Experimental Determination of the De-excitation Cross Section for the 6^3P_1 State of Mercury by Thallium*

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The de-excitation or quenching cross section of thallium for the 6^3P_1 state of mercury has been obtained over a range of temperatures of 800 to 900°C. Comparison of the resonance fluorescence from a mercury-thallium vapor mixture with the resonance fluorescence from a pure mercury vapor at the same number density, as determined by line absorption, allows the calculation of the de-excitation cross section. The present data together with Garrett's value of the lifetime of the 6^3P_1 state of mercury and existing data on the vapor pressures of mercury and thallium were used to obtain values of the de-excitation cross section. Values of the cross section range from 108 ± 41 Å^2 at 800°C to 80 ± 19 Å^2 at 900°C. The corresponding quenching-rate coefficients varied from $(5.1\pm1.9)\times10^{-7}$ cm^3/sec at 800°C to $(4.0\pm0.9)\times10^{-7}$ cm^3/sec at 900°C.

SINCE the advent of the continuous gas laser there has been renewed interest in the processes whereby excitation energy can be transferred from one atom to another. Two manifestations of the transfer of excitation energy from one atomic species to another are: (a) a decrease in emission by the initially excited species, sometimes called quenching, and (b) an increase in emission from the species receiving the excitation energy, sometimes called sensitized fluorescence. Although there have been numerous studies of sensitized fluorescence of the mercury-thallium system,[1] no measurements of the de-excitation cross section of thallium for the mercury 6^3P_1 state have been made. Few measurements of the *absolute* values of any excitation-transfer cross sections have been attempted,[2] but Anderson and MacFarland[3] have given *relative* transfer cross sections for the mercury-thallium system. The present study is concerned with the determination of the de-excitation cross section for transfer of energy out of the 6^3P_1 state of mercury as a result of collisions with unexcited thallium atoms.

ANALYSIS

Figure 1 gives a schematic diagram of the pertinent portions of the energy-level scheme for the mercury-thallium system. The mercury 6^1S_0, 6^3P_0, and 6^3P_1 states are represented by the indices 0, 1, and 2, respectively, and the thallium $6^2P_{1/2}$, $6^2P_{3/2}$, $7^2S_{1/2}$, $7^2P_{1/2}$, $7^2P_{3/2}$, $6^2D_{3/2}$, $6^2D_{5/2}$, and $8^2S_{1/2}$ states are represented by the indices 0, 1, 2, 3, 4, 5, 6, and 7, respectively. When irradiated with 2537-Å mercury resonance radiation, mercury atoms in a mixture of mercury and thallium vapor may absorb radiation and be raised to the 6^3P_1 state. These excited atoms may leave the excited state by spontaneous emission of 2537-Å radiation or by a radiationless transition resulting from collisions.

In treating the phenomena involved, it is desirable to adopt the following simple notation for the rates (number per unit volume per unit time) for the processes involved:

(1) $R_{ij}{}^a$ is the rate of an emission or absorption process by which atoms of species a, a for mercury and b for thallium, are transferred from state i to state j.

(2) $R_{ijkl}{}^{ab}$ is the rate of a collision process between atoms of species a and species b, in which the atom of species a is transferred from state i to state j, while the atom of species b is transferred from state k to state l.

(3) $R_{ij}{}^{aw}$ is the rate for a process involving collisions of atoms of species a with the walls in which the initial and final states of the atom are i and j.

Under the experimental conditions encountered in the present work, the absorption rate $R_{02}{}^a$ depends only on the intensity of the incident 2537-Å radiation and the number density $n_0{}^a$ of ground-state mercury atoms. The rate of spontaneous radiative transitions of mercury atoms from state 2 to state 0 is given by,

$$R_{20}{}^a=n_2{}^aA_{20}{}^a=n_2{}^a/\tau_{20}{}^a, \quad (1)$$

where $A_{20}{}^a$ is the probability of spontaneous emission and $\tau_{20}{}^a$ is the radiative lifetime for state 2. The rate at which excited mercury atoms in collision with ground-state thallium atoms transfer the thallium atoms to the lth state and transfer the mercury atoms to the ground state is given by,

