Laser induced optical pumping measurements of cross sections for fine and hyperfine structure transitions in sodium induced by collisions with helium and argon atoms

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ABSTRACT
Optical pumping of the ground states of sodium can radically alter the shape of the laser induced fluorescence excitation spectrum, complicating measurements of temperature, pressure, etc., which are based on these spectra. Modeling of the fluorescence using rate equations for the eight hyperfine states of the sodium D manifolds can be used to quantify the contribution to the ground state pumping of transitions among the hyperfine excited states induced by collisions with buffer gas atoms. This model is used here to determine, from the shape of experimental spectra, cross sections for $\Delta F$ transitions of the $P_{3/2}$ state induced by collisions with helium and argon atoms, for a range of values assumed for the $P_{3/2}, \Delta F$ cross sections. The hyperfine cross sections measured using this method, which is thought to be novel, are compared with cross sections for transitions involving polarized magnetic substates, $m_f$, measured previously using polarization sensitive absorption. Also, fine structure transition ($\Delta J$) cross sections were measured in the pumped vapor, giving agreement with previous measurements made in the absence of pumping.

PACS Categories:
Fine and hyperfine structure (32.10.Fn), Fluorescence... (32.50.+d), ...optical pumping (32.80.Bx), Scattering of atoms... (34.50.-s)
I. INTRODUCTION

Narrow band frequency-scanning lasers have been used with trace sodium vapor in low pressure gases to measure temperature and pressure from the lineshape of the sodium D resonances observed in fluorescence [1,2,3,4]. Similarly, the Doppler shift of the resonance has been used in flow fields to measure velocity along the laser beam [1,2,3,5]. However, Walkup et al. [6] demonstrated that optical pumping of the hyperfine ground state populations of the sodium can lead to radical changes in the shape of the fluorescence excitation spectrum, thus complicating such measurements. The behavior of the pumped spectrum was quantified using what was in practice a three state model of the sodium. We have used this same basic approach to make temperature measurements and to demonstrate residence time measurements in a laboratory cell using pumped sodium vapor [7,8].

We present here measurements made from laser induced fluorescence excitation spectra of cross sections for transitions among the hyperfine levels of the sodium excited state induced by collisions with helium and argon atoms. These measurements were made possible by expanding the three state model to include explicitly hyperfine structure within, and coupling between, the two excited states. This permits quantification of the contribution to ground state pumping of the excited state hyperfine transitions. As a result, the cross sections for these transitions may, to a degree, be inferred from the shape of the excitation spectrum from pumped vapor. To our knowledge, this is the first use of the fitting of pumped spectra for cross section measurement. We also report the measurement of fine structure cross sections in the pumped vapor, based on signal ratios. These cross sections have previously been measured in the absence of pumping [9].

The hyperfine structure cross section of interest is for collisions which thermalize the distribution over the magnitude of the total angular momentum, $F$, of the sodium atoms in the excited state for the case in which the magnetic substates are evenly populated. Previous measurements of cross sections for collisions which affect $F$ have been based on preferential orientation of the directions of the momenta using circularly polarized light [10,11]. It is expected, however, that the hyperfine, or $\Delta F$, cross sections would be similar in size to the depolarization, or $\Delta m_f$, cross sections [8,9,11]. Error bars for the depolarization measurements have been estimated as typically order of magnitude [11]. For the hyperfine cross section measurements here, the $P_{3/2}$ cross section, denoted $\sigma_{P_{3/2}}$, is
determined from pairs of D1, D2 excitation spectra, not absolutely, but relative to the $P_{1/2}$ cross section, denoted $\sigma_{P_{1/2},\ell}$. For a given $\sigma_{P_{1/2},\ell}$, the accuracy of the $\sigma_{P_{1/2},\ell}$ determination, estimated from the scatter in the results, is typically a few tens of percent.

