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Search for the stabilizing effect of magnetic field on a gas of He_2^* excitations in liquid helium phases

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The absorption spectra of triplet He_2^* *metastable molecules in normal liquid* ^3He *under various pressures have been observed for the first time. Molecules are generated as a result of recombination of positive and negative ions extracted from discharge regions near sharp tips operated in field-emission/field-ionization modes. We have shown experimentally that this method allows one to create measurable concentrations of triplets without essential overheating of normal liquid in the cell.*

The decay of the molecules after the current through the cell is turned off have been studied in various magnetic fields up to 5.5 T. Experimental values of the mutual recombination coefficient agree with the theory of diffusion-limited recombination of excimers and show no effect of the magnetic field. The magnetic relaxation time is found to exceed 200 msec under all conditions studied.

1. INTRODUCTION

The main known properties of triplet $^4He_2^*$ metastable molecules in liquid ^4He are:

The molecule is formed as a result of interaction between two helium atoms, one in the 2^3S state and another one in the normal state. The lower atomic triplet state is metastable with the radiation lifetime[1,2] of about 8000 sec, but in condensed helium phases the 2^3S atom traps the 1^1S atom and forms the triplet molecule very fast[3] ($\sim 10^{-5}$ sec). The lower molecule triplet state is also metastable (with the radiation lifetime[4,5] of about 10 sec) and is the most long-lived neutral excitation in condensed helium.

The molecule forms a bubble in liquid helium with the diameter[6] of about 10 Å due to the repulsion of the excited electron from surrounding

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helium atoms.

The molecules in liquid helium have extremely slow rotational and vibrational relaxation: the vibrational relaxation time exceeds 100 msec and does not depend on pressure, the rotational relaxation time[7,8] is of the order of 10 msec which is 6 – 7 orders of magnitude slower than, for example, for the Na_2 molecule, which also forms a bubble in liquid helium.[9] The magnetic relaxation time[7] is larger than 60 msec, the polarization of triplets was not observed yet.

The lifetime of molecules in the liquid is limited by a mutual recombination process, and the generation of an observable concentration of excimers ($\sim 10^{12} \div 10^{13}\,\mathrm{cm}^{-3}$) decreases the lifetime to milliseconds. The bilinear recombination process

$$\mathrm{He}_2^*(\mathrm{a}^3\Sigma_\mathrm{u}^+) + \mathrm{He}_2^*(\mathrm{a}^3\Sigma_\mathrm{u}^+) \begin{array}{l} \nearrow 3\mathrm{He}(1^1\mathrm{S}) + \mathrm{He}^+ + \mathrm{e}^- \\ \searrow 2\mathrm{He}(1^1\mathrm{S}) + \mathrm{He}_2^+ + \mathrm{e}^- \end{array}$$

has to be prohibited for molecules with equal non-zero projections of spin. So, in the case of full polarization,[4] the lifetime of molecules must be of the order of 1 sec at concentrations $\sim 10^{17}\,\mathrm{cm}^{-3}$. This long-lived state would be of great interest as a new bose gas of excitations. There are some reasons for the magnetic relaxation in liquid ^{4}He to be slow: possible relaxation channels, *e.g.* spin-orbit or spin-lattice interactions are very weak in this case.

In the present work we attempted to observe polarization of molecules in liquid ^{3}He under high pressures. In ^{3}He there exists a much stronger mechanism of magnetic relaxation – the fluctuation of the local magnetic field induced by diffusion motion of surrounding ^{3}He atoms. Reichert and Jarosik[10] have observed the polarization of the excess electrons in liquid ^{3}He and found the magnetic relaxation time to be of the order of 10 msec under high pressure ($20 \div 30$ bar).

2. EXPERIMENTAL TECHNIQUE

There are a few ways to excite liquid helium and generate molecules, *e.g.* bombarding by electrons or discharge near a sharp tip. We have used a technique described in our previous works.[7,8] Molecules are formed in a counterflow of positive and negative ions extracted from discharge regions near two sharp tips operating in field-emission/field-ionization modes. The recombination of ions leads to the triplet molecule appearance with high probability.