$$R_{200l}{}^{ab}=2n_2{}^an_0{}^b\sigma_{200l}{}^{ab}v_{20}{}^{ab}, \quad (2)$$

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[1] J. Franck and G. Cario, Z. Physik **17**, 202 (1923); S. Mrozowski, Acta Phys. Polon. **6**, 58 (1937); R. Swanson and R. MacFarland, Phys. Rev. **98**, 1063 (1955); E. K. Kraulinya, A. E. Lezdin, and Yu. A. Silin, Opt. i Spektroskopiya (USSR) **9**, 154 (1965) [English transl.: Opt. Spectry. **19**, 84 (1965)]. For a review of sensitized fluorescence up to 1934 see Ref. 4.
[2] A. B. Callear, Applied Optics, Suppl. 2, 164 (1965).
[3] R. A. Anderson and R. H. McFarland, Phys. Rev. **119**, 693 (1960); Ph.D. dissertation, Kansas State University, 1959 (unpublished).

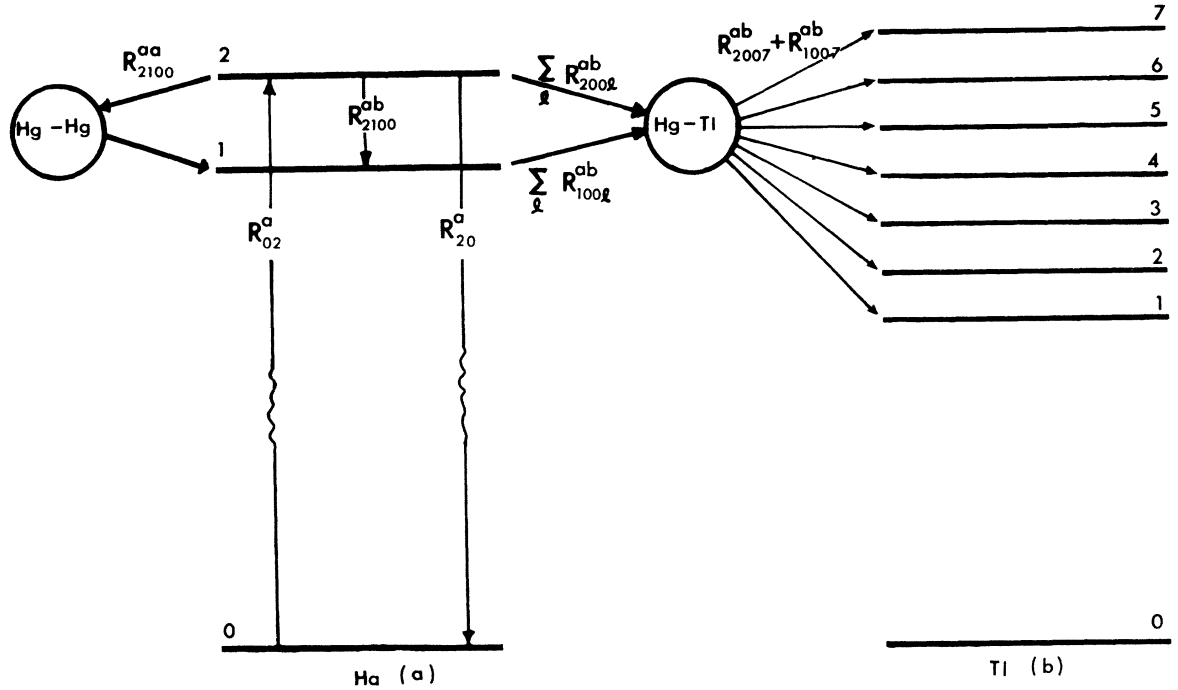

FIG. 1. Schematic energy level diagram indicating processes of interest. Circles represent collision processes.

where $\sigma_{200l}{}^{ab}$ is the cross section[4,5] for this process, and the relative velocity of the collision is

$$v_{20}{}^{ab}=[(2RT/\pi)(1/M_a+1/M_b)]^{1/2}. \quad (3)$$

If the number density of mercury atoms in the ground state $n_0{}^a$, the temperature T, and the intensity of the incident 2537-Å radiation are constant, then the absorption rate $R_{02}{}^a$ with thallium present is equal to the absorption rate ${}^0R_{02}{}^a$ without thallium present, provided the partial pressure of thallium vapor is so low that pressure broadening of spectral lines is negligible and provided the intensity of the incident radiation is so low that $n_1{}^a$ and $n_2{}^a$ are negligible compared with $n_0{}^a$. Under these conditions, which are easily met, we may write,

$${}^0R_{20}{}^a+{}^0R_{2100}{}^{aa}+{}^0R_{20}{}^{aw}=R_{20}{}^a +R_{2100}{}^{aa}+\sum_l R_{200l}{}^{ab}+R_{20}{}^{aw}. \quad (4)$$

