Electric field enhancement of depolarization of excited states

Munir H. Nayfeh, G. B. Hillard,* and W. L. Glab*

Department of Physics, University of Illinois at Urbana-Champaign, 1110 W. Green Street, Urbana, Illinois 61801 (Received 2 November 1984; revised manuscript received 6 August 1985)

Our calculations show that an external dc electric field can enhance by many orders of magnitude the depolarization cross section of highly excited atoms by charged particles. The enhancement is due to the fact that the electric field extends and shifts the electronic charge distribution along its direction, thus effectively creating a giant electric dipole in the atom.

There has been recent interest in the effect of dc electric fields on the structure^{1,2} and collisional interaction of highly excited atoms in the regions near and below the classical ionization limit E_c and near $E=0.^{3-10}$ Investigations of the effect of such fields on half collisions, that is, on the photoionization spectrum of atoms showed that the electric field induces ionizing states in the region near the photoionization threshold, whose electronic charge is highly polarized along the external field.^{1,2}

Understanding the properties of highly excited states in an external field near E_c is not only important for the understanding of the ionization of these atoms, but also important for the understanding of the inverse process, i.e., electron-ion recombination. This is due to the fact that these highly excited states are the gateway for such recombination. Because of the large degree of mixing of the *l* quantum numbers of highly excited states caused by the field it is expected that even a weak external field may affect this process. Several articles in Ref. 1 on dielectronic recombination—a type of electron-ion recombination which is of great current interest— indeed show that weak-field effects on Rydberg states can cause large changes in this collision cross section.⁸

In this paper we present a semiclassical investigation of depolarization of highly excited atoms by charged particles in the presence of dc electric fields. We find for the first time that the electric field enhances by many orders of magnitude the cross section for this process. This enhancement is due to the fact that the electric field extends the electronic charge distribution of the atom along its direction thus effectively creating a giant dipole moment in the atom which then interacts with the long-range field of the charge. The interaction is so long range that the excitation duration (pulse width) governs the time over which the interaction takes place. Even though this long-range dipole may live less than 10^{-11} s and the excitation pulses are less than 10 ns, the interaction is strong enough to cause appreciable depolarization even at number densities as low as 10⁸ cm³.

The size of this giant dipole in complex atoms can be a few thousand angstroms; however, only a few percent (~6% at 5 kV/cm) of the charge distribution of the final state molds into the giant dipole. In addition, this percentage depends very weakly on the strength of the electric field ($F^{1/4}$ dependence). Moreover, these giant dipole atoms are unstable with respect to ionization^{1,2} (a few pi-

coseconds lifetime). Our recent calculations and experiments, however, have shown that this percentage can be enhanced in atomic hydrogen.⁹ Moreover, contrary to complex atoms, it is possible to excite selectively giant dipoles in hydrogen with much longer lifetimes (on the order of 1 ns) at negative energies.⁹ These enhancements of the excitation strength and the lifetime which are only possible in atomic hydrogen would make it easier to experiment with Stark resonant states in which the highly excited electron spends appreciable time upfield (or downfield).

The dipole interaction is the longest range interaction possible between a point charge and a neutral atom and it can be expected to dominate scattering at large distances. Such an interaction has been used to describe scattering of electrons by highly polar molecules by representing the molecule in a first approximation as a rigid rotator possessing a dipole moment.¹⁰

Consider an atom in the presence of a uniform static electric field directed along the z axis, $\mathbf{F}=F\mathbf{\hat{z}}$, colliding with a charged particle of charge e. The Hamiltonian of the system is $H=H_0+V_c$ where $H_0=H_0'+V_s$ is the sum of the Hamiltonian of the unperturbed atom H_0' and its interaction with the static electric field V_s , and V_c is the collisional interaction between the charge and the atom. The effect of the field on the structure of the projectile is negligible.

