Spectral properties and the dynamics of excimer He$_2^*$ molecules in ^{4}He crystals

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The absorption spectrum of solid helium in the presence of a β-source has been measured in the wavelength range 7000–10 000 Å at pressures of 25, 31.2, and 40.5 atm. The absorption lines found correspond to the (0–0) and (1–0) $c^3\Sigma_g^+ \leftarrow a^3\Sigma_u^+$ transitions of He$_2^*$ molecules. It was established that the $a^3\Sigma_u^+$ state is formed as a result of ion recombination and that the probability of formation of an excimer molecule in the act of recombination is close to unity. The lifetime of the $a^3\Sigma_u^+$ state in a crystal was measured as $\tau_0 \approx 15 \pm 1$ s. An additional mechanism for excimer decay was also found, produced by the interaction of excited molecules with charges. The corresponding capture cross section of a molecule by an ion is $\sigma \approx (4\text{–}6) \times 10^{-13}$ cm^2.

1. INTRODUCTION

The energy spectrum of helium crystals has so far been studied almost exclusively in the range not exceeding the Debye frequency. Apart from that and at higher frequencies, starting from the infrared region and up to the vacuum ultraviolet, helium crystals should have a rather rich spectrum of excited states, both charged and electrically neutral. The results of experimental[1,2] and theoretical[3,4] investigations of the excited states of liquid helium provide evidence for this.

One of the most interesting results of these investigations was the discovery of long-lived $a^3\Sigma_u$ triplet states of molecular (He$_2^*$). According to Keto *et al.*,[1] the lifetime of such excimers at all concentrations which could be measured directly is determined by their mutual recombination; the natural lifetime, determined by radiative processes or, possibly, by non-radiative decay was in any case not less than 0.1 s. Compared with $a^3\Sigma_u^+$ all other neutral excitations, both molecular and atomic, were short-lived. It is for this reason that $a^3\Sigma_u^+$ excimers are the only real candidates for the role of carriers of the so-called "neutral currents" in liquid helium, first found by Surko and Reif[5]: the lifetime of the carriers of these currents, according to Mehrotra *et al.*[6] can reach ~ 10 s.

It would be very interesting to compare these results with results on free excimer He$_2^*$ molecules. Unfortunately such results are extremely meager. The only experimental work on this subject which we know of[7] gives an estimate below 0.05 s for the lifetime of the $a^3\Sigma_u^+$ excimer in rarefied gas. The very recent first attempt at a calculation of the radiative lifetime of the $a^3\Sigma_u^+$ state[8] gives an estimate of ~ 10 s. We do not know of any theoretical work on calculations of the lifetimes of these states in condensed helium.

An electron gun generating an electron beam with energies ~ 100 keV is customarily used as the source of the excitations in studies of the excited states of liquid helium (see Ref. 1). Such electrons are stopped in a path of 1–10 mm in liquid helium, forming a track consisting of ions and excited helium atoms. The kinetics of the reactions which take place in such a system is extremely complicated and has so far been quite insufficiently studied; in any case, several competing mechanisms can be indicated, leading to the formation of triplet He$_2^*$ states. In total these mechanisms are fairly effective: according to Keto *et al.*,[1] for a pulsed irradiation of helium by a beam of 160 keV electrons about 10% of the total beam energy goes into the formation of $a^3\Sigma_u^+$ states.

The actual process which leads to the formation of metastable states was not established in these experiments. The capture of a neighboring atom by an excited He* (2^3S) atom with the formation of an excimer He$_2^*$ molecule, although it occurs, is apparently not the main mechanism for excimers to arise. It is possible, at the same time, that in condensed helium excited by an electron beam or by some other means, together with He$^+$ ions and e$^-$ there are appreciable amounts of He$_2^+$ and He$_3^+$ ions. He$_2^*$ molecules may be formed as a result of the recombination of such ions with negative charges.

Going over to the question of the investigation of molecular excitations in solid helium, we note that only one attempt of this sort is known to us.[9] In this experiment with solid helium irradiated by an electron beam, He$_2^*$($a^3\Sigma_u^+$) molecules were observed; their lifetime was measured not directly, as in the same authors' work on the liquid,[1,2] but indirectly using the time dependence of the intensity of vacuum ultraviolet radiation, and without spectral resolution. The conclusion drawn on the basis of these results, that the kinetics of the decay of excimer molecules in a crystal was practically the same as in the liquid, was surprising even to the authors themselves. We note that under the experimental conditions[9] the possibility cannot be ruled out that the working region of the crystal melted under the action of the electron beam.

In the present work we study the spectral properties of He$_2^*$($a^3\Sigma_u^+$) excimer molecules in ^{4}He crystals, the kinetics of their formation and decay, and the interaction of these molecules with charge carriers in helium crystals. Special attention is paid to the elucidation of the role of recombination of charges in the formation of excimers. A short communication was published earlier on these experiments.[10]

2. EXPERIMENTAL METHOD

Special thin-walled windows are essential for introducing an electron beam into the cryostat; this fact highly complicates the use of electron beams when working at elevated pressures. For this reason, in experiments with solid helium, which require pressures of not less than 25 atm, we preferred to use β-sources frozen directly into the crystal being studied. The main drawback of such a source is the impossibility of controlling its intensity, which at first sight prevents any transient measurements. However, it turned out that in electric fields of $\sim 10^4$–10^5 V/cm the rate of formation of ex-
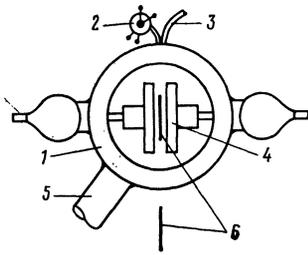
FIG. 1. Construction of the experimental container and the positions of the light beams for measuring the absorption, view along the optic axis of the system: 1) case, 2) thermometer, 3) filling capillary, 4) electrodes, 5) cold finger, 6) position of the beams.

cimers by such a source varies appreciably (see the next section for more detail on this); this makes it possible to control effectively the source. In practice the rapidity of such a means of control was not high and for the present apparatus amounted to about 0.3 s.

