

# Quantum diffusion of He<sup>3</sup> impuritons in solid He<sup>4</sup>

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Using a special technique to reduce the effective spin-lattice relaxation time  $T_1$ , the pulsed NMR method is used to measure the diffusion coefficient  $D$  of He<sup>3</sup> in solid He<sup>4</sup> with He<sup>3</sup> concentration  $6 \times 10^{-5} \leq x \leq 1.2 \times 10^{-3}$ , at a temperature 0.4-1.4 K and a molar volume  $V = 20.95 \text{ cm}^3/\text{mole}$ . The results indicate that  $D$  increases sharply ( $D \propto T^{-9}$ ) with decreasing temperature at an He<sup>3</sup> concentration less than  $10^{-3}$ . This indicates that impuriton scattering by the phonons of the solid-helium lattice plays the decisive role. It is shown that at  $T < 0.7 \text{ K}$ , the diffusion coefficient is inversely proportional to the He<sup>3</sup> concentration in a wide range of concentrations ( $6 \times 10^5 - 2.17 \times 10^{-2}$ ). The width of the impuriton energy band is estimated by various methods at  $\sim 10^{-4} \text{ K}$ .

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A prominent place in the study of the properties of quantum crystals is occupied by a phenomenon predicted by Andreev and I. Lifshitz, namely quantum diffusion, consisting in the fact that the diffusion transport in such crystals at low temperatures is effected by quasiparticles (impuritons) that constitute delocalized impurities moving inside the crystal lattice like a gas. This circumstance leads to substantial singularities in the temperature dependence of the diffusion coefficient  $D$  of impurities in quantum crystals. Whereas in ordinary crystals  $D$  decreases exponentially with decreasing temperature, the temperature dependence of the impurity diffusion coefficient in quantum crystals takes a much more complicated form.<sup>[1,2]</sup>

Figure 1 shows schematically a curve that reflects the most characteristic singularities of  $D(T)$  for a quantum crystals. It follows from the figure that with decreasing temperature the diffusion coefficient decreases exponentially only until its thermal-activation mass-transport mechanism that governs the diffusion becomes comparable with the mechanism caused by tunnel exchange of impurity atoms with the matrix atoms. This can be followed by two possibilities:

1. At extremely low impurity concentration, a power-law increase of  $D$  should be observed with decreasing  $T$ , like  $D \propto T^{-9}$ , owing to the decreased effectiveness of the interaction between the impurity quasiparticles and the thermal phonons of the lattice.<sup>[1]</sup>

2. At higher impurity concentrations, such that the decisive role is played by impuriton collisions with one another, the exponential decrease of the diffusion coefficient gives way to a region where  $D$  is independent of temperature and is inversely proportional to the impurity concentration.<sup>[2-4]</sup> The appearance of the plateau on curve 1 at low temperature is due also to the limit imposed on the impuriton mean free path by their collisions with one another.

Questions connected with the unusual character of the diffusion in quantum crystals, and primarily in solid helium, have been considered in a number of theoretical works.<sup>[2-13]</sup> A few years ago, quantum diffusion was experimentally registered in an investigation, by an NMR method, of diffusion in the hcp phase of solid solutions

of helium isotopes. It was shown that the diffusion coefficient of He<sup>3</sup> impurities is independent of temperature at  $T < 1 \text{ K}$ <sup>[14-17]</sup> and increases in inverse proportion to the He<sup>3</sup> impurity concentration<sup>[14-19]</sup> i.e., in full agreement with the regularities that reflect the second of these cases.

The first experimental indication that the diffusion coefficient increases with decreasing temperature is contained in<sup>[14]</sup>, although the low measurement accuracy did not make it possible at that time to investigate this effect reliably. We have recently reported briefly<sup>[20,21]</sup> observation of impuriton-phonon scattering typical of the quantum-diffusion mechanism in the case of extremely low He<sup>3</sup> impurity concentrations of He<sup>3</sup> in solid He<sup>4</sup>. The present paper is devoted to an exposition of the results of further investigations of the temperature dependence of the diffusion coefficient in solid solutions of helium isotopes.