The collisional effect will be calculated by solving for the density matrix of the atom plus Stark field described by H_0 for a single collision and then averaging over all collisions. The effect of the field on the velocity of the projectile is discussed later. We use basis states which are eigenstates of the sum of the unperturbed Hamiltonian and the interaction Hamiltonian of the Stark field. A semiclassical treatment of the problem of an electron in Coulomb and electric fields of comparable strength has recently been solved.1 We will give here the necessary features for our study. The nonrelativistic, timeindependent Schrödinger equation for a single spinless electron in combined Coulomb and electric fields $\mathbf{F} = F\hat{\mathbf{z}}$ in the z direction can be separated in the parabolic coordinate system $\xi = r + z$, $\eta = r - z$, $\phi = \tan^{-1}(y/x)$ by setting $\psi = u_1(\xi)u_2(\eta)\exp(im\phi).$

For an electron excited to an energy very near the ionization limit, the electric and Coulomb fields are of comparable importance in the determination of its motion.

<u>32</u> 3324

Thus neither field's effects may be treated as a perturbation. However, since the electron is in a highly excited state, a Wentzel-Kramers-Brillouin (WKB) treatment of its motion is applicable. Applying the transformation $\chi_1 = u_1 \sqrt{\xi}$ to the ξ equation of the separated Schrödinger equation and using the Langer approximation of $m^2 - 1 \rightarrow m^2$ gives the form of a one-dimensional Schrödinger equation with energy E/4 and effective potential energy

$$V(\xi) = -Z_1/2\xi + m^2/8\xi^2 + F\xi/8 .$$

Examination of this equation shows that for any energy the electron is bound in this coordinate, with classical turning points $\xi_1 \cong 0$ and $\xi_2 \neq 0$. A similar treatment in the η coordinate leads to another one-dimensional equation with energy E/4 and potential energy

$$V(\eta) = -Z_2/2\eta + m^2/8\eta^2 - F\eta/8 .$$

For energies greater than E_c classical escape occurs. The motions in the two coordinates are coupled by the condition on the separation parameters $Z_1+Z_2=1$. Applying the WKB method to the equation governing the motion in the ξ coordinate yields a Bohr-Sommerfeld quantization condition:

$$\int \{2[\frac{1}{4}E - V(\xi)]\}^{1/2} d\xi = (n_1 + \frac{1}{2})\pi,$$

where the limits of integration are ξ_1 and ξ_2 , n_1 is the quantum number associated with this motion. Similar quantization of the motion in the η direction gives the quantum number n_2 . Thus states are labeled by the three quantum numbers $(n_1, n_2, |m|)$. Moreover, for energies above E = 0, this picture predicts the presence of resonances with energies given by the properties of the motion in ξ .¹¹

This semiclassical treatment predicts a set of resonances for excited states of any m embedded in the continuum that is above E = 0, since the motion in ξ is bounded for all energies and m.¹ Each of these resonances is a superposition of a few parabolic states. The largest contribution comes from an opening of a new channel which may be labeled by quantum numbers: n_1 , $n_2=0$, and m, that is the Stark components which are shifted up the most (bluest components) where n_1 is the quantum number of the ξ motion. The resonance structure, therefore, comes from this new channel, where as the other channels contribute a smooth yield. The ratio of the difference to the average of the maximum and the minimum of the modulation defines the percentage of the height of the modulations or simply their heights which implies (by definition) that the maximum value that they can have is 200 percent. The height, as defined, reflects the partial cross section of the new channel at that energy. The height of the states decreases monotonically with increasing |m|. For a given sum $n_1 + |m| + 1$, the energies of the bluest components differ since $V(\xi)$ depends on |m|, with the variation depending also on the strength of the Stark field. Numerical calculations indicate that these differences are not larger than 5 cm^{-1} in the range of field strength appropriate for the occurrence and detection of this phenomenon (2-10 kV/cm). Because the collisional energies are much larger than 5 cm⁻¹, the variations in the energy of bluest components of a given sum $n_1 + |m| + 1$ but different n_1 and |m| and same $n_1 + |m| + 1$ can be neglected.

