Reflection of Metastable Atoms by a Glass Wall in a Positive Column Discharge Plasma

by

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Abstract

The leser-induced-fluorescence-spectroscopy (LIFS) method has been applied to a dc discharge plasma, and the radial density distribution of metastable ($2\ ^1S$) helium atoms has been measured. It was found that the density did not tend to zero at the discharge tube wall. Rather, the density was about 20 - 30% of the value on the tube axis. By using the collisional-radiative model, we interpreted the result. As the origin of this finite population at the wall we considered two possibilities, namely, 1) the helium ions which recombine at the wall converted partly into the metastable atoms and 2) the metastable atoms were not quenched completely at the wall, but were reflected by a certain amount. We found that the first process could not reproduce the experimental population distribution, and that the second process with the reflection coefficient of the metastable atoms of 80 - 90% accounted for the experiment.

§ 1. Introduction

It is commonly assumed that, in a weakly ionized plasma confined in a glass cell, ions recombine with electrons upon collisions with the glass surface to form ground-state atoms or molecules, and that metastable atoms and molecules are quenched completely. As its result, the population density of these excited or ionized atoms or molecules tends to zero at the glass surface. In other words, if we find that a population density of some of these species has a finite value at the surface, it means that at least one of the above assumptions is incorrect.

There is a report[1] that metastable atoms in a helium discharge plasma and in a neon discharge plasma have population densities which are not zero at the discharge tube wall. These authors ascribe these finite populations to the reflection of these metastable atoms, and they deduce the reflection coefficient. The coefficient sometimes amounts to 0.9. This experiment strongly suggests that, at least under certain conditions, the above assumptions are not justified.

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Recently, investigations of the solid surface by using a metastable atomic beam have revealed that a metastable atom "sees" only the first monolayer of atoms on the surface. This means that if the solid has adsorbed atoms or molecules on its surface, reflection or quenching of metastable atoms is determined by the outer monolayer atoms or molecules, not by the bulk solid material.

It is well known that, in many cases, the charge transfer recombination of ions by collisions with neutral atoms results in excited neutral atoms, not in the ground-state atoms. Thus, there is a possibility that the recombination of an ion, which diffuses out of the plasma to the wall and there picks up an electron, results in an excited species. This process would lead to a finite population as well of these excited atoms or molecules at the tube wall.

In the following, we report the result of our preliminary experiment in which we investigate the above possibilities.

§ 2. Experiment

The discharge tube was made of fused quartz having an inner radius of R= 6 mm and a length of 6 cm. A dc discharge was run through helium by a brush cathode made of tungsten needles. This use of the special cathode was for the purpose of increasing the stability of the discharge. By using the microwave-cavity resonance method we measured the electron density (n_e) and its temporal fluctuation. The cavity had a deameter of 55.9 mm and a length of 50 mm, and the resonance frequency of the TM_{112} mode was 9.25 GHz when the discharge plasma was absent. Details of the method are given in refs. 2 and 3. Figure 1 (a) shows n_e which is determined from the shift of the microwave resonance peak as a function of the discharge current. Figure 1 (b) shows the region of the discharge conditions under which the plasma is quiet, or the fluctuation of n_e is less than 1%. In the followig experiment we chose the pressure of 1.5 torr, because at this pressure the plasma is quiet over a wide range of discharge current.

The emission radiation from the plasma was observed by a monochromator (Nikon G 250, $f/4.5$, 25 cm focal length and 34 A/mm linear reciprocal dispersion) with the entrance and exit slits set parallel to the discharge tube axis. Their widths were 1 mm and the heights were 10 mm. The absolute population density of the metastable 2 ^1S atoms was determined by the self ablorption method; a concave mirror was placed at the opposite side of the monochromator to focus the image of the plasma on the plasma itself. We measured the emission

