Polarization spectra of excited-state-Mg(3p)—rare-gas-atom optical collisions

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Experimental, polarization-dependent excitation spectra for excited-state-Mg—rare-gas-atom optical collisions are reported. In these first studies of the process, polarized Mg atoms in the 3p \( ^1P_1 \) level are produced by absorption of linearly polarized light tuned to the 3s \( ^1S_0 \rightarrow 3p \ ^1P_1 \) resonance transition at 285.2 nm. Detuning-dependent, collision-induced polarization spectra are measured in a ±200-cm\(^{-1} \) range around the Mg 3p \( ^1P_1 \rightarrow 5s \ ^1S_0 \) transition at 571.2 nm. The spectra correspond to probing transient Mg-Ne and Mg-Ar molecules on 3p \( ^1\pi \rightarrow 5s \ ^1\Sigma^+_0^+ \) and 3p \( ^3\Sigma^+_0 \rightarrow 5s \ ^1\Sigma^+_0 \) electronic transitions. Measurements of these excited-state polarization spectra for Mg-Ne optical collisions reveal that for detunings to the red of the atomic Mg 3p \( ^1P_1 \rightarrow 5s \ ^1S_0 \) transition, electronic linear polarization greater than 50% survives far into the molecular regime. This represents a direct measure of the polarization important to alignment-dependent inelastic processes in alkaline-earth-metal—rare-gas-atom collisions. The polarization spectra are discussed in terms of existing information on the interatomic potentials and through an axial recoil limit for the polarization degree for parallel and perpendicular molecular transitions. Rate coefficients \( k \) for disalignment of Mg 3p \( ^1P_1 \) atoms by collisions with Ar \( [k = 9.4/(5) \times 10^{-10} \) cm\(^3\)/s] and with Ne \( [k = 6.5/(7) \times 10^{-10} \) cm\(^3\)/s] are also extracted from the data.

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I. INTRODUCTION

Studies of optical and radiative collisions, whereby correlated radiative and collisional interactions are necessary for the processes to occur, have proved to be a useful tool for learning about electronic multipole dynamics in binary atomic collisions. Experiments and calculations involving collisions between unpolarized ground-state atoms have investigated the dynamics of electronic orientation and alignment, and explored the effects of nonadiabatic electronic energy transfer occurring during the collision [1-4]. Recently, measurements of polarization dynamics by two-photon optical collisions have been reported [5,6]. In these fractional collision experiments, an electronic polarization is injected into a colliding system by absorption of polarized light during a collision, and probed by absorption of a second photon during the same collision. Generally related studies of photodissociation and photoassociation have also investigated the effects of electronic polarization on the experimental observables [7-9]. Due to the short time scale associated with typical thermal energy collisions \( (\sim 1 \) ps), light intensities used in most experiments correspond to a weak-field regime, where the response of the colliding system is linear in the strength of the exciting and probing radiation fields. On the other hand, experiments involving cold-atom collisions explore an alternate dynamical regime, where the time scale of the collisions is orders of magnitude larger and where the dynamics can be strongly affected by the strength of the radiation field [10].

In this paper experimental investigation of an extension of the optical-collision approach is reported. In the process, resonant 3s \( ^1S_0 \rightarrow 3p \ ^1P_1 \) absorption of polarized light by a dilute gas of magnesium atoms [11,12] produces an ensemble of electronically excited and polarized atoms. Collisions between polarized and excited Mg atoms and ground-state Ar and Ne atoms are studied by absorption of light from a second polarized nonresonant radiation field tuned in the vicinity of an atomic Mg electronic transition from the resonant 3p \( ^1P_1 \) level to the more highly excited 5s \( ^1S_0 \) level. The experiments reveal that a large amount of the electronic polarization survives dynamical evolution into the Mg—rare-gas diatomic molecular regime. The experiments directly determine the polarization degree important to alignment-dependent inelastic collisions in alkaline-earth-metal—rare-gas-atom collisions [13,14]. Further, and as will be discussed later in this paper, the results indicate that it will be possible to optically inject electronic alignment directly into a group of molecular terms in such a way that the polarization dependence of the branching into nondegenerate final electronic levels may be directly studied. It is pointed out that the results of this report correspond to a limiting case of the fractional collision results [5,6] published previously. However, due to the much larger signals possible for resonant excitation of the 3p \( ^1P_1 \) level, a considerably wider range of detunings may be investigated in this case.

