COMMENTS

Comments are short papers which criticize or correct papers of other authors previously published in **Physical Review B.** *Each Comment should state clearly to which paper it refers and must be accompanied by a brief abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

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

Dimitar I. Pushkarov*
Institute of Solid State Physics, Bulgarian Academy of Sciences, Sofia 1784, Bulgaria
(Received 3 December 1998; revised manuscript received 30 March 1999)

In this Comment I show that the experimental data on quantum diffusion of ^{3}He impurities in solid ^{4}He can be explained using the adopted quasiparticle theory. The contention by E. G. Kisvarsanyi and N. S. Sullivan in Phys. Rev. B **48**, 16577 (1993) as well as in their Reply [Phys. Rev. B **55**, 3989 (1997)] to Grigor'ev's Comment [Phys. Rev. B **55**, 3987 (1997)] that ''Pushkarov's theory of phonon scattering fails to fit the data by very large factors'' may result from an arithmetical error. This means that the phonon-impurity scattering mechanism of diffusion is consistent with experiment, and its neglect by Kisvarsanyi and Sullivan makes their results questionable.

The temperature dependence of the impurity diffusion coefficient in solid helium can be determined by two types of thermal excitations—phonons and vacancies. Both of them have been considered in a number of works (see, e.g., Refs. 1 and 2). For low concentrations and temperatures $T \leqslant 1$ K the theory based on the impurity-phonon scattering has been confirmed by experiment (cf., e.g., Refs. 1, 3, and 4 and the references therein) and used for determination of quasiparticle characteristics. The quantitative agreement was obtained in Ref. 5 (cf. also Refs. 6 and 7), and since that time the theory has not been subjected to major changes.

Kisvarsanyi and Sullivan (KS) argue in their work[8] and in their Reply[9] to Grigor'ev's Comment[10] that they have proposed ''a new theoretical treatment of the temperature dependence of the diffusion of isotopic impurities in solid ^{4}He'' as well as that my theory of phonon scattering ''fails to fit the data by very large factors.'' The latter assertion is very important because it concerns the generally accepted self-consistent approach to the diffusion in quantum crystals (see, e.g., Refs. 11, 12, 5–7, 13–16, 1, 3, and 4). It has been used as the only argument to neglect fully the phonon-impurity scattering mechanism, and turn back to the vacancy controlled impurity diffusion.

KS have evaluated in Ref. 8 the factor A in the temperature dependence of the diffusion coefficient $D = AT^{-9}$. They have found A ''to be in the range $A \sim 10^{-4} - 10^{-5}$'' while my theoretical prediction[5–7,12–14] is $A \sim 10^{-7}$ cm^2 s^{-1} K^{-9} and the experimental values are $A \approx 10^{-7}$ (Ref. 1) or $A \approx 2.4 \times 10^{-7}$ (Ref. 4). They concluded that ''the phonon scattering is too weak by a factor of at least 100 to explain the observed diffusion'' (Ref. 8, p. 16 579). KS have used in their calculation Eqs. (15) and (16) given below as Eqs. (1) and (2), respectively:

$$D = \frac{1}{3} z a^2 J^2 \tau \approx 4.3 \times 10^{-15} J^2 \tau, \tag{1}$$

$$\tau = \left[\frac{96\pi^9}{5} (6\pi^2)^{2/3} s^2 \right]^{-1} \left(\frac{\Theta_D}{T} \right)^9 \omega_D^{-1} \approx 0.79 \times 10^{-17} \Theta_D^8 T^{-9}, \tag{2}$$

where Θ_D is the Debye temperature, $\omega_D = k\Theta_D/\hbar$, $a = 3.27 \times 10^{-8}$ cm is the interatomic distance, J is the tunneling frequency, and $s = 1/3$. Hence,

$$A = 3.4 \times 10^{-32} J^2 \Theta_D^8. \tag{3}$$

Although these expressions do not exactly coincide with those used in my works, they should give a correct order of magnitude for the diffusion coefficient and, in particular, the order of A. KS have obtained for $\Theta_D = 30$ K and $J = 2.5$ MHz the value $A = 6.0 \times 10^{-5}$ cm^2 s^{-1} T,[9] while the right value [obtained by substituting the same numbers into Eq. (3)] is $A = 1.4 \times 10^{-7}$. The issue is not that the above expressions are the most fundamental or give the best value of A, but rather that they yield A values consistent with experiment. The value obtained by KS and leading to the rejection of the phonon scattering mechanism (cf., Ref. 8, p. 16 579) may come, therefore, from an arithmetical error.

