Magnetic polarons in low-density solid ^{3}He

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We review the properties of polarons nucleated by vacancies in solid ^{3}He and compare the magnetostatic contribution to the recently observed anomalies in the susceptibility for densities close to the melting curve. We show that the experimental results can be understood in terms of polaron effects if the experiments were sensitive only to the bulk paramagnetic solid and could not see the spins bound to the polarons. We also discuss the thermal relaxation of the spins in the polaron cloud and its importance for the analysis of the experiments. The magnetic contribution to the pressure $P(H,T)$ due to the presence of polarons is calculated and new experiments are proposed to test for the predicted polaron properties.

I. INTRODUCTION

The presence of vacancies in solid helium leads to a number of striking and unusual effects that are intimately related to the dominance of quantum effects in these "quantum crystals." Only some of the predicted properties have been verified and the purpose of this work is to discuss the effects of vacancies on the nuclear magnetism in solid ^{3}He. The feature which distinguishes "quantum solids" (such as the solid heliums and solid hydrogens) from ordinary solids is the large quantum zero-point motion of the atoms. The interatomic forces are weak, the masses light, and the atoms are only weakly localized with respect to their crystal lattice sites. This affects the properties of vacancies in general in two ways. Firstly, because of the kinetic zero-point motion of the atoms, an atom may tunnel into a vacant site. The vacancies are therefore delocalized, they should propagate through the crystal as "vacancy waves" and form a band of energies whose width is directly proportional to the vacancy mobility. The vacancies tunnel through the lattice with frequencies of the order of 10^{10} Hz. The second consequence is that, as opposed to ordinary solids, the vacancies make a very substantial contribution to the thermal properties of the solid; namely the heat capacity, the thermodynamic pressure, nuclear spin-lattice relaxation, and the scattering of lattice vibrations.[1] As a result of the large zero-point motion of the quantum solid, the mobility of the vacancies also dominates the properties of the solid close to melting, rather than the thermal vibrations. Finally, in solid ^{3}He, the vacancies can at low temperatures lead to profound changes in the magnetism of the ^{3}He atoms in their surroundings. The reason for this is that the high kinetic energy associated with the vacancies can be reduced if the nuclear spins of the surrounding atoms are allowed to become polarized. This effect, first proposed by Andreev[2] and later by Lederer and coworkers,[3] results in a self-trapped bound polarization state which is split-off from the extended vacancy bands. The analogy with polaron formation in ionic solids is self-evident and the vacancies surrounded by their polarization cloud are therefore referred to as magnetic polarons.

The thermodynamic effects of polarons are due to (a) the thermal population of the vacancy states[4] and (b) the temperature dependence of the size and shape of the polarization cloud. The former is relatively small [although some experimental techniques (Ref. 5) are sensitive to thermal populations as low as 10^{-10}]. Some of the most significant effects are due to the temperature dependence of the nuclear spin polarization which increases with decreasing temperature. These observables are in fact associated with the zero-point population of the vacancies. We refer here to the low-temperature saturation of the vacancy concentration, x_v, which is observed experimentally to scale exponentially with the activation energy $\Phi(V)$. The molar volume dependence at low densities, i.e., close to the maximum molar volume, is given by $\Phi(V) \sim (V_{mc}-V)^{1/2}$, and the dependence for the vacancy fraction is $x_v \sim \exp(V_{mc}-V)^{1/2}$. The critical molar volume $V_{mc}=24.95$ cm^3/mole.

Magnetic polaron effects can only be observed over a limited temperature range. If the number of vacancies is large, e.g., in the immediate neighborhood of the melting pressure minimum, the temperature range over which the solid is stable is very small. As a result the solid melts before the polarons become large enough to be visible by most experiments. One needs to have access to sufficiently low temperatures and low densities for the observation of significant polaron effects. The limiting molar volume and temperature ranges are (see Fig. 1) then $V_{mc} > V > 24.0$ cm^3/mole and $100 > T > 16$ mK.[5,6] The susceptibility measurements of Kirk *et al.*[7] lie in this range but very few experiments had explored this region prior to their studies and they did not use the high-precision techniques of Kirk *et al.* There is clearly a need for more systematic experimental investigations of low-density high-purity solid ^{3}He in this temperature range.

