Observation of Collisional Transfer from Alignment to Orientation of Atoms Excited by a Single-Mode Laser

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The first experimental evidence of transfer from alignment to orientation of laser-excited atoms due to anisotropic collisions is reported. The experiment has been made with respect to neon atoms in the $2p_4$ state, excited by a single-mode laser, which collide with the ground-state neon atoms, and good agreement between experiment and theory is found.

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In this Letter, we report on the first experimental observation of collisional transfer from alignment (electric quadrupole moment) to orientation (magnetic dipole moment) of atoms excited by a single-mode laser, which has been theoretically predicted in our previous paper.[1] In the experiment, we excited neon atoms in a discharge from the $1s_5$ metastable state to the $2p_4$ state by a single-mode dye laser, and monitored the alignment and orientation created in the $2p_4$ state by detecting the linearly and circularly polarized components of fluorescence originating from the $2p_4$ state.

The transfer between alignment and orientation, as well as the relaxation of these multipole components, is due to the collisions of the laser-excited atoms with the ground-state neon atoms. The transfer between alignment and orientation is caused by anisotropic collisions which are due to the anisotropic velocity distribution of atoms excited by a single-mode laser.

Under such an anisotropic situation, if the quantization axis is taken along the direction of the laser propagation, the relaxation and transfer of multipole components of excited-state atoms can generally be expressed as[1]

$$d\rho_q^k/dt = -\sum_{k'} \Gamma_q^{kk'} \rho_q^{k'}, \qquad (1)$$

where ρ_q^k is the density-matrix element corresponding to the q component of the 2^k-pole moment, i.e., ρ_0^0 is the population, ρ_q^1 is the magnetic dipole moment or "orientation," ρ_q^2 is the electric quadrupole moment or "alignment," and so on. In Eq. (1), the transfer between different multipole components is represented by the off-diagonal element $\Gamma_q^{kk'}$ with $k \neq k'$, which does not appear in the conventional theory[2] of isotropic collisions. As described in Ref. 1, the effect of the transfer from alignment to orientation is expected to appear in magnetic depolarization signals in optical-pumping experiments with use of a single-mode laser. When atoms are excited by linearly polarized light, the fluorescence emitted by initially aligned atoms must partially be polarized circularly because of the transfer from alignment to orientation caused by anisotropic collisions.

The experimental setup is schematically shown in Fig. 1. A linearly polarized single-mode light beam from a cw dye laser was applied into the positive column of a glow discharge tube filled with neon of natural abundance at the pressure of 1.3 Torr. To excite atoms velocity selectively from the metastable state $1s_5$ to the $2p_4$ state, the wavelength of the dye laser was locked to 594.5 nm by means of a neon Faraday filter.[3] The static magnetic field $\vec{H}$ was applied along the x direction and swept from -19.3 to $+19.3$ G through zero by a pair of Helmholtz coils. Earth's

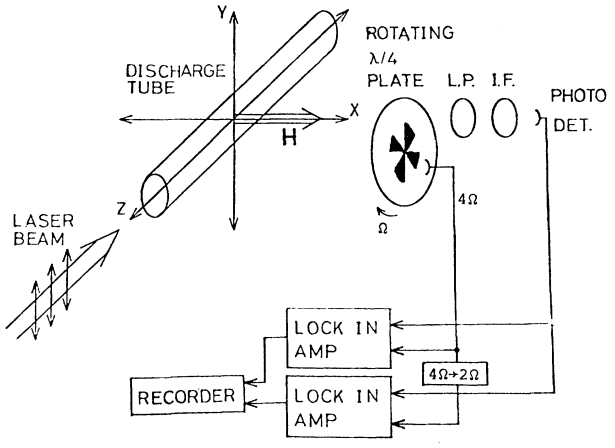

FIG. 1. Schematical experimental setup for simultaneous observations of the transfer from alignment to orientation and the ordinary Hanle signal. The fluorescence at 609.6 nm is selectively detected by passing through an interference filter (I.F.).

magnetic field and other stray fields were compensated by three pairs of Helmholtz coils. The incident laser beam linearly polarized along the y axis was propagated along the z direction. The position of the discharge tube was arranged so that the portion just behind the entrance of the positive column was placed at the center of the Helmholtz coils. We observed fluorescence emitted from this portion to the x direction. In order to avoid the instrumental scattering, we detected the fluorescence at 609.6 nm, which is due to the transition from the $2p_4$ state to the $1s_4$ state.