With a gas sample in a volume sufficiently far removed from the walls, radiative decay or collision with another atom is far more likely for an excited atom than collision with the wall. Under such conditions the rates ${}^0R_{20}{}^{aw}$ and $R_{20}{}^{aw}$ are negligible compared with the other rates in (4) and can be eliminated from consideration.

According to Winan's[6] partial J selection rule, it is highly probable that $\sigma_{2100}{}^{aa}$ is much smaller than $\sigma_{200l}{}^{ab}$. However, even if $\sigma_{2100}{}^{aa}\cong\sigma_{200l}{}^{ab}$, the rates $R_{2100}{}^{aa}$ and ${}^0R_{2100}{}^{aa}$ will be negligible with respect to the remaining rates, since for the conditions of the experiments reported here $n_0{}^b\gg n_0{}^a$, while ${}^0n_2{}^a$ is the same order of magnitude as $n_2{}^a$ and $v_{20}{}^{aa}\cong v_{20}{}^{ab}$. For these reasons the self-quenching rates of mercury can be eliminated from consideration.

The intensity of resonance-fluorescence radiation is related to the number density of atoms in the excited state through the relationship,

$$I_{20}{}^a=Kn_2{}^a, \quad (5)$$

where K is a quantity depending on the response of the monochromator and detecting system, the geometry, and the transition probability for the emission of resonance radiation. Subject to the conditions discussed above and provided K is kept constant we may make use of the relationships (1), (2), (3), and (5), to write Eq. (4) in the form,

$$\sigma_d=(\tau_{20}{}^a n_0{}^b)^{-1}[(8RT/\pi)(1/M_a+1/M_b)]^{-1/2} \times({}^0I_{20}{}^a/I_{20}{}^a-1), \quad (6)$$

where, $\sigma_d=\sigma_{2100}{}^{ab}+\sum_l \sigma_{200l}{}^{ab}$ is called the de-excitation cross section and is the quantity of interest in the present study. Equation (6) is the Stern-Volmer relationship.

The quantities in Eq. (6) are known or can be readily determined. The number density of ground-state thallium atoms $n_0{}^b$ can be obtained from the temperature of the coolest part of the container and the corre-

[4] A. G. C. Mitchell and M. W. Zemansky, *Resonance Radiation and Excited Atoms* (Cambridge University Press, New York, 1934), p. 155.

[5] J. B. Hasted, *The Physics of Atomic Collisions* (Butterworths Scientific Publication, Washington, 1964), p. 452.

[6] This process violates Winan's partial J sum rule. J. Winans, Rev. Mod. Phys. **16**, 175 (1944).

sponding vapor pressure of thallium. The lifetime of the 6^3P_1 state $\tau_{20}{}^a$ has the value of 1.08×10^{-7} sec as determined by Garrett.[7] The ratio of the intensity of resonance fluorescence radiation from a pure mercury vapor to that of mercury vapor at the same density with added thellium was determined experimentally in the present study.

EXPERIMENTAL ARRANGEMENT

A schematic diagram of the experimental arrangement is shown in Fig. 2. A cubical cell of fused quartz C is located at the center of a furnace MF. The source of 2537-Å radiation is a temperature-controlled "Penray"[8] low-pressure mercury lamp L. The 2537-Å radiation from the lamp is collimated by a quartz lens and is incident on the cell through a window cut in a ceramic mask MA. The fluorescence radiation leaves the cell at right angles to the incident radiation through a window in the ceramic mask and is incident on the entrance slit of a Bausch and Lomb 500-mm grating monochromator M_1, equipped with a 1P28 photomultiplier PM_1. The transmitted 2537-Å radiation leaves the cell through another window in the ceramic mask and is incident on the entrance slit of another Bausch and Lomb 500-mm grating monochromator M_2, also equipped with a 1P28 photomultiplier PM_2. The output of the lamp L is monitored by a third photomultiplier PM_3 after passing through a Baird-Atomic 2537A Type A11 interference filter F which has a 250-Å bandwidth at half-peak transmission.

