Matrix isolation by solid helium

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Abstract

The new technique for embedding micro-impurities into solid helium, based on impurity-helium gas jet sedimentation on the top surface of He solid continuously moved by helium exhaust from the bottom, has been elaborated. The guest particle density of 3(1019 per cc and doped crystal growth rate of 1 mm per 20 s were achieved. CARS study of deuterium stabilized in solid He showed they mainly formed small (300 - 1000 molecules) clusters, very small aggregates were present as well.

Key words: solid helium; impurities; CARS; deuterium

1. Introduction

Embedding micro-impurities into solid helium represents an extremely intriguing experimental problem from viewpoints of spectroscopy of atoms and molecules isolated in the most inert and simultaneously quantum matrix[1] and revealing the properties of solid He in restricted geometry. Unlike the single-temperature experiments performed with molecules captured in liquid helium droplets[2], the study of impurities embedded in solid helium may be carried out in a wide range of temperatures and pressures. Unlike more heavy rare gases commonly used for matrix isolation, the introduction of impurities into solid helium seemed to be a very sophisticated problem. Only successful attempt has been known where by using the two-step laser ablation from a metal placed into solid helium the steady concentration of rubidium atoms in solid helium of about 10^8 per cc was achieved[3].

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2. Experiment

Our approach to the problem is as follows[4]. He crystal was grown in a vertical cylindrical cell, placed in the bath of a pumped LHe cryostat, $T = 1.5$ K. The upper part of the cell is connected to the high-pressure (3 MPa) helium gas supply. Then we will allow helium to leave permanently the cell from its bottom, the crystal moves down as a whole simultaneously restoring from the top by He gas input, and the crystal surface level is kept constant. In this way, in spite of steadiness of crystal shape and position, its content changes at a rate determined by the helium outflow. Because the supply tube has a small orifice as inlet at high enough outflow the gas inflow forms a needle-shaped jet[5]. Provided the traces of particles under study were added to the supplied helium gas they have been quickly transported to the condensed helium surfaces. The temperature of inlet orifice was kept high enough to prevent the impurities from freezing out, and their content was low enough to suppress their coalescence in a gas.

The cell body was sapphire tube, providing simultaneously optical transparency and good thermal contact with the main bath. The mixture source consisted

of two coaxial thin-walled stainless steel tubes of 3 and 10 mm diameter, the inner capillary has an orifice of 0.25 mm soldered at its bottom. To create impedance for the outflow an aluminum oxide mesh was installed at the bottom of the cell. A stainless steel capillary is connected the cell bottom with an out-of-cryostat needle valve to control the outflow. A flow-meter was used to control the outflow.

In some experiments a driven by outer magnet light Nickel cap was used to mark, being frozen inside solid, the rate of sample growth. The last was equal to that determined by mass outflow. Because of high heat removal capacity from the region of gas condensation through superfluid helium, the sample preparation rate was up to 1.3 mm per minute. Up to impurities concentration 1000 ppm the doped helium crystals were nicely transparent.

3. Results and Discussions

Due to large matrix shift (around 10 cm^{-1} to the red for simple molecules) and narrowness of vibration-rotational lines detected (1.4 cm^{-1} in our experiments) CARS (Coherent Anti-Stockes Raman Scattering) allows to distinguish separate molecules and their clusters and even to estimate the size of clusters. Indeed, the position of line belonged to surface molecules in a cluster should be near the half of distance between matrix and gas positions. It is easy to show that taking into account the intensity of CARS signal is proportional to square of concentration in approximation of spherical large clusters

$$\frac{\int \sqrt{I_s}\mathrm{d}\nu}{\int \sqrt{I_s}\mathrm{m}\nu} = \frac{3}{N} \qquad (1)$$

where I_s and I_m are the contributions to signal intensity from the surface and bulk, correspondingly, and N is number of layers in a cluster. The experimental line for mixtures contained 30 - 300 ppm of D_2 indeed consists of rather narrow line in a position close to that in D_2 molecular crystal and a broad blue satellite centered 4 cm^{-1} to the blue from the center. Non-zero intensity extends up to gas line position, even small peak is seen there. It proves the presence of very small D_2 oligomers in a sample. In average the estimation by (1) gives $N \simeq 4$. Under the slow frontal melting of a crystal by pressure decrease the clusters aggregated to macroscopic fractal feature just at liquid-solid interface.

The study of CH_4 molecules having much less diffusion coefficient in solid helium is now in progress.

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