Collisional modification of the atomic-Na $3s\ ^2S_{1/2}-5s\ ^2S_{1/2}$ two-color two-photon polarization spectrum

L. Cook, D. Olsgaard, M. Havey, and A. Sieradzan
Physics Department, Old Dominion University, Norfolk, Virginia 23529
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An experimental investigation of the atomic-Na $3s\ ^2S_{1/2}$–$5s\ ^2S_{1/2}$ two-color two-photon polarization spectrum has been made. Measurements of the linear polarization degree of the two-photon transition were made for one laser tuned within a $\pm 100\ \mathrm{cm}^{-1}$ range of the atomic-Na $3p\ ^2P_j$ levels, with the second adjusted to satisfy the two-photon resonance condition. The relatively short-pulse (~ 0.5 ns) lasers used eliminate effects on the spectrum due to atomic-Na hyperfine interactions and radiation trapping, while greatly reducing depolarization due to collisions with Ar gas. In the absence of collisions, the measured polarization spectrum is in excellent agreement with calculations. Measurements of the Ar pressure dependence of the spectrum revealed strong depolarization in the vicinity of resonant excitation through the $3p\ ^2P_{3/2}$ level. A collisional depolarization cross section of 200(30) Å for this process was deduced from the data.

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I. INTRODUCTION

Studies of multiphoton processes in atoms and molecules provide detailed information both on the structural properties of the particles and on the dynamical interaction of intense electromagnetic radiation with matter [1–3]. One such elementary process is two-photon absorption, where two photons from a single light source or one photon from two separate light sources are absorbed. In the second case, the frequency and polarization of each source may be separately varied, along with the relative wave vectors of the radiation. This two-color two-photon approach provides a flexible tool for studying a number of atomic structural and dynamical properties, including excited-state hyperfine splittings, collisional depolarization cross sections, and atomic lifetimes [4–10]. While these processes are on the atomic energy shell, recent studies of two-photon absorption during a binary collision, a so-called fractional collision [11,12], provide an example of a multiphoton process that energetically requires a collision to proceed.

An important factor in two-photon excitation is the time scale associated with the process. In studies employing pulsed light sources, the scale may be determined by the temporal pulse width, the time interval between the two pulses, the detuning of the source frequencies from intermediate states, or the spectral distribution and degeneracies of contributing intermediate states. For collision studies the interval between absorption of one photon and absorption of the second, which may be less than 50 fs for the case of fractional collisions, is a critical parameter in the process.

In this paper we are concerned with two-color two-photon excitation in the vicinity of the atomic-Na$3_s\ ^2S_{1/2}-3p\ ^2P_j-5s\ ^2S_{1/2}$ stepwise resonances. Here $j=\frac{1}{2}$ or $\frac{3}{2}$. The excitation spectra obtained show strong variations as a function of the frequency and polarization of the light sources [13,14]. These are due to the multiple coherent pathways through the different fine and hyperfine components available for optical excitation. We have measured the two-photon polarization spectrum as a function of laser frequency, relative linear polarization direction, and argon gas pressure. In a cell containing Na vapor only, excellent agreement between theory and experiment is obtained over a $\pm 100\text{-cm}^{-1}$ range of detunings around the $3p\ ^2P_{3/2}$ level. The Ar gas-pressure dependence of the linear polarization degree upon stepwise $3s\ ^2S_{1/2}-3p\ ^2P_{3/2}-5s\ ^2S_{1/2}$ excitation leads to a cross section for disalignment of the Na $3p\ ^2P_{3/2}$ level free of effects due to the atomic-Na nuclear spin [15,16]. Disalignment collisions are among the most basic of atomic collision processes, and provide a global measure of the Coriolis and spin-orbit interactions responsible for the atomic depolarization. Recent experiments have demonstrated the feasibility of measurement of dynamical evolution of the alignment during the course of a collision [11]. Numerous experiments have also shown that evolution of electronic alignment during a collision has a pronounced effect on branching into energetically separated atomic levels [17,18]. However, the total depolarization cross section is an important quantity in that it provides the overall scale for the disalignment process.

In the following sections we first present some details of our experimental arrangement and technique. This is followed by the experimental results and analysis, which are then discussed and compared with available theoretical and experimental data.

