Dichotomy of the Hydrogen Atom in Superintense, High-Frequency Laser Fields

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We study the behavior of atomic hydrogen in a monochromatic radiation field of high frequency ω and high intensity I, when its structure depends only on the parameter $\alpha_0 = I^{1/2}\omega^{-2}$ a.u., and when multiphoton ionization is quenched. At large α_0 the ground-state binding energy undergoes a drastic reduction. This is coupled to an unprecedented stretching of the (oscillating) electron wave function, culminating in its separation into two parts (dichotomy) for $\alpha_0 > 50$ a.u.

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Impressive advances in laser technology have made possible the generation of high-frequency radiation, in the uv and beyond, of extreme intensity, in some cases in excess of 1 a.u. $I_0 = 3.5\times10^{16}$ W/cm^2.[1] A nonperturbative high-frequency theory was recently developed by Gavrila and Kaminski[2] to study electron-atom scattering in such fields.[3] The formalism has been extended since to cover atomic structure and ionization under similar conditions.[4] We now present the first accurate calculation within the theory for the structure of atomic hydrogen.[5]

We assume that the radiation can be represented by a monochromatic plane wave (frequency ω), linearly polarized (real polarization vector $\mathbf{e}$), and take the electrodynamic potentials in the dipole approximation $\mathbf{A} = a\mathbf{e}\times\cos(\omega t)$, $\phi = 0$. The semiclassical Schrödinger equation in the momentum gauge, describing the interaction dynamics in the laboratory frame of reference, was transformed by Kramers (see also Henneberger)[6] into the form[7]

$$\{(1/2m)\mathbf{P}^2 + V[\mathbf{r}+\boldsymbol{\alpha}(t)]\}\psi = i\hbar\,\partial\psi/\partial t, \tag{1}$$

by applying the time-dependent translation $\mathbf{r}\rightarrow\mathbf{r}+\boldsymbol{\alpha}(t)$. Here $V(\mathbf{r})$ is the atomic potential and

$$\boldsymbol{\alpha}(t) = \alpha_0\mathbf{e}\sin(\omega t), \quad \alpha_0 = -(ea/mc\omega), \tag{2}$$

represents the quiver motion of a classical electron in the field. In atomic units (Bohr radii)

$$\alpha_0 = I^{1/2}\omega^{-2}, \tag{3}$$

where I is the (time-averaged) beam intensity. Equation (1) characterizes, in fact, the dynamics in a moving frame of reference which follows the quiver motion of the classical electron, and which we shall call the "Kramers reference frame."

By application of the Floquet method of solution,[2,4] Eq. (1) was cast into a system of coupled differential equations in coordinate space for the Floquet components of the wave function Ψ, containing a (in general complex) quasienergy parameter E. The system was supplemented by appropriate boundary conditions to describe the steady decay by multiphoton ionization of an initial state *in the field.* An iterative procedure of solution was devised, valid at sufficiently high frequencies. To lowest order in the iteration (the high-frequency limit), the set of differential equations reduces to a single one

$$[(1/2m)\mathbf{P}^2 + V_0(\alpha_0,\mathbf{r})]\psi_0 = E\psi_0, \tag{4}$$

for the zeroth Floquet component ψ_0, and $\psi(\mathbf{r},t) \cong \psi_0(\mathbf{r})\exp(-iEt/\hbar)$. Equation (4) contains the "dressed potential" $V_0(\alpha_0,\mathbf{r})$, which depends on ω and I only through α_0.[8] It has obviously real eigenvalues E, showing that *in the high-frequency limit the atom is stable* against multiphoton ionization.[9,10] The *frequency condition* under which this should hold was shown to be $\omega \gg |E_0^m(\alpha_0)|$, where $E_0^m(\alpha_0)$ is the lowest eigenvalue having the same magnetic quantum number m as the initial state of the atom in the field. Equation (4) was obtained earlier by Henneberger,[6] and by Gersten and Mittleman using other approaches.[11]

