PHYSICAL REVIEW A VOLUME 42, NUMBER 7 1 OCTOBER 1990

Short-range potential model for multiphoton detachment of the H^- ion

W. Becker, S. Long, and J. K. McIver
Center for Advanced Studies and Department of Physics and Astronomy, University of New Mexico, Albuquerque, New Mexico 87131

(Received 1 June 1990)

The H^- ion is modeled by an effective one-electron potential specified by a regularized three-dimensional δ-function potential. With the help of quasienergy methods, the multiphoton-detachment rate of this model ion by a plane-wave field of arbitrary elliptical polarization can be expressed as a one-dimensional integral that has to be evaluated numerically. The results exhibit a pronounced dependence on the polarization of the laser: circular polarization yields a rate that depends smoothly on frequency and intensity with barely noticeable thresholds. In contrast, linear polarization generates marked thresholds as well as structure in between. All of the calculated thresholds closely obey the Wigner threshold law. There is good agreement with the available data where the ponderomotive energy is kept constant while the frequency varies.

The negative hydrogen ion is a unique system for the investigation of multiphoton detachment. The ion has no excited bound states and, in contrast to ionization of atoms, the detached electron is not subject to the long-range Coulomb potential. These two circumstances greatly facilitate the comparison between experiment and theory. Moreover, the H^- ion is the simplest two-electron system, and this latter aspect is of strong fundamental interest far beyond multiphoton physics.

Due to its importance in astrophysics, the H^- ion was studied in the thirties. The first accurate calculations of the one-photon detachment were performed by Chandrasekhar[1] and carried to a high degree of complexity by Geltman.[2] This line of work consisted of more and more elaborate variational two-electron calculations. However, it was noticed by Ohmura and Ohmura[3] and by Armstrong[4] that a zero-range one-electron approximation yielded very satisfactory agreement with the experimental measurement by Smith and Burch[5] of the one-photon detachment cross section for photon energies up to 2 eV (as well as with the result of the variational calculations). The latest measurements by Sharifian[6] confirm this agreement up to photon energies of several eV. The fact that a one-electron description based on a short-range potential gives quite good results is perhaps not too surprising, since the two electrons in H^- are equivalent, there is no excited bound state, and the lowest continuum resonances do not occur below about 9.5 eV. For a review of H^- one-photon detachment up to 1974, see Risley;[7] more recent two-electron calculations have been carried out by Broad and Reinhardt,[8] Fink and Zoller,[9] Park *et al.*,[10] and Saha.[11]

Recently, the interest in the H^- ion has been revived in the context of multiphoton physics. Tang *et al.*[12] measured total multiphoton detachment rates of H^- ions using a relativistic H^- beam colliding with a linearly polarized CO_2 laser beam.[12,13] The angle between the two beams was continuously varied corresponding to a photon energy between 0.13 and 0.42 eV in the rest frame of the H^- ion. A characteristic feature of these experiments is that the ponderomotive potential $e^2\langle \mathbf{A}^2\rangle/2m \sim I/\omega^2$ (where $\mathbf{A}$, I, and ω are the vector potential, intensity, and frequency of the laser, respectively) is a constant independent of the angle, since it is a relativistic invariant. The magnitude of the ponderomotive potential is $\lesssim 0.1$ eV which is a sizable fraction of the detachment energy, $|E_0| = 0.754$ eV. The detachment rates exhibit a pronounced threshold behavior whenever a new channel (two-photon detachment, three-photon detachment, etc.) opens up. Moreover, there is evidence of structure between the thresholds.

