PHYSICAL REVIEW A VOLUME 31, NUMBER 5 MAY 1985

Collisional redistribution of circularly polarized light in barium perturbed by argon

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(Received 3 December 1984)

We have measured the orientation of the Ba $6p\ ^1P$ level produced by collision-induced excitation from the ground state by circularly polarized light. The detuning dependence of the far-wing excited-state orientation can be interpreted in terms of reorientation of molecular orbitals which occur during the collision. Effects due to rotational coupling are seen to occur at large blue-wing detunings. We have also determined the collisional rate for destruction of orientation by measuring the pressure dependence of the excited-state orientation.

I. INTRODUCTION

The subject of collisional redistribution has received attention both experimentally[1–6] and theoretically[7–11] over the past few years. In a previous paper[4] we have reported results of a collisional redistribution experiment using linearly polarized light to produce aligned excited-state atoms. In that experiment both the excited-state population and alignment were monitored as a function of the excitation-laser frequency. By comparing the redistributed line shape with that of the alignment we were able to describe the qualitative form of the interatomic potentials. Thus redistribution experiments of this type are useful for obtaining information about adiabatic potentials. In addition, the alignment of the excited state has been shown[9–12] to contain information about the dynamics of the collision, especially the mixing at large internuclear separation. Although the interatomic potentials are not known for most of the systems studied to date, Julienne[13] has found that redistribution experimental results are sensitive to the form of the potentials (especially the long-range part of the potential) and therefore make good tests of theoretical predictions.

The topic of this paper is the collisional production of oriented excited states using circularly polarized light. The experiment is schematically illustrated in Fig. 1(a). A near-resonant laser (excitation laser) is incident upon an atom undergoing collisions with perturbers. A second laser (probe laser) is tuned to resonance with a transition from the excited state produced by collisions to a higher excited state. Fluorescence from this higher excited state is monitored. (Note: In contrast to earlier experiments,[3,4] for this work the excited-state population and polarization are probed rather than monitored in fluorescence. Optical-depth problems would preclude monitoring the weak fluorescence signals associated with the circularly polarized light in the forward direction.)

We have studied barium undergoing collisions with argon atoms. The Ba-Ar system has been studied in linear-polarization redistribution experiments[4] and the results presented here are intended to complement the previous results. The relevant barium states are shown in Fig. 1(b). The circularly polarized excitation laser is tuned near the 5535-Å resonance line. The $6p\ ^1P$ level is populated in the presence of collisions and, due to the circular polarization of the excitation laser, the excited state is oriented. The orientation of the excited state is probed with a laser tuned to the $8s\ ^1S$–$6p\ ^1P$ line at 6131 Å. The fluorescence of the $8s\ ^1S$ down to the $6p\ ^3P_1$ is observed and is directly proportional to the probed m_J-state population. The probe-laser helicity may be varied so that we can form the quantity P_c, the circular polarization,