The effects discussed below refer to the zero-point concentration of the vacancies. Whether these are somehow frozen in (e.g., defects that have not been able to diffuse to a surface) or whether they refer to the intrinsic low temperature zero-point saturation of the vacancy concentration, x_{v0}, cannot be conclusively decided on the basis of the experimental evidence that is currently available. We

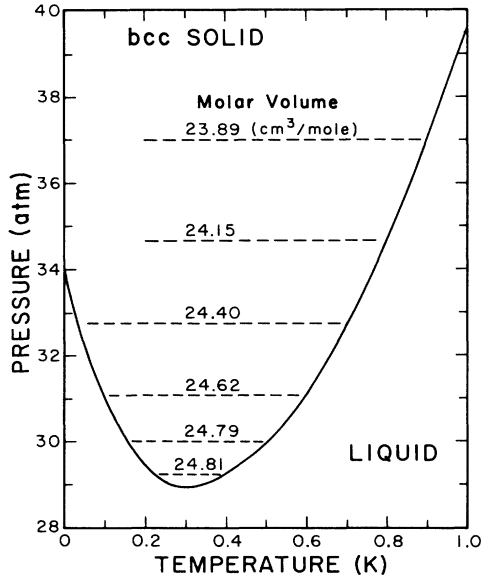

FIG. 1. Isochores on the phase diagram of bulk solid ^{3}He showing the typical trajectories followed by the experiments discussed in the text. [The melting curve appropriate for the experiments of Kirk *et al.* (Ref. 7) differs from the curve for bulk solid because of the effects of constrained geometries on their samples.]

assume that x_{v0} is intrinsic in the following. The evidence supporting this assertion is derived from the low temperature NMR measurements.[5] At higher temperatures, the spin diffusion in magnetic field gradients[8] and the nuclear spin-lattice relaxation rates (T_1^{-1}) both scale as $\exp(-\Phi/k_BT)$ in their temperature dependence. Also seen in x-ray studies[6] and recent careful measurements of the thermodynamic pressure,[9] this temperature dependence is interpreted as due to the thermal vacancies whose activation energy, shown in Fig. 2 as a function of molar volume, is reminiscent of vacancies. At lower temperatures, $(T_1)^{-1}$ scales linearly with T and has been interpreted variously as due to a single-phonon process[10] or due to the motion of vacancies.[5] Additional support in favor of the latter mechanism comes from Fig. 3. Using data from Ref. 5 we have plotted T_1T as a function of $\Phi(V)$. The clear exponential dependence (the values for 24.6 cm^3/mole correspond to a reanalysis of the data reported in Ref. 5) supports the interpretation in terms of a vacancy mechanism and suggests a temperature-independent x_v. Furthermore, the analysis of the x-ray scattering studies[6] indicate an approximately constant concentration of vacancies with $x_v(\text{melt}) \sim 5\times 10^{-3}$ over a very substantial portion of the melting curve ($3.0 > T > 0.3$ K). The magnetic polarons are therefore expected to be abundant in a low-density solid and constitute an assembly of noninteracting, heavy, but nevertheless mobile carriers of polarization. In the appendix, we have calculated the internal modes of a magnetic polaron. The energy scales for the internal energies are typically 0.3 K and therefore negligible at millikelvin temperatures. This means that the polaron dynamics can be treated as that of a rigid body. The internal modes may be important in determining the frequency dependence of $(T_1)^{-1}$ but we defer that analysis for the moment.

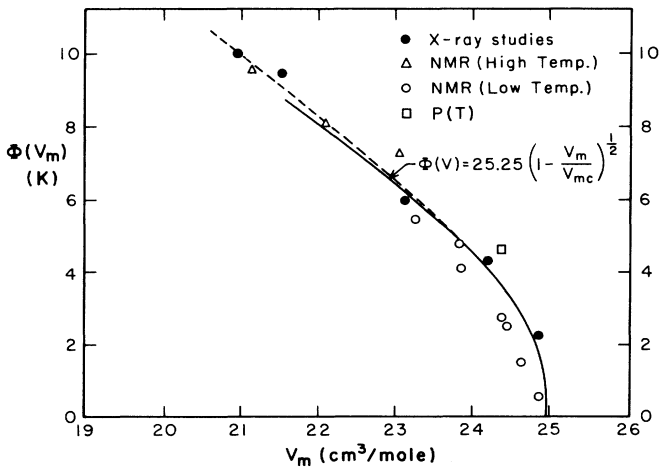

FIG. 2. Volume dependence of the vacancy formation energy $\Phi(v)$ observed in different experiments; NMR (open circles, Ref. 5 and triangles, Ref. 18, x-ray studies (solid circles, Ref. 6), and pressure studies (open square, Ref. 9).

The principal motivation for the present study stems from the recent susceptibility measurements[7] of Kirk *et al.* (for high molar volume samples of solid ^{3}He) who observed an anomalous *decrease* in the susceptibility below the Curie-Weiss law at low temperatures. This anomaly can be understood[11] in terms of the formation of magnetic polarons and the theoretical $T^{-3/5}$ deduction agrees well with the experiments, using reasonable values for the parameters.