The electronic charge distributions of the bluest components are highly distorted. They are elliptical with the major diameter extending over thousands of angstroms, while the minor diameter extends over a few angstroms. The major diameter is defined by the turning points one of which is very close to the nucleus. The orientation of the orbit depends on |m|; for example for m=0, the orbit contains the z axis (direction of the electric field), and for large |m| it is almost normal to z. The far turning point for m=0 is given by $\xi=\sqrt{4/F}$ or $r\sim 1/\sqrt{F}$. Since for other m values, the centrifugal potential is negligible at large ξ we expect the turning points to differ from those of m=0 by very small amounts.

The distribution of the partial cross section of the various parabolic channels, at a given field strength, depend on the energy of the system. For the region below E = 0, the nonrelativistic hydrogenic treatment predicts that one channel dominates because the resonances are quite narrow there, whereas in the E > 0 region other channels may contribute. The above results derived from the nonrelativistic hydrogenic theory were modified in the case of complex atoms due to the additional electron-electronic core interaction. This interaction mixes the hydrogenic channels and their ionization rates, resulting in the modification of the distribution of the partial cross sections of the various parabolic channels.

The height or the partial cross section giving rise to the resonance structure of the hydrogenic Stark-induced resonances having giant dipoles in the region $E \ge 0$ is 15 percent of the underlying smooth ionization yield at 4 kV/cm.² In complex atoms, the interaction of the electron with the non-Coulombic ionic core results in a reduction of this percentage by the factor $\cos(2\pi\mu_l)$ where μ_l is the quantum defect of the *l*th final continum channel. If more than one channel is involved, then the analysis will have to involve a superposition of them.² Typically the height drops to 0-10%. The dependence of the height of the resonances on the electric field strength is a rather slow power law dependence of the form $F^{1/4}$.

Recently our studies on atomic hydrogen have shown, contrary to complex atoms, that the percentage of the charge distribution that can be molded into the giant dipole from the otherwise free electronic charge of the highly excited state (or alternatively the height of the resonances) can be enhanced dramatically.⁹ This can be done by using multistep excitation via resonant intermediate states instead of a single-step excitation from the ground state.

Because of near l degeneracy (the largest splitting is 0.365 cm⁻¹ in n=2), the n=2 states of hydrogen become effectively pure parabolic at fields as low as 10 kV/cm, having very distinct dipole charge distributions that are parallel or antiparallel to the Stark field, whereas the ground state, unaffected by the field, stays spherically symmetric. By appropriate choice of a multistep excitation process and choice of the intermediate states, the overlap and hence the excitation cross section of giant di-

poles extended either upfield or downfield can be greatly enhanced. We present here calculations of the photoionization cross section as a function of the quantum number n of the initial state and as a function of its parabolic quantum numbers n_1 , n_2 , and |m| (defined earlier) and give the dependence of the depth of the Stark-induced resonances on these quantum numbers.

The percentage of the depth of the broad modulations at $E \ge 0$ for excitation from the ground state, the n=2state, 100 and 010 states, and the n=3 states 200, 110, and 020 are presented in Table I. Moreover, the results for excitation from some spherical states are shown in the same table. These results show that for the n=2,100 initial state the predicted depth of modulation is more than twice that for the ground state, while that for the n=2,100 state is so small as to be practically unobservable. The situation is even more pronounced for the n=3states.

Comparison between excitation from initial spherical and parabolic states is interesting since the spherical case is applicable to excitation in complex atoms. Because of the large splittings between the various low-lying angular momentum states in complex atoms, the electric field cannot mix them completely and hence pure parabolic states cannot be prepared using laboratory field. Hydrogen, on the other hand, is a special case where the splittings are small enough to be overcome by the external field in n = 2, and higher. Our calculations in Table I show that for excitation from the ground states both cases give the same result as it should. However, for excitation from excited states we have drastic differences between the two cases. We find an enhancement of the depth of the Stark-induced modulation in the region $E \ge 0$ when the initial excited state is a pure m = 0 blue state, and disappear almost completely when the initial state is a pure m = 0 red state.