For the Coulomb case $V(r) = -e^2/r$, the dressed potential has the form

$$V_0(\alpha_0,\mathbf{r}) = -(2e^2/\pi)(r_+r_-)^{-1/2} \times K[2^{-1/2}(1-\hat{\mathbf{r}}_+\cdot\hat{\mathbf{r}}_-)^{-1/2}], \tag{5}$$

where $\mathbf{r}_\pm = \mathbf{r}\pm\alpha_0\mathbf{e}$ (the origin of the coordinates is kept at the center of V) and K is the complete elliptic integral of the first kind.[12] V_0 has $r^{-1/2}$ type singularities at the points characterized by $+\alpha_0\mathbf{e}$ and $-\alpha_0\mathbf{e}$, and a logarithmic singularity along the segment in between; it is axially symmetric around an axis of direction $\mathbf{e}$ passing through the origin and has even parity. Thus, only the magnetic quantum number m associated with this axis

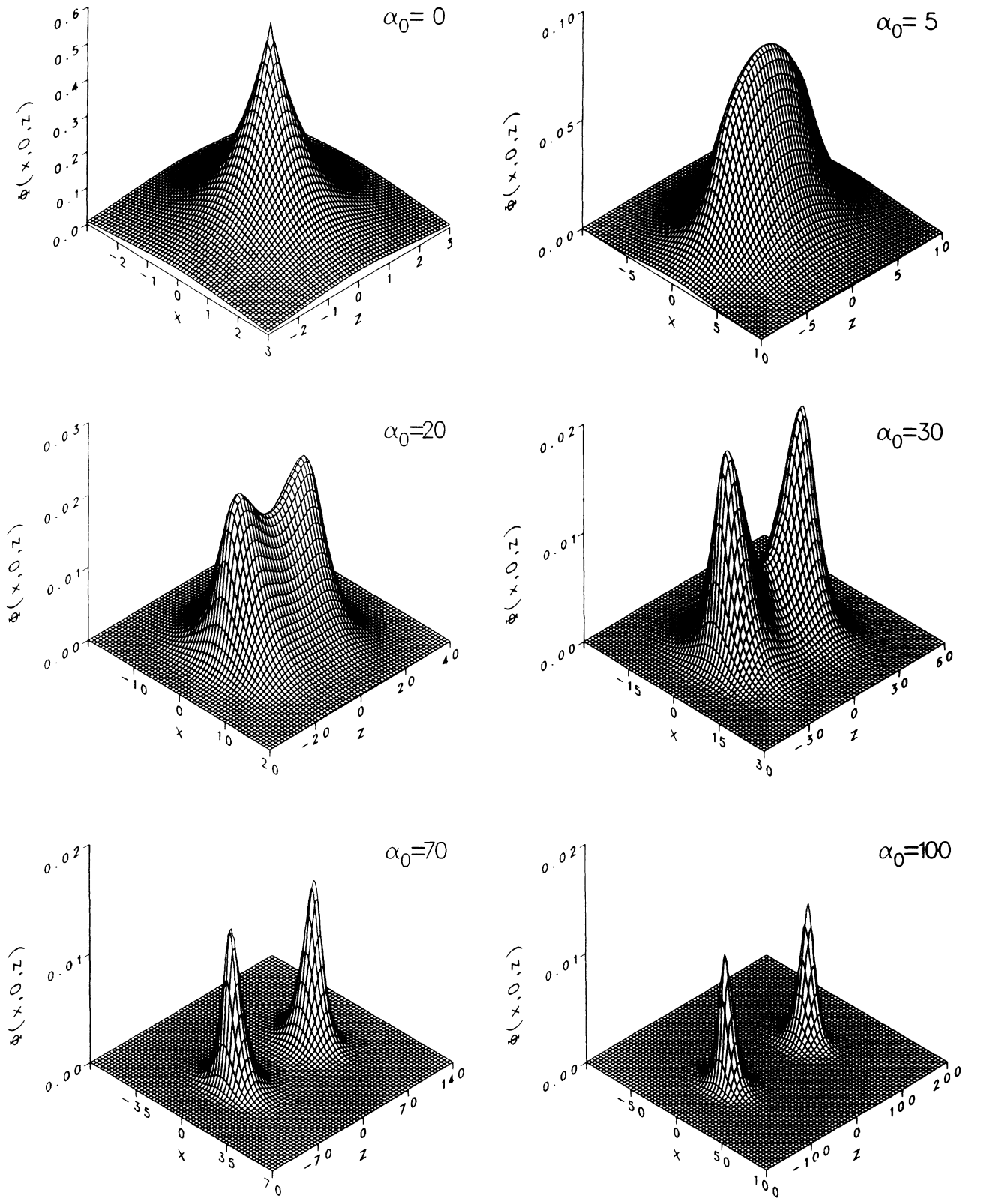

FIG. 1. Evolution of the normalized ground-state $(1s)\sigma_g$ wave function of atomic hydrogen in the Kramers frame of reference, for increasing α_0, Eq. (3). $\phi(x,0,z)$ is the wave function in an xz plane, where the z axis is chosen along the axis of symmetry of the dressed potential, Eq. (5), and the x axis is arbitrary. The length unit is a_0 (the Bohr radius), and the unit for $\phi(x,0,z)$ is $a_0^{-3/2}$.

and the parity π (g or u) remain good quantum numbers. Because the symmetry is identical to that of homonuclear diatomic molecules ($D_{\infty h}$), the same type of classification is adopted for the states. For example, the ground state, which evolves from the $1s$ state of the unperturbed atom, becomes a σ_g state ($m=0$, even parity) and will be denoted by $(1s)\sigma_g$.

The eigenvalue problem, Eq. (4), was solved by diagonalization of the Hamiltonian matrix in a multicenter Gaussian basis. The symmetry classes investigated were σ_g, σ_u, π_g, and π_u.[13] The lowest-lying level in each symmetry class was obtained to five significant figures accuracy.