II. THEORY

The sodium D manifolds involve two hyperfine ground states and six hyperfine excited states, two in the $P_{1/2}$ state, and four in the $P_{3/2}$ state [12], in which each hyperfine state contains magnetic substructure. For the case in which the magnetic substates may be neglected, the rate equations for the D manifolds are

$$
\dot{n}_i = -(\sum_j \beta_j + \sum \gamma_j + D)n_i + \sum \gamma_j n_i + \sum R_{ij}n_j + Dn_i^0
$$

$$
\dot{n}_j = -(\sum_j \gamma_j - \sum \gamma_j + D)n_j + \sum \gamma_j n_j + \sum \beta_j n_i + Dn_j^0
$$

(1)

The $n_i$ are the population number densities of the individual hyperfine states; the index $i = a,b$ refers to the ground states ($F=1,2$); the index $j = 1,2$ ($P_{1/2}, F=1,2$) and $j=3-6$ ($P_{3/2}, F=0,3$) refers to the excited states; $\nu$ is the optical frequency; the $\nu_j$ are the individual hyperfine resonances; $V(\nu - \nu_j)$ is the Voigt profile of the resonance; $P_0$ is the laser power, $a$ is an effective spot size such that the laser irradiance is $P_0/a$; $c$ is the speed of light; $B_j$ is the Einstein absorption coefficient; $R_{ij}$ is the relaxation rate; $\gamma_j$ is the collision induced transition rate; and $D^{-1}$ is the residence time of the sodium in the laser beam. For the spectra treated here, $D$ is set by thermal diffusion, and scales with temperature, $T$, and pressure, $p$, as $D \propto T^{3/2}/p$ [6,13].

The fluorescence excitation spectrum, $f(\nu)$, is determined from the excited state populations, given by numerical solution of Equation 1 for steady-state:

$$
f(\nu) = \left[ n_{\nu_0} P_0 \right]^{-1} \sum_{i=a,b} \sum_{j=1,\Delta} n_j(\nu) A_j h \nu_j
$$

(2)
where the fluorescence has been normalized to the sodium number density $n_{Na}$ and the incident laser power; $A_{\mu}$ is the Einstein coefficient for spontaneous emission and is equivalent to $R_{\mu}$ in the absence of quenching—true for noble gases—and with the neglect of stimulated emission.

The rate of collision induced transitions between any pair of $F$ levels in the two excited states is given by $C_{\mu,\nu}$. For the measurements here, the fine structure transition rate is the total rate per atom of $P_{3/2} \rightarrow P_{1/2}$ transitions, and the hyperfine structure transition rate is the total rate per atom of $F \rightarrow F'$ transitions within a given $P$ state. For reference, the relation between the transition rate and the cross section is, in general, given by [13]

$$C_{\mu} = \sigma_{\mu} \gamma_{\text{Gam}} \rho / \sqrt{T} \quad (3)$$

For $C_{\mu}$ in MHz, $\sigma_{\mu}$ in $\text{Å}^2$, $\rho$ in Torr, and $T$ in °K, the numerical values of $\gamma_{\text{Gam}}$ for sodium with helium and argon, respectively, are 7.61 and 3.68.

As described by Walkup et al. [6] the populations of the two ground states depart from their equilibrium values, $n_{0}^0$, when the laser power, still well below saturation, is large enough so that individual sodium atoms undergo multiple fluorescence cycles while resident in the beam. The cycle number is given by

$$\rho = \left[ \beta_0 / D \right]_{\text{max}} \quad (4)$$

and when it is comparable to or greater than unity, the excitation spectrum becomes distorted, because of repartitioning of the ground state populations. Walkup et al. have shown that, if the internal structure of the $P$ states is neglected, the shape of the pumped spectrum is uniquely determined by temperature, pressure and cycle number.

The effect of the collision induced transitions within (and between) the excited states is to alter the branching ratios for the pumping of the ground states [6,8,11]. In particular, thermalization, or mixing, of the excited state populations tends to erode the comparative isolation of the optical connections to ground. The most striking example of this is laser excitation out of the $F = 2$ ground state into the $P_{3/2}$ state, in which fully 70% of the atoms are excited to the $F = 3$ level. Since the transition from $P_{3/2}, F = 3$ to $S_{1/2}, F = 1$ is electric dipole forbidden [12], these atoms have no
relaxation path, in the absence of mixing, except back to the source ground state. This is an extreme case, but the net result, considering all the transitions involved, is that mixing improves the pumping efficiency of cycling.

