Observation of excimer He_2 molecules in an He^4 crystal

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The He_2 molecules in the $a^3\Sigma_u^+$ state with a lifetime of 15 s have been detected in solid helium in the presence of a β source.

The excited helium atoms and molecules in liquid He^4 have been studied extensively.[1,2] Of greatest interest are the excimer He_2 molecules, whole lifetime in the $a^3\Sigma_u^+$ state was found to be, in contrast with the atomic excitations, rather long. According to the data of optical measurements,[2] this lifetime, which may be at least as high as a tenth of a second, is determined by the mutual recombination rate of these molecules. In veiw of this circumstance, the possibility that molecular excitations are carriers of the so-called "neutral currents" in liquid helium,[3] whose carriers can have a lieftime as high as 10 s, has been discussed in the literature. Direct optical detection of states with such a long lifetime in liquid helium is difficult because it involves dealing with very low excimer concentrations. A solid helium, whose mobility and hence recombination rate of excimer molecules are expected to be much lower compared with liquid helium, are considerably more promising in this respect. In addition, the production of excimer molecules and their study in helium crystals are of interest in their own right.

The measurement of the absorption of light in the near-IRregion ($\lambda \approx 0.9\mu$ corresponds to the transition $a^3\Sigma_u^+ \to c^3\Sigma_g^+$) was found to be the most useful method of detecting excited molecules in an He^4 crystal. Helium crystals were grown in a container which is shown schematically in the inset in Fig. 1. This ferrochromium cylinder with a 15-mm inside diameter had end-faces made from optically polished chemical glass. (The cryostat was also equipped with optical windows). As an excitation source we used disks with a β-active TiT_2 coating which were similar to those used in the measurement of the mobility of charges in helium.[4,5] Such disks (ϕmm) were embedded into two electrodes which were spaced 1.4 mm apart. The electrode planes were oriented vertically and parallel to the optical axis of the system. A voltage of up to 25 kV of both polarities was applied between the electrodes and the current flow through the crystal was measured. The saturation current of a given cell was 2.4×10^{-8} A and both sources had the same intensity.

The standard procedure was used to record the absorption. The light from a ribbon-filament lamp was transmitted through a MDR-2 grating monochromator and then divided into two beams. One beam was focused between the electrodes, as shown in Fig. 1, and the other beam was focused below the cylindrical container. The width of the beams in the apparatus was 0.2–0.3 mm in the horizontal direction and 6–7 mm in the vertical direction. Both beams were focused on the FD-24 photodiode. The rotating disk chopper passed the beams alternately. The signal from the photodiode

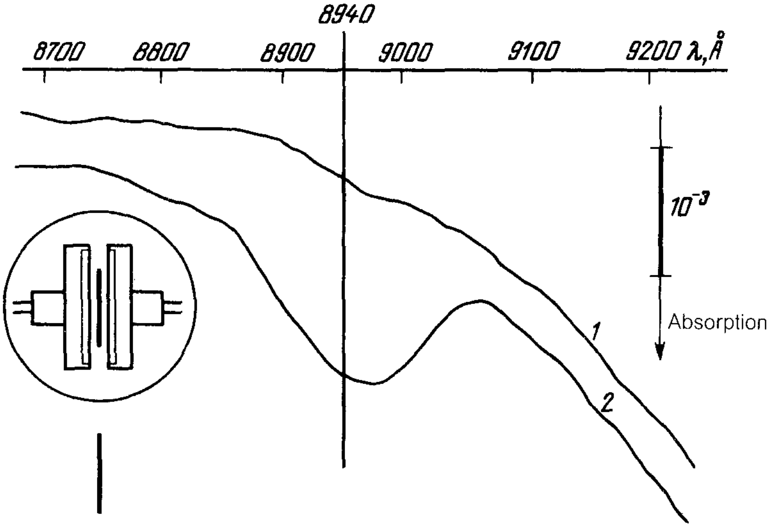

FIG. 1. Experimental trace of the absorption spectrum. 1—Measurements for a liquid; 2—measurements for a crystal.

was detected synchronously at the chopping frequency (300 Hz). The synchronous detector therefore detected the difference in the beam intensities which was recorded on an x-y plotter as a function of the wavelength or time. An example of an experimental trace with scale values of absorption and wavelength is shown in Fig. 1.

We see that the crystal has an absorption line with a maximum ~8940 Å, which corresponds to the transition $a^3\Sigma_u^+ \rightarrow c^3\Sigma_g^+$ of an He_2 molecule[1,2] and a width ~150 Å. As expected, no noticeable absorption was observed in the liquid; the curve in Fig. 1 matches, within the error of experimental measurements, the instrumental function of the detector. This result corresponds to the data of Ref. 2, according to which the concentration of the excimer molecules in the liquid is limited by their mutual recombination, and the intensity of the β source used by us turns out to be insufficient to generate a noticeable excitation.