We report here only on the ground state $(1s)\sigma_g$. The eigenvalues at various α_0 are given in Table I. Striking is the *drastic decrease in binding energy* with increasing α_0: at $\alpha_0=30$ a.u. (a value attained in experiment[13]), it has already dropped by a factor of about 10 with respect to the unperturbed value.[14] This suggests a strong distortion of the atom, which we now analyze. To this end we display the evolution of the (normalized) ground-state wave function $\psi_0=\phi(x,y,z)$ with increasing α_0. Because of the axial symmetry of the problem, it is sufficient to give its values in a plane passing through the symmetry axis z, as done in Fig. 1. When α_0 increases from 0 (case of the unperturbed atom), the wave function ϕ elongates in the z direction, following the elongation of the line of singularities of the dressed potential (see Fig. 1 of Ref. 2). However, as α_0 approaches 20 a.u., a saddle sets in, and by $\alpha_0=30$ a.u. two pronounced maxima appear around the endpoints $\pm\alpha_0\mathbf{e}$ of the line of singularities. As we go on to $\alpha_0=70$ and 100 a.u., the *dichotomy* of the wave function is almost complete. Each of the endpoints is surrounded by a total charge of approximately $e/2$. The interval between the two split parts being about $2\alpha_0$; at $\alpha_0=100$ a.u. one is dealing with truly Rydberg-type sizes.

TABLE I. Energy of the ground state $(1s)\sigma_g$ of atomic hydrogen.

α_0 (a.u.)	E (a.u.)
0	−0.500 00
1	−0.402 37
5	−0.201 95
10	−0.130 09
20	−0.079 871
30	−0.059 705
50	−0.041 883
70	−0.033 335
100	−0.026 183

To understand the dichotomy mechanism, we outline the following argument.[15] If at large α_0 the wave function ϕ is indeed concentrated around the end points $\pm\alpha_0\mathbf{e}$, as suggested by Fig. 1, i.e., if ϕ has significant values only in the regions characterized by $(r_+/\alpha_0)\ll 1$ and $(r_-/\alpha_0)\ll 1$, then the potential $V_0(\alpha_0,\mathbf{r})$ can be reduced to a simpler form. Indeed, as easily seen from Eq. (5), for $(r_-/\alpha_0)\ll 1$, V_0 becomes

