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Quantum diffusion of ^{3}He impurities in solid ^{4}He

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A theoretical analysis is presented for the temperature dependence of the diffusion of ^{3}He impurities in solid ^{4}He in the regime where the coherent impurity motion at low temperatures is interrupted by scattering from vacancies or phonons. It is shown that the observed temperature dependences can be accurately described by a model involving vacancy scattering.

I. INTRODUCTION

The defining characteristic of quantum crystals such as solid helium and solid hydrogen is the weak localization of the atoms or molecules about their equilibrium lattice sites. This characteristic is a consequence of the weak binding interactions and the light masses of the particles. The root-mean-square displacement associated with this quantum zero-point motion is as low as 18% in solid H_2 and as high as 35% of the lattice spacing in solid ^{3}He at the highest molar volume.[1] Due to a significant overlap of the single-particle wave functions in these quantum solids there is a relatively large probability that neighboring particles can exchange sites. The exchange rates are of the order of 10 MHz in solid ^{3}He (Ref. 2) and 1 kHz in solid H_2 (Ref. 3).

Defects in the form of isotopic impurities can also exchange sites with neighbors in a quantum crystal. It is, however, expected that the exchange rate of an impurity will be appreciably lower than the exchange rate of particles in the host matrix because of the lattice distortion around the isotopic defect. The increased zero-point motion of a ^{3}He impurity in a ^{4}He host leads to a compression of the ^{4}He lattice surrounding the impurity and the strain field must accompany the ^{3}He atom in an exchange process. Further exchange with other host atoms results in a quantum diffusion of the impurity that needs no thermal excitation. The diffusion is far from classical and, as a result of the strong delocalization, Andreev and Lifshitz [4] and others[5-10] have suggested that the impurities propagate coherently as wavelike excitations, often called "impuritons". Direct evidence for this wavelike propagation has not been obtained, but the analyses of the NMR observations[8-12] are consistent with this hypothesis.

One regime of the diffusion process that has not been completely understood is that of intermediate temperatures ($0.7 < T < 1.2$ K) for which elementary excitations (e.g., vacancies and phonons) are thermally excited and consequently interfere with the purely quantum diffusion observed at low temperatures. In the coherent propagation model, the impurity waves are scattered by the thermal excitations and the diffusion constant is reduced.

This is contrary to the behavior expected for classical propagation for which thermal excitations would enhance the diffusion rate. As the temperature of solid helium is increased above the intermediate temperature range to $T > 1.2$ K, the number of thermally excited vacancies becomes significant, and a new vacancy-induced diffusion is observed which is associated with impurity-vacancy exchange. In this latter regime, the diffusion constant increases exponentially with temperature, corresponding to the increase in the total fraction of thermally excited vacancies given by $x_v = \exp[-G_F/k_B T]$, where G_F is the free energy of formation of a vacancy.

The purpose of this paper is to analyze the behavior for ^{3}He impurity diffusion in solid ^{4}He at intermediate temperatures. Values of the vacancy-impurity scattering cross section have been calculated in order to compare the observed temperature dependence of the diffusion with that expected for this model concerning the diffusion of impurities in quantum crystals. A similar analysis carried out for HD diffusion in solid H_2 showed that this model provided a better description of the experimental data than models invoking phonon scattering. The theory of impuriton propagation is reviewed in Sec. II, and the results compared with experimental observations for ^{3}He motion in solid ^{4}He in Sec. III.

II. THEORY OF IMPURITY MOTION

As a result of the possibility of quantum tunneling discussed above, isotope impurities can migrate through a quantum crystal by exchanging sites with neighboring atoms. The impurity moves through a periodic potential and the good quantum number for describing its motion is the "quasimomentum" **k**. In addition, the energy $E(k)$ associated with the motion is a periodic function of the quasimomentum. There are two important characteristics of this quasiparticle representation: (i) the width $\Delta \simeq zJ$ of the energy band of the spectrum $E(k)$, where J is the tunneling frequency and z the coordination number, and (ii) the characteristic velocity of the motion given by the group velocity $v \simeq |\partial E/\partial k| \simeq a_0 J$, where a_0 is the nearest-neighbor separation.

