Further studies of H^- photodetachment in electric fields

P. G. Harris, H. C. Bryant, A. H. Mohagheghi, and C. Tang
The University of New Mexico, Albuquerque, New Mexico 87131

J. B. Donahue, C. R. Quick, and R. A. Reeder*
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

Stanley Cohen
Cohen Mechanical Design, Broomall, Pennsylvania 19008

W. W. Smith
The University of Connecticut, Storrs, Connecticut 06268

J. E. Stewart
Western Washington University, Bellingham, Washington 98225

Carol Johnstone
Fermilab, Batavia, Illinois 60510
(Received 11 September 1989; revised manuscript received 22 January 1990)

The results are presented of further studies of the effects of electric fields on the photodetachment cross section of H^-, for both π and σ polarization. Data are fitted to the theory of Rau and Wong [Phys. Rev. A **37**, 632 (1988)], which predicts a modulation about the zero-field cross section. Agreement is good for π polarization, but the predicted profile for σ polarization does not appear to match the data quite as well.

The recent experimental verification[1,2] of the theoretical prediction[3,4] of electric-field-induced modulations in the photodetachment cross section of H^- near the single-electron threshold spurred further theoretical efforts to describe more accurately the shape of the cross section, as a function of photon energy, in static electric fields.[5–7] (An earlier sighting of this effect, in the photodetachment cross section of Rb^-, was not recognized as such until recently.)[8,9] We give here the results of a further investigation, with two important differences from that reported previously: first, this experiment was carried out in a well-determined electric field; and second, emphasis was focused on the region very close to the zero-field threshold, where any effects due to the residual polarized neutral hydrogen atom should be felt most strongly. These results are fitted to the theory of Rau and Wong.[5] In addition, we present data from an earlier experiment[10] in which the field strengths were considerably higher, but the light was exclusively σ polarized.

The experimental method was similar to that previously described in some detail.[2] A relativistic ($\beta = 0.786$) beam of H^- ions, produced at the high-resolution atomic beam (HIRAB) experimental area at the Clinton P. Anderson Meson Physics Facility (LAMPF) of Los Alamos National Laboratory was allowed to intersect a Nd:YAG (yttrium aluminum garnet) laser beam. Varying the angle of intersection of the two beams, by means of a system of mirrors mounted on a turntable as shown in Fig. 1, changes the center-of-mass photon energy according to the relativistic Doppler-shift formula

$$E = E_0\gamma(1+\beta\cos\alpha) , \tag{1}$$

where β and γ are the usual relativistic parameters, α is the angle of intersection of the laser and particle beams such that $\alpha = 0$ is head on, and E_0 is the laser photon energy in the lab (1.164 88 eV). Thus the cross section for photodetachment can be measured as a function of photon energy.

In order to produce a uniform electric field, two polished stainless-steel plates were mounted, 1 cm apart, on either side of the interaction volume. With a potential difference of up to 60 kV between these plates, barycentric electric fields of up to 96 kV/cm became available. Both π (electric vector of laser light parallel to external electric field) and σ (perpendicular) polarizations were obtainable in this manner.

This method was in contrast to that used in the previous experiment, where the laser light was passed through the pole pieces of an electromagnet in order to obtain both π and σ polarizations. The relativistic transformation of the magnetic field produced a large electric field in the barycentric frame. The magnetic field, however, was nonuniform, and its strength was not very well known. The electric plates had the additional advantage that their electric field was not angle dependent, unlike the field produced by the electromagnet, which was fixed with respect to the laser beam. Other differences are