Kirk[12] has put forward an alternative proposal, based on the onset of degeneracy in the susceptibility of magnetic polarons. He argues that the formation of a magnetic polaron in fact lowers the degeneracy temperature for vacancies to $T_F \sim 100$ mK by raising their effective mass. The degenerate magnetic polarons then respond with a Pauli susceptibility, much smaller than the Curie-Weiss law. It was also claimed that the fermion behavior of magnetic polarons leads to the linear dependence $T_1T = \text{const}$, akin to the Korringa relation for electrons.[13]

The temperature range over which the linear dependence is observed, however, is much higher (typically, $T > 180$ mK) than the temperature for which anomalies have been seen ($T < 60$ mK) in the susceptibility. At these higher temperatures the vacancies presumably remain undressed (very few polarized spins surround the vacancy) and as stated above, the observations can be interpreted either in terms of an exchange-vacancy coupling or an exchange-phonon coupling.

The principal disagreement with Kirk's model lies in whether the lost spins respond as a giant moment or whether they constitute a degenerate Fermi liquid. Fortunately, there is a simple test that will help decide between the two models. If the lost spins form a giant moment, it should be easy to saturate them at fields of the order of a few kilogauss. The resulting effects are observable in the finite field response of the susceptibility and/or the isochores $P(H,T)$. If the lost spins make up a

Fermi liquid, no saturation effects should be seen.

We have also explored the possibility of superparamagnetism for the polarons. The giant moments can interact with each other and may lead to an ordered state. The interaction between polarons is assumed to be a classical dipole-dipole interaction. It is important to recognize however, that the polarization of the polaron cloud saturates due to the exchange interaction at around 10^3 spins/polaron. The dipolar interaction for the largest moment leads to an interaction energy scale less than 1 mK. Since the typical temperatures vary from 10—100 mK, polaron-polaron interactions remain negligible. Superparamagnetism is preceded by the appearance of the ordered state in the bulk solid ^{3}He at 1 mK.

The outline of this paper is as follows. In Sec. II, we summarize the principal properties of the magnetic polarons. Section III discusses both the ac and dc susceptibilities. Section IV includes a discussion of finite field isochores $P(H,T)$ and also the melting pressure $P(T)$. Finally, Sec. V provides a summary of our conclusions.

II. THEORETICAL OVERVIEW

The thermodynamic properties of a magnetic polaron can be determined from a free energy functional $F(T)$,

$$F(T)=F_{\rm bulk}(T)+x_v f_p(T)\ , \tag{1}$$

with

$$F_{\rm bulk}(\sigma,T)=\tfrac{1}{2}zJ\sigma^2-\sigma H-T\Sigma(\sigma) \tag{2}$$

and

$$f_p(T)=-t(s)+\pi^2 t(s)\left[\frac{a}{R}\right]^2 -\frac{4\pi}{3}nR^3\{T[\Sigma(s)-\Sigma(\sigma)] -\tfrac{1}{2}zJ(s^2-\sigma^2)+\mu_N H(s-\sigma)\}\ . \tag{3}$$

The quantity $F_{\rm bulk}(\sigma,T)$ is the energy functional for the bulk solid ^{3}He without vacancies. The polarization of bulk ^{3}He, $\sigma_0(T)$, is obtained by minimizing this free energy. Here J denotes the antiferromagnetic exchange interaction with each of z nearest neighbors, H is the external magnetic field, and $\Sigma(\sigma)$ the entropy. μ_N is the ^{3}He nuclear magnetic moment. The vacancy concentration x_v arises from the thermodynamics of vacancies and is in general temperature dependent. As discussed above, it saturates at low temperatures and we focus on this range of temperature. We are thus dealing with only the zero-point population of the vacancies.

The magnetic free energy density (per vacancy), $f_p(T)$, consists of two classes of terms. In the first group, the first two terms refer to the formation energy $t(s)$ which depends on the environment polarization s, and the kinetic energy of localization which scales inversely as the square of the localization radius R. The lattice constant is a. (The nearest-neighbor separation $a_0=a\sqrt{3}/2$.) The second group of terms represents the free energy gain in the formation of magnetic polarons. Here n is the solid ^{3}He density. The magnetic polaron can be visualized as a polarization cloud within which the vacancy is localized. The entropy terms $\Sigma(\sigma)$ and $\Sigma(s)$, represent the entropy functionals for solid ^{3}He and the polaron with average polarization s, respectively. While $\Sigma(\sigma)\simeq\ln 2-\sigma^2/2C$ is a fair approximation (C is the Curie constant), $\Sigma(s)$ should be different and constructed such that the expected magnetic polaron polarization can be obtained by minimization of $f_p(s)$ with respect to s.

