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Magnetic-field dependence of the collisional disalignment of neon $2p^53p$-configuration atoms

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Laser-induced-fluorescence spectroscopy has been applied to neon and helium-neon discharge plasmas in magnetic fields lower than 10 T. Disalignment-rate coefficients of the neon $2p_2$ and $2p_7$ atoms ($2p^53p$ configuration, Paschen notation) have been determined for neon and helium collisions in a field strength that is lower than the critical field at which the collision time is equal to the inverse Larmor frequency. The $2p_7$ atoms have rate coefficients independent of the magnetic-field strength, whereas the $2p_2$ atoms have a field-dependent rate coefficient for neon collisions.

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I. INTRODUCTION

Effects of a magnetic field on atomic collisions have attracted the interest of researchers. In particular, theoretical and experimental investigations have been focused on the collisional excitation-transfer processes between the magnetic substates of excited atoms. Gay and Schneider [1] presented a theory based on the impact-parameter method. They treat the cases in which the interaction is an electrostatic long-range force; the first of these cases is the resonance collisions between atoms of the same species, and the second is the collisions between atoms of different species in which the van der Waals force is responsible. In both the cases, the effect of a magnetic field on collisions is appreciable only when the condition $\omega T_c > 1$ is satisfied, where ω is the Larmor frequency and T_c is the collision time defined by $[\sigma/(\pi v^2)]^{1/2}$. (v is the relative atom velocity and σ is the collision cross section.)

Experimental studies corresponding to the above theory have been made mainly in the region of $\omega T_c > 1$. An experiment on the resonance collisions is reported for $\mathrm{Hg}(6\,^3P_1)+\mathrm{Hg}(6\,^3S_0)$ and $\mathrm{Na}(3\,^2P)+\mathrm{Na}(3\,^2S)$ pairs [2,3], and the observed decrease in the excitation-transfer rates between the magnetic substates with an increase in the field strength (B) is well reproduced by the theory. The foreign gas collision has been studied on $\mathrm{Hg}(6\,^3P_1)$ atoms with the collision partner of rare-gas atoms or several species of molecules [4,5]. The agreement between the experimental results and the theoretical predictions is rather poor: only the observed B dependence of $g^{(11)(10)}/g^{(1-1)(10)}$ for xenon and krypton perturbers is approximately reproduced by the theory, where $g^{(11)(10)}$ stands for the excitation-transfer rate from the $J=1, m_J=0$ state to the $J=1, m_J=1$ state. For other cases, the experimental results are not consistent with theoretical predictions. It is suggested that interactions other than the van der Waals force, e.g., a repulsive force, may be dominant in these collisions. It is noted that the criterion $\omega T_c \gtrsim 1$ is still valid in these cases.

In the region of $\omega T_c \ll 1$, Hirabayashi, Okuda, and Fujimoto [6] have found a strong magnetic-field dependence of the excitation-transfer rates for $\mathrm{He}(3\,^1P$ or $^1D)+\mathrm{He}(1\,^1P)\rightarrow[\mathrm{He}(3\,^3D)+\mathrm{He}(1\,^1S)]$; these excitation-transfer processes apparently violate Wigner's spin conservation rule. Recently, Matsumoto, Shiozawa, and Fujimoto [7] have found that, up to $B=10$ T, these magnetic-field-dependent rate coefficients follow very closely the field-dependent degree of mixing of the spin wave functions of the $3\,^1D$ and $3\,^3D$ levels. This fact suggests that this magnetic-field effect is not caused by collision dynamics but by the wave functions. Therefore, these collision processes are not regarded as violating the criterion $\omega T_c \gtrsim 1$.

There have been reports that, for the $^2P_{1/2}$ resonance state atoms of alkaline metals, the disorientation cross section, which corresponds to $g^{(1/2\mp1/2)(1/2\pm1/2)}$, increases with an increase in the magnetic-field strength less than 1 T [8–10]. An explanation is proposed that is based on the admixture of the $^2P_{3/2}$ wave function into the $^2P_{1/2}$ wave functions due to the magnetic field [11]. However, there appears to be a confusion concerning this phenomenon; i.e., no appreciable increase in the cross section has been observed for the case of potassium $4\,^2P$ atoms in collisions with helium or neon for the magnetic-field strength between 2.2 and 13.1 T [12].

