SEPARATED-ATOM THEORY OF LASER-INDUCED COLLISIONAL IONIZATION OF Cs BY Sr

S. Geltman *
Joint Institute for Laboratory Astrophysics
National Bureau of Standards & University of Colorado
Boulder, Colorado 80309 USA

Abstract A semiquantitative theoretical description is given for the observed laser-induced collisional ionization (LICI) of Cs by Sr atoms. This is done in the separated-atom picture in which the resonant interaction of the atoms with the radiation is fully taken into account and the collision is treated perturbatively. The basic intensity dependence of the cross section and its spectral width are well explained, but the distinctive observed line asymmetry is accounted for only qualitatively.

1. INTRODUCTION -- THE Sr-Cs LICI EXPERIMENT

An intriguing experiment was carried out by Bréchignac et al. in which they irradiated a Sr-Cs vapor mixture with tunable laser radiation centered at the Sr $5s^2 1S_0 - 5s5p 1P_1$ resonance line and observed the production of Cs$^+$ ions resulting from Sr-Cs collisions. The Cs$^+$ were monitored by certain fluorescences in the recombination spectrum. After accounting for several possible competing mechanisms they managed to extract line shapes for the relative probability of Cs$^+$ formation as a function of

*Staff Member, Quantum Physics Division, National Bureau of Standards.
the detuning $\delta$, and also the variation with laser intensity of the relative probability at exact resonance and its FWHM width. The Sr resonance line is at $\lambda = 460.7$ nm, which is only 67 cm$^{-1}$ from the Cs 6s $^2S_{1/2} - 7p$ $^2P_{1/2}$ transition at $\lambda = 459.3$ nm. In spite of this potential overlap, they have convincingly shown that the two-photon ionization of isolated Cs atoms via the 7p $^2P_{1/2}$ intermediate state resonance produces a negligible contribution to their Cs$^+$ signal, which is then taken to be a pure LICI signal. The experimental results showed very asymmetrical line shapes for the LICI cross section $\sigma(\delta)$ (high for $\delta < 0$ and low for $\delta > 0$), and basically $\sigma(0) \sim I$ and $\Delta\sigma_{1/2} \sim I^{1/2}$ (with some possible systematic deviations at the highest intensity, 15 MW/cm$^2$).

A theoretical study of this process was made by Crance and Feneuille$^2$ using a dressed quasimolecule formalism together with Landau-Zener curve crossing theory. They obtain asymmetric line shapes which appear to have the same asymmetry as the observed line shapes and which also have strongly shifted maxima. They also find the intensity variations $\sigma(0) \sim I^{2/5}$ and $\Delta\sigma_{1/2} \sim I^{1/2}$.

We present here a theoretical study from a rather different point of view. The presence of an exactly resonant field for the Sr atom and one that is somewhat off resonance for the Cs atom means that the Rabi cycling between states in the isolated atoms is a dominant feature of their condition between collisions. For two atoms under those conditions, the meaning of molecular potential curves in describing their interaction is questionable. For example, while the meaning of van der Waals potentials between atoms in asymptotic stationary states is clear,
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how can one construct an effective time-independent van der Waals potential between two atoms which are each in time-dependent superposition states as a result of the resonant or near-resonant external field? For example, under laser intensities of the order of 10 MW/cm² the Sr atom will oscillate between its 5s² ¹S and 5s5p ¹P states with a period of the order of 10⁻¹² s, which is comparable to a collision time.

Because of conceptual questions of this kind with the quasimolecular picture, we will use an approach based upon the separated atoms. Their description as free atoms in a resonant field is almost exact (within the rotating wave approximation), and any description of their interaction through an expansion in the complete set of free atom states is also formally exact. However, in practice such expansions must be severely truncated, as we shall see later. This would also be the case for any rigorous expansion based on molecular states. The intrinsic simplicity of the atomic basis will provide a qualitative understanding of all aspects of the measurement in terms of the basic physical parameters such as atomic oscillator strengths, laser intensity, average relative velocity, and other necessary assumptions, such as classical trajectory and short-range behavior of the interaction between atoms.

