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The density of He (2 ^{3}S) atoms in a high-pressure pulsed helium–neon laser discharge

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Abstract. The absorption of the helium 388·9 nm line by a high-pressure pulsed helium–neon laser discharge has been measured. The density and decay rate of density of He (2 ^{3}S) metastable atoms in the discharge have been estimated. While the accuracy of the decay rate measurements is of the order of 5%, that of the density measurements is probably not better than 20%, because of the difficulty of determining the shape of the collision-broadened absorption line. The measured decay rates of He (2 ^{3}S) atoms are given for mixtures with helium and neon partial pressures in the range 80 to 260 torr and 4 to 30 torr respectively. The decay rate at a fixed neon pressure is shown to be independent of the helium pressure, and to be proportional to the neon pressure at a fixed helium pressure. Agreement with values obtained by extrapolating results from low-pressure measurements of Benton *et al.* in 1962 is good. Measurements of the peak density of He (2 ^{3}S) as a function of the pulsed excitation voltage of the laser discharge are given and agree well with calculated values given by Langdon in 1968. The energy conversion efficiency for the production of He (2 ^{3}S) atoms deduced from the foregoing results is between 15 and 30%.

1. Introduction

The pulsed helium–neon laser is capable of producing peak output powers of 100 to 200 w in about 1 μs pulses (Boot and Clunie 1963). At this power level the focused output may be used for micromachining (Boot *et al.* 1963, Jackson *et al.* 1967). A detailed knowledge of the operation of the laser is desirable, particularly inasmuch as it may lead to improvements in performance. Typically the lasing tube is two metres long and two centimetres in diameter and contains helium and neon at partial pressures of 200 and 10 torr, respectively. A pulsed electric discharge is excited in the gases by means of external electrodes strapped round the tube. A number of laser transitions between the 2s and 2p levels are possible (see figure 1), but the principal ones correspond to output wavelengths of approximately 1·12, 1·15 and 1·21 μm. The upper states of the neon lasing transitions are populated principally by resonant transfers of excitation from helium atoms in the 2 ^{3}S metastable state. One of the parameters determining the output power of the laser will therefore be the density of helium 2 ^{3}S atoms.

It can be shown (Langdon 1968) that, when the helium partial pressure is high and that of the neon low, as in the present case, the principal mechanism of formation of helium metastable 2 ^{3}S atoms is direct electron excitation of neutral helium atoms. It is also possible to estimate by calculation (Langdon 1968) the density of metastable helium atoms formed during the excitation pulse. This paper describes a technique for the measurement of this density of metastable atoms and also its decay rate with time. The measured densities are compared with estimates obtained by calculation (Langdon 1968).

Egorov *et al.* (1964) and Egorov and Tibilov (1965) have measured the time variation of the He (2 ^{3}S) density in the afterglow of a high-current pulsed discharge in helium by the anomalous dispersion method. The theory of this method has been given by Mitchell and Zemansky (1961). Although the metastable density in Egorov's work was similar to that occurring in the pulsed helium–neon discharge, the time resolution of 3 to 4 μs of their technique is too long for investigations of the latter. The density of metastable atoms is estimated from measurements of the absorption, by the metastable atoms in the discharge gas, of spectral lines from a helium lamp. This method has been used by Phelps (1955) to

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measure the decay rates of He (2 ^{3}S) and He (2 ^{1}S) metastable atoms in the afterglow of a pulsed discharge in pure helium. A special time-sampling technique with phase-sensitive detection was developed (Phelps and Pack 1955, Phelps and Molnar 1953) to improve the signal-to-noise ratio of the measurements for small values of absorption. This time-sampling technique has also been used (Benton *et al.* 1962) to measure the decay rates of metastable atoms in a low-pressure helium discharge containing small quantities of neon and other gases.

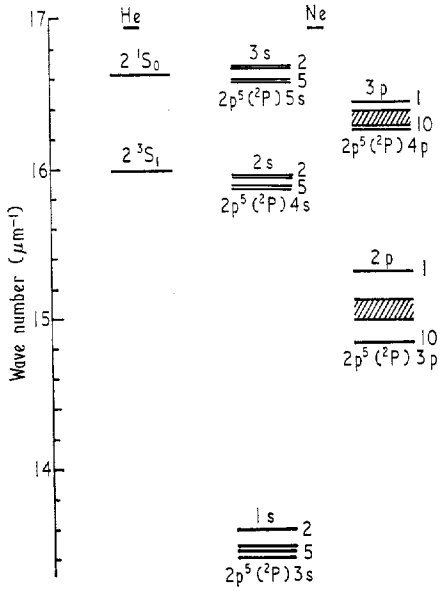

Figure 1. Energy levels of the helium–neon system.

