

therm. One monolayer of  $\text{He}^3$  is  $109.3 \text{ STP cm}^3$  of gas. The fractional monolayer coverages for *both* isotopes throughout this Letter are fractions of this number; monolayer completion coverages are certainly not known to better than 5%.

<sup>8</sup>Most parts for the flux detector were obtained commercially from the SHE Manufacturing Corp., 3422 Tripp Court, La Jolla, Calif.

<sup>9</sup>J. C. Wheatley, O. E. Vilches, and W. R. Abel,

Physics (Long Is. City, N. Y.) 4, 1 (1968).

<sup>10</sup>J. M. Cotignola, F. de la Cruz, M. E. de la Cruz, and R. P. Platzek, *Rev. Sci. Instrum.* 38, 87 (1967).

<sup>11</sup>For a review of the thermodynamic properties of liquid  $\text{He}^3$  see, for example, W. E. Keller, *Helium-3 and Helium-4* (Plenum, New York, 1969), p. 183 and following.

<sup>12</sup>H. M. Guo, D. O. Edwards, R. E. Sarwinski, and J. T. Tough, *Phys. Rev. Lett.* 27, 1259 (1971).

## Dynamics of Atomic and Molecular Metastable States Produced in Electron-Bombarded Superfluid Helium\*

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We have studied the production, destruction, and lifetimes of the  $\text{He}_2(a^3\Sigma_u^+)$  and  $\text{He}(2^3S)$  metastable states of helium that are produced in electron-bombarded superfluid helium. The characteristic lifetime of a metastable molecule in the superfluid is shown to exceed 0.1 sec, while the lifetime of a metastable atom is only about  $15 \mu\text{sec}$ . The production rates for the  $\text{He}_2(a^3\Sigma_u^+)$  and  $\text{He}(2^3S)$  metastable states are about 500 and 450 per incident 160-keV electron, respectively.

The existence of electronic metastable states of superfluid helium was established by Surko and Reif, but they were unable to identify the states involved.<sup>1</sup> The recent electron bombardment of superfluid helium has made it possible to observe the optical emission and absorption spectra of the excited liquid, and these experiments have shown that large concentrations of  $\text{He}_2(a^3\Sigma_u^+)$  and  $\text{He}(2^3S)$  metastable states of helium may be produced in the superfluid.<sup>2-5</sup> However, these experiments and the theory of Hickman and Lane have emphasized the steady-state nature of the excited liquid.<sup>6</sup>

We report the results of an experimental investigation of the production, destruction, and lifetimes of  $\text{He}_2(a^3\Sigma_u^+)$  and  $\text{He}(2^3S)$  metastable states produced in electron-bombarded superfluid helium. In these experiments the liquid is repetitively excited with a pulse of 160-keV electrons. The peak current could be varied between 0.1 and  $4.0 \mu\text{A}$ , and the pulse duration was typically 3 msec with rise and fall times less than 10 nsec. The populations of the metastable states were monitored during and after the electron excitation by observing the absorption of a monochromated light beam focused through the excited liquid. The absorption signals were recorded using conventional signal averaging techniques. A description of the experimental apparatus has been published elsewhere, and the reader is referred

to the work of Hill, Heybey, and Walters for a summary of the optical absorption spectrum of electron-bombarded superfluid helium.<sup>3,5</sup>

The decreasing population of the  $a^3\Sigma_u^+$  state following a pulse of electrons was observed by monitoring the  $2.1\text{-}\mu\text{m}$  ( $b^3\Pi_g - a^3\Sigma_u^+$ ) molecular absorption band.<sup>3</sup> As shown in Fig. 1 and insert A, the reciprocal of the concentration of  $a^3\Sigma_u^+$  molecules,  $1/M$ , increases linearly with time for as long as 0.1 sec after the pulse of electrons, and the loss rate increases as the temperature is reduced from 2.09 to 1.62°K. In addition, the steady-state concentration of metastable molecules,  $M_0$ , increases with the square root of the peak beam current, as shown in insert B. This current dependence was noted previously by Hill, Heybey, and Walters when they suggested the bilinear collision process  $a^3\Sigma_u^+ + a^3\Sigma_u^+$  as the loss mechanism for the metastable molecule.<sup>3</sup> This reaction implies that the concentration of metastable molecules,  $M$ , is described by the equation