2. THEORETICAL FORMULATION AND MODEL

We base our present formulation on our previous work in laser-induced ionizing collisions (reference 3, hereinafter referred to as I). In that work two alkali atoms were assumed to be initially in excited n²P resonance states and finally, after the absorption of one photon
during a collision, one atom was ionized and the other was
de-excited to the n²S ground state. This was the lowest
order process that could result in ionization since the
ionization potential of the excited atoms was greater than
the n²P–n²S energy separation.

There are two basic differences in the present case
from that previous one. First is the above-mentioned ne-
cessity for including the effect of the resonant field on
at least the Sr atom. One could take the point of view
that the detuning from the Cs resonance (∼67 cm⁻¹) is
large enough so that the Cs atom may be regarded as being
entirely in its ground state before the collision. This
would then require the absorption of a laser photon as
well as the effective de-excitation of the Sr atom in or-
der to have enough energy to ionize the Cs. An alternate
point of view, and the one we will presently take, is that
the Cs atom responds to the near-resonance of the applied
field by also being in a time-dependent superposition
state, although of course with much lower average proba-
bility of being in the upper state than is the case for
the Sr atom. Allowing thus for the real but weak popu-
lation of the Cs 7p \(^2\)P\(_{1/2}\) state by the applied field, the
second difference from the previous treatment follows
from the fact that a Sr–Cs collision can lead to Cs\(^+\) with-
out the absorption of any additional photons. The de-
excitation energy of Sr is by itself enough to ionize
Cs 7p \(^2\)P\(_{1/2}\).

This latter fact allows us to ignore the presence of
the radiation field beyond its role in the preparation of
the two colliding atoms in Rabi superposition states. We
know of course that the field continues to act in all orders
as evidenced by the fact that Cs$^+$ will be produced by two-photon absorption of isolated ground-state Cs atoms in this spectral range.$^4$ This process is maximized for radiation tuned exactly to the Cs $6s\,^2S_{1/2} - 7p\,^2P_{1/2}$ energy difference, where many more Cs$^+$ ions would arise from it than are produced in the present collisional process. However, at the detuning of 67 cm$^{-1}$ it is a negligible background to the present collisional process, as shown experimentally and theoretically (in following Sec. 6).

The initial state prior to a collision is the product of the two atom superposition states,

$$\psi_o(r_a, r_b, \tau) = \phi_{a0}(r_a, \tau)\phi_{b0}(r_b, \tau)$$  \hspace{1cm} (1)

where each $\phi_o$ is the superposition state (in the rotating wave approximation),

$$\phi_o(r, \tau) = \frac{2}{\sqrt{\prod_{i, i' = 1}^{2} B_{ii'} \epsilon_i \epsilon_{i'}}} \exp\left[\frac{-i(a_{i, i'} + \eta_i + \eta_i') \tau}{2}\right] \times u_i(r).$$  \hspace{1cm} (2)

Here $\tau$ denotes time measured from the onset of the laser pulse, $\epsilon_i$ and $u_i$ are the unperturbed atomic energies and wave functions, $\eta_1 = 0$, $\eta_2 = \delta = \omega - \omega_{21}$, $a_{1,2} = \frac{1}{2} \left[ -\delta \pm (\delta^2 + \Omega^2)^{1/2} \right]$, $\Omega = E_o z_{12}$ (or $\frac{1}{\sqrt{2}} (x + iy) z_{12}$ for circularly polarized radiation), and

$$B = \frac{1}{2(\delta^2 + \Omega^2)^{1/2}} \left( \begin{array}{cc} \delta + (\delta^2 + \Omega^2)^{1/2} & \delta - (\delta^2 + \Omega^2)^{1/2} \\ \Omega & -\Omega \end{array} \right)$$

(see for example reference 5 for details). We will use atomic units throughout unless otherwise specified.