The peak value of absorption in the work to be described is about 70%, and so the time-sampling technique of Phelps is unnecessary. The absorption can be measured adequately by direct observation of the photomultiplier output displayed on an oscilloscope as a function of time. One difference from the Phelps method is the need to remove the effect of the side-light radiation from the laser tube. This is done by measuring the total emission intensity, corresponding to the absorbed line, from the discharge in the absence of radiation from the helium lamp. This measurement is then subtracted from that of the apparent absorption.

2. The absorption measuring procedure

Calculation (Langdon 1968) indicates that only the 388·9 nm line originating on the He (2 ^{3}S) level (see figure 2) would produce a measurable absorption in the shortest path available, namely the tube diameter of 2 cm. This line was therefore selected for the measurements.

The apparatus is shown schematically in figure 3. The probing beam from the helium lamp was limited to a diameter of the tube by a mask with a 5 mm wide and 2 cm long slit.

The rise time of the photomultiplier (Mullard, 150 AVP) and oscilloscope was of the order of 10^{-8} s. Tests showed that the input-intensity–output-voltage characteristic was linear over the range of intensities used in the measurements.

The 388·9 nm emission was selected by an interference filter, with a pass band of the order of 7 nm. The response of the filter–photomultiplier combination to the various lines emitted by the lamp was checked with a monochromator. Errors due to unwanted lines, the nearest significant line being at 400 nm, were estimated to be below 1% and were neglected.

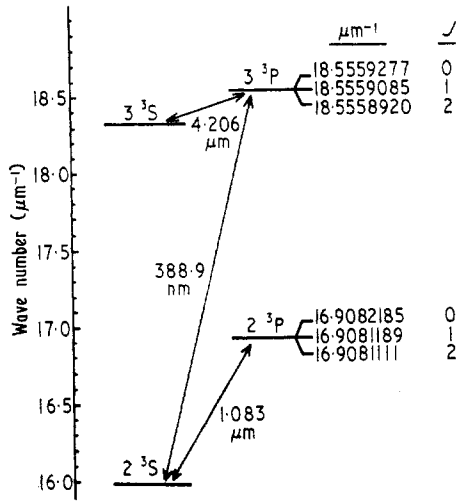

Figure 2. Helium transitions for absorption measurements.

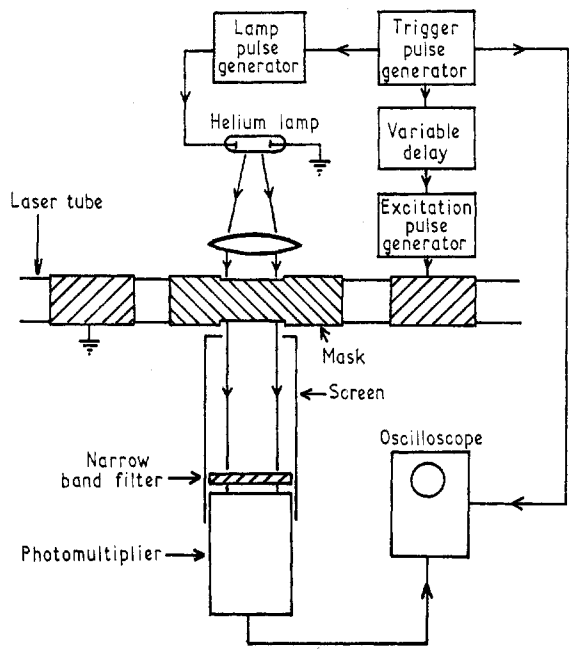

Figure 3. Absorption measuring system.

The light received at the detector contained two components: the intensity from the helium lamp transmitted through the discharge tube and the light emitted from the side of the discharge tube at the absorption wavelength. The intensity of the latter was measured separately and subtracted from the total in order to obtain the transmitted intensity. Reasonable accuracy of measurement required that the intensity from the lamp should be at least as great as that from the laser discharge tube. An appreciable sidelight intensity at the absorption wavelength was produced immediately following the excitation pulse, presumably as a result of recombination. In order to make the source intensity higher than this it was necessary to operate the helium lamp with a high current density. The lamp was pulsed with a low-duty cycle, partly to ensure a low density of metastable atoms and so reduce the effects of self-reversal and partly to keep the gas temperature low. In these

circumstances the width of the emission line should approximate to the Doppler width at room temperature. Linewidth measurements (see § 3) showed that this assumption was substantially valid.