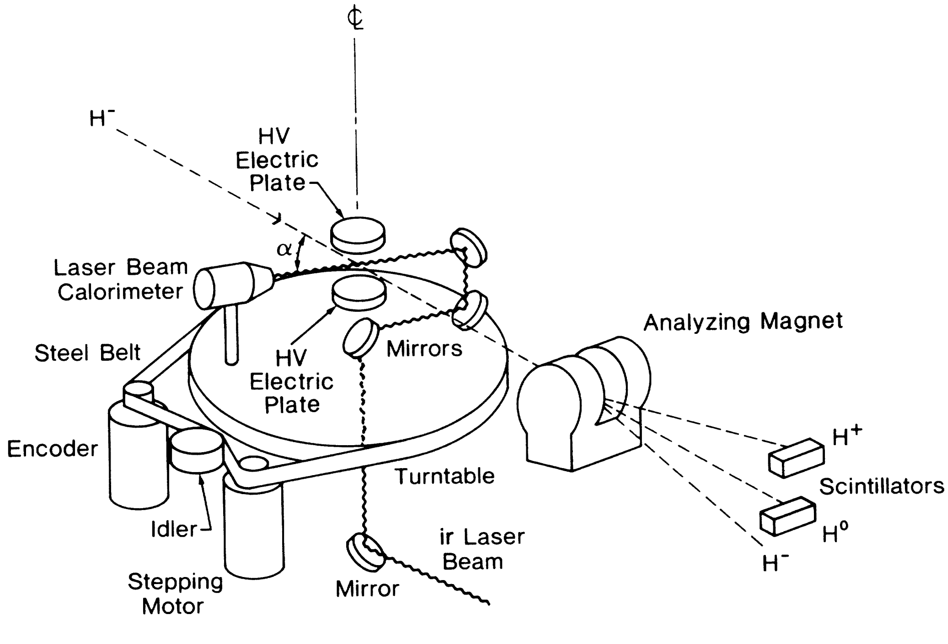

FIG. 1. Schematic of experimental apparatus.

that, in the previous experiment, the laser was focused with a cylindrical lens into the plane of the laser and particle beams, primarily in order to let it pass through the pole pieces of the magnet; and the laser was not operated in Q-switched mode as was the case in the most recent experiment, so the peak power was much lower.

In the 1985 experiment, the electric field was also produced by the relativistic transformation of a magnetic field; however, the magnet consisted of a pair of Helmholtz coils, which were energized in very short pulses in coincidence with the laser; thus high fields were obtained for short periods of time, but the geometry of the system permitted only σ polarization of the light.

The neutral hydrogen atoms from photodetachment were separated magnetically from the primary H^- beam and detected, downstream of the interaction region, in a scintillator, also shown in Fig. 1.

The resulting yields were converted into relative cross sections by applying a multiplicative factor, $\sin\alpha/(1+\beta\cos\alpha)$, which allows for the relativistic variation of intensity with angle as well as the change in overlap volume of the two beams. The energy scale for the 1988 (electric field) experiment was calibrated by fitting a series of near-threshold curves at zero field—as shown in Fig. 2—to find the angle of the threshold itself; it was also checked by exciting transitions in neutral hydrogen from $n=4$ to $n=12, 13, 14$, and 15. The uncertainty in energy is less than 1 meV.

In the absence of any electric field, the photodetachment cross section depends upon photon energy E according to a power law

$$\sigma = \frac{16\pi}{3(137)} \frac{E_0^{1/2}(E-E_0)^{3/2}}{E^3} \frac{1}{1-k_B r_{\text{eff}}} . \tag{2}$$

E_0 here represents the threshold energy, 0.7542 eV; $k_B^2 = 2E_0$, and the effective range, r_{eff}, of the potential is $2.646a_0$. Du and Delos[7] point out that the factor $1/(1-k_B r_{\text{eff}})$, numerically equal to 2.65, was absent from the paper (although included in the calculation) of Rau and Wong;[5] it was also omitted from our previous paper.[2]

We may make an estimation of the absolute cross section: with a pulsed H^- beam crossing a pulsed laser beam, the number of photodetachments per pulse is given by