## MEASUREMENT METHOD AND TECHNIQUE

The most important factor in NMR investigations of solid helium containing small amounts of He<sup>3</sup> is to ensure sufficient sensitivity of the NMR apparatus. The latter depends on numerous factors and primarily on the intensity of the NMR signal in the tank circuit. The maximum amplitude of the emf induced in the receiving coil by the processing magnetization vector is given

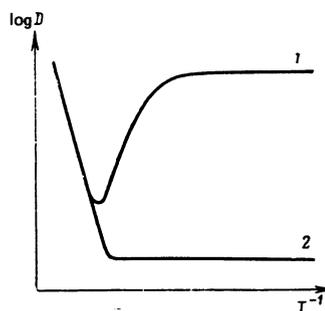


FIG. 1. Temperature dependence of the diffusion coefficient of impurities in quantum crystals (scheme): 1—impuriton scattering by phonons predominates, 2—scattering of impuritons by one another predominates.

by [22]

$$U \sim \nu_0 H_0 \chi^2 d^2 n Q, \quad (1)$$

where  $\nu_0 = \omega_0/2\pi$  is the NMR frequency,  $H_0$  is the intensity of the constant magnetic field,  $\chi$  is the magnetic susceptibility of the  $\text{He}^3$ - $\text{He}^4$  solution,  $d$  and  $n$  are the diameter and number of turns of the measuring coil, and  $Q$  is the quality factor of the tank circuit.

It follows from (1) that the intensity of the NMR signal can be increased by various methods, but analysis shows that the most effective of them entails raising the NMR frequency. It should be noted that increasing the NMR frequency in the case of weak  $\text{He}^3$ - $\text{He}^4$  solutions leads to the need of overcoming considerable difficulties that result from an abrupt increase of the spin-lattice relaxation time  $T_1$ . For temperatures below 1K the magnetic relaxation rate  $T_1^{-1}$  of  $\text{He}^3$  impurities in solid  $\text{He}^4$  (molar volume  $V = 21 \text{ cm}^3/\text{mole}$ ) is described by the relation [19]

$$T_1^{-1} = k \exp(-\omega_0/\omega_c), \quad (2)$$

where  $k = 77 \text{ sec}^{-1}$  and  $\omega_0/2\pi = 1.17 \text{ MHz}$ . An estimate of  $T_1$  for a concentration  $x = 10^{-4}$  and for  $\omega_0/2\pi = 10 \text{ MHz}$  leads to  $T_1 \approx 7 \times 10^5 \text{ sec}$ .

Since the use of the pulsed NMR method under equilibrium magnetization conditions calls for pauses  $\Delta t \geq T_1$  to follow each application of the high-frequency pulses to the sample, large values of  $T_1$  hinder the use of high NMR frequency for the investigation of weak  $\text{He}^3$ - $\text{He}^4$  solutions. Preliminary saturation of the spin system [23,24] is sometimes used under these conditions; this method eliminates in part the difficulties connected with large values of  $T_1$ . The signal  $h(0)$  observed in this case, however, is smaller than the equilibrium value  $h_0$ :

$$h(0) = h_0 [1 - \exp(-\Delta t/T_1)]. \quad (3)$$

It is easily seen that in the case of  $\text{He}^3$ - $\text{He}^4$  solutions with  $x \sim 10^{-4}$  the measured signal is less than 1% of the equilibrium magnetization signal  $h_0$  even at  $\Delta t \sim 10^3 \text{ sec}$ . Thus, when working by the usual method, [23,24] increasing the frequency produces no effect and the concentration region  $x \leq 10^{-4}$  remains practically inaccessible to NMR investigations.

The advantages offered by high frequencies can be utilized by artificially accelerating the restoration of the equilibrium magnetization. There are devices of decreasing  $T_1$ , either by introducing paramagnetic impurities into the sample [22,25] or by using an additional mechanism of relaxation on the walls of the container in which the investigated sample is located. The last procedure is successfully used in the case of liquids, particularly in the investigation of liquid  $\text{He}^3$ , [26] when the rate of magnetic relaxation is given by

$$1/T_{1m} = 1/T_{10} + 1/T_{1w}. \quad (4)$$

Here  $T_{1m}$  is the measured spin-lattice relaxation time,  $T_{10}$  is the intrinsic relaxation time of the spin system, and  $T_{1w}$  is the time of relaxation on the container walls.