$$-P_c=\frac{I_{\parallel}-I_{\perp}}{I_{\parallel}+I_{\perp}}\ , \tag{1}$$

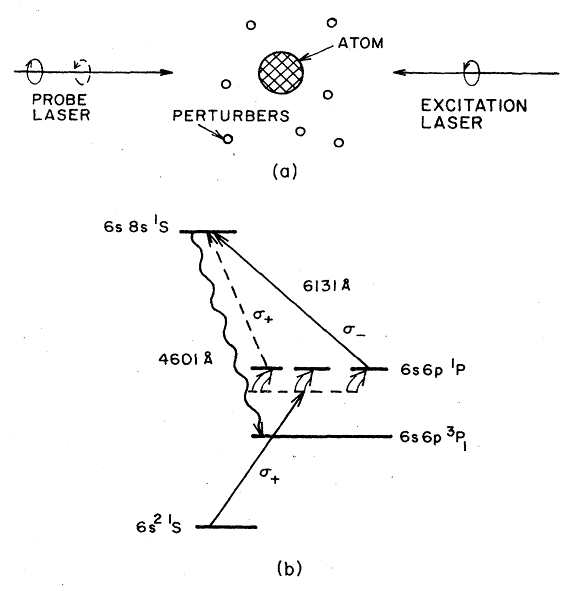

FIG. 1. (a) Schematic of experiment. Note that the probe laser propagates antiparallel to the excitation laser with a helicity parallel or antiparallel to the excitation laser. (b) Relevant Ba I energy levels. The $6s\,8s\ ^1S$ level is given as $6p^2\ ^1S$ by Moore (Ref. 21) but has been shown (Ref. 22) to actually be $6s8s$. The state in question has an energy of 34 371 cm^{-1}. Double arrows denote the collisional excitation of the $6s6p\ ^1P$ level.

where $I_{\parallel}$ ($I_{\perp}$) is the $8s\,^2S$—$6p\,^3P_1$ fluorescence intensity when probing with a helicity parallel (antiparallel) to the excitation-laser helicity. By parallel (antiparallel) we mean the sense of rotation of the electric field is the same (opposite) to the excitation laser. The overall minus sign in Eq. (1) is due to the fact that if we preferentially excite the $^1P(m_J=+1)$ state with σ_+ polarization, then this state is probed with σ_- polarization, not σ_+.

II. THEORY

When the detuning of the excitation laser $\Delta\omega=\omega_L-\omega_0$ is small compared to the inverse duration of a collision time τ_c^{-1}, we are in the impact region of the line shape. The impact region of the line shape can be described in terms of rates which depend on the average effects of collisions.[14] Using the general redistribution expressions of Burnett and Cooper,[8] one can show that for $\Delta\omega \ll \tau_c^{-1}$ the circular polarization, in the absence of depolarizing effects such as subsequent collisions, is given by

$$P_c(\Delta\omega \ll \tau_c^{-1})=\frac{6-3\gamma_c^{(1)}/\gamma_c}{6-\gamma_c^{(2)}/\gamma_c} . \tag{2}$$

Here γ_c is the impact-broadening rate and $\gamma_c^{(1)}$ ($\gamma_c^{(2)}$) is the collisional rate of destruction of orientation (alignment). In this limit the rate at which excited atoms are produced is proportional to the impact-broadening rate. The effects of subsequent collisions which occur during the lifetime of the excited state on the excited-state orientation (alignment) are determined by $\gamma_c^{(1)}$ ($\gamma_c^{(2)}$).

For detunings such that $\Delta\omega \gg \tau_c^{-1}$, we are in the quasistatic region of the line profile. Excitation occurs during the collision at the instant(s) when the transient molecule is in resonance with the incident light (Franck-Condon principle). Redistribution theories for such detunings must describe the excitation to molecular states at small internuclear separations followed by the evolution of the system into well-separated atoms. Using the results of Burnett and Cooper[8] one can show that the circular polarization is given by

$$P_c(\Delta\omega)=\frac{3\alpha^{(1)}(\Delta\omega)\gamma_N/(\gamma_N+\gamma_c^{(1)})}{2+\alpha^{(2)}(\Delta\omega)\gamma_N/(\gamma_N+\gamma_c^{(2)})} , \tag{3}$$

where $\alpha^{(1)}(\Delta\omega)$ and $\alpha^{(2)}(\Delta\omega)$ are, in general, complicated expressions describing the evolution of the system through the "half-collision" (from molecular excitation to well-separated atoms)[12] and γ_N is the radiative lifetime of the excited state. For large detunings, $\Delta\omega \gg \tau_c^{-1}$, one can derive rather simple expressions for $\alpha^{(1)}(\Delta\omega)$ and $\alpha^{(2)}(\Delta\omega)$ by assuming that after excitation the molecular orbitals rotate so as to follow the internuclear axis until a "decoupling radius" is reached. At that point the orbital decouples from the internuclear axis and becomes fixed in space. This "reorientation model" was used to interpret the results of the linear-polarization experiment.[4] The results of the model are

