Depolarization of excited Ne*($2p^5 3p$; J=1) atoms due to He atom collisions

Hiraku MATSUKUMA, Cristian BAHRIM[1)] and Masahiro HASUO
Department of Mechanical Engineering and Science, Kyoto University, Kyoto 606-8501, Japan
[1)] *Department of Chemistry and Physics, Lamar University, Beaumont, TX 77710-10046, USA*

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In a temperature controlled glow discharge of a mixture of neon and helium gases at 77 and 292 K, a linearly polarized laser pulse produces polarized neon atoms on the $2p_5$ (in Paschen notation) level (atomic alignment) from the $1s_3$ level. We measure the direct fluorescence from the $2p_5$ level to the $1s_2$ level with its polarization components resolved. From the temporal evolution of the intensities for these components, we determine the disalignment rate. We report the rate coefficients due to helium-neon collisions determined from the dependence of the disalignment rate on the helium atom density. We compare our experimental rate coefficient with the theoretical value obtained from quantum close-coupling calculations based on the model potential for the Ne*($2p^5 3p$)-He system proposed by Hennecart and Masnou-Seeuws in 1985, and find that the rate coefficients predicted by our calculations are about 90 % at 77 K and 60 % at 292 K larger than the experimental values. New calculations based on a modified model potential, which includes an additional static dipole polarizability term for the Ne*($2p_5$) atom to the model potential proposed by Hennecart and Masnou-Seeuws, show agreement with our experimental data within 30 % for both temperatures.

Keywords: polarization, atomic collisions, disalignment, glow discharge, laser-induced fluorescence spectroscopy, atom-atom potential, quantum cross section

1. Introduction

The importance of atomic polarization in plasmas is recognized through the study of the anisotropy in plasmas. Plasma polarization spectroscopy (PPS) and the collisional-radiative model including polarization effects have been recently developed [1]. Depolarization effects may also be important for quantitative analysis of PPS. In high density plasmas, atomic collisions can induce the depolarization of atoms.

On the other hand, the depolarization due to atomic collisions is known to offer accurate information about anisotropic potentials between colliding atoms [2,3]. We have measured the depolarization of neon Ne*($2p_i$; J=1) atoms (in Paschen notation) excited on levels of the $2p^5 3p$ configuration due to collisions with helium (He) atoms based on a method which resolves polarized fluorescence using a laser-induced fluorescence spectroscopy (LIFS) technique [2,4,5]. Here J is the total angular momentum quantum number. Also, we have compared our experimental results with full quantum calculations [4] based on a model potential for the Ne*($2p^5 3p$)-He system [6]. For the depolarization of Ne*($2p_2$ and $2p_7$) atoms excellent agreement between experiment and theory is found at temperatures between 77 and 600 K when a static dipole polarizability for each Ne*($2p_i$) level is added to the long-range potential from [3,6]. It appears that the static dipole polarizability is stronger for the $2p_2$ level [7] than for the $2p_7$ level.

Here, we report our measurements for the depolarization of Ne*($2p_5$) atoms due to He atom collisions at 77 and 292 K, and compare with quantum calculations including the polarizability of the $2p_5$ level, which is expected to have a strong contribution to the disalignment rates as in the case of the $2p_2$ level.

2. Experiment

A schematic diagram of the experimental set-up for the LIFS measurement is shown in figure 1. A temperature controlled glow discharge cell is made of fused quartz. The discharge tube is composed of a cylindrical discharge channel (5 mm inner diameter and 190 mm length), its surrounding layer for the temperature control of the channel and a vacuum layer for the thermal isolation from outside. Detailed explanation of the temperature controlled discharge cell is given in [4]. The discharge channel is filled with pure neon gas or a mixture of neon and helium gases. We set the temperature at 77or 292 K. We measure the gas pressure by a ceramic capacitance manometer (ULVAC, CCMT-100) at room temperature. The DC discharge current is 1.00 mA. We define the (x,y,z) coordinate as shown in figure 1.

