PHYSICAL REVIEW A, VOLUME 65, 031405(R)

Observability of the dynamic stabilization of ground-state hydrogen with superintense femtosecond laser pulses

M. Dondera,[1] H. G. Muller,[2] and M. Gavrila[2,3]

[1]*Department of Physics, University of Bucharest, Bucharest-Magurele MG11, 76900 Romania*

[2]*FOM Institute for Atomic and Molecular Physics, Amsterdam, 1098 SJ, The Netherlands*

[3]*Institute for Theoretical Atomic and Molecular Physics, Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts 02138*

(Received 13 June 2001; revised manuscript received 6 November 2001; published 22 February 2002)

Theoretical information on dynamic stabilization (DS) of the ground state of H is still fragmentary, while its detection has remained open because of a lack of lasers with the required characteristics. The problem has been reactivated by the new light sources that are being developed, such as VUV-FEL's or attosecond pulses from high-harmonic generation, which will offer adequate frequencies, intensities, and pulse durations. We are now presenting a mapping out of DS over an extended range of high frequencies, for pulses with various envelopes, peak intensities, and durations. We find prominent DS under conditions where our nonrelativistic, dipole approximation, and calculation should be valid. There is a marked dependence of DS on the pulse shape. The effect of quasistationary stabilization (of the rates) on DS is analyzed. We comment on the impact of our results on the detection possibility. We conclude that ground-state DS for H should be observable with the new light sources in a state-of-the-art experiment.

DOI: 10.1103/PhysRevA.65.031405 PACS number(s): 42.50.Hz, 73.21.−b, 73.43.−f, 85.35.Be

Research on superintense-field atomic stabilization has not led to a positive answer to the problem: Is the phenomenon detectable for the ground state of H? After an early theoretical start a decade ago, the issue was abandoned because of computational limitations and lack of incentive, as none of the required experimental prerequisites were available: high-frequency lasers, in operation at high intensities, delivering very short pulses. (For an overview on stabilization, see [1].) However, a different situation has developed recently with the advent of the VUV-FEL light sources, now in test operation (HASYLAB at DESY, [2]) or under construction (BNL, ANL). Some of these sources (e.g., HASYLAB) are expected to deliver in a few years pulses with photons of energies in excess of 200 eV at high intensities (more than 10^{18} W/cm^2). Another promising development is the production of attosecond pulses from high-harmonic generation (HHG) [3], that are now under intense study. For example, the production of 100 asec pulses with 90-eV photons appears to be a realistic possibility, and the good beam quality of harmonic generation should allow diffraction-limited focusing to very high intensities as well. In view of the modified circumstances and of the fundamental interest in the problem, we are now presenting the comprehensive theoretical survey of stabilization in the ground state of H, and reassessing the possibility of its experimental detection.

There are two aspects of the phenomenon that need to be distinguished (see [1]). Stabilization was originally identified in Floquet theory as a *property of the ionization rates*: beyond some critical value of the field amplitude E_0, the rates $\Gamma(E_0)$ manifest a decreasing trend [4]. Following [1], we shall denote this property as "quasistationary stabilization" (QS). It refers to an idealized radiation field of constant amplitude, whereas in reality superintense fields are produced in the form of short pulses, sometimes of several cycles only (see [5]). Stabilization was discovered, however, also for wave-packet solutions of the time-dependent Schrödinger equation [6]; here one is dealing with atomic *ionization probabilities* P_{ion} at the end of a pulse, reflecting the experimental ionization yields. For pulses of fixed shape and duration, but increasing peak values E_0, at some point P_{ion} may start to decrease overall, or flatten out at a value $P_{ion}<1$. This has been termed "dynamic stabilization" (DS). A large numerical effort was devoted to its study, but most of it focused on one-dimensional (1D) models. There have been few dedicated computations for the ground state of real 3D hydrogen, [7–9], none of them recent. The conclusion reached at the time on the possibility of observing DS for the ground state was pessimistic (e.g., [8], a study of the case $\omega=1$ a.u.). Attention was shifted thereafter to Rydberg states, for which the phenomenon could eventually be demonstrated in two experiments [10]; comparison with theory was made in [11]. A recent endeavor on high-intensity ionization [12] bears little resemblance to our undertaking.

We are concerned here with DS of the ground state of H. We are studying the phenomenon for linear polarization, and pulses of various envelopes (sech, Gaussian, cos^2), over extended ranges of high frequencies ($0.51<\omega<8$ a.u.), electric-field amplitudes ($0<E_0<80$ a.u., depending on ω), and full width at half maximum (FWHM) pulse durations τ_p ($1<\tau_p<100$ cycles, depending on ω). Our calculations are done in the nonrelativistic dipole approximation.