$$\alpha^{(1)}(\Delta\omega \gg \tau_c^{-1})=\frac{\langle \cos\Omega_\Pi \rangle}{2+g(\Delta\omega)} , \tag{4}$$

$$\alpha^{(2)}(\Delta\omega \gg \tau_c^{-1})=\frac{\frac{1}{6}+\frac{1}{10}\langle 2\cos\Omega_\Pi+\cos(2\Omega_\Pi)\rangle+g(\Delta\omega)[\frac{1}{30}+\frac{1}{10}\langle\cos(2\Omega_\Sigma)\rangle]}{\frac{2}{3}+\frac{1}{3}g(\Delta\omega)} , \tag{5}$$

where $g(\Delta\omega)$ is the ratio of excitation probabilities for the $^1\Sigma$ state to that for the $^1\Pi$ state, and $\langle \cdots \rangle$ denotes an average over collisions.[11,12] Note that for Ba-Ar the excited molecular state can be $^1\Sigma$ or $^1\Pi$. The quantity Ω_Π (Ω_Σ) is the angle through which the molecular orbital is rotated after $^1\Pi$ ($^1\Sigma$) excitation to the point at which "decoupling" occurs. In certain regions of the Ba-Ar line shape excitation occurs to only one molecular state. In this case Eqs. (3)—(5) yield

$$P_c(\Delta\omega \gg \tau_c^{-1},\,^1\Sigma)=0 , \tag{6}$$

$$P_c(\Delta\omega \gg \tau_c^{-1},\,^1\Pi)=\frac{\langle 5\cos\Omega_\Pi\rangle}{\langle 7+\cos\Omega_\Pi+\cos^2\Omega_\Pi\rangle} . \tag{7}$$

The zero result for $^1\Sigma$ excitation can be explained as follows: A $^1\Sigma$ state is a "dumbbell" orbital oriented along the internuclear axis with an angular-momentum projection of zero onto the internuclear axis. The angular-momentum projection being zero means that a $^1\Sigma$ state cannot retain any information about the helicity (± 1) of the excitation.

It is often the case in line broadening and redistribution that for moderate detunings, $\tau_c^{-1} \ll \Delta\omega \ll kT$ (kT =thermal energy), one can assume straight-line trajectories (SLT) for collision partners. Assuming SLT, one late the average rotation angle of the molecular orbital to the excitation radius R_c (Condon point) and the decoupling radius $R_{\rm dec}$. This yields

$$P_c(\Delta\omega \gg \tau_c^{-1},\text{SLT},\,^1\Pi)=\frac{25X}{55+5X+3X^2} , \tag{8}$$

where

$$X=\frac{R_c}{R_{\rm dec}} . \tag{9}$$

It is interesting to note that X can be related to the circular and linear polarizations for the case of SLT:

$$\frac{R_c}{R_{\rm dec}}=\frac{6P_c}{3-P_l} , \tag{10}$$

where P_l is given by the right-hand side of Eq. (1) with $I_{\parallel}$ ($I_{\perp}$) referring to the probe laser being parallel (perpendicular) to the incident linear polarization. In Ref. 4 an expression analogous to Eq. (8) is given for the linear polarization. Thus the circular polarization can be predicted from the linear polarization assuming SLT.

The polarizations discussed in Eqs. (2), (7), and (8) assumed no depolarizing effects to occur after excitation. From Eq. (3), however, we know depolarizing effects due to subsequent collisions can occur. Thus the data taken at finite argon pressure needs to be corrected by extrapolating to "zero pressure." Assuming γ_N, $\gamma_c^{(1)}$, and $\gamma_c^{(2)}$ are known, this is a straightforward extrapolation. For Ba-Ar, $\gamma_N = 1.19\times10^8$ and $\gamma_c^{(2)} = (1.30\pm0.08)\times10^{-9}$ $\text{sec}^{-1}/\text{cm}^{-3}$ by previous measurements.[4] Since $\gamma_c^{(1)}$ is not known, we have to determine the value experimentally. Using Eq. (14) of Ref. 4 and Eq. (3) one can show