The shape of the spectrum can therefore be influenced by the excited state hyperfine mixing rate, despite the fact that the hyperfine splittings are not resolved. Moreover, when the mixing time is comparable to the excited state lifetime (≈16 ns), the shape of the spectrum will vary with the mixing time. This effect is distinctly weaker than the influence of cycle number, but it can be significant. For the D2 manifold in 1 Torr of He at 400°K, for example, the change in the spectrum from one mixing asymptote to the other corresponds to a change in cycle number of about a factor of two. The key point is that both mechanisms, cycling and mixing, affect the spectrum in essentially the same way. For this reason it is convenient to characterize the pumped spectrum by a parameter which uniquely specifies the degree of pumping, independently of the mechanisms.

This may be accomplished with the ground state depopulation ratio, $G$, defined as the mean ratio of pumped ground state populations normalized to the thermal population ratio, sampled at the center frequencies of the individual hyperfine transitions of Equation 1:

$$G = \frac{1}{N} \sum \left( \frac{n_a(v_a)n_0^a}{n_b(v_b)n_0^b} + \frac{n_b(v_b)n_0^b}{n_a(v_a)n_0^a} \right)$$

For D1, $N = 4$, and for D2, omitting the two forbidden transitions, $N = 6$. In the general case, then, the shape of the spectrum is uniquely determined by temperature, pressure and the depopulation ratio, and the depopulation ratio is determined by cycle number and mixing rate. This situation is illustrated in Figure 1, which shows calculations from the eight state model for the D2 manifold ($\nu = 0$ at $f_{\max}$). Each of the two spectrum shapes in Figure 1 corresponds to a distinct value of $G$ and each can be produced with different combinations of the excited state mixing rate, $C_{\nu'}$, and the cycle number, $\rho$. By contrast, the D1 spectrum is comparatively insensitive to mixing. The change in cycle number corresponding to the change in $G$ between mixing asymptotes is only about 15% (again, at 1 Torr and 400°K).
III. EXPERIMENT

The experimental spectra for the collision induced transition cross section measurements were collected from a static cell enclosed in a laboratory furnace. A Coherent 699-21 ring dye laser, pumped by a Coherent Innova 300 argon ion laser, was used to induce the fluorescence in a windowed vacuum chamber containing sodium metal and filled to low pressures with inert buffer gases. The laser power was varied using calibrated ND filters. The temperature, roughly in the range from 450 to 500 °K, was measured with a type-J thermocouple adjacent to the probe volume, and the pressure was measured using a capacitance manometer situated in a cold leg [7,8].

In order to measure the collision cross section for fine structure transitions, narrow-band interference filters (FWHM = 0.3 nm) were used to measure, at separate photomultipliers, the signals from the two \( P \) states. These cross sections are of interest in themselves and are also of interest here because these transitions contribute to the mixing of the hyperfine levels of the \( P \) states.

Because of the sensitivity of the depopulation ratio to the cycle number, accurate measurement of the laser power, which was used to set the cycle number (as per Equation 4), is critical to the determination of the mixing cross section. For this reason, two independent measurements of the (relative) laser power in the probe volume were made, and, as indicated below, agreement between these two measurements was a factor in the hyperfine cross section measurements.

Helium and argon were used, separately, as buffer gases, and the pressure was between about 0.1 Torr and 1 Torr. This pressure range was chosen to produce excited state mixing times comparable to the excited state lifetime, based on the depolarization cross sections from the literature (see Table 3) [10,11]. As indicated by Equation 3, pressures above 1 Torr would also be useful, but this was precluded by the (available) transducer.

Strictly speaking, measurement of the hyperfine cross section defined in the introduction requires that the magnetic substates remain evenly populated independently of collisions. For collimated (anisotropic) incident light, this will be the case if there is sufficiently rapid magnetic mixing, due to an external magnetic field transverse to the propagation vector [14]. This requires a field of a few gauss for the excited state, and, for the absorption rates of interest (\( \beta_n = 10 \) to 100 kHz), on order of \( 10^{-2} \) gauss for the ground state [14,6,8]. Although no magnetic field measurements were
made in the laboratory cell, the local geomagnetic field is about 0.52 gauss directed approximately 76° from the propagation vector. In any case, because the laser was linearly polarized, only alignment, rather than orientation, is possible, and this effect is expected to produce a subpercent change in $G$ if the $S$ states, but not the $P$ states, are mixed, and no more than a few percent change in the complete absence of magnetic mixing [14,8]. Also of note is that the Zeeman splittings for fields on the order of a gauss are on the order of a MHz [4], and are thus negligible in comparison with the Voigt linewidth.