The time-dependent Schrödinger equation is integrated in the velocity gauge using a highly efficient numerical program optimized in all respects [13(a)]. Although the program has already been checked in low-frequency problems [13(b)], we have undertaken a thorough testing of its performance also at high frequencies with excellent results. The laser pulses we have chosen are of the form $\mathbf{A}(t)=A_0(t)\mathbf{e}\sin(\omega t)$, where $\mathbf{e}$ is the linear polarization vector, $A_0(t)=A_0 f(t)$ is the pulse envelope with A_0 its peak value, and $f(t)$ its shape function. The latter was taken as

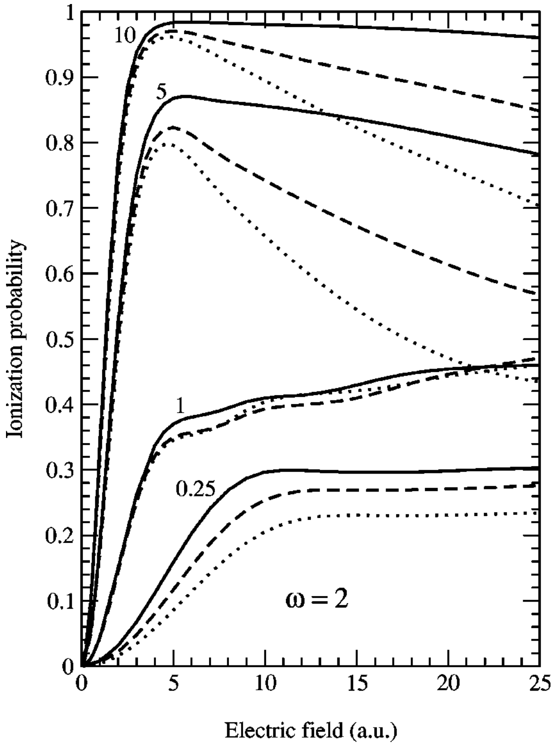

FIG. 1. Ionization probability of ground-state H exposed to pulses with $\omega=2$ a.u. and one of the envelopes in Eq. (1), at various FWHM pulse durations τ_p (in cycles), and varying peak fields E_0 (in a.u.). Sech pulses: solid lines; Gaussian pulses: dashed lines; $\cos^2$ pulses: dotted lines. The value of τ_p, in cycles, is specified per triplet of shapes (all of which have the same behavior at small E_0).

$$f_{sh}(t)=\mathrm{sech}(1.763t/\tau_p),$$

$$f_g(t)=\exp[-(1.177t/\tau_p)^2], \qquad (1)$$

$$f_c(t)=\cos^2(1.144t/\tau_p), \quad |t|<\pi\tau_p/2.288,$$

where τ_p represents the FWHM for $\mathbf{A}^2$ [15]. These forms are very much alike in their central parts $(-\tau_p/2<t<+\tau_p/2)$, but differ substantially in the shape of their wings. Their choice has an exploratory character, as it is not clear what the pulses of the prospective high-frequency sources will look like. Our pulses satisfy, for *any* τ_p, the experimental requirements on $\mathbf{A}(t)$ and $\mathbf{E}(t)$, leading to the vanishing of the displacement $(\delta\mathbf{r})$ and the drift momentum $(\delta\mathbf{p})$ acquired by a *free* classical electron during the pulse, $(\delta\mathbf{r})=(\delta\mathbf{p})=0$ (see [1]). In all cases we have kept the phase of the carrier wave fixed, as specified [14].

We have obtained P_{ion} by the standard procedure of computing the survival probabilities in discrete states at the end of the pulse, and taking the complement of their sum to 1. The accuracy on P_{ion} should be of better than 1%. We present in Figs. 1–3 samples of our results at $\omega=2,4,8$; a.u. are used. A nominal electric peak field E_0 was introduced for reference, via the plane-wave connection, $E_0=(\omega A_0/c)$. The values of E_0 considered extend up to a point at which relativistic corrections are expected to be small. As guidance we have used results of model calculations, which have shown that the corrections are negligible at $\omega=2$ and $E_0=20$ [16], and the fact that classical relativistic corrections scale as E_0/ω (see also [17(b)]).

