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Spin Polarons in Solid ^{3}He: Suggestions for Further Experiments

H. J. M. van Bemmel and W. van Saarloos

Institute Lorentz, University of Leiden, P. O. Box 9506, 2300 RA Leiden, The Netherlands

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A proposal is made for a simple way to test experimentally whether anomalies in the low temperature thermodynamic properties of solid ^{3}He are due to metastable trapped *vacancies in spin polarons. The idea is to perform susceptibility measurements before and after applying a strong magnetic field for some time, with the aid of a SQUID or NMR. If this* magnetic annealing *reduces the susceptibility, it provides evidence for the existence of metastable trapped vacancies in the original sample. With this procedure it may also be possible to clarify the issue of the existence of a "vacancy solid." Finally, the effects of the tendency of vacancies to cluster are discussed.*

1. INTRODUCTION

In this article we make a simple but, to our knowledge, new proposal for a way to experimentally investigate the existence of ferromagnetic spin polarons in solid ^{3}He. Arguments in favor of the formation of these ferromagnetically ordered regions around vacancies were already put forward over 25 years ago by Andreev[1] and Héritier and Lederer[2] on the basis of the description of the motion of a vacancy in solid ^{3}He in terms of a Hubbard model (in this case, the usual hopping parameter t is the hopping amplitude of vacancies and U is of the order of the energy of a vacancy-interstitial pair). The basic idea underlying these arguments is the result of Brinkman and Rice[3] that the density of states for the vacancy in a given spin background is narrower in the paramagnetic and antiferromagnetic phases than in the ferromagnetic phase. As a result, in the limit $t/U \ll 1$ in which the effective spin interaction $J = 2t^2/U$ is much smaller than t, a vacancy can lower its kinetic energy by surrounding itself with a ferromagnetically ordered region—a spin polaron. Although a model with higher order interactions is needed to describe the spin ordering of solid ^{3}He in the milliKelvin range,[4] this basic feature of polaron formation for $t/J \gg 1$ remains in all earlier approaches. A particularly thorough discussion of the theoretical work on spin polarons in ^{3}He as well as of the experimental evidence for polarons up to 1982 was given by Montambaux *et al.*[5] Their main conclusion was that some anomalies in the static susceptibility and in the specific heat between a few mK and $40 \sim 50$ mK could well be consistent with the existence of polarons, but that the number of vacancies needed to explain the data they considered (which were taken away from the melting line), was larger than one would expect. Possibly, they suggested, these vacancies are trapped in the ^{3}He crystals; some experiments that they discussed indeed suggest that the number of vacancies decreases upon annealing.

To our knowledge, a systematic investigation[6] of spin polarons has not been undertaken, even though the polaron concept has occasionally been invoked to explain certain data (as will be discussed later, it is believed that the susceptibility data by Kirk *et al.*[7] show evidence of a partial melting of the crystal[8,9] not of the existence of spin polarons, as was suggested earlier[10]). From a different perspective, Lengua and Goodkind[11] have recently also stressed the need to understand the behavior of vacancies in solid He at higher temperatures.

From the theoretical side, there is renewed interest in the old subject of magnetic textures around holes in the Hubbard model, in the context of high T_c materials. These textures include polarons, domain walls[12] and vortices.[13] The notion of phase separation of holes, which had already been introduced in the context of the discussion of polarons in He,[14] was recently discussed for such models by Emery *et al.*[15] There are several reasons why it is useful to study these textures in ^{3}He. First, the long range Coulomb interaction that could cause the actual behavior in high T_c materials to be different from that of the Hubbard model (e.g. it could prevent phase separation of holes), does not exist between vacancies in ^{3}He. Also, the ratio U/t as estimated for ^{3}He[5] is a factor of five larger than for the high T_c materials[16] or any other material for which the Hubbard model yields an appropriate description of the electronic degrees of freedom. In fact the extent to which the one band Hubbard model incorporates the essential features of the high T_c materials is still subject of debate.[16] Therefore, studying solid ^{3}He as an example of a correlated Fermi system is very attractive as it appears to provide the *cleanest experimental realization* of a Hubbard type model in the extreme large U limit. This is the parameter region in which polarons could exist, while for smaller U/t domain walls are energetically more favorable. One should keep in mind however one important difference between most theoretical analyses of the Hubbard model and related models and of the studies of polarons in ^{3}He: the former are mostly focussed on the $T=0$ ground state properties in two dimensions, while the latter are most relevant in the paramagnetic phase at temperatures T above the Néel ordering temperature T_N. In this case the formation of a ferromagnetic polaron results from the balance of the reduced kinetic energy of the vacancy and the decrease in spin entropy associated with the ferromagnetic spin ordering, while at $T=0$ the kinetic energy of the vacancy is balanced with the increase in exchange energy resulting from the breaking of the antiferromagnetic bonds.