In our experiments we used a pumped ^{3}He cryostat and windows which were transparent in the visible and near-IR region.

The helium crystals were grown in a container shown in Fig. 1. The ferrochromium case had internal diameter 18 mm. Windows of optically polished chemical glass, 16 mm in diameter, were stuck into fitting places on its ends with Stycast-1266 epoxy cement. The container was 17 mm long from glass to glass. A 4 mm diameter copper cold finger was soldered through the container wall; its asymmetrical positioning was associated with convenience in passing the light beam through the apparatus for measuring the absorption. A carbon resistance thermometer was placed on a copper wire also soldered through the wall. The mountings of the two electrodes were fixed to the body of the container through ferrochromium–chemical glass seals. The 10 mm diameter electrode plates, positioned vertically at a distance of 1.4 mm from one another, were covered on the inner side with β-active titanium tritide. Similar sources were used earlier in measurements of the mobility of charges in helium.[11,12] The range of the β-electrons emitted by the tritium in condensed helium is several microns; in this way a thin layer of partially ionized and excited helium is formed near the surface of the electrodes. The intensities of both sources were identical, and the total saturation current was 2.4×10^{-8}A. The heat dissipation due to the slowing down of energetic β-particles in the helium was less than 10^{-6} W for the sources used, and does not lead to any noticeable heating of the container. We note that 10^5 times more power was dissipated in experiments[1,2] with irradiation by an electron beam in liquid helium; strong spectra could be produced in this way. In our experiments, as has already been reported,[10] it was not possible to observe any absorption in the liquid, since the intensity of the excitation source used was considerably less than in those other experiments.[1,2]

We were able to apply a voltage from 0 to 30 kV of both polarities between the electrodes and record the electric current passing through the specimen. The electrical power, UI, dissipated in the crystal did not exceed 3×10^{-4} W in all experiments. The presence of two β-sources made it possible to transfer charges of both signs from the pre-electrode layers to the bulk of the crystal and, by measuring the absorp-

tion spectrum, to establish the products of their recombination. Sufficient spatial resolution was achieved in the optical measurements, owing to the large width of the interelectrode gap, so that the absorption could be measured separately in the pre-electrode layers and in the middle of the gap.

At $P \approx 25$ atm the crystals were grown from superfluid liquid at constant pressure and temperature, at $P = 31.2$ and 40.5 atm from normal liquid at constant pressure in a temperature gradient. The container filled with crystal in ~ 1 h, and optical measurements were only carried out on those specimens in which there were no visible bulk and surface defects.

In the first experiments we measured the radiation spectrum from our apparatus filled with liquid and solid helium. An MDR-2 monochromator was used for this and a cooled FÉU-79 photomultiplier, working in the photon counting regime. When measuring the radiation, light was collected from the whole interelectrode space, i.e., there was no spatial resolution; current was not passed through the helium. The results obtained at $T \approx 1.2$ K and for various pressures are shown in Fig. 2. As can be seen from Fig. 2, raising the pressure leads to a sharp reduction in the intensity of the lines. This effect was already observed before[1,2] and was associated by the authors with an amplification of the nonradiative de-excitation as the density of the medium is increased. As can be seen, it was only possible to record the radiation from the solid phase at the limit of the sensitivity of the apparatus. In such a situation an appreciably more sensitive means of recording the excited states is the measurement of the absorption lines corresponding to transitions from the metastable state $a^3\Sigma_u^+$ at higher energy.

For measuring the absorption the light from a ribbon-filament incandescent lamp, which passed through the MDR-2 monochromator was split by a vertical bilens into
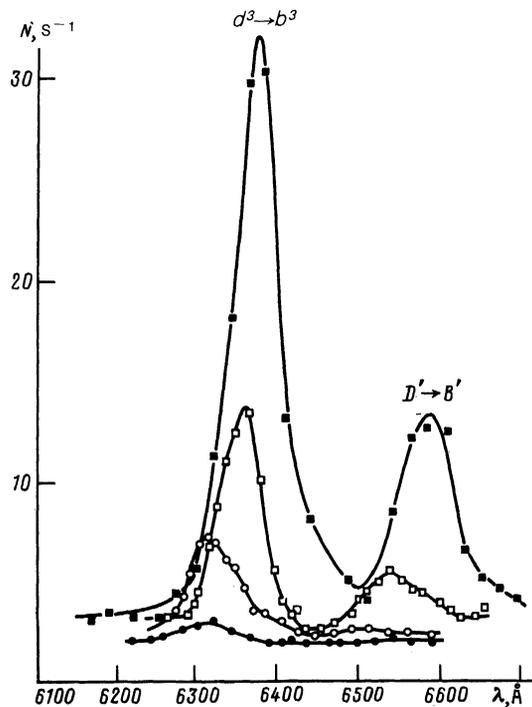

FIG. 2. Radiation spectrum from liquid and sold helium at different pressures: ■) 0.5 atm; □) 12 atm; ○) 24.5 atm; ●) 25 atm, crystal. Collecting time in the liquid was 100 s, in the crystal 500 s; N is the number of photons per second.

two beams, which passed through the apparatus as shown in Fig. 1 and then came together at the FD-24 photodiode. The beams were focused in the middle of the cross-section of the experimental container and in this place their dimensions were usually ≈ 0.3 mm in the horizontal direction and ≈ 7 mm in the vertical. The beams were chopped in turn at a frequency of 300 Hz, and the signal from the photodiode was detected at this frequency and was recorded as a function of wavelength or time. The use of the indicated source of illumination and photodetector produced the most efficient recording of the absorption in the range 7000–10 000 Å. The accuracy of the measurements was limited mainly by vibrations of the cryostat. The minimum absorption which could in that case be recorded, when the beam passing through the crystal did not touch the electrodes, was $(2-3) \times 10^{-5}$.