Starting with the case of *longer pulses*, we find a dependence of P_{ion} on E_0 similar to that emerging from 1D cal-

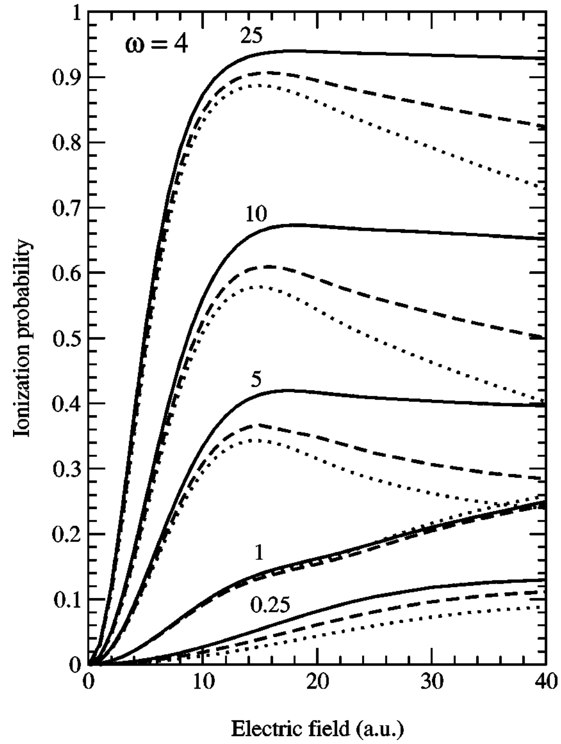

FIG. 2. Same as for Fig. 1, except that $\omega=4$ a.u.

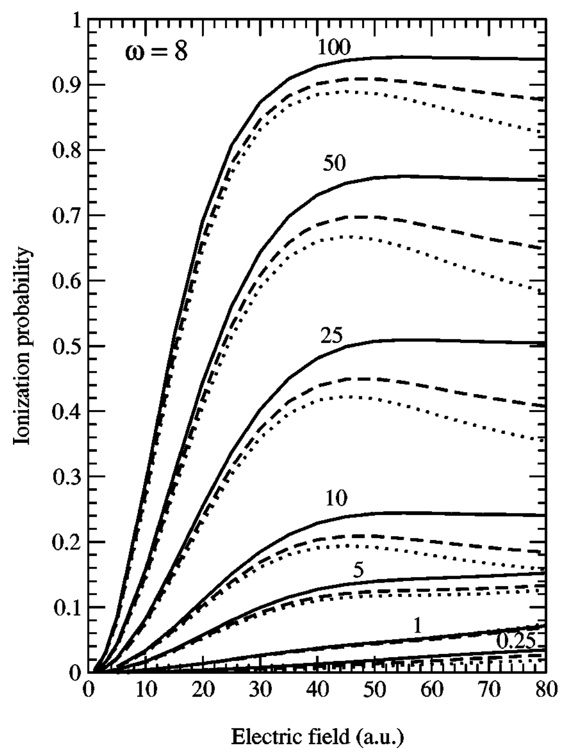

FIG. 3. Same as for Fig. 1, except that $\omega=8$ a.u.

culations: an incipient growth with E_0 followed by a maximum and then by a monotonic decrease (for sech pulses this is more similar to a plateau). The latter regime is that of DS, quite evident in all cases. In contrast to the 1D case (e.g., see [18]), P_{ion} has no oscillations in the DS regime. Its dependence on the pulse shape is quite marked. Since these shapes basically coincide in their central parts, the difference stems from the shapes of their wings. DS manifests itself by a decreasing behavior of P_{ion} with E_0 for the $\cos^2$ pulses (finite extension) and Gaussian (infinite extension but rapidly decreasing wings), and by a flat behavior of P_{ion} (with $P_{ion}<1$) for the sech pulses (infinite extension but more slowly decreasing wings). The dependence of P_{ion} on ω is quite strong, leading to more prominent DS and atomic survival as ω increases, but on the other hand, requiring larger peak values E_0 for DS to set in. The largest τ_p leading to DS at a detectable P_{ion} (i.e., not too close to 1) is in the femtosecond range and increases slowly with ω. *Short pulses* ($\tau_p<1$) yield a different picture. For the extreme case of $\tau_p=0.25$, P_{ion} becomes practically constant at large E_0 (more slowly at larger ω). For somewhat longer pulses (e.g., $\tau_p=1$), this situation has not been fully developed in the E_0 range shown and P_{ion} is still in a growing stage.

Let us now analyze the physical interpretation of DS as emerging from our results. This is best understood by expanding the evolving wave packet $\Psi(\mathbf{r},t)$ in Floquet states corresponding to the instantaneous value of the field envelope $E_0(t)$, a procedure termed "multistate Floquet theory," see [1].