If the concentration of impurities is sufficiently small that the impurities are outside the effective range of any strain interaction most of the time, then they can be treated as a rarified gas of quasiparticle excitations, and in analogy with the kinetic theory of dilute gases, the self-diffusion constant is given by

$$D \simeq vl \ , \tag{1}$$

where l is the mean free path for the impurities. Neglect-

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ing phonons and other thermal excitations,

$$l \simeq (n\sigma)^{-1} \simeq a_0^3/\sigma x \ , \tag{2}$$

where n is the number of impurities per unit volume, x is the impurity concentration, and σ is the scattering cross section of a propagating impurity by a defect or another impurity in the crystal. The diffusion constant in this model is

$$D \simeq \frac{J a_0^4}{\sigma x} \ . \tag{3}$$

Assuming a nearest-neighbor tunneling mechanism only, the energy spectrum $E(k)$ can be calculated for a given lattice, enabling one to determine the mean-square speed $\langle v^2 \rangle$ and the diffusion constant if the scattering mechanism is known. The latter calculation is the most difficult and the most important for a direct test of the quantum diffusion model by comparison with the experimental observations.

The quasiparticle energy spectrum $E(k)$ can be determined using a second quantization representation of the Hamiltonian describing the impurity tunneling. For a nearest-neighbor tunneling frequency J, the energy dispersion relation is

$$E(k) = \hbar J \sum_{\{\delta\}} e^{i\mathbf{k}\cdot\boldsymbol{\delta}} \ , \tag{4}$$

where $\{\boldsymbol{\delta}\}$ denotes the set of vectors specifying allowed nearest-neighbor exchanges per unit cell, and $\hbar$ is Planck's constant. For a bcc lattice, with one atom per unit cell,[3]

$$E(k) = 8\hbar J \cos[\tfrac{1}{3}k_x a_0] \cos[\tfrac{1}{3}k_y a_0] \cos[\tfrac{1}{3}k_z a_0] \ , \tag{5}$$

where $\mathbf{k} = (k_x, k_y, k_z)$. In an hcp lattice, there are two particles per unit cell,[3] and

$$\frac{E(k)}{\hbar J} = \eta \pm 2(3+\eta)^{1/2} \cos\left[\frac{\sqrt{2}}{3} k_z a_0\right] \ , \tag{6}$$

with

$$\eta = -2 + 4\cos[\tfrac{1}{2}k_x a_0] \times \left[\cos[\tfrac{1}{2}k_x a_0] + \cos\left[\frac{\sqrt{3}}{2} k_y a_0\right]\right] \ .$$

The mean-square velocity of the quasiparticle in the hcp lattice is given by the average[3] (over all components of $\mathbf{k}$),

$$|\Delta_k E|^2 = 18 a_0^2 \hbar^2 J^2 \ . \tag{7}$$

Following the treatment of Widom and Richards,[11] diffusion in the presence of scattering is given in the very dilute case by

$$D = \tfrac{1}{3} \langle v^2 \rangle \tau_c \ , \tag{8}$$

where τ_c is the mean free time between collisions, and can be determined from the phase shifts $\delta(E)$ describing the scattering. τ_c is determined from a thermal average over the full energy band and is given by[12]

$$\tau_c^{-1} = \frac{2x}{\pi \hbar \rho_0} \sum_\alpha g_\alpha \overline{\sin^2 \delta_\alpha} \ , \tag{9}$$

where α designates the representations with degeneracy g_α of the states that diagonalize the scattering matrix. Because the bandwidth of the state $E(k)$ is very small, $\Delta \leq 0.1$ K, the effective temperature dependence of τ_c would not be significant for the temperatures at which experiments have been carried out to date. It would, however, be very interesting to carry out new experiments at very low temperatures that would probe only the contribution for the lowest energy states. The two-particle density of states (assumed constant in this calculation) is represented by ρ_0. The impurity-impurity scattering cross section is

$$\sigma_{\mathrm{II}} = 2a_0^2 \sum_\alpha g_\alpha \overline{\sin^2 \delta_\alpha} \tag{10}$$

and the diffusion constant is given by[12]

$$D_{\mathrm{II}} = \frac{\pi}{8} \frac{J a_0^4}{x \sigma_{\mathrm{II}}} \ , \tag{11}$$

using $\rho_0 = 1/(48\hbar J)$ for the hcp lattice.