II. EXPERIMENT

The basic two-photon two-color excitation scheme is illustrated in Fig. 1, where the excitation and spontaneous decay pathways of interest, along with the pertinent atomic-Na energy levels are indicated. In the scheme, separate pulsed laser light sources produced photons of frequency f_1 and f_2 such that $f_1+f_2=f_0$, where f_0 is

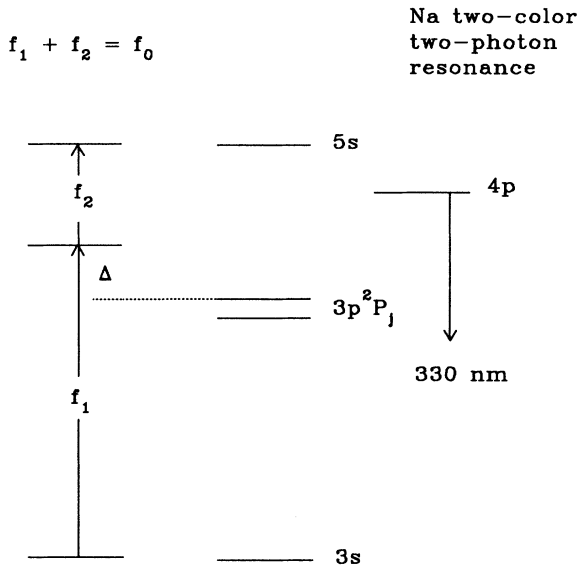

FIG. 1. Partial energy-level diagram for atomic Na, illustrating excitation and fluorescence pathways relevant to the two-color two-photon excitation scheme.

the atomic-Na $3s$-$5s$ frequency interval. Each laser has a bandwidth of about 0.5 cm^{-1}, much larger than the hyperfine splitting of any of the levels in Fig. 1. As f_1 and f_2 are separately varied, the virtual level indicated by a dashed line in Fig. 1 is moved relative to the $3p\,^2P_j$ levels. The detuning $\Delta = f_1 - f_{3/2}$ is defined as the energy defect between the laser-1 frequency and the atomic-Na $3s\,^2S_{1/2}$–$3p\,^2P_{3/2}$ resonance transition at 16 973.4 cm^{-1}. In the experiments described here, Δ spans a range of ± 100 cm^{-1}. Detection of the two-photon resonance is made by observing the induced $4p\,^2P_j \rightarrow 3s\,^2S_{1/2}$ cascade fluorescence at 330 nm.

The experimental apparatus is schematically shown in Fig. 2. Details of this setup have been described previously [19], so only an overview is presented here. In the arrangement, two N_2-pumped dye lasers provide separately tunable light pulses having an effective duration of 0.53(5) ns in a bandwidth of about 0.5 cm^{-1}. Laser 1 is tuned in the vicinity of the atomic-Na resonance transition around 590 nm, while the laser-2 output frequency is varied around the $3p\,^2P_j \rightarrow 5s\,^2S_{1/2}$ transition near 616 nm. Each laser produces a maximum output power of about 15 μJ collimated to a beam size of 2 mm. For each setting of laser 1, the frequency of laser 2 is varied to satisfy the two-photon resonance condition. This is determined by maximizing the $4p\,^2P_j - 3s\,^2S_{1/2}$-induced fluorescence signal at 330 nm.

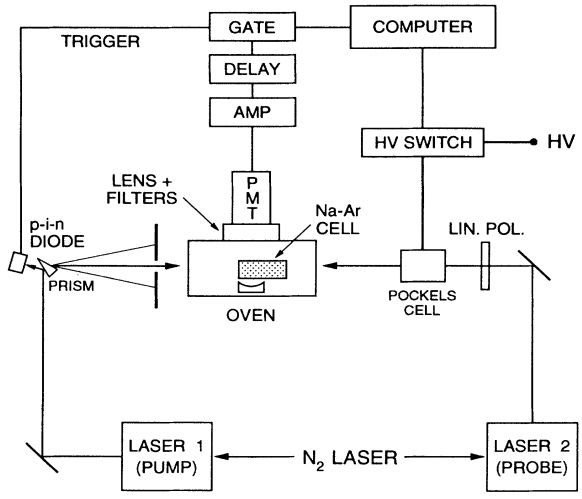

FIG. 2. Block diagram of the experimental apparatus.

