Spin diffusion in solid He^3-He^4 solutions

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Spin diffusion in a solid He^3—He^4 solution containing 0.75% He^3 is measured in the 1.3—2.0°K range. Spin diffusion is found to be significantly different in the hexagonal close-packed and body-centered cubic phases. The temperature dependence of the spin diffusion coefficient in both cases can be described by the classical exponential dependence.

Much attention has been paid of late to the properties of quantum crystals, particularly to effects connected with tunneling of isotopic impurities in solid helium. The most reliable information of these phenomena can be obtained by measuring the diffusion in weak He^3—He^4 solutions. In this paper we report the results of measurements of the coefficient D_S of spin diffusion in a solid solution of helium isotopes containing 0.75% He^3, in the temperature interval 1.3—2.0°K.

The published data on D_S[1-3] pertain mainly to solid He^3, for which the following has been established:

1) At high temperatures, the usual exponential decrease of the diffusion coefficient with decreasing temperature is observed:

$$D_s = D_0 e^{-\Delta/T}. \qquad (1)$$

The values of Δ range from 8 to 17°K when the molar volume V is varied from 24 to 19 cm³/mole.

2) At $T < 1$°K, the spin-diffusion coefficient of He^3 ceases to depend on the temperature down to the very lowest investigated temperatures (0.06°K according to the data of Thompson et al.[2]). Depending on the molar volume, the value of D_S in the body-centered cubic lattice of He^3 takes on values from 1.4×10^{-7} cm²/sec at $V = 24$ cm³/mole to 10^{-8} cm²/sec at $V = 20$ cm³/mole.

3) The values of D_S along the melting curve of He^3 turn out to be approximately constant and equal to 2×10^{-7} cm²/sec.

In the only published paper devoted to diffusion in He^3—He^4 solid solutions containing more than 2% of He^3 [3], direct measurements were made only at temperatures for which D_S exceeds 10^{-8} cm²/sec. It was concluded in[3] on the basis of measurements along the melting curve that D_S^{melt} is independent of the concentration, phase, and molar volume. At high temperatures and at a constant molar volume, the behavior of the diffusion coefficient is described by relation (1) with values of Δ and D_0 close to those of He^3.

The independence of the spin-diffusion coefficient of the temperature can be attributed to the fact that at $T < 1$°K, owing to the strong decrease of the diffusion mobility of the atoms, the main contribution to the magnetization transport is made by flip-flops of the neighboring spins under the influence of the exchange interaction. The independence of the exchange interaction of the temperature leads to constancy of the spin-diffusion coefficient, which in this case may not coincide with the mass diffusion coefficient D.

In this situation, particular interest attaches to an investigation of spin diffusion in solutions with low concentrations of He^3. The exchange interaction and the associated flip-flop processes are significant only for nearest neighbors, and their contributions to the spin diffusion at sufficiently low He^3 concentrations should therefore decrease, so that D_S should again coincide with D. We can then hope to observe the recently predicted singularities of the behavior of D in quantum crystals[4,5] by measuring the spin-diffusion coefficient. We note that the quantum diffusion phenomenon itself should become manifest only at sufficiently low impurity concentration[4].

We measured D_S by the spin-echo method using a previously described setup[6]. The samples were crystallized at constant volume by the capillary blocking method. The molar volume was determined from the pressure in the chamber, as measured with a capacitor pickup that registers the deformation of the bottom of the sample chamber. The spin-diffusion coefficient was determined from the dependence of the echo-signal amplitude on the magnetic-field gradient.

As is well known, at low temperatures one can obtain He^3 and He^4 crystals having BCC or HCP structures, and in the case of He^4 the phase with BCC structure exists in a rather narrow range, 1.78—1.42°K and 30—26 atm. In solutions, the region of existence of this phase is larger.

We have measured the diffusion coefficient both in the pure BCC and HCP phases of the solution, and along the liquid-crystal and BCC-HCP transition curves. The results are shown in Figs. 1 and 2. The experimental data indicate that in the investigated temperature region, both in the HCP phase (Fig. 1a) and the BCC phase (Fig. 1b) of the solution, the spin-diffusion coefficient varies with temperature in accordance with the classical exponential dependence (1) with parameters $\Delta = 13.9 \pm 1$°K and $D_S = (8 \pm 4) \times 10^{-6}$ cm²/sec for the HCP phase and $\Delta = 15.9 \pm 1$°K and $D_0 = (3 \pm 2) \times 10^{-3}$ cm²/sec for the BCC phase.

As already noted, the classical behavior of the diffusion in solid helium in this temperature region was observed earlier[1,3]. In our case, however, there are many singularities not previously observed in investigations of He^3 and of solutions with large He^3 content[1-3]. Attention is called first to the significant difference between the diffusion coefficients in the BCC and HCP phases at close values of the densities and identical temperatures[1]. This difference remains in force also for coexisting phases, as confirmed by direct measurements along the melting curve and the BCC-HCP phase transition curve.

In Fig. 2, which shows the results of measurements along the melting curve, one can see a sharp decrease

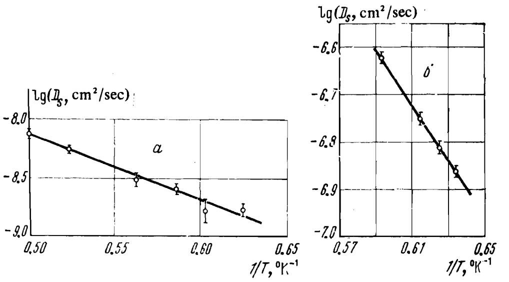

FIG. 1. Temperature dependence of the spin-diffusion coefficient: a–HCP phase, V = 20.4 ± 0.1 cm³/mole; b–BCC phase, V = 21.0 ± 0.05 cm³/mole.

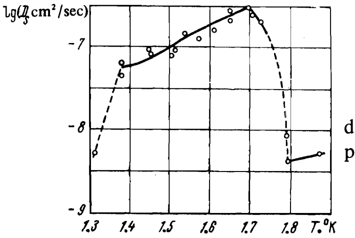

FIG. 2. Dependence of the spin-diffusion coefficient D_S on the temperature along the melting curve.

of D_S in the polymorphic transition region. This result contradicts the already mentioned statement, made by Myoshi et al.[3], that D_S^{melt} is independent of the phase. This contradiction cannot be due to the difference between the concentrations, since our measurements of the 16% solution have also shown a large difference between the values of D_S^{melt} of the BCC and HCP phases. Since the values of D_S of the HCP phase are lower than 10^{-8} cm²/sec, the limiting value that could be measured in[3], it can be assumed that the values of D_S cited in[3] pertain only to the BCC phase or to a mixture of phases (in[3] they cite only one value of D_S pertaining to the HCP phase).

Unlike the data by others[1,3], we observed in the solution investigated by us also a noticeable temperature dependence of D_S along the melting curve of the BCC phase. Unfortunately, owing to the strong increase of T_1 and the decrease of T_2, quantitative measurements of the diffusion coefficient at low temperatures are extremely difficult, and the data indicate only that D_S does not exceed a value on the order of 10^{-8} cm²/sec down to 0.4°K. This means that in the investigated solution there is no significant increase of the diffusion as a result of the effects predicted by Andreev and Lifshitz[4].

To interpret the results and to ascertain the conditions under which impurity quantum diffusion can be observed, it is important to perform detailed investigations, particularly at lower He^3 concentrations and at lower temperatures.

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1) It should be noted that an increase of the diffusion coefficient, by a factor of several times ten, was observed also in a number of metals in the FCC-BCC transition [7,8].

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