The sample cell is a 25-mm cube with a side arm reservoir 75 mm in length and was sealed after vacuum distillation of the desired amounts of mercury and thallium into the cell. Prior to the final distillation and sealing, the cell was outgassed at pressures of less than 5×10^{-8} Torr and at temperatures above 1000°C. Each cell employed contained a small, fixed amount of mercury and a copious supply of thallium. At cell temperatures above 500°C the number density of mercury atoms was nearly constant and the number density of thallium atoms was determined by the temperature of the side arm, which was controlled by an auxiliary furnace SAF, in Fig. 2. In normal operation the side arm was maintained at a temperature 50°C below that of the main cell. With this temperature difference between the cell and the side arm, thallium condensation on the cell windows was eliminated.

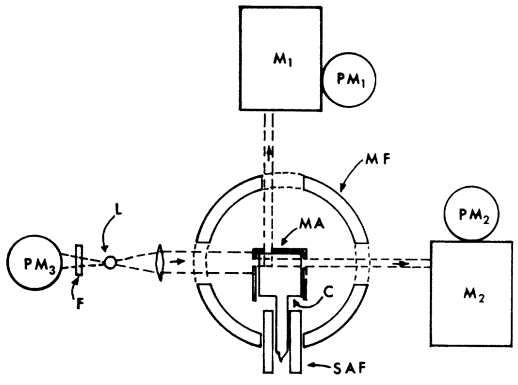

FIG. 2. Experimental arrangement. C—cell; F—2537A filter; L—mercury lamp; M_1,M_2—monochromators; MA—ceramic mask; MF—main furnace; PM_1, PM_2, PM_3—photomultipliers; SAF—side arm furnace.

The ceramic mask determined the geometry of irradiation within the cell and the volume from which the fluorescence radiation was detected. The dimensions of the windows in the mask are 10 mm by 1 mm and permit the vapor in the cell to be irradiated about 0.5 mm from one wall of the cell, as indicated in Fig. 2. The fluorescence radiation leaves the cell from a region about 1 mm from the point at which the incident 2537-Å radiation enters the cell. At temperatures above 700°C, the 5350-Å radiation from thallium was clearly visible from a well-defined region along the path of the incident 2537-Å radiation. The dimensions and locations of the windows in the ceramic mask were chosen in such a manner as to ensure (1) that the 2537-Å fluorescence radiation was primary resonance radiation and (2) that collisions of excited mercury atoms with the walls could safely be neglected.

The spectral slit widths of the monochromators were sufficiently large to pass all the radiation in the Doppler-broadened spectral lines being measured. The outputs of the photomultipliers PM_1 and PM_2 were amplified with an Engis S01-10 dual-channel photomultiplier readout amplifier.

The number density of thallium atoms in the cell was determined from the known vapor pressure of thallium[9] at the temperature of the side arm with appropriate ideal-gas-law correction for the 50°C higher temperature of the cell. The number density of mercury atoms was determined by comparison of the line absorption in the sample cell with line absorption in a secondary cell containing pure mercury vapor at known pressures. The secondary cell was identical with cell C in Fig. 1; however, its side arm contained an excess of pure mercury. The side arm of the secondary cell was surrounded by a quartz jacket through which liquid nitrogen or water at temperatures in the range 0–30°C could be circulated. With the side arm at liquid-nitrogen temperature, the vapor pressure of Hg was negligibly low; with water in the jacket surrounding the side arm, the mercury vapor pressure was controlled by adjusting the temperature of the circulating water. The temperature of the secondary cell itself was maintained at 850°C. All temperatures were determined by the use of Leeds & Northrup Chromel-Alumel thermocouples and the manufacture's calibration curves were em-

[7] Measurements of $\tau_{20}{}^a$ are reviewed in Ref. 4, pp. 123–126.

[8] Model 11SC-1, without handle, Ultra-Violet Products, San Gabriel, California.