The excitation light source is a dye laser (ELTO, LT1233; DCM dye solved in methanol) pumped by a frequency-doubled YAG laser (Spectra Physics, GCR-100). The pulse duration of the laser light is 5ns (FWHM) and the pulse repetition rate is 50Hz. We detect a small part of

author's e-mail: hiraku@t04.mbox.media.kyoto-u.ac.jp

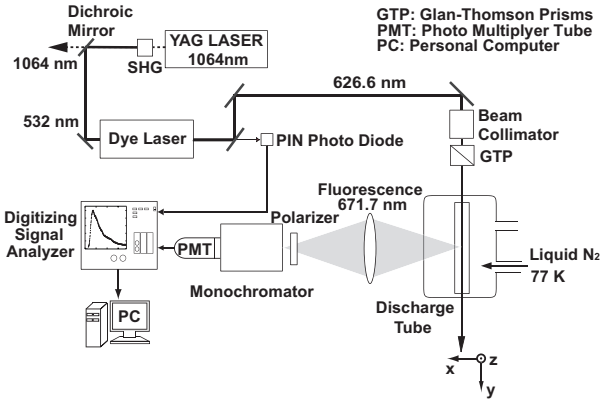

Fig.1 Schematic diagram of the LIFS set-up

the laser light by a PIN photo diode (Hamamatsu, S3883) and use its output as the monitor of the excitation energy and also as the trigger of a digitizing signal analyzer (Tektronix, DSA601). The main part of the laser light is linearly polarized along the z direction by Glan-Thomson prisms located in front of the discharge cell's window. Because we define the quantization axis along the z direction, the excitation laser light is π-polarized. The light beam goes through the discharge channel along its central axis (y direction). The diameter of the light beam is about 2 mm at the position of observation. In order to excite the neon atoms from the $1s_3$ (J = 0) level to the $2p_5$ (J = 1) level, we set the wavelength of the laser at 626.6 nm.

We observe the fluorescence (of 671.7 nm) for the transition from the $2p_5$ (J=1) level to the $1s_2$ (J = 1) level through a slit located on the side wall of the discharge channel along the x direction in figure 1. We analyze the fluorescence with a linear polarizer located in front of the entrance slit of a monochromator (Nikon, G-250). This allows to resolve the σ- and π-components of the fluorescence. We set the entrance slit parallel to the discharge channel. The width and height of the entrance and exit slits are 0.5 and 15 mm, respectively. We measure the intensity of the fluorescence with a photomultiplier tube (Hamamatsu, R3237-01) and record the temporal evolution of its output signal by using a digitizing signal analyzer with 1 ns temporal resolution. The digitized signals are accumulated over laser excitation pulses. The number of accumulation is 512. We transfer the recorded signal data to a personal computer through GPIB.

In order to determine the relative sensitivity of π- to σ-component in our measurement system, we polarize the excitation light along the x-direction. In this case, the fluorescence intensity is independent of its polarization direction around the x-axis. From the comparison of the measured fluorescence intensities between the π- and σ-components, we determine that the relative sensitivity is 0.644 at the wavelength of 671.7 nm.

3. Results and discussion

Figure 2(a) shows an example of the temporal evolution of the fluorescence intensities. The intensities of

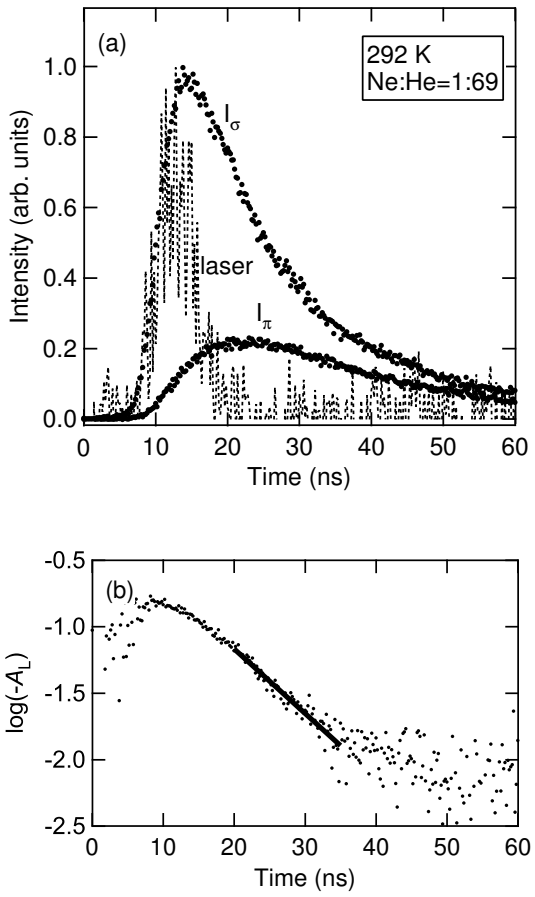

Fig.2 (a) An example of the observed temporal evolution of the fluorescence intensities with the π- and σ-components resolved, and the excitation laser pulse. The temperature is 292 K and the partial pressures of neon and helium gases are 0.05 and 3.47 Torr, respectively. (b) The temporal evolution of the longitudinal alignment calculated using data from (a).

