Vacancy-impurity nanoclusters in solid solutions
$^3\text{He} - ^4\text{He}$

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Abstract. Structural effects streaming from the quantum nature of the vacancies in solid helium are considered. As known, the point defects in quantum crystals are delocalized and turn into quasiparticles – defectons (vacancions, impuritons etc). An important feature is that the bottom of the quasiparticle energy band lies lower compared to the activation energy of a localized defect – as a rule by a half band width. However, the delocalization requires good periodicity and identity of the atoms around the defect. This may result both in repulsion between a vacancy and impurities, and in attraction between them. This leads to different effects – removal of impurities, new type of vacancy-impurity nanoclusters, creation of nuclei of new phases, vacancy induced local phase transition, change in the phase separation line in solid solutions. Experimental results are analyzed.

1. Introduction
The main property of the quantum crystals is the relatively large amplitude of zero-point vibrations of their atoms. This means that the wave functions of nearest atoms overlap and a nonzero probability of tunnel transitions appears. So, a new type of motion appears, and new carriers of this kind of motion arise. These are the new quasiparticles (defectons, impuritons, vacancions, interstitials, crowdions etc.). They appear due to a plain physical reason. The state of an ideal crystal with a localized defect cannot serve as a ground state because of the degeneracy with respect to the defect position (all lattice sites are equivalent). The bottom of the energy band of the delocalized defect lies lower than the activation energy of a localized defect. In most cases the lowering is by a half bandwidth, $\Delta/2$. In helium crystals with largest molar volumes the vacancion bandwidth $\Delta$ is of the order of several degrees and may even be comparable with the activation energy of an isolated vacancy. Therefore, the energy reduction is sufficient. In contrast, the bandwidth, e.g. of $^3\text{He}$ impuritons in h.c.p. $^4\text{He}$ is $10^4$ times less [1]. The situation described is closely dependent on the identity of lattice cites. This means not only that the geometrical structure must be well periodic, but also that the corresponding lattice sites are occupied by identical particles in identical states. For example, a $^3\text{He}$ atom delocalizes in $^4\text{He}$ matrix, but a $^4\text{He}$ (or a vacancy) can delocalize in $^3\text{He}$ matrix only if the spins of all $^3\text{He}$ are parallel. Otherwise, the localized vacancy will try to orient the nearest spins. Andreev [4] has supposed that a vacancy should be surrounded by such magnetic region and turn into "ferrovacancy". All attempts to observe this structure have failed (we shall discuss the reason later on, see [10] as well). We shall not discuss these works here in detail because the magnetic effect mentioned has not been confirmed yet and concerns pure helium, not solid mixtures. The point is that the vacancy behavior is different in $^4\text{He}$ and $^3\text{He}$ crystals.
Another kind of effects of vacancions on the crystal lattice structure in solid solutions were predicted by this author [7, 8, 9, 10]. It was shown that in the presence of impurities, a vacancy cleans the surrounding region. We called this removal effect or rid effect [7]. Another effect is the vacancy induced local phase transition (see below). More complicated effects were discussed in [10].

Another important phenomena is the phase separation of solid solutions $^3$He $– ^4$He. The nature of this phenomena is still not well understood. Of special interest for us will be solutions with small concentration of $^4$He.

At first glance, a delocalized vacancy should have a ferromagnetic structure proposed by Andreev [4]. We show, however, that vacancies ‘prefer’ to attract $^4$He atoms and create vacancy-impurity clusters (VIC) instead orienting spins of the nearest host atoms, $^3$He.

The paper is organized as follows. In section 2 we give some information on quantum vacancions in helium crystals necessary for further understanding, in section 3 we consider effects in a rare solid solutions far from the phase separation transition (the removal effect) and an effect on impuriton diffusion, in section 4 a simple model for vacancy-impurity clusters in phase separated mixtures is considered, and a comparison with the experimental data is made, in section 5 a semi-quantitative analysis is given and another effect, the local vacancy stimulated phase transition, is considered. Results are discussed in Conclusion.