The helium lamp (Phillips, type 93098E) had a discharge tube about 8 mm in diameter and 4 cm long. One ampere current pulses of 5 to 10 μs duration were applied to the lamp. A delay was incorporated between the pulsing of the lamp and the pulsing of the laser to allow the lamp to attain its maximum brightness before the absorption measurements commenced. Also the helium lamp was triggered at twice the repetition rate of the laser, so that alternate traces on the oscilloscope recorded the absorbed and unabsorbed intensities. For each measurement a double exposure was taken on the oscilloscope camera: one exposure recorded the absorbed and unabsorbed intensities from the lamp, together with the sidelight emission from the laser tube, and the other recorded the sidelight emission only (lamp off). The magnitude of the absorbed intensity was obtained by subtraction of the sidelight from the absorption trace, and this was then expressed as a fraction of the unabsorbed intensity. In this way the fractional absorption as a function of time was obtained.

3. The relationship between the absorption and the metastable density

The integral of the absorption coefficient k_ν is related to the population densities N_2 and N_1 in the upper and lower levels by (Mitchell and Zemansky 1961, p. 141)

$$\int k_\nu \, \mathrm{d}\nu = \frac{c^2}{8\pi {\nu_0}^2} \frac{g_2}{g_1} N_1 A_{21} \left(1 - \frac{g_1 N_2}{g_2 N_1}\right) \tag{1}$$

where ν_0 is the centre frequency of the line. In this experiment the lower level is metastable, so the population of this level will be much higher than the population of any of the higher excited levels which decay to it. That is, $N_1 \gg N_2$ and $g_1 N_1 / g_2 N_2$ may be neglected in comparison with unity. The value of A_{21} used in the calculations was $9{\cdot}5 \times 10^6 \ \mathrm{s}^{-1}$, which corresponds to the oscillator strength of 0·0645 computed by Schiff and Pekeris (1964). Allowing for the branching ratio of 8·6 in favour of the $2\ ^3\mathrm{S}$ state given by Gabriel and Heddle (1960) this agrees well with the total decay rate of 91 ± 8 ns (Fowler *et al.* 1964).

However, it is not the integral of the absorption coefficient that is directly measured. Let the intensity of the source lamp be $E_\nu \, \mathrm{d}\nu$, the absorption coefficient of the laser discharge be k_ν and the path length through the discharge be l. Then the intensity transmitted is

$$I_t = \int E_\nu \exp(-k_\nu l) \, \mathrm{d}\nu. \tag{2}$$

The fractional absorption is

$$A = 1 - \frac{\text{transmitted intensity}}{\text{incident intensity}}$$

so

$$A = \frac{\int E_\nu \{1 - \exp(-k_\nu l)\} \, \mathrm{d}\nu}{\int E_\nu \, \mathrm{d}\nu}. \tag{3}$$

Provided that the dependence of both E_ν and k_ν on frequency is known (i.e. the line shape of the emission and the absorption), it is possible to proceed numerically from the measured value of A to k_ν and hence N_1.

The line is broadened because the upper level is a triplet, $3\ ^3\mathrm{P}$. Each component of the triplet is broadened by the Doppler and pressure effects. Using the data given by Griem *et al.* (1962), the Stark broadening at the expected electron density of less than $10^{13} \ \mathrm{cm}^{-3}$ is two orders of magnitude smaller than the Doppler width.

The emission linewidth was measured with a Fabry–Pérot etalon and found to be 0·32 cm^{-1}, in agreement with figure 4, which shows the line shape to be expected if the Doppler width is 0·17 cm^{-1}, which corresponds to room temperature. The linewidth will vary during the lamp pulse because of self-reversal as the metastable density increases. That this effect is small can be deduced from the closeness of the observed linewidth to that expected from pure Doppler broadening; but also it was shown to be small experimentally. By varying the delay between the lamp pulse and the laser pulse, the absorption at a given time after the start of the laser pulse—which should not vary—can be measured at different times from the start of the lamp pulse. A change in the width of the lamp pulse would produce a change in the measured absorption. The observed change was small (e.g. 50% to 45% for a delay change of 6 μs). The measured absorption was always referred to a fixed time after the start of the lamp pulse.