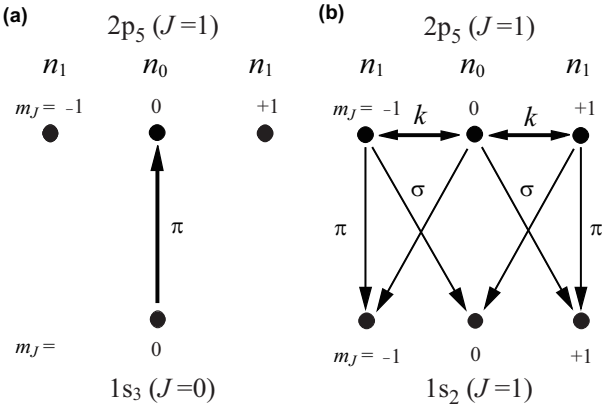

Fig.3 Kastler diagrams (a) for the $1s_3$-$2p_5$ transition showing the π-light excitation and (b) for the $2p_5$-$1s_2$ transition showing the emission and the intramultiplet excitation transfer processes.

the π- and σ-components are calibrated with their relative sensitivity. Since the excitation is done with the π-polarized light in the $J = 0$ to $J = 1$ transition, the initial population of the upper level is produced only on the $m_J = 0$ sublevel as shown by figure 3(a), where m_J is the magnetic quantum number. During the lifetime of the upper level, excitation transfer from the $m_J = 0$ sublevel to $m_J = +1$, -1 sublevels, or disalignment, may take place. In consequence, the π–component appears in the fluorescence (figure 3(b)).

After the cessation of the excitation laser pulse (for $t >$ 20 ns in figure 2(a)), the rate equations for the sublevel populations in the upper $2p_5$ level are

$$\frac{dn_0(t)}{dt} = -(2k+\gamma)n_0(t) + 2kn_1(t) \qquad (1)$$
$$\frac{dn_1(t)}{dt} = kn_0(t) - (k+\gamma)n_1(t),$$

where n_0 and n_1 are the populations of the $m_J = 0$ and $m_J = +1$ (or $m_J = -1$) sublevels, respectively, of the $2p_5$ level; k is the excitation transfer rate between the $m_J = 0$ and $m_J = +1$ (or $m_J = -1$) sublevels; and γ is the depopulation rate of the Ne*($2p_5$) atoms. Because of the symmetry of the excitation, the sublevels with $m_J = +1$ and $m_J = -1$ are equally populated and therefore, we need to include only n_1 in equation (1).

The intensity of the fluorescence of each polarized component is given by

$$I_\sigma(t) = C\frac{1}{2}[n_0(t) + n_1(t)] \qquad (2)$$
$$I_\pi(t) = Cn_1(t),$$

where C is a constant. We define the longitudinal alignment for the emitted radiation as

$$A_L(t) \equiv \frac{I_\pi(t) - I_\sigma(t)}{I_\pi(t) + 2I_\sigma(t)} = \frac{1}{2}\frac{n_1(t) - n_0(t)}{n_0(t) + 2n_1(t)}. \qquad (3)$$

The second equality in (3) is easily found by substitution of the π– and σ–components from equation (2). Using equation (1), one can find the temporal evolution of A_L expressed as

$$A_L(t) = -\frac{1}{2}e^{-3kt}. \qquad (4)$$

The decay constant, $3k$, is the disalignment rate. Figure 2(b) shows the temporal evolution of $\log[-A_L(t)]$ calculated from the fluorescence intensities presented in figure 2(a). From the slope of the linear fit calculated with equation (4) and applied to $\log[-A_L(t)]$, as shown in figure 2(b), we determine the disalignment rate. The fit is done for a time period running from the cessation of the laser pulse (t = 20 ns) till the time when the scatter of the data points becomes too large, which is t =35 ns in the case of figure 2(b).