These experiments raise the interesting question of whether multiphoton detachment of the H^- ion (as opposed to one-photon detachment) is still predominantly a one-electron process. For multiphoton ionization of rare gases, all evidence to date seems to speak[14] for sequential ionization, i.e., one electron at a time is involved in the process of ionization. The situation may be different, however, for H^- whose energy spectrum is so vitally dependent on the presence of two electrons. In order to obtain a preliminary answer to these questions we present in this Rapid Communication calculations of the multiphoton detachment rate in a one-electron zero-range potential model and compare our results with preliminary experimental findings of Tang *et al.*[12] We employ the three-dimensional regularized δ-function potential

$$V(\mathbf{r}) = \frac{2\pi}{\kappa m}\delta(\mathbf{r})\frac{\partial}{\partial r}r \tag{1}$$

that supports one bound state with energy $|E_0| = \kappa^2/2m$ and wave function $(\kappa/2\pi)^{1/2}e^{-\kappa r}/r$ (we use units such that $\hbar = c = 1$). This potential was first introduced by Fermi[15] in the context of neutron scattering in hydrogenous substances and has been referred to as the Fermi contact potential. The regularizing operator $(\partial/\partial r)r$ makes sure that the potential $V(\mathbf{r})$ applied to the wave function of the bound state yields a well-defined expression. Because of this regularization the potential is nonlocal and, in fact, repulsive when acting on continuum states. However, its salient features are the ultrashort range, the existence of just one bound state, and the fact that it only depends on the one parameter $\kappa = (2m \times |E_0|)^{1/2}$. The δ-function potential (1) allows for a comparatively simple, largely analytical calculation of quasienergies and quasienergy wave functions in the presence of an external plane-wave field in the dipole approximation for circular polarization[16] and arbitrary elliptic polarization.[17]

The first nonperturbative calculation of multiphoton detachment of H^- was carried out by Reiss.[18] It was based on what is now referred to as the Keldysh-Faisal-Reiss (KFR) approximation and used the same zero-range potential wave function for the ground state that we discussed above. A similar approach including corrections to the KFR approximation and using a cutoff Coulomb potential in place of the zero-range potential was recently followed by Mu.[19] Shakeshaft and Tang[20] applied Floquet theory to multiphoton detachment of a Yukawa potential modeling the H^- ion. Seven-photon detachment of H^- was considered by Mercouris and Nicolaides[21] in terms of two-electron Floquet theory with some evidence for electron correlation effects. Our calculations agree well (within 20%) with the results of Shakeshaft and Tang[20] within the (quite limited) range of parameters where we could compare. The qualitative agreement with the Keldysh-type calculations[18,19] is also good. This is not surprising, since it has been shown[22] that for the δ-function potential (1) the Keldysh approximation is very good.

In solving the model we will use the electric-field gauge so that we will look for quasienergy solutions of the Schrödinger equation

$$i\frac{\partial}{\partial t}\Psi(\mathbf{r},t)=\left[-\frac{1}{2m}\nabla^2+\frac{2\pi}{\kappa m}\delta(\mathbf{r})\frac{\partial}{\partial r}r-e\mathbf{r}\cdot\mathbf{E}(t)\right]\Psi(\mathbf{r},t)\ . \tag{2}$$

Quasienergy wave functions have the Floquet form

$$\Psi(\mathbf{r},t)=e^{-iEt}\phi(\mathbf{r},t)\ , \tag{3}$$

where $\phi(\mathbf{r},t)=\phi(\mathbf{r},t+2\pi/\omega)$ has the period of the external field and

$$E=\mathrm{Re}(E)-\frac{i}{2}\Gamma \tag{4}$$

is the quasienergy. For the quasienergy of the ground state, $\mathrm{Re}(E)=-|E_0|+\Delta$, with Δ a real level shift which is small for not too high intensities. The real quantity Γ is the total multiphoton detachment rate which we want to determine. When the external field $\mathbf{E}(t)$ is turned off, Γ and Δ vanish.

For an external field with general elliptic polarization, viz.,

$$\mathbf{E}(t)=\omega a[\sin(\omega t)\hat{x}-\xi\cos(\omega t)\hat{y}]\ , \tag{5}$$

the following integral equation holds for the quasienergy E,

$$\left[\frac{-E'}{|E_0|}\right]^{1/2}-1=\left[\frac{i}{4\pi|E_0|}\right]^{1/2}\int_0^\infty d\tau\,\tau^{-3/2}\exp(-iE'\tau)\left\{\exp\left[-i\frac{4U}{\omega^2\tau}\sin^2\left[\frac{\omega\tau}{2}\right]\right]J_0(z(\tau))-1\right\}\ , \tag{6}$$