IV. FINE STRUCTURE CROSS SECTION

The cross section for collision induced transitions between the fine structure levels of the sodium is determined, not from the shape of the spectrum, but rather from the ratio of fluorescence signals from the two $P$ states, denoted as $I_{3/2}$ and $I_{1/2}$. In general, the optical pumping plays a role here. For example, the change in $I_{3/2}$ for D1 vs. D2 tuning depends upon both $p$ and $C$. Thus this ratio, $I_{3/2}(D1)/I_{3/2}(D2)$, which is attractive because it may be measured on a single detector, varies by about 20% for the two inside spectra of Figure 1.

On the other hand, the ratio of signals from the two detectors for tuning to a single manifold is not sensitive to the pumping. For D1 tuning, the transition rate is related to this ratio by

$$C_{p3/2-p1/2} = \frac{A + D}{2e^{-\Delta \delta/kT} I_{1/2}/I_{3/2} - 1}$$

This is derived from a three state model, detailed in Ref. 8, which assumes $F$ degeneracy in all three states, and a similar equation holds for D2 tuning. It is also useful to combine the equations for the two tunings, so that the cross section may be determined independently of the laser normalization as well. This is straightforward in principle, but there was a minor complication for the data here. Because of the low pressure, spurious signals due to incomplete discrimination between the D lines by the filters became significant. This was corrected for by using multiple spectra [8].

The results for helium and argon buffer gases are given in Figure 2; the accuracy of the measurements is estimated from the scatter in the results. Table 1 compares the present measurements with other work [9].
V. HYPERFINE STRUCTURE CROSS SECTION

For the hyperfine cross section measurements, a number of spectra have been fitted using a function minimization routine which minimizes the difference between experimental and theoretical spectra by choosing particular variables as fit parameters. The fitting procedure, which has likewise been used to make measurements of temperature and cycle number in pumped vapor, is described in Refs. 7 and 8. For the hyperfine cross section, the important fit parameters are cycle number and mixing rate, and the fundamental problem is the need to distinguish, experimentally, between cycling and mixing. As illustrated in Figure 1, a single spectrum will uniquely determine $G$, but not $\rho$ or $C_{A'F}$.

This difficulty may be partially overcome by collecting the spectra in D1, D2 pairs, sequentially scanning the two manifolds. Since the ratio of cycle numbers for the two spectra is known from the independent measurements of temperature, pressure and laser power, there are now three unknowns, the two mixing cross sections and one $\rho$, for two spectra. In this way, it is possible to determine one cross section in terms of the other; that is, given $G(\rho_{D1}, \sigma_{P_{1/2,F}^2})$, $G(\rho_{D2}, \sigma_{P_{3/2,F}^2})$ and $\rho_{D1}/\rho_{D2}$, all four parameters, $\rho_{D1}$, $\sigma_{P_{1/2,F}^2}$, $\rho_{D2}$ and $\sigma_{P_{3/2,F}^2}$, are determined if one cross section is known.

