Decay of metastable H atoms in intense excimer lasers

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(Received 30 December 1991)

Decay widths of metastable hydrogen atoms subjected to the intense field of the currently available KrF^* laser ($\lambda = 248$ nm) and ArF^* laser ($\lambda = 193$ nm) are calculated by solving the full $(3+1)$-dimensional Schrödinger equation in the space-translated frame of reference. The critical intensities, above which the suppression of ionization decay sets in, are found to be $I_c \simeq 2.5\times10^{14}$ and 7×10^{14} W/cm^2 for $\lambda = 248$ and 193 nm, respectively.

PACS number(s): 32.80.Rm

Studies of highly perturbed quantum systems in intense laser fields revealed in the past a number of unexpected phenomena such as above-threshold ionization (ATI) [1,2] and multiharmonic generation [3,4]. Gersten and Mittleman [5] and Gavrila and co-workers [6] used a time-averaging approximation (applicable rigorously at asymptotically large frequencies) to suggest the possibility of another unexpected phenomenon: stability or suppression of ionization of atoms against photoionization with increasing intensity of the field. Voss and Gavrila [7] have recently emphasize the significance of stabilization in adiabatically switched laser fields as providing the possibility of producing strongly deformed atoms with intensity-tunable states for experimentation. They also applied the time-averaging approximation to obtain the lifetime of certain Rydberg states. Floquet calculations by Shakeshaft and collaborators [8] using extensive L^2 basis sets have shown that the time averaging and the high-frequency approximation are not strictly necessary requirements for the occurrence of the stability phenomenon. This has been found also to be the case by Marte and Zoller [9] and Kulander, Schafer, and Krause [10]; the latter authors employed a time-dependent method of calculation for pulsed radiation. Such behaviors at nonassymptotic frequencies have been first suggested by Su, Ebelry, and Javanainen [11], using a one-dimensional (1D) model calculation. Additional 1D model calculations by Burnett *et al.* [12] further illustrated these behaviors in strong laser fields. A recent 3D time-dependent study of Pont and Shakeshaft [13] suggests that the adiabatic switching condition is necessary for observing stabilization with above-threshold frequencies since the associated bandwidth should be small enough to avoid Raman coupling to low-l states which may be unstable. Since the centrifugal barrier also tends to keep the electron away from the nucleus, use of the circularly polarized field which tends to populate high-lying (lm) states can have a similar effect.

At present it appears to be very difficult to test such predictions for the ground-state hydrogen atom (or for noble gas atoms) due primarily to a lack of appropriate high-frequency lasers. The first excited (metastable) state of the H atom may, however, be investigated with the currently available intense KrF^* excimer laser ($\lambda = 248$ nm) [14] and/or the ArF^* laser ($\lambda = 193$ nm [15]. As yet no theoretical results for these available laser frequencies and intensities are known. The purpose of this Brief Report is to describe the behavior of the metastable H atom in the field of these two lasers and provide accurate theoretical values for the decay width and its dependence on the laser intensity under the adiabatic condition. This is achieved by solving the full $(3+1)$-dimensional Schrödinger equation of the system in the space-translated frame [15,16]:

$$i\frac{\partial}{\partial t}\psi(\mathbf{r},t) = \left[-\tfrac{1}{2}\Delta^2 - \frac{1}{|\mathbf{r}-\boldsymbol{\alpha}_0(t)|} \right] \psi(\mathbf{r},t) \ . \quad (1)$$

$\boldsymbol{\alpha}_0(t) = -(1/e)\int_{-\infty}^{t} \mathbf{A}(t)dt$ is the instantaneous "quiver radius" and $\mathbf{A}(t)$ is the vector potential which we choose to be circularly polarized. The $(3+1)$-dimensional partial differential equation (1) poses a formidable problem for direct integration due to the large space-time grid necessary for determining the rates in the adiabatic switching on and off conditions. We therefore apply the recently developed radiative close-coupling method [16,17,9] according to which the partial differential Eq. (1) can be converted by a joint Floquet plus partial wave expansion into an infinite set of ordinary differential equations for the coupled radial wave functions for the reaction channels $\{nlm\}$; the latter are characterized by the photon and the angular momentum quantum numbers:

$$\left[\frac{d^2}{dr^2} + k_n^2 - \frac{l(l+1)}{r^2} - \frac{2z}{r} \right] F_{nlm}(r) = -2z \sum_{n',l',m'} V_{nlm}^{n'l'm'}(r) F_{n'l'm'}(r) \ , \quad (2)$$

where $V_{nlm}^{n'l'm'}(r)$ are the channel-coupling potentials [16,17,9]. We note that if one is restricted only to the diagonal set of equations (2) diagonal in n, then Eq. (2) would correspond to the time-averaging approximation mentioned above. No such restriction is made in the present calculation and results reported are obtained by starting with a finite set of equations and extending it successively until convergence is secured for a given set of laser parameters. Retention of up to 15 $\{nlm\}$ channels has been found necessary and sufficient, for the results reported here.