where

$$z(\tau)=\frac{(ea)^2}{2m\omega}(1-\xi^2)\sin\left[\frac{\omega\tau}{2}\right]\times\left[\cos\left[\frac{\omega\tau}{2}\right]-\frac{2}{\omega\tau}\sin\left[\frac{\omega\tau}{2}\right]\right]\ , \tag{7}$$

$$E'=E-U\ , \tag{8}$$

and

$$U=\frac{(ea)^2}{4m}(1+\xi^2) \tag{9}$$

is the ponderomotive (quivering) energy. For circular polarization, $\xi=1$, we have $z(\tau)=0$, and Eq. (6) simplifies considerably. Equation (6) is exact for circular polarization. In general, the quasienergy is exactly obtained by equating to zero an infinite determinant with elements much like the right-hand side of Eq. (6). Hence for any polarization other than circular, Eq. (6) is an approximation whose relative accuracy has been estimated[17] to be of the order of $(\omega/4|E_0|)^2$. As long as Γ and Δ are small compared with $|E_0|$ we may, on the right-hand side of Eq. (6), replace E by $-|E_0|$, i.e., the value in the absence of the field. The total detachment rate is then given by

$$\Gamma=\mathrm{Re}\left\{\left[\frac{4(|E_0|+U)}{i\pi}\right]^{1/2}\int_0^\infty d\tau\,\tau^{-3/2}\exp[i(|E_0|+U)\tau]\left\{\exp\left[-i\frac{4U}{\omega^2\tau}\sin^2\left[\frac{\omega\tau}{2}\right]\right]J_0(z(\tau))-1\right\}\right\} \tag{10}$$

which we evaluate numerically.

For circular polarization, $\xi=1$, the integral representation (10) of the total detachment rate can be rewritten as the double sum[16]

$$\Gamma=4(|E_0|+U)\sum_{s=1}^{\infty}\sum_{n=-s}^{s}(-1)^{n+s}\left[\frac{4U(|E_0|+U)}{\omega^2}\right]^s\frac{\{[n\omega/(|E_0|+U)]-1\}_+^{s+1/2}}{(2s+1)(s-n)!(s+n)!}\ , \tag{11}$$

where $x_+=x$ for $x>0$ and $x_+=0$ for $x<0$.

The one-photon detachment rate is obtained from the integral (10) as the lowest-order term in an expansion with respect to the intensity, that is, the parameter U. For one-photon detachment, the rate is independent of the polarization, so that we can use the sum (11). The corresponding one-photon detachment cross section is

$$\sigma_1=\frac{\omega}{I}\Gamma_1=\frac{8}{3}r_0\lambda\frac{|E_0|^{1/2}(\omega-|E_0|)^{3/2}}{\omega^2}\ , \tag{12}$$

with $r_0=e^2/mc^2$ the classical electron radius and λ the laser wavelength. This agrees with the result of effective range theory as given in Eq. (5) of Ref. 4 except for the effective range factor. The cross section (12) should be multiplied by a factor of 2 to account for the presence of two equivalent electrons. The following numerical results include neither this factor of 2 nor the effective range factor[4] $f^2=1.3$.

The results of our numerical calculations are shown in Figs. 1–3. The integral (10) was evaluated numerically for linear polarization while the sum (11) was used for circular polarization. In each case the detachment rate Γ is plotted versus the photon energy in the rest frame of the H^- ion. Figure 1 shows the detachment rate for both circular and linearly polarized laser fields for three different laser intensities. The thresholds are visible for each channel opening and the thresholds are shifted by the ponderomotive potential as the laser intensity is increased. It is interesting to note that the structure is largely independent of the intensity. For higher intensities the thresholds and other features are shifted to higher photon energies and the overall slope decreases but the shape of the detachment-rate curves show little change while the intensity is increased by nearly 1 order of magnitude. The detachment rate for a linearly polarized field is greater than that for a circularly polarized field except in the one-photon region where they are equal and in portions of the two- and three-photon regions where the rate for circular polarization is greater. This is in qualitative agreement with results derived using the Keldysh model.[18,19] Figures 2 and 3 are magnified views of the two- and three-photon thresholds, respectively, for a linearly polarized laser. In each case the threshold is at $\hbar\omega=[|E_0|+(ea)^2/2m]/n$, where n is the number of photons in the new channel. After the three-photon threshold the detachment rate rises 1 order of magnitude to a peak and then decays 30% before reaching the two-photon threshold. For a ponderomotive potential of 0.05 eV the detachment rate peaks at 6.8×10^{10} per sec at a photon energy of 0.32 eV. This is in quantitative agreement with the data from Tang *et al.*[12] The rate also peaks after the two-photon threshold but the peak is not as pronounced, the rate decays 10% before the one-photon threshold. This is also in agreement with the experimental results.