[9] K. K. Kelley, U. S. Bureau of Mines Bulletin No. 383, 1 (1935).

ployed. For temperatures below 100°C the thermocouple emf was recorded to ±0.01 mV, which gives an uncertainty in the temperature of ±0.25°C. For temperatures above 100°C the thermocouple emf was recorded to ±0.25 mV, which gives an uncertainty of ±6.25°C for the high-temperature readings. The number densities of mercury atoms in the side arm were computed from the known mercury vapor pressure[9] at the temperature of the side arm; the number density of mercury in the secondary cell was computed from the side arm temperature and the temperature of the secondary cell by the use of the Knudsen effusion relationship.

The results of the measurements of absorption and resonance fluorescence of pure mercury vapor in the secondary cell are shown in Fig. 3. Abscissa for both urves is the number density of mercury atoms in units of 10^{12} atoms/cm^3, while the ordinate of curve A represents percent absorption and the ordinate for curve B gives the ratio of resonance fluorescence to the intensity of the beam transmitted by the evacuated cell. The symbol ${}^0I_t$ represents the intensity of the transmitted beam of 2537-Å radiation with mercury present but with no thallium present. The symbol I_0 represents the transmitted beam intensity with neither vapor present; i.e., with the mercury in the side arm frozen out by liquid nitrogen. The curve marked $(1-{}^0I_t/I_0)$ represents the percentage of radiation which is absorbed by the mercury vapor in the cell, while the curve marked $({}^0I_{20}{}^a/I_0)$ represents the ratio of the resonance fluorescence to the intensity of the beam transmitted by the evacuated cell. The near linearity of $({}^0I_{20}{}^a/I_0)$ as a function of number density of mercury atoms indicates that radiation imprisonment is unimportant.

