Andreev-Pushkarov's vacancion clusters in phase-separated solid ^{4}He - ^{3}He mixtures

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Abstract

An anomalous behaviour of pressure found in phase-separated mixture of ^{4}He in ^{3}He at cycling the temperature explained by formation of pure ^{4}He clusters around vacancies. The thermodynamic calculation was carried out for the cluster parameters taking the change of the initial mixture concentration at finite ammount of vacancies and the surface tension at the cluster boundary into account.

Key words: quantum crystal; solid helium mixture; phase separation; vacancion; cluster.

In 1976 Andreev [1] predicted the existence of vacancion polarons (VP) in solid ^{3}He, i.e. the formation of ^{3}He clusters with the same direction of nuclear spins around vacancies. The VP formartion is expedient due to the increase of the vacancy localization area and the lowering of its energy. Some later, Pushkarov [4] paid attention that pure ^{4}He clusters can be formed around the vacancies in solid mixtures of ^{4}He in ^{3}He. Such complexes are called vacancion impurity clusters (VIC). The cluster radius was calculated in [2], but taking into account only entropy contribution to the thermodynamic potential (TP).

This paper presents results of a more consistent calculation of VIC parameters considering the change of the initial mixture concentration at finite amount of vacancies and the surface tension at the cluster boundary. We describe also the surprising effects, discovered during precise pressure measurements in phase-separated dilute mixtures of ^{4}He in solid ^{3}He.

Let y_0 be the initial ^{4}He concentration in mixture, in which VIC with radius r formed around the vacancies with concentration x_v. After the VIC formation, the ^{4}He concentration is y. Using chemical potentials of ^{3}He and ^{4}He in mixtures from [3], one can find the difference in TP for volume per a vacancy, $\Delta\Phi$, between mixture with VIC and one, separated according to phase diagram into BCC and HCP. The Fig.1. shows dependences $\Delta\Phi(y)$ calculated for $P = 33$ bar at the following values of parameters: $x_v = 10^{-4}$, $y_0 = 0.028$, the width of vacancion band $\Delta = 4$ K [4] and the surface tension at the cluster boundary $\sigma = 5.5 \cdot 10^{-3}$ erg/cm^2 [5]. One can see the well-defined minimums,

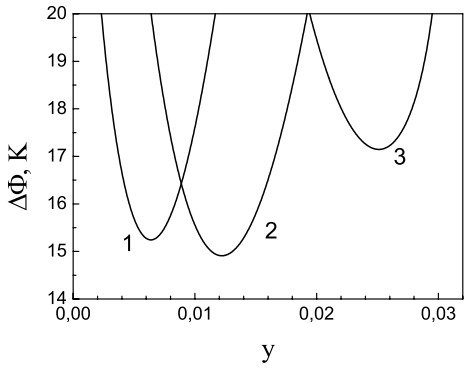

Fig. 1. The concentration dependence of $\Delta\Phi$ for different temperatures: (1) 150 mK, (2) 170 mK, (3) 200 mK.

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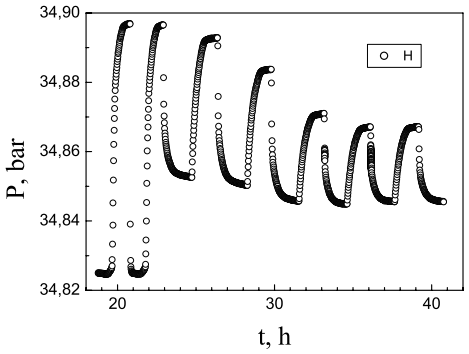