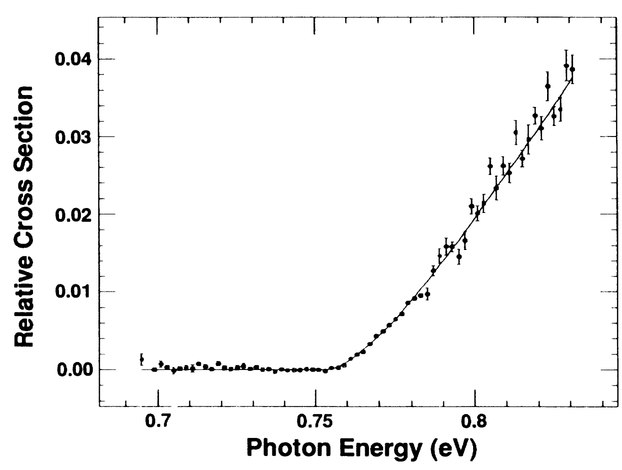

FIG. 2. H^- photodetachment cross section at threshold in zero electric field. Five runs are combined here. The solid line is a simplex fit to the theoretical threshold power law, as given in the text. The fit has a χ^2/ν value of 1.9. The error bars are statistical only; they represent the standard deviation of the mean number of signal counts per laser shot for each angle. The apparent sudden changes in the sizes of the error bars occur because not all runs span the entire photon energy range.

$$N = V_0 T_0 \rho \sigma \Phi \left[\frac{1+\beta\cos\alpha}{\sin\alpha} \right] , \tag{3}$$

where V_0 and T_0 are the volume (at $\alpha=90°$) and time, respectively, of interaction of the pulses, ρ is the number density of H^- in the pulse, σ is the cross section, and Φ is the photon flux. The photon flux Φ and particle density ρ are assumed constant over the volume V_0 and time T_0 of integration. Using our estimated typical values of $N=30$ counts per pulse at a center-of-mass photon energy of 0.82 eV, with $V_0=6\times10^{-2}$ cm^3, $T_0=10^{-9}$ s, $\rho=500$ H$^-$/cm^3, $\alpha=140°$, and a photon flux of $1.6\times10^{27}/(\text{cm}^2\,\text{s})$, we obtain $\sigma=10^{-18}$ cm^2, in comparison with the expected value of 7×10^{-18} cm^2. This discrepancy is a cause of minor concern; however, the volume of overlap, the particle density, and the photon flux have only been estimated approximately, each perhaps within a factor of 2.

With π-polarized light, "ripples" appear on this cross section. This effect may be understood in a simple way by considering just the wave packet of the ejected electron, spreading preferentially "upstream" and "downstream" in the electric field (σ polarization, in contrast, would produce preferential spreading transverse to the field); the part of the wave packet that spreads "upstream" will reflect from the potential barrier of the external field and will return to interfere with the wave spreading "downstream," thus producing the characteristic interference pattern.[1]

The frame-transformation approach of Rau and Wong[5] predicts that the zero-field cross section will be modified by a modulating factor $H^F(k)$,

$$\sigma^F(k) = \sigma^{F=0} H^F(k) = \sigma^{F=0}(k) \int_{-\infty}^{k^2/2} d(q^2/2) |U_{q1}^F|^2 , \tag{4}$$

where

$$U_{q1}^F = (3\pi/k^3)^{1/2} (16F)^{1/6} \text{Ai}'[-q^2/(2F)^{2/3}] . \tag{5}$$

The expression for the cross section ignores the final state interaction of the electron with the neutral atom.[6] This is a good approximation, since the electron-scattering phase shifts are small ($\delta \le 0.01$ rad) for H^- in this energy range. The overall effect of δ is to replace $H^F(k)$ by

$$H^F(k)/[\cos^2\delta + H^F(k)^2 \sin^2\delta] ,$$

producing a fractional deviation of the order of 10^{-5}. The simpler formulation has therefore been used in this analysis.