$$\tilde{V}_0(\alpha_0,\mathbf{r}_-)=-(2e^2/\pi)(2\alpha_0 r_-)^{-1/2}\times K[2^{-1/2}(1-\hat{\mathbf{r}}_-\cdot\mathbf{e})^{1/2}], \quad (6)$$

whereas for $(r_+/\alpha_0)\ll 1$ it becomes $\tilde{V}_0(\alpha_0,-\mathbf{r}_+)$. Hence, in the vicinity of the end point $+\alpha_0\mathbf{e}$, Eq. (4) reduces to one containing Eq. (6) as a potential. By introduction of the scaled variable $\boldsymbol{\xi}=\mathbf{r}_-/\alpha_0^{1/3}$, this equation takes the form $\mathbb{H}(\xi)\varphi(\xi)=W\varphi(\xi)$, where $\mathbb{H}(\xi)$ is independent of α_0 and $W=\alpha_0^{2/3}E$. Since the square in-

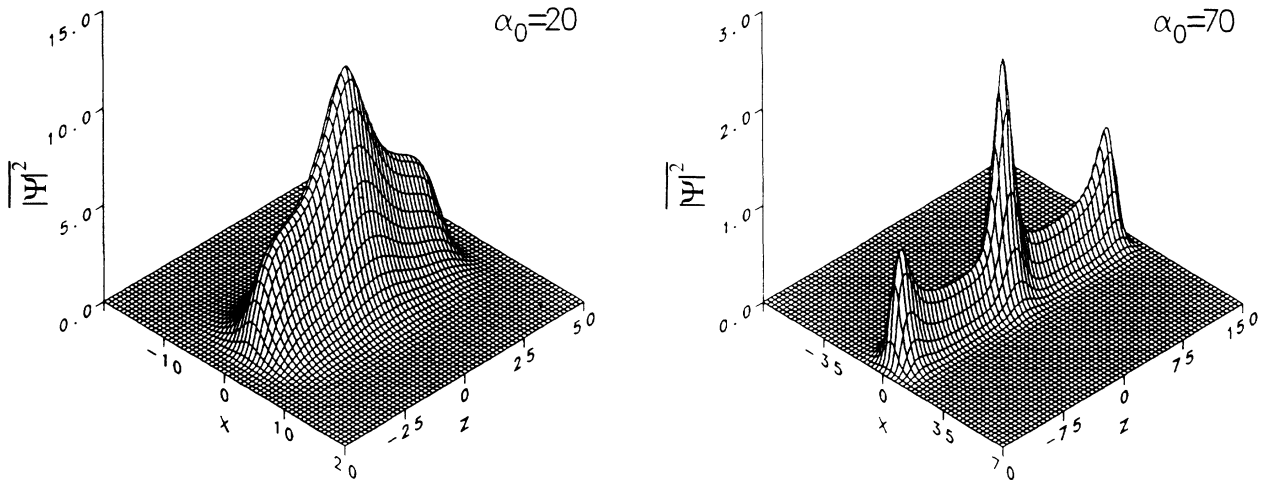

FIG. 2. Time-averaged position probability density $\overline{|\Psi|^2}$ for the ground state $(1s)\sigma_g$ of atomic hydrogen in the laboratory frame of reference [from Eq. (8)], for $\alpha_0=20$ and 70 a.u. The axes are defined as in Fig. 1, the length unit is a_0, $\overline{|\Psi|^2}$ is in units of $10^{-5}a_0^{-3}$.

tegrable function $\varphi(\xi)$ extends essentially over a finite region in the space of the ξ variable, it follows that for the corresponding region in the r_- variable we have $(r_-/\alpha_0)=(\xi/\alpha_0^{2/3})\ll 1$. This confirms that indeed (to lowest order in $1/\alpha_0$) the potential Eq. (6) can support wave functions concentrated around the end point $+\alpha_0\mathbf{e}$.[16] We find further that the (high) α_0 dependence of the eigenvalues $E_n(\alpha_0)$ of Eq. (4) is given by

$$E_n(\alpha_0)=\alpha_0^{-2/3}W_n[1+O(\alpha_0^{-2/3})], \tag{7}$$

where W_n are eigenvalues of H. For the ground state, the α_0 dependence is borne out by the last two eigenvalues of Table I.