The two laser beams are directed colinearly into a sealed Pyrex cell containing Na metal and up to 100 Torr Ar gas. The cell is mounted in an oven heated with a coaxially wound element to a temperature of about 413 K, corresponding to a Na density of about 10^{11} cm^{-3}. The output of laser 1 is spectrally purified by a rutile prism ($n \approx 2.5$). The laser-1 output is horizontally polarized, while that from laser 2 is made to be either parallel or perpendicular to this by computer-controlled application of a half-wave voltage to a Pockels cell in the beam path. The linear polarization degree is defined in the usual manner, with $P_L = (I_\parallel - I_\perp)/(I_\parallel + I_\perp)$. The intensities $I_\parallel$ and $I_\perp$ are the average number of photons counted for each run in the two channels. Extinction at the 10^{-4} level of the linear polarization output from either laser with a crossed linear polarization analyzer is routinely achieved.

The $4p$-$3s$ fluorescence signal was detected by an EMI 9817 bialkali-cathode photomultiplier tube (PMT) operated in a gated-photon-counting mode. The PMT was mounted with a 330-nm interference filter (10-nm bandwidth) and a 1-mm-thick Schott UG-11 colored-glass filter. These served to remove background due to the scattered light and atomic-Na D-line resonance fluorescence. The signal was enhanced by a concave mirror mounted inside the oven but opposite the PMT and a lens combination which focused the fluorescence onto the PMT cathode. Photons were counted in a 500-ns gate opened 30 ns after the lasers fired. The delay eliminated significant electronic pickup otherwise counted as photon pulses by the photon-counting electronics. The counting rate was limited to typically less than one photon for every ten laser pulses. This minimized dead-time corrections, which were made as required.

Computer control of the experiment and data acquisition allowed for elimination of systematic effects associated with long-term variations in the signal. Switching the polarization of laser 2 typically every five laser shots eliminated bias of the polarization measurements whereby one polarization direction would have higher average power than the other. The photon-counting data were accumulated and displayed for each laser-2 polarization direction in real time. A histogram of the time distribution for each run, normally consisting of 5000 laser shots, was displayed for diagnostic purposes. Final polarization results were obtained by combining the dead-time corrected data for $I_\parallel$ and $I_\perp$ from all runs and combining the individual errors in quadrature.

III. RESULTS AND DISCUSSION

A. Collisionless polarization spectrum

Results for the linear polarization spectrum in the absence of collisions are shown in Fig. 3. There it is seen that the linear polarization degree of the two-photon $3s$-$5s$ resonances is near 100% when the detuning is large compared to the atomic-Na $3p\ ^2P_j$ fine-structure doublet splitting of 17.2 cm^{-1}, but varies rapidly in the vicinity of $\Delta=0$. The structure is a consequence of the two intermediate levels contributing to the two-photon transition amplitude. Interference between the two pathways is responsible for the strong variations of $P_L(\Delta)$, and also for the previously reported two-photon transparency at $\Delta=-11.6$ cm^{-1}, which occurs simultaneously with $P_L=-100\%$ [13,14]. In a time-dependent picture, the dynamical electronic alignment produced by coherent excitation of the $3p\ ^2P_{jm}$ states precesses under the influence of the spin-orbit interaction. When $\Delta=-11.6$ cm^{-1}, the average alignment has rotated so as to be perpendicular to the initial alignment axis. Thus there is no scattering of the second, z-axis polarized photons, and the polarization is -100%, for the transition moment is in the x-y plane. The polarization spectrum also goes smoothly through the value of $P_L=60\%$ anticipated at $\Delta=0$, when the $3p\ ^2P_{3/2}$ level is resonantly excited and probed. The absence of hyperfine-structure effects is a direct results of the short time scale (~ 0.5 ns) associated with the pump-probe approach used in the experiment. This is most evident for resonance excitation of the atomic-Na $3p\ ^2P_j$ levels, where the important time scales are the radiative decay time of 16.3 ns and the hyperfine precession time of ~ 20 ns for $j=\frac{3}{2}$ alignment quantum beats. Here it is the interaction of the orbital angular momentum with the unobserved electron spin which leads to reduction of the polarization from 100% expected from an s-p transition in a spinless system to the 60% value obtained for well-separated fine-structure components. The $j=\frac{1}{2}$ level cannot be aligned and so no hyperfine quantum beats associated with that level can be observed in the experiments described here. Thus negligible radiative decay or hyperfine precession occurs in the ~ 0.5-ns pump-probe time and these effects may be ignored. When Δ is away from atomic resonance the most important time scales are those associated with the two-photon excitation amplitudes, $|\Delta|^{-1}$ and $|\Delta+17.2|^{-1}$. Finally, when $|\Delta| \gg 17.2$ cm^{-1} all spin interactions become negligible and $P_L \rightarrow 100\%$.