Du and Delos[7] have produced an equivalent formula that may be closely approximated by a simple analytic function for energies somewhat above the zero-field threshold; to be specific, they define a parameter

$$x = \frac{2^{1/3}(E-E_0)}{F^{2/3}} , \tag{6}$$

and give as the cross section

$$\sigma = 0.3604 \frac{F}{E^3} D(x) a_0^2 , \tag{7}$$

where

$$D(x) = \int_{-\infty}^{x} \left[\frac{d}{dz} \text{Ai}(-z) \right]^2 dz , \tag{8}$$

Ai(z) being the standard Airy function. For sufficiently large x,

$$D(x) \approx \frac{1}{4\pi} [\tfrac{4}{3} x^{3/2} + \cos(\tfrac{4}{3} x^{3/2})] . \tag{9}$$

The difference between the function $D(x)$ defined in (8) and the analytic approximation (9) is 2% for $x=2$, and 0.2% for $x=4$ (corresponding to photon energies of 0.78 eV and 0.80 eV, respectively, at a field of 64 kV/cm). Thus, the analytic formula is extremely good once past the first "ripple".

Figures 3–6 show the results of some typical runs, taken with electric fields of 64, 80, 83.2 and 96 kV/cm. Individual sets of data from all runs are tabulated in full in a Los Alamos report.[11] The theoretical prediction is represented in each figure by the solid line; the data shown include error bars that are the standard deviation of the mean signal at a given angle (energy). The confidence levels of the fits are low; 29% for the data taken at 64 kV/cm, and < 1% for the other data; we suspect that this indicates that the error bars are too small, since they do not allow for systematic effects such as changes in the temporal overlap of the laser and particle beams, and a fluctuating background from "spray" with the particle beam. Visually, the curves appear to match well. However, it is interesting to note a tendency for the theoretical curve to lie above the data in the "toe" region, and to lie below it at lower energies as the cross section tends to zero.

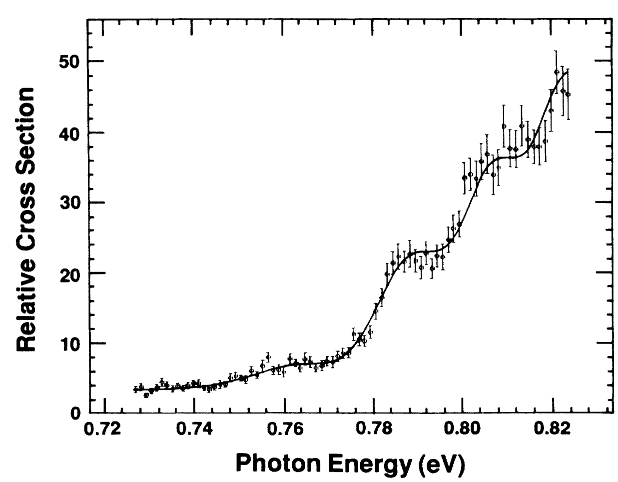

FIG. 3. Relative photodetachment cross section near threshold in an electric field of 64 kV/cm with π-polarized laser light; four data sets combined. The solid line represents the best fit to the theory of Rau and Wong; $\chi^2/\nu = 1.08$.

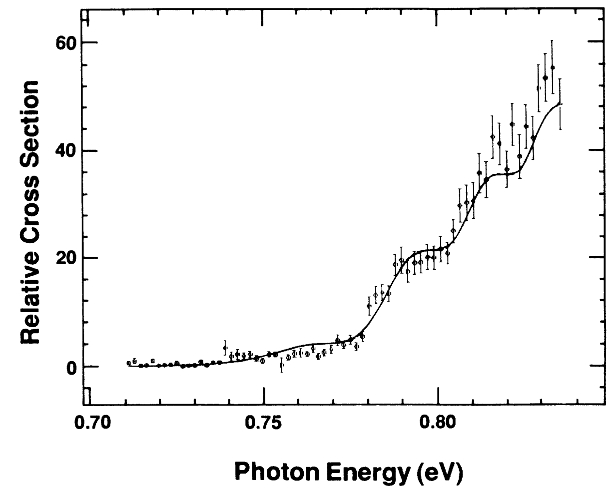

FIG. 4. Relative photodetachment cross section, with π polarization, near threshold in an electric field of 80 kV/cm; three data sets combined. $\chi^2/\nu = 2.32$.

