Threshold effect during dissolution of $^3$He inclusions in solid $^4$He

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A pressure jump has been found at the onset of the dissolution of bcc inclusions in separated solid $^3$He - $^4$He mixture if the crystal is overheated above a certain critical value. This effect can be explained in the framework of a multistage dissolution process model.

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1. INTRODUCTION

The interest in the investigation of the phase separation kinetics of solid $^3$He - $^4$He mixtures has been remaining for a long time. This interest is stimulated by the prospect to discover new additional features of the $^3$He impurities quantum motion in solid helium. In particular, the diffusion coefficient of impurities during the phase separation can differ from one measured in NMR experiments in a homogeneous crystal due to the influence of a finite concentration gradient and the distinction of U-processes contribution. However, the available experimental data on the phase separation kinetics appear to be badly reproducible and do not allow one to make any comparisons with the results obtained in the study of the quantum diffusion.

A good reproducibility of the experimental data was attained quite recently. The correlation was established between the results obtained in the investigation of the separation kinetics and in the NMR experiments in a homogeneous mixture. Nevertheless, it was shown in that the correlation of these data has a more complicated character. The impurity motion mechanism during the separation of solid $^3$He - $^4$He mixtures remains uncertain. This calls for continuing the investigation and in particular clearing up
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the bcc phase inclusions dissolution kinetics, which has been scarcely studied. The revealed in essential difference between inclusions growth and dissolution processes adds interest to such investigations.

This work is devoted to a detailed investigation of the \(^3\)He inclusions dissolution, which takes place at sharp warming the separated solid mixture.

2. KINETICS OF DISSOLUTION OF \(^3\)He INCLUSIONS IN \(^4\)He MATRIX

The time dependences of the pressure \(P\) at a constant volume in a sample after heating has been measured. The experimental setup and technique are discussed in detail in \(^2\). The initial concentration of the mixture was 2.05 % of \(^3\)He. The molar volume of the sample before separation was 20.44 cm\(^3\)/mole (\(P = 33.4\) bar). All the experiments were carried out with the sample after stabilization of its properties by the many times repeated cycles of the inclusions growth (at \(T \approx 103\) mK) and dissolution (at \(T \approx 230\) mK) with achievement of equilibrium at each temperature.

The main set of experiments included measurements of dependences \(P(t)\) at sharp (within several seconds) heating of the sample from the same initial temperature \(T_i =103\) mK to various final temperatures \(T_f = 108 \div 690\) mK. Results are partially presented in Fig.1. The shape of these curves is essentially conditioned by the value of \(T_f\). On small heating (Fig.1a) the pressure change \(\Delta P\) proved to be exponential as in the most of experiments on the separation kinetics of solid \(^3\)He - \(^4\)He mixtures:

\[
\Delta P = \Delta P_0 \, e^{-t/\tau},
\]

where \(\Delta P_0\) is difference of equilibrium pressure values in the sample at the initial and final temperatures.

At intermediate values of \(T_f = 127 \div 230\) mK (Fig.1b) two different parts of the dependence \(P(t)\) are observed: almost vertical part (corresponding time is of the order of the time of reaching thermal equilibrium) and smoother one described by the time constant \(\tau_{eff}\), which is of the same order as that observed at small \(T_f\). The time constant \(\tau_{eff}\) is some effective value obtained at fitting the smooth part of \(P(t)\) by one exponent. In fact, this portion of \(P(t)\) can be described by two exponents within our experimental accuracy. And finally, the pressure relaxation occurs very fast at high temperatures of \(T_f\) (Fig.1c).

The ratio of the pressure jump to the total pressure change versus \(\Delta T = T_f - T_i\) is of threshold character (see Fig.2). A pressure jump appears at some critical value \(\Delta T_c\). However, the obtained value \(\Delta T_c \approx 25\) mK is
Fig. 1. Time dependence of pressure change in the sample during $^3\text{He}$ inclusion dissolution at various temperatures: a) $T_f = 110$ mK b) $T_f = 150$ mK c) $T_f = 570$ mK. The dashed lines are fits to Eq.(1) with the corresponding time constants $\tau$ shown in this figure.
likely to be characteristic only of $T_i = 103$ mK. As it has been found in two experiments carried out with this sample that at warming the crystal from 150 mK to 230 mK the dissolution occurs without a pressure jump (see Fig.3) in spite of the fact that the value of $\Delta T$ is essentially more than 25 mK. From these two experiments it follows that it makes no sense to consider a critical chemical potential difference since in this case it is several times larger than that corresponding to the obtained $\Delta T_c$.