As discussed in Ref. 1, the intensity difference $I_{\sigma+} - I_{\sigma-}$ between the circularly polarized components σ_+ and σ_- of the fluorescence originates in the orientation transferred from the alignment by anisotropic collisions, and the intensity difference $I_{\parallel} - I_{\perp}$ between the components polarized linearly along the y and z axes originates in the alignment itself. To detect simultaneously both the orientation and alignment signals, $I_{\sigma+} - I_{\sigma-}$ and $I_{\parallel} - I_{\perp}$, the fluorescence was passed through a rotating quarter-wave plate and then through a linear polarizer, as shown in Fig. 1. When the angular frequency of rotation of the quarter-wave plate is Ω, the orientation signal $I_{\sigma+} - I_{\sigma-}$ is modulated at 2Ω while the alignment signal $I_{\parallel} - I_{\perp}$ (ordinary Hanle signal) is modulated at 4Ω. The orientation and alignment signals were obtained by applying the output of a solar cell to lockin amplifiers tuned at 2Ω and 4Ω, respectively.

In a cw dye laser with a Faraday filter inside the cavity, the laser frequency can be varied in the vicinity of the absorption line by changing the strength of the magnetic field applied to the Faraday filter. In this experiment, we used the Faraday filter of natural neon (see Ref. 3). As the magnetic field applied to the Faraday filter is increased, the laser starts to oscillate in a single mode at a frequency between the 609.6-nm lines of Ne^{20} and Ne^{22} and the frequency is shifted toward the higher-frequency side. The relative frequency shift from the starting frequency was measured by a scanning Fabry-Perot interferometer. The detuning of the starting frequency from the line center of Ne^{20} is expected to be 600 ± 100 MHz from our theoretical estimation.

Typical recorder traces of the orientation signal $I_{\sigma+} - I_{\sigma-}$ and the alignment signal $I_{\parallel} - I_{\perp}$ obtained simultaneously for the case that the detuning $\Delta\nu_{20}$ from the line center of Ne^{20} is 1500 MHz are shown in Fig. 2 as functions of the magnetic-field intensity. The upper trace ($I_{\sigma+} - I_{\sigma-}$) is magnified by forty times as compared with the lower trace ($I_{\parallel} - I_{\perp}$). We see in Fig. 2 that the orientation signal shows a dispersionlike dependence on the magnetic-field intensity, as expected from the theory. Similar recorder traces were obtained for the detuning $\Delta\nu_{20}$ of 960, 1140, 1320, 1500, and 1680 MHz. If the birefringence of the walls of the cylindrical cell is not small enough, the alignment signal might appear in the circu-

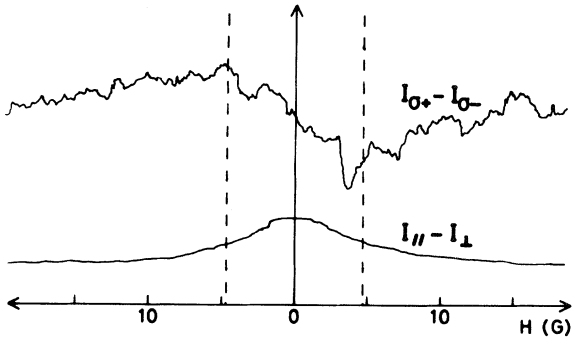

FIG. 2. Typical recorder traces showing the alignment signal $I_{\parallel} - I_{\perp}$ (lower trace) and the orientation signal $I_{\sigma+} - I_{\sigma-}$ (upper trace) as functions of the magnetic-field intensity. The orientation signal is magnified by forty times as compared with the alignment signal.