When the photon energy is slightly above the n-photon threshold the detachment rate for the H^- ion should obey the Wigner threshold law[23]

$$\Gamma\propto E_e^{l+1/2}=(n\omega-|E_0|-U)^{l+1/2}\ , \tag{13}$$

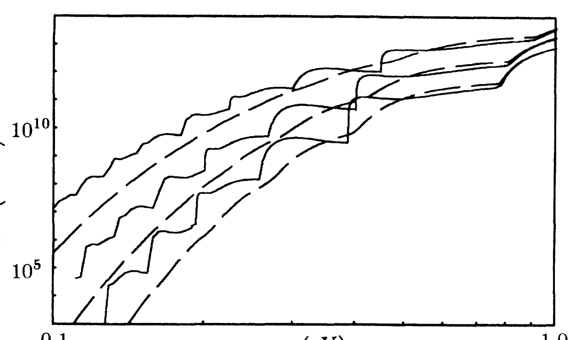

FIG. 1. Detachment rate Γ as a function of the photon energy ω in the ion frame for fixed ponderomotive energies; top 0.15 eV, 0.05 eV, and 0.02 eV bottom; for linear (solid) and circular (dashed) polarization.

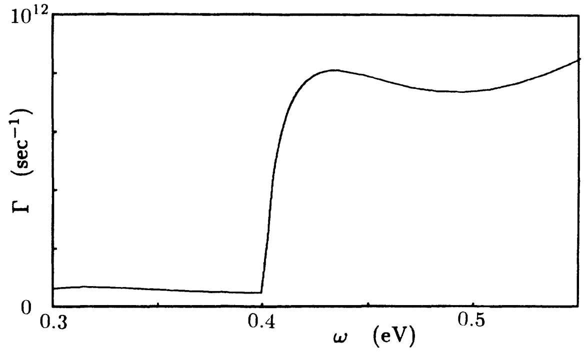

FIG. 2. The two photon threshold in the detachment rate Γ for linear polarization as a function of the photon energy ω in the ion frame with the ponderomotive energy fixed at 0.05 eV.

where l is the angular momentum, and E_e is the kinetic energy of the detached electron. For n-photon detachment in a circularly polarized field it is easy to see that the detachment rate obeys Wigner's law. The angular momentum of the electron is n and the lowest-order term in the expansion of the detachment rate [Eq. (11)] is $\Gamma\propto(n\omega-|E_0|-U)^{n+1/2}$. For a linearly polarized field the final electron can have any even angular momentum between zero and n for n even or any odd value between one and n for n odd. When the excess energy is small enough only the lowest-order term will contribute. The contribution of the new channel to the detachment rate will be proportional to $(n\omega-|E_0|-U)^{3/2}$ for photon energies just above an odd threshold and proportional to $(n\omega-|E_0|-U)^{1/2}$ just above an even threshold. This is clearly visible in Fig. 4 which is a plot of the logarithmic derivative of the detachment rate near the first six thresholds.