In the following sections an overview of the physical method and the experimental scheme is described. These are followed by presentation of the experimental results and analysis. Discussion of the results is presented in terms of available Mg—rare-gas potentials and in terms of an axial recoil limit for the polarization dependence of
the processes. The connection of the results to future experiments on electronic energy transfer in excited Mg levels is then discussed.

II. EXPERIMENTAL APPROACH

The basic physical process is depicted in Fig. 1, where relevant atomic Mg energy levels [11] and qualitative Mg–rare-gas interaction potentials are presented [15–18]. In the scheme, Mg atoms are promoted to the $3p^1P_1$ level by absorption of linearly polarized light (laser 1) tuned to the $3s^1S_0 \rightarrow 3p^1P_1$ resonance transition at 285.2 nm. The linear polarization direction defines a z axis, whereby only the $m_f=0$ state of the $3p^1P_1$ level is populated. In the absence of collisions, a second probe light source (laser 2) can promote these atoms to the $5s^1S_0$ level only when tuned to the atomic Mg $3p^1P_1 \rightarrow 5s^1S_0$ transition at 571.2 nm. Satisfaction of the resonance condition is monitored by measurement of the intensity of the $4p^1P_1 \rightarrow 3s^1S_0$ cascade fluorescence at 202.6 nm. The polarization of the intermediate $3p^1P_1$ level is then determined by measuring the intensity $I_z$ when laser 2 is linearly polarized along the z axis and comparing that to the intensity $I_x$ measured when laser 2 is polarized perpendicular to the z axis. The measurements are conventionally parametrized by the polarization ratio

$$P_L = (I_z - I_x) / (I_z + I_x).$$

The total excitation strength is given by $I_f = I_z + 2I_x$. In the absence of depolarizing mechanisms (collisions, hyperfine interactions, external magnetic fields), $P_L = 100\%$ for stepwise resonant excitation.

When collisions can occur, additional excitation takes place even when the second light source is not tuned to atomic resonance. The excitation is due to optical transitions taking place in the transient Mg–rare-gas molecule and between the indicated molecular terms associated with the symptomatic atomic levels. According to the classical Franck-Condon principle [1–4], the transitions take place at localized stationary phase points at internuclear separations $R$. Defining a detuning $\Delta = h_f - h_f^{20}$, where $h_f$ is the energy of laser-2 photons and where $h_f^{20}$ is the energy of the atomic Mg $3p^1P_1 \rightarrow 5s^1S_0$ transition, transitions occur when $\Delta = V_n(R) - V_f(R)$ is satisfied. Here, $V_n(R)$ and $V_f(R)$ are potentials of the upper and lower molecular terms connected by laser 2. Since the atomic Mg $3p^1P_1$ is electronically polarized, the resultant Mg–rare-gas molecule is also. Qualitatively, the molecular polarization depends on the integrated effect of rotational interactions from the commencement of the excited-state collision to the stationary phase point or points associated with optical transitions to the $5s^1S_0^+$ Mg–rare-gas electronic state. Measurement of the polarization $P_L$ proceeds as described in the previous paragraph. However, the initial $5s^1S_0$ population is now produced by dissociation of optically produced $5s^1S_0^+$ Mg–rare-gas molecules.