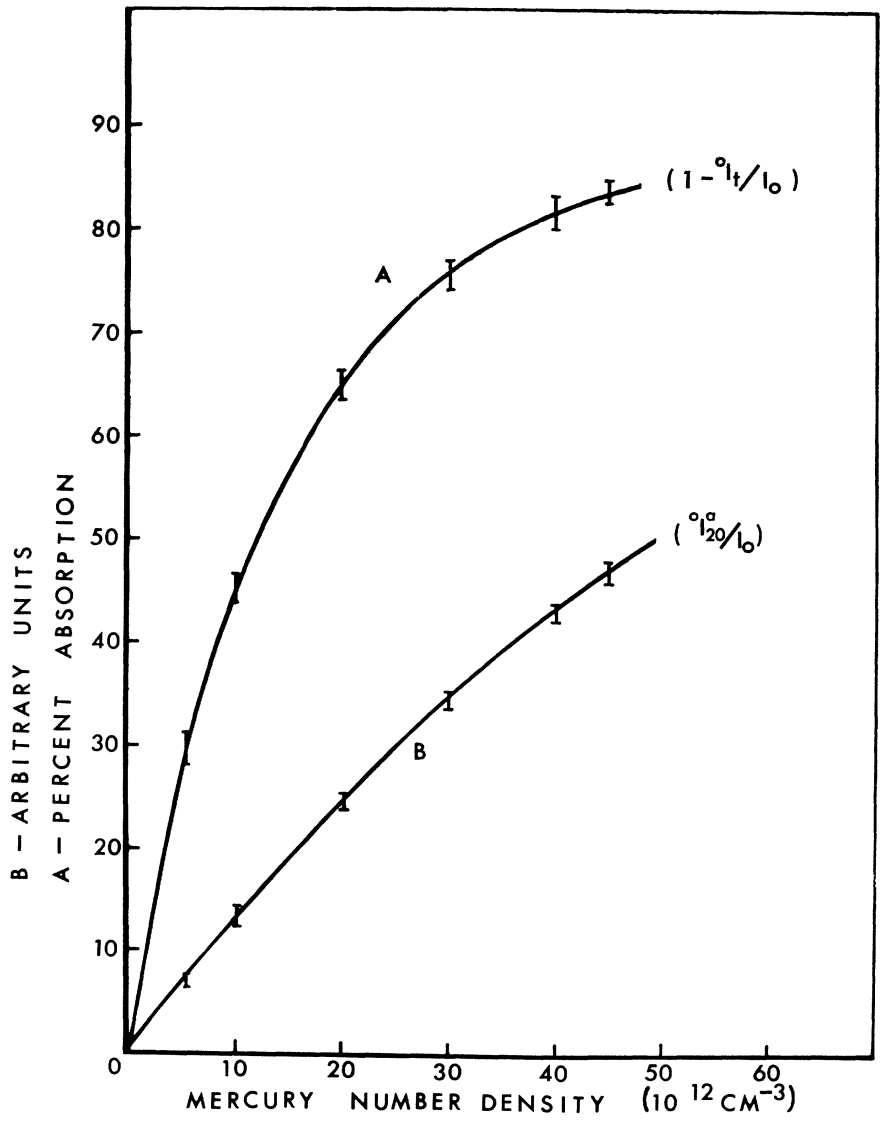

FIG. 3. Resonance fluorescence and line absorption from a cell containing pure mercury vapor. The vertical bars indicate the extreme range of the measurements for three independent measurements.

RESULTS

Typical results of measurements on a cell containing a mercury-thallium vapor are shown in Fig. 4. The ordinate for curve A represents percent absorption and for curves B and C is the ratio of resonance-fluorescence intensity to the intensity of the beam transmitted by the evacuated cell, while the abcissa is the number density of thallium atoms in units of 10^{12} atoms/cm^3 plotted on a logarithmic scale. Curve A, marked $(1-I_t/I_0)\times 100$, and curve B, marked (I_{20^a}/I_0), are the measured results from a cell containing a mercury-thallium vapor, while curve C, marked $({}^0I_{20^a}/I_0)$, gives the relative intensity of resonance fluorescence from a cell containing pure mercury vapor at the same mercury number density. Curve C in Fig. 4 is determined by using the value of $(1-I_t/I_0)\times 100$ given in curve A and finding from Fig. 3 the value of $({}^0I_{20^a}/I_0)$ to be expected in the absence of thallium.

The increasing absorption of mercury with increasing thallium number density or increasing temperature as seen in curve A of Fig. 4 was a common feature of all the measurements. This increase, which was not completely reproducible for different runs on a given cell, depended on the distribution of thallium in the side arm, the greater the thallium surface area the greater the slope of the absorption curve. This increased absorption was interpreted as a true increase in number density of mercury atoms in the interaction region of the sample cell due to the release, as the temperature increased, of mercury atoms adsorbed on the thallium. This phenomenon necessitated the monitoring of mercury number density by line absorption.

Data similar to those shown in Fig. 4 were obtained with five cells, each of which contained a different amount of mercury. The results of the measurements are given in Table I. The first column of Table I gives the cell identifications. The second column gives the cell temperatures, which were measured to ± 6°C. The third column lists the number density of mercury atoms in the cell as determined by absorption measurements of the type presented in Fig. 3. The vapor-pressure data[9] were considered to contribute no error, but the uncertainty in the temperature measurements on which Fig. 3 is based introduced a $\pm 3\%$ uncertainty in the mercury number density. The fourth column of the table gives the number density of thallium atoms in the cell as determined from the side-arm temperature. Because of the greater uncertainty in the measurement of high cell temperatures mentioned above, the uncertainty in the thallium number density is $\pm 12\%$. The fifth and sixth columns tabulate, respectively, the values of (I_{20^a}/I_0) and $({}^0I_{20^a}/I_0)$. The intensity ratio (I_{20^a}/I_0) was determined from chart recordings of the output of the photomultiplier readout amplifiers, while the ratio $({}^0I_{20^a}/I_0)$ was determined as described above. The ratios reported for I_{20^a}/I_0 and ${}^0I_{20^a}/I_0$ in Table I are averages for three sets of readings and have an uncertainty of ± 0.3.

The seventh column lists the values of $({}^0I_{20^a}-I_{20^a})/I_{20^a}$ as computed from the intensity ratios of columns five and six. The eighth column tabulates the expected uncertainty in the values of $({}^0I_{20^a}-I_{20^a})/I_{20^a}$. This expected error was computed by taking the square root of the sum of the squares of the errors of the separate ratios as the error in the difference of the ratios $[({}^0I_{20^a}/I_0)-(I_{20^a}/I_0)]$. The expected error in $[({}^0I_{20^a}/I_0)-(I_{20^a}/I_0)]$ was converted to a percent error and the percent error in (I_{20^a}/I_0) was added to it giving the expected error in $[({}^0I_{20^a}/I_0)-(I_{20^a}/I_0)]/(I_{20^a}/I_0) = ({}^0I_{20^a}-I_{20^a})/I_{20^a}$, which is listed in column 8 of the table.