In the absence of a magnetic field, $\sigma\to 0$. The function $t(s)$ is monotonic and largest at $s=1$. This determines the polarization of the polaron. Clearly, the strong discontinuity in polarization at the polaron boundary is an approximation. If the polarization varies less rapidly, there might be surface energy terms that should be included in the following. We however ignore these terms for the moment, with the understanding that the radius of polaron may in reality, be larger than the estimates given below. The corrections are however, not expected to be important for large polarons.

The effective polaron free energy therefore becomes (leaving aside the constant shifts)

$$f_p(T)=\pi^2 t\left[\frac{a}{R}\right]^2+\frac{4\pi}{3}nR^3(T\ln 2+\tfrac{1}{2}zJ-\mu_N H)\ . \tag{4}$$

Minimization with respect to R yields a polaron size

$$R^5=\frac{\pi}{4}\frac{ta^2}{n}(T\ln 2+\tfrac{1}{2}zJ-\mu_N H)^{-1}\ . \tag{5}$$

Equations (4) and (5) were given in Ref. 3. The resulting polaron consists of $N_{\rm sp}=4\pi nR^3/3$ polarized spins. In effect then, $x_v N_{\rm sp}$ spins respond as a polaron whereas $1-x_v N_{\rm sp}$ spins maintain their paramagnetic response. The polarons behave as a giant rigid body. As mentioned above and in the Appendix, the internal excitations of a polaron are much too high in energy. The translational motion of this rigid body, however, is hindered by the large mass associated with the number of spins increasing with decreasing temperature. The problem is indeed very similar to self-trapping (hence the name). Nevertheless, the polarons hardly become sufficiently dense to form a degenerate gas. The effective Fermi energy depends on temperature

$$k_B T_F=\frac{\hbar^2 k_F^2}{2m^*}=\frac{\hbar^2(3\pi^2 x_v)^{2/3}}{2m-\varepsilon_b}\ . \tag{6}$$

where ε_b is the binding energy of the magnetic polaron.

There is a saturation of the polaron size. The radius R in Eq. (5) decreases to a saturation value R_m, for $T\leqq T_0=zJ/(2\ln 2)$. (The Curie-Weiss temperature $\Theta=[6\ln(2s+1)/s(s+1)]T_0$.) This maximum size of the polaron plays an important role in determining bounds on various properties. Equation (6) gives a lower bound to the Fermi energy at approximately 10^{-4} K. The degeneracy remains elusive until the system is cooled to a tem-

perature below that bound.

We now consider the energy of interaction between the polarons. Given that the number of polarons is temperature independent, the energy of interaction increases with decreasing temperature because of the increase in polaron moment,

$$E_d = \frac{m^2}{r^3} \simeq x_v N_{\rm sp}{}^2 \varepsilon_d \ . \tag{7}$$

ε_d is the dipolar energy for interaction between two ^{3}He spins in a lattice ($\sim 10^{-6}$ K). The largest value of E_d is obtained at saturation and is of the order of (or less than) 1 mK. In the temperature range where polaron effects are expected to be present (and observed if our interpretation is correct) neither the degeneracy, nor superparamagnetism appears to be an important effect.

III. MAGNETIC RESPONSE

Given the enhanced polarization in the neighborhood of vacancies, the effect of vacancies should be most easily observed in response to applied magnetic fields. In this section we discuss the magnetic response including the susceptibility and the nuclear spin lattice relaxation time. We remark that one of the most characteristic signatures of polarons, noted originally by Delrieu,[14] is the saturation of the polaron moment by relatively low fields. This saturation field is given by

$$H_c(T) = \frac{k_B T}{M_{\rm pol}} = \frac{k_B T}{\mu_N N_{\rm sp}} \simeq A(T+T_0)^{3/5} \ . \tag{8}$$

At high $T(\gg T_0)$, $H_c(T) \propto T^{8/5}$ and as the polaron size saturates near $T \simeq T_0$, the dependence becomes linear with $H_c(T) \propto T$. In contrast, if the polaron gas becomes degenerate, the saturation field $H_c \propto T_F (\sim T^{-3/5})$ and has a much weaker temperature dependence. The effect of this saturation field on the field dependence of the isochores $[P(H,T)]$ is discussed in the following section. Here we calculate the spin susceptibility and spin-lattice relaxation time T_1.