From this it is seen that for monochromatic radiation exactly on the Sr resonance the average probability that Sr is in its upper state is 1/2 at any intensity while for
toy parameters p, η, φ (all defined in I through the
particular argument v, they also depend on the traje-
ctory a function of the atom a, in addition to their dependence on the ex-
siting a straight-line trajectory for the relative motion
the Porter transformations of the dipole-dipole operator a-
appeared of 1. The I (v) are essential components of
r and I (v) are defined in the
operator component. The a and I (v) are the atomic matrix element of the dipole
and 

\[ \langle 22 | \hat{D} | 22 \rangle = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & -\eta & -\eta & 0 \\ 0 & -\eta & -\eta & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \]

where \( \mathbf{r} \) is the center of a collision.

\[ (3) \]

The first-order transition amplitude to the specified
state is

\[ \langle 1 | \hat{D} \hat{c}^{\dagger} \hat{c} \mid 1 \rangle = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & -\eta & -\eta & 0 \\ 0 & -\eta & -\eta & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \]

The interaction (usual e.g., (6)), and (1), (1) of
and (6) of the dipole-dipole
interaction under the restriction of the dipole-dipole

interaction much lower laser intensities.

sensible at this power level, but could be questionable at
approximation of assuming monochromatic radiation is true
with the stark width \( \Delta \omega = 3.6 \, \text{cm}^{-1} \) in our

experiment, laser bandwidth is 6 \( \Delta \omega \). Thus on the

experimentally, we may also note at this time that the

at 1 \( \text{nm/cm}^2 \). We will use a and \( \Delta \) to designate the SR and GS

\[ \frac{\Delta \omega}{\Delta \omega} = \frac{\lambda(\omega_2 \omega_2)}{\lambda(\omega_2 \omega_2)} \]

as it is.

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trajectory integrals

\[ T_1 = \int_{-\infty}^{\infty} dt \frac{e^{i\Delta t}}{R^3} = \frac{2}{v \rho^2} zK_1(z) \Delta^+ \frac{2}{\Delta^+} \]

\[ T_2 = \int_{-\infty}^{\infty} dt e^{i\Delta t} \left( \frac{v \rho}{R} \right)^2 = -\frac{2z}{3v \rho} [\frac{3K_1(z) + zK_2(z)}{\Delta^+ \frac{2}{3v \rho} ^2} \]

\[ T_3 = \int_{-\infty}^{\infty} dt e^{i\Delta t} \frac{v \rho}{R^5} = \frac{2i}{3v \rho} z^2 K_2(z) \Delta^+ \[0 \]

\[ T_4 = \int_{-\infty}^{\infty} dt e^{i\Delta t} \frac{v \rho}{R^5} = \frac{2}{3v \rho} z^2 K_2(z) \Delta^+ \frac{4}{3v \rho} ^2 \]

where \( z = \rho \Delta / v \). (Note: These expressions are somewhat more condensed than those given in (A.4) of I.)

Since the above transition amplitude applies to any one specific collision resulting in the excitation of a specific state in the Cs continuum, one must sum over all continuum states (energies of the ejected electron) and then average over \( \tau_c \), \( \theta \), and \( \phi \) (instant of collision within the laser pulse and direction of relative motion referred to a fixed coordinate system). Averaging over \( \tau_c \) eliminates cross terms within \( \sum_{i} \), leaving

\[ \langle |a(1)|^2 \rangle_{\tau_c} = \frac{2 \Omega^2 \omega^2}{4(\delta^2 + \omega^2)(\delta^2 + \omega^2)} \sum_{i,i'} \sum_{\lambda} \Gamma_{\lambda}'(\zeta_{i11}) \times \left| \sum_{\lambda} \Gamma_{\lambda}'(\zeta_{i11}) \langle 1f | d_{\lambda} | 22 \rangle \right|^2 \quad . \quad (4) \]

