

Adiabatic stabilization of a circular state: theory compared to experiment

Bernard Piraux¹ and R. M. Potvliege²

¹*Institut de Physique, Université Catholique de Louvain, 2 Chemin du Cyclotron, B-1348 Louvain-la-Neuve, Belgium*

²*Physics Department, University of Durham, Science Laboratories, Durham DH1 3LE, England*

Abstract

The probability that an atom of hydrogen, initially in the $5g(m = 4)$ state, survives a 90-fs pulse of 620 nm wavelength is calculated both by direct integration of the time-dependent Schrödinger equation and by a Floquet calculation. The two methods give virtually identical results. The survival probability calculated for a one-electron model of neon, for the same initial state, pulse duration and wavelength, is in fair quantitative agreement with the experimental data of van Druten *et al.* [Phys. Rev. A **55**, 622 (1997)].

To be published in Physical Review A, June 1998

As is well known, Fermi's golden rule predicts that an atom irradiated by a weak laser field decays by photoionization at a rate proportional to the intensity of the field if the photons energy exceeds the binding energy. This simple perturbative result is true, however, only at low intensity. It is now widely accepted that the rate of ionization in an intense high-frequency laser field increases *less* rapidly with intensity than a linear power law, and may even *decrease* above a certain critical intensity (before increasing again at ultra-high intensity when the coupling with the magnetic field of the incident wave becomes important). This remarkable reduction in the photoionization rate is often referred to as “adiabatic stabilization”, even though the atom never becomes completely stable against photoionization, because the quasi-stationary wave packet describing the atom at high intensity can be traced adiabatically to its field-free, initial state; in particular, its formation does not depend critically on fast variations of the intensity [1].

Adiabatic stabilization was first studied in depth in the late 1980s, by M. Gavrila, M. Pont and co-workers [2], and subsequently by many others. Numerical calculations focused initially on the stabilization of the ground state or low-excited states of hydrogen or model systems [3]. These were extended in 1991 and later to the analysis of atoms initially in an excited state with a high magnetic quantum number with respect to the polarization direction [4]. While results interpreted by several authors as casting doubts on the existence of the effect were also reported [5], the numerical calculations in general showed that the ionization rate indeed decreases at sufficiently high intensity. In particular, the calculations for atoms initially prepared in an excited circular state ($|m| = l \approx n - 1 \gg 1$) demonstrated stabilization at intensities, frequencies and pulse durations that could be achieved in the laboratory. This prompted two experiments — the only experimental test of adiabatic

stabilization so far — in which the photoionization of the $2p^55g(m=4)$ state of neon was studied at a wavelength of 620 nm and intensities up to 2.3×10^{14} W/cm² [6,7]. Onset of stabilization was observed at high intensity, and, to the extent that a comparison could be made, the experimental results were found to be consistent with the results of calculations done at the same wavelength for the $5g(m=4)$ state of hydrogen.

The present work aims at a closer comparison of theory with the experimental data of Ref. [7]. The quantities measured in the experiment (the yield in photoelectrons and the residual populations at the end of the pulse) can be calculated, for a simplified model of the atom, either by direct integration of the time-dependent Schrödinger equation or by treating the system as being quasi-stationary and using Floquet methods. We first show that, in the present case, these two approaches give virtually identical results.

Both the Floquet and the time-dependent calculations were performed in the dipole approximation, with the field described by the vector potential $\mathbf{A}(t) = A_0(t) \sin \omega t \hat{\mathbf{z}}$. For A_0 constant and with Siegert boundary-conditions imposed on the wave function, the quasienergy of the (Floquet) dressed state, E , is a complex eigenvalue of a system of time-independent differential equations. We solved this system numerically in the velocity gauge, using a basis of spherical harmonics and complex radial Sturmian functions [8]. The quasienergy was also obtained by Padé-summing its Rayleigh-Schrödinger perturbative expansion in powers of the intensity. In the high frequency regime, the latter technique gives the same results as a full non-perturbative calculation for a small fraction of the cost in CPU time. The rate of photoionization, $\Gamma = -2 \text{Im} (E/\hbar)$, does not depend on the normalization of the Floquet wave function and is a function of the vector potential amplitude, A_0 , and of the other parameters of the incident field. The variation of the ionization rate with intensity is shown in Fig. 1, for the dressed $5g(m=3)$ and $5g(m=4)$ states of hydrogen at 620 nm wavelength: Γ increases in proportion of the intensity in weak fields, in agreement with the golden rule, but eventually reaches a maximum and then decreases monotonically as stabilization sets in. The maximum occurs at a higher intensity for $m=3$ than for $m=4$, as is discussed in Ref. [4d]. Note that in Fig. 1, Γ remains small compared to the inverse of a laser period, $\omega/2\pi = 0.012$ a.u., and can therefore be interpreted in the usual sense, as a cycle-averaged ionization rate. The process being non-resonant, one may expect that the atom follows the dressed states adiabatically if A_0 varies slowly instead of being constant. Our Floquet calculations thus relied on representing the atom by a single dressed state, i.e. the one that develops from the unperturbed initial state as A_0 increases. Accordingly, the probabilities that the atom survives a (not too short) laser pulse and is left in its original state, P_{surv} , or that it is photoionized, P_{ion} , were taken to be