The range of electrons emitted by the β source is on the order of several microns in helium. These electrons therefore excite the helium in the thin layer near the electrode. We were able to transmit a light beam through the crystal in such a way that it would touch one of the electrodes. In such a geometry we measured the absorption in the temperature interval 0.8–1.4 K for various values of applied electric voltage. The absorption was found to be independent of the temperature. It does, however, depend on the current flow through the crystal, as shown in Fig. 2. The voltages necessary to produce the same current at various temperatures in this case may differ by an order of magnitude.[4,5] At zero current the absorption occurs in a layer which is much thinner than the beam width, so that it probably reaches values of order 1 in this layer.

We were able to place the beam transmitted through the crystal at the center of the gap and to measure the absorption in the bulk of the crystal. The absorption in this case was also found to depend on the current that flowed through the helium, although this dependence was of a different nature. Figure 2 shows the results of mea-

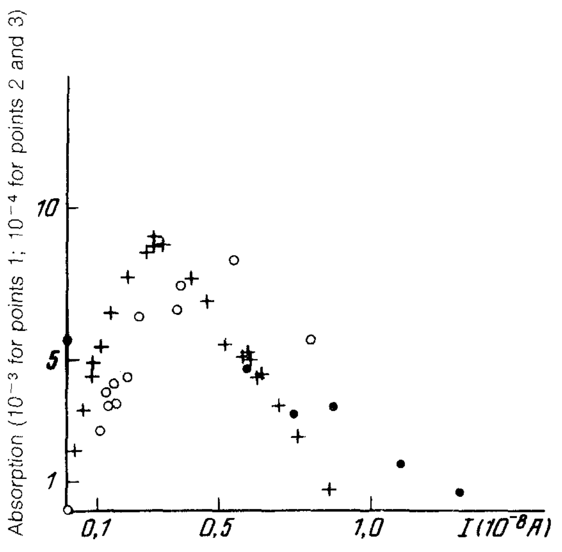

FIG. 2. Absorption versus the electric current. 1(●)—Near the electrode at $T = 1.1$ K; 2(○)—in the crystal at $T = 0.95$ K; 3(+)—absorption in the crystal at $T = 1.4$ K.

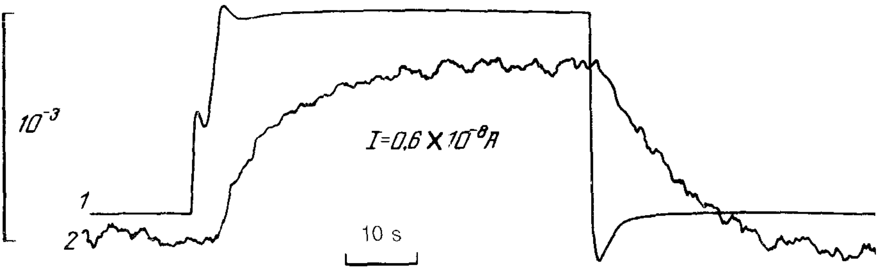

FIG. 3. The current (1) and the absorption in the crystal (2) versus the time during which the voltage is applied and the time during which it is removed.

surements at two temperatures. The absorption at the maximum $\sim 10^{-3}$ corresponds, based on our estimates, to the excimer concentration in a crystal $\sim 10^{-12}$ cm^{-3} in volume. In the calculation of the absorption cross section the oscillator strength for a given transition was assumed to be equal to 0.1 (see Ref. 2).

We wish to emphasize that at zero current we saw no absorption in the cell in the bulk of the crystal, with the exception of thin layers adjacent to the sources. In other words, we observed no excimer diffusion at distances comparable with the spacing between the electrodes.

In addition to the spectral measurements, we obtained an absorption-versus-time curve upon the application of the voltage and after removing it (Fig. 3). At the same time we measured the current. The time required to produce the necessary current in the temperature and voltage intervals indicated above varies from a second to ten seconds, in agreement with the data on the charge mobility in helium crystals.[4,5] The appreciable increase in the absorption did not begin immediately after the voltage was applied but after a time delay of several seconds which corresponded roughly to the time it took to bring up the current. The rise and fall of the absorption was assumed to obey, within the error of the experimental measurements, and exponential law, and the corresponding time constants could be calculated.

The rise time of the signal, which was ~ 15 s at low currents, decreased to 8–10 s at currents $\sim 10^{-8}$ A, regardless of the temperature. The reason for this decrease so far is unclear. The signal decayed with a time constant $\tau \approx 15$ s, which did not depend either on the current, the temperature, or the initial absorption level. This circumstance allows us to assert that the loss of excimer molecules does not stem from their mutual recombination. Measurements of the time dependence of the absorption near the electrodes showed that even there both time constants are close to 15 s.

The time constant $\tau \sim 15$ s should therefore be attributed to the intrinsic lifetime of He_2 excimers in solid He^4.

With regard to the mechanism for the formation of molecules in the $\alpha^3\Sigma_u^+$ state, we can assume on the basis of the available experimental data that they are produced as a result of recombination of the charges. At low voltages and low currents the absorption in this case should be proportional to the current (all the charges that leave the electrodes are recombined in the crystal). The probability for the formation of an He$_2$ excimer molecule upon the recombination of ions can thus be estimated from the slope of the initial part of the curves drawn through points 2 and 3 in Fig. 2. This probability turned out to be close to unity.

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