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Possibility of breakdown of atomic stabilization in an intense high-frequency field

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We have calculated the ionization yield for a hydrogen atom irradiated by a short (5-cycle) intense pulse, with the atom initially in the 1*s* or 2*p* ($m \neq 0$) state. We numerically solved the time-dependent Schrödinger equation, first making the dipole approximation, and then including the photon momentum. At sufficiently high intensities we find significant differences between making and not making the dipole approximation, suggesting that atomic stabilization may break down in superintense fields.

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The possibility that an atom becomes stable against ionization in a very strong high-frequency field has aroused considerable interest — see, e.g., Refs. [1–11]. However, up to now all calculations have been performed making the dipole approximation, wherein the photon momentum is neglected. While each photon carries only a very small momentum, i.e., $\hbar\mathbf{k} \equiv (\hbar\omega/c)\hat{\mathbf{x}}$, where $\hbar\omega$ is the photon energy and $\hat{\mathbf{x}}$ is a unit vector along the pulse propagation direction, many photons are scattered from the atomic electron. Moreover, during the rise and fall of the pulse, scattering in the forward direction is a *stimulated* process, i.e., photons scatter from and into occupied modes within the energy bandwidth of the pulse. Stimulated photon scattering results in a significant energy transfer, ΔE, to the electron — this is the ac shift. Since an energy transfer ΔE must be accompanied by a momentum transfer of $(\Delta E/c)\hat{\mathbf{x}}$, and since, at high frequencies and high intensities, ΔE is of the order of the peak ponderomotive energy, P_0, the net momentum imparted to the electron by the radiation field is comparable to $(P_0/c)\hat{\mathbf{x}}$. If the rate at which this momentum is transferred to the electron is comparable to the characteristic atomic binding force, one may wonder whether stabilization still occurs. Indeed, the breakdown of stabilization due to radiation pressure was recently suggested by Katsouleas and Mori [12].

It is perhaps worth recalling that the photoionization of an atom by a weak field can be treated within the dipole approximation as long as the wavelength, λ, is large compared to the characteristic size of the atom; in this case the momentum imparted by the radiation is h/λ. However, in strong fields we must multiply h/λ by the number of photons that participate in the dressing of the atom, and this number is typically of order $P_0/(\hbar\omega)$.

A proper treatment of the interaction of an atom with a very strong radiation field should be given within the framework of relativistic quantum mechanics, but the numerical solution of the time-dependent Dirac equation is presently beyond our scope. Thus, as a preliminary investigation we have numerically solved the time-dependent nonrelativistic Schrödinger equation for the hydrogen atom, taking into account the photon momentum. (Some aspects of the computation are sketched in the Appendix.) We find that stabilization can indeed break down. Furthermore, we find that significant population transfer between bound states can occur, enhanced by a rotational coupling which appears only beyond the dipole approximation. We view these results as a warning that predictions based on the use of the dipole approximation may be invalid at intensities lower than one might have expected [13].

For simplicity we assume the light to be linearly polarized, along the z axis, and the intensity to be spatially homogeneous. Letting H_{a} denote the Hamiltonian of the bare atom, the Hamiltonian of the atom interacting with the pulse is

$$H(t) = H_{\mathrm{a}} - \frac{e}{\mu c}\mathbf{A}(\mathbf{r},t)\cdot\mathbf{p} + \frac{e^2}{2\mu c^2}|\mathbf{A}(\mathbf{r},t)|^2, \tag{1}$$

where e, μ, and $\mathbf{p}$ are the charge, mass, and canonical momentum of the electron, and where $\mathbf{A}(\mathbf{r},t)$ is the vector potential of the field. Within the dipole approximation, the vector potential is