It is interesting to note that in weak liquid  $\text{He}^3$ - $\text{He}^4$

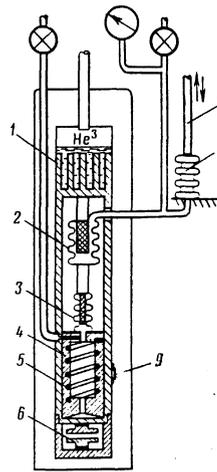


FIG. 2. Measuring cell: 1—refrigerator with pumped-on  $\text{He}^3$  vapor; 2, 8—auxiliary bellows with liquid  $\text{He}^4$ ; 3—bellows for compression of the investigated solution; 4—epoxy-resin insert; 5—NMR coil of 10 mm diameter, 6—capacitive manometer, 7—rod, 9—carbon resistance thermometer.

solutions  $T_{10}$  is also very large and for  $x \sim 10^{-4}$  the intrinsic relaxation time is  $T_{10} \sim 10^5 - 10^6 \text{ sec}$ . [27] However, such large  $T_1$  are in practice never observed in experiments with liquid helium, since the  $\text{He}^3$  atoms rapidly reach the container walls by diffusion and relax there, so that the measurements yield  $T_{1m} \sim T_{1w}$ . A detailed examination of this question has shown [28] that  $T_{1w}$  depends on the wall material and on the container dimensions, but for most experimental installations it does not exceed  $\sim 30 \text{ sec}$ .

Bearing these circumstances in mind, we developed a procedure that made it possible to obtain a practically equilibrium magnetization in solid samples. It consists of melting the sample prior to each NMR-signal measurement by lowering the pressure, maintaining the liquid state for some time to obtain equilibrium magnetization, and then allowing the sample to crystallize. This was followed by measurement of the spin-echo signal, remelting the sample, and repeating the entire procedure at different magnetic-field gradients.

To effect this procedure, additional units were introduced into the previously described [24] cryogenic installations and made it possible to vary the pressure in the cell (Fig. 2). The sample crystallization was now effected by compressing a bellows 3 with the aid of auxiliary bellows 2 and 8 filled with Liquid  $\text{He}^4$  and in which the pressure could be raised by compressing one of them (8) with a rod 7. The ratio of the cross section areas of bellows 2 and 3 was approximately 2:1, so that the  $\text{He}^4$  was prevented from crystallizing in bellows 2 and 8 at a cell pressure  $\sim 30 \text{ atm}$ . The described system ensured the possibility of crystallizing the sample within about two minutes. Increasing the duration of the process revealed no changes of any of the measured characteristics of the samples.

An essential factor in this method of obtaining solid-helium crystals is the reproducibility of the density and concentration of the  $\text{He}^3$  in the investigated samples after each melting and crystallization cycle. The sam-

ple density was monitored against the pressure in the cell with the aid of capacitive manometer  $\delta$ <sup>[24]</sup> whose sensitivity was  $\sim 0.02$  atm. The scatter of the values of the molar volume of the solid-helium sample in different experiments did not exceed  $\pm 0.05$  cm<sup>3</sup>/mole. Particular attention was paid to the reproducibility of the concentration in the investigated samples. Since the crystallization and the friction heat release in bellows 3 could give rise to a temperature gradient in the cell, the concentration of the He<sup>3</sup>-He<sup>4</sup> solution could change as a result of the thermomechanical effect in the liquid phase.

It is well known<sup>[30]</sup> that in the  $T \leq 0.08$  K region, in which the main experiments on crystallization and melting were performed,  $dP/dt$  is close to zero, so that the heat of the phase transition is negligibly small. This was confirmed, in particular, by the readings of the resistance thermometer  $\vartheta$ , which revealed no changes, accurate to  $10^{-3}$  K, of the temperature in the course of the crystallization.

The He<sup>3</sup> concentration in the sample was estimated by several independent methods: from the value of the magnetic susceptibility of the liquid and solid He<sup>3</sup>-He<sup>4</sup> solutions and from the value of the diffusion coefficient in the liquid, which varies like  $x^{-1}$  under these conditions.<sup>[31]</sup> All the methods yielded compatible values within the limits of error, which decreased (30–10%) with increasing concentration.

The solid samples for the measurement of the diffusion were prepared by two methods. In the first, the crystallization was effected at  $T \approx 0.4$  K, the sample temperature was raised to the specified value (0.4–1.4 K), and the NMR measurements were made. In the second, the measurements were made at the crystallization temperature. The values of the diffusion coefficient agreed in both cases within 15–20%, but in the former case, owing to the longer times  $T_1$  in the He<sup>3</sup>-He<sup>4</sup> solid solutions, the amplitude of the NMR signal remained larger and corresponding to a larger equilibrium magnetization at  $T = 0.4$ , thereby improving additionally the signal/noise ratio.