In the Brinkman-Rice[3] approach on which the polaron theory[1,2,5] is based, quantum fluctuations in the antiferromagnetic order are ignored and as a result the vacancy motion in an antiferromagnetic background is found to be diffusive. Recent numerical and theoretical results[17] on the $t-J$ model with $t/J=\mathcal{O}(1)$ show that the quantum fluctuations make the motion coherent, as a quasiparticle band of width of order J is found. If these findings extrapolate to smaller values of J then they do not necessarily, however, invalidate the earlier arguments for the existence of polarons for very small J/t: for in this limit the decrease in kinetic energy of the hole in a region of ferromagnetic order $(\sim t)$ always exceeds the kinetic energy of the coherent quasiparticle motion in the antiferromagnetic or paramagnetic background $(\sim J)$. Moreover, there have been arguments[17] that the bandwidth for the hole motion is exponentially small for $t/J \gg 1$, which is the limit relevant for ^3He.

Our main purpose in this paper is to advocate a simple way to test experimentally whether *metastable* trapped vacancies are present in a sample. The point is that polarons around vacancies have been invoked to explain anomalies in low-temperature thermodynamic properties sufficiently away from the melting line, and that more vacancies are needed than one would expect in equilibrium.[18] In this picture, the reason for this excess of vacancies is that polarons are effectively very heavy, and are trapped in the crystal when the temperature is reduced.[5] This scenario can be tested, in the following way. We propose to perform systematic measurements in the temperature and molar volume range of the susceptibility[19,20] and specific heat[21,22] measurements whose behavior was interpreted in terms of polarons by Montambaux *et al.*[5] If these measurements show behavior consistent with the earlier experiments, and hence with the existence of metastable trapped vacancies, a strong magnetic field (of several Tesla) should be applied for some time (we shall estimate how long later). Since their mobility is a strongly increasing function of the polarization (as the vacancies can move coherently when the polarization approaches unity) this allows the density of vacancies to approach the equilibrium density at the temperature at which the measurement is done. Then, after having switched off the magnetic field, the measurement should be repeated. If the anomalies are then reduced, one can conclude that their occurrence in the first measurement was indeed due to polarons around metastable trapped vacancies. This procedure comes down to *magnetic annealing*. Furthermore, if polarons are found to be stable rather than metastable according to these measurements, we estimate that they can be oriented in fields of the order of 0.5 Tesla (depending, of course, on the temperature). This translates into a nonlinear susceptibility that should start to decrease in fields higher than these values, an effect that should be easily measurable.

Following the suggestion of Castaing and Nozières,[23] the melting of highly polarized solid ^{3}He has been used as a way to obtain strongly polarized liquid ^{3}He. In a number of experiments of this type, a relatively sharp break in the polarization dependence of the melting pressure has been observed.[24] Again, it has been speculated that this behavior might be associated with the existence of polarons in the solid phase.[5,25] After having discussed in Sec. 2 in which experiments in zero field the existence of spin polarons could be visible and how the results would change after magnetic annealing if there was an excess of vacancies before, we shall briefly comment in Sec. 3 on the particular suggestion of the existence of a "magnetic vacancy solid" of Bouchaud and Lhuillier.[25] It seems appropriate to test whether the effects in the melting behavior for which this new thermodynamic phase is invoked, could be due to (trapped) vacancies. Therefore, resolving the question of the existence of polarons in low temperature ^{3}He crystals would at least settle the issue of the appropriateness of using polarons as one of the ingredients for an explanation of the change of the melting behavior that is observed.