If the scattering particle forms bound states, Locke[12] has shown that $\sigma_{\mathrm{II}} \simeq z a_0^2$, while if no binding occurs, $\sigma_{\mathrm{II}} \simeq a_0^2$. The diffusion constant in the low-temperature regime is temperature independent and inversely proportional to the impurity concentration. If the scattering cross section is known, the tunneling frequency can be determined from the product of the diffusion constant and the impurity concentration for a series of different concentrations. Allen, Richards, and Schratter[7] assumed values of the tunneling frequency J_{34} of ^{3}He impurities in ^{4}He to be the same as those for in pure ^{3}He and from the observed diffusion constants estimated values of $\sigma_{\mathrm{II}}/a_0^2$ to vary from 48 to 173. As outlined above, we expect $J_{34} \simeq 0.1 J_{33}$ and the inferred values of σ_{II} to be reduced by a factor of 10, corresponding to the values expected for binding.

As the temperature is increased, scattering by thermally excited vacancies and phonons becomes important. Vacancies in solid helium are highly mobile and travel through the lattice by vacancy-host particle exchange with exchange rates ω_v approximately one thousand times faster than the host ^{4}He-^{4}He exchange rate.[13] The density of states for collisions with vacancies is $\rho_0 = 1/(2z\hbar\omega_v)$ and if the vacancy-impurity scattering cross section is σ_{IV}, the diffusion constant is given by[12]

$$D_{\mathrm{IV}} = \frac{\pi}{4} \frac{J^2 a_0^4}{x_v w_v \sigma_{\mathrm{IV}}} \ , \tag{12}$$

where x_v is the vacancy concentration.

Although the tunneling observed at low temperatures is a quantum effect, the actual scattering of the impurities by defects is different and can be treated classically.[14] In this approximation, the scattering cross section is computed using the standard central force approach as given by Goldstein.[15] Using a Lennard-Jones 6–12 potential to describe the interaction with parameters given for solid ^{4}He,[1] we find $\sigma_{\mathrm{IV}} = 1.40 a_0^2$. From simple diffusion theory, the effective overall diffusion in the presence of quantum tunneling and scattering by vacancies is given by

$$D_{\mathrm{Ieff}}^{-1}=D_{\mathrm{IV}}^{-1}+D_{\mathrm{II}}^{-1} \ . \tag{13}$$

At low temperatures D_{II} dominates, but on increasing the temperature the number of vacancies increases as well, and the contribution from the first term in Eq. (13) results in an exponential decrease of the effective diffusion constant. The exponential behavior appears in the temperature dependence of the vacancy concentration.

If the scattering is due to phonons, Andreev and Lifshitz[4] have shown that the temperature dependence varies as T^9 and not exponentially as it does for vacancy activation. These two temperature dependencies are very difficult to distinguish within the precision of the data, and we must rely on an absolute calculation of the phonon scattering rate to compare the theory with the data. For HD scattering in solid H$_2$, it was found[16] that the phonon scattering amplitude was too weak by a factor of 20 to account for the observed temperature dependence.

The T^9 dependence for phonon scattering can be understood very simply in terms of the arguments of Andreev and Lifshitz. At low temperatures in quantum crystals, for which $T<\theta_D$ (the Debye temperature), collisions with long-wavelength acoustic phonons play the dominant role, and the collision cross section σ_{IP} is proportional to the fourth power of the frequency, i.e., $\sigma_{\mathrm{IP}}\simeq a_0^2(qa_0)^4$, where q is the phonon wave vector. The frequency of the collisions is $\tau^{-1}\simeq N_{\mathrm{ph}}c\sigma_{\mathrm{IP}}$, where c is the velocity of sound and $N_{\mathrm{ph}}\simeq(T/a_0\theta_D)^3$ is the number of phonons, $qa\simeq(T/\theta_D)$, and $\tau^{-1}\simeq(\theta_D/\hbar)(T/\theta_D)^7$. The phonon momentum is, however, much smaller than the impuriton momentum, and the transport time τ_{tr} differs from τ (the time between collisions) by the factor $(\theta_D/T)^2$. We therefore find

$$D_{\mathrm{IP}}\simeq\frac{\hbar a_0^2J^2}{\theta_D}\left[\frac{\theta_D}{T}\right]^9 \ . \tag{14}$$