The NMR spectrometer operated at 9.25 MHz.<sup>[24]</sup> The Carr-Purcell<sup>[32]</sup> method and the Hahn stimulated-echo method<sup>[33,34]</sup> were used. In the Carr-Purcell method we used a sequence of three  $90^\circ$ - $180^\circ$ - $180^\circ$  pulses, and the interval  $\tau_1$  between the  $90^\circ$  and the first  $180^\circ$  pulses was chosen small enough ( $\sim 200$   $\mu$ sec) so that the amplitude of the first echo signal was independent of the magnetic field gradient  $G$  in either the solid or the liquid phase. As a result, the first signal was in fact proportional to the magnetic susceptibility of the investigated He<sup>3</sup>-He<sup>4</sup> reproducibility of the sample concentration.

The ratio of the amplitudes of the two echos produced in this sequence is described by the expression<sup>[23]</sup>

$$\frac{h_2}{h_1} = \exp \left[ -\frac{2t-2\tau_1}{T_2} \right] \exp \left[ -\frac{2}{3} \gamma^2 G^2 D (t-\tau_1)^3 \right], \quad (5)$$

where  $h_1$  and  $h_2$  are the amplitudes of the signals of the first and second echo,  $t$  is the time interval between

the two  $180^\circ$  pulses,  $T_2$  is the spin-spin relaxation time, and  $\gamma$  is the gyromagnetic ratio. In the experiment we measured  $h_1/h_2$  as a function of  $G$ . The calculation of the diffusion coefficient was by least squares using expression (5) are the corresponding expression for the stimulated-echo method.<sup>[23,24]</sup>

The described procedure yielded information on the diffusion coefficient up to values  $D \approx 10^{-10}$  cm<sup>2</sup>/sec. The error in  $D$  varied with the He<sup>3</sup> content and amounted to 15–20%.

## RESULTS AND DISCUSSION

The results of the measurements of the temperature dependence of the diffusion coefficient of He<sup>3</sup> in the hcp phase of weak He<sup>3</sup>-He<sup>4</sup> solutions are shown in Fig. 3. The curves 1, 2, and 3 correspond here to concentrations  $6 \times 10^{-5}$ ,  $5.2 \times 10^{-4}$ , and  $1.2 \times 10^{-3}$  He<sup>3</sup> in solid He<sup>4</sup>, and were obtained in the present study, while curves 4, 5, and 6, which correspond to solutions with He<sup>3</sup> contents  $2.5 \times 10^{-3}$ ,  $7.5 \times 10^{-3}$ , and  $2.7 \times 10^{-2}$  He<sup>3</sup> correspond to the previously measured data.<sup>[17,24]</sup> It is clearly seen that the character of the temperature dependence of  $D$  for the weakest He<sup>3</sup>-He<sup>4</sup> solid solutions (curves 1 and 2) differ radically from the analogous relations for more concentrated solutions (curves 4–6).<sup>1)</sup>

The exponential decrease of the diffusion coefficient on curves 4–6 gives way at high temperatures to constance of  $D$  with decreasing temperature, thus characterizing a transition from the vacancy diffusion mechanism to the mechanism of quantum diffusion limited by impuriton-impuriton collisions, in full accord with the theoretical curve in Fig. 1. With decreasing He<sup>3</sup> concentration, the temperature region of the vacancy diffusion narrows down and the plateau region broadens.

At a concentration  $\sim 10^{-3}$ , a new scattering mechanism comes into play and determines the growth of the diffusion coefficient with decreasing temperature (curve 3). With further decrease of the He<sup>3</sup> impurity concentration

