies are neglected).

$$H_0 = -t \sum_{i,j} c_{i\sigma}^+ c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} \quad (t > 0). \quad (1)$$

In the first term, the sum is taken over nearest neighbour sites (i) and (j). $c_{i\sigma}^+$ ($c_{i\sigma}$) is the creation (annihilation)

(*) Laboratoire associé au C.N.R.S.

lation) operator of a ^3He atom Wannier state on site i with spin σ . The second term, in which $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$, represents the fermion hard core repulsion.

In this simple model, the nearest neighbour *direct* (as opposed to the indirect one discussed here) exchange constant is $J = 2 \frac{t^2}{U}$. Here, $J \sim 1 \mu\text{K}$, whereas t is certainly at least as large as in the bulk solid at the melting pressure : $t \gtrsim 100 \text{ mK}$ [10]. Thus we are in a situation where $t \ll U$. In fact, the real physical origin of the antiferromagnetic direct exchange might be different from this superexchange term, but this is not important for our purpose.

Furthermore, we allow for exchange of particles with the liquid, by introducing an interaction term :

$$H_i = \sum_{\mathbf{q}, i} t_{si}(\mathbf{q}) e^{i\mathbf{q} \cdot \mathbf{R}_i} a_{q\sigma}^\dagger c_{i\sigma} + \text{h.c.} \quad (2)$$

where $\mathbf{R}_i$ is the position of the lattice site (i), $a_{q\sigma}^\dagger$ is the creation operator of an ^3He atom state in the liquid with wave vector $\mathbf{q}$ and spin σ , and h.c. means the hermitic conjugate of the first sum. The strength of this coupling $|t_{si}(\mathbf{q})|$ is likely to be of the order of t . The adsorbed monolayer is a 2 D quantum crystal in which the amplitude of the zero-point vibrations of ^3He atoms is not negligible compared to d (probably of the order of $d/2$). Therefore, the probability for an atom to escape in the liquid has a finite spatial extension, and $t_{si}(\mathbf{q})$ cannot be considered as constant. It is maximum at $\mathbf{q} = 0$ and decreases appreciably over the first Brillouin zone. The chemical potential Φ of the ^3He atoms in the liquid, measured from the ground state of H_0 , depends on the strength of the Van der Waals attraction, and thus on the given substrate. It seems to range from 5 K to 50 K [11, 12]. Thus, the following inequalities among the various physical quantities are quite plausible :

$$t \sim |t_{si}(q)| \ll \Phi \ll U \quad (3)$$

Then, a straightforward third-order perturbation calculation (to first-order in t/Φ and second-order in t_{si}/Φ) gives a contribution to the energy :

$$\Delta E^{(3)} = \sum_{i,j} \tilde{J}_{ij} \left(\mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{4} \right) \quad (4)$$

where the sum is taken over nearest neighbour sites and

$$\tilde{J}_{ij} = 4 t \sum_{\mathbf{q}} \frac{|t_{si}(\mathbf{q})|^2 \cos \mathbf{q} \cdot (\mathbf{R}_i - \mathbf{R}_j)}{(\Phi + \varepsilon(q))^2} [1 - f(\varepsilon(q))]. \quad (5)$$

Here $\varepsilon(\mathbf{q})$ is the Hartree-Fock energy of a ^3He atom with wave vector $\mathbf{q}$ in the liquid, measured from Φ ,

and $f(\varepsilon)$ is the Fermi distribution function. In this crude approximation, correlation effects in the liquid are neglected. The sum appearing in (5) is positive so that the coupling is antiferromagnetic. The reason why $\tilde{J}_{ij}$ is positive is that the largest terms in absolute value appearing in (5) correspond to small values of q , and thus, to positive vacancy tunnelling matrix element [13]. If, in the intermediate state, an interstitial instead of a vacancy, was present, the tunnelling matrix element would be negative [13] at low q (in a bonding state, the interstitial has a negative kinetic energy), and the coupling would be ferromagnetic. Indeed, such a coupling induced by virtual interstitials exists but has a much lower strength than the virtual-vacancy induced one, because the energy of the virtual state is much larger ($U \gg \Phi$).