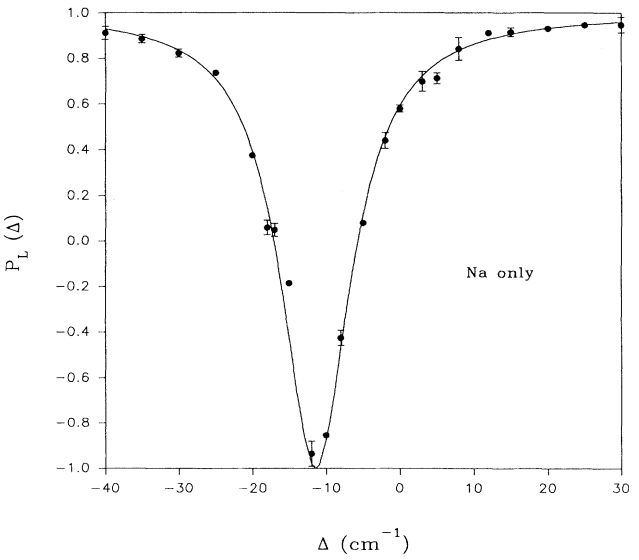

FIG. 3. Two-color two-photon polarization spectrum in the vicinity of the Na D lines for a pure Na sample. The solid line represents the result of a four-level calculation of the polarization spectrum. The only parameters in the calculation are the positions of the atomic-Na $3p\ ^2P_j$ fine-structure levels.

The solid curve in Fig. 3 is the result of a calculation of the polarization spectrum assuming a weak-field, four-level atomic model and ignoring hyperfine and spontaneous radiative processes. A rotating wave approximation is used, along with $f_{1/2}=16\,956.2$ cm^{-1} and $f_{3/2}=16\,973.4$ cm^{-1} [20]. There are no adjustable parameters in the model, and the agreement is seen to be excellent over the entire range of detunings.

Note also that our measured polarization spectrum is equivalent to the polarization dependence of Rayleigh scattering as reported by Tam and Au [21]. The polarization of scattered light near the atomic $D2$ transition has also been measured by Walkup, Migdall, and Pritchard [22]. In that experiment the spectral variation of the alignment parameter, which is simply related to P_L [23], was determined by measurements with a narrow-band, continuous-wave laser. Under those circumstances, decreases in P_L due to the unresolved excited-state hyperfine structure were important. If the excited-state hyperfine splitting had been resolved, an interference structure similar to that reported here would have been observed, but on the spectral scale of the hyperfine level separations. For detunings large compared to the hyperfine structure, but such that $\Delta \ll 17.2$ cm^{-1}, the polarization rose in those experiments to the approximately 60% level determined (for near $D2$ resonant excitation) by the well-resolved $3p\ ^2P_j$ fine-structure splitting; this is the polarization degree measured in our experiments.

B. Collisionally modified polarization spectrum

The two-photon polarization spectrum as in Fig. 3 has been measured as a function of Ar gas pressure up to 100 Torr. Spectra such as those shown in Fig. 4 have also been recorded at 40 Torr Ar and 7 Torr Xe [24]. All the spectra have qualitatively the same structure except near direct excitation of the Na resonance levels. Collisions are found to modify the spectrum in two ways. First, in the vicinity of $D1$ resonance excitation around $\Delta=-17.2$ cm^{-1} a mild inflection in the P_L spectrum is seen. This is an instrumental effect, and is due to increased near-resonant pumping and probing around $\Delta=-17.2$ cm^{-1} combined with uncertainties in tuning the relatively broadband dye lasers used in the experiments reported here. Around the $D2$ pump transitions at $\Delta=0$, a strong