The predictions for the effects of a diminishment of the number of vacancies are rather clear-cut. In Sec. 4, we analyze the behavior that could arise because holes in the Hubbard model have the tendency to cluster. Clustering could be especially important if there was no excess of vacancies to begin with, or if the applied field in magnetic annealing is too weak or is applied for too short a time that the vacancies can leave the crystal. These effects are more subtle, and possibly difficult to test directly in experiments. Yet it is important to discuss them, because we need to know how they affect the interpretation of measurements before and after magnetic annealing with respect to the number of vacancies. Moreover it would be of fundamental importance if one would be able to demonstrate experimentally that clustering exists.

2. SPIN POLARONS AND TESTS FOR THEIR EXISTENCE

2.1. Spin Polarons

For an introduction to and overview of the properties of spin polarons in ^{3}He we refer to the paper by Montambaux *et al.*[5] The main idea is that a vacancy has most freedom to move, and thus the lowest kinetic energy, in a ferromagnetic region.[26,3] The background is paramagnetically ordered in the case of ^3He above T_N (or, in the analogous case of the Hubbard model at $T=0$, antiferromagnetically ordered). This leads to a competition, and in a simple continuum picture of a particle in a deep well with sharp walls, one has to minimize an expression for the free energy of the form[1,2,5]

$$F=\frac{a}{R^2}+bR^d \tag{1}$$

where the first term arises because the vacancy is localized in a ferromagnetic region of size R, and the second term accounts for the fact that the spin background is broken down over a d-dimensional volume. Formula (1) applies at $T=0$ with the free energy replaced by the energy if one has holes in a $t-J$ or large U Hubbard model; then $a \sim t$ and $b \sim J$. For the case of ^3He above T_N, the competition is between ferromagnetic ordering of the spins around vacancies and the paramagnetic background. Then the main contribution to the second term in (1) comes from the entropy difference, while the first term still represents the vacancy kinetic energy of order t, the vacancy hopping amplitude. From (1) one finds $R_{\rm min} \sim (a/b)^{1/(d+2)}$; for the $t-J$ or large-U Hubbard model with $J=2t^2/U$, this gives $R_{\rm min} \sim (t/J)^{1/(d+2)}$, which illustrates that the continuum picture becomes self-consistent for large t/J. The estimate of Montambaux *et al.*[5] for ^3He, after putting in the geometrical factors and accounting for several corrections to the simple picture implied by (1), is that such a polaron contains 30–50 spins. Much work has been done on the Hubbard model and ^3He since; the feature that is of importance here, the preference of a hole to have a ferromagnetic surrounding, remains in all these more sophisticated approaches, although the exponent with which the radius scales as a function of J remains subject of debate.[17] It is important to emphasize that the size of a polaron (if it exists) is not necessarily the same in all experiments, since we believe t to be strongly dependent on the molar volume.[27] Moreover, the estimate $t \approx 50$ mK that Montambaux *et al.* used[5] appears somewhat low; we believe t is likely to be larger in most experiments,[28] and this would tend to give larger polaron sizes.

2.2. Thermodynamic Properties

The fact that polarons act as clusters with giant moments has consequences for thermodynamic properties. For the susceptibility one finds[5]

$$\chi=\frac{\mu^2}{k_B(T+\Theta)}(1-Nx)+x\frac{N^2\mu^2}{3k_BT} \tag{2}$$

where x is the vacancy concentration and N is the number of spins per polaron (it is assumed that there is one vacancy per polaron; we shall come back to this point in Sec. 4). In addition, there is a loss of entropy due to the alignment of the nuclear spins. The expression given in Ref. 5 for the case of three dimensions is

$$\Delta S = -x k_B \ln 2 \left(\frac{\pi t}{k_B T_N + 2 k_B T \ln 2} \right)^{3/5} \tag{3}$$

In this expression the term between brackets is the number of spins per polaron N (in terms of the general formula (1) this term is of the form $(a/b)^{d/(d+2)}$). These relations have been used by Montambaux *et al.*[5] to explain the enhanced static susceptibility between 1 and about 5 mK, found by Bernier and Delrieu[19] and by Prewitt and Goodkind[20] in a SQUID measurement, and the anomalies in the entropy measurements of Halperin[21] and in the specific heat data of Dundon and Goodkind,[22] both below 20 mK.