$$\frac{3-P_l(M)}{P_c(M)} = \frac{1}{\alpha^{(1)}(\Delta\omega)}\left[1+\frac{1}{\gamma_N}\left[\frac{\gamma_c^{(1)}}{N_{\text{Ar}}}\right]N_{\text{Ar}}\right] , \tag{11}$$

where $P_l(M)$ and $P_c(M)$ refer to measured linear and circular polarizations.[15] This predicts that a plot of $[3-P_l(M)]/2P_c(M)$ versus argon pressure (for a fixed detuning) will yield a straight line with a pressure intercept independent of detuning which will yield $\gamma_c^{(1)}/N_{\text{Ar}}$. Figure 2 shows our experimental results for Ba-Ar (to be discussed below).

In addition to depolarizing collisions, there is also the depolarizing effects due to the odd isotopes of barium. 82% of naturally abundant barium have zero nuclear spin, while 18% have a nuclear spin of $\frac{3}{2}\hbar$ (^{135}Ba, ^{137}Ba). The size of the hyperfine interaction is such (see Baird *et al.*[16]) that the hyperfine structure has time to develop almost fully before decay ($\Delta\omega_{\text{hfs}}\tau_N \gg 1$), while, on the other hand, it does not play a role during the collision ($\Delta\omega_{\text{hfs}}\tau_c \ll 1$). One can calculate the correction using Eq. (61) of Ref. 10 and one finds

$$\alpha_M^{(1)}(\Delta\omega) = \alpha^{(1)}(\Delta\omega)(1-\tfrac{26}{45}q) , \tag{12}$$

$$\alpha_M^{(2)}(\Delta\omega) = \alpha^{(2)}(\Delta\omega)(1-\tfrac{113}{150}q) , \tag{13}$$

where q is the fraction of $I=\frac{3}{2}$ atoms, 18%, and the subscript M denotes measured quantities, i.e., $\alpha_M^{(1)}(\Delta\omega)$ and $\alpha_M^{(2)}(\Delta\omega)$ can be obtained from the experimental circular and linear polarizations.

III. EXPERIMENT

The experimental setup is illustrated in Fig. 3. An Ar^+ laser pumps two dye lasers. The exciting dye laser is a

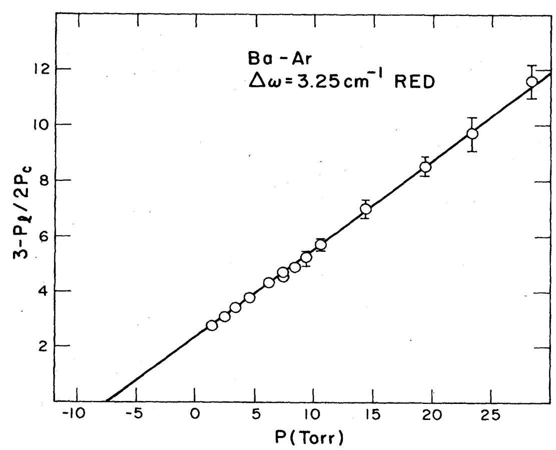

FIG. 2. Argon pressure dependence of the quantity $(3-P_l)/2P_c$. Using Eq. (11) the pressure intercept yields the collisional rate for destruction of orientation.