3. MODEL OF THE DISSOLUTION PROCESS

It is possible to explain qualitatively the observed effects, assuming that the $^3$He inclusions dissolution process consists of several stages. The essential feature of this model is the presence of a strain layer in the vicinity of the inclusions boundary due to the great difference between molar volumes of the inclusions and the matrix. The elastic potential gradient appearance may result in the mismach of energy levels in neighboring lattice sites and a restriction on impurities quantum diffusion. In particular, this circum-
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Fig. 3. Time dependence of pressure change in the sample during $^3$He inclusion dissolution under warming from $T_i = 150$ mK to $T_f = 230$ mK. The dashed line is fit to the equation: $\Delta P/\Delta P_0 = 0.0472e^{-t/\tau_1} + 0.2619e^{-t/\tau_2}$ with $\tau_1 \approx 85$ s and $\tau_2 \approx 390$ s.

stance is revealed at low temperatures when the effective diffusion coefficient, which characterizes the separation process, turns out much less than the quantum diffusion coefficient measured in NMR experiments. Therefore, quick enough substance transport near the droplets is possible only after the elimination of the strain layer.

It can be realized at the first stage of the process, when the $^3$He inclusions saturation with $^4$He atoms occurs. The diffusion penetration of $^4$He inward the $^3$He inclusion, considered as a sphere of radius $R$, will occur for the characteristic time

$$\tau_4 = \frac{R^2}{\pi^2 D},$$  \hspace{1cm} (2)

where the corresponding diffusion coefficient $D$ describes $^4$He atoms transport in solid $^3$He by random tunnel jumps. $D$ may be presented as

$$D \sim J_{43} a^2,$$  \hspace{1cm} (3)

where $a$ is the distance between the nearest neighbours and $J_{43}$ is the exchange integral, which characterizes the tunneling frequency of $^4$He atoms in
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$^3$He matrix. One would expect the magnitude of $J_{43}$ to be close to the tunneling frequency of $^3$He in $^4$He crystal $J_{34}$, which is precisely determined in quantum diffusion experiments. Extrapolating well-known values of $J_{34}$ to the molar volume $V \approx 24 \text{ cm}^3/\text{mole}$, which is characteristic of droplets, one can find $J_{34} \approx J_{43} \approx 2 \times 10^7 \text{ s}^{-1}$. It gives the magnitude of $D \sim 10^{-8} \text{ cm}^2/\text{s}$.

Then, for $R \sim 2 \mu\text{m}$ (data of S.C.J. Kingsley et al.), we obtain $\tau_1 \sim 0.1 \text{ s}$.

Hence, the first stage of the dissolution is very fast. But the pressure change $\Delta P$ at this stage must be very small because it is proportional to the product of the concentration change of $^4$He in a droplet (according to the separation phase diagram) times the volume fraction occupied by the inclusions. The corresponding value of $\Delta P$ is estimated to be not larger than 2% of the full pressure change $\Delta P_0$.

Nevertheless, this stage can strongly affect the droplets dissolution process. When leaving a strained layer around droplets, $^4$He atoms destroy it and make, therefore, possible free motion of the impuritons, which determines the second stage of the process. The characteristic time for this stage is determined by the velocity of the $^3$He atoms egress from inclusions which can be estimated by analogy with evaporation of liquid. The number of atoms, evaporating from a unit of surface per unit of time, is determined by

$$\alpha \sim \vartheta N,$$  \hspace{1cm} (4)

where $\vartheta$ is the mean velocity of atoms in vapor and $N$ is density of the atoms.

In our case $\vartheta \sim a \Delta_3$ (see) ($\Delta_3$ is impuritons bandwidth in hcp phase). The density of $^3$He atoms in mixture $N_3$, which is proportional to the concentration, serves as $N$.

$$N_3 = N_m x,$$ \hspace{1cm} (5)

where $N_m$ is density of the atoms of the mixture.

Then, it is possible to find out evaporation time for a droplet of radius $R$, neglecting the difference in molar volumes of the inclusions and surrounding mixture

$$\tau \sim \frac{R}{3 \Delta_3 a x},$$ \hspace{1cm} (6)

and assuming in Eq.(6) $R \sim 2 \mu\text{m}$ (data of S.C.J. Kingsley et al.), $\Delta_3 \sim 10^7 \text{ s}^{-1}$ (from) and $x = 10^{-2} \%$ (from separation phase diagram) gives $\tau \sim 1 \text{ s}$. This stage turns out quick enough too.