It may be noted that all the above experiments except for those of Refs. 6 and 7 are based on measurement of signal intensities for stationary optical excitation. On the contrary, observation of a time-dependent signal following a pulsed excitation is simple and straightforward in its principle and interpretation, and this alternative method is expected to give less ambiguous results.

In the following, we report our time-dependent laser-induced-fluorescence spectroscopy on collisional disalignment of excited neon atoms, a kind of excitation transfer between the magnetic sublevels, and show the first obvious violation of the above criterion.

II. EXPERIMENT AND RESULTS

The experimental setup was basically the same as that described in Ref. 6 with several modifications. The main body of experimental data was taken by using a superconducting magnet (Oxford instruments, S10-76.5 (60RT)/-13) which was capable of producing magnetic fields up to 10 T at a current of 100.1 A. The bore diameter of the solenoid was 74 mm and the height was 320 mm. The field inhomogeneity was claimed to be less than 0.1% over the 10-mm-diam sphere. The field strength at the center of the solenoid bore was calibrated with the Gauss meter (F. W. Bell 610) against the voltage drop of the current through a shunt resistor (0.1998 T/mV). This magnet was located in a cryostat that had a cylindrical-shape free space of 60 mm diam. The inside wall of the bore was painted black so as to avoid reflection of light. A discharge tube with a 10-mm inner diameter made from pyrex glass was placed on the axis of the solenoid. The basis pressure was 6×10^{-7} Torr, and 1–7 Torr neon was introduced for an experiment of neon collisions. A mixture gas of neon and helium was used for a helium collision experiment. A dc mild glow discharge was produced with a current of 0.8–2.2 mA. By careful alignment of the discharge tube axis along the magnetic field, we obtained a stable discharge. A nitrogen-laser pumped dye laser produced a laser pulse of 5-ns duration and a repetition rate of 26 pps. This laser light was transmitted through an optical fiber into the free space of the cryostat, reflected by a prism, focused by a lens, which illuminated the plasma from the direction perpendicular to the magnetic field.

Some of the neon $1s_3$ (Paschen notation, $J=0$) metastable atoms in the discharge plasma were excited by the 616.4-nm light to the $2p_2$ ($^3P_1$) level, where we observed the fluorescence light of the 659.8 nm [$1s_2$ $(J=1)-2p_2$] line. For observation of the $2p_7$ ($^3D_1$) atoms, we used the 653.2-nm ($1s_3-2p_7$) light excitation and observed the 638.3-nm [$1s_4$ $(J=1)-2p_7$] line. For both cases, therefore, we used the $(J=0)\rightarrow(J=1)\rightarrow(J=1)$ excitation-observation scheme. Under a magnetic field, the lines for the laser excitation were Zeeman split, and for $B>2$ T we excited only the central π component, producing a population only in the $m_J=0$ magnetic substate of the upper level.

The fluorescence light emanating from the plasma at right angles to the directions of the magnetic field and the laser beam was reflected by two mirrors and focused by a lens onto the entrance slit of the monochromator. The arrangement of the mirrors was such that no elliptical polarization was created by the reflections by these mirrors. The monochromator (Nikon G250) had a focal length of 250 nm and $f/5$. The linear reciprocal dispersion was 3.4 nm/mm. To observe all the components of the Zeeman-split emission line, the entrance and exit slit widths were 1 and 2 mm, respectively, and the height was 10 mm. The observed spectral range was so wide that other spectral lines were included. However, since we observed only the effect of the laser excitation, and the excitation transfer by atomic collisions to other $2p_j$ levels is slow [13], these spurious emission lines did not affect the observed signal. The photomultiplier was Hamamatsu R928 with a special potential distribution in the dynode chain so as to draw a high current [14], and a gating of 4 μs was applied to the first and third dynodes. The tem-