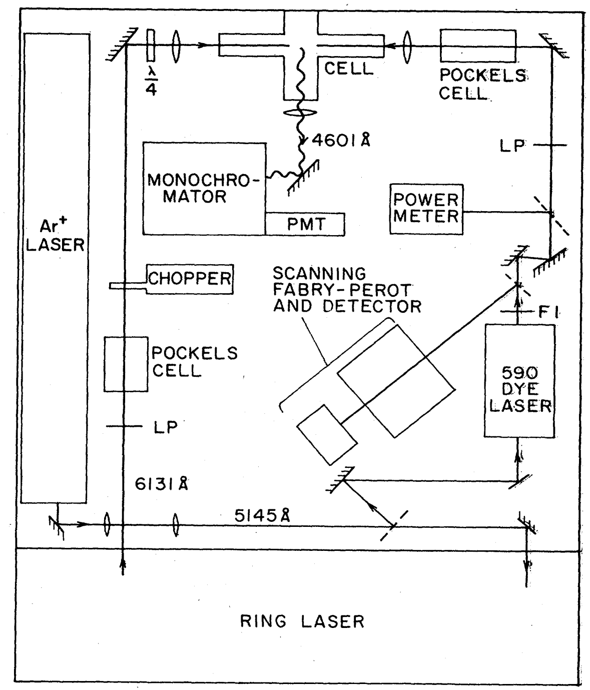

FIG. 3. Experimental setup. LP denotes linear polarizer. $\lambda/4$ denotes a quarter-wave plate. $F1$ is a filter to prevent ring laser reflection off the 590 dye-laser output coupler.

Coherent model no. 590 using a birefringent tuner and a thin étalon as tuning elements. The bandwidth of the laser is $\sim$3 GHz. The probe laser is a homemade single-mode ring dye laser which is locked to an external cavity. The output of each laser is linearly polarized. The excitation laser is made circularly polarized by applying the quarter-wave voltage to a Pockels cell. The probe laser passes through a Pockels cell to give either vertically or horizontally polarized light which is then sent through a quarter-wave plate to produce circularly polarized light. The degree of circular polarization is determined by measuring the linear polarization of the circularly polarized light and using the relation $P_c^2+P_l^2=1$. Typically we find $P_c \gtrsim 99.0\%$. In addition, we also check the setup by exciting with circularly polarized light (orienting the excited state) and probing the residual alignment with linearly polarized light, which should inherently give zero linear polarization. This typically yields linear polarizations of $\lesssim 1\%$.

The detuning of the excitation laser is measured using a scanning Fabry-Perot. All data is normalized to the excitation-laser intensity which is measured using a power meter. The observed signal is found to be nonlinear in the probe-laser intensity (signal $\sim I_{\text{red}}^{1/10}$) and because of the weak power dependence the probe-laser intensity is not monitored. One might have expected that saturation effects due to the red laser would effect the measured polarizations. Experimentally, however, we have determined that we are in a region of red laser intensity where this is not the case since we measured the same linear polarizations with this setup as previously[4] measured. In addition, the measured polarizations are independent of red laser intensity over at least 4 orders of magnitude in intensity. The 4601 Å $8s\,{}^1S-6p\,{}^3P_1$ fluorescence is spectrally resolved from other wavelengths by a 0.2-m monochromator. The throughput of the monochromator is detected by a cooled photomultiplier whose output is sent to a photon-counting system. The filter $F1$ shown in Fig. 3 prevents the probe laser from being reflected off the excitation-laser output coupler and reentering the cell. The cell is a heated stainless-steel cross with a cold finger in the middle of the cross. The cell is typically heated to $\sim 600\,^{\circ}\mathrm{C}$ while the finger is 80—100 °C cooler. In contrast to earlier designs a reentrant window was not needed.

IV. RESULTS AND DISCUSSION

As noted in Sec. II the collisional rate of destruction of orientation can be obtained by measuring the pressure dependence of the circular and linear polarizations. Using the linear-polarization results obtained previously[4] and the circular-polarization results reported here we obtain Fig. 2. From the pressure intercept we have

$$\frac{\gamma_c^{(1)}}{N_{\mathrm{Ar}}}=(1.46\pm0.10)\times10^{-9}\ \mathrm{sec}^{-1}/\mathrm{cm}^{-3}\ .$$

Using our previously obtained value for $\gamma_c^{(2)}/N_{\mathrm{Ar}}$ [$(1.30\pm0.08)\times10^{-9}\ \mathrm{sec}^{-1}/\mathrm{cm}^{-3}$], we find

$$\frac{\gamma_c^{(1)}}{\gamma_c^{(2)}}=1.12\pm0.10\ .$$

It is interesting to note that this agrees with the result obtained by Berman and Lamb[17] for a van der Waals interaction (C_6/R^{-6}) assuming[18] $C_6({}^1\Sigma)/C_6({}^1\Pi)=4$.