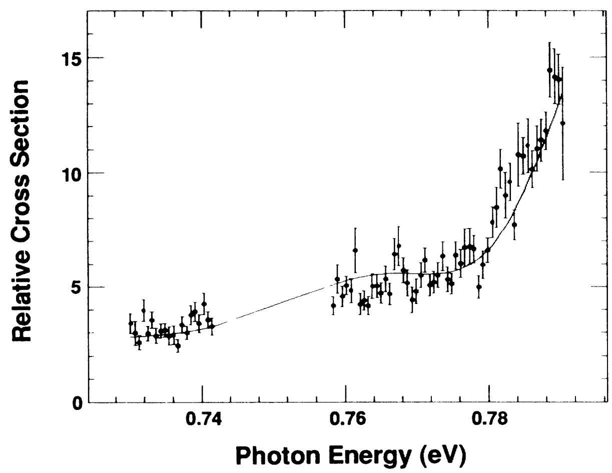

FIG. 6. Relative photodetachment cross section, with π polarization, near threshold in an electric field of 96 kV/cm; one data set. The absence of data in the region between 0.741 and 0.758 eV was caused by an equipment failure. $\chi^2/\nu = 1.98$.

The apparent nonzero cross section below threshold is due in part to back-reflected photons from the laser-beam calorimeter, which intersect the H^- beam at a very forward angle and thus have a barycentric energy well above threshold. The cross section at these high energies is, however, relatively flat, and provides a constant background over the small angular range of the near-threshold measurements.

It should be pointed out that magnetic effects cannot be ruled out; the pure electric field in the laboratory produced a magnetic field in the barycentric frame of up to 250 G, which, although small, may not be negligible. The Landau levels[12] (resonances due to quantized cyclotron orbits) are unlikely to be the cause of such effects, as their energy spacing is 2.9 μeV at 250 G, which is well below our resolution of 0.5 meV.

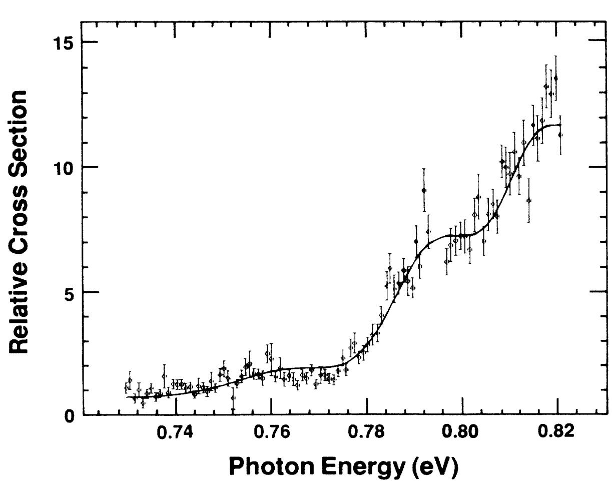

FIG. 5. Relative photodetachment cross section, with π polarization, near threshold in an electric field of 83.2 kV/cm; three data sets combined. $\chi^2/\nu = 1.87$.

Although Du and Delos[7] do not give a formula for the photodetachment cross section in σ-polarized light, Rau and Wong[5,6] predict that there are once again oscillations on the zero-field cross section. These, however, have a considerably smaller amplitude than their π-polarization counterparts, and therefore may only be seen in very strong fields, such as those of our 1985 experiment. The modulating factor in this case is again $H^F(k)$,

$$\sigma^F(k) = \sigma^{F=0} H^F(k) = \sigma^{F=0}(k) \int_{-\infty}^{k^2/2} d(q^2/2) |U_{q1}^F|^2 , \tag{10}$$

where, for σ polarization,

$$U_{q1}^F = (3\pi/2k)^{1/2} (4/F)^{1/6} (1 - q^2/k^2)^{1/2} \times \mathrm{Ai}[-q^2/(2F)^{2/3}] . \tag{11}$$

Unfortunately, the oscillation cannot be visualized in a simple manner as can the π-polarization ripples, except that the largest effect is a field-induced increase in the cross section in the threshold region due to the lowering of the potential barrier. Figure 7 shows the near-threshold photodetachment cross section as a function of photon energy, for an electric field of 1.32 MV/cm. The solid line is a fit [using the computer routine MINUIT (Ref. 13)] to the theory of Rau and Wong; the dashed line is the zero field cross section. Although data above 0.9 eV are available, these were not used as there is a possibility that our detectors may have begun to saturate somewhere above this value. Data below 0.9 eV are believed to be reliable.