$$P_{\text{surv}} = \exp \left(- \int_{-\infty}^{+\infty} \Gamma[A_0(t)] dt \right) \quad (1)$$

and $P_{\text{ion}} = 1 - P_{\text{surv}}$ (thus neglecting excitation of other dressed states).

The method used for numerically integrating the time-dependent Schrödinger equation, for atomic hydrogen, has been described in Ref. [9]. Briefly, the wave function, represented on a basis of spherical harmonics and (real or complex) radial Sturmian functions, is propagated in time using a very stable predictor-corrector algorithm based on implicit Runge-Kutta methods. The photoionization yield is obtained as the complement to unity of the population remaining bound to the nucleus at the end of the pulse, which is calculated by projection

on all atomic bound states with $n \leq 25$ and $l \leq 18$. When using real Sturmians, the basis must span a region of space large enough to contain the wave packet until the end of the pulse. Much smaller bases are sufficient when employing complex Sturmians, as they smoothly absorb the outgoing flux before it leaves the region where the atom is represented accurately. The results quoted below have been obtained by using complex bases, of typically 15 spherical harmonics (with ℓ varying from 4 to 18) and, for each partial wave, 120 complex radial Sturmians. We have verified that at high field intensity, 2×10^{14} W/cm², identical results (to three significant figures) are obtained with a large basis of 600 real Sturmians per angular momentum and 50 angular momenta.

In order to compare the Floquet and the time-dependent methods, we calculated the probability that an hydrogen atom, initially in the $5g(m = 4)$ state with respect to the polarization direction, is ionized by a 120-cycle pulse. The pulse duration (90 fs, FWHM in intensity) was the same as in the experiment. For convenience, its envelope was taken to be $A_0(t) = A_0 \sin^2(t/\tau)$ [10]. The results are shown in Table I. Only insignificant ($< 0.0001\%$) residual populations in bound states other than the $5g$ state were found in the time-dependent calculation. Adiabatic stabilization manifests by a *decrease* in the ionization probability above about 1×10^{14} W/cm² peak intensity. The decrease is less pronounced in the ionization probability than in the ionization rate, but is still remarkable. Also noteworthy is the 4-digit agreement between the Floquet results and the time-dependent results. The agreement is better than had been previously reported for much shorter pulses [11]. It confirms that the one-state Floquet approximation is reliable for describing ionization in high frequency pulses, and makes it possible to assign, unambiguously, the reduction in the ionization probability observed in the time-dependent results to adiabatic stabilization [12].

The probability that the atom remains in the original $5g(m = 4)$ state is also the same in the two methods, since there is no net population transfer to other bound states. This is illustrated by Fig. 2. The figure also gives the survival probability for a pulse of same width as above but with a sech^2 intensity profile: the probability is a little smaller for the sech^2 -pulse than for the finite-duration pulse because for the same fluence the former irradiates the atom at (relatively) weak intensities for a longer time compared to the latter. A simple calculation shows that for a fixed pulse length and increasing fluence, the survival probability tends asymptotically to a constant smaller than 1 for a sech^2 -pulse, assuming of course adiabaticity and a continuing decrease of Γ at high intensity. However, it decreases to zero for pulses with, for example, a Lorentzian profile or increases to 1 for finite-duration pulses. Thus adiabatic stabilization (the decrease in the ionization rate) does not necessarily translate into a decrease in the ionization probability.