Finally, Fig. 5 shows the detachment rate for fixed frequency as a function of the ponderomotive energy which is proportional to the intensity. For linear polarization, pronounced thresholds are visible at the ponderomotive energies $(ea)^2/4m=n\omega-|E_0|$, where the n-photon channel closes. In each case if one follows the graph from lower to higher intensities the channel closing is anticipated by a precipitous drop in the detachment rate. A better way of looking at the situation is to read the graph from higher to lower intensities. Then a new channel opens up whenever the intensity goes through a threshold, and the rate tem-

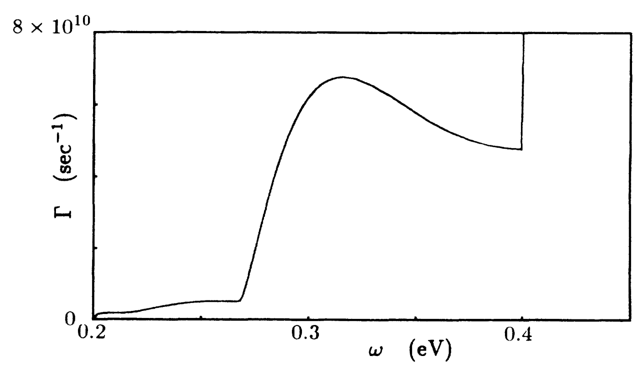

FIG. 3. The three-photon threshold in the detachment rate Γ for linear polarization as a function of the photon energy ω in the ion frame with the ponderomotive energy fixed at 0.05 eV.

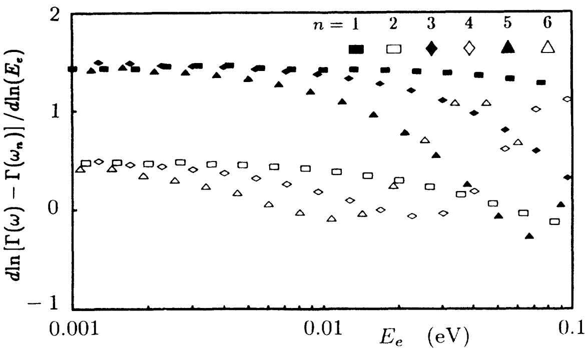

FIG. 4. The logarithmic derivative of $\Gamma(\omega)-\Gamma(\omega_n)$ vs the kinetic energy of the detached electron, $E_e = n\omega - |E_0| - U$, in a linearly polarized field, where ω_n is the energy of the n-photon threshold.

porarily rises. In no event does the rate follow a simple I^n power law. The same behavior has been observed in a one-dimensional numerical simulation[24] and in a Floquet approach.[25] No thresholds are visible for circular polarization. This is again readily understood with the help of the Wigner threshold law. The channel closings in the figure correspond to $n \geq 3$, and therefore, for circular polarization, to $l \geq 3$. The onset of channels with angular momenta this high is hardly noticeable.

Note added. After this work was completed we became aware of several closely related references. Faisal, Filipowicz, and Rzążewski,[26] in a time-dependent solution of the potential (1) with a circularly polarized field, also observed the smooth dependence of the total detachment rate as a function of intensity and conjectured a more ragged behavior for linear polarization. Geltman[27] calculated n-photon detachment rates for a zero-range potential. His results exhibit many of the features observed in our model. A quantitative comparison is cumbersome since he displays his results as generalized multiphoton cross sections. Finally, Dörr, Potvliege, Proulx, and

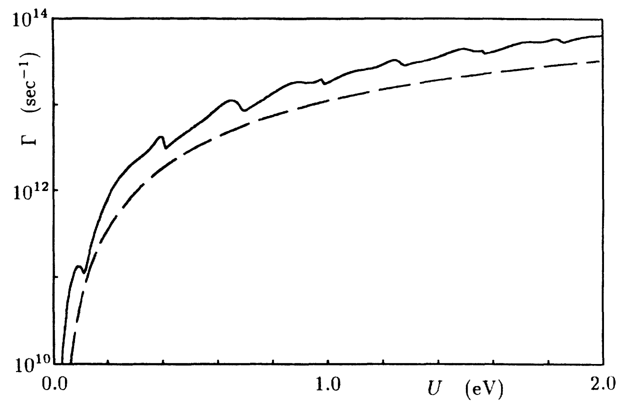

FIG. 5. Detachment rate Γ for linear (solid) and circular (dashed) polarization as a function of the ponderomotive potential U for a fixed photon energy, $\omega = 0.29$ eV, in the ion frame.