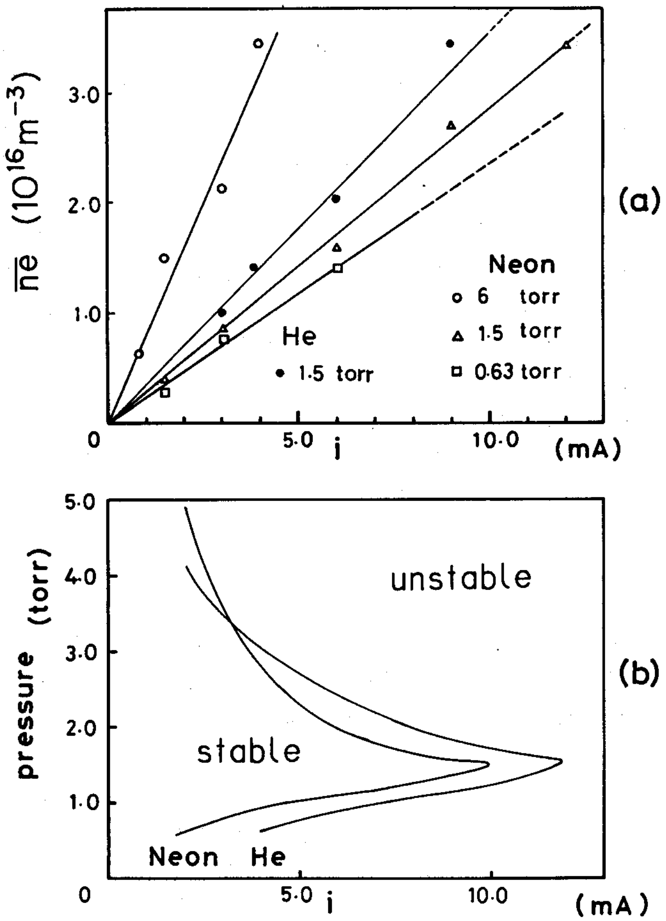

Fig. 1. (a) Averaged electron density $\overline{n_e}$ as determined from the shift of the microwave-cavity resonance frequency. (b) The region of discharge conditions under which the discharge is quiet, i.e., the fluctuation of $\overline{n_e}$ is less than 1%.

intensities of the 5016 A (2 ^1S–3 ^1P) line with and without the mirror. The population density of the lower level averaged over the tube diameter was determined from the quantity called the line absorption, or the intensity ratio, where the Voigt line profile was assumed. Besides the thermal Gaussian component, we included the effect of the natural width and the resonance broadening[4] for the 3 ^1P level. Figure 2 shows the result.

A nitrogen-laser-pumped dye laser delivered a 5016 A light pulse. The

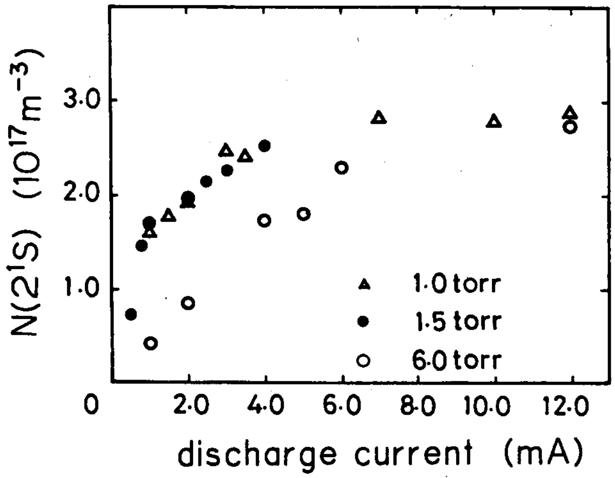

Fig. 2. Averaged population density of the helium metastable 2 ^1S atoms determined by the self absorption metod.

duration was about 5 ns and the repetition rate was 26 pulses per second. The spectral width was estimated to be the order of 0.1 A. The laser light was transmitted through a 20 m multimode optical fiber in order to delay the light pulse, and to depolarize the partially polarized laser light. The laser light emerging from the optical fiber was focused with a microscope objective lens. The beam diameter was determined by the core diameter of the fiber and the focal length of the lens. It was about 0.8 mm at the location of the plasma which was observed by the monochromator. We observed the direct fluorescence intensity of the 5016 A light. We recorded the peak fluorescence intensity immediately after the laser irradiation. A small contribution from the scatter of the laser light by the tube wall was corrected. For a constant laser intensity, the fluorescence intensity was proportional to the lower-level population. (Actually, the laser intensity was so strong that the pumped upper level population was nearly saturated, and a fluctuation of the laser intensity little affected the result.) The effect of absorption of the fluorescence on its way through the plasma was corrected. The discharge tube was moved laterally while the fluorescence intensity was measured. Figure 3 shows the population-density distribution over the tube radius, which is normalized to the value on the tube axis. The striking feature is that it does not tend to zero at the tube wall.