$$\mathbf{A}^{(0)}(t) = f(t)\sin(\omega t)\hat{\mathbf{z}}, \tag{2}$$

where $f(t)$ is a slowly varying function of the time t on the scale of one cycle $2\pi/\omega$. We took the intensity profile of the pulse to be Gaussian: $f(t) = f_0 e^{-t^2/t_p^2}$. The inclusion of the photon momentum gives rise to a magnetic field, $\mathbf{B}(t)$, which we can express as $\mathbf{B}(t) = \hat{\mathbf{x}}\times\mathbf{E}(t)$, where $\mathbf{E}(t)$ is the electric field and where $\hat{\mathbf{x}}$ is a unit vector along the x axis. We can evaluate $\mathbf{E}(t)$ within the dipole approximation; we have $\mathbf{E}(t) = -(1/c)d\mathbf{A}^{(0)}(t)/dt$. Thus, writing $\mathbf{A}^{(0)}(t) \equiv A^{(0)}(t)\hat{\mathbf{z}}$, we have $\mathbf{B}(t) = (1/c)(d/dt)A^{(0)}(t)\hat{\mathbf{y}}$, and now writing $\mathbf{B}(t) = \nabla\times\mathbf{A}^{(1)}(\mathbf{r},t)$ we see that the leading correction to $\mathbf{A}^{(0)}(t)$ is

$$\mathbf{A}^{(1)}(\mathbf{r},t) = -\frac{x}{c}\frac{d\mathbf{A}^{(0)}(t)}{dt}. \tag{3}$$

Note that $\mathbf{A}^{(1)}(\mathbf{r},t)$ is of order v/c smaller than $\mathbf{A}^{(0)}(t)$, where v is the (instantaneous) speed of the electron. Dropping the contributions in $|\mathbf{A}^{(0)}(t)|^2$ and $|\mathbf{A}^{(1)}(\mathbf{r},t)|^2$

to the term in $|\mathbf{A}(\mathbf{r},t)|^2$ (since the first contribution is spatially independent and the second contribution is relativistic) we have

$$H(t) = H_\mathrm{a} - \frac{e}{\mu c}[\mathbf{A}^{(0)}(t) + \mathbf{A}^{(1)}(\mathbf{r},t)]\cdot\mathbf{p} + \frac{e^2}{\mu c^2}\mathbf{A}^{(0)}(t)\cdot\mathbf{A}^{(1)}(\mathbf{r},t). \tag{4}$$

The Lorentz force, $\mathbf{F}(t)$, on the electron due to its quivering with a velocity $\mathbf{v} \equiv (e/\mu)\int^t dt'\ \mathbf{E}(t') = -(e/\mu c)\mathbf{A}^{(0)}(t)$ in the magnetic field is $\mathbf{F}(t) = (e/c)\mathbf{v}\times\mathbf{B}(t)$, which, up to a sign, is just the spatial gradient of the last term on the right-hand side of Eq. (4). It is useful to express $\mathbf{F}(t)$ as

$$\mathbf{F}(t) = \frac{1}{c}\frac{dP(t)}{dt}\hat{\mathbf{x}}, \tag{5}$$

where

$$P(t) = (e^2/2\mu c^2)|\mathbf{A}^{(0)}(t)|^2, \tag{6}$$

and where the cycle average of $P(t)$ is the ponderomotive energy, $P_0(t) \equiv (e^2/4\mu c^2)f^2(t)$, whose peak value we denote as $P_0 \equiv (e^2/4\mu c^2)f_0^2$. In photon terms $\mathbf{F}(t)$ is mediated by stimulated photon scattering, as noted above. This force transfers drift momentum to the electron, and pushes the electron in the direction that the pulse propagates. In contrast, the electric field force $e\mathbf{E}(t)$ drives the electron back and forth along the polarization axis, and therefore distorts the atom along this axis.

In order to carry out calculations at very high intensities — which demand considerable computer time — we made a further approximation: We dropped the term $-e/(\mu c)\mathbf{A}^{(1)}(\mathbf{r},t)\cdot\mathbf{p}$ from $H(t)$ in Eq. (4). This term takes into account the Lorentz force associated with the *cycle-averaged motion* of the electron in the magnetic field; it is proportional to the square-root of the intensity, and at sufficiently high intensities is dwarfed by the Lorentz force associated with the quiver motion, which is proportional to the intensity — the cycle-averaged motion is much slower than the quiver motion. This further approximation enables us to cast the Hamiltonian into a form that is similar to that obtained when the dipole approximation is made. To this end we first introduce a gauge transformation which effects a shift in the canonical momentum by $(1/c)P(t)\hat{\mathbf{x}}$; replacing $|\Psi(t)\rangle$ by $e^{(i/\hbar c)P(t)x}|\Psi(t)\rangle$ in the Schrödinger equation $i\hbar(d/dt)|\Psi(t)\rangle = H(t)|\Psi(t)\rangle$ results in a modification to the Hamiltonian:

$$H(t) = H_\mathrm{a} - \frac{e}{\mu c}\mathbf{A}'^{(0)}(t)\cdot\mathbf{p}, \tag{7}$$

where $\mathbf{A}'^{(0)}(t) = \mathbf{A}^{(0)}(t) - (1/e)P(t)\hat{\mathbf{x}}$, and where we have dropped the spatially-independent term $(e^2/2\mu c^2)|\mathbf{A}'^{(0)}(t)|^2$. After some algebra we can express $\mathbf{A}'^{(0)}(t)$ as

$$\mathbf{A}'^{(0)}(t) = \beta(t)A^{(0)}(t)[-\hat{\mathbf{x}}\sin\alpha(t) + \hat{\mathbf{z}}\cos\alpha(t)], \tag{8}$$

where $\alpha(t)$ and $\beta(t)$ are defined by

$$\beta(t) = \sqrt{1 + \frac{P(t)}{2\mu c^2}}, \tag{9}$$

$$\cos\alpha(t) = 1/\beta(t). \tag{10}$$

We now rotate the coordinate system about the y axis through the angle $\alpha(t)$, that is, we transform $|\Psi(t)\rangle$ to $e^{(i/\hbar)\alpha(t)J_y}|\Psi(t)\rangle$, where J_y is the component of the angular momentum operator along the y axis; the Hamiltonian becomes

$$H(t) = H_\mathrm{a} - (e/\mu c)\beta(t)\mathbf{A}^{(0)}(t)\cdot\mathbf{p} + \dot{\alpha}(t)J_y, \tag{11}$$

where $\dot{\alpha}(t)$ is the time derivative of $\alpha(t)$. The Hamiltonian of Eq. (11) differs from the Hamiltonian obtained in the dipole approximation only through (i) the presence of the rotational coupling term $\dot{\alpha}(t)J_y$ and (ii) the magnification of the vector potential by $\beta(t)$. Note that $\beta(t)$ differs from unity by a relativistic correction, and it follows that the ionization yield, integrated over all angles of emission, differs from the dipole approximation result only by a relativistic correction. (This also follows from the fact that the yield is not sensitive to a reversal in the direction of propagation of the light pulse, and so the correction arising from the inclusion of the photon momentum must be even under the change $c \to -c$.) Nevertheless, as we see below, the rotational coupling can have a significant effect even when $P_0/(\mu c^2) \ll 1$.

In Fig. 1 we show the ionization yield, vs the peak intensity, after a hydrogen atom initially in the $2p_x$ state has been irradiated by a pulse whose frequency is 0.5 a.u. and whose intensity profile has a width, $\sqrt{2\ln(2)}t_p$, of 5 cycles. We see that the ionization yield begins to decline when the intensity exceeds about 2×10^{16} W/cm^2, i.e., when $P \approx \hbar\omega$ (a criterion which applies rather generally [5, 6, 10]). Within the dipole approximation, the magnetic quantum number m is conserved, so the atom cannot deexcite to the ground state, and we see that the yield continues to decline as the intensity increases further. However, beyond the dipole approximation, the rotational coupling term mixes states with different m; the degenerate $2p_x$ and $2p_z$ states are strongly mixed, and the atom can deexcite to the ground state, from which it

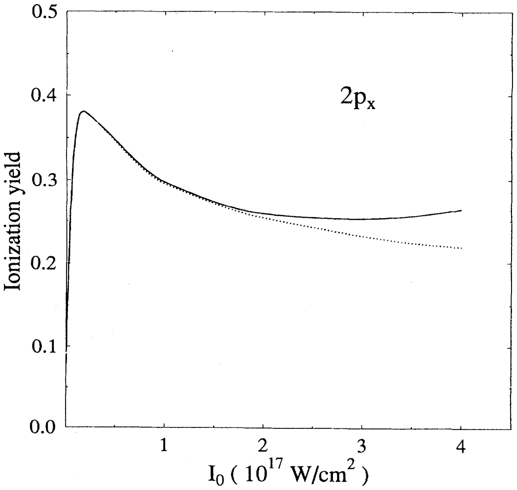

FIG. 1. Ionization probability at the end of the pulse (5 cycles, $\omega = 0.5$ a.u.) vs peak intensity I_0, for hydrogen initially in the $2p_x$ state. Dotted line: dipole approximation; solid line: incorporating corrections to the dipole approximation.