We now compare our theoretical results with the experimental data. We model the neon atom by a single electron moving in a central potential, $V(r)$, that accounts for the partial screening of the nuclear attraction by the inner electronic shells. In atomic units [13],

$$V(r) = -\frac{1}{r}[2f_0(\alpha_1 r) + 2f_0(\alpha_2 r) + 5f_1(\alpha_3 r) + 1] \quad (2)$$

where the parameters $\alpha_1 = 13.06$, $\alpha_2 = 7.2$ and $\alpha_3 = 3.68$ were obtained by fitting the energy levels of $V(r)$ to experimental values, and

$$f_\ell(\alpha r) = e^{-\alpha r} \sum_{j=0}^{2\ell+1} \left(1 - \frac{j}{2\ell+2}\right) \frac{(\alpha r)^j}{j!}. \quad (3)$$

Not surprisingly, the short-range part of $V(r)$ is of very little importance for states with $m = 3$ or 4 . The rates of ionization from the dressed $5g(m = 3)$ and $5g(m = 4)$ states of the model atom are within 0.02 % and 0.0005 %, respectively, from those based on a pure Coulombic potential (shown in Fig. 1).

The fraction of the atomic population left in the initial state at the end of a 90-fs pulse is shown in Fig. 3. The surviving fraction was measured directly in the experiment, and was also estimated indirectly as the complement to unity of the measured ionized fraction. The two sets of data are consistent and do not follow the rapid decrease at high fluence that a naive application of the golden rule would predict (see the dotted line in the figure, or Fig. 8 of Ref. [7]). The solid line in Fig. 3 represents the result of the Floquet calculation for the sech^2 -pulse of above. We assumed a uniform distribution of intensity since in the experiment the target atoms were prepared in the initial excited state only in the central part of the 620 nm beam. We also assumed that the system was initially in a pure $5g(m = 4)$ state while in the experiment the atoms were initially in a superposition of several correlated states with a small (at most 13 %) admixture of states having a $5g(m = 3)$ character. The importance of this admixture was tested by repeating the calculation for an incoherent superposition of the $5g(m = 4)$ and $5g(m = 3)$ states in a 87 % / 13 % ratio. The resulting survival probability, represented by the dashed line, is smaller than for the pure $5g(m = 4)$ state, since ionization proceeds faster from the $m = 3$ state, but the difference is minor. As seen from the figure, the calculated probabilities and the data are in fair quantitative agreement. This, of course, tails well with the conclusion of van Druten *et al.* [7], that the large survival probability at high fluence observed in the experiment can be attributed to adiabatic stabilization.

The authors thank J. F. McCann for his critical reading of the manuscript. B.P. is supported by the “Fonds National de la Recherche Scientifique”.