A schematic diagram of the experimental apparatus is shown in Fig. 2. There two pulsed dye lasers are pumped by the green second-harmonic output of a neodymium yttrium aluminum garnet (Nd:YAG) laser. The grating incidence dye lasers [19] produce 200-µJ, 6-ns pulses having an approximately 3 GHz bandwidth. The output of each laser is amplified by about a factor of 10. Laser-1 pulses are then frequency doubled by an angle-tuned potassium dihydrogen phosphate (KDP) crystal, producing radiation at the atomic Mg $3s^1S_0 \rightarrow 3p^1P_1$ transition at a wavelength of 285.2 nm. The fundamental radiation is removed from the beam by a 1-mm-thick Schott UG-11 colored-glass filter, which passes more than 85% of the desired ultraviolet light. The resultant ultraviolet pulse energies of about 30 µJ are then attenuated to a pulse energy of about 1 µJ in order to avoid saturation of the resonance transition. The output of laser 1 is strongly linearly polarized in a horizontal plane. The laser-2 beam has a wavelength around 571.2 nm and consists of approximately 1-mJ pulses having a temporal width of about 6 ns. Broadband fluorescence from laser 2 and its

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**FIG. 1.** Schematic Mg–rare-gas interatomic potentials. Note that the relative separations of the $4p^1P_1$, $5s^1S_0$, and $4d^1D_2$ levels are to scale, as are the approximate relative shapes of the interatomic potentials.

**FIG. 2.** Block diagram of the experimental apparatus.
amplifier is removed from the beam by passing it through a rutile-prism–iris combination. The fluorescence otherwise produced a spurious signal, when laser 2 was tuned off atomic resonance, due to resonant absorption of the residual fluorescence within the width of the $3p^1P_1 \rightarrow 5s^1S_0$ transition. The laser-2 beam is then passed through a Babinet-Soleil compensator which randomizes the beam polarization across its cross section. Passage of the laser-2 beam through a rotatable linear polarizer completes the polarization analyzer, which has an analyzing power of greater than 0.98. The laser beams propagate collinearly, but in opposite directions, through the Mg vapor cell. Each beam has a cross section of about 4 mm in the interaction region of the cell. When laser 2 is tuned to stepwise atomic resonance, its intensity is decreased by about $10^4$, in order to avoid saturation of the transition.

A detailed description of the sample cell has been given in an earlier report [6], and will be only briefly described here. It consists of a 250-cm$^2$ Pyrex sphere mounted with four side arms in a T-shaped configuration. Quartz windows are attached to the ports by high-vacuum epoxy. Magnesium vapor is generated in the cell by heating, with an internal coaxial heater, a stainless-steel boat packed with Mg metal chips and located just below (~ 1 cm) the interaction region of the cell. The boat is mounted on a quartz tube, which thermally isolates the hot oven from the much cooler cell walls. The boat temperature of about 580 K is monitored by a Chromel-Alumel thermocouple attached to the outside of the boat. A commercial oven controller provides both heater power and feedback from the thermocouple output, resulting in a temperature stability of ±0.2 K. At the usual boat temperature, the Mg density is roughly $10^{11}$ cm$^{-3}$ in the interaction region of the cell. The effective temperature in the interaction region of the cell is approximately 330 K, determined by the mean wall temperature of the cell. The entire sample cell arrangement is attached to an ion pumped vacuum system allowing evacuation of the cell to less than $10^{-6}$ Torr. The vacuum system also permits addition of a rare gas into the cell. A capacitance manometer having a rated accuracy of 0.1 Torr is used to determine the rare-gas pressure. The electronic manometer is calibrated against an oil manometer; the two measurements agree to within the combined uncertainty of about 0.2 Torr.

Population produced in the Mg 5s $^1S_0$ level is monitored through the $4p^1P_1 \rightarrow 3s^1S_0$ cascade fluorescence at 202.6 nm. The fluorescence is collected at 90° from the laser beams and detected by a photomultiplier tube (PMT) having a bialkali cathode and a quartz window, permitting detection of ultraviolet light. In order to eliminate the intense scattered laser light and other atomic emission from the cell, the PMT was mounted with well-blocked interference filters which passed about 2% of the desired fluorescence light, and which eliminated nearly all other light from the cell, this being predominately the intense $3p^1P_1 \rightarrow 3s^1S_0$ resonance fluorescence at 285.2 nm. The filters passed a residue of this light amounting to less than 10% of the weakest excited-state optical-collision signals reported in this paper. The PMT output, integrated over a 350-ns gate opened after the laser-2 pulse exited the cell, was digitized and stored in a laboratory computer for further analysis.