For *longer pulses* or not too high E_0, one may expect that the evolution is adiabatic, i.e., the expansion will contain essentially a single, slowly changing Floquet state [rate $\Gamma(E_0(t))$], namely that corresponding to the initial state. P_{ion} can then be calculated from the adiabatic approximation formula

$$P_{ion}^{(ad)}=1-\exp\left[-\int_{-\infty}^{+\infty}\Gamma(E_0(t))dt\right]. \qquad (2)$$

Conversely, the fact that the dynamic P_{ion} coincides with $P_{ion}^{(ad)}$ is an indication that the evolution is adiabatic, see also [11]. For rapidly varying envelopes $E_0(t)$, several Floquet states need be included in the expansion of the wave packet and P_{ion} will differ from $P_{ion}^{(ad)}$. To illustrate the limits of adiabaticity, we have represented in Fig. 4, P_{ion} and $P_{ion}^{(ad)}$ at $\omega=2$, for the case of some pulses with larger τ_p; the corresponding $\Gamma(E_0)$ was obtained to high accuracy from our program [19]. It is apparent that, for the pulse shapes considered, the evolution of the atom is remarkably adiabatic up to quite high values of E_0 (e.g., $E_0<10$), even for short pulses ($\tau_p\cong5$). The agreement extends well into the DS regime, a situation already encountered in the 1D case [20]. In the E_0 range where this happens, DS is a direct consequence of the QS for $\Gamma(E_0)$, i.e., of the fact that $\Gamma(E_0)$ passes a maximum (at $E_0=3.60$, for $\omega=2$) and then decreases. However, as emphasized, the extent to which QS manifests itself dynamically on DS depends on the pulse shape. This is because $\Gamma(E_0)$ has to be compounded with the pulse shape in

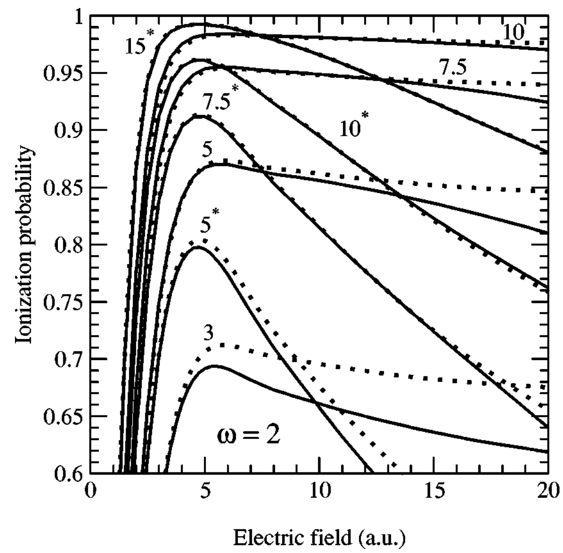

FIG. 4. Comparison at $\omega=2$ a.u. of E_0 dependence of the ionization probability of ground-state H, according to the computation (full lines) and the adiabatic approximation Eq. (2) (dotted lines). The values of τ_p (in cycles) are given for pairs of such curves. Sech and $\cos^2$ pulses are represented; $\cos^2$ pulses are distinguished by an asterisk on their τ_p value.

order to get $P_{ion}^{(ad)}$ from Eq. (2). At larger E_0, beyond the adiabatic regime, Fig. 4 shows that $P_{ion}<P_{ion}^{(ad)}$; this is due to the fact that part of the population has been projected from the ground Floquet state to excited states by the shock of the rising envelope ("shake up"), from where it ionizes more slowly.

In the case of *very short pulses* $\tau_p<1$, or very large E_0, the behavior of P_{ion} in Figs. 1–3 can be explained by the fact that high-energy continuum Floquet states are excited in the expansion of $\Psi(\mathbf{r},t)$. The electron is driven similar to a free particle, with negligible influence from the nucleus, so that the increase of E_0 will practically no longer affect the ionization ("strong-field limit," see [17(a)]).

We shall now make a few comments on the possibility of an *experimental detection of* DS as emerging from our predictions. A presently envisaged goal of the DESY-HASYLAB VUV-FEL machine is of some 30 fs pulses, by making use of advanced manipulation of the electron beam before entering the undulators [21]. This duration would be inacceptably long for a DS experiment. Fortunately, the machine was designed to be seedable with XUV radiation, which opens the possibility of implementing chirped pulse amplification. This would allow a one order-of-magnitude reduction of the pulse duration, bringing it into the desired range. Obviously, attosecond pulses from HHG offer in this respect a very favorable starting point. Another delicate experimental issue is the inevitable intensity distribution in the laser focus, which could completely blur the DS effect. The stabilization experiment on Rydberg atoms [10] countered this effect by preparing the target atoms in a sufficiently small part of the focus, and a strategy based on the same principle of strongly localized measurement can be applied in the present case.

In conclusion, we have mapped out P_{ion} for the ground state of H, and have found prominent DS in field ranges where our nonrelativistic calculation should be valid. We find a strong dependence of DS on the shape of the pulse envelope. The evolution of the atom is remarkably adiabatic up to large peak fields, and down to short pulses. Our results indicate that DS should be detectable by experiments at the different VUV-FEL light sources, or with attosecond pulses generated by HHG, in a state-of-the-art experiment.

This work is part of the research program of FOM, which is a division of NWO, the Netherlands organization for scientific research. One of us (M.G.) is grateful for the support from the I.R. Foundation.

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