A. Susceptibility

The magnetic moment of solid ^{3}He, containing ferromagnetic polarons is given by

$$M = M_{\rm CW} + x_v M_{\rm pol} \ , \tag{8a}$$

$$M_{\rm CW} = \mu_N^2 H (T+\Theta)^{-1}(1 - x_v N_{\rm sp}) \ , \tag{8b}$$

$$M_{\rm pol} = \mu_N N_{\rm sp} x_v L[\mu_N N_{\rm sp} H/kT] \ , \tag{8c}$$

where the Curie-Weiss law [Eq. (8b)] takes into account only the paramagnetic spins. Since the polaron is large we have treated its field response as that of a Langevin function. The consequences are the following:

(1) The saturation field, as mentioned above, is small and has a specific temperature dependence.

(2) The Curie-Weiss susceptibility of the paramagnetic spins is altered in its temperature dependence. If the polaron moment is invisible to the experimental probe, either because of long relaxation times or because of frequency shifts, the observed susceptibility would be given by Eq. (8b). As shown in Fig. 3, it agrees very well with the currently available data of Kirk *et al.*[7]

(3) When the polaron response is visible, the static susceptibility is given by

$$M = \frac{\mu_N^2}{T+\Theta}[1 - x_v N_{\rm sp} + x_v (\mu_N N_{\rm sp})^2/kT] H \ . \tag{9}$$

In this case the important temperature-dependent correction is the last term which is proportional to $N_{\rm sp}^2 \sim (T+T_0)^{-6/5}$. The susceptibility is now enhanced with a $T^{-11/5}$ dependence, as opposed to the reduction seen in (2) above which scales as $T^{-3/5}$. Both of the responses, in Eqs. (2) and (3) saturate near $T_0 \simeq zJ/\ln 2$.

Equation (8b) can be rewritten in the form of a change in susceptibility $\delta\chi$ such that

$$\frac{\delta\chi}{\chi_{\rm CW}} = 1 - \frac{\chi}{\chi_{\rm CW}} = x_{v0} N_{\rm sp} = x_{v0} A \left[\frac{t}{T+T_0}\right]^{3/5} , \tag{10}$$

χ is the susceptibility of paramagnetic spins subject to the caveat given above. In Fig. 4, we compare[11] Eq. (10) with the experiments[7,12] for two molar volumes, $V_m = 24.42$ cm^3/mole and $V_m = 24.21$ cm^3/mole. The high-temperature limit of Eq. (10) is limited by rather small polarons. Experimental curves yield

$$\frac{\delta\chi}{\chi_{\rm CW}} = c[(T+0.012)^{-3/5} - 5.2] \ , \tag{11}$$

where $c = 0.054$ and 0.033 for $V_m = 24.42$ and 24.21 cm^3/mole, respectively. Evidently, the polaron size saturates at $T_0 = 12$ mK. Recovering the exchange constant

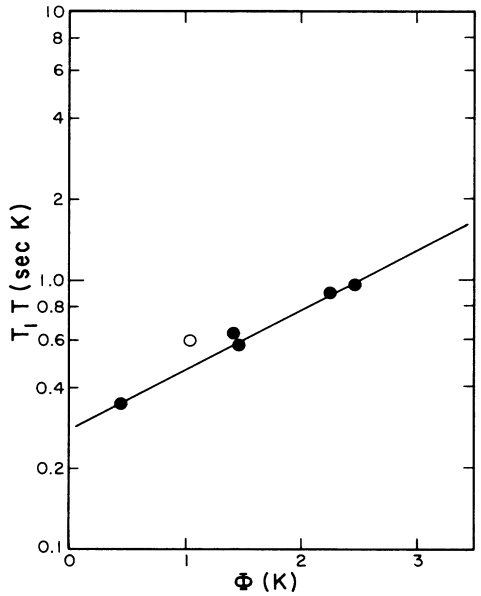

FIG. 3. Variation of the product $T_1 T$ with the vacancy formation $\Phi(v)$. The relaxation times T_1 given in the figure refer only to the regime where a linear temperature dependence ($T_1 \propto T^{-1}$) has been observed (see Ref. 5). The solid circles are taken from Ref. 5 and the open circle corresponds to a reanalysis of the data of Chapellier *et al.* for molar volume $V_m = 24.62$ cm^3/mole ($T_1 T = 0.61$ s K and $\Phi = 1.05$ K).

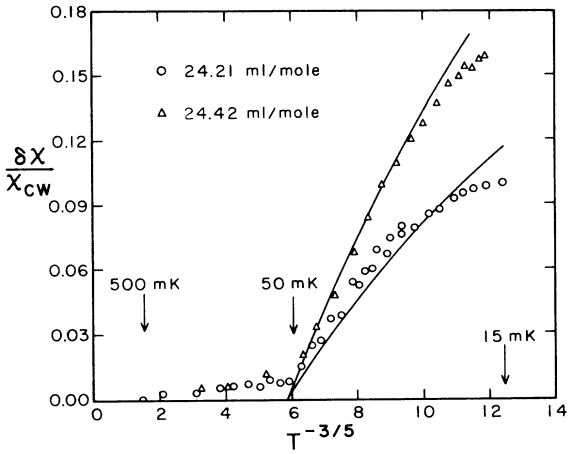

FIG. 4. Comparison of the observed temperature dependence of the deviation $\delta\chi$ of the nuclear spin susceptibility from the Curie-Weiss value with the anticipated variation $(T+T_0)^{-3/5}$ for a rigid polaron model.