We note that NMR measurements by Kirk *et al.*[7] show an anomaly opposite to those of Bernier and Delrieu[19] and of Prewitt and Goodkind,[20] i.e. a *decrease* in the susceptibility upon lowering the temperature. Kumar and Sullivan[10] have argued that the particular form of NMR used in those experiments could well probe only the first contribution to the susceptibility in (2), as the polaron contribution from the second term could be shifted out of the NMR frequency window due to local field effects. If this assumption is made, one can fit the data of Kirk *et al.* in terms of a polaron model. However, Bernier and Suaudeau[8] have put forward as a more likely explanation of Kirk's data a partial melting of the solid, as the experiments were done close to the melting line, and since the susceptibility of the liquid is much smaller than that of the solid. Later measurements of Bernier *et al.*[9] strongly support this suggestion. We will therefore concentrate our discussion only on those measurements that show an *increase* of the susceptibility (or specific heat), an effect that can not be attributed to a partial melting of the sample.

We finally note that for polarons consisting of N spins, the magnetization will saturate at fields large enough that $N\mu H/k_B T \gtrsim 4$. For N of the order of 30 and T of the order of 5 mK the crossover occurs for fields of the order of 0.3 Tesla. If polarons are larger, the resulting decrease of the susceptibility will of course occur at even smaller field values. Hence, if one still observes an enhancement of the susceptibility after the magnetic annealing discussed below, one can test whether this is due to the existence of stable polarons by measuring whether the nonlinear susceptibility indeed decreases for fields larger than about 0.5 Tesla.

2.3. Magnetic Annealing and the Detection of a Diminishment of the Number of Vacancies

Although Montambaux *et al.*[5] may have overestimated the number of vacancies in the experiments whose data they analyzed,[18] the vacancy concentration they needed to account for the anomalies is consistent for the various measurement, but higher than would be expected for thermally activated vacancies at the temperatures ($T \lesssim 40$ mK) at which the experiments are performed. (Except very close to the melting line, vacancy activation energies are of the order of 1 K, so that at low temperatures one expects only a very low fraction of thermally activated vacancies.) The reason why there could be many vacancies in a metastable state is the following. Once the magnetic polarons form at a certain temperature and pressure, they are trapped in the lattice: because many spins are involved, the polarons are effectively very heavy.[17] Iordanskiĭ[29] has discussed the possibility of polaron motion by the propagation of spin waves through the ferromagnetic region. This gives a larger mobility than if only motion by flipping spins at the boundary is considered, but it is much lower than the mobility of a bare vacancy. As already noted by Montambaux *et al.*,[5] a magneto-elastic coupling to the lattice would make the polarons even heavier,[30] and possibly make them self-trapped.

This seems a plausible argument for the explanation of the anomalies mentioned, and it is consistent with a number of observations discussed by Montambaux[5] that their occurrence depends on the way the sample is prepared. We propose to test this scenario directly as follows. If, in a certain sample a susceptibility increase (or change in the specific heat) is found, a large magnetic field should be applied for some time, while the temperature is kept fixed. If the sample becomes almost completely polarized, the reason why the excess of vacancies was trapped is taken away, because it is a *magnetic* trap. After sufficient time to reach the equilibrium number of vacancies, the field should be switched off. Then the original measurement should be repeated after the sample is depolarized again. If the previously measured effect has diminished, this would indicate that the proposed explanation is correct. If one does not find a reduction, the assumption that *metastable* trapped vacancies are the cause of the effects would be proven incorrect. As discussed in the previous section, one should in that case investigate the nonlinear susceptibility.

How strong does the field have to be for such a magnetic annealing, and how long does it need to be applied? It is difficult to give a precise answer to this question, but the following estimates indicate that annealing in a field of several Tesla at a few mK should suffice:

1. In the Brinkman–Rice picture, the effective hopping amplitude t_{eff} of a polaron is equal to the bare vacancy hopping amplitude t times the

overlap of the two spin states which differ by the shift of the polaron by one lattice unit. If the background is paramagnetic, the overlap of these two states is of order $(1/2)^{N_s}$, where N_s is the number of spins on the surface of the polaron. Hence the effective hopping amplitude $t_{\text{eff}} \approx (1/2)^{N_s}\, t$ is indeed extremely small in the absence of a field, if the polaron is not too small ($N_s \gtrsim 20$, say). If the background has a polarization Δ in the presence of a field, the effective hopping becomes $t_{\text{eff}} \approx ((1+\Delta)/2)^{N_s}\, t$. This shows that if the polarization becomes appreciable (60% or more), the hopping amplitude and hence the mobility of the polarons becomes large enough that they can leave the crystal. Note that a polarization of about 60% can be obtained in a field of about 7 Tesla at 6 mK.[31] With fields of the order of 10 Tesla, one can get a Δ of order 90% at about 4 mK.