2. Vacancion spectrum in b.c.c. and h.c.p. $^4$He

The defecton bandwidth $\Delta$ is proportional to the tunnel probability amplitude $A$ (called sometimes exchange integral $J$; we prefer notation $A$ because of some uncertainty in defining $J$ in the literature). In simple lattices $\Delta = zA$ ($z$ being the number of the nearest neighbors in the lattice). The bandwidth for $^3$He impuritons in h.c.p. $^3$He as determined from the diffusion experimental data is $\Delta_{imp} \approx 10^{-4}$ K [1, 11]. Unfortunately, there is still no reliable experimental data for vacancions. First theoretical estimations [2, 3, 12] gave $A \approx 1$ K and this value has been used till now as a good approximation. The vacancion dispersion law for b.c.c. $^4$He has the form:

$$\epsilon_b(k) = \Phi + 4A_b \left( \frac{k_x a}{2} \cos \frac{k_y a}{2} \cos \frac{k_z a}{2} \right)$$

(1)

where $\Phi$ is the activation energy of a localized vacancy, and $k$ is the quasivavevector. The bandwidth is therefore $\Delta_{bcc} = 8A_{bcc}$.

The dispersion relation for h.c.p. lattice is more complicated [13, 12]:

$$\varepsilon(k)_{ac} = \varepsilon_0 + A \left\{ 7 - 4 \cos \frac{k_1 a_1}{2} \cos \frac{k_2 a_2}{2} \cos \frac{k_3 a_3}{2} \right\}$$

$$\mp \cos \frac{k_c}{2} \left[ 1 + 8 \cos \frac{k_1 a_1}{2} \cos \frac{k_2 a_2}{2} \cos \frac{k_3 a_3}{2} \right]^{1/2} \right\}$$

(2)

where

$$a_1 = a [1, 0, 0], \quad a_{2,3} = a \left[ -\frac{1}{2}, \pm \frac{\sqrt{3}}{2}, 0 \right], \quad c = a \left[ 0, 0, \frac{\sqrt{8}}{3} \right]$$

(3)

are the basic lattice vectors. This spectrum consists of two branches called conditionally acoustic and optical. The dispersion law (2) is written in a form which fixes the bottom of the acoustic band at $k = 0$, so that $\varepsilon(0) = \varepsilon_0$ is its lowest level. The bandwidth of the acoustic branch is $\Delta_{ac} = 7A$ while that of the optical one is $\Delta_{opt} = 5A$. The bottom of the optical branch lies higher by $3A$. Hence, the total width of the two partially overlapping branches is $\delta_h = 8A$. The form of the energy surfaces $\varepsilon = \varepsilon(k)$ at different fixed values of $k_z$ for h.c.p. structure are given on figures. 1, 2, 3, 4.
Since we consider low temperatures typical of the phase separation quasiparticles from the acoustic branch can only participate. The corresponding effective mass is

\[ m_{\text{hcp}} = \frac{\hbar^2}{2Aa^2} \]  

while for the b.c.c. case one has

\[ m_{\text{bcc}} = \frac{\hbar^2}{A_{\text{bcc}}a^2}. \]  

As to the activation energy \( \varepsilon_0 \) its value varies sufficiently (see e.g. [14]) since there are no direct measurements. For h.c.p. \(^4\)He of largest molar volumes a more or less reliable value is 6 K.

The tunneling amplitude as well as the vacancion bandwidth are larger in the b.c.c. phase. This may have effect on the b.c.c.–h.c.p. phase transition [7, 8, 9, 10]. Let us consider pure \(^4\)He near the b.c.c.–h.c.p. phase transition line. The free energy in the two phases has one and the same value. If a vacancy appears in h.c.p. phase the energy increases by \( \varepsilon_0 \). However, the energy of the same vacancy in b.c.c. phase is lower because of the larger bandwidth and hence, larger
reduction of the activation energy (see figure 5). As a result, a cluster with b.c.c. structure can appear in the h.c.p. phase (local phase stratification) [7]. This reflects in a hysteresis in the phase transition and other properties when crossing the phase separation line [15]. We shall turn to this effect later on.