Normalizing \( \langle 1f \rangle \) for the density of states, we obtain the sum over final continuum states using (from eq. (18) in I)

\[ \int_{0}^{\infty} d\epsilon_b f^*_{\lambda'}(\zeta_{i11}) f_{\lambda}(\zeta_{i11}) = \int_{0}^{\infty} d\epsilon_{ii11}^* \Gamma_{\lambda}'(\zeta_{i11}) \Gamma_{\lambda}(\zeta_{i11}) = 2\pi \omega_{\lambda} \quad . \quad (5) \]
We also use the fact that the $\epsilon_{bf}$ variation in $\langle 1f | d_{\lambda} | 22 \rangle$ is slow and this factor may be extracted from the integral at $\zeta_{11}^* \equiv 0$. The order of performing the operations in (4) and (5) should be reversed, but one can easily see that this would not affect the result. Carrying out the angular averages (using definitions in the Appendix of I), we have

$$\overline{W}_{\lambda\lambda'} = \frac{\pi}{\nu \rho} \xi_{\lambda\lambda'}, \quad (6)$$

where the only nonvanishing coefficients $\xi_{\lambda\lambda'}$ are $\xi_{11} = 33/64$, $\xi_{12} = -15/64$, $\xi_{13} = -9/32$, $\xi_{22} = 81/320$, $\xi_{23} = -3/160$, $\xi_{33} = 3/10$, $\xi_{44} = 9/64$, $\xi_{55} = 27/160$, and $\xi_{66} = 9/32$. (Note that values given in I for $\xi_{12}$, $\xi_{13}$, and $\xi_{23}$ have been corrected. I am indebted to Mr. J. J. Malasse for pointing out these errors to me.)

The resulting expression for the average first-order transition probability is now

$$p(1) = \frac{\pi^2}{2 \nu \rho^5} \frac{\Omega_a^2 \Omega_b^2}{(\delta_a^2 + \Omega_a^2)(\delta_b^2 + \Omega_b^2)} \times \sum_{\lambda\lambda'} \xi_{\lambda\lambda'}, \langle 1f | d_{\lambda} | 22 \rangle^* \langle 1f | d_{\lambda'} | 22 \rangle \quad (7)$$

One must also average over initial m-states (which will depend on whether the applied radiation is of linear or circular polarization) and sum over final m-states. This form explicitly exhibits the dependence of the transition probability on $\nu$, $\rho$, $I$ (through $\Omega$'s), the atomic properties in the transition matrix elements and in the initial superposition state (again through $Q$'s), and the effect of the assumed trajectory and dipole-dipole force through the
\( \xi_{\Lambda \Lambda} \)'s and \( d_{\Lambda} \)'s. Thus the effects of the electronic motion within each atom, the relative motion of the atoms, and the applied field are all coupled to each other. The magnitude of the applied field enters only in a simple way here, essentially only by its effect in populating the upper resonance state of each atom, because of the simplification of (5) being independent of \( \xi_{11} \). We will see later that this simplification no longer exists in higher order terms, and a very complicated coupling occurs between the trajectory and the applied field via the Stark states of the atoms.

3. ESTIMATES OF CROSS SECTIONS

To evaluate the cross section for this LICI process it is necessary to integrate the transition probability over all impact parameters. However, it is clear that our expression for \( P^{(1)} \) is valid only for large \( \rho \) because of our approximations — (1) keeping only the dipole-dipole interaction between atoms, (2) treating its effect in only first-order perturbation theory, and (3) assuming only straight-line trajectories. For sufficiently small \( \rho \) it is clear that all three of these approximations would break down, and the evaluation of \( P \) becomes quite intractable. Thus it is necessary to introduce a cut-off impact parameter \( \rho_o \), for which we will assume that \( P(\rho) = P(\rho_o) \) if \( \rho < \rho_o \). Our procedure here is somewhat different from that used in I, where we related \( \rho_o \) to the initial van der Waals potential. This is not adequate here since the van der Waals potentials are not well defined in the presence of the external resonance field. We must therefore choose \( \rho_o \) more arbitrarily.
In the limit of very high field a fortunate simplification occurs, as discussed in I. First we note that the intensity dependence contained in (7) may be explicitly exhibited in the form

\[ p^{(1)} = \frac{\alpha}{\nu} \frac{I^2}{(I_a+I)(I_b+I)} \]  