2. In addition to the above polarization dependence of the effective single polaron hopping amplitude, the polaron mobility will also be increased by a field due to two collective effects. First of all, the increase of polarization will tend to increase the attractive force between polarons[5]; this effect, which Montambaux *et al.* estimate will become significant at fields of a few Tesla in the mK range, will enhance clustering of polarons. Secondly, in fields of a few Tesla the size of a polaron will increase, since the entropy difference between the ferromagnetic spin state in the polaron and the polarized state outside it, decreases. E.g., in fields of the order of 7 Tesla at 4 mK, the entropy difference per spin is reduced by a factor of two,[31] and this increases the number of spins in the polaron by some 50%. Depending on the original concentration of vacancies and the field, such an increase may lead to the breakdown of the independent polaron picture. E.g., if the vacancy concentration is of order 10^{-3}, and if each vacancy gives rise to a zero field polaron of some 50 spins, the total fraction of spins in polarons in the above field increases to 6%; at even somewhat higher values, such an estimate would lead to a percolating polaron network. Clearly, in this regime the independent polaron picture is not justified.

3. It is useful to also consider the other limit. In a very strong field, so that the polarization is almost 100%, the concept of a spin polaron has no meaning. At low temperatures, vacancies then move coherently with a mean free path l which will be much larger than the lattice spacing a: $l/a \gg 1$. For their diffusion coefficient D_v we then have $D_v = (t/h \cdot a) l = (l/a)(t/h \cdot a^2) \approx 4 \cdot 10^{-7} \cdot (l/a)(cm^2/s)$. As a result, for a sample size of the order of centimeters, a reasonable fraction of the vacancies can diffuse to the walls in an hour if $l/a > 10$.

Based on these observations, we believe that annealing in fields of several Tesla for a good fraction of an hour should be sufficient to test the idea of *magnetic annealing.*

Unfortunately, there do not appear to be sensitive other experimental probes of the formation of polarons besides measurements of the specific heat and susceptibility (either with NMR or with a SQUID). Spin polarons in magnetic semiconductors[32] can be probed with magneto-electric effects, but we lack the coupling to charge in ^3He. Likewise, magneto-optical effects will be much weaker in ^3He than in materials like EuTe[33,34] because we deal with nuclear spins instead of electronic ones. A different possibility to test for the existence of spin polarons in ^3He are the resonances predicted by Meĭerovich,[35] which are due to the curvature of the wave function of the vacancy at the origin, when it is localized in a polaron. Such experiments appear to be difficult, however, and, to our knowledge, they have not been performed.

3. MELTING BEHAVIOR, SPIN POLARONS AND THE VACANCY SOLID

As an explanation of rather sharp breaks in the dependence of the melting pressure on the polarization during rapid melting of ^3He,[24] Bouchaud and Lhuillier[25] assumed the existence of a new phase: the magnetic vacancy solid. The possibility of having zero point vacancies in He crystals near the minimum of the melting curve where the formation energy of vacancies appears to go to zero[36] has been speculated about often,[37] especially for ^4He.[38] Bouchaud and Lhuillier build on this and the idea of spin polarons by speculating that for nonzero magnetization m of the solid there might be a stable vacancy solid ^3He phase. The phase diagram they propose is sketched in Fig. 1(a). There are two reasons why we consider this proposal not tenable. First of all, the phase diagram is thermodynamically inconsistent. As pointed out to us by Nozières,[39] if one tries to keep track of the relative stability of the three phases involved in Fig. (1), one finds out that it is not possible to get the order correct everywhere. The lines cross in an inconsistent way: between two lines that separate two phases, there should be the extrapolation of a line that separates a different set of phases, so that upon circling the triple point, one crosses a dashed line each time after a full line is crossed. A thermodynamically consistent way to draw the phase diagram is sketched in Fig. 1(b), but this phase diagram would not explain the breaks in the data for the melting pressure as a function of polarization. Our second reason to question the proposed explanation of these data is that the measurements are performed relatively far away from the minimum of the melting curve, where the formation energy of vacancies tends to zero, and so where zero point vacancies are most likely to be found, if they do exist.