3. Quantum vacancy far from phase separation

In this section we consider a simple model of a quantum vacancy appropriate for $^4$He with a small concentration $x_3$ of $^3$He impurities. The presence of impurities breaks the lattice periodicity, so the vacancy should clean the surrounding region from them. This results in its localization in a region of radius $R$ (measured in interatomic distances $a_0 = (V_m/N_A)^{1/3}$, $V_m$ and $N_A$ being the molar volume and the Avogadro number, respectively). The lowest energy $E_0$ of a vacancion in such a region can be estimated by means of the Heisenberg uncertainty principle. This yields, having in mind relations (5):

$$E_0 = \pi^2\hbar^2/mR^2 = \pi^2A/R^2. \quad (6)$$

The removal reduces the entropy by an amount of $4/3\pi R^3S$ with $S = x\ln(e/x_3)$ being entropy per unit lattice site. Hence, the change of the free energy is

$$F = E - TS = \Phi - \Delta/2 + \pi^2A/R^2 + 4\pi R^3TS. \quad (7)$$

This expression has minimum at

$$R = \left(\frac{\pi A}{2TS}\right)^{1/5}. \quad (8)$$

The volume of the cleaned region is hence

$$V_0 = \frac{4}{3}\pi \left(\frac{\pi A}{2TS}\right)^{3/5}. \quad (9)$$

This result is restricted by the requirement that the free energy be not lower than the bottom of the vacancion zone, $\varepsilon_0 = \Phi - \Delta/2$, as well as by the obvious condition $R > a$. The latter is stronger and reads $TS < A$. In addition, the energy of the first quantum level (6) must be less than $\Delta/2$. The minimal value of $F$ is

$$F_{\text{min}} = \Phi - \frac{\Delta}{2} + \frac{5}{2}TSV_0. \quad (10)$$

Hence, the variation of $F$ depends linearly on the number of the lattice sites, $V_0$, in the cleaned region. This number is limited by the condition $5TSV_0 < \Delta$. 

Figure 5. Even if the activation energy $\phi$ is larger, the bottom of the vacancy band is lower.
The above consideration does not take into account the change of the entropy outside the cleaned region due to the change of impurity concentration. A straightforward analysis \cite{8, 10} yields

\[
R = \left( \frac{\pi A}{2T x_3'} \right)^{1/5}, \quad V = \frac{4}{3} \pi \left( \frac{\pi A}{2T x_3'} \right)^{3/5}
\]

(11)

where \(x_3'\) is the impurity concentration in the solution after cleaning. The free energy \(F_{\text{min}}\) becomes then

\[
F_{\text{min}} = \Phi - \frac{\Delta}{2} + \frac{5}{2} T x_3' V = \Phi - \frac{\Delta}{2} + \frac{5}{2} T n_x
\]

and one sees that the free energy depends on the number of impurities removed by one vacancy, \(n_x = x_3' V\). This results also in a decreasing of the equilibrium vacancy concentration, \(x_v\), compared to its value in an impurity free crystal, \(\tilde{x}_v\):

\[
x_v = \exp \left\{ -\frac{\Phi - \Delta}{2} T - \frac{10 \pi}{3} \left( \frac{\pi A}{2T} \right)^{3/5} x_3^{2/5} \right\}
\]

\[
= \tilde{x}_v \exp \left\{ -\frac{5}{2} n_x \right\}.
\]

(13)

In fact, the number of the removed impurities by a vacancy is \(n_x \sim 2 \div 5\) and the change of the concentration \(\delta x_3/x_3\) is of the order of several percents. However, this can lead to a decreasing of the equilibrium vacancy concentration by more than 2 orders of magnitude. For typical values \(A = 1\) K, \(T = 0.1\) K and \(x_3 = 1\%\), one finds from (9) \(V = 346\) lattice sites. The dependence of the number of atoms in the cleaned region on temperature (at constant \(x_3')\) corresponding to formulas (9) and (21) are shown in figure 6.