(8)

Since \( p^{(1)} \) should not exceed 1, as it represents a normalized probability, we will find a saturation impact parameter \( \rho_s \),

\[ \rho_s = \left[ \frac{\alpha}{\nu} \frac{I^2}{(I_a+I)(I_b+I)} \right]^{1/5} \]  

(9)

which will exceed \( \rho_o \) at some high intensity. Assuming that \( p^{(1)} = 1 \) for all \( \rho < \rho_s \) is quite reasonable (one would not expect a decrease in the ionization probability at smaller \( \rho \)), and this leads to the simple cross section

\[ \sigma^{(1)} = \frac{5}{3} \pi \rho_s^2 = \frac{5}{3} \pi \left[ \frac{\alpha}{\nu} \frac{I^2}{(I_a+I)(I_b+I)} \right]^{2/5} \]  

(10)

if \( \rho_s > \rho_o \). The result at lower intensities where \( \rho_s < \rho_o \) follows from our previous assumption to be

\[ \sigma^{(1)} = \frac{5}{3} \pi \rho_o^2 p^{(1)}(\rho_o) \]  

(11)

The values of \( I_a \) and \( I_b \) depend on the detuning \( \delta_a \) (which determines \( \delta_b \)). For the experimental range of \( I \), for which \( I \ll I_b \), it follows that

\[ \sigma^{(1)}(\delta_a=0) \sim I \quad \text{and} \quad \Delta \sigma_{1/2} \sim I^{1/2} \]  

which qualitatively agree with the experimental results.
In figures 1 and 2 we show the measured results as well as the calculated results for $\sigma^{(1)}(\delta_0=0)$ and $\Delta\sigma^{(1)}_{1/2}$ for linearly polarized radiation and various choices of $\rho_0$. Also indicated in figure 1 is $\sigma^{(1)}_S(\delta_0=0)$ which varies roughly as $I^{2/5}$ (from eq. (10)). It is seen that the measured relative results of Bréchignac et al. for $\sigma(\delta_0=0)$ as a function of intensity are roughly fit by any choice of $\rho_0 > 2$. We have normalized their relative measurements to

\[ \text{FIGURE 1} \quad \text{Calculated } \sigma^{(1)}(\delta_0=0) \text{ for various values of the cut-off impact parameter, } \rho_0. \text{ The line for } \rho_0 = 0 \text{ is the limiting high intensity result } \sigma^{(1)}_S(\delta_0=0). \text{ The points are the measured relative cross section normalized to our } \sigma^{(1)}(\delta_0=0) \text{ result for } \rho_0 = 4 \ a_0. \]
FIGURE 2  Calculated widths (FWHM) $\Delta \sigma_{1/2}$ for various values of the cut-off impact parameter, $\rho_0$. The line for $\rho_0 = 0$ is the limiting high intensity result $\Delta \sigma_{1/2}$. The points are the measured absolute widths.

our calculation with $\rho_0 = 4$ in figure 1. In the subsequent discussion of line shapes we will indicate why we think this is perhaps the most reasonable choice for $\rho_0$ to use in our model of the process. Note that for this choice of $\rho_0$ the intensity at which $\sigma_s^{(1)}$ would begin to apply is $\sim 5 \times 10^9$ W/cm$^2$, far beyond the range of the present experiment.