Fig. 2. The kinetics of pressure change due to phase separation at cycling the temperature from $T_m = 180$ mK to 100 mK. ($P_0 = 34.82$ bar; $y_0 = 2.8$ %)

at which the concentration corresponds to equation, which obtained from condition $\frac{\partial \Delta\Phi}{\partial y} = 0$

$$A(1-y)^2 + T_s ln y + \delta_4 + \frac{2^{7/3}}{3^{5/3}}\pi^{8/3}\Delta\left(x_v\frac{1-y}{y_0-y}\right)^{5/3} - \left(\frac{32\pi}{3}\right)^{1/3}\frac{a^2\sigma}{k}\left[\frac{(1-y)x_v}{y_0-y}\right]^{1/3} = 0, \quad (1)$$

where a is the interatomic distance (r is determined in a units). According to [3] $A = 0.76$ K and $\delta_4(P = 33$ bar$) = 27$ mK.

The experiments were carried out in the temperature range of $100-300$ mK under pressures $32.8-35.5$ bar with samples grown by blocked capillary technique from gas helium mixture, containing about 2 % ^{4}He. The samples were inside the metal cell and had shape of a disk 9 mm in diameter and 1.5 mm high. The measurement technique was described in [6]. After homogenization by cycling the temperature from the region of homogeneous mixture (~ 270 mK) to almost complete phase separation (~ 100 mK), the cycling were carried out from the minimum temperature T_f to temperature T_m which is a bit less than the phase separation temperature T_{s0} for the initial mixture. In this case, the amplitude of the pressure change $\Delta P = P_f - P_m$ ($P_i \equiv P(T_i)$) decreased at first, and then got stable (see Fig.2). The revealed behavior can be explained by assumption of appearance of excess vacancies at heating, which is accompanied by sharp pressure drop. The quasiequilibrium VIC were forming around them. As results, the surrounding mixture had weakened and the usual nuclei of new phase had turned up oversaturated and began to dissolve. After several cycles, the only VIC remained in the matrix and caused the behavior of the system.

The pressure measurements at stepwise cooling down ($\Delta T \approx 10-15$ mK) and similar heating were carried out for quantitative approval of this hypothesis. In some cases, this procedure was repeated several

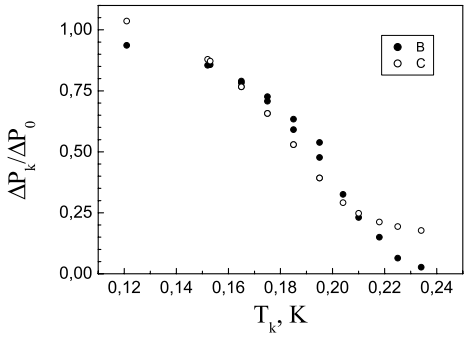

Fig. 3. The comparison between (•) experimantal and (◦) calculated relative pressure change.

times and took tens of hours. The pressure values obtained at the same temperatures were reproducible within a few mbars. These values $\Delta P_k = P(T_k) - P_0$ (P_0 is pressure in homogeneous mixture, $P(T_k)$ is equilibrium pressure at the k-th step) can be related to the change of concentration in the matrix, using the Mullin's formula [7]

$$\frac{\Delta P_k}{\Delta P_0} = \frac{y_0 - y_k}{y_0} + \frac{60x_v}{y_0}, \quad (2)$$

where ΔP_0 is the pressure change at complete phase separation. The second term in (2) is the contribution of vacancies to pressure (the estimation showed the contribution from the appearance of a vacancy is ≈ 60 times more than from the transfer of an atom to cluster). In the framework of the considering hypothesis, the values of $\frac{\Delta P_k}{\Delta P_0}$ can be calculated, using $y_k(T_k)$ from solution of (1) at various x_v, which provide the describing the experimental data in the best way. The comparison between experimental and calculated (at $x_v = 4.9 \cdot 10^{-5}$) data indicates about good agreement (see Fig.3).

The qualitative approval of the hypothesis of VIC formation is the fact, that the equilibrium value of P_0 recovered only after heating the sample to temperature, which is essentially higher ($50-70$ mK) than T_{s0}. The vacancion clusters are slow-moving and disappear only if their radius is close to a.

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