There is another important property that distinguishes the hydrogen system from all other atoms, that at E < 0but $E > -2\sqrt{F}$, one can populate quasibound states (with "giant dipoles"), whose potential barriers are large enough (tunneling small enough) to render their lifetimes quite long on the time scale of the experiment. Hydrogen is a special case, because these states are not mixed with less stable ones so their "giant dipoles" survive long enough to be studied.

The collisional mixing among the degenerate m states occurs by j·L coupling of the electronic angular momen-

TABLE I. Effect of initial state (from Ref. 9).

n	Parabolic states (n_1n_2m)			%	Spherical states	%
1	0	0	0	20.4	1s	20.4
2	1	0	0	57	2 <i>s</i>	20.4
	0	1	0	2.3	2 <i>p</i>	38
3	2	0	0	89.7	3 <i>s</i>	18.8
	0	2	0	0.3	3 <i>p</i>	39.6
	1	1	0	11.0	-	

tum j with the orbital angular momentum L associated with the relative motion of the colliding particles. Because the external electric field F is strong, then the velocity of precession of j about the field direction is greater than the velocity of precession of the orbital and spin angular momenta about j, hence an uncoupling takes place as in the Paschen-Bach effect. Therefore $j \cdot L = m'_s L$ $+m'_{l}L$ where m'_{s} and m'_{l} are the projection of the spin and angular momenta along L. Note that m'_1 is different from m used above for the projection of l along the electric field. Because we are interested in mixing of the Stark states we will only calculate the effect of the m'Linteraction here (where the subscript l is dropped from m'_l to simplify the notations). Thus we write V(t) = A[R(t)]Lm' where A(R) is a function of the interparticle distance R which depends on the nature of the interaction between the particles.

We consider here charges whose velocities are along the direction of the electric field, therefore the angular momentum of the collision L is perpendicular to the direction of F. As a result the interaction V(t) is not diagonal since the axis of quantization is taken along the direction of the Stark field; the Stark states are eigenstates of the projection of their orbital angular momenta along the direction of the field (*m* is a good quantum number).

Consider an atom with n distinguishable substates $|m\rangle$ where m is the projection of the angular momentum l along the electric field. If only the population probabilities of these states are of interest, then an n-dimensional vector of components given by the density of atoms in the various states is adequate. Such a vector can of course be expanded as a linear combination of n Cartesian basis vectors.

This population representation is not generally applicable especially when one is interested in representing the same physical phenomenon in terms of states defined with a different axis of quantization. Such a representation arises in the problem at hand; in these cases we use an $n \times n$ density matrix ρ instead of the above *n*-dimensional vector. The matrix, written in "Liouville space," is of n^2 dimensions. A spherical basis will be used in this space. The components of those are matrix elements of irreducible tensor operators T_{LM} . Possible symmetries of the system can be conveniently studied in the Liouville space because the multipoles of the system are just the coefficients of the expansion of the density matrix ρ in terms of T_{LM} :

$$\rho = \sum_{L,M} \rho_{LM} T_{LM} \ . \tag{1}$$

We use an impact parameter treatment where the position of the charge from the atomic nucleus is $R = (b^2 + v^2 t^2)^{1/2}$ where b is the impact parameter and v is the velocity of the charge. A cross section for relaxation or decaying of the multipole ρ_{LM} is defined as follows:¹²

$$\sigma_{LM} = -2\pi \int_{0}^{\infty} b \, db \, \Delta_{LM} , \qquad (2)$$

$$\Delta_{LM} = \sum_{\substack{n,n' \\ m,m'}} (-1)^{n-m} \begin{bmatrix} l & L & l \\ -n & M & n' \end{bmatrix} \begin{bmatrix} l & L & l \\ -m & M & m' \end{bmatrix} \times \langle n \mid S \mid m \rangle \langle n' \mid S \mid m' \rangle^{*} , \qquad (3)$$

where we have used the 3-j symbols. The S matrix is given by

$$S = \exp\left[-i\tilde{n}^{-1}\int_{-\infty}^{\infty} V(t')dt'\right].$$
(4)