J, we find a value of 2 mK, in order of magnitude comparable to the independent[15] susceptibility measurements. The constant A in Eq. (10) consists of natural constants and is given as approximately 10. In order to deduce the vacancy hopping energy t from the constant c in Eq. (11), we need x_v. Using $x_v = 5\times 10^{-3}$, the maximum value reported so far by x-ray studies near the melting curve, we find $t=1$ and 0.44 K for $V_m = 24.42$ and 24.21 cm^3/mole, respectively. Again there is quantitative agreement with known values. Theoretical estimates based on the overlap of distorted Gaussian wave functions yield values $t\sim 1$ K (Refs. 1 and 16) and these are consistent with measurements of spin diffusion by Reich[8] and analyses of the heat capacity studies by Hetherington.[17] Measurements of the nuclear spin relaxation times at high temperatures[18] however, imply much smaller values, with t as low as 50 mK and these latter values are not understood. The relaxation studies differ from the other experimental probes in that they are sensitive only to the motion of vacancies and measure the fluctuation rates at the nuclear Larmor frequency (typically 100 MHz). The interpretation of the T_1 results therefore hinges crucially on the models assumed for the dependence of the spin-spin correlation functions on the hopping time t. At the present time there is insufficient experimental data to determine whether the discrepancy lies in the models used or on a subtle variation of the dynamics across the vacancy bandwidth. T_1 studies over a broad range of frequencies could help resolve this dilemma.

The agreement between known parameter values and the numbers derived from the fit is encouraging. This is especially so in view of the rather qualitative nature of the calculation. At high temperatures, the anomalous contribution to the susceptibility $\delta\chi$ in Eq. (11) is observed to decrease to zero rather abruptly at 60 mK—leading to the constant 5.2. If we nonetheless apply Eq. (10) at $T=60$ mK, there are only approximately 50 spins in the polaron. It is precisely for this number of spins that our approximation of a constant polarization across the polaron must break down. There are a number of effects which become important in the limit of small polaron size. A first attempt to include size effects for small polarons has been made by Montaumbaux *et al.*[3] These authors estimated the critical radius for the formation for a polaron cloud to be $R_c = 1.8a_0$ (a_0 being the nearest-neighbor separation). If we interpret the apparent cutoff of the anomaly in the susceptibility seen at 60 mK as due to the fact that the radius has dropped to R_c, we find R_c (apparent)$\simeq 1.9\ a_0$ at $T=50$ mK using $t=1$ K. This is in good agreement with the estimate of Montaumbaux *et al.* and indicates that the interpretation of both the temperature dependence ($T^{-3/5}$ for $50>T>16$ mK) and the cutoff are self-consistent, and both features give values of t consistent with other experimental data.

B. Nuclear spin lattice relaxation

The spin-lattice relaxation rate T_1^{-1} has been found proportional to T by Chapellier *et al.*[5] in the temperature range $T>60$ mK. In this temperature range, the polaron is small and its effects are negligible. In other words, the analysis by Chapellier *et al.* using energy bands for vacancy motion is quite adequate. When the polaron does become large, the dominant relaxation mechanism is completely different and ferromagnetic polarons play no role. This is because the solid, at least partially, melts. If the melting curve is lowered below the bulk value due to the effects of constrained geometries in the experiments of Kirk *et al.*[7] so that a large amount of solid is still present, we expect an additional spin diffusion constant derived from the dipolar interaction between the polarized clusters. This diffusion constant is much smaller, of the order of $(\mu_N N_{sp})^2 x_v/J$ compared to the ^{3}He spin diffusion constant. To our knowledge, no experiments report the existence of such a mechanism.

It is worth pointing out that spin diffusion away from the polaron cloud is strongly bottlenecked by the high polarization P of the spins in the cloud. The flip-flop rate between spins in the polaron varies as $1-P^2$ and since $P\sim\tanh(\mu_N H_{int}/2k_BT)$, where H_{int} is the polarizing field in the cloud ($\sim$0.1 kG), this polarization barrier reduces the relaxation rate by several orders of magnitude. It will lead to an exponential temperature dependence $(1-P^2)\sim\exp(-\mu_N H_{int}/k_BT)$. This has not been observed, probably because the relaxation times become very long, rendering the polarons invisible to the experiments employed. This inability to observe the spins in the cloud (without special precautions) is the central premise in our interpretation of the data of Kirk *et al.*[7]

In the model of Chapellier *et al.*,[5] the relaxation is assumed to proceed via a sequential coupling of four energy baths: Zeeman (Z)→Exchange (E)→Vacancies (V) →Phonon bath (B). The weak link is the exchange-vacancy coupling and this leads to not only the linear temperature dependence (for "frozen in" or ground state vacancies), but also to a very strong volume dependence arising from the coupling of the exchange to vacancies.