The measured widths in figure 2 are absolute, and we see that they are about one-half the magnitude given by the $\rho_0 = 4$ theoretical curve. This could be partially due to the higher-order effects discussed in §5.
4. ATOMIC MATRIX ELEMENTS AND LIGHT POLARIZATION

The polarization of the applied radiation will affect \( \sigma^{(1)} \) only through the matrix element \( \langle 1f|d_A|22 \rangle \) in (4). Since the ground state of Sr is an s-state, the only populated m-states of the upper resonant level 5s5p \( ^1P \) (designated by \( |2\rangle \)) are \( m = 0 \) and 1 for linear and right circular polarization, respectively. In Cs we take into account fine structure (since it is only the 7p \( ^2P_{1/2} \) upper level which is closest to resonance) so the allowed \( m_j \) states are \( \pm \frac{1}{2} \) (over which the resulting transition probability must be averaged) and \( +\frac{1}{2} \), respectively, for the above two polarizations.

The final continuum states of Cs which are dipole connected with the 7p \( ^2P_{1/2} \) initial state are \( \epsilon s \) \( ^2S_{1/2} \) and \( \epsilon d \) \( ^2D_{3/2} \), or a total of six final \( m_j \)-states which must be summed over. We use quantum defect values for bound-bound and bound-free radial matrix elements.

Carrying out the atomic part of the calculation as indicated above we find that for circular polarization \( \sigma^{(1)}(\delta_A = 0) \) is reduced from its value for linear polarization by a factor of about 0.86, for any choice of \( \rho_0 \). The widths \( \Delta \sigma_{1/2} \) are determined essentially by the Lorentzian factor \( \frac{\Omega_A^2}{(\delta_A^2 + \Omega_A^2)} \) in (4) and hence remain the same for both polarizations. An experimental test of these predictions of our model would be most welcome.

5. LINE SHAPE AND ASYMMETRY — EFFECT OF HIGHER ORDERS

A striking feature of the relative line shapes for \( \sigma(\delta_A) \) observed by Bréchignac et al. is a strong asymmetry in the direction of being high for \( \delta_A < 0 \) and low for \( \delta_A > 0 \). In
their paper they attribute this to the relative shapes of interaction potential curves of the transient SrCs molecule, similar to the explanation for far-wing asymmetries by the statistical theory of line broadening.

Since we saw in figure 2 and §3 that the basic width of the line is determined by the ac Stark width $2\Omega_a$, we feel that other features such as asymmetries should also be explainable on the basis of our present model, even though this may be a more complex procedure than the phenomenological framework of the quasimolecule picture. One immediate setback is the fact that the asymmetry in the first-order result (7) is opposite to that observed. This is seen by noting that since $\delta_a - \delta_b = 67$ cm$^{-1}$, it follows that as $\delta_a$ increases positively $|\delta_b|$ decreases and $(\delta_b^2 + \Omega_b^2)^{-1}$ increases. This has the effect of increasing $\sigma^{(1)}(\delta_a)$ over its Lorentzian value for $\delta_a > 0$ and decreasing it for $\delta_a < 0$, which is opposite to the measured asymmetry.

We must therefore search elsewhere for the explanation of the asymmetry in our present model, and we now look at the effects of higher orders of perturbation theory applied to the dipole-dipole term of the collisional interaction. We note first that if our first-order matrix element $<1f|d_\lambda|22>$ is nonvanishing, then any second-order term which still connects equal energy initial and final states, $\sim<1f|d_\lambda|jk><jk|d_\lambda|22>$, must vanish since no $|j>$ state of Sr can be dipole connected with both $<1>$ = Sr($5s^2\ 1S$) and $|2>$ = Sr($5s5p\ 1p$). Hence the third-order amplitude is the next nonvanishing term, and one can show that
\[ a^{(3)} = \frac{i}{4} \sum_{ii'} \sum_{jk} \sum_{\lambda \mu \nu} \sum_{j'k'} \frac{\Gamma_i^{1', i}}{c} B^{(a)}_{\mu} B^{(b)}_{\lambda} \]