The ninth column gives the values for the de-excita-

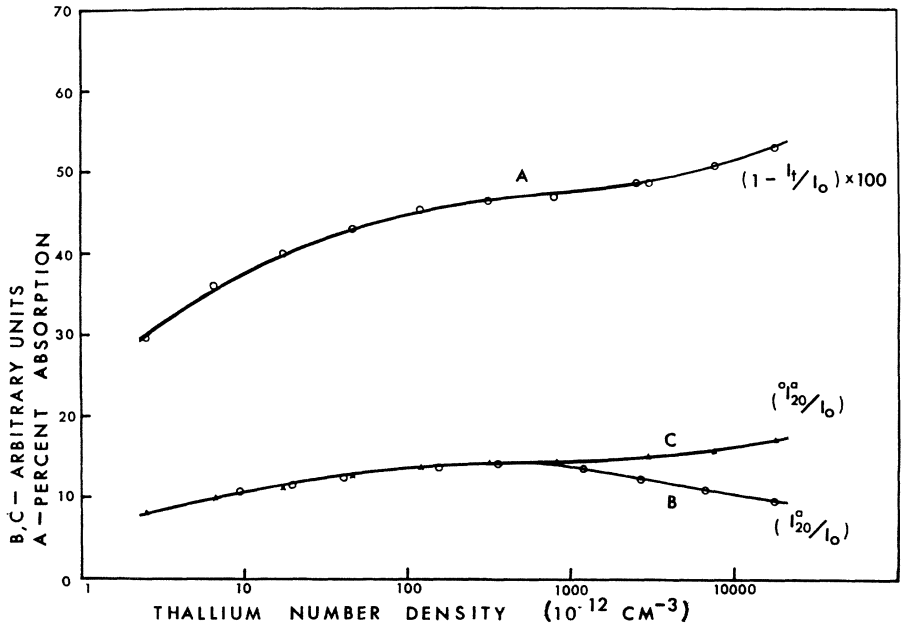

FIG. 4. Resonance fluorescence and line absorption from a cell containing a mercury-thallium vapor mixture. The points are average values for three measurements or calculations and have an uncertainty of ± 0.3 units. The curve marked C is found by the use of Fig. 3 and the value of the percent absorption of the cell from curve A. Curve C is corrected for the difference in cell window transmission by a constant factor determined at a thallium atom number density of about 10^{14} atoms/cm^3.

TABLE I. Experimentally determined values of the de-excitation cross section σ_d of thallium for the 6^3P_1 state of mercury.

Cell	Temperature (°C)	$n_0{}^a$ (10^{12} cm^{-3})	$n_0{}^b$ (10^{12} cm^{-3})	$I_{20}{}^a/I_0$	${}^0I_{20}{}^a/I_0$	$({}^0I_{20}{}^a-I_{20}{}^a)/I_{20}{}^a$	Uncertainty in $({}^0I_{20}{}^a-I_{20}{}^a)/I_{20}{}^a$ (%)	σ_d(Å^2)	Rate coefficient $\langle\sigma_d v\rangle$ (10^{-7} cm^3/sec)
A	800±6	8	2700	10.3±0.3	11.1±0.3	0.078	±53	58.9±38.2	
B	805±6	10	2300	11.8±0.3	13.7±0.3	0.161	±24	141.7±50.9	
C	800±6	11	2300	13.6±0.3	15.5±0.3	0.140	±23	123.1±43.3	
D	805±6	11	2900	12.6±0.3	14.9±0.3	0.182	±19	127.4±33.8	
E	800±6	12	2600	14.4±0.3	16.0±0.3	0.111	±27	86.6±33.7	
								Av. 108±41	5.1±1.9
A	850±6	8.5	5800	9.3±0.3	11.2±0.3	0.258	±20	87.6±28.1	
B	860±6	10	6200	11.0±0.3	13.8±0.3	0.254	±17	80.9±28.1	
C	855±6	12	4800	12.2±0.3	16.1±0.3	0.320	±12	131.2±31.5	
D	855±6	12	7700	11.2±0.3	15.7±0.3	0.402	±12	103.2±24.8	
E	850±6	13	6200	13.1±0.3	16.7±0.3	0.275	±13	87.2±22.0	
								Av. 99±25	4.8±1.2
A	890±6	9.5	14 000	7.6±0.3	13.2±0.3	0.736	±11	108.9±25.2	
B	900±6	10	16 000	9.8±0.3	13.7±0.3	0.388	±13	46.8±11.8	
C	900±6	13	15 000	10.3±0.3	17.2±0.3	0.670	±9	85.9±18.2	
D	900±6	13	16 000	9.8±0.3	17.1±0.3	0.745	±8	89.8±17.8	
E	890±6	14	15 000	11.3±0.3	17.6±0.3	0.557	±9	71.3±14.9	
								Av. 80±19	4.0±0.9