Let us note that the cleaned regions are not clusters. They consist of solvent atoms and do not have well expressed boundaries, surface energy etc. They have, however effect on the kinetic properties. The removal effect can be considered as an effective repulsion which prevents impurities and vacancies to be closer to each other. The impuriton energy band is very narrow (\(\sim 10^{-4}\) K) and impuritons are scattered even on a small barriers. The corresponding cross-sections are very large \cite{2, 1, 11}. A rough estimation can be made using \(R\) as a measure of the scattering distance. Then, the cross-section is of the order of \(\sigma \sim \pi R^2 \sim \pi (\pi A/2T x_3) T^{2/5} \sim 60 a^2\) (at \(T = 100\) mK). In fact, the cross-section can be smaller, because it depends on the way the wave function vanishes at distance \(R\) and this asymptotic can be smooth. The diffusion coefficient, however, has a specific dependence on temperature and concentration:

\[
D = D_0 \frac{T^{2/5}}{x_3^{3/5}}.
\]

(14)
If such dependencies could be confirmed experimentally this will be an evidence for a new mechanism of impuriton-vacancion scattering.

Concluding this section let us emphasis once more that all the consideration was made for concentrations far from the saturation concentration $x_s$. If $x' > x_s$ then a new phase of impurities appears. The effect of vacancies will be analyzed in the next section.

4. Vacancy-impurity clusters

4.1. A simple model

Let us consider a solid solution of $^4$He impurities with a concentration $x = n/N$ where $n$ is the number of impurities, and $N$ is the number of solvent ($^3$He) atoms. If $x < x_s$ the Gibbs potential $G$ may be written in the form [20]

$$G_1 = N\mu_0 + nT\ln\frac{n}{N_v} + n\psi$$

(15)

where $\mu_0$ is the chemical potential of the pure solvent, and $\psi = \psi(P, T)$. The chemical potentials $\mu_3$ and $\mu_4$ of the solvent atoms and impurities are derivatives of $G$ with respect to $N$ and $n$:

$$\mu_3 = \mu_0 - Tx, \quad \mu_4 = T\ln x + \psi.$$ 

(16)

If one moves $\delta n$ impurities from the solution to their "pure" condensed phase, the change of the Gibbs potential is

$$\Delta G = -\delta n(T\ln x + \psi) + \delta n\mu_4^0.$$ 

(17)

The chemical potential $\mu_4^0$ of the pure phase corresponds in equilibrium to a saturation concentration $x_s$ (otherwise this phase will dissolve). Hence, $\mu_4^0 = T\ln x_s + \psi$, and

$$\Delta G = -\delta nT\ln\frac{x}{x_s} = -NT\delta x\ln\frac{x}{x_s}, \quad \delta x = \delta n/N.$$ 

(18)

Therefore, the entropy difference per lattice site is

$$\Delta S = -\delta x\ln\frac{x}{x_s}.$$ 

(19)

Suppose that all $\delta n$ impurities are confined to $n_v = x_vN$ vacancies. Then $\delta n = N x_v V$ where $V = 4/3\pi R^3$ is the vacancy cluster volume (the number of impurities around one single vacancy) and $\Delta S = -4/3\pi R^3 x_v \ln N$. With this expression for the entropy the free energy (c.f. (7)) takes the form

$$F = x_v \left( \Phi - \frac{\Delta}{2} + \frac{\pi^2 A}{3 R^2} \right) + \frac{4}{3} \pi R^3 x_v T |\ln x - x_s|$$