### III. RESULTS AND DISCUSSION

A single data run consisted of recording the signals from 2000 laser shots with laser 2 polarized first along $z$ ($I_z$) and then along $x$ ($I_x$). Residual background levels were obtained for each single run and subtracted from the total signals before obtaining $P_L$ as described above. Leakage of resonance fluorescence through the interference filters was the dominant source of background level. Spurious signals due to direct pumping of the atomic Mg $3p^1P_1 \rightarrow 5s^1S_0$ transition by laser-1 or laser-2 fluorescence were negligible. For each detuning $\Delta$, results from two to eight (depending on the signal-to-noise ratio) single data runs were averaged to obtain the polarization at that detuning. Measured polarization values were independent of Mg density and the intensity of either laser 1 or laser 2. As described previously, systematic effects on the measured polarization due to the operation of the polarimeter are smaller than 2%. As discussed in an earlier report [6], hyperfine and Zeeman depolarization of the signals is negligible, even for direct stepwise excitation of the $5s^1S_0$ level.

The main systematic effect on the measured values of $P_L$ is collisional depolarization of the Mg $3p^1P_1$ level by rare-gas collisions prior to the excited-state optical collisions. Essentially, the mean polarization in that level is reduced by collisional mixing of the initial $m_f=0$ population, produced by laser-1 optical pumping, into the $m_f=\pm1$ states. As the main body of data is taken at an Ar or Ne pressure of 20 Torr, the measured polarization is less than it would be in a single collision limit. To account for this effect, pressure-dependent measurements of $P_L$ in the range 5–30 Torr were made. The measurements were made with both lasers tuned to stepwise resonance, and also with laser 2 set at several nonresonant detunings. Within the statistics of the measurements, the resonant and nonresonant values were in agreement with each other, both for Ar and for Ne collisions. Typical data for several Ne pressure runs are shown in Fig. 3, where it is seen that the inverse polarization $P_L^{-1}$ increases quite linearly with increasing pressure. To analyze the pressure dependence, the laser pulse temporal profiles were modeled as rectangular pulses having widths of 3 and 6 ns for laser 1 and laser 2, respectively. Then temporal overlap for our experiment is such that the two laser pulses overlap in the interaction region of the cell for the full length of the pump pulse; the probe pulse then ends approximately 2.5 ns after the pump pulse. Accounting for spontaneous decay of the $3p^1P_1$ level (2.1 ns lifetime) and weak fields leads to the following expression for the pressure dependence of the normalized alignment $\langle A_0 \rangle$ and the $P_L^{-1}$:

$$\langle A_0 \rangle = -1.192/x - (0.833/x^2)(1-e^{1.43x})(e^{-2.62x})$$,

$$P_L = -3 \langle A_0 \rangle / (2 - \langle A_0 \rangle)$$.
Here, \( x = 1 + \Gamma d \tau \), where \( \Gamma d \) is the collisional disalignment rate for the \( 3p \ 1P_1 \) level and \( \tau \) the radiative lifetime of that level [11]. The results of fitting the measurements to the above model expression are summarized in Table I, which contains the average rate coefficients for collisional depolarization of the Mg \( 3p \ 1P_1 \) level by Ar and Ne. Also presented are the results of fitting the data to a triangular pulse model, which yields quite complicated expressions for the pressure dependence of the polarization. The results for this model are seen to be in good agreement with those of the rectangular pulse one and within the error in the fit in each case. We thus quote the average value obtained from the two models, with the much larger uncertainty obtained from one of them. The average rate coefficients are used to extrapolate the polarization spectra taken at 20 Torr to a single collision limit. To our knowledge, the depolarization rate coefficients reported here are the only values available for the Mg \( 3p \ 1P_1 \) level. Experimental data on depolarization of the resonance transition in Sr–rare-gas and in Ba–rare-gas depolarizing collisions have been reported [20,21]. Not unexpectedly, these values are on the same order of magnitude as those reported in Table I.