can ionize more easily since $\hbar\omega$ is not large compared to the ground-state binding energy. Indeed, the yield begins rising again at intensities above about 3×10^{17} W/cm^2. We also find that significant population is transferred to excited s states, even though at the intensity 4×10^{17} W/cm^2 we have $P_0/\mu c^2 \approx 10^{-3}$. In Fig. 2 we show the ionization yield when the frequency is 2.0 a.u., and we consider the initial state to be either the $1s$ or $2p_x$ state. The frequency bandwidth is now about 0.25 a.u., which is larger than the unperturbed binding energy of the $2p$ state, so ionization from the $2p$ state can take place via Raman scattering. Hence, stabilization does not occur when the atom is initially in the $2p_x$ state, even within the dipole approximation [6]. However, we see, in Fig. 2(b), that the probability for ionization from the $2p_x$ state is more than twice as large when the dipole approximation is not made. When the atom is initially in the $1s$ state stabilization does occur, and we find very little change in the ionization yield when we go beyond the dipole approximation; see Fig. 2(a). However, plotting the population in states other than the ground state reveals that (due to the rather large bandwidth) there is significant Raman scattering into excited bound states, and that the population in these excited states is strongly mixed by the rotational coupling.

Our inclusion of the photon momentum is equivalent to expanding $e^{i\mathbf{k}\cdot\mathbf{r}}$ in a Taylor series through the second term. The multipole expansion of $e^{i\mathbf{k}\cdot\mathbf{r}}$, while probably more accurate, leads to matrix elements that are considerably more complicated to evaluate, which is why we chose the Taylor series. We have neglected some relativistic corrections and retained others, but we think it unlikely that a fully relativistic treatment will drastically alter our principal findings; nevertheless, these findings must be viewed cautiously. We hope that the present work will stimulate further inquiry into whether stabilization breaks down in superintense fields.

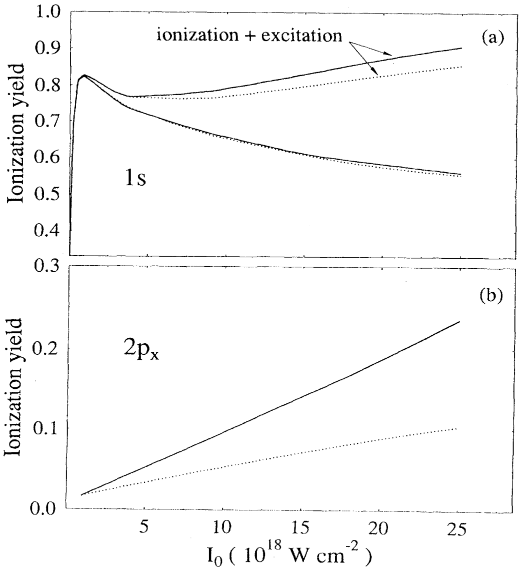

FIG. 2. Ionization probability at the end of the pulse (5 cycles, $\omega = 2.0$ a.u.) vs peak intensity I_0, for hydrogen intially in (a) $1s$ state, and (b) $2p_x$ state. Dotted line: dipole approximation; solid line: incorporating corrections to the dipole approximation. In (a) we also show the result of including excitation, i.e., $1 - P(1s)$, where $P(1s)$ is the probability for the atom to be in the $1s$ state (at the end of the pulse).