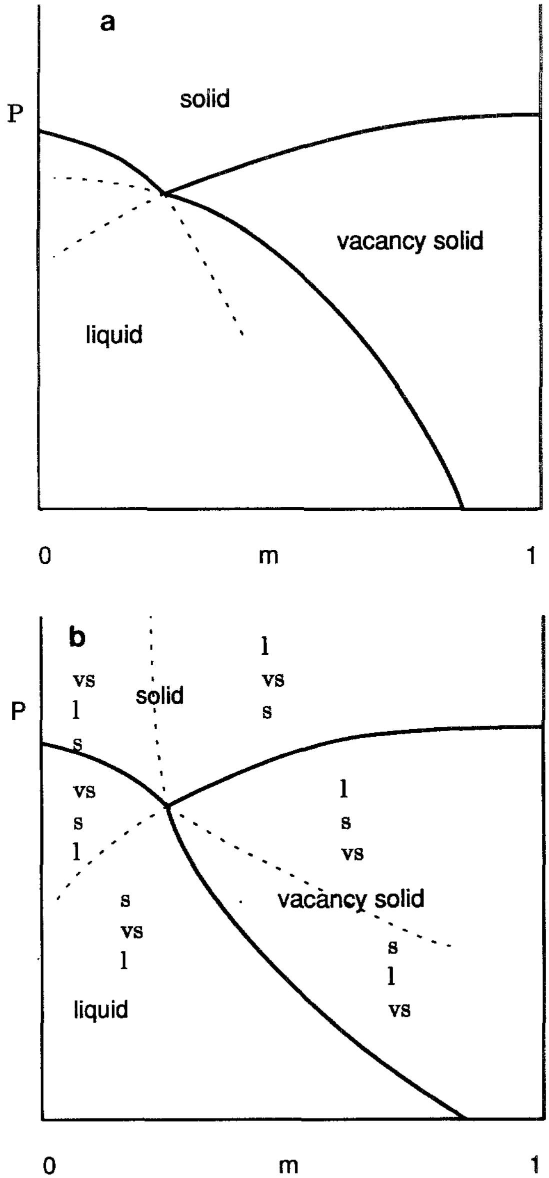

Fig. 1. (a) The phase diagram as proposed by Bouchaud and Lhuillier. (b) Thermodynamically consistent phase diagram; in each part of the diagram, the relative order of the energies of the phases is indicated, the lowest position corresponding to the lowest energy. In (a), a consistent labeling is impossible.

In spite of these caveats, the suggestion of Bouchaud and Lhuillier[25] that the observed anomalies result from some effect in the solid phase associated with polarons near (trapped) vacancies, appears worth pursuing, especially since the possibility of an anomaly in the liquid has recently been ruled out.[24] In fact Montambaux *et al.*[5] had already suggested that these effects could be associated with the saturation of the magnetization due to polarons at higher fields (if so, the effect would have to be strongly dependent on the temperature) or some collective polaron effect. If the solid is inhomogeneous due to the existence of polarons, it is conceivable that the ferromagnetic regions near polarons melt slower (later) than the regions with a lower polarization. In this respect, it is worth mentioning that Frossati *et al.*[31] have observed kinks in the relaxation of the magnetization after rapid melting of ^3He crystals. We intend to investigate in the future whether these could be interpreted in terms of the slow melting of regions with high magnetization.

Whether the anomalies in the melting behavior are due to *metastable* trapped vacancies can not be tested as directly as in the case of susceptibility. It would be interesting, however, to investigate whether their appearance depends on sample preparation. For example, one could do measurements on a number of samples which are grown in the same way, but of which some have been magnetically annealed. In particular if phase separation of vacancies occurs (as has been predicted theoretically[15]), this might affect the melting behavior.