An example of an experimental trace of an absorption spectrum in the wavelength region 8500–9300 Å is given in Fig. 3. The magnitude of the absorption, as already reported,[10] depended appreciably on the electric current flowing through the specimen. By switching on and off a high voltage, we recorded on a two-pen plotter the change in current and the absorption as a function of time. A typical experimental trace of this type was shown in Fig. 3 of the earlier publication.[10] In this way we could measure the magnitude of the absorption in two ways. In the first case the absorption was recorded as a function of wavelength for fixed current and other parameters (Fig. 3). The second method consisted of measuring the difference in the stationary absorption levels, established after switching the voltage on and off for fixed wavelength. The second method turned out in practice to be somewhat more accurate; it was in this way that the absorption spectrum in a crystal, shown below, and most of the points in the dependence of absorption on current, were obtained. The wavelength was then fixed for measuring the current dependences at the maximum of the absorption line, while for measuring the spectrum the current was turned on and off, which achieved the maximum absorption in the bulk
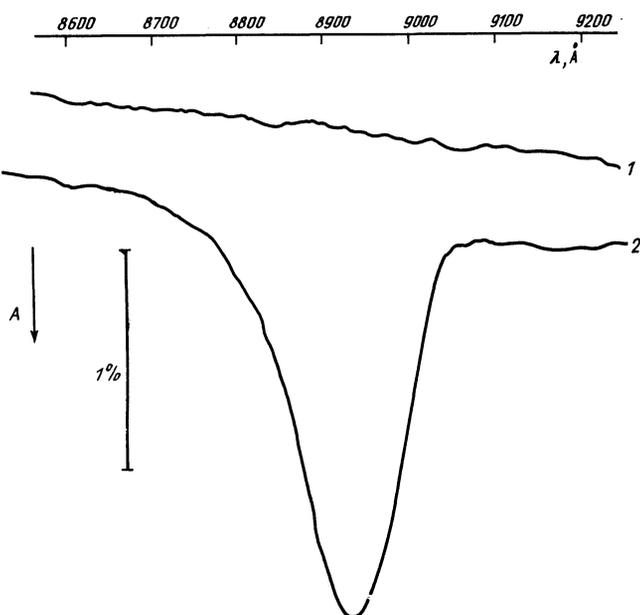
of the crystal (for measuring the spectrum the beam was located in the middle of the interelectrode gap, the corresponding current dependence of the magnitude of the absorption is discussed in the following section).

It should be noted that the electrometer which measured the magnitude of the electrical current had an intrinsic time constant $\tau = 0.87$ s. Several recorded curves of the establishment of the current $I(t)$ on a large time scale and the true time dependences of the current established by numerical analysis using the relation

$$I_{\text{tr}}(t) = I(t) + \tau \dot{I}(t)$$

are shown in Fig. 4. Such an analysis permits a more distinct separation from one another of the capacitance current pulse (the first maximum on the curves) and the moment of the transition from the rise part to the constant value corresponding to the time of flight of charges across the interelectrode space. The drift velocity of charges calculated from the curves of the establishment of the current, $10^{-2}-10^{-1}$ cm/s, agrees within an order of magnitude with the results of extrapolating into our region the experimental parameters for the mobility of both positive and negative charges, measured earlier in somewhat lower fields and at lower temperatures.[12] The maximum which appeared on the curves of Fig. 4 (Ref. 13) was interpreted as the arrival at the collector of a higher-density leading edge of charges. As can be seen, in
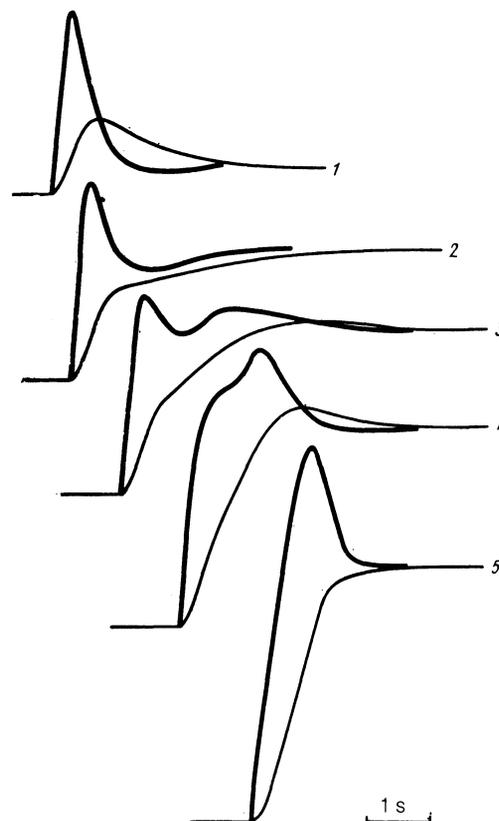

FIG. 4. Experimental curves of the establishment of current for $P = 25$ atm, $T \approx 0.9$ K and different pressures (narrow lines) and the I_{tr} (t) dependences established by numerical analysis (thick lines): 1) $U = 13.8$ kV, $I = 0.10 \times 10^{-8}$ A; 2) $U = 16.5$ kV, $I = 0.45 \times 10^{-8}$ A; 3) $U = 17.3$ kV, $I = 0.54 \times 10^{-8}$ A; 4) $U = 18.5$ kV, $I = 0.65 \times 10^{-8}$ A; 5) $U = 22.0$ kV, $I = 0.85 \times 10^{-8}$ A.