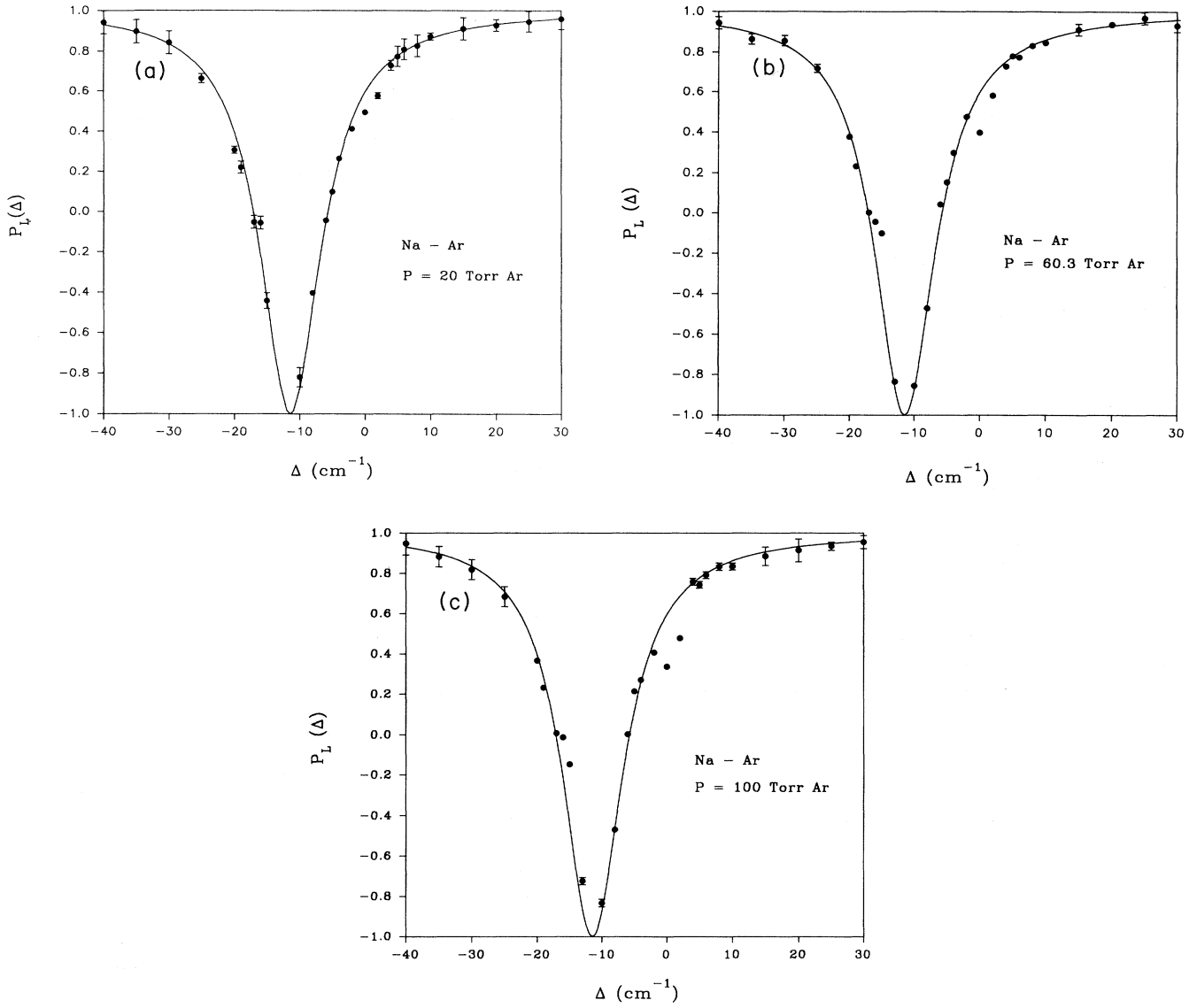

FIG. 4. Collisionally modified two-color two-photon polarization spectra in the vicinity of the Na D lines. The solid line represents in each case the result of a four-level calculation of the collision-free spectrum. (a) $P=20$ Torr Ar. (b) $P=60$ Torr Ar. (c) $P=100$ Torr Ar.

reduction is seen in P_L as the Ar gas pressure is increased. This is due to collisional disalignment [15] of the $3p\,^2P_{3/2}$ level. The cross section for this process is approximately 200 Å^2. As discussed in the following section, we have analyzed the Ar pressure dependence of fine-structure mixing and collisional depolarization in order to extract a disalignment cross section for the $3p\,^2P_{3/2}$ level independent of hyperfine or external field effects.