$$dM/dt = -\alpha(T)M^2 \text{ or } 1/M = M_0 + \alpha(T)t, \quad (1)$$

where  $\alpha(T)$  is the bilinear reaction coefficient and  $t$  is the time after the end of the beam pulse. During the time the electron beam is on, Eq. (1) can be written as  $dM/dt = KI - \alpha(T)M^2$ , where the steady-state concentration of the  $a^3\Sigma_u^+$  state is given by  $M_0 = [KI/\alpha(T)]^{1/2}$ , with  $K$  equal to the number of metastable molecules produced per

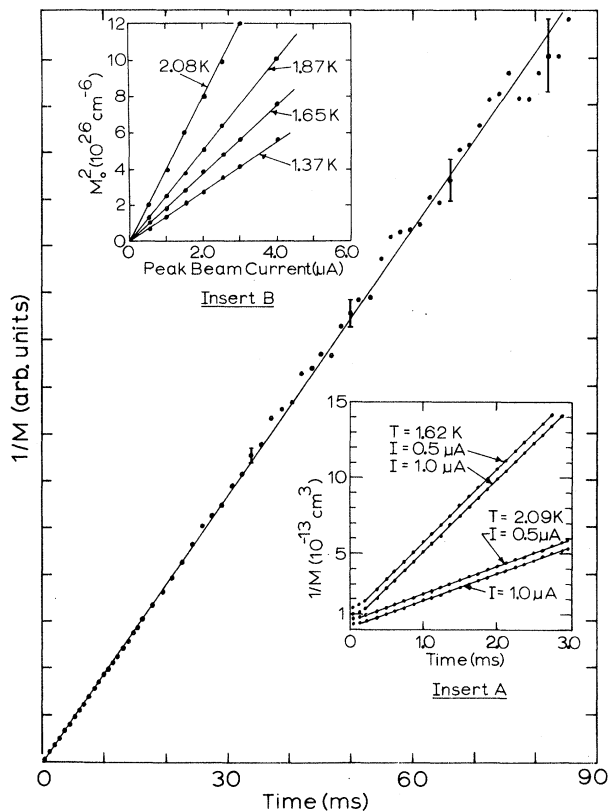


FIG. 1. Inverse concentration of ( $v=0$ )  $\alpha^3\Sigma_u^+$  molecules, for  $T=2.08^\circ\text{K}$ , as a function of time after the beam is off; insert A shows the temperature and beam-current dependences. The initial nonlinear variation is due to the 0.25-msec response time of the PbS detector. Insert B, beam-current dependence of the steady-state concentration at various temperatures.

$\text{cm}^3/\text{sec}$  by the beam current  $I$ . This description agrees well with the results shown in Fig. 1.

The temperature dependence of the bimolecular reaction rate is indicated by the slopes of the lines in Fig. 1 (insert A). These slopes were determined for temperatures between 1.4 and  $2.08^\circ\text{K}$  and the results are shown in Fig. 2 where  $\ln\alpha(T)$  is plotted as a function of  $1/T$ . The error bars represent the range of the measured values for the different beam currents. The variation of  $\alpha(T)$  with temperature suggests the bimolecular reaction is limited by  $\text{He}_2(\alpha^3\Sigma_u^+)$ -roton collisions. In this case the reaction rate would be inversely proportional to the number density of rotors, or proportional to  $e^{\Delta/T}$ , where  $\Delta$  is about  $8.6^\circ\text{K}$ .<sup>7</sup> The deviation from this simple roton model at the lower temperatures is not understood; however, it is not due to possible minute concentrations of  $^3\text{He}$  as shown by the effect observed upon adding 1% molar concentration of  $^3\text{He}$  to the sam-