The spherical layer, which is rich in $^3$He, is formed around the droplet after this stage. For $x \sim 10^{-2} \%$ $^3$He the mean free path of $^3$He impuritons in the mixture will be $\lambda_3 \sim 10^2 a$ (from). The formation of such a layer with the estimated thickness of the order of several $\lambda_3$ ($\sim 10^{-5} \text{ cm}$) can provide
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de the value of the pressure jump $\Delta P_j$ observed in the experiment. One can see from Fig.2 that $\Delta P_j \approx 0.4 - 0.5 \Delta P_0$, where the full pressure change $\Delta P_0$ is proportional to the concentration change $\Delta x$ after heating the two-phase crystal of $\Delta T$. According to the phase separation diagram $\Delta x \approx 0.2 \%$ $^3$He for $\Delta T \approx \Delta T_c$. So, about 10 $\%$ of the $^3$He atoms in the crystal transfer from the inclusions to the matrix and this leads up to the pressure decrease $\Delta P_0$. Therefore, the observed jump $\Delta P_j$ can be provided by only about 4 $\%$ $^3$He atoms.

The propagation of $^3$He on a larger distance should occur by diffusion due to the great probability of U-processes at impurities collisions. The characteristic time for this third stage of the dissolution will be of the same order of magnitude as that obtained in other situations of the phase separation of solid $^3$He - $^4$He mixtures (see, for example, $\text{[1]}$).

The threshold effect in the proposed model takes place when the strain layer around the $^3$He droplet preventing diffusion is completely eliminated as a result of the first stage, namely the dissolution of $^4$He. Since the quantity of dissolved $^4$He is uniquely determined by the equilibrium concentration difference at the initial and at final temperatures, the critical conditions correspond to such a difference of temperatures $\Delta T_c$ at which the concentration difference $\Delta x$ provides the full elimination of the strain layer.

The equilibrium $^4$He concentration difference in the inclusion at the final temperature $T_f \sim 130$ mK, which corresponds to the critical pressure jump and the initial temperature $T_i \approx 103$ mK is about 0.2 $\%$. The spherical layer of thickness $\delta R \approx 0.7 \cdot 10^{-3} R \approx 1.4 \cdot 10^{-7}$ cm must be dissolved in the inclusion for providing such a value of the concentration difference. The critical value $\Delta T_c$ is achieved by dissolution of $3 \div 4$ atom layers. It is a value reasonable enough value for the layer thickness in which the quantum diffusion may be impeded. It should be pointed out that dissolution of $^4$He is connected with decreasing density, and it provides some additional removal of strains.

The second necessary condition to observe the pressure jump is a impurities long mean free path in the mixture at the initial concentration (temperature). The result of the above mentioned experiments with the sample warmed from 150 mK testifies in favor of this confirmation. The equilibrium concentration of $^3$He at 150 mK is about one order of magnitude more than that at 100 mK. Accordingly, the mean free path of impurities is of the same times less, and the process of inclusions "evaporation" cannot cause the perceptible pressure jump. Apparently it speeds up dissolving the inclusions essentially and provides the effect comparable with the following diffusion as a result the dependence of $\Delta P(t)$ turns out a sum of two exponents, which is illustrated in Fig.$\text{[3]}$. 

4. CONCLUSION

Thus, dissolving droplets under heating from $T_i \simeq 100$ mK to $T_f = 125 \div 230$ mK can be considered to consist of three stages: $^4$He dissolution in bcc $^3$He inclusion, which removes the strain layer around the droplet; formation of a rich $^3$He region around the inclusion; diffusion dissolution of $^3$He. The first and the second stages last for several seconds, but the third one lasts for about the time observed in other cases.

The first stage of the process will not have the above indicated concentration limitation and can provide the perceptible pressure change under higher overheating, when the sample is outside the phase separation region. It has been confirmed experimentally that in this case $\Delta P(t)$ dependence is described by a sum of two exponents.

Unequal processes of the droplets growth and dissolution are natural enough in the context of the considered model because the first of them includes only the slowest stage. In doing so, the quantum diffusion process turns out restricted at the expense of the strain layer near the droplet boundary. In this connection it should be noted that the lack of the sharp pressure jump in the experiment on dissolving the droplets, which is illustrated in Fig.6, is related with warming the sample much slower (as compared with the procedure used in the present work).

At present the experimental data which contradict the supposed dissolution model are obviously unknown. One will be able to say with a fair degree of confidence about its reality only after the verification of predictions made on its basis. Such predictions may be the following: expected decrease in the pressure jump and gradual disappearance of this jump owing to reducing the mean free path of impurities at increasing $T_i$; increase in the value of $\Delta T_c$ for the liquid $^3$He droplets (at a lower pressure). It must cause increasing the strain near the droplet boundary due to the density difference enhancement.

Our experiments will be continued in this direction. A theoretical treatment of the questions considered would be desirable.

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