To illustrate this, and to demonstrate the need, in general, for mixing in interpreting the experimental spectra, Figure 3 gives an example of the fitting for paired spectra. The discrete points are an experimental D2 spectrum (D2 tuning) taken in argon, the same in both (a) and (b). The solid curves are theoretical spectra. Although not shown in the figure, a D1 spectrum taken under the same conditions was also fitted. Figure 3 (a) is a fit from the three state model, in which $G$ is determined solely by $\rho$, and the fitted value of $\rho$, when compared with the value from the D1 fit, gives a laser power ratio (D1/D2) of about 1.4. The actual ratio in this case, known from direct measurements, is 1.02. Figure 3 (b) is a fit from the eight state model (Equations 1 and 2) which, when compared with the corresponding D1 fit, gives the correct ratio. $\sigma_{P_{1/2,F}^2}$ was set arbitrarily to 88 Å$^2$, a value given for the depolarization cross section [10, 11], and this fixes $\rho$ for the D2 spectrum. $\sigma_{P_{3/2,F}^2}$ was then chosen so that the resulting $\rho$ for the D1 spectrum would produce the known ratio, $P_0(D1)/P_0(D2)$. 
The results presented here are based on this approach. Because $G$ is particularly insensitive to $C_{\lambda F}$ for D1, $\sigma_{\rho_{1/2},\lambda'}$ was selected as the starting point. $\sigma_{\rho_{1/2},\lambda'}$ is determined from paired D1, D2 spectra by assigning to $\sigma_{\rho_{1/2},\lambda'}$ a range of arbitrary values. An example of this is given in Figure 4 for a specific $\sigma_{\rho_{1/2},\lambda'}$ in helium buffer gas. The procedure is essentially that of Figure 3 (b), but, in Figure 4, both manifolds are given explicitly. The discrete points are the experimental spectra and the solid curves are the theoretical spectra, from Equation 2. In Figure 4 (a), the D1 spectrum is fitted for $\rho$ assuming $\sigma_{\rho_{1/2},\lambda'} = 500$ Å², resulting in $\rho = 5.13$. This value for $\rho$, adjusted for ($B_\parallel$ and) any changes in $P_0$, $T$, and $\rho$, is then used in Figure 4 (b) to fit the D2 spectrum, giving $\sigma_{\rho_{1/2},\lambda'} = 71.1$ Å². Similarly, the process was repeated beginning with $\sigma_{\rho_{1/2},\lambda'} = 50$ Å², resulting in $\rho = 5.48$ and $\sigma_{\rho_{1/2},\lambda'} = 35.2$ Å², and then again for three additional choices of $\sigma_{\rho_{1/2},\lambda'}$. Each resulting pair of theoretical spectra was sensibly indistinguishable from that of Figure 4. For each $\sigma_{\rho_{1/2},\lambda'}$, then, $\sigma_{\rho_{1/2},\lambda'}$ may be averaged over several pairs of spectra.

Because of the comparative insensitivity of the spectrum to mixing, even for D2, the cross section results are especially sensitive to errors in the fitting. For this reason, several parameters which quantify errors for individual pairs of spectra were used in the data reduction. These parameters, which are defined and discussed in Ref. 8, were essentially (1) the error bar for the direct measurement of laser power; (2) the error bar for the lineshape measurement of temperature; and (3) the merit function for the curve fit. In calculating averages and standard deviations for the cross sections over multiple spectrum pairs, each term is weighted by the inverse product of these parameters [15]. Spectrum pairs for which one or more of these parameters exceeded a reasonable limit (here, about 3%) were excluded entirely.

A more fundamental restriction on the spectra comes from the cycle number. Because the spectrum is only sensitive to mixing for appreciable cycle numbers, a reasonable lower limit must be imposed for $\rho$. This limit was chosen here, from general calculations [8], as $\rho \geq 3$ for the D1 fitting. The specific pressure ranges resulting from this and the quality screening of the data were from about 50 to 800 mTorr for He, and from about 350 to 750 mTorr for Ar.
Table 2 gives the results for the mean $\sigma_{PL/2,AF}$, in Å², for five values of $\sigma_{PL/2,AF}$. A group of from three to six pairs of spectra was used for each cross section measurement and the root mean square (RMS) standard deviation, also in Å², for the group is given. For comparison, values for the depolarization cross sections from the literature are given in Table 3 for several alkali metals in several noble gases.

The hyperfine structure cross section is difficult to measure accurately, and the error bars reflected in Table 2 are significant. Moreover, since $\sigma_{PL/2,AF}$ is not known a priori, the absolute value for $\sigma_{PL/2,AF}$ is indeterminate. If the assumption is made that the $P_{L/2}$ depolarization cross section for Na compares with those of the heavier alkali atoms much as in the case of $P_{3/2}$, the $P_{1/2}$ cross section would be, from Table 3, about 4 Å² for both helium and argon. If the depolarization cross section is then, in turn, used to approximate $\sigma_{PL/2,AF}$, the result from Table 2 is $\sigma_{PL/2,AF} \approx 23$ and 14 Å², respectively, for helium and argon. It should be noted that for the smaller values of $\sigma_{PL/2,AF}$, the mixing rate is (see Equation 3) fairly small compared to the spontaneous emission rate, $A_{J''}$, and the sensitivity of the spectrum to mixing is diminished. On the other hand, if $\sigma_{PL/2,AF}$ is comparatively large, Table 2 implies it is larger than $\sigma_{PL/2,AF}$, which was not expected [11]. In either case, but especially for the smaller cross sections, data at somewhat higher pressures (e.g., 1 to 10 Torr) would be useful.