Detuning-dependent polarization spectra for Mg-Ne and Mg-Ar excited-state optical collisions, which represent the main results of this paper, are displayed in Figs. 4 and 5. In each case, measured 20-Torr polarization spectra and \( P_L(\Delta) \) spectra extrapolated to a single collision limit are presented. The error bars on the vertical scale represent the statistical error in the measurements, compounded in the low-pressure limit with the statistical uncertainty in the rate coefficient used in the extrapolation. The uncertainty in the horizontal detuning scale is about 1 cm\(^{-1}\), negligible on the scale of the Figures.

For both positive and negative detunings, the qualitative shape of the spectra is similar in Mg-Ne and Mg-Ar. In the zero detuning, stepwise resonance, limit the polarization rises to 100\%, within the statistics of the measurements. This is what is expected in the impact limit, when the inverse detuning is larger than the mean time scale for the collisions. For negative detunings the polarization remains quite high out to \( \Delta = -200 \text{ cm}^{-1} \), persisting at a higher level of about 50\% for Mg-Ne, while dropping to around 10\% for Mg-Ar. For positive detuning values, the polarization drops quite sharply to small values for both molecules, but again is persistently higher for Mg-Ne.

**TABLE I.** Rate coefficients at \( T=330 \text{ K} \) for collisional disalignment of the Mg \( 3p \ 1P_1 \) level by Ar and Ne atoms.

<table>
<thead>
<tr>
<th>( k \times 10^{-10} \text{cm}^3/\text{s} )</th>
<th>Mg-Ar</th>
<th>Mg-Ne</th>
<th>Model for calculations</th>
</tr>
</thead>
<tbody>
<tr>
<td>9.3(5)</td>
<td>6.3(7)</td>
<td>Square pulse</td>
<td></td>
</tr>
<tr>
<td>9.5(1)</td>
<td>6.7(1)</td>
<td>Triangular pulse</td>
<td></td>
</tr>
<tr>
<td>9.4(5)</td>
<td>6.5(7)</td>
<td>Average</td>
<td></td>
</tr>
</tbody>
</table>

FIG. 3. Ne pressure dependence of the inverse polarization \( P_L^{-1} \) measured at several probe detunings.

FIG. 4. Excited-state optical-collision polarization spectra for Mg-Ne optical collisions. Data taken at 20-Torr Ne and an extrapolation to a low-pressure limit are presented.

FIG. 5. Excited-state optical-collision polarization spectra for Mg-Ar optical collisions. Data taken at 20-Torr Ar and an extrapolation to a low-pressure limit are presented.
To interpret these general features, good qualitative potentials are required. Fortunately, rotationally resolved spectra of Mg-Ne and Mg-Ar have provided quantitative information on the shape of the $3s^1\Sigma^+_0$ and $3p^1\pi_1$ electronic potentials [15,16]. Lambda doubling in the observed spectra has also allowed some qualitative information on the $3p^1\Sigma^+_0$ state to be extracted. To our knowledge, no experimental or theoretical information is available on the shape of the $5s^1\Sigma^+_0$ electronic potential. However, calculations on the Mg-Ne $4s^1\Sigma^+_0$ potential by Hliwa and Daudey [17], along with expectations obtained from spectroscopy of alkali-metal–rare-gas-atom $\Sigma$ states, indicate that the nodal structure in the metal atom electronic wave function generates a barrier in the electronic potential at long range and that significant binding occurs at shorter range. Thus the calculated potentials of Hliwa and Daudey [17] may plausibly be used for qualitative discussion. In the absence of a full set of quantitative potentials to use in interpretation of our results, we base a general qualitative discussion on the information given in the papers indicated above. Development of an extension of the model of Lewis et al. [22] that is applied to the experimental results given in this paper is reported by Kuprianov [23].