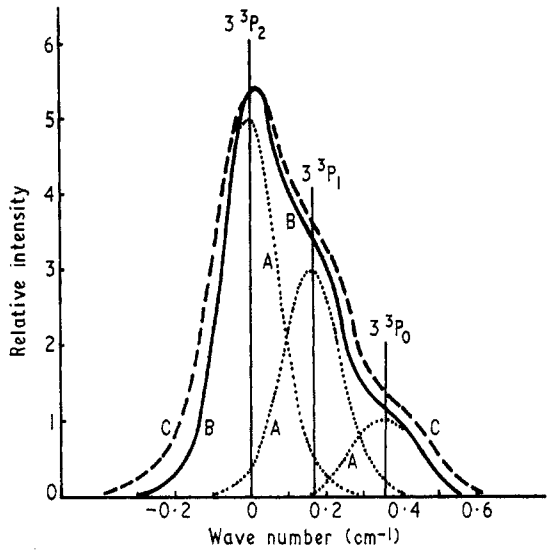

Figure 4. Line shape of 388·9 nm emission from helium lamp when Doppler-broadened at 300 °K: curve A, line shape of each component; curve B, total line shape; curve C, line shape transmitted through etalon.

It was less easy to estimate the effect of collision broadening on the absorption line Pittack (1964) has measured the broadening of some triplet lines (2 ^3P–4 ^3D, 2 ^3P–4 ^3S, 2 ^3P–3 ^3D) at a pressure of 3 atmospheres and found the widths to be 1·75, 1·3 and 1·2 cm^{-1}. If we assume the pressure broadening of the 2 ^3S–3 ^3P is roughly given by the average of these values, we obtain a broadening at the pressure used (180 torr He) of 0·11 cm^{-1}. We neglect the broadening due to the pressure of 4 torr of Ne. We assume that the broadening is likely to be less than twice this amount, that is less than 0·22 cm^{-1}. It should be noted that the broadening could be greater—the broadening of one of the singlet series (2 ^1S–3 ^1P) is very similar to the values quoted above, but the broadening of the 2 ^1P–4 ^1D lines is much greater (Pittack 1964).

Figure 5 shows the result of a numerical integration of equations (1), (2) and (3) for various amounts of pressure broadening. If the pressure broadening is taken as between zero and 0·22 cm^{-1}, as suggested above, it will be seen that there is an uncertainty of about $\pm 20\%$ in the value of the metastable density.

4. Experimental measurements of the metastable atom density and decay time

Although there is this uncertainty in the absolute value of the He (2 ^3S) density, the decay time constant of the density deduced from the time variation of the absorption will not be

affected, to a first order of approximation, by a lack of knowledge of the absorption linewidth. Figure 6 shows a plot of the decay of the He (2 ^{3}S) density in a dischage containing 180 torr of helium and 4 torr of neon. The density of the He (2 ^{3}S) atoms has been plotted using the two extremes of broadening given in figure 5, that is, 0 and 0·66 cm^{-1}. In both cases the experimental points lie close to an exponential curve and the decay time constants are the same to within 5%.

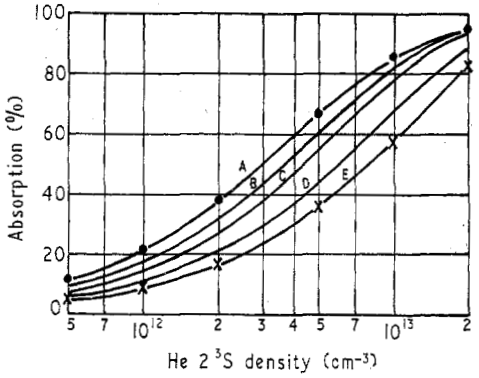

Figure 5. Absorption of He 388·9 nm line by a 5·1 cm path length of He 2 ^{3}S metastable atoms. Both source and absorbed lines Doppler-broadened by a temperature of 300 °K. Absorbed line pressure-broadened with full width at half maximum of: curve A, 0 cm^{-1}; curve B, 0·11 cm^{-1}; curve C, 0·22 cm^{-1}; curve D, 0·44 cm^{-1}; curve E, 0·66 cm^{-1}. Source line not pressure-broadened.