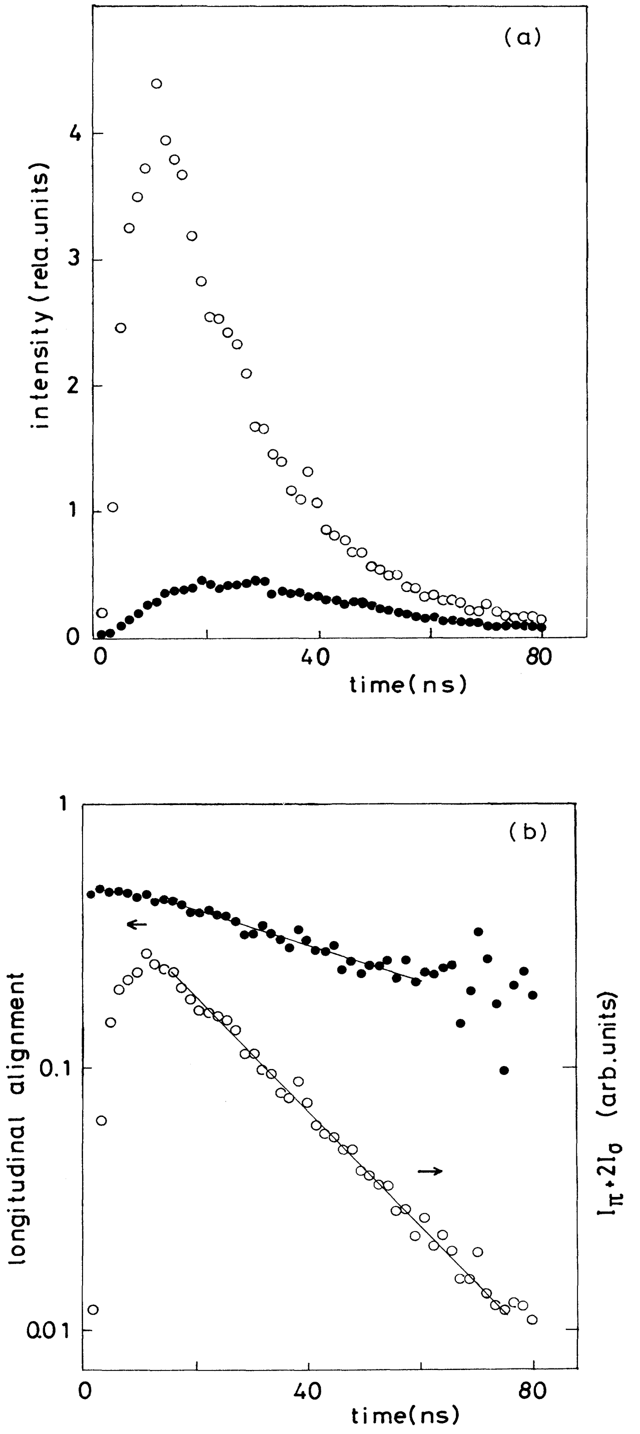

FIG. 1. (a) The temporal development of the π component (●) and the σ component (○) of the fluorescence light λ659.8 nm (Ne, $1s_2-2p_2$) following the laser excitation of λ616.4 nm (Ne, $1s_3-2p_2$). The magnetic field is 7 T, the pressure of pure neon gas is 2.1 Torr, and the discharge current is 1.6 mA. (b) The temporal development of the upper-level population (○) and the longitudinal alignment (●) of the fluorescence light (sign reversed).

poral development of the output current was sampled with a 1.6-ns time resolution, averaged and processed by a boxcar averager system (EG&*G* PAR Model 4402, 4420, and 4421).

By placing a polarizer for photographic cameras in front of the entrance slit of the monochromator, we observed the π and σ components of the fluorescence light separately. The relative sensitivity of our detection system to these components was calibrated by the following method: we removed the cryostat and applied to our discharge plasma a weak magnetic field about 40 G in the direction of observation. Due to the Larmor precession of the dipole, which might be created by the plasma itself (i.e., the self-alignment [15]), the continuous emission radiation was expected to be unpolarized (Hanle effect). We measured and compared the output current from the photomultiplier for the π and σ components.