In fact, $\Theta_D = 30$ K is the upper limit and corresponds to a molar volume $V_m = 19.8$ cm^3 while the experimental values cited above are for $V_m = 21$ cm^3 with $\Theta_D = 26$ K.[17] This yields $A = 0.44 \times 10^{-7}$. An ambiguity appears with the notation $J = 2.5$ MHz (p. 16 579) and $J_{34}/2\pi = 2.3 \times 10^5$ s^{-1} $= 0.23$ MHz (p. 16 580) used for one and the same quantity. I suppose that J is given in rad/s. Otherwise, it should have been written in the form $J/2\pi = 2.5$ MHz (as for J_{34}) and would be more than 10 times larger than J_{34} leading to $J = 2\pi \times 2.5 \times 10^6 \hbar/k_B = 1.2 \times 10^{-4}$ K and to an energy band width $\Delta = 2zJ \approx 3 \times 10^{-3}$ K in a drastic disagreement with all known experiments. On the other hand, the value $J_{34} \approx 1.10 \times 10^{-5}$ K is typical of the exchange integral of ^{3}He atoms in solid ^{4}He and its substitution into Eq. (3) gives the

correct order of magnitude for A. Finally, even if the unrealistic value $J=2\pi\times 2.5\approx 15.7$ MHz were used in Eq. (3) the result for A differs from that of KS by more than an order of magnitude.

It is clearly seen, therefore, that expressions (1) and (2) (not necessarily the best ones) are in agreement with the experimental data. This can be easily verified by substituting J and Θ_D. Therefore, the argument that ''the diffusion constant calculated for this theory fails to fit the experimental data by a factor of 100'' (Ref. 9) fails. There are no experimental data to require any drastic change in Eqs. (1) and (2).

It is not accurate to cite my paper[5] in Ref. 8 as if it were in support to the values of A ''in the range $10^{-4}-10^{-5}$.'' The corresponding value is $A=2\times 10^{-7}$ as follows from Eqs. (1) and (10) in Ref. 5. It is worth noting in addition that the idea of vacancy controlled mechanism could not be called *new* because it was considered about 20 years ago (cf., e.g., Ref. 2) and was found not satisfactory (cf., e.g., Ref. 1) to explain the temperature dependence of the diffusion coefficient.

As a consequence, the good fit of the vacancy mechanism reported by KS after neglecting the phonon-impurity scattering is doubtful. There are also other circumstances which call the impurity diffusion description by KS (Ref. 8 into, question, some of them being listed in Ref. 10 (see also Ref. 16). They will be considered elsewhere.

I am not concerned with the problem of impurity diffusion in solid hydrogen as it is not relevant to the immediate discussion. If the theory of KS does not work well for helium, it obviously cannot be a ''universal theory'' for both quantum solids, as claimed in Ref. 9.

*Electronic address: dipushk@issp.bas.bg

[1] A.R. Allen, M.G. Richards, and J.R. Schratter, J. Low Temp. Phys. **47**, 289 (1982).
[2] D.P. Locke, J. Low Temp. Phys. **32**, 159 (1978).
[3] V.A. Mikheev, B.N. Essel'son, V.N. Grigor'ev, and N.P. Mikhin, Fiz. Nizk. Temp. **3** 385 (1977) [Sov. J. Low Temp. Phys. **3**, 186 (1997)].
[4] B.N. Essel'son *et al.*, in Proc. 20th Soviet Conf. on Low Temp. Phys. NT-20, v.2 (1979), p. 236; J. Phys. (Paris), Colloq. **39**, C6-119 (1978).
[5] D.I. Pushkarov, Pisma Zh. Éksp. Teor. Fiz. **19**, 751 (1974) [JETP Lett. **19**, 386 (1974)].
[6] D.I. Pushkarov, Fiz. Nizk. Temp. **1**, 581 (1975) [Sov. J. Low Temp. Phys. **1**, 280 (1975)]; *ibid.* **1**, 586 (1975) [**1**, 283 (1975)].
[7] D.I. Pushkarov, Zh. Éksp. Teor. Fiz. **68** 1471 (1975) [Sov. Phys. JETP **41**, 735 (1976)].
[8] E.G. Kisvarsanyi and N.S. Sullivan, Phys. Rev. B **48**, 16 577 (1993).
[9] E.G. Kisvarsanyi and N.S. Sullivan, Phys. Rev. B **55**, 3989 (1997).
[10] V.N. Grigor'ev, Phys. Rev. B **55**, 3987 (1997).
[11] D.I. Pushkarov, Zh. Éksp. Teor. Fiz. **59**, 1755 (1970) [Sov. Phys. JETP **32**, 954 (1971)].
[12] D.I. Pushkarov, Ph. D. thesis, Moscow University, Moscow, 1972 (in Russian).
[13] D.I. Pushkarov, *Quasiparticle Theory of Defects in Solids* (World Scientific, Singapore, 1991).
[14] D.I. Pushkarov, *Defektony v Kristallakh* . . . (Defectons in Crystals. Quasiparticle Approach to Defects in Solids) (Nauka, Moscow, 1993) (in Russian) (first edition, JINR P17-87-177, Dubna, Russia, 1987).
[15] D.I. Pushkarov, D.Sc. thesis, Dubna, USSR, 1986 (in Russian); JINR Report No. 17-86-223, Dubna, Russia, 1986.
[16] V.N. Grigor'ev, Fiz. Nizk. Temp. **23**, 5 (1997) [Sov. J. Low Temp. Phys. **23**, 1 (1997)].
[17] D.O. Edwards and R.C. Pandorf, Phys. Rev. **140**, A816 (1965).