The circular-polarization data extrapolated to zero pressure and corrected for hyperfine structure is shown in Fig. 4. The point shown at 1 cm^{-1} (red or blue) detuning represents our estimate of the impact polarization based on measurements at 0.6 cm^{-1} to the red and blue. The blue detuning value was slightly ($\sim 2\%$) higher than the red due to terms on the order of $\Delta\omega\tau_c$ which were neglected in obtaining Eq. (2). Thus we took the average of the blue and red points to obtain the impact value. Note that τ_c^{-1} for Ba-Ar is $\sim 5.2\ \mathrm{cm}^{-1}$ ($\gg 0.6\ \mathrm{cm}^{-1}$). The impact linear and circular polarizations obtained in this way are

$$P_l(\Delta\omega \ll \tau_c^{-1})=(69.3\pm1.6)\%\ ,$$

$$P_c(\Delta\omega \ll \tau_c^{-1})=(64.6\pm2.0)\%\ .$$

Using Eq. (16) of Ref. 4 and Eq. (2) we obtain

$$\frac{\gamma_c^{(1)}}{\gamma_c}=0.880\pm0.043\ ,$$

$$\frac{\gamma_c^{(2)}}{\gamma_c}=0.798\pm0.036\ .$$

Berman and Lamb[17] obtained $\gamma_c^{(1)}/\gamma_c=0.849$ and $\gamma_c^{(2)}/\gamma_c=0.758$ for a van der Waals potential assuming $C_6(\Sigma)/C_6(\Pi)=4$. [Note that for $C_6(\Sigma)/C_6(\Pi)=\frac{7}{4}$ one would predict an impact linear polarization of $\sim 81\%$, which clearly disagrees with the data.[19]]

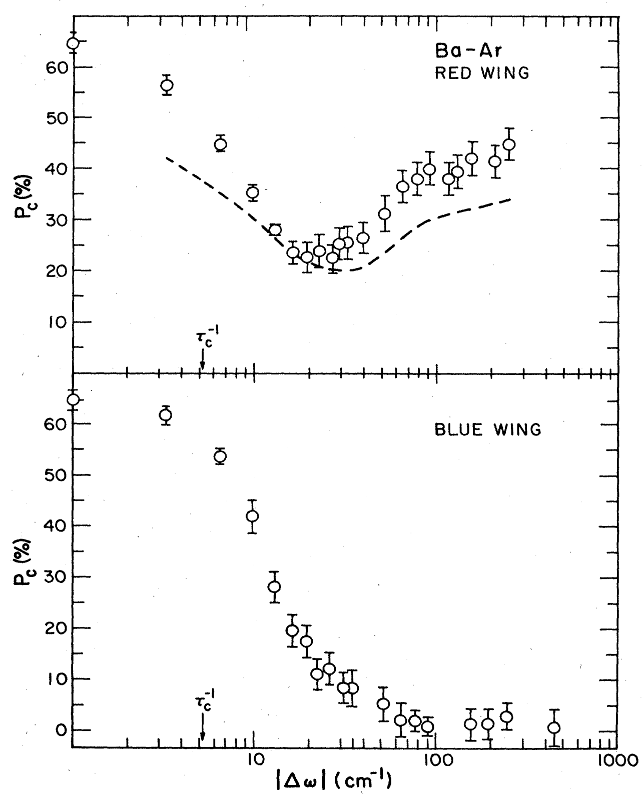

FIG. 4. Circular polarization [see Eq. (1)] as a function of excitation-laser detuning for the red and blue wings. The dashed curve is a prediction of the circular polarization using the linear-polarization results of Ref. 4 and Eq. (8). For this prediction we have assumed straight-line trajectories and used the reorientation model.