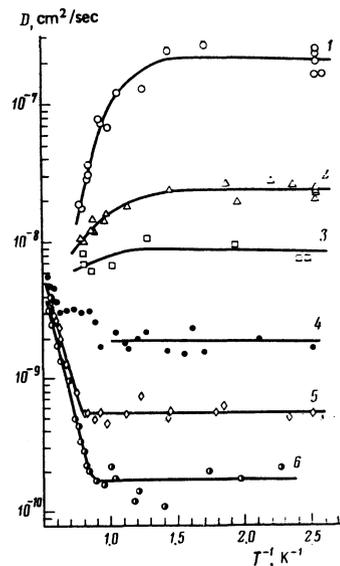


FIG. 3. Temperature dependence of the diffusion coefficient of He<sup>3</sup> in solid He<sup>4</sup> at various He<sup>3</sup> concentrations: 1 -  $x = 6 \cdot 10^{-5}$ ; 2 -  $x = 5.2 \cdot 10^{-4}$ ; 3 -  $x = 1.2 \cdot 10^{-3}$  (present results,  $V = 20.95$  cm<sup>3</sup>/mole); 4 -  $x = 2.5 \cdot 10^{-3}$ ; 5 -  $x = 7.5 \cdot 10^{-3}$ ; 6 -  $x = 2.17 \cdot 10^{-2}$  (results of earlier work,<sup>[24]</sup>  $V = 20.7$  cm<sup>3</sup>/mole).

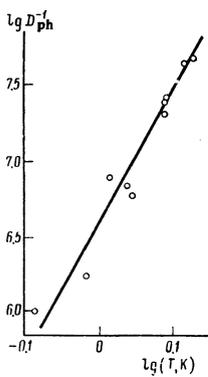


FIG. 4. Temperature dependence of phonon component  $D_{ph}$  of the diffusion coefficient at  $x = 6 \times 10^{-5}$ . The straight line corresponds to the equation  $\log D_{ph}^{-1} = 9 \log T + 6.613$ .

the region where  $D$  increases is observed more and more distinctly (curves 1, 2, and Fig. 3). At the same time, there appears in the low-temperature a section characterized by independence of the coefficient of diffusion of the temperature. For the smallest of the investigated concentrations in the temperature region 1.3–0.7 K, the diffusion coefficient increases approximately tenfold. The experimentally registered growth of the diffusion coefficient is best compared with the available theoretical calculations for the scattering of impurities by phonons.

For a quantitative comparison of theory and experiment, we reduced the experimental  $D(T)$  data under the assumption that the collisions of the impurities with one another are independent of the collisions with the lattice phonons.<sup>[2, 14]</sup> Then

$$D^{-1} = D_{imp}^{-1} + D_{ph}^{-1}, \quad (6)$$

where  $D_{imp}$  is the low-temperature diffusion-coefficient limit determined by the impuriton scattering;  $D_{ph}$  is the temperature-dependent part of  $D$  determined by the impuriton-phonon interaction. The values of  $D_{ph}$  were calculated from (6) using the experimental data on the diffusion coefficient.

The result of the calculations for  $x = 6 \times 10^{-5}$   $\text{He}^3$  plotted in coordinates  $\log D_{ph}^{-1}$  and  $\log T$ , is shown in Fig. 4, from which it follows that  $D_{ph}$  has a power-law dependence on the temperature

$$D_{ph}^{-1} = BT^n,$$

where  $B = (4.1 \pm 0.5) \times 10^6$  and  $n = 9 \pm 1$ . The obtained data are in full agreement with the theoretical conclusion<sup>[1]</sup> that the diffusion coefficient depends on temperature if the impuriton-phonon interaction is decisive. The experimental value of this dependent was first established by us in<sup>[20, 21]</sup> and was subsequently confirmed by Allen and Richards.<sup>[35]</sup> The value obtained here for the constant  $B$  also agrees with the previously obtained analogous value.<sup>[14]</sup>

Figure 5 shows a plot of  $D(x)$  in the plateau region and gives, besides the present data of the present study, also our earlier results.<sup>[17, 24]</sup> It should be noted that in a range where the concentration changes by almost three orders of magnitude all the experimental values

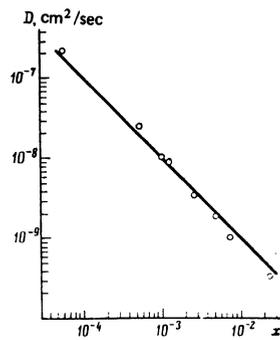


FIG. 5. Concentration dependence of the coefficient of diffusion of  $\text{He}^3$  in solid  $\text{He}^4$  ( $V = 20.95$  cm<sup>3</sup>/mole) in the region of the impuriton–impuriton interaction. The data for  $x > 1.2 \times 10^{-3}$  were taken from our earlier papers.<sup>[17, 24]</sup>

of  $D_{imp}$  satisfy well the relation  $D_{imp} = Ax^{-1}$ . The constant  $A = (1 \pm 0.2) \times 10^{11}$  cm<sup>2</sup>/sec agrees within the limits of errors with the earlier data.<sup>[17-19, 24]</sup>