(20)

and reaches its minimum at

$$R = \left( \frac{\pi A}{2T|\ln(x/x_s)|} \right)^{1/5}, \quad V = \frac{4}{3} \pi R^3.$$ 

(21)

Equation (21) is restricted by the conditions $T|\ln(x/x_s)| < A$ and $x \neq x_s$. When the concentration $x$ approaches the saturation concentration $x_s$ the entropy term vanishes and the vacancy is fully delocalized inside the impurity phase. This effect could be observed as a rapid increase of its mobility and growth of $^4$He domains. In terms of a phase transition from localized to delocalized motion, the domain size $R$ may be considered as an order parameter. The phase transition is continuous and may find analogy with the observed transitions in [16, 14] as well as to the revealed fast travelling gap nonphonon excitations.
4.2. Experimental Evidence

Although the vacancy-impurity clusters were predicted by this author already in 1978 [7] they were discovered experimentally by the Kharkov group only in 2001 [17] (see also [18, 19, 21]) when investigating phase separation in solid helium mixtures by precise pressure measurements. In their experiments Ganshin et al. [17] found an essential difference between the pressure behavior upon temperature cycling in phase separated $^4$He-$^3$He solid solutions in the two limiting cases: i) small concentration of $^4$He, and ii) small concentration of $^3$He. In the latter case the appearance of solid domains of $^3$He was accompanied with pressure lowering. Results were reproducible with temperature cycling. If the cycling starts from a solution with liquid $^3$He inclusions, then the pressure increases monotonically with the cycle number.

In contrast, the pressure amplitude at temperature cycling in the case of $^3$He with $^4$He inclusions decreases, and after several cycles stabilizes with an amplitude approximately 3 times less than the initial one. The initial pressure amplitude was restored only after heating the system to a temperature appreciably higher (by several tens mK) than the phase-separation temperature. The qualitative explanation was given in the frame of the above theory. The first cooling starts separating and some $^4$He rich domains appear. This results in increasing pressure. On subsequent heating these domains dissolve and the pressure rapidly decreases initiating creation of nonequilibrium vacancies. If a vacancy appears inside or on the boundary of the domain, a vacancy-impurity cluster arises. This cluster is more stable and survives (if temperature is not much higher the separation temperature, $T_s$) in contrast to the usual domains which dissolve. The dissolution process is stimulated in addition by the lowering of the concentration of $^4$He atoms in the solution due to the capturing atoms in VIC. After several cycles almost all impurities are sucked out. In order to restore the initial situation the clusters must dissolve at $T > T_s$.

5. Quantitative analysis

The simple model of VIC described in section II gives a correct order of magnitude for the number of atoms involved in a cluster. Equation (21) can be simplified for rough estimations. If the initial concentration is $x_0$, then the actual concentration is $x = x_0 - x_v - n_s/N \approx x_s - x_v V$ where $n_s$ is the number of atoms separated in the pure phase and the small concentration of solvent atoms, $^3$He in the pure phase $^4$He is neglected. The initial concentration drops as it should be in a case of a phase separated mixture in equilibrium. Then

$$|\ln(x)| = |\ln \left(1 - \frac{x_v}{x_s} \frac{4}{3\pi R^3} \right)| \approx \frac{x_v}{x_s} \frac{4}{3\pi R^3};$$

and (21) takes the form

$$R = \left(\frac{3A x_s}{8T x_v} \right)^{1/8}. \tag{22}$$

For $T = 150 \text{ mK}$, $x_v = 10^{-5}$, $x_s = 5 \times 10^{-3}$ one has $R \approx 2.5$ and $V \approx 60$ ($x_v V/x_s = 0.12 \ll 1$). Unfortunately this approximation requires $x_v V \ll x_s$ and is not applicable to the most interesting low temperature case where $x_s$ can be smaller than $x_v V$.