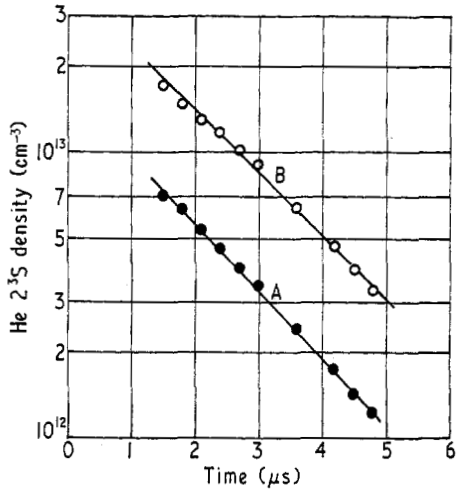

Figure 6. Decay of He 2 ^{3}S density in a He–Ne mixture of limiting values of absorption linewidth: curve A, Doppler-broadened at 300 °K only; curve B, Doppler-broadened at 300 °K and pressure-broadened with full width of 0·66 cm^{-1} at half maximum.

It was important to know whether the presence of impurities in the discharge would be likely to affect the decay rate of the metastable atoms. In order to estimate the density of impurities in the discharge, the decay of the He (2 ^{3}S) atoms in a pure helium discharge at a pressure of 180 torr was observed. The decay rate was about 1/30 of that shown in figure 6. A comparison (Langdon 1968) of the measured decay rate with that predicted by the measurements of Phelps (1955) yielded an estimate of the impurity concentration of the order of (0·5 to 5) $\times 10^{-3}$ torr. An impurity present in this concentration will certainly have no effect on the decay rate of He (2 ^{3}S) atoms in the helium–neon mixtures used experimentally.

The results of measurements of the decay time constant of He (2 ^{3}S) atoms in the helium–neon mixture as a function of the neon partial pressure is shown in figure 7. The time constant was found to be inversely proportional to the neon partial pressure over the range of pressures indicated. The time constant was also measured as a function of the helium pressure while keeping the neon pressure fixed. No significant change ($\sim 5\%$) in the decay time was observed over the range of helium pressures from 100 to 300 torr.

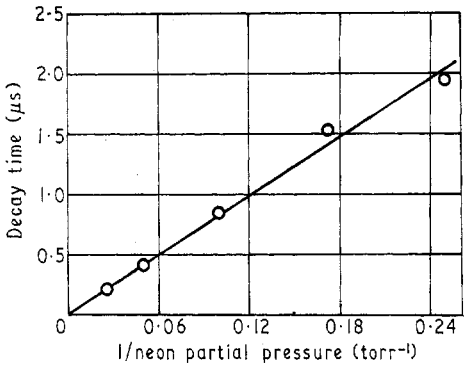

Figure 7. Variation of He 2 ^{3}S decay time with neon pressure. Partial pressure of helium, 180 torr.

The decay times of a few microseconds observed in the helium–neon mixtures are much shorter than those for the pure helium discharge. The decay of the He (2 ^{3}S) atoms is, therefore, almost entirely the result of collisions with neon atoms. Benton *et al.* (1962) have measured the total destruction cross section of the He (2 ^{3}S) atoms in collision with neon atoms by observing the decay of He (2 ^{3}S) atoms in a pulsed low-pressure helium–neon discharge. From their results the decay rate of He (2 ^{3}S) atoms as a function of the neon partial pressure can be estimated as $1{\cdot}4\times10^5\ P_{Ne}\ s^{-1}$, where P_{Ne} is in torr. The value of this quantity obtained from figure 7 is $1{\cdot}2\times10^5 P_{Ne}\ s^{-1}$. While these values are reasonably close, the discrepancy between them cannot be wholly attributed to experimental error. A discussion of this discrepancy has been given by Langdon (1968).

Since the decay time of the He (2 ^{3}S) atoms in the helium–neon discharge is considerably longer than the excitation time, of the order of 10^{-6} s in comparison with 10^{-7} s, the maximum density of He (2 ^{3}S) atoms occurs at the end of the excitation pulse. A value for this maximum density can therefore be found by extrapolating the measured He (2 ^{3}S) decay curve to coincide with the end of the excitation pulse. The maximum densities obtained in this way are plotted as a function of the h.t. voltage† in figure 8. Also shown in figure 8 are the results of calculations (Langdon 1968) of the densities of He (2 ^{3}S), He (2 ^{1}S) and excited neon atoms. It can be seen that there is a fair measure of agreement between the experimental and calculated results for the He (2 ^{3}S) atoms, if we bear in mind the uncertainty factor of 20% in the absolute values of the density of He (2 ^{3}S) atoms determined experimentally.