The P_L spectra of Fig. 4 are qualitatively similar to those measured by Zei *et al.* [14], but extend over a significantly larger range of Δ. The similarity is notable because of the great difference in experimental conditions under which the spectra were obtained. In our measurements a Na density of $\sim 10^{11}$ cm^{-3} was typical, while in those of Zei a density of $\sim 10^{16}$ cm^{-3} was indicated. Under those more severe conditions a number of atomic and molecular ionization and collisional processes occur which are unimportant in our experiment. To the extent that these processes effect $P_L(\Delta)$, the rate associated with them must be smaller than the inverse time scale important to the unperturbed shape of the polarization spectrum. Apparently, only near $\Delta=0$ and -17.2 cm^{-1}, where the time scale for the radiative excitation process can be much longer, does the influence of the collisional processes play a significant role in determining $P_L(\Delta)$. Under the conditions of the experiments reported here, only Na-Ar collisions play a role in modifying the polar-

ization spectrum. This makes possible the detailed quantitative analysis of the collisional depolarization near $\Delta=0$ presented in the following section.

C. Measurement of the $3p\ ^2P_{3/2}$ depolarization cross section

When $\Delta=0$, the $3p\ ^2P_{3/2}$ level is directly excited and aligned by the linearly polarized laser 1. As described previously, laser 2 probes the alignment through measurement of P_L. In spite of the ~ 0.5-ns pump-probe time scale, collisional depolarization of the $3p\ ^2P_{3/2}$ level does occur. This is illustrated in Fig. 5, which shows the dependence of the inverse polarization P_L^{-1} on Ar gas pressure.

The functional dependence of P_L^{-1} on pressure has been obtained through a rate-equations analysis of the $3p\ ^2P_{3/2}$ m_j-state populations. The analysis assumes that the pump and probe lasers produce rectangular temporal pulses, though similar calculations using triangular and Gaussian pulse shapes led to indistinguishable final results within the statistical uncertainty of the measurements. Further, for the largest collision rate and time scale of the results reported here (when $P=100$ Torr Ar), fine-structure transitions have only a few percent effect on the relative $3p\ ^2P_{3/2}$ m_j-state populations. Fine-structure transitions thus have a negligible effect on the measured polarization for the lower-pressure data used to obtain the depolarization cross section. For rectangular pulses, P_L is then given by

$$P_L=3[x-1+e^{-x}]/[2x^2+x-1+e^{-x}]\ , \qquad (1)$$

where $x=\Gamma T$, Γ is a disalignment rate, and T the temporal width of the laser pulses. A least-squares fit of the 0–60-Torr data in Fig. 5 yielded a value of the coefficient x/P of $=0.028(3)$ Torr^{-2}, where P is the Ar pressure. The usual rate coefficient is given by $k=x/PT$.

In order to extract a rate coefficient from these measurements, T must be found. To accomplish this, auxiliary pump-probe measurements of fine-structure mixing [25] were made by pumping the $D1$ transition and measuring the population in each fine-structure level. Similar measurements were also done, but pumping instead the $D2$ transition. These measurements were made with the probe linear polarization at the magic angle of 54.7° relative to that of the pump, so that alignment effects were eliminated from the population measurements [23]. Both pump and probe transitions were unsaturated for the measurements, which were made at an Ar pressure of 100 Torr. The experiments yielded ratios $I(D1)/I(D2)=0.22(2)$ for $D2$ pumping and $I(D2)/I(D1)=0.55(6)$ for $D1$ pumping. Rate-equations analysis of the $3p\ ^2P_{3/2}$ and $3p\ ^2P_{1/2}$ populations were made assuming rectangular pump and probe laser pulses of width T, and a fine-structure mixing rate coefficient [26] of $K_{12}=10.7(1.3)\times 10^{-10}$ cm^3 s^{-1}. As before, other pulse shapes led to polarization results statistically indistinguishable from the rectangular-pulse-shape ones. The analysis yielded an average $T=0.53(5)$ ns for the two measurements. The average value of T was then combined with the results from the depolarization measurements to give a Na($3p\ ^2P_{3/2}$)-Ar depolarization rate coefficient $k=1.66(25)\times 10^{-9}$ cm^3 s^{-1}. Further assuming that $\langle \sigma v\rangle$ may be factored so that $\langle \sigma v\rangle=\sigma_d\langle v\rangle$, a cross section of 200(30) Å^2 was obtained. Here $\langle\ \rangle$ denotes an average over a Maxwell-Boltzmann distribution of relative velocities at the cell temperature of 413 K, for which $\langle v\rangle=8.4\times 10^4$ cm s^{-1}.