An example of the observed temporal developments of the intensities of the polarized components of the fluorescence light is shown in Fig. 1(a). This is for $2p_2$ and neon collisions. From the intensities of the π and σ components, I_π and I_σ, respectively, we calculate the relative total population of the upper level $(I_\pi+2I_\sigma)$ and the lon-

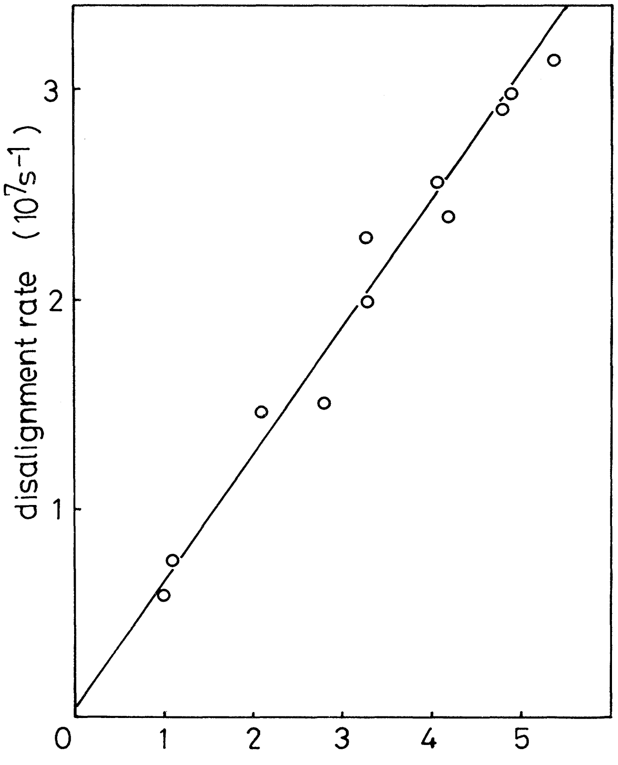

FIG. 2. The pressure dependence of the disalignment rate of the $2p_2$ atoms due to collisions with neon atoms. The solid line is the result of the least-squares fit to the experimental data. The magnetic field is 7 T.