An interesting comparison can be made between the peak energy stored in the He (2 ^{3}S) atoms and the electrical energy dissipated in the discharge. The total number of He (2 ^{3}S) atoms created was calculated by assuming that the 2 ^{3}S atoms were uniformly distributed over all regions of the discharge. A measurement of the distribution of sidelight radiation emitted from all points of the discharge indicated that uniformity of excitation over the whole of the discharge could be assumed. The total energy stored was then obtained by multiplying the total number of excited atoms by the excitation energy (19·8 ev) of the

† This is the voltage on the primary side of a pulse transformer. The laser tube excitation voltage is obtained from the secondary of the tranformer which has a voltage step-up ratio of 4:1. The h.t. voltage is a measure of the stored energy which is to be dissipated in the discharge and is therefore a convenient parameter to describe the degree of excitation of the discharge.

state. This energy was then expressed as a fraction of the input energy actually dissipated in the discharge. The method of calculating this energy by numerical integration with respect to time of the product of the voltage across the gas with the current through it has been given by Langdon (1968). The results are shown in figure 9. The energy conversion efficiency for the production of He (2 ^{3}S) atoms is between 15 and 30%, and so it can be concluded that this process is not the major cause of the low overall power conversion efficiency in this type of laser. The main reason for the low power conversion efficiency is that the energy required to excite the laser levels ($\sim$20 ev) is very much larger than the laser photon energy ($\sim$1 ev).

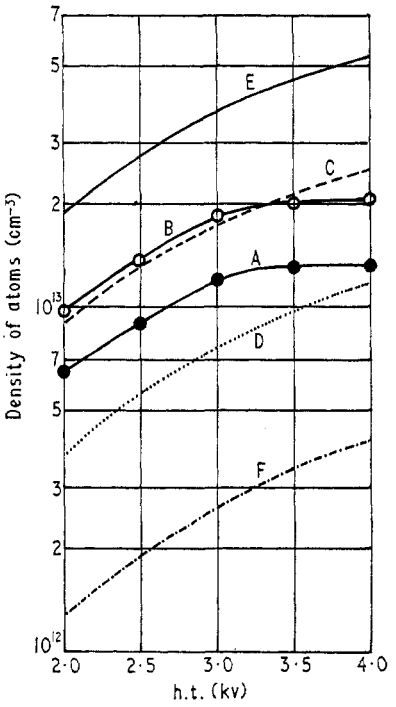

Figure 8. Calculated and measured energy level populations. He 2 ^{3}S density from absorption measurements: curve A, Doppler-broadened at 300 °K only; curve B, Doppler-broadened at 300 °K and pressure-broadened with full width of 0·22 cm^{-1} at half maximum. From calculations (Langdon 1968): curve C, He 2 ^{3}S density; curve D, He 2 ^{1}S density; curve E, total helium excitation; curve F, total neon excitation. Partial pressure of neon, 4 torr; partial pressure of helium, 180 torr.

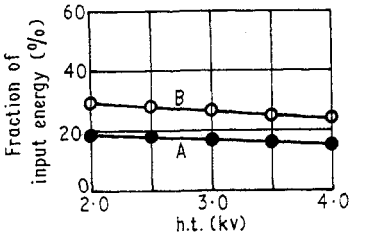

Figure 9. Fraction of input energy converted into potential energy of He 2 ^{3}S atoms: curve A, Doppler-broadened at 300 °K only; curve B, Doppler-broadened at 300 °K and pressure-broadened with full width of 0·22 cm^{-1} at half maximum. Partial pressure of neon, 4 torr; partial pressure of helium, 180 torr.

5. Conclusions

An optical absorption technique for the measurement of the density of He (2 ^{3}S) metastable atoms in a high-pressure helium–neon discharge has been described. The determination of the absolute magnitude of this density by this technique is uncertain to about 20%. The cause of this uncertainty is the difficulty of the determination of the shape and width of the 388·9 nm absorption line of the laser discharge. The accuracy of the measurement of the decay rate of the density of metastable atoms is, however, not limited by the lack of precise knowledge of the absorption line shape and linewidth and is determined to an accuracy of about 5%.

The results presented show how the decay rate of the density of He (2 ^{3}S) atoms depends on the partial pressures of neon and helium in a laser discharge and agree well with the rate predicted from low-pressure measurements. The result of an experimental estimation of the maximum density of He (2 ^{3}S) atoms, as a function of the degree of excitation of the discharge, is also presented. This result is compared with the results of calculations given elsewhere. The efficiency of conversion of input excitation energy into potential energy of He (2 ^{3}S) atoms is also estimated as a function of the degree of excitation.

Acknowledgments

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