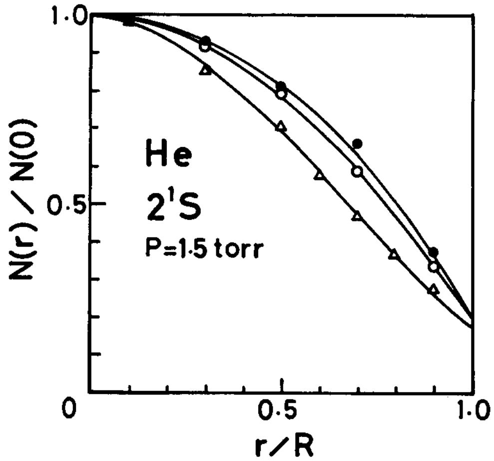

Fig. 3. The population density distribution of the helium $2\ ^1S$ metastable atoms determined by the LIFS method. The distribution is normalized to the value on the tube axis. △ : $\bar{n}_e = 1.3 \times 10^{15}\ m^{-3}$, ○ : $\bar{n}_e = 1.3 \times 10^{16}\ m^{-3}$, ● : $\bar{n}_e = 6 \times 10^{16}\ m^{-3}$.

§ 3. Theory

We assume a cylindrical geometry of the plasma and a stationary state. The diffusion equation of the metastable atoms is given as

$$\frac{\partial}{\partial t} n_{S(T)}(r) = -D_{S(T)} \frac{1}{r} \frac{\partial}{\partial r} \left[r \frac{\partial n_{S(T)}(r)}{\partial r} \right] + K_{S(T)}(r) \tag{1}$$

where $D_{S(T)}$ is the diffusion coefficient for the singlet (triplet) metastable atoms $2\ ^1S$ ($2\ ^3S$), $n_{S(T)}(r)$ is the population density at the radial position r and $K_{S(T)}$ is the collision term. The latter quantity is given from the collisional-radiative-model calculation as[1)]

$$K_S = k_{1S} n_g n_e + k_{TS} n_T n_e - k_S n_S n_e - k_m (n_S + n_T) n_S \tag{2}$$

for the singlet state and a similar equation for the triplet state with S replaced by T. Here k_{1S}, k_{TS} and k_S are the effective rate coefficients for the electron collisions, including indirect processes through intermediate states. k_{1S} is for the excitation from the ground state to $2\ ^1S$, k_{TS} is for the excitation $2\ ^3S \rightarrow 2\ ^1S$, and k_S is for the depopulation from $2\ ^1S$. k_m is the ionization rate coefficient for metastable–metastable collisisons, where we have assumed that the singlet and triplet metastable atoms behave equally. The radial dependence of the rate coefficients and of the densities are omitted in the expression of Eq. (2).

By assuming the Maxwellian distribution for electron velocities, we performed the collisional-radiative-model calculation for helium.[5] For the loss mechanism of the helium ions the ambipolar diffution was assumed. From a balance of the neutral and ionized helium densities, we fixed the electron temperature (T_e). T_e was about 4 eV over the range of our experimental conditions. We assumed the zero-th order Bessel function $J_0(2.405\,r/R)$ for the spatial distribution of elctrons (see later), and by combining this distribution with the absolute n_e value obtained from the microwave-cavity resonance method (Fig. 1 (a)), we determined the electron density at the radial position r to be $n_e(r)$.

We divided the entire plasma column into 15 concentric cylindrical layers. For each layer we calculated the various effective rate coefficients which appeared in Eq. (2). The differential equation (1) reduced to a difference equation, and we solved this equation with the boundary condition described below.