The prefactor in Eq. (14) needs to be calculated accurately in order to make a reliable comparison with experimental data. Bernier and Landesman[17] have carried out this calculation carefully for an hcp lattice and find

$$D_{\mathrm{IP}}=\tfrac{1}{3}za_0^2J_{34}^2\tau_{\mathrm{IP}} \ , \tag{15}$$

where the collision frequency

$$\tau_{\mathrm{IP}}^{-1}=\left[\frac{96\pi^9}{5}(6\pi^2)^{2/3}s^2\right]\left[\frac{T}{\theta_D}\right]^9\omega_D \tag{16}$$

and $s=\frac{1}{3}$ for ^{3}He in ^{4}He, $s=\frac{1}{2}$ for HD in H_2, and $\omega_D=k_B\theta_D/\hbar$ is the Debye frequency. Writing Eqs. (15) and (16) as $D_{\mathrm{IP}}=AT^{-9}$, we find $A\simeq6.0\times10^{-5}$ cm^2 sec^{-1} K^9 for $\theta_D\simeq30$ K and $J\simeq2.5$ MHz. There is considerable variation in the calculated value of the prefactor given by the large parentheses of Eq. (16),[10,18,19] but the values of A are in the range 10^{-4}–10^{-5}. In order to fit the experimental data however, Grigor'ev *et al.*[8] find $A\simeq2.4\times10^{-7}$ cm^2 sec^{-1} K^9 and Allen, Richards, and Schratter[7] find $A\simeq1.0\times10^{-7}$ cm^2 sec^{-1} K^9. Similar to the case of solid H$_2$,[16] we find that the theoretical value of the phonon scattering is too weak by a factor of at least 100 to explain the observed diffusion. In the following, therefore, we consider only vacancy scattering.

At the highest temperatures ($T\geq1.2$ K), the number of vacancies becomes high (approaching approximately 1% at melting) and diffusion is dominated by impurity-vacancy exchange. In this high-temperature regime, the diffusion constant is given by

$$D_v=\tfrac{1}{6}a_0^2x_vw_v \tag{17}$$

and the diffusion rate increases exponentially on approaching the melting point with

$$x_v=\exp[-\Phi_v/(k_BT)] \ , \tag{18}$$

where Φ_v is the vacancy formation energy. We assume here, as usual, that the entropy associated with vacancy formation is zero, and $\Phi_v=G_F$.

The overall diffusion constant describing the full temperature range is given by

$$D_{\mathrm{TOT}}=D_v+D_{\mathrm{Ieff}} \ . \tag{19}$$

The diffusion constants are simply added because the two contributions are independent. In the following section we compare the experimental results for very low ^{3}He concentrations with the predictions of Eq. (19).

III. ANALYSIS OF THE EXPERIMENTAL OBSERVATIONS

Three temperature regimes have been observed experimentally.[6–9] They are characterized by (i) temperature independent quantum tunneling at low temperature, (ii) the reduction of diffusion at intermediate temperatures, and (iii) vacancy-mediated diffusion at high temperatures. We will consider here only the data for the lowest ^{3}He concentrations which have been studied quite extensively as a function of temperature and molar volume, by Allen, Richards, and Schratter[7] ($10^{-3}<x_3<10^{-4}$). It is in this low-concentration regime, where the impurities are weakly interacting, that the low-temperature diffusion constants scale as $D\propto x_3^{-1}$ for coherent gaslike propagation. At higher concentrations, where the impurities are more strongly interacting, the low-temperature diffusion is then more liquidlike because the impurities are in continuous interaction. From simple models of the range of the interaction potential, the diffusion is estimated to scale as $D\propto x_3^{-4/3}$.[20,21] It is this latter regime that is appropriate for the experiments on the diffusion of HD impurities in solid para-H$_2$.[3,16]

Results obtained by Allen, Richards, and Schratter[7] from direct NMR measurements of the diffusion constant are shown in Fig. 1 for a molar volume $V_m=21.0$ cm^3 and ^{3}He concentrations of 1×10^{-4}, 2×10^{-4}, and 5×10^{-4}. The experimental results are given by the symbols and the solid lines are fits to the data using Eq. (13). Two regimes are clearly distinguished: (i) the temperature-independent regime at low temperatures