\[ \times <lf| d_\lambda |jk> <jk| d_\mu |j'k'> <j'k'| d_\nu |22> \]

\[ \times \Gamma_{\lambda}^{(1)} (\omega f; jk) \Gamma_{\mu}^{(2)} (\omega j; j'k') \Gamma_{\nu}^{(2)} (\omega j'k'; 22 - \Delta_{ii'}) , \]

where \( \Delta_{ii'} \) is the composite Stark shift \( \delta_a + \delta_b + \alpha_{a1} + \alpha_{b1} \). Its full evaluation would be a horrendous task, but we would like to evaluate it at some level of approximation to see the kind of effects it would produce in interfering with \( a^{(1)} \). Let us therefore retain only one of the six components of \( d_\lambda \), namely \( d_3 = z_a z_b \), and chose the two pairs of intermediate states \( jk \) and \( j'k' \) that minimize the energy arguments of the \( \Gamma_{\lambda}^{(1)} (\Delta) \). Recall that the \( \Gamma_{\lambda}^{(1)} \)'s are linear combinations of the trajectory integrals \( T_n \), which tend to peak at \( \Delta = 0 \) (except for \( T_3 \)).

Since \( \varepsilon_{a1} + \varepsilon_{bf} \equiv \varepsilon_{a2} + \varepsilon_{b2} \) by the required overall energy conservation, let us take \( |j\rangle = |a2\rangle = Sr(5p) \) and \( |k\rangle = |b2\rangle = Cs(7p_{1/2}) \), and this would then require \( \varepsilon_{aj'} + \varepsilon_{bk'} \equiv \varepsilon_{a2} + \varepsilon_{b2} \), and \( |aj'\rangle \) and \( |bk'\rangle \) to be dipole connected with \( |a2\rangle \) and \( |b2\rangle \), respectively. A suitable choice would be \( |aj'\rangle = Sr(10s \, 1S) \equiv |a3\rangle \) and \( |bk'\rangle = Cs(6s_{1/2}) \equiv |b1\rangle \), with the energy defect

\[ \varepsilon_{aj'} + \varepsilon_{bk'} - (\varepsilon_{a2} + \varepsilon_{b2}) = 49 \, \text{cm}^{-1} . \]

With these simplifications made both in \( a^{(1)} \) and \( a^{(3)} \), their sum is
\[ a^{(1)} + a^{(3)} = -i \sum_{i' = 1}^{2} e^{i \text{Re}_{ij'} \tau^c B_{(a)} B_{(b)} \text{B}_{2i} \text{B}_{2i'} \text{B}_{3\text{i}}} <1f|d_{3}|22> \times \{ \Gamma^3(\xi_{i'}) - \frac{1}{4} <22|d_{3}|31><12|\} \times \Gamma^3(\xi_{22} - \Delta_{ij'}) \} \]  

When this is squared and only the cross term is retained (since the \(|a^{(3)}|^2\) term is of same order as the neglected \(a^{(1)}_a(a^{(5)}\) term), and the integration over \(\epsilon_{bf}\) is carried out, one can define a multiplicative correction factor, \(A\), in terms of which the new transition probability is

\[ P^{(1,3)} = A(\rho, \nu, I_a \delta_a) P^{(1)} \]

We do not attempt to write down here the very complicated form for \(A\), but it involves more complex \(\theta, \phi\) averages and additional trajectory integrals of the form

\[ \int_{-\infty}^{\infty} dt e^{\lambda t} \rho^{n}(vt) \frac{4-n}{R^{10}} \]

which reflect the higher-order effects in the dipole-dipole interaction. It satisfies the limit \(A \rightarrow 1\), showing the negligible effect of \(a^{(3)}\) for large impact parameters.