Hence the matrix elements of S are given by

$$\langle n | S | m \rangle = \exp \left[-i\hbar^{-1} \int_{-\infty}^{\infty} V_{nm} dt' \right], \qquad (5)$$

$$V_{m} = \langle n | V | m \rangle = c \quad A(R) \qquad (6)$$

$$V_{nm} = \langle n \mid V \mid m \rangle = g_{nm} A(R) , \qquad (6)$$

where A(R) is the matrix element of the radial motion; it is a function of the internuclear distance only, and g_{nm} is the matrix element of the angular motion in θ .

We now discuss the physical meaning of the multipoles ρ_{LM} . The system of the *n* states is considered spherically symmetric if $\rho_{LM} = 0$ for all $L \ge 1$, i.e., $\rho_{LM} \ne 0$ for only L = 0. In this case all *n* states will be equally populated,

and the system is unpolarized. If any of the higher multipoles, $L \ge 1$ is nonzero, the system is polarized with the *n* states being unequally populated. The multipoles ρ_{1M} and ρ_{2M} for example designate the orientation and alignment of the system, respectively. If for any L, $\rho_{LM}=0$ for $|M| \ge 1$, that is it is nonzero for M=0 only, then the system is cylindrically symmetric; otherwise it is not cylindrically symmetric and it is said to have coherence.

To evaluate g_{nm} explicitly, we expand the states $|m\rangle$ and $|n\rangle$ in terms of the $|m'\rangle$ eigenstates. The mathematical determination of this expansion is rather laborious¹³

$$|m\rangle = \sum_{m'} \beta_{m'm}^{(l)}(\theta) |m'\rangle , \qquad (7)$$

where

$$\beta_{m'm}^{(l)}(\theta) \sum_{p=0}^{\infty} (-1)^p \frac{\left[(l+m')!(l-m')!(l+m)!(l-m)!\right]^{1/2}}{(l+m'-p)!(l-m-p)!(p+m-m')!p!} (\cos\frac{1}{2}\theta)^{2l-2p+m'-m} (\sin\frac{1}{2}\theta)^{2p-m'+m}$$
(8)

which is a homogeneous polynomial of degree 2l in $\cos \frac{1}{2}\theta$ and $\sin \frac{1}{2}\theta$. Note that $\beta_{m'm}^{(l)}(\theta)$ generates (finite) rotations about the axis that is perpendicular to the axes defining *m* and *m'*. For small values of *l* these polynomials have only a few terms. Taking $\theta = \pi/2$ gives $\cos \frac{1}{2}\theta = \sin \frac{1}{2}\theta = 1/\sqrt{2}$; thus

$$\beta_{m'm}^{(l)} = \sum_{p=0}^{\infty} (-1)^p \frac{\left[(l+m')!(l-m')!(l+m)!(l-m)! \right]^{1/2}}{(l+m'-p)!(l-m-p)!(p+m-m')!p!} (\frac{1}{2})^l .$$
⁽⁹⁾

For l=0, we only have m=m'=0 and $\beta_{00}^0=1$. On the other hand for l=1, m can be 1, 0, and -1, and m' can also be 1, 0, and -1. In this case and for $\theta=\pi/2$, the matrix $\beta_{m'm}^{(l)}$ is

$$\beta_{m'm}^{(l)} = \frac{1}{2} \begin{bmatrix} 1 & -\sqrt{2} & 1\\ \sqrt{2} & 0 & -\sqrt{2}\\ 1 & \sqrt{2} & 1 \end{bmatrix}$$
(10)

and thus the explicit expansions of the $|m\rangle$ states in terms of the $|m\rangle'$ states are the following:

$$|1\rangle = \frac{1}{2} |1\rangle' - \frac{1}{\sqrt{2}} |0\rangle' + \frac{1}{2} |-1\rangle',$$

$$|0\rangle = \frac{1}{\sqrt{2}} |1\rangle' - \frac{1}{\sqrt{2}} |-1\rangle',$$

$$|-1\rangle = \frac{1}{2} |1\rangle' + \frac{1}{\sqrt{2}} |0\rangle' + \frac{1}{2} |-1\rangle'.$$