The eigenfunction ϕ for the ground state, as well as the corresponding solution of Eq. (1), $\psi(\mathbf{r},t)\cong\phi(\mathbf{r})\times\exp(-iEt/\hbar)$, refer to the Kramers frame. In the laboratory frame, the dichotomized wave function of Fig. 1 and the corresponding charge density oscillate with (high) frequency ω. The "observable" charge density is obtained, as usual, by our averaging $|\Psi(\mathbf{r},t)|^2=|\phi[\mathbf{r}-\alpha(t)]|^2$ over a period, where ψ denotes the wave function in the laboratory frame.[17] The result is shown in Fig. 2 for two characteristic values of α_0. At $\alpha_0=70$ a.u., when dichotomy has set in, the averaged charge density has a typical *trident* shape, of extension $4\alpha_0$ in the z direction, with a central peak roughly twice the height of the lateral ones. This can be easily explained by our taking into account the harmonic nature of $\boldsymbol{\alpha}(t)$, Eq. (2).

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[4]M. Gavrila, in *Atoms in Unusual Situations,* edited by J. P. Briand, NATO ASI Ser. B, Vol. 143 (Plenum, New York, 1987), p. 225, and to be published.

[5]An exploratory calculation was carried out previously by M. Pont and M. Gavrila, Phys. Lett. A **123**, 469 (1987), with a simplified form of the "dressed potential," Eq. (5).

[6]H. A. Kramers, *Collected Scientific Papers* (North-Holland, Amsterdam, 1956), p. 866; W. C. Henneberger, Phys. Rev. Lett. **21**, 838 (1968).

[7]In passing from the Schrödinger equation in the momentum gauge to Eq. (1), we have also removed the term $e^2\mathbf{A}^2(t)/2mc^2$ by a phase transformation [see Ref. 4, Eq. (7)].

[8]The physical meaning of $V_0(\alpha_0,\mathbf{r})$ was discussed in Refs. 2 and 4.

[9]The energy spectrum of Eq. (4) is shifted from that of the Schrödinger equation in the momentum gauge by $(-e^2a^2/4mc^2)$, see Refs. 7 and 17.

[10]Multiphoton ionization becomes possible in the next order of the iteration, when the quasienergy E becomes complex. The expressions of the n-photon decay amplitudes were given in Ref. 4.

[11]In the heuristic approach of Henneberger [Ref. 6; see also C. K. Choi, W. C. Henneberger, and F. C. Sanders, Phys. Rev. A **9**, 1895 (1974)] the high-frequency character of the approximation was missed, whereas it was recognized by J. I. Gersten and M. H. Mittleman, J. Phys. B **9**, 2561 (1976) (although they formulated the frequency condition too restrictively, see Ref. 5).

[12]A graphical representation of V_0 was given in Fig. 1 of Ref. 2.