FIG. 3. Experimental trace of absorption in the liquid (1) and in the crystal (2) for measurements near an electrode.

our case for currents $I \gtrsim 0.6 \times 10^{-8}$ A the additional charge density in the leading edges have the same order of magnitude as the density in the stationary flow.

In the majority of cases, after the high voltage is switched off, apart from a pulse of capacitance current, a pulse of current of the opposite sign was observed, evidently associated with a redistribution of charge in the interelectrode space. However, the current dropped to practically zero after a time $\gtrsim 1$ s. The charges remaining after this in the bulk of the crystal recombined, according to estimates, within several seconds or faster (this depends on temperature and on the initial current).

3. RESULTS OF THE MEASUREMENTS

An absorption spectrum in a crystal at $P \approx 25$ atm is shown in Fig. 5. The distance between the points ≈ 30 Å corresponds approximately with the spectral width of the beam. No new details in the spectrum appeared on reducing the spectral width of the beam (by narrowing the entrance and exit slits of the monochromator). Comparison with the absorption spectrum reported earlier shows that in the crystal, unlike the liquid, at a sufficient concentration there are only molecules in the zero vibrational state $a^3\Sigma_u^+$. Evidently, under conditions of a small lifetime, the excited states which are observed in the liquid are not in thermal equilibrium with the surrounding helium.

The distance between the lines (0–0) and (1–0) of the transition $c^3\Sigma_g^+ \leftarrow a^3\Sigma_u^+$ agree within an experimental accuracy of $\approx 2\%$ with the vacuum value 1478 cm^{-1} (Ref. 14). The position and shape of the lines did not depend on temperature; the shape and width of the line (0–0) of the transition $c^3 \leftarrow a^3$ were also close to that observed in liquid helium.[1] Raising the pressure to 31.2 and 40.5 atm leads to a shift of the lines in the direction of shorter wavelengths, while their shape and width remain unchanged. The position of the maximum of the main line is shown in Fig. 6 a as a function of crystal density. Results obtained in the liquid[1,2] are also shown there. It can be seen that the results of measurements in liquid and solid helium lie on the same straight line.

The accuracy in determining the position of the lines $D\,^1\Sigma_u^+ \to B\,^1\Pi_g$ and $d\,^3\Sigma_u^+ \to b\,^3\Pi_g$ in Fig. 2 is not high, especially for radiation from the solid phase; within the limits of this accuracy the earlier results[1,2] and the results of our experiments for the liquid and crystal can also be described by a single linear dependence (see Fig. 6 b).

The magnitude of the absorption in a crystal and its dependence on the electric current were mainly determined by whether the excited pre-electrode helium layer intercepted the light beam traversing the specimen or the beam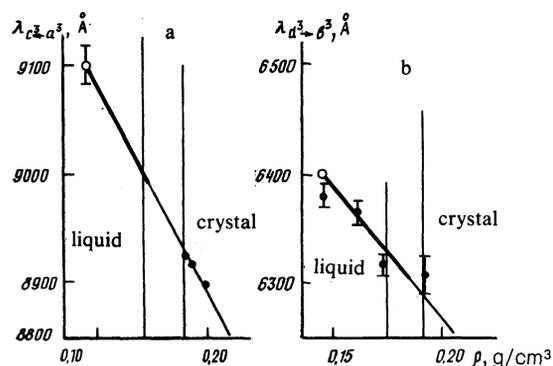 traversed the middle of the interelectrode space. The dependences of the magnitude of the absorption on current for one and the same means for the beam to traverse the crystal, as reported earlier,[10] were similar over the whole range of temperatures studied (from ~ 0.8 K to T_m) and pressure.

Typical curves of the absorption in the middle of the interelectrode gap are shown in Fig. 7. If the beam was positioned symmetrically with respect to the electrodes the absorption was independent of the polarity of the high voltage. If, however, the beam was shifted from the center of the gap towards one of the electrodes (generally approximately by the beam width), but still did not fall on the pre-electrode layer then, as shown in Fig. 8, the dependences of the absorption on current, measured for different polarities of the applied voltage, were noticeably different. Unfortunately, the existing spatial resolution did not permit a detailed measurement of the concentration distribution of excimers in the interelectrode space; we could only differentiate three beam positions qualitatively: in the middle of the interelectrode space; asymmetrical of the type described above, and near an electrode when it caught an excited layer.

The following means of passing the light beam was convenient for a quantitative analysis in the case of measurements near an electrode. The beam fell on the surface of an electrode at a small glancing angle and was reflected from it; the angle of incidence was chosen so that the beam width ≈ 0.3 mm was somewhat smaller than the transverse dimensions of the electrode seen at that angle (i.e., the angle of incidence was ≈ 0.03 rad). All the light then traversed a path of known length (to the same accuracy with which the thickness of the layer and the angle of incidence are known) in the excited pre-electrode layer, and the concentration of excimer molecules in the layer could be estimated from the

FIG. 6. The density dependence of the position of the maximum of lines: a) (0–0)$c^3\Sigma_g^+ \leftarrow a^3\Sigma_u^+$ absorption and b) $d\,^3\Sigma_u^+ \leftarrow b\,^3\Pi_g$ radiation. The values for the liquid, shown by open circles and thick lines, are taken from Ref. 1 and 2.
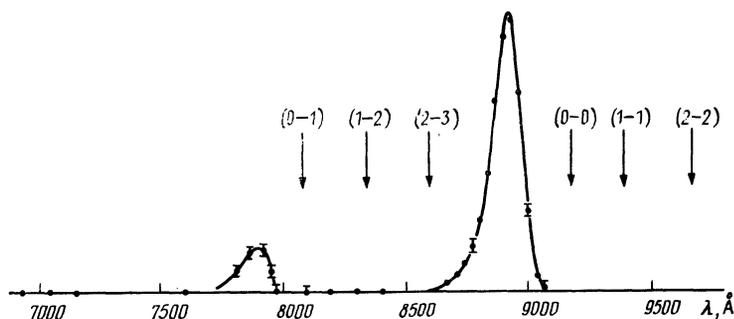