The possibility that a second solid layer exists has been discussed, especially on a Grafoil substrate [9, 11]. If this is the case, the same kind of indirect exchange is still present in the first layer. The only difference is that, in the virtual intermediate state, the atom emitted from the first layer is in an interstitial position in the second layer. The same kind of perturbation treatment still applies. The same orders of magnitude are expected for t_{si} and Φ , and thus for $\tilde{J}$.

Because the effect of the fermion repulsion is much weaker in the liquid (or in the second solid layer) than in the first solid layer ($\varphi \ll U$), the indirect coupling $\tilde{J}$, although due to a third-order process, is non negligible compared to the second-order superexchange $J = 2 t^2/U$ for acceptable values of the parameters. For example, taking $t \simeq 200 \text{ mK}$, $|t_{si}(q)| \simeq 100 \text{ mK}$ and $\Phi \simeq 10 \text{ K}$, we obtain $\tilde{J} \simeq 40 \mu\text{K}$, while $J \simeq 3 \mu\text{K}$. In this case,

$$\frac{\tilde{J}}{J} \sim \frac{U}{t} \times \left(\frac{t_{si}}{\Phi} \right)^2$$

is much larger than 1 because of the exceptionally high value for $U/t \sim 10^5$. We are not aware of another system of strongly interacting fermions in which such a large value of U/t can be found.

It is clear that this indirect coupling is, by all means, different from the RKKY-like indirect coupling, which has been proposed recently [7]. The latter is long ranged, and ferromagnetic at short distances ; in our simple model, this is a fourth-order interaction, of order t_{si}^4/Φ^3 , and, thus probably much smaller than the one discussed in this letter. However in a more complete model, other exchange mechanisms between the solid layer and the liquid could exist, with possibly, an exchange strength J_{si} larger than t_{si}^2/U . With the above values for t , t_{si} and Φ , these two opposite indirect interactions would be of the same order if $J_{si} \sim 15 \text{ mK}$, which seems to be a quite unrealistic value. Whatever the conditions, such a large exchange strength has never been found in the ^3He physics. The real value of J_{si} is quite probably much smaller. Therefore, we expect $\tilde{J}$ to

be larger than this RKKY-like coupling by about two orders of magnitude.

Another exchange interaction, namely the cyclic three-spin exchange, was suggested to be important in these compact triangular structures. This mechanism, which is ferromagnetic (cyclic permutation of an odd number of fermions [14]) has been invoked to interpret the tendency to ferromagnetism observed in ^3He in confined geometries [6]. Such an explanation would require a strength of the three-spin interaction two orders of magnitude larger than the two-spin direct exchange. Such a large term was not detected in a recent N.M.R. study of one complete ^3He monolayer adsorbed on Grafoil [2]. The measured exchange constant had the expected order of magnitude of the two-spin exchange for such an interatomic distance. In fact, this experiment puts an upper limit of $3\text{ }\mu\text{K}$ for the strength of any exchange mechanism which does not require the presence of the liquid, in particular to the triple exchange in the solid layer. Furthermore, the sharp increase of the exchange constant when a second layer begins to be formed [2] constitutes a strong indication that an indirect exchange (*via* the second layer) rapidly predominates over the direct one.

It was also proposed that vacancies [4] or interstitials [5] could favour ferromagnetism in 2 D quantum crystals. However, although interstitials actually induce a ferromagnetic coupling in any lattice, vacancies do so only in alternate lattices [5]. In a triangular structure, they probably favour a magnetic vortex ordering. In the very dense layers considered here, a loose-packed alternate structure and a sizeable concentration of vacancies or interstitials are quite unlikely. Therefore, we do not expect an important ferromagnetic coupling due to this mechanism.