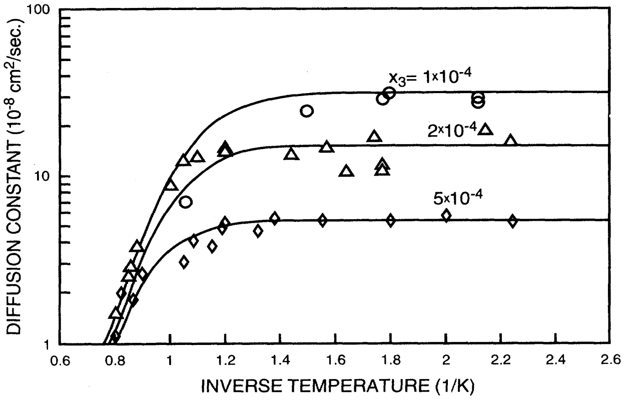

FIG. 1. Temperature dependence of the diffusion of ^{3}He impurities in hcp ^{4}He at molar volume $V_m=21$ cm^3. The experimental data are from Fig. 3 of Ref. 7: squares: $x_3=1.0\times10^{-4}$; triangles: $x_3=2.0\times10^{-4}$; circles: $x_3=5.0\times10^{-4}$. The solid lines are theoretical fits to the data as described in the text for a model in which the strong temperature dependence at intermediate temperatures is attributed to vacancy-impurity scattering.

($T<0.7$ K), and (ii) an intermediate-temperature regime ($0.7<T<1.2$K) where thermal excitations reduce the diffusion constant.

The solid lines in Fig. 1 are the calculated values of the diffusion constant using Eqs. (11)–(13). The strong dependence for $0.8<T^{-1}<0.9$ (K^{-1}) corresponds to a thermal activation energy $\Phi_v=11.5$ K, which is in good agreement with the vacancy activation energies determined by Allen, Richards, and Schratter[7] for comparable molar volumes. All three of the theoretical fits to the data were obtained using this value of Φ_v, an exchange rate $J_{34}/2\pi=2.3\times10^5$ Hz, and the experimentally observed values for $D_{\rm II}$ given by the low-temperature limits for the diffusion constant. No other adjustable parameters were used for this fit.

The collision cross section used was the theoretical value $\sigma_{\rm IV}=1.40a_0^2$ as discussed in Sec. II above. For the vacancy tunneling rate we used $\omega_v=1.2\times10^9$ sec^{-1}, corresponding to the average reported by Allen, Richards, and Schratter[7] for comparable molar volumes. The fits to the experimental data are very good, and although as shown by Allen, Richards, and Schratter,[7] a T^9 dependence provides an equally good fit within the precision of the data, one does not need to invoke a phonon interaction scattering rate 100 times stronger than the theoretical estimates.

The values of the impurity-impurity scattering cross section $\sigma_{\rm II}$ deduced from the low-temperature limits of $D_{\rm II}$ are given in Table I for $J_{34}/2\pi=2.3\times10^5$ Hz. This value of the exchange constant provided the best overall fit to the data in the intermediate temperature regime for the vacancy parameters given above. The values of $\sigma_{\rm II}$ inferred from the values of $D_{\rm II}$ are close to a factor of 2 higher than the values ($\sigma_{\rm II}\simeq z$) expected for ^{3}He-^{3}He binding into short-lived molecular states. It is important to note that Sacco *et al.*[9] were able to interpret the unusual frequency dependence of the nuclear spin-lattice relaxation times assuming that ^{3}He-^{3}He "molecular" bound states existed. In their model, the experimental results were best described with an exchange rate $J_{34}\simeq0.1J_{33}$. As discussed in the introduction, we expect $J_{34}<J_{33}$ because the impurity exchange must also involve the distortion around the impurity in the exchange process. This reduces J_{34} with respect to J_{33}. For these values of J_{34}, the scattering cross sections $\sigma_{\rm II}\simeq9a_0^2$, which are slightly smaller than the values for binding. This would require only a small reduction in $\Phi_v\rightarrow11.0$ K to obtain a good fit to the data. We therefore infer that in the absence of an independent measure of J_{34}, the analysis presented here and that of Sacco *et al.*[9] is consistent with a model in which ^{3}He-^{3}He binding does occur. Experiments to test this directly would be very valuable.

TABLE I. Parameters deduced from the low-temperature values of D (for $J_{34}\simeq0.3J_{33}$).