FIG. 5. Absorption spectrum of a crystal at $P = 25$ atm. The positions of the lines of the vibrational structure of the $c^3\Sigma_g^+ \leftarrow a^3\Sigma_u^+$ transition in vacuo[14] are shown by the arrows.
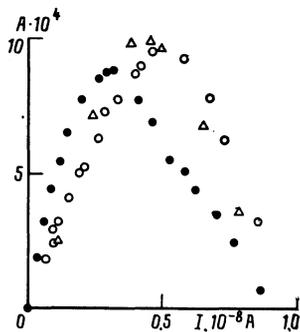
FIG. 7. The current dependence of the absorption in the middle of the interelectrode gap: ●) $P = 25$ atm, $T = 1.4$ K; ○) $P = 25$ atm. $T = 1.1$ K; △) $P = 40.5$ atm, $T = 1.3$ K.
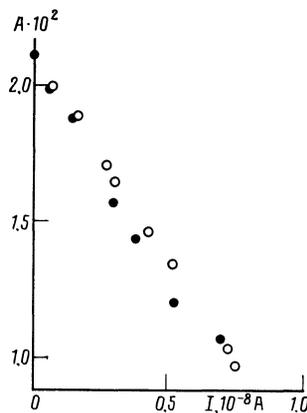

FIG. 9. Current dependence of the absorption near an electrode; $P = 25$ atm, $T = 1.07$ K. The dark circles correspond to negative voltage applied to the electrode illuminated by the beam, the open circles to positive voltage.

magnitude of the absorption. A typical current dependence of the absorption near an electrode in shown in Fig. 9. The absorption $A_0 \approx 2 \times 10^{-2}$ corresponds to an excimer concentration $m \sim 10^{15}$ cm^{-3}. We should remember that the concentration in the bulk of a crystal at the absorption maximum was estimated[10] at $m \sim 10^{12}$ cm^{-3}.

The systematic difference between the locations of the points for different polarities in Fig. 9 is most likely due to the fact that in measurements near the electrode one records also molecules outside the near-electrode layers, so that the interelectrode gap is asymmetrically illuminated. Note that in Fig. 9 the difference between the points for different polarities does not exceed the absorption at the middle of the interelectrode gap.

The increase and decrease of the absorption when the voltage was reversed were exponential within experimental accuracy; the corresponding time constants are shown in Fig. 10. Exceptions were situations when it was impossible to separate distinctly in time the buildups of the current and of the absorption signal. These include, first, the case of small currents ($\sim 1 \cdot 10^{-9}$) at sufficiently low temperatures ($\lesssim 1 \cdot 1$ K), when the current buildup time becomes too long (> 5 s). The second case was that of large currents ($\gtrsim 8 \cdot 10^{-9}$ A), when the current-buildup curve showed a distinct maximum due to the passage of the dense charge front; the absorption-signal buildup can in this case become even nonmonotonic.

4. DISCUSSION OF THE RESULTS

Some conclusions about the mechanisms for the formation and decay of He_2^* molecules in solid helium can be drawn from the experimental results presented. Mutual recombination, which determines the lifetime of excimer molecules in liquid helium, is not manifest in the crystal. On the contrary, the intrinsic lifetime of excimers is observed in solid helium $\tau_0 = 15 \pm 1$ s which is independent of temperature, of pressure and of molecular concentration (Figs. 10 b and c).

The reduction in the excimer lifetime when an electric current flows through the crystal (Figs. 10 a and b) is evidence of the existence of an additional mechanism for the decay of excitations, associated with the motion of electric charges through the helium. We assume that the interaction of molecules with charges is described by a capture cross section σ (Ref. 15). When a current I flows, the lifetime τ_I of excimers is determined by the relation

$$\frac{1}{\tau_I} = \frac{1}{\tau_0} + \frac{j\sigma}{e},$$

where e is the electron charge and $j = I/S$ is the current density. As can be seen from Figs. 10 a and b, the observed reduction in the time τ_I corresponds to a cross section $\sigma = (4-6) \times 10^{-13}$ cm^2.

The formation of excited molecules evidently takes place mainly as a result of recombination of charges of different signs. This suggestion explains in a natural way the reduction in the number of excimer molecules in the pre-electrode layers with increasing current: those charges which
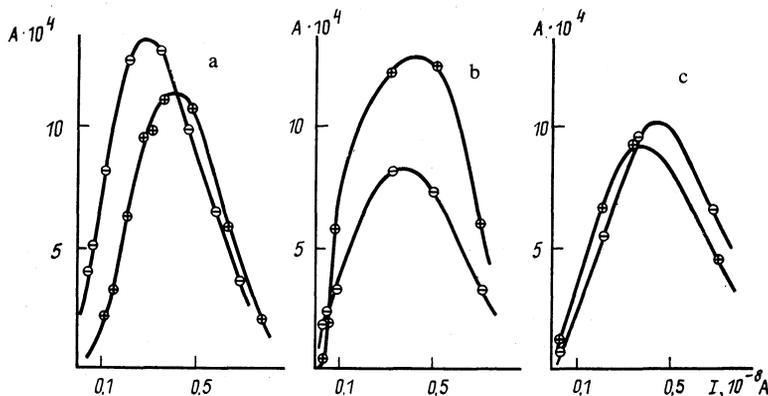