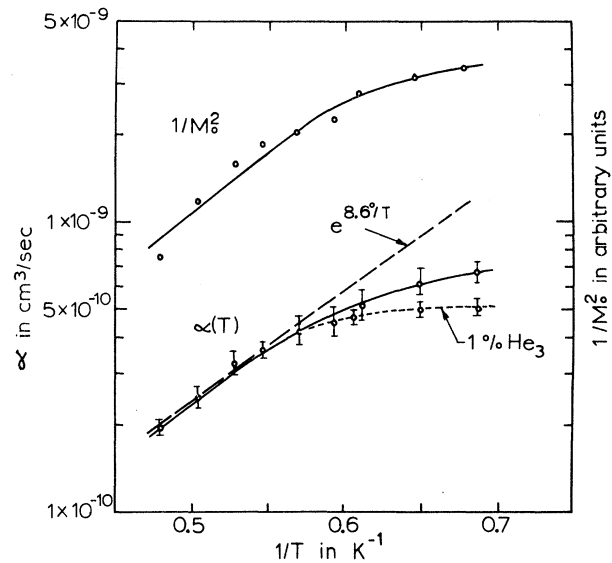


FIG. 2. The bilinear reaction rate  $\alpha(T)$  as a function of inverse temperature. The dotted slope corresponds to the activation energy for rotors. The effect of 1%  $^3\text{He}$  doping of the sample is shown. Also shown is the temperature variation of  $1/M_0^2$  for the case of  $1\text{-}\mu\text{A}$  peak beam current.

ple. As shown also in Fig. 2, the measured values of  $(1/M_0)^2$  exhibit the same variation with temperature as  $\alpha(T)$ ; thus the steady-state production rate for the metastable molecule is essentially independent of the temperature.

The previous discussion is based upon the relative variations of the  $b \rightarrow a$  absorption signal. Absolute values of the reaction coefficient and production rates may be determined by relating the percent optical absorption of the sample to the absolute concentration of metastable molecules. Our measurements, plus an assumed radiative lifetime of  $10^{-7}$  sec for the  $b \rightarrow a$  transition, suggest that the steady-state concentration of metastable molecules is about  $2 \times 10^{13}/\text{cm}^3$  for  $1 \mu\text{A}$  of 160-keV electrons stopped in  $0.04 \text{ cm}^3$  of superfluid helium at  $2.08^\circ\text{K}$ .<sup>8</sup> This estimate has been used to scale the ordinates of Figs. 1 and 2. The absolute values of  $M_0$  and  $\alpha(T)$  may be combined to show that about 500 metastable helium molecules are produced per incident 160-keV electron. The lack of any significant deviation from the bilinear loss of metastable molecules for decay times as long as 0.09 sec establishes a lower limit of about 0.1 sec for the radiative lifetime of the  $\alpha^3\Sigma_u^+$  state in superfluid helium.<sup>9</sup> In addition, the concentration of impurity atoms which could quench the metastable state must

have been less than about 5 parts in  $10^{10}$ . The intensity of the light used for the extended decay measurements is estimated to result in an optical shortened lifetime of about 20 msec for the  $a^3\Sigma_u^+$  state. Thus the induced  $b-a$  transition does not result in a net loss of metastable molecules. This is of particular interest since the optical emission spectrum of the excited liquid indicates that the  $b^3\Pi_g$  state is nonradiatively quenched by the surrounding superfluid.<sup>2</sup>

The lifetime of the metastable  $2^3S$  atom in the superfluid was determined by measuring the transient decay of the  $2^3P-2^3S$  optical absorption at  $10830 \text{ \AA}$ .<sup>3</sup> The response time of the optical detection system was measured to be about  $3 \mu\text{sec}$ . As shown in Fig. 3(a), the number density of  $2^3S$  atoms decays exponentially with a lifetime of  $15 \pm 1 \mu\text{sec}$ . This result is independent of the temperature and beam current to within the indicated uncertainty. In view of the discrepancy between our measurement and a previously reported value, we carefully checked for possible systematic errors in the measurement.<sup>10</sup> We established that the bandpass of the transmitted light

was sufficiently narrow to reject nearby molecular absorption bands; that light emitted by the excited liquid was not detected along with the atomic absorption signal; and that the experimental conditions corresponded to an impurity concentration of no greater than 5 parts in  $10^{10}$ . Assuming a radiative lifetime of  $10^{-7}$  sec for the  $2^3P-2^3S$  transition, we estimate that the concentration of  $2^3S$  atoms is about  $10^{12}/\text{cm}^3$  for  $1 \mu\text{A}$  of electrons.<sup>8</sup> Combining this result with the measured lifetime, we estimate that about 450 metastable  $2^3S$  atoms are produced per incident 160-keV electron.