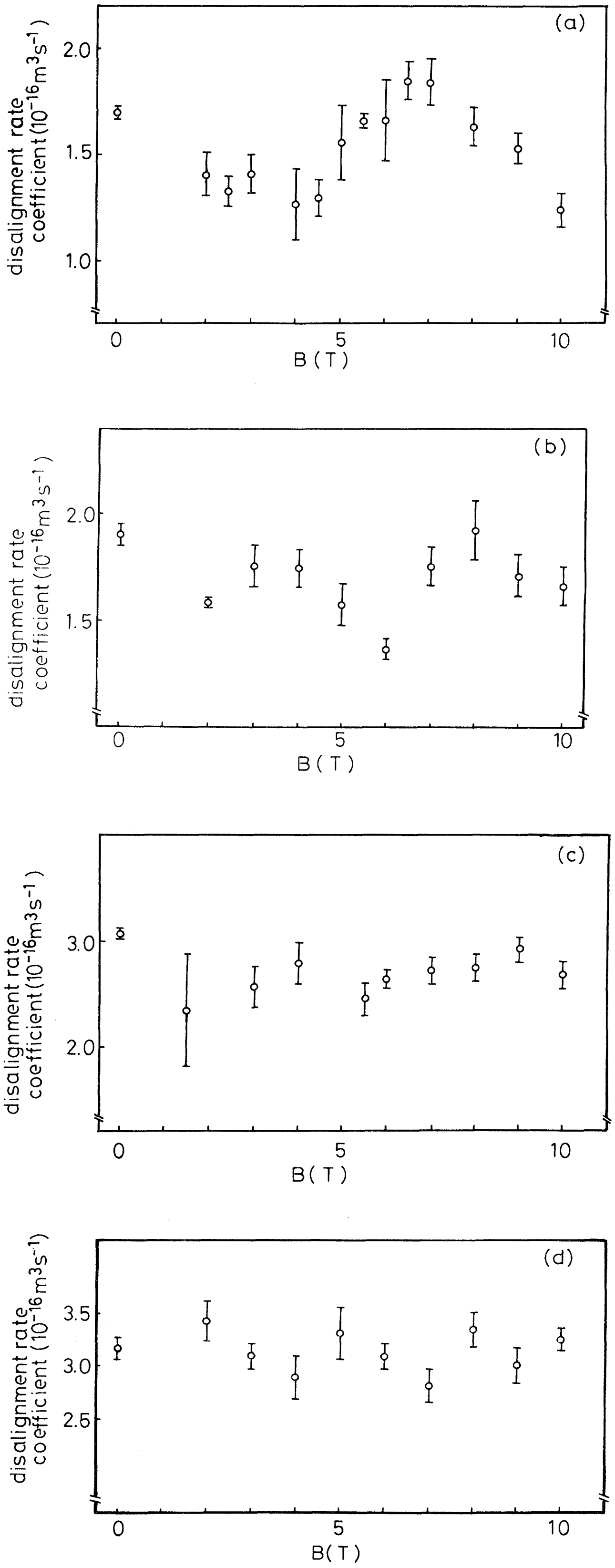

FIG. 3. The magnetic-field dependence of the disalignment-rate coefficient due to atomic collisions (a) $Ne(2p_2)+Ne$; (b) $Ne(2p_2)+He$; (c) $Ne(2p_7)+Ne$; (d) $Ne(2p_7)+He$.

gitudinal alignment $\alpha=(I_\pi-I_\sigma)/(I_\pi+2I_\sigma)$. These quantities are shown in Fig. 1(b). (The sign of α has been reversed.) From the slope of the temporal decay of the longitudinal alignment, we determine the disalignment rate. By changing the filling gas pressure and applying a similar procedure, we obtained the pressure dependence of the disalignment rate. Figure 2 shows an example. From the slope of the least-squares-fitted line, we determine the disalignment-rate coefficient for atomic collisions. (The small disalignment rate at zero pressure will be discussed later.) By repeating the above procedure for various magnetic fields, we obtained the field dependence of the disalignment-rate coefficient. The result is shown in Fig. 3(a). This whole procedure is repeated for another level $2p_7$ [Fig. 3(c)]. In the experiment of the helium collisions, a helium gas of variable pressure was added to a neon gas of 1 Torr, and the dependence of the disalignment rate on the helium pressure was determined. The result of the disalignment-rate coefficient against the magnetic-field strength is shown in Figs. 3(b) and 3(d).

The disalignment-rate coefficient in the field free case was determined by following another procedure as described in Ref. [16]. We used another discharge tube, and applied the π-polarized light excitation. We observed the π and σ components.

Figure 3 also includes the experimental data taken by using a smaller magnet, which was capable of producing magnetic fields lower than 5.7 T. The experimental setup was almost the same except that the fluorescence light was transmitted through a bundle of optical fibers, which almost completely destroyed the polarization. We compared the results by these two different configurations and found very good agreement between them.

III. DISCUSSION

As is seen in Fig. 2 of the pressure dependence of the disalignment rate, the best fitted line tends to a finite value at zero pressure. This finite disalignment is ascribed to the effect of radiation trapping, or the opacity effect. Each of the $2p_2$ and $2p_7$ levels has four emission lines, two of which terminate on the metastable levels, $1s_3$ ($J=0$) and $1s_5$ ($J=2$). The populations of these levels are expected to be rather high, and the effect of radiation trapping may be appreciable for these lines. In the field free case, disalignment by trapped radiation is observed and explained quantitatively (see Fig. 1 of Ref. [17]). Under a magnetic field, the line terminating on the $1s_3$ level is split into three components, and reabsorption of any of these three components does not cause disalignment. Another line terminating on the $1s_5$ level has a complicated Zeeman pattern, and its reabsorption, even if it were slight, may cause disalignment.