The red-wing circular polarization is seen in Fig. 4 to decrease as the detuning leaves the impact region, reach a minimum at $\Delta\omega\approx 23\ \mathrm{cm}^{-1}$, and then increase for $\Delta\omega \gtrsim 27\ \mathrm{cm}^{-1}$. This behavior can be understood in terms of the reorientation model given in Sec. II. From previous results[4] it is known that the red wing is due almost entirely to ${}^1\Pi$ excitation for $\Delta\omega \gtrsim 10\ \mathrm{cm}^{-1}$. According to Eqs. (3)—(5) the polarization decreases as the average rotation angle Ω_Π increases. Thus for SLT one expects the red-wing circular polarization to monotonically decrease as $\Delta\omega$ increases. However, previous results[4] suggest that for $\Delta\omega \gtrsim 30\ \mathrm{cm}^{-1}$, curved trajectories are important. The effect of curved trajectories (off the repulsive core interaction) is to decrease the average angle of rotation of the ${}^1\Pi$ orbital.[4,11] Hence, as we tune further into the red wing and look at "harder" collisions, the average rotation angle decreases leading to an increase in the polarization.

It is also known that the blue wing is due almost entirely to ${}^1\Sigma$ excitation.[4] Thus for $\Delta\omega \gg \tau_c^{-1}$ we expect the circular polarization to be zero [see Eq. (6)]. Figure 4 shows that as we increase the detuning out of the impact region the polarization decreases such that for $\Delta\omega \gtrsim 100\ \mathrm{cm}^{-1}$ the polarization is a constant value close to zero. The reason for the nonzero polarization in the far blue wing is that we never quite have a pure ${}^1\Sigma$ state. Because the atom and perturber have a nonzero relative velocity there will always be a small amount of rotational coupling between the $^1\Sigma$ and $^1\Pi$ states. This rotational coupling can be estimated as the ratio of the Coriolis coupling to the $^1\Sigma$-$^1\Pi$ difference at the excitation radius:

$$\frac{\hbar(b\bar{v}/R_{\rm exc}^2)}{|V_\Sigma(R_{\rm exc})-V_\Pi(R_{\rm exc})|} ,$$

where b is the impact parameter and $\bar{v}$ is an average velocity. Thus the state we excite to in the far blue wing has a small $^1\Pi$ component which produces a small nonzero circular polarization.

It was noted in Sec. II that for straight-line trajectories the circular polarization can be predicted from the linear polarization, within the reorientation model. Using our previous linear-polarization measurements one would predict the dashed curve shown in Fig. 4 for the red-wing polarization. The agreement is rather good considering that the range of validity for this prediction is 10 $\text{cm}^{-1} \lesssim \Delta\omega \lesssim 30\ \text{cm}^{-1}$ (for $\Delta\omega \lesssim 10\ \text{cm}^{-1}$ the reorientation model breaks down since we are no longer in the quasistatic region and for $\Delta\omega \gtrsim 30\ \text{cm}^{-1}$ we have curved trajectories).

From Eq. (9) we can predict the ratio of the excitation to the decoupling radius from the linear and circular polarization if we assume straight-line trajectories. Since it appears that the SLT assumption is valid for $\Delta\omega \lesssim 30\ \text{cm}^{-1}$, we can predict X using the linear- and circular-polarization values at, say, 20 cm^{-1} to the red:

$$P_l(20\ \text{cm}^{-1},\ \text{red}) \approx 22.5\% ,$$

$$P_c(20\ \text{cm}^{-1},\ \text{red}) \approx 44\% .$$

This yields $R_{\rm exc} \approx \frac{1}{2} R_{\rm dec}$. Assuming $R_{\rm exc} \sim 5$ Å we obtain $R_{\rm dec} \sim 10$ Å which is comparable to the Weisskopf radius of ~ 9 Å.[4] One would expect the decoupling radius to be within a factor of 2 of the Weisskopf radius.[20] Thus the decoupling radius obtained from these results using the reorientation model is consistent with expectations. This was not the case for Ba-Xe for reasons discussed in Ref. 4.