The particle flow of metastable atoms at position r is given by

$$\Gamma(r) = -D\partial n(r)/\partial r \tag{3}$$

At the plasma boundary, or at the wall surface, the flow out of the plasma is expressed as

$$\Gamma_+ = \frac{n(R)v}{4} - \frac{D}{2}\frac{\partial n(r)}{\partial r}\Big|_{r=R} \tag{4}$$

where $n\ (R)$ and $\partial n(r)/\partial r\,|_{r=R}$ are the metstable population and its gradient, respectively, at the wall. Here, v is the thermal speed of the atoms. If some of the metastable atoms are reflected at the surface with the reflection coefficient R_e, and if some of the ions which recombine at the surface convert into the metastable atoms with the conversion efficiency c, the inward particle flow is given as

$$\Gamma_- = R_e\Gamma_+ + c\Gamma_i \tag{5}$$

where Γ_i is the outward flow of ions at the wall, and this is given as

$$\Gamma_i = \pi R^2(2.405/R)^2 D_A n_i \tag{6}$$

Here, D_A is the ambipolar diffusion coefficient, and n_i is given by n_e from the condition of the charge neutrality. The net flow of the metastable atoms at the wall is given as

$$\Gamma(R)=\Gamma_{+}-\Gamma_{-}$$
$$=(1-R_e)\Gamma_{+}-c\Gamma_i \qquad (7)$$

In our calculation, we took Eq. (7) as the boundary condition with the variable parameters R_e and c.

An example is shown in Fig. 4 of the results of the calculation of the

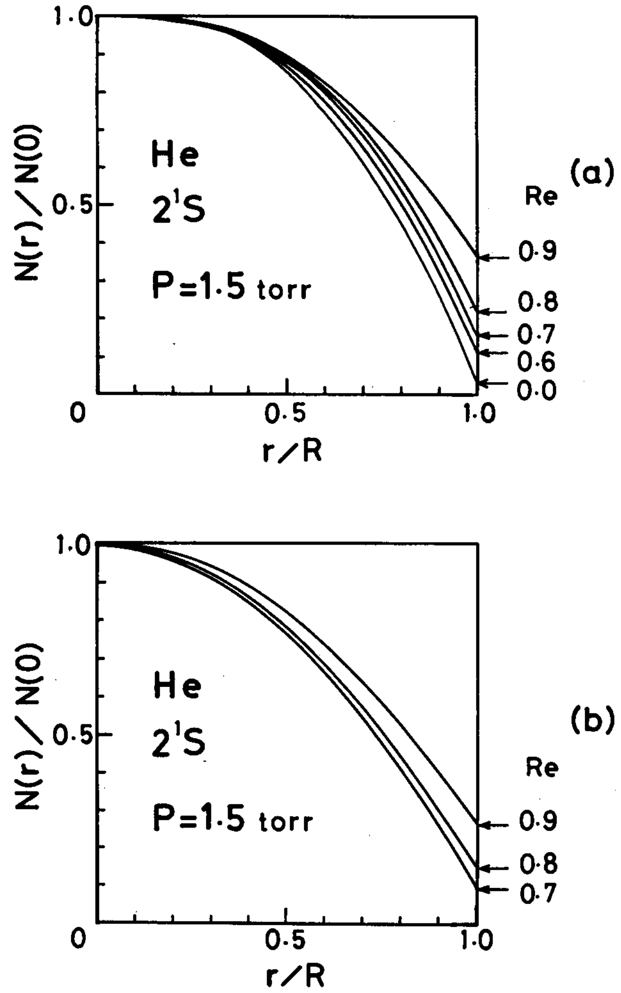

Fig. 4. Calculated population density distribution of the helium 2 ^1S metastable atoms. The reflection coefficient R_e has been varied as a parameter. The conversion coefficient of the ions is set equal to 0. (a) $\overline{n_e}=6\times10^{16}\,\mathrm{m}^{-3}$, (b) $\overline{n_e}=1\times10^{15}\,m^{-3}$.

population density distribution of the metastable atoms over the tube radius. This is the normalized distribution to the value on the tube axis. We find that the effect of c, the conversion of the ion flow, does not appreciably change the result. This is because, as is seen in Figs. 1 and 2, n_i is about an order smaller than the metastable population, and the diffusion coefficients for the ions and the metastable atoms are of the same order. On the other hand, as is seen in Fig. 4, reflection of the metastable atoms has a substantial effect on the density distribution.