The rapid and temperature-independent destruction of metastable atoms is not consistent with a mechanism which involves  $2^3S-a^3\Sigma_u^+$  collisions. Since a significant radiative or collision induced  $2^3S-1^1S$  transition rate is not expected, we considered the loss of  $2^3S$  atoms as a source of  $a^3\Sigma_u^+$  molecules.<sup>9</sup> The nearly equal production rates for  $2^3S$  and  $a^3\Sigma_u^+$  states led us to search for a delay in the initial growth of the  $2.1\text{-}\mu\text{m}$  absorption band. Our results, shown in Fig. 3(b), curve 1, indicate a delay of only  $3 \mu\text{sec}$ , and this corresponds to the response time of the detector and not to production by the destruction of  $2^3S$  atoms. However, Hill, Heybey, and Walters report two unidentified absorption bands at about  $1.90$  and  $1.98 \mu\text{m}$ .<sup>3</sup> We observe a substantial delay in the growth of both of these bands; our results for the  $1.90\text{-}\mu\text{m}$  band are shown in Fig. 3(b), curve 2. In addition, we observe the strength of the  $1.90\text{-}\mu\text{m}$  band to vary linearly with beam current and to decay exponentially with a temperature-independent time constant of  $32 \pm 4 \mu\text{sec}$ . As shown in Fig. 3(b), the growth of the  $1.90\text{-}\mu\text{m}$  band may be predicted by assuming a lifetime of  $32 \mu\text{sec}$  for the absorbing state and a production rate equal to the loss rate of  $2^3S$  atoms.

We suggest that the  $1.90\text{-}$  and  $1.98\text{-}\mu\text{m}$  absorption bands are due to the selective population of very high vibrational levels of the  $a^3\Sigma_u^+$  state. This suggestion is based upon the following two arguments: (1) Recent estimates of the  $a^3\Sigma_u^+$  and  $b^3\Pi_g$  molecular potential curves indicate that transitions corresponding to wavelengths between  $1.9$  and  $2.0 \mu\text{m}$  would be consistent with the classical Frank-Condon principle if they occurred between the high levels of the  $a^3\Sigma_u^+$  state and intermediate vibrational levels of the  $b^3\Pi_g$  state.<sup>11</sup> (2) The vacuum-uv emission spectrum of electron excited superfluid helium indicates the rapid and selective population of one high vibrational level of the  $\text{He}_2(A^1\Sigma_u^+)$  state. It has been suggest-

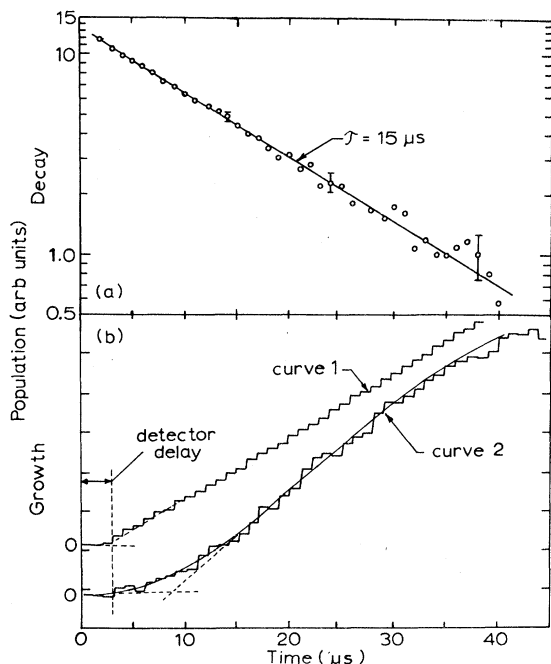


FIG. 3. (a) Logarithmic plot of the  $\text{He}(2^3S)$  concentration for times after the beam is off. (b) Curve 1 shows the growth of the  $(v=0)$   $a^3\Sigma_u^+$  concentration for times after the beam is turned on. The apparent delay is due to the PbSe detector. Curve 2 shows the delayed growth of the  $1.90\text{-}\mu\text{m}$  absorption band. The solid line indicates the predicted delay assuming the absorbing species are formed by the destruction of metastable atoms.

ed that the population of this particular level corresponds to the destruction of the metastable  $\text{He}(2^1\text{S})$  atom.<sup>5</sup> Thus, we suggest that the destruction of metastable  $\text{He}(2^3\text{S})$  atoms in superfluid helium results in the population of high vibrational levels of the corresponding  $a^3\Sigma_u^+$  molecular state. It is clear that further investigation of this phenomenon is necessary.