The energy scale in the σ-polarization experiment[10] is less well determined than for the π-polarization runs, and it may be offset by a few meV. This offset was therefore allowed to vary as a free parameter in the fit, the result indicating that a shift of -8.7 meV was appropriate; the data shown already incorporate this energy shift. The only other free parameters were the amplitude and a background.

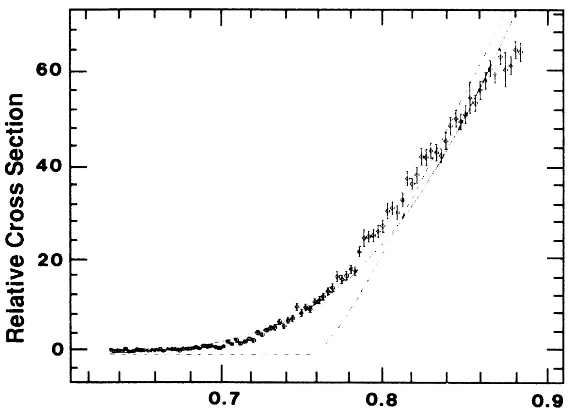

FIG. 7. Relative photodetachment cross section in an electric field of 1.32 MV/cm, with σ-polarized light. The solid line is the best fit to the theory of Rau and Wong. $\chi^2/\nu=6.5$; the dashed line is the zero-field cross section.

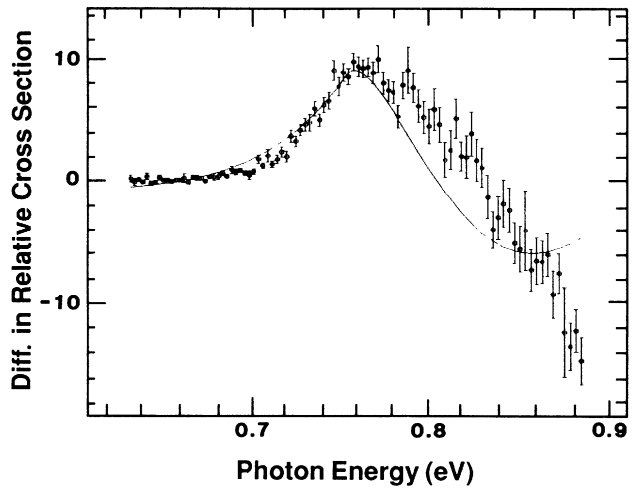

FIG. 8. Difference between relative photodetachment cross section in an electric field of 1.32 MV/cm and the calculated zero-field cross section. The solid line is again the best fit to the theory of Rau and Wong.

It appears that the theoretically predicted shape of the cross section oscillates about the data. This, with the oscillatory nature of the cross section itself, becomes more apparent in Fig. 8, which shows the difference between the data and the calculated zero-field cross section. (The difference between the fitted theory and the zero-field cross section is represented by the solid line.) Since the fit only had three free parameters, namely the scale factor, the background, and a small energy scale offset, we must conclude that the actual shape of the cross section is different from that predicted, although the difference is small. The fit has a χ^2 of 6.5 per degree of freedom. As with the π-polarized light, the theoretical curve appears to be too high just above threshold, and to be too low at lower energies; in fact, the data do not seem to fall off with the exponential tail that would be expected from a model of field-assisted tunneling below the zero-field threshold.

Data also exist for lower fields; these are published elsewhere.[10] The effects are seen most clearly in this high-field example. The lower-field data also have fewer points below the threshold, reducing the quality of the fits.