The main shortcoming in the derivation of (21) is that it does not take into account the surface energy. There are at least three problems with its introducing into consideration: i) VIC can appear both inside an existing domain, and as a separate inclusion, ii) the small size of the VIC makes its definition not clear enough, and iii) the structure of the VIC is not known. A quite sophisticated analysis was made in [21]. The authors supposed that the cluster has h.c.p. structure, and that the number of clusters equals the number of vacancies. They found for the cluster radius in the temperature region considered $R \approx (2,1 \div 4.4)\,a$. These values may have slightly changed because in the evaluation of the vacancion energy inside
the cluster. The authors used the $E_0 = \pi^2\Delta/R^2$ instead of $\pi^2A/R^2$ (see (6)). As a result, the first quantum level $E_0$ turns out to be out of the potential well (with depth $\Delta/2$), i.e. $\pi^2\Delta/R^2 > \Delta/2$ for all values of $R$ obtained and arbitrary $\Delta$. As for the surface energy and vacancy concentration fitted with $\Delta = 4$ K, the best fit has been obtained for a surface tension $\sigma = 1.43 \times 10^{-2}$ erg/cm$^2 \approx 2 \times 10^{-2}$ K/Å$^2$ and $x_v = 8 \times 10^{-5}$.

All this consideration was made assuming that VICs have h.c.p. structure while the solvent is in b.c.c. phase. However, as shown in [12] the bandwidth for a vacancion in b.c.c. $^4$He is larger than that in the h.c.p. phase. In addition, the activation energy for a localized vacancy in b.c.c. lattice is about a half of the activation energy in the h.c.p. one.[24] Hence, a vacancy can gain more energy for delocalization if the cluster undergos a phase transition from h.c.p. to b.c.c. [7]. The cubic lattice of VIC is more favorable also because it coincides with the solvent structure with the corresponding reduction of the surface energy. An estimation in [17] shows a reduction more than two orders of magnitude: $\sigma < 10^{-4}$ erg/cm$^2 \approx 10^{-3}$ K/particle. With neglecting surface energy and using the same procedure as in section II one obtains easily [7] for the number of particles in the VIC:

$$V = \frac{4}{3}\pi \left(\frac{\pi A}{2q}\right)^{3/5}$$

(23)

where $q = T\delta S$ is the latent heat per particle, and $\delta S$ is the entropy change at the transition. Extrapolating the data from [22, 23] one finds $\delta S \approx 10^{-2}$, and $q \approx 10^{-3}$ K. This yields for the radius of VIC $R \approx 4.4$ a and for the total number of atoms in a cluster $V \approx 350$. Let us note that the local phase transition into b.c.c. phase will appear even if the free energy is lower in the h.c.p. phase because the bottom of the vacancy band is lower in b.c.c. [7]. In our opinion these were the clusters observed by the Kharkov group.

6. Conclusion

We considered in this work several effects arising from the quantum nature of vacancies in $^4$He – $^3$He solid solutions. For small concentration of $^3$He impurities in $^4$He quantum effects lead to an effective repulsion between vacancies and impurities which may express itself in cleaning the surrounding region from impurities, as well in significant disturbing the impurity diffusion. Near the phase separation line in $^4$He – $^3$He mixtures with a small fraction of $^4$He atoms vacancies form clusters. Unlike the intuitive feeling that a vacancy will delocalize creating a well ordered ferromagnetic region of $^3$He atoms with aligned spins, it prefers to create a cluster (VIC) built from impurities ($^4$He). The VIC formation undergoes a local phase transition to b.c.c. structure favorable due to the larger vacancion bandwidth and smaller surface tension. This makes the cluster even more stable. The evaluated cluster size is in good agreement with the experimental observations. On the phase separation line the cluster size tends to infinity, vacancies become fully delocalized and can move fast inside the $^4$He separated phase. The effect of vacancy assisted local phase stratification should be observable and can be useful for explaining hysteresis effects in phase transitions between different solid phases as well as at liquid-solid ones.

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