§ 3. Results and Discussion

When we compare carefully the result of the calculation, Fig. 4, with that of the experiment, Fig. 3, we find that the experimental profile is more "peaked" near the tube axis than the calculation. This inconsistency is probably due to the failure of the assumptions: 1) the Bessel function distribution of electron density, 2) the Maxwellian distribution of electron velocities, or both. A deviation of electron density from the Bessel function is well known in the case of the neon discharge as the phenomenon "contraction". We actually found that the population density of the neon metastable atoms are more peaked near the

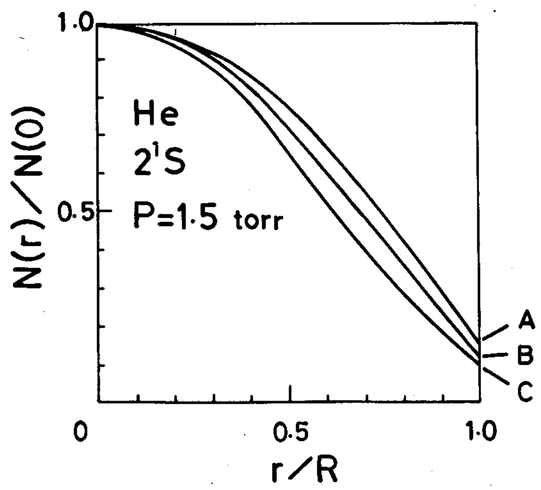

Fig. 5. Calculated population density distribution under the assumption that the electron density distribution is described by the powers of the zero-th order Bessel function J_0 $(2.405\,r/R)$.
(a) $n_e \propto J_0$ (b) $n_e \propto J_0^2$, (c) $n_e \propto J_0^3$; $\overline{n_e} = 1 \times 10^{15}\,m^{-3}$.

tube center than the present case of helium. (See Appendix.) The second deviation is also well known. The high-energy part of the velocity distribution is lower than the thermal Maxwellian distribution. There is a report[6] that this deviation is more pronounced near the tube wall than near the center. For the purpose of effectively taking these effects into account in our calculation, we made calculations in which the electron density distribution is assumed to be more peaked near the tube center, i. e., the density distribution is proportional to the powers of the zero-th order Bessel function. Figure 5 shows an example of the results. In these calculations, the reflection coefficient R_e is assumed to be 0.8. It is seen that these population distributions fit the experimental ones much better than the original distributions in Fig. 4.

We conclude that a direct comparison of the theoretical and experimental profiles is rather inadequate for the purpose of determining R_e. Instead, we followed the following alternate method.

The boundary condition, Eq. (7), is rewritten as

$$1-\Gamma(R)/\Gamma_+=R_e+c\Gamma_i/\Gamma_+ \tag{8}$$

From Figs. 2 and 3, we can determine $\Gamma(R)$ by using Eq. (3), and Γ_+ by using Eq. (4), and Γ_i is given from Fig. 1 and Eq. (6). Figure 6 shows the plot of Eq. (8). From the intercept and from the slope we determine R_e and c, respectively. As expected, c is too small to be detected, and R_e is about 0.9.

We draw a conclution that under our particular discharge condition, about

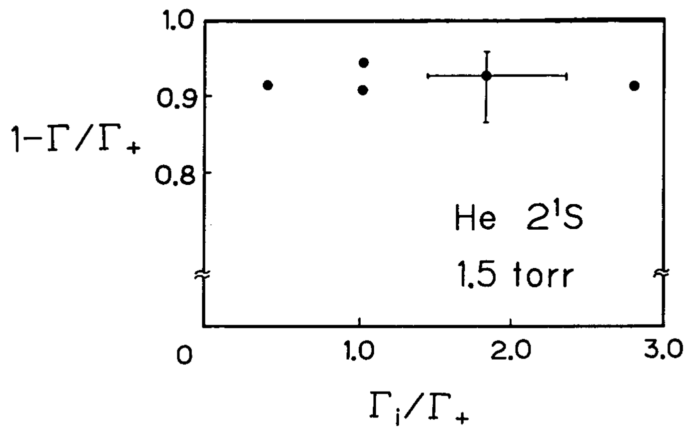

Fig. 6. Experimental plot of Eq. (8). The intercept gives the reflection coefficient R_e, and the slope gives the conversion oefficient c.

90% of the singlet metastabel helium atoms are reflected back to the plasma.