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APPENDIX

We represented $H(t)$ by a matrix, using a basis formed from linear combinations of $S^{\kappa}_{nl}(r)Y_{lm}(\hat{\mathbf{r}})$, where $S^{\kappa}_{nl}(r)$ is a radial Sturmian function, behaving for large r as $r^n e^{i\kappa r}$, and where $Y_{lm}(\hat{\mathbf{r}})$ is a spherical harmonic (defined with respect to a polar axis along the z axis). In order to describe both closed and open channels we chose the wave number κ to lie in the upper right quadrant of the complex κ plane [14]. We constructed the basis to exploit the full time- and spatial-reflection symmetries of $H(t)$, in the following way: The Hamiltonian $H(t)$ is invariant under a reflection, $\mathcal{I}_y$, in the xz plane. The Hamiltonian of the complete system, atom plus radiation field, is invariant under time-reversal, but we are treating the field as external. If $\mathcal{T}$ denotes a time-reversal operator which acts only on the atomic coordinates, $\mathbf{p}$ changes sign under the action of $\mathcal{T}$, and $H(t)$ is not invariant. However, the direction of propagation of the pulse can be reversed by a reflection, $\mathcal{I}_x$, in the yz plane. Introducing $\mathcal{S}_x \equiv \mathcal{T}\mathcal{I}_x$, and observing that H_a commutes with both $\mathcal{T}$ and $\mathcal{I}_x$, and that $f(t) = f(-t)$, we have

$$\mathcal{S}_x H(t) \mathcal{S}_x = H(-t). \tag{A1}$$

Note that $\mathcal{S}_x$ is antiunitary (since $\mathcal{T}$ is antiunitary while $\mathcal{I}_x$ is unitary) and therefore $\mathcal{S}_x$ complex conjugates c numbers. We use the Condon-Shortley phase convention for $Y_{lm}(\hat{\mathbf{r}})$, so the linear combination

$$\mathcal{Y}^{\pm}_{lm}(\hat{\mathbf{r}}) \equiv Y_{lm}(\hat{\mathbf{r}}) \pm Y^{*}_{lm}(\hat{\mathbf{r}}) \tag{A2}$$

is an eigenfunction of $\mathcal{S}_x$ with eigenvalue $(-1)^m$. Furthermore, $\mathcal{Y}^{\pm}_{lm}(\hat{\mathbf{r}})$ transforms to $\pm\mathcal{Y}^{\pm}_{lm}(\hat{\mathbf{r}})$ under the action of $\mathcal{I}_y$. Introducing the ket $|nlm;\pm\rangle$, where $\langle \mathbf{r}|nlm;\pm\rangle = S^{\kappa}_{nl}(r)\mathcal{Y}^{\pm}_{lm}(\hat{\mathbf{r}})$, we have

$$\mathcal{S}_x |nlm;\pm\rangle = |nlm;\pm,*\rangle, \tag{A3}$$

where $\langle \mathbf{r}|nlm;\pm,*\rangle = (-1)^m [S^{\kappa}_{nl}(r)]^{*} \mathcal{Y}^{\pm}_{lm}(\hat{\mathbf{r}})$. We also have

$$\mathcal{I}_y |nlm;\pm\rangle = \pm|nlm;\pm\rangle. \tag{A4}$$

We represented $H(t)$ by the matrix $\mathbf{H}(t)$ whose elements are $\langle n'l'm';\pm,*|H(t)|nlm;\pm\rangle$; in view of Eq. (A4), the elements $\langle n'l'm';\mp,*|H(t)|nlm;\pm\rangle$, i.e., those elements off-diagonal in the quantum number associated with $\mathcal{I}_y$, vanish, so $\mathbf{H}(t)$ consists of two uncoupled diagonal blocks. Since $\mathcal{S}_x$ is antilinear we have, for any two kets $|\phi\rangle$ and $|\psi\rangle$, that $\langle\phi|(\mathcal{S}_x|\psi\rangle)= \langle\psi|(\mathcal{S}_x|\phi\rangle)$. Noting that $\mathcal{S}_x = \mathcal{S}_x^\dagger$, it follows from Eq. (A1) that, with the tilde denoting the transpose,

$$\tilde{\mathbf{H}}(t) = \mathbf{H}(-t). \tag{A5}$$

Making use of these symmetries of $\mathbf{H}(t)$, we solved the time-dependent Schrödinger equation using a method described previously [15].

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