FIG. 8. Electric current dependence of the absorption for the case of asymmetrical positioning of the beam at $P = 25$ atm and different temperatures: a) $T = 1.35$ K, b) $T = 1.05$ K, c) $T \approx 0.9$ K. The symbols ⊕ and ⊖ correspond to the polarity of the voltage applied to the electrode nearer to the region of the crystal studied.
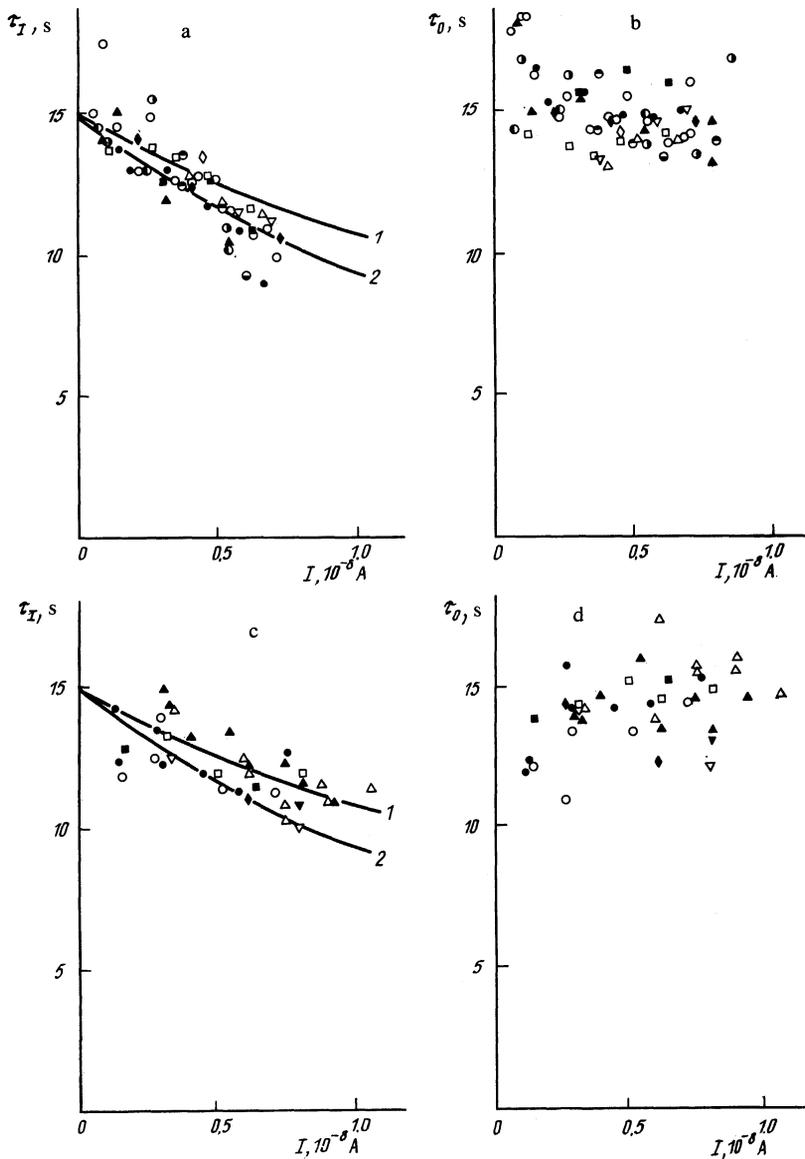
FIG. 10. The dependence of the time constants of the intermediate processes in the absorption on the value of the current I established after the voltage has been switched on (τ_I) and after it has been switched off (τ_0); a and b—measurements in the middle of the interelectrode gap ($P = 25$ atm: O) 1.23 K, ▲) 1.23 K, ◑) 1.1 K, ●) 1.1 K, ◐) 1.23 K, ◒) 1.1 K, ◓) 0.95 K, ▽) 1.27 K, △) 1.32 K; $P = 31.2$ atm: □) 1.43 K, ■) 1.6 K; $P = 40.5$ atm; ◆) 1.35 K, ◇) 1.3 K); c and d— measurements near an electrode ($P = 25$ atm; ●) 1.33 K, O) 1.07 K, ■) 1.33 K; $P = 31.2$ atm; ▲) 1.16 K, △) 1.61 K, ▼) 1.78 K, ▽)1.29 K, ◆) 0.87 K; $P = 40.5$ atm: □) 1.42 K). The calculated curves shown in Figs. a and c describe the shortening of the lifetime of excimers due to their capture by charges. The capture cross section is $\sigma = 4 \times 10^{-13}$ cm², for curve 1 and $\sigma = 6 \times 10^{-13}$ cm² for curve 2.

make a contribution to the current do not take part in the recombination. The passage of an electric current leads not only to a reduction in the number of excimers in the cell, but also to their redistribution over the crystal volume. From the magnitudes of the absorption near an electrode and in the bulk (Figs. 7–9) it is easy to estimate, however, that the fraction of molecules which are formed within the pre-electrode layers is relatively small. For this reason measurements near an electrode give, in fact, the current dependence of the total number of He_2^* molecules in the cell; the difference may be about the same as the difference in the absorption for voltages of different polarities (see Fig. 9).