The value obtained for the cross section is generally in excellent agreement with more recent determinations of this quantity [27–29], as seen by the summary in Table I. Our measurements are unique in that they are not subject to the usual slowing down of the relaxation rate [15,16,30] by the hyperfine interaction. This effect normally needs to be removed by either analysis or by performing the experiments in a magnetic field large enough that the nuclear spin is decoupled from the electronic angular momentum. That hyperfine interactions are negligible in our measurements is seen directly from the measured value of $P_L=58(2)\%$ for resonant excitation of the $3p\ ^2P_{3/2}$ level in the absence of collisions. Averaged over the 16.3-ns lifetime of that level, hyperfine precession

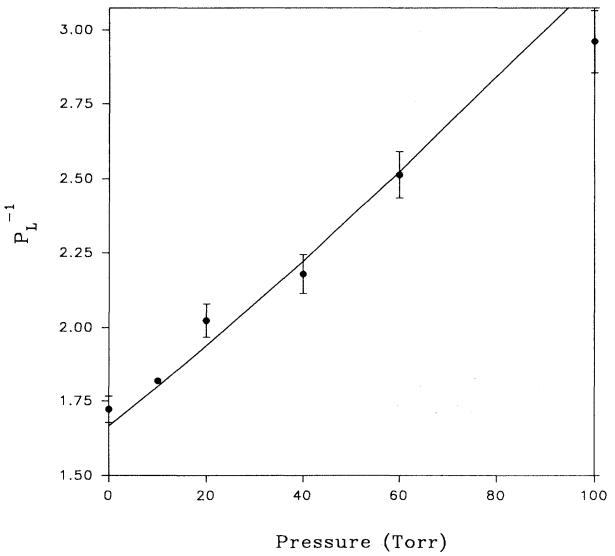

FIG. 5. Dependence of the inverse polarization P_L^{-1} for resonant $D2$ excitation on Ar gas pressure. The solid line represents the results of a fit to a square-pulse pump-probe model of the collisional depolarization process.

TABLE I. The disalignment cross section σ_d for the Na $3p\ ^2P_{3/2}$ level for Na-Ar collisions. Values are in Å^2 units. Numbers in parentheses refer to the uncertainty.

Source	σ_d (Å^2)
This work	200(30)
Gay and Schneider [27] expt.	228(13)
Elbel *et al.* [28]	308(31)
Behmenburg *et al.* [29] calc.	229

would reduce this from the theoretically expected value of 60% to about 21%. Second, we have made measurements of the polarization hyperfine quantum beats [8,30–32] as a function of delay time between the pump and probe lasers. These measurements, which are planned to be presented elsewhere, show that about 2 ns must pass before significant precession of the electronic angular momentum occurs in the Na nuclear magnetic field. Thus the result presented here for σ_d represents a direct measurement of that quantity.

IV. CONCLUSIONS

An experimental study of the two-color two-photon $3s\ ^2S_{1/2}-5s\ ^2S_{1/2}$ excitation spectrum of atomic Na has been made. In the measurements the $3p\ ^2P_j$ fine-structure doublet levels are the dominant intermediate levels. The linear polarization degree shows strong spectral variations due to interference of the excitation amplitudes from the two intermediate levels. In the absence of collisions, excellent agreement between the measurements and a simple four-state model of the polarization spectrum is found. The effect of Ar collisions on the spectrum is evident in their depolarizing influence near resonant excitation of the $3p\ ^2P_{3/2}$ level, and a depolarization cross section was extracted from the data.