4. THE EFFECTS OF CLUSTERING

In the previous sections, we have described what the main effects are if the mobility is increased sufficiently by the field that metastable trapped vacancies can move to the surface of the crystal and disappear, so that their concentration goes down. We will now discuss how a clustering of vacancies in larger polarons would affect the proposed measurements,[40] in the simplest approximation based on the picture discussed in Sec. 2.1.

The first issue is the reason why this tendency exists. If one calculates in a simple continuum picture what the minimum (free) energy is of i vacancies in a polaron of size R, including the penalty for change in spin energy or entropy in this region, one has to minimize

$$E_i = \frac{a_i}{R^2} + bR^d \qquad (4)$$

which is a generalization of formula (1). The a_i's depend on which levels (of spinless fermions in a box) have to be filled (for example, for square polarons in two dimensions, we obtain $a_2 : a_1 = 7 : 2$, for circular ones it is

$a_2 : a_1 = (2.41^2 + 3.83^2) : (2.41^2)$). One finds that the total energy of more vacancies in a polaron is always lower than the total energy of separate polarons with fewer vacancies each. The fact that the background has to be broken down only once, is more important than the need to fill higher levels. (As an illustration we show in Fig. (2) how the energy *per hole* depends on i, for the two-dimensional t-J model. In this case the hole energy is proportional to $\sqrt{tJ}$. The limit of large i gives the same value as one obtains using the picture of phase separation (see Emery *et al.*[15])).

Suppose in an experiment there are initially trapped vacancies, mainly in separate ferromagnetic polarons, with one vacancy per polaron. Then, by an applied magnetic field, their mobility increases. There will be a tendency to cluster, because that lowers the energy. If there was no excess of vacancies (but the vacancies were trapped in their positions), or if the field is applied for a short time, which gives the vacancies insufficient time to leave the crystal, this clustering will dominate. If this is the case, fewer but bigger polarons will exist after the magnetic annealing.

The consequences for the thermodynamic properties are as follows. From (4) we find

$$R_i \sim \left(\frac{a_i}{b}\right)^{1/(d+2)} \tag{5}$$

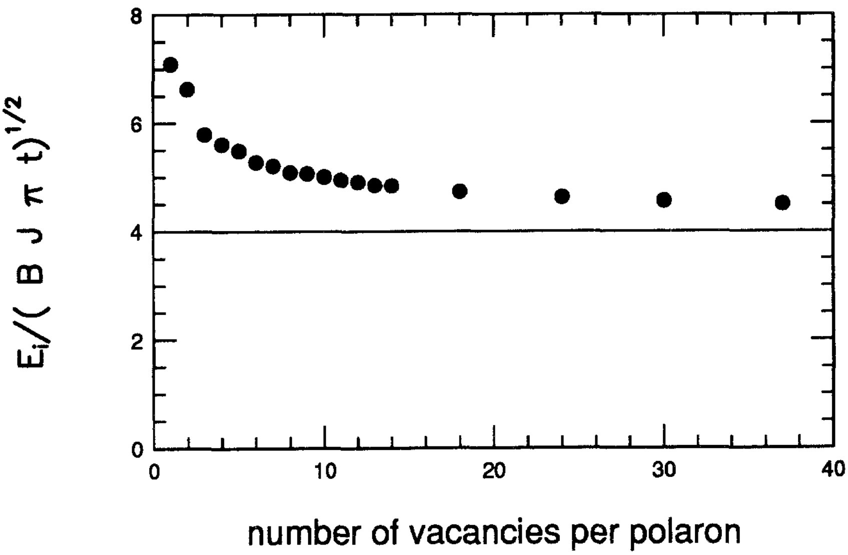

Fig. 2. Energy per hole, for i holes in an infinitely deep square well in two dimensions, in the t-J model, illustrating the tendency cluster. The prefactor of $\sqrt{BJ\pi t}$ tends to four for large i, which is the value in the case of phase separation (see Emery *et al.*[15]).

where R_i is the radius of a polaron with i vacancies, and d is the dimension. Then the number of spins per polaron goes as

$$N_i \sim \left(\frac{a_i}{b}\right)^{d/(d+2)} \tag{6}$$

and thus we can see what happens to the thermodynamic properties. For the entropy loss due to polarons, we find

$$\Delta S_i \sim \frac{x}{i}\left(\frac{a_i}{b}\right)^{d/(d+2)} \tag{7}$$

where x is the vacancy concentration. The susceptibility enhancement in the second term of Eq. (2) is now

$$\Delta\chi_i^+ \sim \frac{x}{i}\left(\frac{a_i}{b}\right)^{2d/(d+2)} \tag{8}$$

while the diminishment in the first term becomes

$$\Delta\chi_i^- \sim \frac{x}{i}\left(\frac{a_i}{b}\right)^{d/(d+2)} \tag{9}$$

just as the entropy contribution. Note that the enhancement of the first term $\Delta\chi^+$ always dominates.