In preparation for a discussion of the experimental results, it is useful to consider a limiting case where the Mg–rare-gas collisions are axial. For this case, only transitions from the lower $3p^1\pi_1$ and $3p^1\Sigma^+_0$ states to the upper $5s^1\Sigma^+_0$ state occur; of course, transitions from the $3p^1\pi^+_1$ state do occur in the general case of nonzero impact parameter. For a Mg atom located at an origin and polarized in the direction $e_1$, and for a rare-gas atom approaching in the direction $n$, the probability of producing a $\Sigma$ and a $\pi$ orbital is given by $(n \cdot e_1)^2$ and $1 - (n \cdot e_1)^2$, respectively. The second laser excites the colliding atoms in the molecular region with probability $(d \cdot e_2)^2$, where $d$ is the transition dipole moment and where $e_2$ is the linear polarization direction of the second laser. One can then readily show that the transition dipole moment is directed along the unit vectors $n$ and $n \times (n \times e_1)/|n \times e_1|$ for $\Sigma-\Sigma$ and for $\Sigma-\pi$ transitions, respectively. The probabilities of two-step excitation are then given by

$$[1 - (n \cdot e_1)^2][e_2 \cdot (n \times (n \times e_1)/|n \times e_1|)]^2$$

for $\pi-\Sigma$ excitation. In the experimental situation, the polarization of the Mg orbital is taken as $e_1 = z$, while $e_2 = z$ or $x$. Averaging the above expressions over collision angles yields the axial recoil polarization values of $P_L(\Sigma-\Sigma) = \frac{1}{9}$ and $P_L(\pi-\Sigma) = \frac{4}{9}$. Note that these results ignore important effects such as molecular axis rotation and interference of the excitation channels. As shown by Kuprianov [23], full theoretical results represent a combination of these axial recoil values with slowly varying and detuning-dependent coefficients. This dependence arises through movement of real Condon points as a function of the detuning of laser 2 from resonance.

Now we qualitatively discuss the experimental results. First, for red detunings ($\Delta < 0$) observed in these experiments, there are, in both Mg-Ne and Mg-Ar, multiple Condon points. The general location of these points seems to be dependent only on the qualitative global features of the potentials, rather than on quantitative ones. They correspond to molecular transitions from both the $3p^1\pi_1$ and the $3p^1\Sigma^+_0$ electronic states to the $5s^1\Sigma^+_0$ state. For Mg-Ar, the Condon points for the $3p^1\pi_1 \rightarrow 5s^1\Sigma^+_0$ transition connect levels in the quite strongly attractive $3p^1\pi_1$ potential and to a region several hundred cm$^{-1}$ below the dissociation limit of the more strongly attractive $5s^1\Sigma^+_0$ potential. Due to the low Ar pressures used in the experiments, these transitions are not expected to contribute strongly to the observed signals. This is supported by the observed linearity of the signal size with laser-2 intensity; any bound-state contribution would be strongly saturated for the intensities used in the experiments and would produce a nonlinear intensity dependence in the observed signals. Thus only the molecular $3p^1\Sigma^+_0 \rightarrow 5s^1\Sigma^+_0$ transition is expected to contribute significantly to the Mg-Ar $P_L$ spectrum when $\Delta < 0$. However, in Mg-Ne the $3p^1\pi_1$ potential is very shallow, and the Condon point for this case corresponds to optical transitions to near the top of the $5s^1\Sigma^+_0$ potential barrier. Then molecular transitions from both $3p^1\pi_1$ and $3p^1\Sigma^+_0$ could plausibly contribute to the observed polarization spectrum when $\Delta < 0$.

The measured $P_L(\Delta)$ spectrum for $\Delta < 0$ is in qualitative agreement with these expectations for both molecules. In the case of Mg-Ar, increasing $\Delta$ corresponds to Condon points located at increasingly smaller internuclear separations, and toward a more repulsive region of the $3p^1\Sigma^+_0$ potential. Accumulated rotation of the polarized orbital then leads directly to the decreased polarization seen for larger red detunings. As expected, the polarization is less than the axial recoil limit of 50% for all $\Delta < 0$. For Mg-Ne the same general phenomenon should occur. However, as discussed in the previous paragraph, a contribution from the $3p^1\pi_1$ electronic state is possible in this case. Previous results have indicated that because of the existence of both in-plane and out-of-plane electronic orbitals for this state, high polarizations occur for optical collisions involving $np^1\pi^\pm$ electronic states. The measured leveling off of $P_L(\Delta)$ around 50% in the Mg-Ne red wing is consistent with this possibility. The axial recoil limit obtained for the $\pi^-$ state would be significantly reduced by molecular axis rotation in this case, the residual polarization arising from the $\pi^+$ state only.