The exchange-vacancy coupling is assured by a Raman process of simultaneous excitation to, and emission from, energy levels in a broad band $N(E)$ of vacancy states. The intrinsic relaxation rate, on summing over all available states, is

$$T_{1(EV)}^{-1}=\frac{2\pi}{\hbar^2}\int\int M^2N(E)[1-N(E)]\rho(E)\rho(E')\delta(E-E'\pm J)dEdE'$$
$$\simeq\frac{2\pi}{\hbar^2}M^2k_BT\rho^2(E_F)\ . \tag{12}$$

M is the matrix element for the coupling of vacancy waves to the exchange system and E_F is the Fermi energy for the vacancy states. The resulting temperature dependence is

$$T_{1(EV)}T\sim\text{const}\ .$$

The density dependence is given by $M\sim[V(\partial J/\partial V)]\sim 18J$. The Grüneisen constant for the intrinsic relaxation rate is then $\gamma_{\text{intrin}}\sim 2\gamma_J$ (the exchange Grüneisen). Experiments actually follow the Zeeman energy which is bottlenecked by the exchange-vacancy coupling and the observed relaxation is

$$T_1^{-1}=\left[\frac{C_E}{C_Z+C_E}\right]T_{1(EV)}^{-1}\simeq 12(J/\omega_L)^2T_{1(EV)}^{-1}\ , \tag{13}$$

where C_x is the heat capacity of bath x, and ω_L is the nuclear Larmor frequency. The observed density dependence therefore has a Grüneisen constant

$$\gamma(T_1)\sim 4\gamma_J$$

(i.e., the same as J^4) where we do not include the volume dependence of the vacancy fraction x_v. Representing the latter by γ_V (which is not known), we finally have

$$\gamma(T_1)\sim 4\gamma_J+2\gamma_V\ .$$

IV. MAGNETOELASTIC EFFECTS

Since the molar volume dependence of the exchange integral is large (e.g., the magnetic Grüneisen constant is 18), the magnetoelastic effects are in general very large in solid ^{3}He. At low densities, the polarons are present and cause some profound shifts of the isochore $P_V(T,H)$. This can be seen by recalling that the magnetic field dependence of $P_V(T)$ can be obtained from the Maxwell relation

$$\left.\frac{\partial P}{\partial H}\right|_V=\left.\frac{\partial M}{\partial V}\right|_T\ .$$

In the absence of ferromagnetic polarons, the molar volume dependence of $M(V)$ arises from the same dependence of the Curie-Weiss constant Θ [i.e., that of $J(V)$]. The resulting $P(H)$ is given by

$$P(H)=P(0)-\frac{C}{(T+\Theta)^2}\frac{\partial\Theta}{\partial V}\frac{H^2}{2}\ . \tag{14}$$

In the presence of ferromagnetic polarons, the magnetization becomes

$$m(v)=m_{\text{CW}}+x_vN_{\text{sp}}m_p\ , \tag{15}$$

where

$$m_p=\mu_NL(\mu_NN_{\text{sp}}H/\mu T)-m_{\text{CW}}\ ,$$
$$m_{\text{CW}}=\mu_N^2(T+\Theta)^{-1}H\ .$$

The vacancy concentration x_v is taken to be $\exp[-\alpha\Phi(v)]$. The corresponding $\partial M/\partial V$ is dominated by the square root singularity in $\partial x_v/\partial V$. Ignoring all other contributions, the effect of ferromagnetic polarons on $P_v(H,T)$ can be expressed as

$$P(H)=P(0)+\frac{\alpha}{4}\frac{x_v}{(V_c-V)^{1/2}}\frac{(\mu_NN_{\text{sp}})^2}{kT}H^2$$
$$\text{for } H<H_c(T) \tag{16}$$

$$P(H)=P(0)+\frac{\alpha}{2}\frac{x_v}{(V_c-V)^{1/2}}(\mu_NN_{\text{sp}})H$$
$$\text{for } H>H_c(T) \tag{17}$$