The current dependence of the absorption near an electrode for different temperatures and pressures normalized to the value of the absorption for zero current A_0 is shown in Fig. 11. Since the magnitude of the absorption depends on the angle of incidence of the light beam on the electrode, the

value of A_0 varies from experiment to experiment within the limits ≈ 1.7–2.1%. For the case of constant lifetime of the excimers, their number in the cell for a current I is determined by the fraction of the charges which is formed under the action of the β-radiation and which recombine near the place where they are formed and therefore do not contribute to the current, i.e.

$$\frac{A(I)}{A_0} = 1 - \frac{I}{I_{sat}}.$$

This dependence is shown in Fig. 11 by the straight line. If, however, $\tau_I (I) < \tau_0$ then as τ_I/τ_0 decreases the equilibrium number of excimers in the crystal also decreases (the dashed curves in Fig. 11). As can be seen, the capture cross section $\sigma \approx (4$–$6) \times 10^{-13}$ cm² describes the experimental results
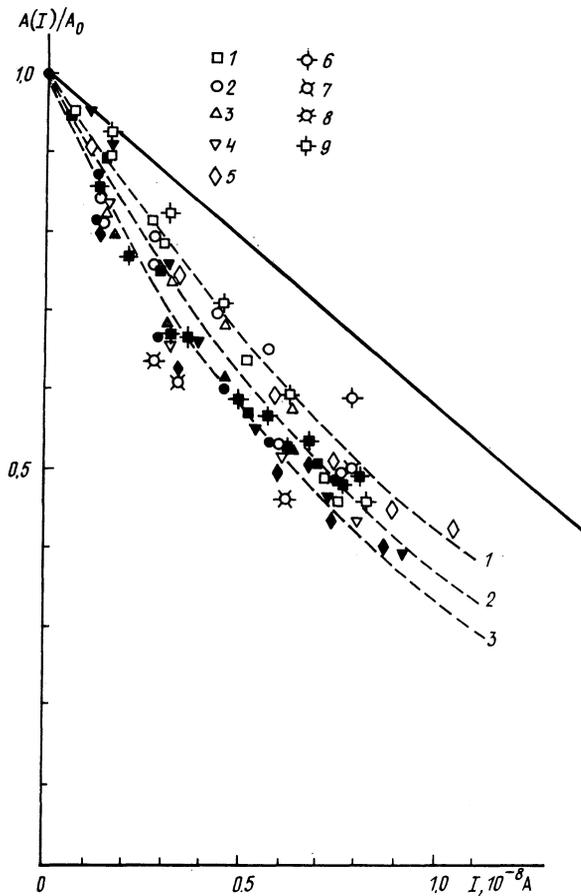
FIG. 11. Normalized absorption near an electrode for different pressures and temperatures. The open symbols correspond to positive voltage applied to the given electrode, the dark symbols to negative; $P = 25$ atm: 1) 1.07 K, 2) 1.33 K, 3) 1.33 K; $P = 31.2$ atm: 4) 1.16 K, 5) 1.61 K, 6) 1.78 K, 7) 1.29 K, 8) 0.86 K; $P = 40.5$ atm: 9) 1.42 K. The solid straight line represents the $A(I)/A_0 = 1 - I/I_{sat}$ relation. The dashed curves are the result of calculations which take account of the capture of molecules by charges and for curve 1) $\sigma = 3 \times 10^{-13}$ cm^2, 2) $\sigma = 4.5 \times 10^{-13}$ cm^2, 3) $\sigma = 6 \times 10^{-13}$ cm^2.

satisfactorily. The value of the corresponding characteristic interaction radius $R = (\sigma/\pi)^{1/2} \approx 35\text{--}45$ Å agrees well with that calculated by Bashkin.[15]

The number of excimer molecules in the pre-electrode ionized layer, $M \approx 3 \times 10^{11}$, can be estimated from the value of the absorption near an electrode in the absence of a current $A_0 \approx 2 \times 10^{-2}$, and the thickness of the layer, which is only known to an order of magnitude, is not important for this. In the stationary state the decay of excimers at a rate $\dot{M} = -M/\tau_0$ should be compensated by their formation upon recombination of charges, $\dot{M} = \gamma I_{int}/2e$. Therefore the probability γ, of the formation of a triplet state on the recombination of charges turns out to be about 0.5. The error in determining this quantity is associated mainly with the fact that the oscillator strength for the transition $c^3 \leftarrow a^3$ is not known accurately; in calculating the absorption cross-section for light by an He$_2^*$($a^3\Sigma_u^+$) molecule, we assumed that the oscillator strength was equal to 0.3 (Ref. 10).

The mechanism proposed for the formation of excimers as a result of charge recombination enables the current dependence of the value of the absorption outside the ionized layers to be described. For this we note that the concentration of excimers in the stationary regime should be equal to

$m = \gamma\tau_f\dot{n}$, where $\dot{n}$ is the rate of charge recombination and the problem comes down to seeking the value of $\dot{n}$ as a function of current. The electric field E, the current density j, and the charge concentrations n_+ and n_- are related by the equations:

$$E_x' = 4\pi e(n_+ - n_-),$$

$$(\mu n_+ E)_x' = -\alpha n_+ n_-,$$

$$j = e\mu E(n_+ + n_-),$$

where x is the coordinate in a direction perpendicular to the plane of the electrodes. The mobilities of positive and negative charges were assumed to be equal and dependent on the strength of the electric field $\mu = \mu(E)$. In the second equation which describes the charge recombination, Langevin's relation $\alpha = 8\pi\mu e$ was used for the recombination coefficient.

It is customary[16] to use as boundary conditions for equations of this type the assumption $E = 0$ at the boundaries of the ionized layers. This condition, however, can only be justified in the limiting case of very small currents $j \ll j_{sat}$. In the current range of most interest to us this condition is clearly not satisfied; this follows, for example, from an analysis of the current-voltage characteristics. As additional conditions necessary for a unique solution, we made use of the current-voltage characteristics $j(U)$ measured in each experiment and, of course, under symmetry conditions corresponding to the symmetry of the measuring cell.