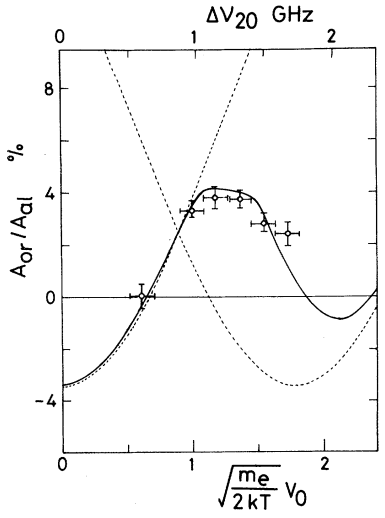

FIG. 3. Measured amplitude ratio of the orientation signal to the alignment signal as a function of the normalized velocity of the excited neon atoms, together with the theoretical values for the natural neon (solid curve) and those for isotopes Ne^{20} or Ne^{22} (dotted curves).

larly polarized component and might overlap the orientation signal. In order to check such a parasitic signal, we measured the birefringence of the walls and found that the conversion of the linear polarization to the circular polarization was less than 0.5%.

In order to compare with theoretical calculations, we measured the ratio of the amplitude of orientation signal to that of alignment signal as a function of the detuning $\Delta\nu_{20}$ and the results are shown in Fig. 3. The error in measurement of the detuning $\Delta\nu_{20}$ is mainly due to the ambiguity in estimating the starting frequency of the dye laser with a Faraday filter, as mentioned already.

Since we used neon gas of natural abundance in the experiment, we have to modify the theory given for the case of a pure isotope. For a pure isotope, the amplitude ratio A_{or}/A_{al} of orientation signal to alignment signal is given by the relation

$$\frac{A_{or}}{A_{al}}=\left(\frac{15}{7}\right)^{1/2}\frac{\Gamma}{\gamma_1^{\,1}}\left(\frac{4\gamma_1^{\,1}\gamma_0^{\,2}\gamma_2^{\,2}}{(\gamma_1^{\,1}\gamma_1^{\,2}+\Gamma^2)(3\gamma_2^{\,2}+\gamma_0^{\,2})}\right)^{1/2} \quad (2)$$

for the observed fluorescence [$2p_4(j=2)\rightarrow 1s_4(j=1)$], where $\gamma_q^{\,k}=\gamma_{nat}+\Gamma_q^{\,kk'}$ and $\Gamma=i\Gamma_{-1}^{\,21}=i\Gamma_{-1}^{\,12}=-i\Gamma_1^{\,21}=-i\Gamma_1^{\,12}$, γ_{nat} being the natural decay rate. In Eq. (2), we have neglected the transfer from orientation or alignment to higher-order multipole moments with $k\geq 3$. Since the relative abundance of Ne^{21} (0.26%) is small compared with those of Ne^{20} (90.92%) and Ne^{22} (8.82%), we will neglect the presence of Ne^{21} in the present discussions. Because of the isotope shift of 1.72 GHz between Ne^{20} and Ne^{22} for the $1s_5$-$2p_4$ transition, the axial velocity of Ne^{22} atoms excited by a single-mode laser is different from that of excited Ne^{20} atoms. This isotope shift corresponds to the difference of 1.78 in the normalized axial velocity $(m_e/2kT)^{1/2}v_0$ for $T=400\,^{\circ}$K. Therefore, the effect of anisotropic collisions on Ne^{22} atoms is not the same as that on Ne^{22} atoms excited by the light with the same frequency. Consequently, the relaxation matrix elements $\Gamma_q^{\,kk'}$ have different values for Ne^{20} and Ne^{22}. Thus, the presence of Ne^{22} modifies the expression for the ratio A_{or}/A_{al} as