Our method allows one to observe molecules far from strongly excited regions and has a reasonable efficiency: a pair of ions, passing through 7 –

8 kV voltage difference forms the molecule with high probability. In addition, the adequate model was developed for the ion's recombination and excimer generation rate.[8]

Molecules were detected by measuring light absorption spectra near 1 μ ($a^3\Sigma_u^+ \rightarrow c^3\Sigma_g^+$) and 2 μ ($a^3\Sigma_u^+ \rightarrow b^3\Pi_g$). The light was modulated with a chopper at 250 Hz and the photodetector output was demodulated by lock-in amplifier. The tip voltage was also modulated at $1 \div 5$ Hz and the lock-in amplifier output was demodulated by a computer. This method allowed us to measure absorption spectra with the accuracy of $3 \cdot 10^{-6}$. To observe the decay of molecules, the current through the cell was modulated, while the photodetector output was amplified and averaged over about 10 000 periods. Experimental curves were fitted by the following equations:[8]

$$dn/dt = BI(t) - \alpha n^2(t)$$

$$n(t+T) = n(t)$$

$$A(t) = l\sigma n(t) + C$$

$$\int_0^T A(t)dt = 0,$$

where $n(t)$ is the concentration of molecules, B is the constant depending on the cell geometry,[8] $I(t)$ is the current through the cell, α is the mutual recombination coefficient, σ is the light cross-section, l is the light path, $A(t)$ is the measured relative absorption. The constant C, which can be found from the last equation, appears as a result of filtering of the DC component of the absorption signal by AC amplifier. α and σ are the parameters of fitting.

3. EXPERIMENTAL RESULTS

3.1. 20% ^{3}He $-^4$ He mixture

With the method described in the previous section we have found the mutual recombination coefficient of a 20% ^{3}He $-^4$ He mixture at 1.7 K to be about $8.5 \cdot 10^{-11}$ cm^3/sec and practically independent on pressure. Hereford[11] has derived the following relation in terms of the diffusion-limited recombination: $\alpha = \alpha_0/(1 + \alpha_0/4\pi R_0 D)$, where R_0 is the distance at which molecules recombine and α_0 is the reaction rate. We can write for the diffusion coefficient $D = 1/3v^2\tau$, where $1/\tau = 1/\tau_{rot} + 1/\tau_i$; τ_{rot} and τ_i correspond to the scattering by rotons and ^{3}He impurities respectively.

Assuming $\tau_{rot} \simeq \tau_{rot}(T)$ and $\tau_i \simeq \beta x$ (x is the ^{3}He concentration), we can obtain the following formula for the effect of the impurity on the

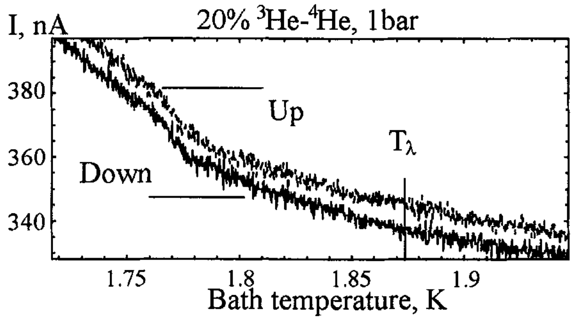

Fig. 1. Measuring the overheat of the cell with respect to the helium bath.

recombination:

$$\frac{1}{\alpha} \simeq \frac{1}{\alpha_0} + \frac{3}{4\pi R_0 \alpha_0 v^2 \tau_{rot}(T)} + \frac{3\beta}{4\pi R_0 \alpha_0 v^2} x \equiv f(T) + g(T)x.$$

Keto et al.[3] have measured the mutual recombination coefficient in superfluid liquid ^{4}He and a 1% ^{3}He $-^4$ He mixture. We have recalculated their data taking into consideration the directly measured light cross-section.[8] Taking the corrected values for 1.7 K ($\alpha(0\%) = 3.7 \cdot 10^{-10}$ cm^3/sec, $\alpha(1\%) = 3.2 \cdot 10^{-10}$ cm^3/sec) we can determine the coefficients f and g at this temperature. Substituting $x = 0.2$, we find $\alpha(20\%) = 9.0 \cdot 10^{-11}$ cm^3/sec, which coincides with our experimental data in the limits of the experimental accuracy. So, the model of diffusion-limited recombination describes correctly both the temperature dependence[3] and the dependence on the impurity concentration.

Heat release in these experiments is about ($\sim$ 2 mW), so we had to take care about overheating of the experimental volume in normal liquid. We have performed the measurement of the temperature difference between the helium bath and the cell both in normal and in superfluid states of the mixture. Applying a constant voltage to the tips, we increased and decreased the helium bath temperature, recording the current through the cell. The kink at the current – bath temperature dependence was treated as the superfluid transition because the temperature dependence of the ion's mobility in normal liquid is weaker than that in the superfluid. The difference between the kink temperature and the tabulated value of the transition gives a value of overheating of the experimental cell with respect to the helium bath. As one can see from the figure, the overheating in normal liquid is equal to the one in the superfluid and is about 0.1 K. This fact shows that the heat exchange is rather due to liquid stirring than to heat conductivity. We have corrected the measured values of the temperature by this shift.