Because of the limitation of the space, we could not measure the lower-level populations in the plasma under a magnetic field, and we could not estimate quantitatively the disalignment rate by the trapped radiation. What we did instead was as follows: We tried to minimize this effect by keeping the population of the low-lying levels as low as possible. This resulted in a low fluorescence intensity that was approximately proportional to the $1s_3$ population, and this was the reason why we used the wide slit widths of the monochromator. During a series of experiments for a particular magnetic-field strength, we adjusted the discharge current so that we had an almost constant peak value of the fluorescence intensity, or a constant population of the $1s_3$ level before the laser excitation. The population of the $1s_5$ level is expected to be approximately proportional to the $1s_3$ population (see Fig. 3 of Ref. [14]). Thus we expect that the degree of radiation trapping is almost constant over this series of observations and so is the disalignment rate by the trapped radiation. This assumption is supported by the fact that the observed apparent depopulation rate, which is more sensitive to the effect of radiation trapping, showed almost constant value or a smooth increase over the pressure (see Fig. 8 of Ref. [14]). We therefore conclude that our result of the collisional disalignment determined from the experimental points in Fig. 3 is free from this effect.

Inhomogeneity of the magnetic field may cause reabsorption of radiation at other parts of the discharge tube. The volume that we observed was a 1-mm thick disk of the cross section of the discharge tube. Thus it is concluded that our result is free from this effect.

An electric field may cause precession of the dipole, and thus could affect the present measurement of disalignment. It is estimated that the static electric field to sustain the discharge is about 1 kV/m, the microfield by the plasma ions (the Holtzmark field) is about 0.5 kV/m, and the motional electric field is less than 5 kV/m. The temporal evolution of the produced alignment due to these electric fields is estimated from Ref. [18], and the "decay time" is found to be about 10 μs for the $2p_2$ atoms. Thus the effect of the electric fields is neglected.

The electron density is estimated [19] to be less than 10^{16} m^{-3} under the present experimental conditions. The disalignment rate due to electron collisions is less than 4×10^3 s^{-1} for the $2p_2$ atoms [18], and that for the $2p_7$ atoms it is estimated from this value and the Stark broadening parameters for the spectral lines [18] to be of a similar magnitude.

The critical-field strength for $\omega T_c=1$ is calculated from Fig. 3 for the neon (helium) collisions; it is 35 T (80 T) for $2p_2$ and 55 T (125 T) for $2p_7$. In this estimate, we have used the excitation-transfer cross section between the $m_J=0$ and $m_J=\pm1$ magnetic substates, which is one-third of the disalignment cross section. Figures 3(c) and 3(d) show that the $2p_7$ atoms have the field-independent rate coefficients, as expected from the criterion. The $2p_2$ atoms, on the other hand, behave differently; Fig. 3(b) appears to suggest a slight magnetic-field dependence, but not conclusive, and Fig. 3(a) shows a clear field-dependent rate coefficient.

At the present moment, we do not have any explanation to the above different field dependences of the disalignment-rate coefficients. It may be interesting, however, to note the following fact: Carrington and Corney [20] measure the temperature dependence of the alignment destruction-rate coefficient for eight neon levels of the $2p^53p$ configuration, and compare the result with the calculation based on the van der Waals interaction. It is found that all the levels but two, $2p_2$ and $2p_6$, follow the calculated temperature dependence of the disalignment-rate coefficient of $T^{0.3}$ for both the neon and helium collisions. These exceptional "ill-behaved" levels show a large positive temperature dependence. The present anomalous magnetic-field dependence of the disalignment cross section of one of these levels may have some relation to its anomalous temperature dependence.

Recently, Hennecart and Masnou-Seeuws [21] have published the model potential calculations for the present pairs of atoms. According to their results, the repulsive potential is much larger than the attractive van der Waals potential; e.g., for $2p_2$ the molecular potential is of the order of 100 cm^{-1} at a nuclear distance of 10 a.u., whereas the van der Waals potential is of the order of -1 cm^{-1}. It would therefore be natural to expect that the disalignment collision is governed by the former potential, and that the magnetic-field dependence of its rate coefficient as well as its temperature dependence is different from those for the van der Waals interaction. This situation concerning the interaction potentials is, however, much the same for the well-mannered level $2p_7$. Therefore, these different magnetic-field dependences and the temperature dependences would be a result of complicated interplay of various factors in collision dynamics. It is hoped that the present experimental result would be interpreted in terms of a theoretical calculation based on the molecular potentials.

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