$$\frac{A_{or}}{A_{al}}=\frac{A_{or}(y)\exp(-y^2)+\alpha A_{or}(y-1.78)\exp[-(y-1.78)^2]}{A_{al}(y)\exp(-y^2)+\alpha A_{al}(y-1.78)\exp[-(y-1.78)^2]}, \quad (3)$$

where α is the abundance ratio of Ne^{22} to Ne^{20}, y is the normalized axial velocity of excited Ne^{20} atoms, which is proportional to the detuning $\Delta\nu_{20}$, and $A_{or}(y)$ and $A_{al}(y)$ are the amplitudes of orientation and alignment signals for pure isotope as functions of the normalized axial velocity of this isotope. In the present case, we can estimate Eq. (3) by using the relaxation-matrix elements $\Gamma_q^{\,kk'}$ given in Fig. 6(b) of Ref. 1. In Fig. 3, we show the theoretical values of A_{or}/A_{al} for natural neon by a solid curve, together with those for pure Ne^{20} and Ne^{22} (the two dotted curves). In these theoretical calculations, we have used the values $\gamma_{nat}=5.9\times10^7$ rad/sec,[4] and $(2\pi/5)n_p B^{2/5}\times\langle v_p^{\,3/5}\rangle=6.8\times10^6$ rad/sec. The latter was estimated experimentally from the collisional broadening of the alignment signal. As seen in Fig. 3, when the laser is tuned around the absorption line center of Ne^{22}, the presence of Ne^{22} gives a significant influence upon the ratio A_{or}/A_{al} in spite of its small relative abundance. The reason is that the collisional transfer rate between orientation and alignment changes drastically with the change of laser detuning. In Fig. 3, we can find a fairly good agreement between theory and experiment, although the $2p_4$ state has the total angular momentum $j=2$, while the theoretical calculation of $\Gamma_q^{\,kk'}$ has been made for $j=1$. This might indicate that the dependence of the collisional relaxation matrix $\Gamma_q^{\,kk'}$ on the axial emitter velocity is not so affected by the value of the total angular momentum j.

We also measured the width of the alignment signal as a function of the detuning of the single-mode laser. But no significant change in the width was observed in the present region of detuning. This comes from the fact that the detuning-dependent part in the decay rate of alignment is only a small fraction of the total decay rate in the case of collisions between atoms with the same mass. In this case, the decay rate of alignment for atoms excited by a single-mode laser is approximately equal to that for atoms excited isotropically. Except for the case in which the perturber is much heavier than the emitter, the use of the theory of isotropic collisions in estimating the alignment-destroying cross section from the experiment with use of a single-mode laser may bring about no serious errors.

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New Observation of Parity Nonconservation in Atomic Thallium

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Refined observations of parity nonconservation in the $6^2P_{1/2} - 7^2P_{1/2}$ transition in $^{203,205}_{81}$Tl are reported. Absorption of circularly polarized 293-nm photons by $6^2P_{1/2}$ atoms in an E field results in polarization of the $7^2P_{1/2}$ state, through interference of the Stark $E1$ amplitude with $M1$ and parity-nonconserving $E1$ amplitudes. Detection of this polarization yields the circular dichroism $\delta = +(2.8 \pm {}^{1.0}_{0.9}) \times 10^{-3}$, which agrees with theoretical estimates based on the Weinberg-Salam model, for $\sin^2\theta_W = 0.23$.

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We report new observations of parity nonconservation (PNC) in the $6^2P_{1/2}$-$7^2P_{1/2}$ transition in atomic thallium (see Fig. 1). The transition amplitude is forbidden $M1$ with measured amplitude $\mathfrak{M} = (-2.1 \pm 0.3) \times 10^{-5} |e\hbar/2m_e c|$.[1] Parity nonconservation causes the $6^2P_{1/2}$ and $7^2P_{1/2}$ states to be admixed with $^2S_{1/2}$ states; thus the transition amplitude contains an additional $E1$ component $\mathcal{E}_p$. This results in circular dichroism, defined by