tion cross section σ_d. The uncertainties listed for σ_d include the uncertainty in $({}^0I_{20}{}^a-I_{20}{}^a)/I_{20}{}^a$, the uncertainty in the number density of thallium atoms and a 5% uncertainty in the value of $\tau_{20}{}^a$. The final values of σ_d as determined by these measurements and their uncertainties are: 108±41 Å^2 at 800°C, 99±25 Å^2 at 850°C, and 80±19 Å^2 at 900°C. In view of the large uncertainties in these values, it is impossible to make valid inferences concerning possible temperature dependence of the cross section.

If we treat the variation of results from one cell to another at the same temperature as a statistical variation, the mean deviations are 25% in the 800°C range, 15% in the 850°C, range and 18% in the 900°C range. Since the mean deviations of σ_d as determined in each temperature range are about two-thirds of the average uncertainty, based on the least count of the measuring instruments, the internal consistency of the data appears satisfactory.

The approximations made in the development of Eq. (6) require that (a) the loss of excited mercury atoms in wall collisions can be neglected, (b) the resonance fluorescence be primary resonance fluorescence, (c) the number density of ground-state thallium atoms greatly exceed the number density of ground-state mercury atoms, and (d) the geometry of the irradiation and detection system be kept constant.

Calculations based on the gas-kinetic cross sections indicate that at the lowest thallium vapor pressures for which cross sections were determined the mean free path in the cell was about 1 mm; this mean free path would thus permit mercury atoms to reach the walls, since the distance between the primary beam and the wall was 0.5 mm. However, at the atomic velocities involved an excited mercury atom travels only about 0.004 mm in one radiative lifetime; hence relatively few excited mercury atoms were lost in wall collisions.

For the measurements summarized in Table I the number density of mercury atoms was about 10×10^{12} atoms/cm^3, corresponding to a pressure of about 2×10^{-4} mm Hg. At this pressure radiation imprisonment should be negligible, in view of the fact that the irradiated region was 0.5 mm from the window through which the fluorescence was measured. This conclusion is supported by the observation that with thallium in the cell the 5350-Å sensitized fluorescence was clearly visible along the entire path of the incident 2537-Å radiation.

The ratio $n_0{}^b/n_0{}^a$ was greater than 100 for all situations for which cross sections were calculated.

The geometry was established by the construction of the sample cells, the use of a ceramic mask, and positioning of the source, cell, and monochromators. No changes were made in the course of the experiments summarized in Table I.

The conditions used in the present experimental work, therefore, meet all the requirements necessary for the proper applicability of Eq. (6).

The de-excitation cross sections reported here are large and are of the order of magnitude of the reported quenching cross sections[10] of large organic molecules for the 6^3P_1 state of mercury. If any appreciable fraction of this cross section is due to processes in which the thallium atoms are excited then it is likely that such collisions may be used to invert the thallium population distribution.

It might be remarked that the present method can be applied to other systems exhibiting sensitized fluorescence, provided the conditions necessary for the applicability of Eq. (6) are met, the lifetime of the excited state is known, and the number densities of the colliding atoms can be determined.

ACKNOWLEDGMENTS

We would like to acknowledge the expert technical assistance of M. Ohno with the fabrication of the quartz cells and of L. G. Phillips with the construction of equipment. We are indebted to Dr. Dudley Williams for his constructive criticism of the manuscript.

[10] Reference 2, p. 161 and p. 167, and Ref. 5, p. 461.