The situation when $\Delta > 0$ (blue wing) seems more complex. The potentials of Fig. 6 generally indicate that excitation from either lower molecular state will become anisotropic in the near blue wing, with the $3p^1\pi_1$ lower state being the dominant one. This is supported by the observation that in each molecule the strength of the signal drops off much more rapidly with detuning than when $\Delta < 0$. How, then, can excitation to the $5s^1\Sigma^+_0$ state via the $3p^1\pi_1$ state lead to the very small polarization measured in the far blue wing? A likely possibility is that one or more of the 4$d$ potential curves are sufficiently deep that amplitude for direct excitation of them becomes important (the asymptotic 4$d$ level lies only about 600 cm$^{-1}$ above the 5$s$ level) for detunings on the order of 100...
cm$^{-1}$ for Mg-Ne and 50 cm$^{-1}$ for Mg-Ar. This observation is consistent with the more attractive potentials expected in the latter case, but would require nonadiabatic mixing into the 5s state to generate the observed signal. Supporting this interpretation is much greater transition strength for excitation to the 4d states compared to the 5s one. Further, quenching of the 4d $^1D_2$ level to the 5s $^1S_0$ level has been observed in an auxiliary experiment, implying that just such nonadiabatic coupling exists. Finally, excitation to the 4d $^1\pi$ state from the 3p $^1\pi_1$ state proceeds via a parallel transition, and thus can lead to small polarizations when probing the lower $\pi$ state. These observations suggest that polarization-dependent mixing and final-state branching into the 4d $^1D_2$ and 5s $^1S_0$ levels may be observed in optical collisions.

A second possibility is that resonant scattering through the centrifugal barrier produces population in the well region which may be trapped for a relatively long time (compared to the usual collision time) within the effective potential. Then collisions subsequent to the resonant one may reorient the original collision plane, leading to rotational depolarization of both 3p $^1\pi_1^+$ and 3p $^1\pi_1^+$ orbitals. This would require a quadratic dependence of the signal size on rare-gas pressure, which cannot be completely excluded by the data available in the far blue wings, where the signal-to-noise ratio is small. Finally, it may be that the 3p $^3\Sigma_0^+$ potential is significantly deeper in the long-range region than existing data indicate. Then a real stationary phase point for excitation from the 3p $^3\Sigma_0^+$ state is feasible; lower polarization values are possible for 3p $^1\Sigma_0^+$ excitation to the 5s $^1S_0$ state.

IV. CONCLUSIONS

Experimental studies of a new process, excited-state optical collisions, have been made on polarized Mg (3p $^1P_1$) in collision with neon and argon atoms. The experiments first directly show that a sizeable fraction of the alignment produced in excitation of an atomic level survives during the collision process well into the molecular regime (of internuclear separations), particularly for Mg-Ne. Experiments [13,14] based on this premise are thus well supported by our observations. The experimental results also indicate that direct observation of alignment-dependence inelastic branching processes may be made for these systems, and experiments in this direction are under way.

With appropriate modeling, additional information concerning the collision dynamics may be extracted from the data. For Mg-Ne and Mg-Ar, collisional disalignment rate coefficients for the 3p $^1P_1$ resonance level have been obtained by modeling of the laser pulse shape in combination with the observed pressure dependence of the polarization. Further modeling of the full polarization spectra, using an extended version [23] of the treatment developed by Lewis et al. [22], is currently under way. This should provide quantitative information on the quantitatively unknown interatomic potentials for the 3p $^1\Sigma_0^+$ and 5p $^1\Sigma_0^+$ states.

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