In our calculations of the mobility of the charges we assumed a power-law function for the electric field $\mu = \mu_k E^k$ and constructed the $\dot{n}(I)$ relation for different k and μ_k. Calculations showed that the current dependence of the rate of recombination was close to that shown in Fig. 7, i.e., the maximum which occurs for $I = (0.3\text{--}0.5) \times 10^{-8}$ A and is practically equal to zero for $I \gtrsim 10^{-8}$ A arises if the mobility grows sufficiently rapidly with increasing field $\mu \propto E^n$, where $n = 2\text{--}4$. The power depends on the temperature: the lower the temperature the higher the power necessary to obtain curves close to the experimental curves. A weaker field dependence of mobility shifts the calculated $\dot{n}(I)$ curves in the direction of larger currents. The calculated recombination rate for a given current also depended strongly on the absolute value of the mobility (i.e., of the coefficient μ_k) used in the calculations. By using values of the mobility corresponding to the times of flight of charges across the interelectrode space, by comparing the calculated recombination rate and the measured concentration of excimers in the middle of the interelectrode gap, the efficiency of excimer formation γ can be estimated. The efficiency $\gamma \approx 0.5$ is in full agreement with the estimate given above.

We should note that the results of the calculations, carried out within the framework of the simplified model discussed, only describe the motion and recombination of charges in the experimental cell in its main features. In particular, the mobility of charges of different signs are in general unequal in magnitude, and this must lead to an asymmetry (relative to the middle of the interelectrode space) in the distribution of recombination rate. Such an asymmetry was, in fact, observed in our experiments (see Fig. 8). The intersection of curves in Fig. 8 corresponding to different polarities indicates that at the given temperatures and applied voltage the mobilities of positive and negative charges coincide.

We note that, judging from Fig. 8 c, there is a temperature region $T < 1$ K (and not only for $P = 25$ atm, but also for $P = 31.2$ and 40.5 atm) within which the mobility of unlike charges is close over the whole current range of interest to us $I \approx (0.1-1.0) \times 10^{-8}$ A.

In this way the suggestion that the main mechanism for the formation of excimers in the state $a^3\Sigma_u^+$ is charge recombination describes well the total available experimental results.

The capture of excited molecules by charges could, in principle, lead to a certain concentration of molecules arising in the bulk of the crystal even for the case when the excimers are not formed by recombination of charges. The simplest possibility is that molecules formed in some way in the pre-electrode layer are captured by charges, are transported from the electrodes together with them, and as a result of recombination collect in the bulk of the crystal. Such a mechanism would lead to qualitatively the same observed consequences, but in this case there is numerical deviation from the experiment: the capture probability of a molecule by a charge on passing through the pre-electrode layer is on average $\sigma M/2S \approx 10^{-1}$ [$\sigma = (4-6) \times 10^{-13}$ cm^2 is the capture cross section, $M \approx 3 \times 10^{11}$ is the number of excimers in the pre-electrode layer, $S = 0.8$ cm^2 is the electrode area], and thus the probability of formation of a free excimer molecule in the act of charge recombination would be $\gamma \ll 1$. In addition, the probability itself of the existence of such a long-lived bound state of a charge and a metastable molecule is highly doubtful, since processes must occur which lead to the decay of the metastable state, for example, superelastic scattering.[17]

The fact that the mutual recombination of excimer molecules does not occur in a crystal, even in the pre-electrode layers where their concentration is high, enables the following estimate to be made. The difference between the time constants τ_0 in the pre-electrode layers and in the rest of the crystal volume is not more than 1 s, from which $\alpha_m m \lesssim 4 \times 10^{-3}$ s^{-1}, where $m \sim 10^{15}$ cm^{-3} is the concentration of excimers in the pre-electrode layer and α_m is their recombination coefficient in solid helium. The recombination of excimers in the crystal is limited by their low mobility; in that case, as shown for example by Hereford,[18] $\alpha_m = 4\pi D R_m$, where D is the diffusion coefficient of molecules in a crystal and R_m is their characteristic radius of interaction. The most probable mechanism for the decay of excimers as they interact with one another is Penning ionization. This process is characterized by a capture radius (or cross section) which, according to measurements in different gases has, in general, a magnitude of the order of $R_{\text{cap}} \approx 10^{-8}-10^{-7}$ cm (Ref. 19), including the mutual recombination of atomic excited He* (2^3S), $R_{\text{cap}} \approx 10^{-7}$ cm (Refs. 17, 19).

The magnitude and temperature dependence of the recombination coefficient of He$_2^*$($a^3\Sigma_u^+$) excimers in liquid helium is well described[1,18] by a temperature-dependent diffusion coefficient, close in value to the diffusion coefficient of charges in the liquid, and $R_m \approx 2 \times 10^{-7}$ cm. The results

of the spectral measurements, namely the coincidence of the shape and width of the (0–0) line of the $c^3 \leftarrow a^3$ transition in the liquid and in the crystal and the continuous dependence of the position of the line on the helium density (Fig. 6), without any break at the transition to the other phase, indicates, evidently, that the microscopic structure of the excitations studied in the crystal is the same as in the liquid (according to Hansen and Pollock[3] and Wisdom et al.[4] the excited states in condensed helium form "bubbles" around themselves). There is, therefore, no reason to expect the interaction radius in the crystal to be very different from $R_m \sim 10^{-7}$ cm. The estimate given above thus indicates that the diffusion coefficient of molecules in solid helium is $D \lesssim 3 \times 10^{-12}$ cm$^{-2} \cdot$s^{-1}, which is about three orders of magnitude smaller than for charges of both signs and for ^{3}He in a ^{4}He lattice at $T > 1$ K (Ref. 1). In this connection we note once more that the mechanisms for the motion of point defects in helium crystals has up until now been studied quite insufficiently.

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