Shakeshaft[28] presented an extensive treatment of H^- multiphoton detachment comparing the exact Floquet results (for a suitable Yukawa potential) with Keldysh-type approximations for one and two electrons. They corroborated the, in general, excellent agreement between the exact Floquet results and the Keldysh approximations. We compared their Yukawa-potential-based Floquet results with ours and found agreement within a factor of 2 between our Fig. 1 and their Figs. 1–3 over a wide range of frequencies ($0.11\ \text{eV} \leq \hbar\omega \leq 0.85\ \text{eV}$) and intensities such that the total detachment rate varied over 10 orders of magnitude.

We would like to thank H. C. Bryant and C. Y. Tang for useful discussions and for showing us preliminary results from their experiment. This work was partially supported by the Division of Chemical Sciences, Office of Basic Energy Sciences, Office of Energy Research, U.S. Department of Energy.

[1]S. Chandrasekhar, Astrophys. J. **102**, 223 (1945).
[2]S. Geltman, Astrophys. J. **136**, 935 (1962).
[3]T. Ohmura and H. Ohmura, Phys. Rev. **118**, 154 (1960).
[4]B. H. Armstrong, Phys. Rev. **131**, 1132 (1963).
[5]S. J. Smith and D. S. Burch, Phys. Rev. **116**, 1125 (1959).
[6]M. H. Sharifian-Attar, Ph.D. thesis, University of New Mexico, 1977 (unpublished).
[7]S. J. Risley, in *Atomic Physics IV, Proceedings of the Fourth International Conference on Atomic Physics, 1974,* edited by G. zu Putlitz, E. W. Weber, and A. Winnacker (Plenum, New York, 1975).
[8]J. T. Broad and W. P. Reinhardt, Phys. Rev. A **14**, 2159 (1976).
[9]M. G. F. Fink and P. Zoller, J. Phys. B **18**, L373 (1985).
[10]C-H. Park, A. F. Starace, J. Tan, and C. D. Lin, Phys. Rev. A **33**, 1000 (1986).
[11]H. P. Saha, Phys. Rev. A **38**, 4546 (1988).
[12]C. Y. Tang *et al.* (unpublished).
[13]C. Y. Tang, P. G. Harris, A. H. Mohagheghi, H. C. Bryant, C. R. Quick, J. B. Donohue, R. A. Reeder, S. Cohen, W. W. Smith, and J. E. Stewart, Phys. Rev. A **39**, 6068 (1989).
[14]N. B. Delone and M. V. Fedorov, Usp. Fiz. Nauk **158**, 215 (1989) [Sov. Phys. Usp. **32**, (1989)].
[15]E. Fermi, Ric. Sci. **7**, 13 (1936).
[16]I. J. Berson, J. Phys. B **8**, 3078 (1975).
[17]N. L. Manakov and A. G. Fainshtein, Zh. Eksp. Teor. Fiz. **79**, 751 (1980) [Sov. Phys. JETP **52**, 382 (1981)].
[18]H. R. Reiss, Phys. Rev. A **22**, 1786 (1980).
[19]X. Mu, Phys. Rev. A **42**, 2944 (1990).
[20]R. Shakeshaft and X. Tang, Phys. Rev. A **36**, 3193 (1987).
[21]Th. Mercouris and C. A. Nicolaides, J. Phys. B **21**, L285 (1988).
[22]W. Becker, J. K. McIver, and M. Confer, Phys. Rev. A **40**, 6904 (1989).
[23]U. Fano and A. R. P. Rau, *Atomic Collisions and Spectra* (Academic, New York, 1986).
[24]W. G. Greenwood and J. H. Eberly, Phys. Rev. A (to be published); W. G. Greenwood, Bull. Am. Phys. Soc. **35**, 1175 (1990).
[25]M. Dörr, R. Potvliege, and R. Shakeshaft, Bull. Am. Phys. Soc. **35**, 1183 (1990).
[26]F. H. M. Faisal, P. Filipowicz, and K. Rzążewski, Phys. Rev. A **41**, 6176 (1990).
[27]S. Geltman (unpublished).
[28]M. Dörr, R. M. Potvliege, D. Proulx, and R. Shakeshaft, this issue, Phys. Rev. A **42**, 4138 (1990).