In summary, (a) the concentration of  $\text{He}_2(a^3\Sigma_u^+)$  molecules in electron-bombarded superfluid helium is limited by collisions between pairs of these molecules. This bimolecular reaction is limited by  $\text{He}_2(a^3\Sigma_u^+)$ -roton collisions and the reaction coefficient is estimated to be about  $2 \times 10^{-10}$   $\text{cm}^3/\text{sec}$  at a temperature of  $2.08^\circ\text{K}$ .<sup>8</sup> (b) The radiative lifetime of the  $\text{He}_2(a^3\Sigma_u^+)$  state in superfluid helium exceeds 0.1 sec. (c) Approximately 500  $\text{He}_2(a^3\Sigma_u^+)$  molecules are produced in the superfluid per incident 160-keV electron. (d) The lifetime of the  $\text{He}(2^3\text{S})$  state in the superfluid is about 15  $\mu\text{sec}$  and approximately 450  $\text{He}(2^3\text{S})$  atoms are produced per incident 160 keV electron. (e) We suggest the destruction of  $\text{He}(2^3\text{S})$  atoms in superfluid helium results in the population of high vibrational levels of the corresponding  $\text{He}_2(a^3\Sigma_u^+)$  molecular state.

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Foundation.

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<sup>6</sup>A. P. Hickman and N. F. Lane, Phys. Rev. Lett. **26**, 1216 (1971).

<sup>7</sup>J. Wilks, *Liquid and Solid Helium* (Clarendon, Oxford, England, 1967), p. 119.

<sup>8</sup>Our values for the concentrations of metastable states agree well with those of Ref. 3, and our results for  $\alpha(T)$  are in good agreement with the single measurement at  $1.7^\circ\text{K}$  of J. C. Hill, O. Heybey, and G. K. Walters, Phys. Rev. Lett. **26**, 1519(E) (1971).

<sup>9</sup>A lower limit of about 0.05 sec for the radiative lifetime of the metastable helium molecule in helium gas at  $300^\circ\text{K}$  has been established previously. See A. V. Phelps, Phys. Rev. **99**, 1307 (1955). The three-body conversion of the  $\text{He}(2^3\text{S})$  atom into a ( $v=0$ )  $\text{He}_2(a^3\Sigma_u^+)$  molecule is also reported in this paper.

<sup>10</sup>Hill *et al.* (Ref. 3) report a lifetime of 1 msec for the  $\text{He}(2^3\text{S})$  state in superfluid helium.

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## Resonant Absorption of Laser Light by Plasma Targets\*

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It is proposed that a resonant mechanism should cause significant absorption of energy from intense laser pulses in plasma targets, and that the energy should be deposited in such a way as to form a very non-Maxwellian high-temperature tail on the electron velocity distribution.

Current interest in the controlled release of nuclear energy from laser-heated pellets of thermonuclear fuel has drawn interest to mechanisms by which laser light might be absorbed in the surface of such pellets. The large pulsed light intensities which are required for this heating and are beginning to be available in absorption experiments put the interaction of the laser radiation with the initially solid target in the approximately collisionless regime. Binary collisions between electrons driven by the wave fields and ions (sometimes called inverse bremsstrahlung)

can cause some absorption and may be quite important in allowing a weak precursor of a pulse to ionize the target and give the surface some thickness as assumed below, but as the intensity increases, the fraction of the light energy absorbed by collisions becomes smaller.

In this Letter we propose a resonant, collisionless absorption mechanism by which an inhomogeneous target plasma can absorb a significant fraction of obliquely incident laser light. We then show from numerical simulations that the energy is deposited in a very non-Maxwellian tail