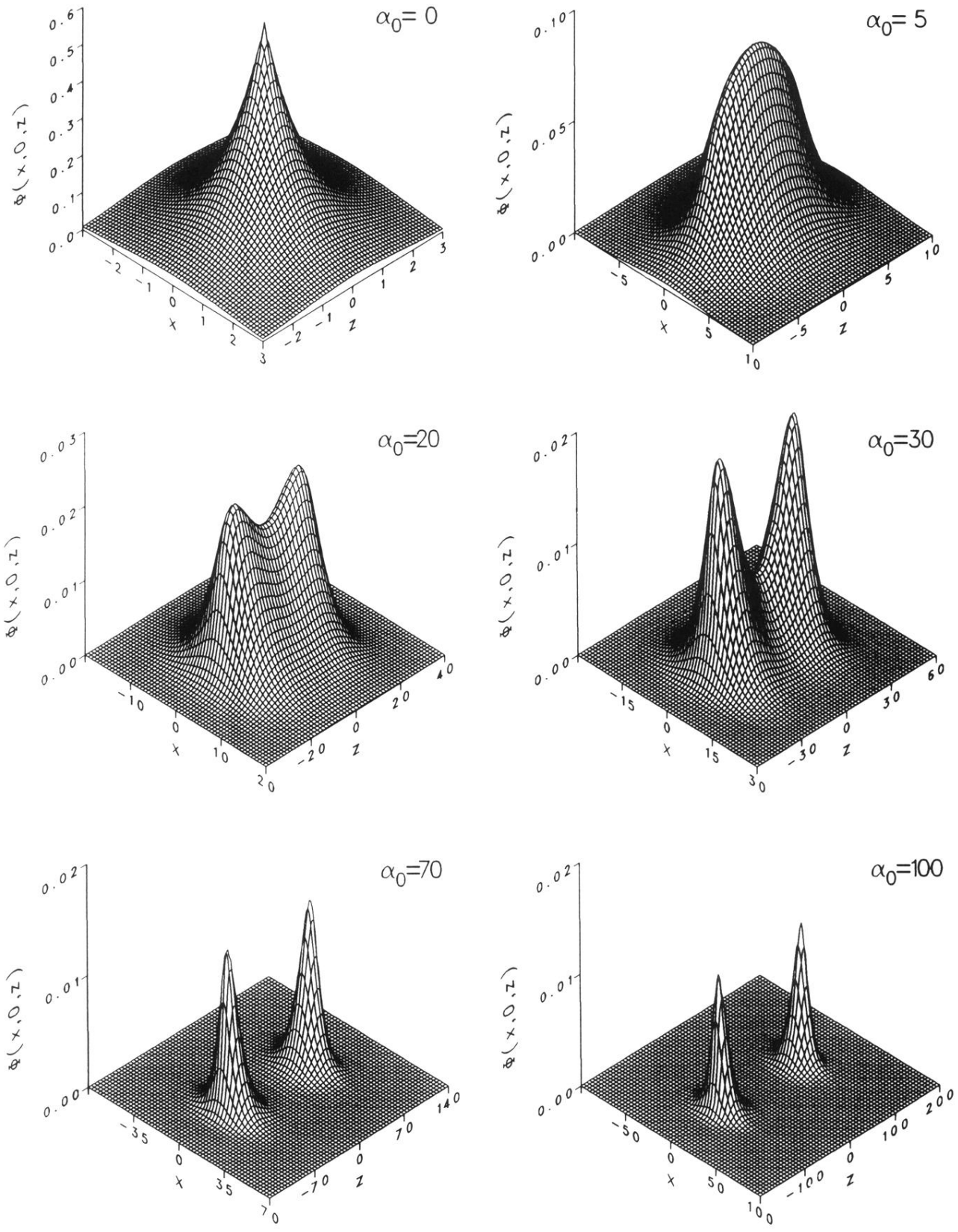
[13]When the excimer laser of Ref. 1 ($\hbar\omega=6.4$ eV) is operated at $I=10^{17}$ W/cm^2, we have $\alpha_0\simeq 32$.

[14]This was already noted in Ref. 5. The accuracy of those calculations decreases with increasing α_0. For $\alpha_0\ll 1$, they are quite accurate; for $\alpha_0=1$, 10, 50, 100 a.u., the respective deviations from the values given in Table I amount to 1%, 8%, 17%, 25%. The calculation of Choi, Henneberger, and Sanders (see Ref. 11) extending up to $\alpha_0=1$, agrees with ours to better than 1% (their results were given in graphical form).

[15]A more rigorous proof of this argument will be given elsewhere (M. Pont, N. R. Walet, and M. Gavrila, to be published).

[16]The full wave function is obtained from $\phi(\mathbf{r})\cong\varphi(\mathbf{r}_+)+(-1)^\pi\varphi(-\mathbf{r}_-)$, where π denotes the parity.

[17]In the laboratory frame the solution of the Schrödinger equation in the momentum gauge is $\Psi(\mathbf{r},t)\cong\phi[\mathbf{r}-\boldsymbol{\alpha}(t)]\times\exp\{-(i/\hbar)[E+(e^2a^2/4mc^2)]t\}$, see also Refs. 7 and 9.



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