In conclusion, we have performed a redistribution experiment using circularly polarized light. The results in the impact region are reasonably consistent with those of a van der Waals potential $[C_6(\Sigma)/C_6(\Pi)=4]$ even though a van der Waals potential is known to be valid over a rather small region in internuclear separation.[4] The results for quasistatic detunings, $\Delta\omega \gg \tau_c^{-1}$, are interpretable using a reorientation model originally used to explain redistribution experiments with linearly polarized light. Previous experiments indicated the far blue wing was due to $^1\Sigma$ excitation. This experiment indicates that a small amount of $^1\Pi$ appears in the far blue wing due to rotational coupling.

ACKNOWLEDGMENTS

We wish to thank P. S. Julienne and I. V. Hertel for stimulating discussions. We also thank G. Nienhuis and other members of the redistribution roundtable discussion at the Seventh International Conference on Spectral Lineshapes, Aussois, France, 1984 for suggesting a circular-polarization experiment. This work was supported by National Science Foundation Grant No. PHY-82-00805 through the University of Colorado and one of us (J.C.) by the Atomic and Plasma Radiation Division of the U.S. National Bureau of Standards.

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[1]J. L. Carlsten, A. Szöke, and M. G. Raymer, Phys. Rev. A **15**, 1029 (1977).

[2]P. Thomann, K. Burnett, and J. Cooper, Phys. Rev. Lett. **45**, 1325 (1980).

[3]W. J. Alford, K. Burnett, and J. Cooper, Phys. Rev. A **27**, 1310 (1983).

[4]W. J. Alford, N. Andersen, K. Burnett, and J. Cooper, Phys. Rev. A **30**, 2366 (1984).

[5]V. Kroop and W. Behmenburg, Z. Phys. A **294**, 299 (1980).

[6]M. D. Havey, G. E. Copeland, and W. J. Wang, Phys. Rev. Lett. **50**, 1767 (1983).

[7]K. Burnett, J. Cooper, R. J. Ballagh, and E. W. Smith, Phys. Rev. A **22**, 2005 (1980).

[8]K. Burnett and J. Cooper, Phys. Rev. A **22**, 2027 (1980); **22**, 2044 (1980).

[9]G. Nienhuis, J. Phys. B **16**, 1 (1983).

[10]P. S. Julienne and F. H. Mies, Phys. Rev. A **30**, 831 (1984).

[11]E. L. Lewis, M. Harris, W. J. Alford, J. Cooper, and K. Burnett, J. Phys. B **16**, 553 (1983).

[12]J. Cooper, in *Spectral Line Shapes,* edited by K. Burnett (de Gruyter, Berlin, 1983), Vol. 2, p. 737.

[13]P. S. Julienne, in *Spectral Line Shapes,* edited by K. Burnett (de Gruyter, Berlin, 1983), Vol. 2, p. 796.

[14]N. Allard and J. Kielkopf, Rev. Mod. Phys. **54**, 1103 (1982).

[15]Equation (11) is easily obtained from Eq. (68) of Ref. 10.

[16]P. E. G. Baird, R. J. Brambley, K. Burnett, D. N. Stacey, D. M. Warrington, and G. K. Woodgate, Proc. R. Soc. London, Ser. A **365**, 567 (1979).

[17]P. R. Berman and W. E. Lamb, Phys. Rev. **187**, 221 (1969).

[18]H. Margenau, Rev. Mod. Phys. **11**, 1 (1939).

[19]Using Eq. (3), p. 31 of Ref. 18, one finds that $C_6(^1\Sigma)/C_6(^1\Pi)=4$ assuming only ^1S states contribute to the van der Waals interaction. A value of $\frac{7}{4}$ assumes ^1S and ^1D states contribute equally. If only ^1D states contribute, a value of $\frac{22}{19}$ is valid.

[20]J. Grosser, J. Phys. B **14**, 1449 (1981).

[21]C. E. Moore, *Atomic Energy Levels,* Natl. Bur. Stand. (U.S.) Circ. No. (U.S. GPO, Washington, D.C., 1958), Vol. III.

[22]J. R. Rubbmark, S. A. Borgström, and K. Bockasten, J. Phys. B **10**, 421 (1977).