where $H_c(T)=kT/\mu_NN_{\text{sp}}(T)$. There are two notable differences between Eqs. (16) and (17) and Eq. (14). The first difference is that the pressure increases with field. We have omitted the Curie-Weiss correction in deriving Eqs. (16) and (17). However, when important, it further adds to the positive field dependence. The second effect is in the magnetic field dependence. Since the polarons can be saturated at $H_c(T)$, the pressure depends linearly on field for $H>H_c(T)$. The Curie-Weiss correction still provides a quadratic field dependence. At low temperatures ($N_{\text{sp}}\gg 1$), polarons dominate and it should be possible to detect their linear field dependence. These effects are dominant near the melting pressure minimum because of the square root divergence in the molar volume dependence. Typically, N_{sp} ranges from 70—700. The scale field $H_c(T)/T$ varies as 20 kG/K. At 50 mK, this leads to a typical critical field $H_c(T)\simeq 1$ kG. The order of magnitude effect in Eq. (17) can then be estimated to be of the order of a few percent.

V. SUMMARY

The analyses presented above show that the recently observed anomalies in the nuclear magnetic susceptibility of ^{3}He at high molar volumes can be understood in terms of the effects of magnetic polarons which are nucleated by delocalized point defects in the solid. The model predicts two features, (i) a $(T+T_0)^{-3/5}$ temperatuare dependence with saturation at low temperatures ($T\leqq T_0\sim ZJ$) due to spin exchange and (ii) a null effect for temperatures $T>T_\mu\sim 60$ mK corresponding to the critical radius for a magnetic polaron. Both effects are seen experimentally, and the required tunneling energy needed to fit the data is given by $t\sim 1$ K which is consistent with other experimental results. New experiments, in particular studies of the contribution to the field dependence of the thermodynamic pressure, are suggested to further test the predictions of the model proposed.

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APPENDIX

In the introduction we refer to the possibility of internal modes for the magnetic polarons. In the following, we derive the energies of these internal modes. The classical calculation of the polaron free energy in Eq. (4) can be rewritten as an expectation value of a quantum Hamiltonian H,

$$E=\pi^2 ta^2\frac{1}{R^2}+\frac{4\pi}{3}R^3 nT\ln 2=\langle H\rangle\ , \tag{A1}$$

with

$$H=\frac{\hbar^2}{2m^*}\nabla^2+gr^3\ . \tag{A2}$$

The mass m^* and the potential strength g can be expressed in terms of the hopping energy t and the entropy terms $nT\ln 2$, respectively. From dimensional analysis, any expectation value of H must scale with

$$E=\left[\frac{\hbar^2}{m^*}g^{2/3}\right]^{3/5}$$
$$\simeq(tT^{2/3})^{3/5}\simeq 0.3\ K \text{ at } T=50\ \text{mK}\ . \tag{A3}$$

One can therefore ignore the internal modes at mK temperatures and the polarons respond as a rigid body. The internal modes can however, affect the frequency dependence of T_1. We outline an approximate calculation of these energy levels.

If the vacancy wave function is $\psi(r)$, the lowest mode can be calculated variationally using $\psi(r)=e^{-\alpha r^2}$ with α as the variational parameter. The lowest state, corresponding to orbital quantum number $l=0$, is given by

$$E=\langle H\rangle=\int dv\left[\frac{\hbar^2}{2m^*}|\nabla\psi|^2+gr^3\psi^2\right]\ , \tag{A4}$$

where ψ is normalized. Equation (A4) when compared to Eq. (A1) leads to the identification

$$\frac{\hbar^2}{2m^*}=\frac{4}{3}\pi^2 ta^2\ ,$$
$$g=\frac{\sqrt{\pi}}{32}\frac{4\pi}{3}nT\ln 2 \tag{A5}$$

The finite angular momentum ($l\neq 0$) states can be obtained from an effective potential

$$V_{\text{eff}}(r)=\frac{l(l+1)\hbar^2}{2m^*r^2}+gr^3$$

and

$$E_l=\tfrac{5}{6}3^{2/5}[l(l+1)]^{3/5}\left[1+\frac{3}{\sqrt{5}}[l(l+1)]^{-1/2}\right]\left[\frac{\hbar^2 g^{2/3}}{m^*}\right]^{3/5}\ . \tag{A6}$$

These states have meaning only when they are below the cutoff $E_c=z[t(1)-t(0)]$. The cutoff describes the evident free mobility of the vacancies in a randomly polarized environment. Indeed, the identification of the binding potential as gr^3 is also questionable and the potential should be cutoff at the energy E_c. This cutoff will further lead to a rise in the eigenvalues Eq. (A6). Thus an internal mode of the vacancy can be excited. The lifetime of this level is expected to be rather short on account of the close proximity to the potential cutoff and the extended state solutions for the vacancy motion.

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