Figure 4 shows the disalignment rate for the Ne*($2p_5$) atoms as a function of the helium atom density at 77 and 292 K. During our measurement at each temperature, the partial pressure of neon gas is fixed at 0.05 Torr, while the helium gas pressure varies from about 0.7 to 6.0 Torr. A linear dependence of the disalignment rate with the helium atom density can be observed.

In our cell experiment, disalignment due to electron collisions, radiation trapping and neon-neon collisions are always present together with the one due to helium-neon collisions. Because in our experimental conditions the electron density is about 10^{16} m^{-3} [8], the disalignment rate due to electron collisions is about 10^4 s^{-1} [9] and therefore can be neglected.

The disalignment effect of the radiation re-absorption for the Ne*($2p_2$) atoms was quantitatively estimated in a similar discharge condition in [10]. Since the transition lines from the $2p_2$ and $2p_5$ levels have Einstein's A coefficients of the same order, the disalignment rate of the Ne*($2p_5$) atoms due to the radiation re-absorption is expected to be similar as for the Ne*($2p_2$) atoms, which is about 10^6 s^{-1}, and therefore, it is one or two orders of

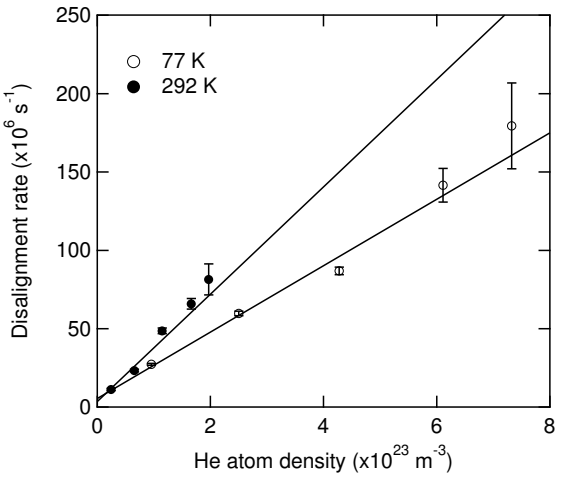

Fig.4 Disalignment rate of the Ne*($2p_5$) atoms as a function of the helium atom density.

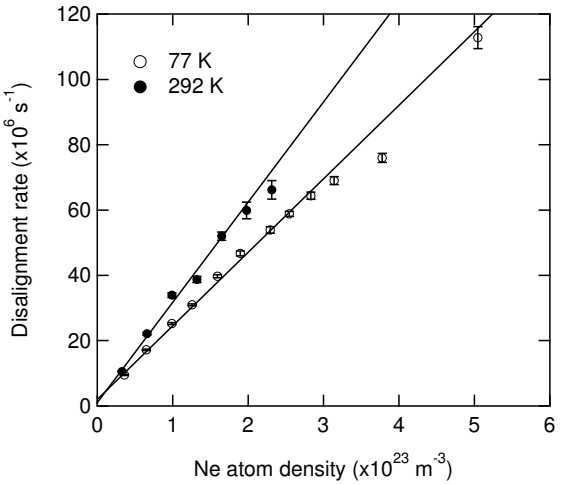

Fig.5 Disalignment rate of the Ne*($2p_5$) atoms as a function of the neon atom density.

magnitude smaller than the rate shown in figure 4.

Since we kept the neon density constant for all the helium densities at each temperature, the disalignment rate due to neon atom collisions is constant for all helium densities. Figure 5 shows the disalignment rate for the Ne*($2p_5$) atoms as a function of the neon atom density determined from the experiment with pure neon gas. A linear dependence on the neon atom density is observed, from which we estimate a disalignment rate coefficient due to neon atom collisions of 2.25 x 10^{-16} and 3.07 x 10^{-16} $m^3 s^{-1}$ at 77 and 292 K, respectively. These rate coefficients give the disalignment rate at 0.05 Torr and 77 K of about 3.4 x 10^6 s^{-1} while at 292 K is 1.5 x 10^6 s^{-1}, which is also one or two order of magnitude smaller than the observed rate in figure 4.

From these considerations, we fit the helium atom density dependence of the disalignment rate shown in figure 4 using a linear function with an intercept which includes the disalignment rates due to radiation re-absorption and neon-neon collisions. We determine the disalignment rate coefficient of Ne*($2p_5$) atoms due to helium-neon collisions as the slope of the linear fit shown in figure 4. The experimental rate coefficients measured at temperatures of 77 and 292 K are shown in figure 6 by the closed circles.