Appendix

We made a similar experiment for a neon discharge, where the population distribution was measured for the metastable $1s_5$ level and the resonance $1s_4$

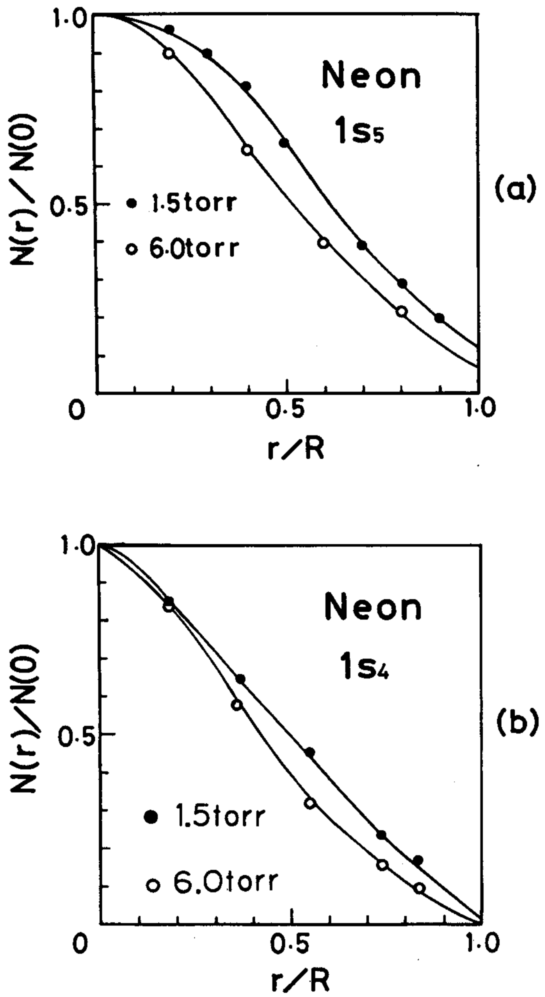

Fig. A 1. (a) The population density distribution of the neon metastable $1s_5$ atoms and (b) that the resonance $1s_4$ atoms as determined by the LIFS method.

level. Fig. A 1 shows the result. In this case, the metastable atoms have a finite population close to the discharge tube wall, while the resonance level has a density distribution which tends to zero at the wall. Fig. A 2 shows the relative population of the metastble levels close to the wall, *i. e.*, at $r = 0.87R$, as a function of the filling pressure (a) and of the discharge current (b).

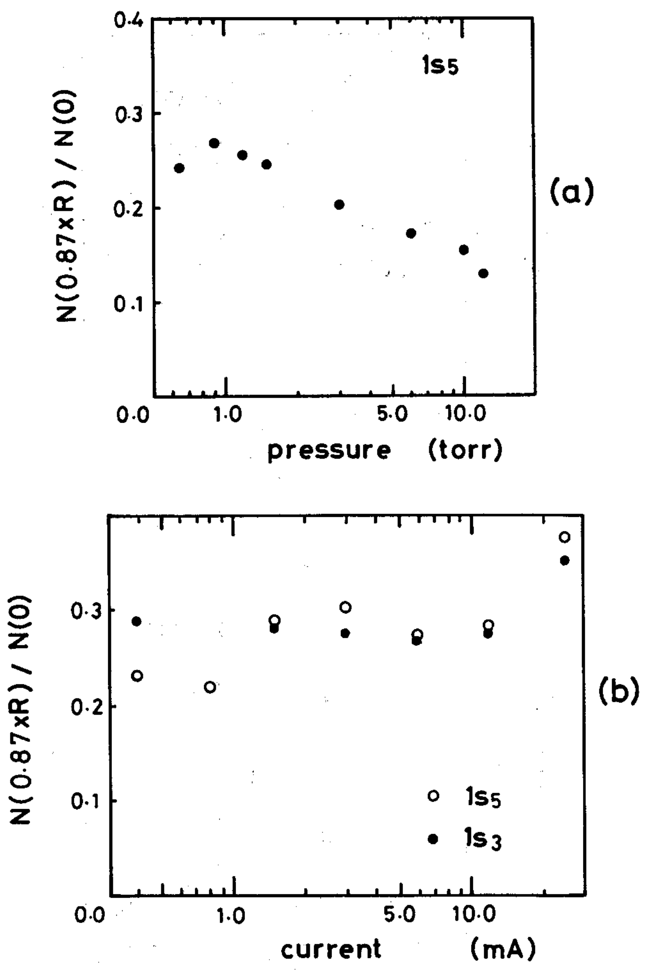

Fig. A 2. (a) The relative population density of the metastable atoms near the tube wall as a function of the filling pressure and (b) that of the discharge current. The discharge current is 0.8 mA for (a) and the pressure is 1.5 torr for (b).

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