(11)

The matrix elements g_{mn} can be calculated using Eqs. (7) and (9) and the fact that V is diagonal in the prime basis:

$$g_{mn} = \sum_{m'} m' \beta_{m'm}^{(l)} \beta_{m'n}^{(l)} .$$
 (12)

For example for l = 1, the explicit elements form the 3×3 matrix:

$$g_{mn} = \begin{bmatrix} 0 & \frac{1}{\sqrt{2}} & 0\\ \frac{1}{\sqrt{2}} & 1 & \frac{1}{\sqrt{2}}\\ 0 & \frac{1}{\sqrt{2}} & 0 \end{bmatrix}.$$
 (13)

We should note that this procedure is equivalent to the diagonalization of the collisional interaction, V(t). The diagonal collisional interaction V(t) implies that the various components of its eigenstates evolve independently, that is all the channels decouple. The same procedure is often made in the study of rotational excitation of molecules by collisions with atoms.¹⁴

We now turn to the calculation of the cross section. We will only calculate it for the case of l=1, the case that arises in one-photon excitation from s ground states. Substituting Eqs. (5) and (6) and Eq. (13) in Eq. (3) and taking L=1 gives $\Delta_{LM}=0$ for M=0,

$$\Delta_{LM} = \frac{4}{3} [\cos(g_{00}\alpha) - 1] \text{ for } |M| = 1 , \qquad (14)$$

and the following for the cross section for relaxation of the multipoles ρ_{11} :

$$\sigma_{11} = 2\pi \int_0^\infty b \, db [1 - \cos(g_{00}\alpha)] \,, \tag{15}$$

where $g_{00} = 1$, α is the phase factor accumulated during the collision:

$$\alpha = \frac{1}{\hbar} \int_{t_1}^{t_2} A(R) dt \tag{16}$$

and t_1 and t_2 are the times between which the interaction takes place. For cases where contributions come from large impact parameters such as in cases of long-range interactions, the duration of the excitation pulse may become comparable to or shorter than the time over which the interaction is effective, and hence become the limiting interaction time.

We are interested in the depolarization of states of low m quantum numbers, e.g., m = 0, 1, and 2. These are the states that usually get populated in the most frequently used radiative processes, one- and two-photon processes. The electronic charge distribution of these states is mostly elongated along the electric field. However, because of the large extent of the electronic charge, it is not appropriate to represent it as a dipole distribution. Thus we write for the interaction

$$A(t) = \frac{e^2}{4\pi\varepsilon_0 v} \{ -(t^2 + b^2/v^2)^{-1/2} + [(t + z_0)^{\theta}v)^2 + b^2/v^2]^{-1/2} \}, \quad (17)$$

where z_0 is the coordinate of the electron, and v is taken along the z axis. Thus for pulse width τ

$$\alpha = \frac{e^2}{4\pi\varepsilon_0 v\hbar} \ln \frac{(z_0/v+t) + [(z_0/v+t)^2 + b^2/v^2]^{1/2}}{t + \sqrt{t^2 + b^2/v^2}} \bigg|_0^{\tau} .$$
(18)