Fabrikant[4] also predicts oscillations for both π- and σ-polarization photodetachment in a field of strength F. He introduces a parameter β,

$$\beta=\frac{2}{3}\frac{m_e^2 v^3}{Fe\hbar}, \quad v=\frac{\hbar k}{m_e} . \tag{12}$$

In terms of this parameter, the formulas for the photodetachment cross section are

$$\begin{aligned} \sigma_\pi &= \sigma_0\left[1+\frac{\cos\beta-1}{\beta}+O(\beta^{-2})\right], \\ \sigma_\sigma &= \sigma_0\left[1+\frac{\Gamma(1/3)}{4\beta^{1/3}}-\frac{\sin\beta}{\beta^2}+O(\beta^{-3})\right], \end{aligned} \tag{13}$$

for π and σ polarizations, respectively, with $l=m=0$. These "ripples" appear to be different in nature from those of Rau and Wong.[5] Unfortunately, Fabrikant's theory only claims to be valid for $\beta>2\pi$, which, in our case, implies photon energies greater than about 1 eV; in this region, our data may be unreliable due to saturation, and we therefore can make no claims for the validity of either theory.

In summary, the theoretical model of Rau and Wong[5] of the threshold photodetachment cross section of the H^- ion in an electric field appears to be accurate for π-polarized light, but the profile seems to fit less well with σ-polarized light in much stronger electric fields. The theoretical prediction of Fabrikant[4] differs from that of Rau and Wong, but its range of validity lies beyond currently available data.

This work was supported by the Division of Chemical Sciences, Office of Basic Energy Sciences, Office of Energy Research, U.S. Department of Energy, and was also funded in part by the U.S. Army Strategic Defense Command. The authors are grateful for the assistance of Jim Knudson, Bob Sander, George Kyrala, and Vincent Yuan, and for technical support from Jim Hontas, Lawrence Quntana, and others at the University of New Mexico and at Los Alamos.

*Present address: The University of New Mexico, Albuquerque, NM 87131.

[1]H. C. Bryant, A. Mohagheghi, J. E. Stewart, J. B. Donahue, C. R. Quick, R. A. Reeder, V. Yuan, C. R. Hummer, W. W. Smith, S. Cohen, W. Reinhardt, and L. Overman, Phys. Rev. Lett. **58**, 2412 (1987).

[2]J. E. Stewart, H. C. Bryant, P. G. Harris, A. H. Mohagheghi, J. B. Donahue, C. R. Quick, R. A. Reeder, V. Yuan, C. R.

Hummer, W. W. Smith, and S. Cohen, Phys. Rev. A **38**, 5628 (1988).
[3]W. P. Reinhardt, in *Atomic Excitation and Recombination in External Fields,* edited by M. H. Nayfeh and C. W. Clark (Gordon and Breach, New York, 1985), p. 85.
[4]I. I. Fabrikant, Zh. Eksp. Teor. Fiz. **79**, 2070 (1980) [Sov. Phys.-JETP **52**, 1045 (1980)].
[5]A. R. P. Rau and H. Wong, Phys. Rev. A **37**, 632 (1988).
[6]H. Wong, A. R. P. Rau, and C. H. Greene, Phys. Rev. A **37**, 2393 (1988).
[7]M. L. Du and J. B. Delos, Phys. Rev. A **38**, 5609 (1988).
[8]C. H. Greene, N. Rouze, Z. Phys. **9**, 219 (1988).
[9]P. Frey, F. Breyer, and H. Hotop, J. Phys. B **11**, L589 (1978).
[10]J. E. Stewart, Ph.D. dissertation, University of New Mexico, 1987 (presented at Los Alamos, Report No. LA-11152-T, 1987).
[11]P. G. Harris, H. C. Bryant, A. Mohagheghi, C. Tang, J. B. Donahue, C. R. Quick, R. A. Reeder, S. Cohen, W. W. Smith, and J. E. Stewart, Los Alamos National Laboratory Report No. LA-UR-89-322, 1989.
[12]L. D. Landau and E. M. Lifshitz, *Quantum Mechanics: Non-Relativistic Theory,* 2nd ed. (Addison-Wesley, Reading, MA, 1965), p. 424.
[13]F. James and M. Roos, Comput. Phys. Commun. **10**, 343 (1975).