$$\delta = \frac{\sigma_+ - \sigma_-}{\sigma_+ + \sigma_-} = \frac{2\,\mathrm{Im}(\mathcal{E}_p \mathfrak{M}^*)}{|\mathfrak{M}|^2 + |\mathcal{E}_p|^2} \simeq \frac{2\,\mathrm{Im}\mathcal{E}_p}{\mathfrak{M}}, \quad (1)$$

where $\sigma_\pm$ are the cross sections for absorption of 293-nm photons, with $\pm$ helicity, respectively. Theoretical estimates of $\mathcal{E}_p$ based on the Weinberg-Salam (W-S) model[2] yield[3-5]

$$\delta_{\mathrm{theor}} = 2\,\mathrm{Im}(\mathcal{E}_{p,\mathrm{theor}})/\mathfrak{M}_{\mathrm{expt}} = (2.1 \pm 0.7) \times 10^{-3} \quad (2)$$

for $\sin^2\theta_W = 0.23$, where θ_W is the Weinberg angle.

The aim of this experiment is to measure δ. The dipole amplitudes $\mathcal{E}_p$ and $\mathfrak{M}$ are observed by their interference with a Stark $E1$ amplitude βE caused by a 215-V/cm electric field E, in the $6P_{1/2}, F = 0$–$7^2P_{1/2}, F = 1$ transition. This causes a polarization $\Delta = -(2\mathfrak{M}/\beta E)(1 \pm \frac{1}{2}\delta)$ in the $7^2P_{1/2}$, $F = 1$ state. The latter is analyzed by selective excitation of $m_F = +1$ or -1 substates to the $8^2S_{1/2}$ state with circularly polarized 2.18-μm light, followed by observation of $8^2S_{1/2}$-$6^2P_{3/2}$ fluorescence at 323 nm. (See Fig. 1.) In a preliminary version of the experiment[6] we obtained the result

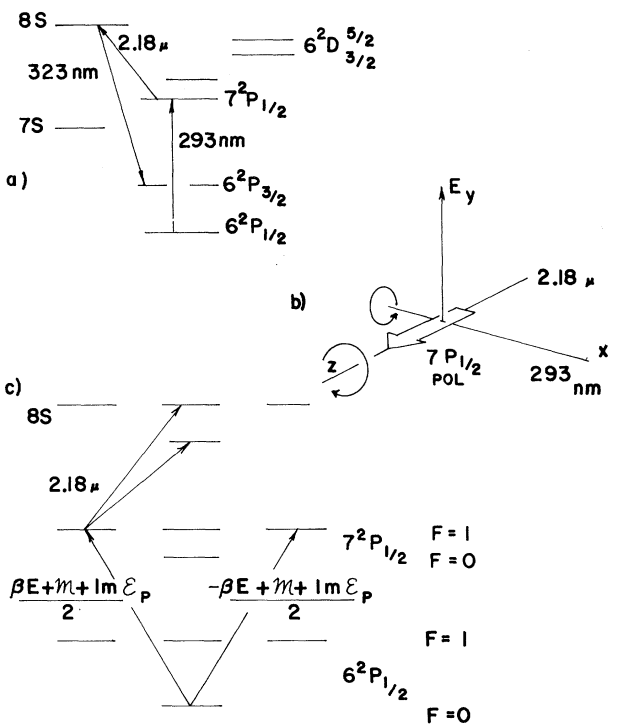

FIG. 1. (a) Low-lying energy levels of Tl (not to scale). (b) Coordinate system, orientation of photon beams, and electric field direction. (c) Schematic diagram indicating production and analysis of $7^2P_{1/2}$ polarization in the 0-1 transition. The transition amplitudes to the $m_F = \pm 1$ levels of $7^2P_{1/2}$ are indicated. The polarization is analyzed by circularly polarized 2.18-μm radiation ($7^2P_{1/2}$-$8^2S_{1/2}$ transition). The $8^2S_{1/2}$ hfs is not resolved.