As indicated above, the fine structure cross section also plays a role in the mixing of the excited states since the destination $P$ state is thermally populated by these transitions as well. The values given in Figure 2 were used for the results presented in this section.

VI. CONCLUSION

We have used an eight state model of the sodium D manifolds to make laser induced fluorescence measurements of cross sections for hyperfine structure transitions induced by collisions with helium and argon atoms. Cross sections for transitions between the two $P$ states were also measured. These cross sections are of interest because they provide information on the interactions between the sodium atoms and the collision partners. They are also of interest, particularly the mixing
cross section, in making cycle number measurements from the spectra [7]. All the spectra used here are from trace sodium vapor in which the ground state populations have been optically pumped by the laser.

The fine structure cross sections have been measured previously in the absence of pumping. The measurements of these cross sections here, which are notable in that they are made at low pressures and in the presence of optical pumping, are in agreement with the previous measurements.

The hyperfine structure cross sections have been determined from the shape of the optically pumped fluorescence excitation spectrum. This technique, which is thought to be novel, is based on the contribution of excited state mixing to ground state pumping and is more viable for the D2 manifold than for the D1 manifold. To demonstrate the technique, we have evaluated the $P_{3/2}$ cross section from least squares curve fitting of experimental spectra by assuming arbitrary values for the $P_{1/2}$ cross section. The measured cross section is specifically for $\Delta F$ transitions among thermally populated $m_r$ states, and the accuracy of the measurements is estimated from about $\pm10\%$ to about $\pm40\%$; this compares with estimated order of magnitude error bars for measurements of depolarization cross sections, which rely on deliberate athermalization of the $m_r$ populations.

REFERENCES

Figure 1. Calculations illustrating the influence of cycling and mixing on the ground state depopulation ratio. The two distinct shapes correspond to two distinct depopulation ratios: $G = 3.8$ for the outer spectra, and $G = 6.4$ for the inner spectra. The discrete points are for $C_{\Delta\nu} / A_{ps} = 10^{-2}$ ($\rho = 7.1$ for the outer spectrum; $\rho = 15$ for the inner spectrum). The solid curves are for $C_{\Delta\nu} / A_{ps} = 10^{+2}$ ($\rho = 3.3$ for the outer spectrum; $\rho = 7.1$ for the inner spectrum). The case is D2 fluorescence in He at $T = 400^\circ$K and $p = 1$ Torr.
Figure 2. Cross section for fine structure transitions in sodium induced by collisions with rare gas
atoms.

◎ helium: $\sigma = 46.16 \pm 2.06 \text{ Å}^2$

□ argon: $\sigma = 66.09 \pm 4.18 \text{ Å}^2$
Figure 3. Fitting of experimental D2 spectrum in Ar using (a) cycling only, and (b) both cycling and mixing. The discrete points are the experimental spectrum and the solid curves are the theoretical spectra. For the three state model (a), the ratio of laser powers from the fitted spectra for D1 (not shown) vs. D2 is 1.44. For the eight state model (b), the ratio is 1.02, which agrees with the direct measurements. The non-unique mixing cross sections used were $\sigma_{pV2,df} = 1250 \text{Å}^2$, $\sigma_{pV2,df} = 88 \text{Å}^2$. 
Figure 4. Hyperfine structure cross section measurement in helium. The discrete points are experimental spectra and the solid curves are theoretical spectra. (a) The D1 spectrum is fitted to determine $\rho$ for the arbitrary choice, $\sigma_{P_{1/2}, \psi} = 500 \text{Å}^2$ (b) This value of $\rho$ is, in turn, used in fitting the D2 spectrum to determine $\sigma_{P_{1/2}, \psi} = 71.1 \text{Å}^2$. 
Table 1. Sodium fine structure cross sections: present work in **bold**; all other values from Ref. 9 (theoretical values in parenthesis)

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Table 2. Cross sections for $P_{3/2}$ hyperfine state transitions in sodium induced by collisions with helium and argon atoms. The cross sections result from curve fitting of pairs of D1, D2 spectra using arbitrary values for the $P_{1/2}$ cross section. The units are $\text{Å}^2$.

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Table 3. Depolarization cross sections for several alkali metals in several noble gases. The units are Å². All values from Ref. 11.

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