We note that the same set of radiative close-coupling equations also describes the time-reversed problem of *stimulated capture* and *scattering* of an electron by a proton in the presence of an intense laser field, which has been investigated by us recently [16]. For the present purpose it is only necessary to replace the S-matrix scattering boundary condition by the well-known Siegert (ionization) boundary condition:

$$F_{ji}(r)_{r\to\infty}=H_j^+(r)T_{ji} ,$$

where $H_j^+(r)$ is the outgoing Coulomb radial wave function in the jth channel and T_{ji} is the corresponding transition matrix element. The energy determining secular determinant is obtained by matching the logarithmic derivative of the inner solutions of the close-coupling equations with the outer solutions at an intermediate radius. The imaginary part of the complex zero E_i which is connected adiabatically to the energy of the initial state provides the total decay rate Γ_i through the relation: $\Gamma_i=-2\text{Im}(E_i)$.

In view of the elaborate nature of the numerical calculations involved in (3+1) dimensions it is worthwhile to be able to test the present algorithm with respect to some known cases. To this end in Fig. 1 we compare the Floquet L^2 basis set calculations by Dörr *et al.* [8] at high frequencies $\hbar\omega=1.0$ and 2.0 (a.u.). The essential agreement both in shape and in magnitude found is especially significant in view of the completely independent algorithms used in the two calculations [18]. In Fig. 1, we also show the results for the ionization rates at the intermediate frequency $\hbar\omega=1.5$ (a.u.), which follow a trend similar to that of the neighboring curves. We may note here that the high-frequency approximation [5,6] tends to overestimate the rates of ionization in this frequency domain but qualitatively follows a similar behavior as the results of the full calculation. Finally, in Fig. 2 we present the results for the decay width of the metastable H atom subjected to the currently available KrF* laser ($\lambda=248$ nm) and the ArF* laser ($\lambda=193$ nm) as a function of the quiver radius $\alpha_0=E/\omega^2$ (a.u.), where E is the peak field strength of the laser. We observe that the maximum width of the H(2s) atom is higher at the KrF* wavelength ($\lambda=248$ nm) than that for the ArF* laser ($\lambda=193$ nm). Note, however, that beyond the maximum, the stabilization rate is faster at $\lambda=248$ nm than at $\lambda=193$ nm. These trends are consistent with the behavior of the ground-state H atom shown in Fig. 1. We observe that the maximum rates of ionization at both of these frequencies are very high. This would require presently unrealistically short pulses if one intends to use stationary atoms. However, use of highly relativistic beams following a variation of the current experiments with hydrogen negative ions by Bryant and co-workers [19] could provide the effective interaction time for this purpose. On the other hand, the predicted behavior of metastable H atoms at subcritical intensities may be tested directly with currently available pulse durations.

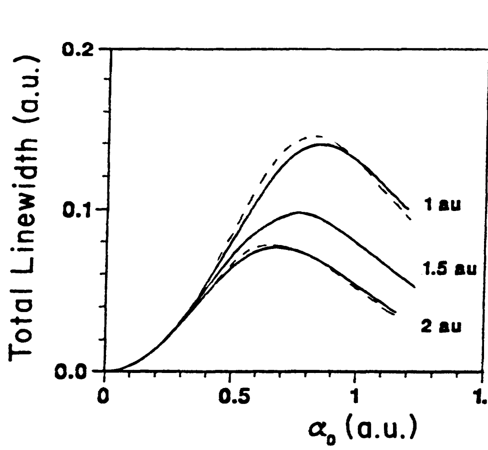

FIG. 1. Rates of ionization (linewidths) of H (1s) atoms at $\omega=1.0$, 1.5, and 2.0 a.u. Solid lines are present results. Dashed lines are estimated from data of L^2 Floquet calculations by Dörr *et al.* [8].

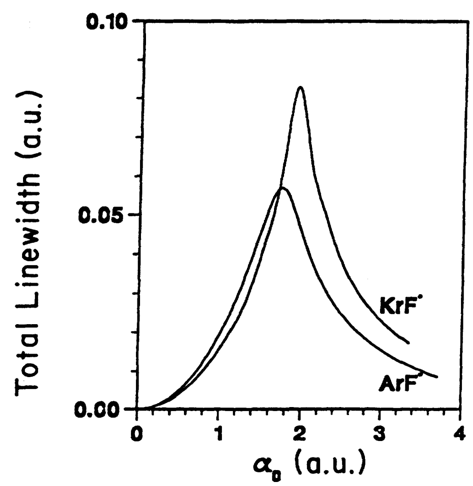

FIG. 2. Rates of ionization of metastable H atoms for available excimer lasers at $\lambda=248$ nm (KrF* laser) and $\lambda=193$ nm (ArF* laser). The critical intensities for the onset of ionization suppression are $I_c\simeq2.5\times10^{14}$ and 7×10^{14} W/cm^2, respectively, at $\lambda=248$ and 193 nm.

To conclude, we have investigated quantitatively the linewidth of the metastable H atom subjected to the intense field of the KrF* laser ($\lambda=248$ nm) and ArF* laser ($\lambda=193$ nm), both of which are currently available in the laboratory. The dependence of the decay widths on the laser intensity is calculated by solving the full (3+1)-dimensional Schrödinger equation of the system in the space-translated frame of reference using the method of radiative close-coupling equations. Critical intensities beyond which the decreasing rate of decay under adiabatic conditions sets in are found to be $I_c\simeq2.5\times10^{14}$ W/cm^2 and 7×10^{14} W/cm^2 for the KrF* laser and the ArF* laser ($\lambda=193$ nm), respectively. The nonperturbative response of the H (2s) atom can be tested using these lasers.

We thank Professor Dirk Feldmann for a useful discussion. This work was supported by Deutsche Forschungsgemeinschaft, under Project No. SFB-216M2.

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