It is known<sup>[14-18]</sup> that a comparison of experimental data on the concentration dependence of  $D$  with the first variant of the theory that takes the impuriton–impuriton interaction into account<sup>[3-4]</sup> has greatly underestimated the exchange integral  $J_{34}$ . The main cause of this phenomenon is the assumption, made in the calculation, that the interaction of the impurities with one another is small. Further development of the theory<sup>[2]</sup> has shown that the impuriton–impuriton scattering cross section is  $\sigma \gg a^2$  ( $a$  is the distance between atoms in the lattice) and a recent semiquantitative calculation of  $\sigma$  with account taken of the strong anisotropy of the impuriton interaction<sup>[36]</sup> yielded a value of  $J_{34}$  that differs from the corresponding value of  $J_{33}$  in pure  $\text{He}^3$  by only approximately one order of magnitude.

To explain the concentration dependence of  $D$  in the interval  $10^{-3} \lesssim x \lesssim 10^{-2}$  a model called the interaction model was proposed.<sup>[10]</sup> According to this theory impurities can be produced in solid helium only at  $x < 10^{-3}$ , and their appearance should be accompanied by a sharp increase of the diffusion coefficient in this region. The linearity of  $D(x)$  in a very wide range of concentration, established in the present study, contradicts the conclusions of the interaction model and demonstrates that it cannot be used to describe the real situation in solid helium in the investigated region.

Using the calculated values of  $A$ ,  $B$ , and  $n$ , the interpolation formula (6) (at  $V = 20.95$  cm<sup>3</sup>/mole) can be written in the form

$$D^{-1} = 10^{11}x + 4.1 \cdot 10^6 T^9 [\text{sec/cm}^2]. \quad (7)$$

Equation (7) describes satisfactorily the experimentally established temperature and concentration dependences of the coefficient of diffusion of  $\text{He}^3$  impurities in solid  $\text{He}^4$  for samples containing  $2.17 \times 10^{-2} - 6 \times 10^{-5}$   $\text{He}^3$ , and describes the influence of various scatterings of the impurity quasiparticles on the behavior of quantum diffusion in a solid.

It can thus be regarded as firmly established by now that the principal theoretically predicted singularities of the diffusion of impurities in quantum crystals ac-

tually exist, namely: the exponential decrease of  $D$ , the power-law growth of the diffusion coefficient with temperature like  $D \propto T^{-9}$ , and the independence of the diffusion of temperature and its inverse proportionality to the impurity concentration.

The presence of phonon and impuriton scattering section on curves 1 and 2 (Fig. 3) makes it possible to calculate from the experimental data the width of the impuriton energy band  $\Delta\epsilon = zJ_{34}$  ( $J_{34}$  is the He<sup>3</sup>-He<sup>4</sup> exchange integral and  $z$  is the number of nearest neighbors) independently for each quasiparticle-interaction mechanism. Estimate of  $\Delta\epsilon$  by Pushkarov's formula<sup>[12]</sup> for the diffusion coefficient, bounded by impuriton-phonon scattering:

$$D_{ph} = 2.4au \left( \frac{\Delta\epsilon}{\Theta_p} \right) \left( \frac{\Theta_p}{T} \right)^9,$$

where  $a$  is the interatomic distance,  $u$  is the speed of sound,  $\Theta_p = \Theta/8$  ( $\Theta$  is the Debye temperature), leads to a value  $\Delta\epsilon \sim 10^{-4}$  K and accordingly to  $J_{34} \sim 10^{-5}$  K. An estimate of  $J_{34}$  for the case of impuriton-impuriton scattering of quasiparticles, given in<sup>[37]</sup>, yields the same order of magnitude.

Attention is called to the fact that the exchange integral  $J_{34}$  obtained in this manner is approximately one-tenth the value of  $J_{33}$  for pure He<sup>3</sup> at the corresponding molar volume.<sup>[38]</sup>

In conclusion, the authors thank B. I. Verkin, V. A. Slyusarev, and M. A. Strzhemechnyi for useful discussions and V. A. Maidanov for help with the experiments.

<sup>1</sup>We note that curves 1-3 and 4-6 correspond to somewhat different molar volumes ( $V=21$  and  $20.7$  cm<sup>3</sup>/mole) of the solid helium. This is immaterial when diffusion mechanisms in different temperature regions are considered.

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