$D_{\rm II}$ (cm^2/sec)	x_3	$D_{\rm II}x_3$ (cm^2/sec)	$\sigma_{\rm II}/a_0^2$
3.0×10^{-7}	1×10^{-4}	30×10^{-12}	27.3
1.4×10^{-7}	2×10^{-4}	28×10^{-12}	25.5
5.5×10^{-8}	5×10^{-4}	27.5×10^{-12}	25.0

In Fig. 2, we show the results for a theoretical fit to the data of Allen, Richards, and Schratter[7] at a molar volume of 20.62 cm^3 for the full temperature range explored. These results clearly show the high-temperature regime where the diffusion is enhanced by vacancy formation. The solid line represents the diffusion constant calculated using Eq. (19). The vacancy formation energy $\Phi_v=10.5$ K was determined from the best fit to the slope in the high-temperature regime, but all other parameters were taken to be the same as for the theoretical fits used for the data of Fig. 1. The overall fit for only one adjust-

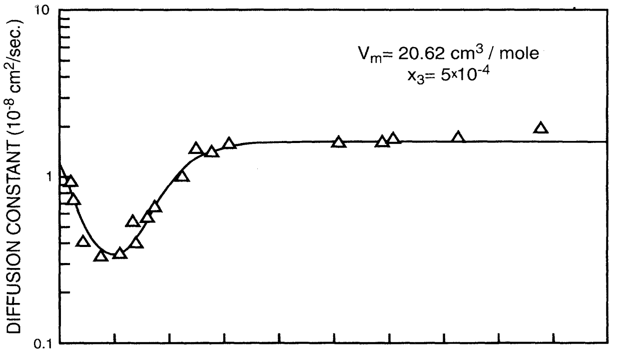

FIG. 2. Temperature dependence of the diffusion of ^{3}He impurities in solid ^{4}He at molar volume $V_m=20.62$ cm.^{3} The experimental data represented by the solid circles are from Fig. 1 of Ref. 7. The solid line is a theoretical fit as described in the text.

able parameter is very good. The minimum occurs at a temperature where the vacancy-impurity exchange and the impurity-host particle exchanges contribute equally to the diffusion rate. This occurs for a vacancy concentration

$$x_v \simeq \left[\frac{3\pi a_0^2}{2\sigma_{\mathrm{IV}}} \right]^{1/2} \frac{J}{\omega_v} , \tag{20}$$

which corresponds to a temperature given by

$$T_{\min} \simeq \Phi_v \Bigg/ \left\{ \ln \left[\left[\frac{2\sigma_{\mathrm{IV}}}{3\pi a_0^2} \right]^{1/2} \frac{\omega_v}{J} \right] \right\} . \tag{21}$$

The variation of $T_{\min}$ with molar volume is therefore relatively weak because of the logarithmic dependence on ω_v/J, both of which increase with increasing molar volume. This is consistent with the observations of Allen, Richards, and Schratter (see Fig. 1 of Ref. 7).

IV. CONCLUSION

The analysis of the temperature dependence of the experimentally observed diffusion rates for very dilute ^{3}He impurities in solid ^{4}He shows that the model in which quantum tunneling is interrupted by scattering from thermally excited vacancies provides an accurate fit to the data. Estimations of the impurity-vacancy scattering cross section using a classical approximation have been used for both ^{3}He scattering in solid ^{4}He and HD scattering in solid para-H_2.[14] In the case of isotopic impurity motion in both solid ^{4}He and solid H_2, we have found that, in a direct comparison of phonon scattering with vacancy scattering, the calculated phonon scattering gives a contribution that is at the very least an order of magnitude smaller than needed to fit the data for the temperature dependence of the diffusion.

The calculations presented show that the model of coherent wavelike propagation for the motion of isotopic impurities in quantum crystals provides a good description of the data. In solid ^{4}He, the analysis of the low-temperature data supports the interpretation of ^{3}He-^{3}He impurity scattering in terms of short-lived bound $(^3\mathrm{He})_2$ molecular states. Direct experimental verification of this molecular binding would provide a valuable test of the theory and understanding of the dynamics in quantum crystals. Such a test could be carried out at ultralow temperatures ($T < 0.1$mK) for which only a fraction of the molecular states would be accessible, and the resulting change in impurity-impurity scattering would lead to an effective change in the diffusion constant. Another important test would be to study the diffusion in very high magnetic fields and low temperatures for which the nuclear spin polarization of the ^{3}He impurity is close to unity. In this case, only one channel among all the spin states of the two scattering particles is available, and a strong polarization dependence on the low-temperature diffusion constant is expected.

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