Let us examine α as a function of b and v for $\tau \simeq 5$ ns, and $z_0 \simeq 1000$ Å. Numerical evaluations show that α stays much less than one for velocities larger than 10⁸ cm/s regardless of the magnitude of b. At lower velocities, $v < 10^7$ cm/s, however, α exceeds one for a wide range of b. In the latter case the integrand in Eq. (15), $1 - \cos(g_{00}\alpha)$, usually oscillates rapidly about an average value of one out to a critical impact parameter b_c which of course depends on m and z_0 and beyond which it drops rapidly to zero. Hence¹² the cross section is given by $\sigma = \pi b_c^2$. The criterion (Weisskopf or Firsov approximation) that is customarily employed to define the boundary between both of these two regions is defined by $\alpha \simeq 1$ rad. An interesting limit of Eq. (18) is the low-speed limit when $z_0 \tau / v \ll b^2 / v^2$. In this limit, the general expression for α simplifies and takes the following form:

$$\alpha = \frac{e^2}{\hbar v} \ln(1 + z_0 v \tau / b^2) = \frac{e^2 z_0 \tau}{\hbar b^2} .$$
 (19)

Taking $\alpha = 1$, then in this limit the cross section becomes

$$\sigma_{11} = \pi b_c^2 = \frac{\pi e^2 z_0 \tau g_{00}}{\hbar} . \tag{20}$$

We note that the cross section is velocity independent at low velocities, hence the corresponding depolarization rates are proportional to the velocity. As we mentioned above at high velocities where α stays small, the $1-\cos(g_{00}\alpha)$ term stays small, hence resulting in a smaller cross section than that of the low-velocity regime.

When testing the low-velocity regime, one should have special consideration with the way the ions are injected or generated in the interaction region. One scheme for achieving this regime is via injection of the ions with their initial velocity opposite to the direction of the field. To avoid acceleration by the field over large distances before it interacts with the atomic system, the ions can be produced in the same region of space by photoionization of a low-pressure impurity using a laser beam that overlaps the beam that produces the highly excited atomic states. The density of ions can be varied by varying the partial density of the impurity or/and the intensity of the photoionizing radiation. Moreover, time-delay measurements between the production of the highly excited atoms and the ions can be made thus effectively, giving the means for variation of the average velocity. In fact, we expect to have some ions present in the interaction region that come from the photoionization of the highly excited atoms themselves.

The turning point of the electronic orbit is 1000 Å in the presence of 5 kV/cm. If we take $z_0 \simeq 1000$ Å, and $\tau = 5$ ns, then $\sigma \simeq 4 \times 10^{-5}$ cm². This is a much larger cross section than one expects from the interaction of an n = 20 state with charged particles in the absence of an electric field. In this case the interaction energy is given by $-C_4/R^4$ where $C_4 = \mu^2 e^2/(\hbar\Delta)$. For a matrix element of 1 D and an energy denominator Δ of 15000 cm⁻¹, we find $C_4 = 5 \times 10^{-58}$ Jm⁴. Hence the cross section for depolarization may be drawn from the corresponding α :

$$\alpha = \frac{1}{\hbar} \int C_4 / R^4 dt = 2C_4 / (\hbar v b^3) .$$
 (21)

This gives $b_c = (2C_4/\hbar v)^{1/3}$ and hence $\sigma = \pi (2C_4/\hbar v)^{2/3} \simeq 10^{-14} \text{ cm}^2$ for $v = 10^6$ cm/s.

Finally it is relevant to talk about the importance of this effect in the collision of neutrals with the excited state. When one is dealing with neutrals, the plane of collision can be in any direction relative to the direction of the electric field. Hence the procedure we used for diagonalization in the charged particle interaction where a single direction of relative velocity was used should be averaged over the direction of the plane of collision. Here, however, we will only calculate the cross section for a single plane of collision. The neutrals can be similar or dissimilar atoms or molecules. If we consider a nonpolar ground state atom of polarizability β at a distance R from the highly excited atom of dipole moment $\mu(F)$, then the induced dipole moment of the ground state and the interaction energy averaged over the possible orientations are βF , and $u = -\beta \mu^2 / R^6$ where F is the electric field produced by the excited atom at the location of the ground-state atom. Taking $\beta = r^3$ where r is of the order of a Bohr radius, gives $u = -\mu^2 r^3 / R^6$.