We have numerically evaluated \(A\) in the above indicated approximation and it is shown in figure 3. It is seen to be highly asymmetric in \(\delta_a\), always in the same direction as the experimentally observed asymmetry. Where it goes negative is obviously unphysical, since all transition probabilities must be positive, and this is probably an indication of the need for still higher orders in the perturbation expansion. This is consistent with \(A\)
FIGURE 3 The asymmetry factor $A$ for various values of impact parameter $\rho$ and intensity $I$.

becoming negative for small $\rho$, where the dipole-dipole interaction is largest. Because of these problems, we do not here attempt to evaluate corrected cross sections. Rather we simply conclude with the point that this very complicated asymmetry correction may well be large enough to overcome the milder asymmetry in the wrong direction which is contained in $P^{(1)}$. Figure 3 also indicates that the asymmetry in $A$ increases as $I$ increases, which is also in qualitative accord with the measurement of line shapes at $I = 0.9$ and $10$ MW/cm$^2$. From figure 3 it may also be seen that in addition to its effect on the line asymmetry, the factor $A$ also would tend to decrease the magnitude of $\sigma^{(1)}(\delta_a = 0)$ and the widths $\Delta_{1/2}^{(1)}$. 

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This might account partially for the lower experimental widths as given in figure 2. It would also mean that any absolute result for \( \sigma^{(1)}(\delta_a=0) \) in figure 1 should be revised somewhat downward.

6. INFLUENCE OF DIRECT TWO-PHOTON IONIZATION OF Cs

As mentioned earlier the laser wavelength that is resonant with Sr 5s^2 1S_0 - 5s5p 1P_1 is only 67 cm\(^{-1}\) from the Cs 6s 2S_{1/2} - 7p 2P_{1/2} transition energy. Since the basic width of the present collisional ionization cross section is the ac Stark broadening of the Sr resonance line, one would expect that at intensities such that \( \Omega_a \sim 67 \text{ cm}^{-1} \), or \( I \sim 7 \times 10^8 \text{ W/cm}^2 \), there is a very strong mixing of the two channels for the production of Cs\(^+\). In fact since the two-photon ionization of Cs proceeds throughout the laser pulse (of about 4 ns in the Bréchignac et al. experiment) it is desirable to estimate its magnitude as a possible background effect.

Let us estimate the magnitude of the ionization produced in the two-photon isolated Cs atom channel and compare this with our present estimate of the collisional ionization in the experimental range of intensities. This would then be another check on the reasonableness of our present cross section estimate. Let us use the absolute measurement of Morellec et al., which was \( \sigma_2 = 7 \times 10^{-49} \text{ cm}^2 \sec \) at a detuning of \( \delta_b = -67 \text{ cm}^{-1} \). At \( I = 10 \text{ MW/cm}^2 \) we find for the probability of two-photon ionization of Cs at the Sr resonance wavelength during a pulse of length 4 ns,

\[
P_2 = \sigma_2(\delta_b = -67 \text{ cm}^{-1}) \tau_p^2 \tau_p \approx 1.5 \times 10^{-6}.
\]
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We compare this with the probability of ionizing Cs via the present collisional process,

\[ P_c = \sigma(\delta_a=0) N_a \frac{\nu}{\tau} \rho \approx 8 \times 10^{-5} \]

or \( P_2 \approx 0.02 P_c \). This 2% background would be insignificant and indeed the wings of the Bréchignac et al. profile for \( \sigma(\delta_a) \) at 10 MW/cm\(^2\) do not drop below 2% of the central maximum. On the other hand, if we had chosen to normalize the measured \( \sigma(\delta_a=0) \) in figure 1 to a lower calculated magnitude, say for \( \rho_0 > 6 \), the resulting lower \( P_c \) would imply a larger fractional background from \( P_2 \), which was not observed. We take this as an added bit of experimental evidence to support our choice of \( \rho_0 = 4 \) in our model and the cross section magnitude given to the data points in figure 1.

REFERENCES