ACKNOWLEDGMENTS

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[1] *Multiphoton Processes,* 6th ed., edited by S. J. Smith and P. L. Knight (Cambridge University Press, Cambridge, England, 1988).
[2] W. Demtroder, *Laser Spectroscopy* (Springer-Verlag, Berlin, 1982).
[3] *Laser Spectroscopy of Atoms and Molecules,* edited by H. Walther (Springer-Verlag, Berlin, 1976).
[4] A. Sieradzan and F. A. Franz, J. Phys. B **17**, 701 (1984).
[5] P. F. Liao, J. E. Bjorkholm, and P. R. Berman, Phys. Rev. A **21**, 1927 (1980).
[6] J. H. Tung, A. Z. Tang, G. J. Salamo, and F. T. Chan, J. Opt. Soc. Am. B **3**, 837 (1986).
[7] W. K. Bischel, P. J. Kelly, and C. K. Rhodes, Phys. Rev. Lett. **34**, 300 (1975).
[8] T. W. Ducas, M. J. Littman, and M. L. Zimmerman, Phys. Rev. Lett. **35**, 1752 (1975).
[9] A. Quattropani and R. Girlanda, Riv. Nuovo Cimento **6**, 1 (1983).
[10] J. Morellec, D. Normand, G. Mainfray, and C. Manus, Phys. Rev. Lett. **44**, 1394 (1980).
[11] D. A. Olsgaard, M. D. Havey, and A. Sieradzan, to appear, Phys. Rev. Lett. (1992).
[12] G. Alber and J. Cooper, Phys. Rev. A **31**, 3644 (1985).
[13] J. E. Bjorkholm and P. F. Liao, Phys. Rev. Lett. **33**, 128 (1974).
[14] D. Zei, R. N. Compton, J. A. D. Stockdale, and M. S. Pindzola, Phys. Rev. A **40**, 5044 (1989).
[15] W. E. Baylis, in *Progress in Atomic Spectroscopy,* edited by W. Hanle and H. Kleinpoppen (Plenum, New York, 1979), p. 246.
[16] Alain Omont, Prog. Quantum Electron. **5**, 69 (1977).
[17] R. L. Robinson, L. J. Kovalenko, and S. R. Leone, Phys. Rev. Lett. **64**, 399 (1990).
[18] L. J. Kovalenko, R. L. Robinson, and S. R. Leone, J. Chem. Soc. Faraday Trans. 2, 939 (1989).
[19] D. A. Olsgaard, M. D. Havey, and A. Sieradzan, Phys. Rev. A **43**, 6117 (1991).
[20] A. A. Radzig and B. M. Smirnov, *Reference Data on Atoms, Molecules, and Ions,* Chemical Physics Vol. 31 (Springer-Verlag, Berlin, 1985).
[21] A. C. Tam and C. K. Au, Opt. Commun. **19**, 265 (1976).
[22] R. Walkup, A. L. Migdall, and D. E. Pritchard, Phys. Rev. A **25**, 3114 (1982).
[23] C. H. Greene and R. N. Zare, Annu. Rev. Phys. Chem. **33**, 119 (1982).
[24] M. D. Havey, in *Proceedings of Symposium on Atomic and Molecular Collisions in Excited States,* edited by B. Stumpf (University of Idaho, Moscow, ID, 1991), p. 55.
[25] L. Krause, in *The Excited State in Chemical Physics,* Advances in Chemical Physics Vol. 28, edited by J. W. McGowan (Wiley, New York, 1975), p. 267.
[26] J. Apt and D. E. Pritchard, J. Phys. B **12**, 83 (1979).
[27] J.-C. Gay and W. B. Schneider, Z. Phys. A **278**, 211 (1976).
[28] M. Elbel, B. Kamke, and W. B. Schneider, Physica **77**, 137 (1974).
[29] W. Behmenberg, V. Kroop, and F. Rebentrost, J. Phys. B **18**, 2693 (1985).
[30] K. Blum, *Density Matrix Theory and Applications* (Plenum, New York, 1981).
[31] Th. Krist, P. Kuske, A. Gaupp, W. Wittmann, and H. J. Andra, Phys. Lett. **61A**, 94 (1977).
[32] W. Yei, A. Sieradzan, and M. Havey (unpublished).