An estimate of the depolarizing cross section can be arrived at by calculating α for this case which we find to be $\alpha = (3\pi\mu^2 r^3)/(8\hbar vb^5)$. The corresponding cutoff for the impact parameter is calculated by taking $\alpha = 1$ making the cross section $\sigma = \pi (3\pi\mu^2 r^3/8\hbar v)^{2/5}$. Taking the velocity for neutrals to be $v = 2 \times 10^2$ m/s, r = 0.5 Å, and

3328

 $\mu = ez_0$ where $z_0 = 1000$ Å, we get $\sigma \simeq 1.5 \times 10^{-8}$ cm².

In conclusion we have shown that the presence of a few kV/cm dc electric field can enhance the depolarization cross section of excited states by charged particles by many orders or magnitude. Such enhancement is a result

*Permanent address: General Electric Company, Nela Park, Cleveland, OH 44112.

- ¹See, e.g., Atomic Excitation and Ionization in External Fields, edited by M. H. Nayfeh and C. Clark (Gordon and Breach, New York, 1985).
- ²See, e.g., D. A. Harmin, Phys. Rev. A 26, 2656 (1982); E. Luc-Koenig and A. Bachelier, J. Phys. B 13, 743 (1980); see, e.g., R. R. Freeman, N. P. Economou, G. C. Bjorklund, and K. T. Lu, Phys. Rev. Lett. 41, 1463 (1978).
- ³W. A. Huber and C. Bottcher, J. Phys. B **13**, L399 (1980).
- ⁴V. L. Jacobs, J. Davis, and P. C. Kepple, Phys. Rev. Lett. **37**, 1390 (1976).
- ⁵A. Burgess and H. P. Summers, Astrophys. J. 157, 1007 (1969).
- ⁶D. S. Belic, G. H. Dunn, T. J. Morgan, D. W. Mueller, and C. Timmer, Phys. Rev. Lett. **50**, 339 (1983).
- ⁷K. LaGattuta and Y. Hahn, J. Phys. B 15, 2101 (1982); Phys. Lett. 84A, 468 (1981); Phys. Rev. Lett. 51, 558 (1983).
- ⁸See articles in Ref. 1 by P. Dittner et al., J. Mitchell et al., J.

of the large elliptical distortion (eccentricity of near unity) of the atomic electronic charge.

This work was supported by the National Science Foundation by (NSF) Grant No. NSF PHY 81-09305.

Kohl et al., G. Dunn et al., and J. Tanis et al.

- ⁹M. H. Nayfeh, K. Ng, and D. Yao, in *Laser Spectroscopy VII*, edited by T. W. Hänsch and Y. R. Shen (Springer, Berlin, 1985), p. 71; W. L. Glab and M. H. Nayfeh, Phys. Rev. A 31, 530 (1985); W. L. Glab, K. Ng, D. Yao, and M. H. Nayfeh, *ibid.* 31, 3677 (1985).
- ¹⁰See for example, B. Jaduszliwer, A. Tino, P. Weiss, and B. Bederson, Phys. Rev. Lett. **51**, 1644 (1983); H. Massey, Proc. Cambridge Philos Soc., **28**, 99 (1931); N. Lane, Rev. Mod. Phys. **52**, 29 (1980); D. W. Norcross and L. A. Collins, Adv. At. Mol. Phys. **18**, 344 (1982).
- ¹¹A. R. P. Rau, J. Phys. B 12, L193 (1979).
- ¹²W. E. Baylis, in *Progress in Atomic Spectroscopy*, part B, edited by W. Hanle and H. Kleinpoppen (Plenum, New York, 1978), pp. 1227-1297.
- ¹³U. Fano and L. Fano, Basic Physics of Atoms and Molecules (Wiley, New York, 1959), pp. 363-373.
- ¹⁴D. Secrest, J. Chem. Phys. **62**, 710 (1975).