Recently, Bahrim and Khadilkar [7] have re-analyzed the long-range potential for the Ne*($2p^5 3p$)-He system proposed by Hennecart and Masnou-Seeuws [6], and calculated the disalignment rate coefficient of the Ne*($2p_2$) atoms induced by helium-neon collisions using a quantum multi-channels close-coupling method. When a more repulsive interaction between the He and Ne*($2p_2$) atoms is incorporated in the model potential from [6] an excellent agreement between experiment and theory at temperatures from 77 K to 600 K is found [7]. This finding led to the hypothesis that static dipole polarizabilities for each $2p_i$ level should be included in the model potential from [6] for collisions between the Ne*($2p_i$) and He atoms. Indeed, when the dipole polarizabilities are added to the long-range potentials of the Ne*($2p^5 3p$)-He system reported in [3], an excellent agreement between theory and experiment is found for the disalignment rate coefficients of the Ne*($2p_2$) and Ne*($2p_7$) atoms induced by collisions with helium.

Figure 6 shows our theoretical calculations for the $2p_5$ level. The dotted line represents calculations based on potentials from [3], while the solid line shows calculations based on an improved model potential which includes an additional static dipole polarizability of the Ne*($2p_5$) atom to the model potential from [3] at internuclear distances larger than $14a_0$, where a_0 is the Bohr radius. The theoretical rate coefficients calculated with the improved model potential are much closer to the experimental data than in the case when the potentials from [3] are used. The difference is now less than 30 % at 77 and 292 K, which is smaller than in the calculations based on the potentials from [3]. In the latter case, the difference reaches 90% at 77K and 60% at 292K. Thus, the inclusion of a static atomic dipole polarizability potential in our quantum calculations decreases significantly the value of the disalignment rate coefficient for the $2p_5$ level; however, this tendency is opposite to the $2p_2$ case shown in Fig. 10 of Ref. [7]. Our experimental results for the $2p_2$ and $2p_5$ levels confirm this opposite tendency.

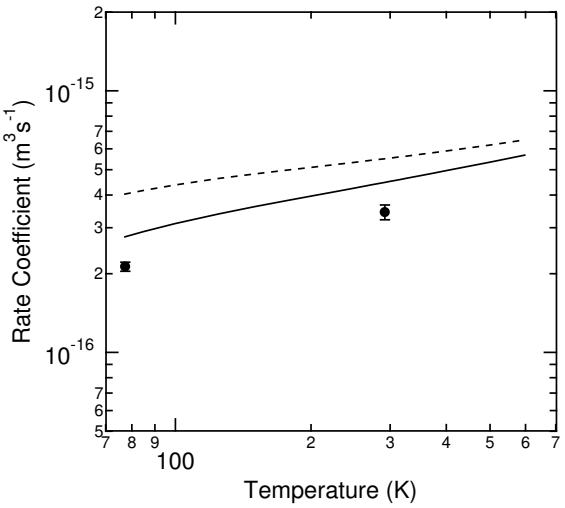

Fig.6 Comparison between our experimental values of disalignment rate coefficient for the Ne*($2p_5$) atoms induced by collisions with helium atoms (closed circles), and quantum calculations based on (1) the model potential for the Ne*($2p^5 3p$)-He system from [3,6] (dashed line), and (2) a modified model potential which includes a static dipole polarizability term for the Ne*($2p_5$) atom added to the potentials from [3] at internuclear distances larger than 14 a_0 (solid line).

4. Conclusion

Using the polarization resolved LIFS with the temperature controlled discharge cell we have determined the disalignment rate coefficient of excited neon atoms on the $2p_5$ level of the $2p^5 3p$ configuration at 77 and 292 K. We found that quantum multi-channels close-coupling calculations based on a modified model potential for the Ne*($2p^5 3p$)-He system, which incorporates static dipole polarizabilities for the neon excited on $2p_i$ levels added to the model potential of Hennecart and Masnou-Seeuws [6], show a good agreement with our present experimental results for the $2p_5$ level. The conclusion of the present study is consistent with the results from [7], where an excellent agreement between quantum calculations and measurements of disalignment of Ne*($2p_2$) atoms induced by collisions with He in a discharge cell at temperatures

between 77 and 600 K was reported.

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