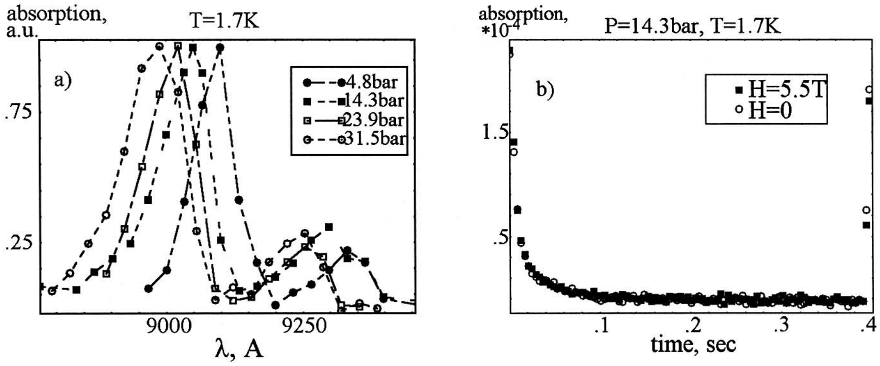

Fig. 2. Absorption spectrum of $^3He_2^*$ molecules in liquid ^3He (a) and the decay of the molecules after the current pulse (b).

3.2. ^3He

We have measured a → c absorption spectra for $^3He_2^*$ molecules at 1.7 K in a wide pressure range (fig.2a). In the previous work of our group,[8] comparing the blue shift of the (0-0) band with the pressure in liquid and solid[5] ^4He, we have assumed, that the position of the (0-0) a → c band is a universal function of helium density. Our present results, however, show that in the case of ^3He this is not justified. The dependencies on pressure are more closer to each other than those on the atomic density. The linewidth dependence on density is similar to the ^4He case.

Moreover, we have observed the decay of the molecules after the current pulse. To make sure that the lifetime of excimers is determined by recombination (but not by fluid motion, for example), we have used different durations and repetition frequencies of the current pulse. In all three cases fitting is satisfactory and gives very close values of the mutual recombination coefficient α: $6.1 \cdot 10^{-11} cm^3/sec$, $6.5 \cdot 10^{-11} cm^3/sec$ and $6.8 \cdot 10^{-11} cm^3/sec$. Nevertheless, we did not observe the suppression of recombination by the magnetic field up to 5.5 T under any pressure (fig.2b). Fitting the experimental data with the following equations

$$\frac{dn_\uparrow}{dt} = \frac{1}{3}BI(t) - \alpha n_\uparrow(n_0 + n_\downarrow) + (n\frac{b^2}{1+b+b^2} - n_\uparrow)/\tau$$

$$\frac{dn_0}{dt} = \frac{1}{3}BI(t) - \alpha n_0(n_\uparrow + n_0 + n_\downarrow) + (n\frac{b}{1+b+b^2} - n_0)/\tau$$

$$\frac{dn_\downarrow}{dt} = \frac{1}{3}BI(t) - \alpha n_\downarrow(n_0 + n_\uparrow) + (n\frac{1}{1+b+b^2} - n_\downarrow)/\tau,$$

where $n_\uparrow$, n_0, $n_\downarrow$ are the concentrations of the molecules with various projections of spin, $n = n_\uparrow + n_0 + n_\downarrow$, $b = e^{2\mu_B H/kT}$ and τ is the magnetic relaxation time, we have established that the magnetic relaxation time of triplets in liquid ^{3}He is greater than 200 msec.

4. CONCLUSIONS

Spectroscopic investigations of He_2^* in a 20% $^3He-^4He$ mixture under various pressures were performed. We have shown experimentally that our way of generation and observation of molecules did not lead to essential overheating of the normal liquid. Our experimental values of the mutual recombination coefficient are in good agreement with the diffusion-limited recombination model.

Absorption spectra of $^3He_2^*$ molecules in liquid ^{3}He in a wide pressure range were measured for the first time. The magnetic relaxation time was found to be greater than 200 msec. This is two orders of magnitude more than those for the trapped electrons.

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