We have reviewed all the exchange mechanisms which have been proposed in the literature to explain the magnetic properties of the ^3He adsorbed layers. All of them are ferromagnetic. Their strengths cannot be expected to be larger than $3\text{ }\mu\text{K}$ without either disregarding the data of ref. [2], or assuming quite unrealistic values of the physical parameters. On the contrary, the coupling we propose here is antiferromagnetic and can be of the order of $40\text{ }\mu\text{K}$.

It may have been observed in a recent experiment. In an N.M.R. study, of the susceptibility of liquid ^3He in contact with carbon black [15], two components of the free induction decay were detected. The fast one, whose amplitude corresponds to about one monolayer, leads to an antiferromagnetic Curie-Weiss temperature $\theta_1 \simeq -90\text{ }\mu\text{K}$. The slow one, corresponding to about five monolayers, leads to a ferromagnetic Curie-Weiss temperature

$$\theta_2 = +350\text{ }\mu\text{K}.$$

They were ascribed respectively to the first adsorbed monolayer and to the high density fluid above the surface [15]. θ_1 corresponds to a nearest neighbour

antiferromagnetic interaction $J = 60\text{ }\mu\text{K}$, much larger than that observed in an ^3He monolayer adsorbed on Grafoil ($J \simeq 3\text{ }\mu\text{K}$) in the absence of the liquid. The simplest explanation would be to assume a larger interatomic distance in the monolayer adsorbed on carbon black. $d \simeq 3.57\text{ }\text{\AA}$ (instead of $3.32\text{ }\text{\AA}$ on Grafoil) would account for the data [9]. We propose another explanation, namely the indirect exchange induced in the presence of the liquid in the carbon black experiment.

Some doubts could already be cast on the models explaining the tendency to ferromagnetism by ferromagnetic interaction in the solid layer, either by multiple exchange (a very small value of J was measured in the absence of the liquid) or by vacancy-induced coupling (the existence of an alternate structure in each experiment would be necessary). If our interpretation of the data is correct, this seems to rule out these models. If the dominant coupling in the solid layer is antiferromagnetic, the ferromagnetic tendency can only be ascribed to the high density liquid near the surface. In this picture, the theory of ref. [9] predicting a favoured Stoner instability seems the most appealing one.

Preplating the substrate with Ne should probably increase the indirect exchange $\tilde{J}$: a reduced holding potential [11, 16] results in less tightly bound adsorbed atoms, therefore in a slight decrease of t (which decreases $\tilde{J}$), but more importantly in an increase of t_{sq} and a decrease of Φ , and thus in a large increase of $\tilde{J}$.

This short range Heisenberg coupling is not able to induce a long range ordering at finite temperature in a 2 D magnetic system. However the adsorbed layer is not really a two-dimensional system since it is coupled to the liquid, in which ferromagnetic interactions predominate near the surface, so that the problem of the long range order in the adsorbed layer is not simple.

Just as there exists a repulsive interaction between ^3He atoms with parallel spins in pure ^3He adsorbed layers, there exists a repulsive interaction between ^4He atoms in an adsorbed layer of ^3He - ^4He mixture. The interaction is of the Ising type, with the same order of magnitude as the Heisenberg interaction between ^3He atoms. It would favour an ordering of the ^3He - ^4He mixture. However, it is probably negligible compared to the direct He-He interactions which, on the contrary, seem to favour an isotopic segregation [11].

A similar antiferromagnetic Heisenberg coupling should also exist in a system of electronic spins. The system equivalent to the ^3He adsorbed layer is a layer of insulating transition metal oxide at the surface of a transition metal. Of course, the orders of magnitude are different. We are not aware of an example in which $\tilde{J}/J$ could be of order unity, and we doubt this could be realized because the ratio U/t in the oxide layer is much smaller than in the

^3He layer. Indeed in the oxide, the intraatomic Coulomb repulsion $U \sim 10$ eV, $t \sim 0.1$ eV, so that $U/t \sim 100$ instead of 10^5 in the ^3He layer. Therefore $\tilde{J}/J \gtrsim 1$ only if $t_{s1}/\Phi \gtrsim 1/10$, a condition which it seems difficult to satisfy.

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