First we consider what happens if the polarons remain small, that is, if we go from single polarons to bipolarons. Then it is important that the second level in the "well" is much higher than the first, and that makes the polarons so much bigger that the effect in $\Delta\chi^+$, which is proportional to N^2, increases, despite the diminishment of the *number* of polarons. The effects proportional to N become a little bit smaller. For square polarons in two dimensions we obtain $\Delta\chi_2^+ = 1/2 \cdot 7/2\, \Delta\chi_1^+ = 1.75 \cdot \Delta\chi_1^+$, and $\Delta S_2 = 1/2 \cdot (7/2)^{1/2}\, \Delta S_1 = 0.94 \cdot \Delta S_1$, the latter factor also applies to $\Delta\chi^-$. The numbers for circular polarons are approximately the same. In three dimensions we obtain $\Delta\chi_2^+ = 1/2 \cdot (9/3)^{6/5}\, \Delta\chi_1^+ = 1.87 \cdot \Delta\chi_1^+$, and $\Delta S_2 = 1/2 \cdot (9/3)^{3/5}\, \Delta S_1 = 0.97 \cdot \Delta S_1$. The precise numbers depend on the shape of the polarons, but the trend is that going from single to bipolarons gives an enhancement of $\Delta\chi^+$, while $\Delta\chi^-$ and ΔS do not change much.

The effect on the resonances in high frequency NMR measurements, predicted by Meĭerovich[35] is qualitatively as follows. The resonances arise because a single vacancy in a polaron is situated primarily at the center, which causes the effective exchange interaction to be inhomogeneous. The frequencies depend on how the wave function is curved at the origin. If there are more vacancies in a polaron, the first effect is that the polaron becomes bigger, which makes the curvature smaller and the resonance frequencies lower. Furthermore, the second vacancy occupies a state with a wave function that is not centered at the origin. The exchange will be much more homogeneous over the polaron, which makes the frequencies lower. It is likely that, because polarons of different shapes and with different amounts of vacancies per polaron form, no clear resonances will be observed at all, if clustering occurs.

Let us finally discuss what happens if very many vacancies go into big ferromagnetic regions. The filling of higher levels has a somewhat different effect than for the case of bipolarons, because the number of levels with a certain energy proliferates for higher energies. We can use the picture of a Fermi pressure for $a_i : a_i \sim \varepsilon_F \sim (i)^{2/d}$. So we obtain for i large

$$N_i \sim (i)^{2/(d+2)} \tag{10}$$

Thus

$$\Delta\chi_i^+ \sim (i)^{4/(d+2)-1} \tag{11}$$

and

$$\Delta\chi_i^- \sim \Delta S_i \sim (i)^{2/(d+2)-1} \tag{12}$$

So one sees that, if vacancies go into very big regions, $\Delta\chi^+$ remains the same in two dimensions and decreases slightly in three, while the other two effects decrease in both dimensions. Although the particular exponents with which these quantities scale may change in a more sophisticated approximation,[17] we expect the tendency to cluster to remain.

5. CONCLUSIONS

In this article, we hope to have convinced the reader that it should be possible to convincingly prove or disprove the existence of spin polarons in ^{3}He crystals with existing experimental techniques: systematic experiments, in particular SQUID or NMR susceptibility measurements (or measurements of the specific heat), together with the proposed magnetic annealing, should be able to show whether polarons exist, and if so whether they are associated with metastable trapped vacancies. If the magnetic annealing does not give rise to a change in behavior, one should study the nonlinear susceptibility. Unravelling this issue may also help understand the origin of some of the anomalies seen upon melting of polarized ^{3}He.

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