REFERENCES

- [1] See, e. g., H. G. Muller, in *Super-Intense Laser-Atom Physics IV*, edited by H. G. Muller and M. V. Fedorov (NATO ASI Series 3, Vol. 13, Kluwer, Dordrecht, 1996), for a review of the various forms of atomic stabilization in intense laser fields.
- [2] M. Gavrilă, in *Fundamentals of Laser Interactions*, edited by F. Ehlotzky (Lecture Notes in Physics, Vol. 229 (Springer, Berlin, 1985); M. Pont, PhD Thesis, University of Amsterdam (1990); M. Gavrilă, in *Atoms in Intense Laser Fields* [Adv. At. Mol. Opt. Phys. Suppl. **1**, 435 (1992)]. The theory has been extended recently to the case of two-electron systems: M. Gavrilă and J. Shertzer, Phys. Rev. A **53**, 3431 (1996).
- [3] The Floquet ionization rate has been calculated in a variety of ways, e. g., (a) M. Pont and M. Gavrilă, Phys. Rev. Lett. **65**, 2362 (1990) (high-frequency Floquet calculations); (b) M. Dörr, R. M. Potvliege, and R. Shakeshaft, Phys. Rev. Lett. **64**, 2003 (1990), and M. Dörr, R. M. Potvliege, D. Proulx, and R. Shakeshaft, Phys. Rev. A **43**, 3729 (1991) (Sturmian-Floquet calculations); (c) P. Marte and P. Zoller, Phys. Rev. A **43**, 1512 (1991), and L. Dimou and F. H. M. Faisal, *ibid.* **46**, 4442 (1992) (Floquet-coupled-channels calculations); (d) M. Dörr, P. G. Burke, C. J. Joachain, C. J. Noble, J. Purvis, and M. Terao-Dunseath, J. Phys. B **26**, L275 (1993) (R-Matrix-Floquet calculations). Stabilization has also been observed in full time-dependent calculations for ultra-short high frequency pulses as a decrease in the ionization probability for increasing peak intensity; see, e. g., J. H. Eberly and K. C. Kulander, Science **262**, 1229 (1993). However, in many of these time-dependent calculations transfer of population to dressed excited states caused by the very fast pulse turn-on may be partly responsible for this decrease.
- [4] (a) M. Pont and R. Shakeshaft, Phys. Rev. A **44**, R4110 (1991); (b) R. J. Vos and M. Gavrilă, Phys. Rev. Lett. **68**, 170 (1992); (c) R. M. Potvliege and R. Shakeshaft, in *Electronic and Atomic Collisions*, edited by W. R. MacGillivray, I. E. McCarthy, and M. C. Standage (Adam Hilger, Bristol, 1992); (d) R. M. Potvliege and P. H. G. Smith, Phys. Rev. A **48**, R46 (1993); (e) A. Buchleitner and D. Delande, Phys. Rev. Lett. **71**, 3633 (1993); (f) F. H. M. Faisal, L. Dimou, H.-J. Stiemke, and M. Nurhuda, J. Nonl. Opt. Phys. Mat. **4**, 701 (1995).
- [5] The case against adiabatic stabilization has been reviewed by S. Geltman, Chem. Phys. Lett. **237**, 286 (1995).
- [6] M. P. de Boer, J. H. Hoogenraad, R. B. Vrijen, L. D. Noordam, and H. G. Muller, Phys. Rev. Lett. **71**, 3263 (1993); M. P. de Boer, J. H. Hoogenraad, R. B. Vrijen, R. C. Constantinescu, L. D. Noordam, and H. G. Muller, Phys. Rev. **50**, 4085 (1994).
- [7] N. J. van Druten, R. C. Constantinescu, J. M. Schins, H. Nieuwenhuize, and H. G. Muller, Phys. Rev. **55**, 622 (1997).
- [8] See R. M. Potvliege and R. Shakeshaft, in *Atoms in Intense Laser Fields* [Adv. At. Mol. Opt. Phys. Suppl. **1**, 373 (1992)], for a more detailed description of the method.
- [9] E. Huens, B. Piraux, A. Bugacov, and M. Gajda, Phys. Rev. A **55**, 2132 (1997).
- [10] A. Fring, V. Kostykin, and R. Schrader, J. Phys. B **29**, 5651 (1996), concluded to the absence of bound-state stabilization for intense pulses with non-vanishing total impulse or classical displacement. The pulses considered in the present work do not fall in that category. See also V. Kostykin and R. Schrader, J. Phys. A **30**, 265 (1997); A. Fring, V. Kostykin, and R. Schrader, *ibid.* **30**, 8599 (1997); C. Figueira de Morisson Faria, A. Fring, and R. Schrader, J. Phys. B **31**, 449 (1998).

- [11] E.g. Ref. [4c]; O. Latinne, C. J. Joachain, and M. Dörr, Europhys. Lett. **26**, 333 (1994); Ref. [9].
- [12] Interference stabilization, which occurs when the widths of adjacent Rydberg states overlap, does not play any role in the present case, since the ionization width of the $5g(m = 4)$ state is two orders of magnitude smaller than its separation from the other ($m = 4$) states. Transient stabilization can also be safely ruled out on the ground that the time-dependent wave function remains very close to the Floquet wave function during the *entire* pulse. A detailed comparison of these wave functions, for various pulse durations, will be reported elsewhere by the authors.
- [13] S. Feneuille, M. Klapisch, E. Koenig, and S. Liberman, Physica **48**, 571 (1970).

TABLES

TABLE I. Probability that an atom of hydrogen, initially in the $5g(m = 4)$ state is ionized by the 120-cycle laser pulse described in the text, as calculated either by full numerical integration of the time-dependent Schrödinger equation or by Floquet methods. The wavelength is 620 nm and the polarization is linear. The reduced mass of the system was taken to be 1 a.u. in both calculations.

Peak Intensity	Time-dependent	Floquet
$0.25 \times 10^{14} \text{ W/cm}^2$	0.10103	0.10105
0.50	0.14122	0.14126
1.00	0.16035	0.16042
1.50	0.15596	0.15599
2.00	0.14705	0.14705

FIGURES

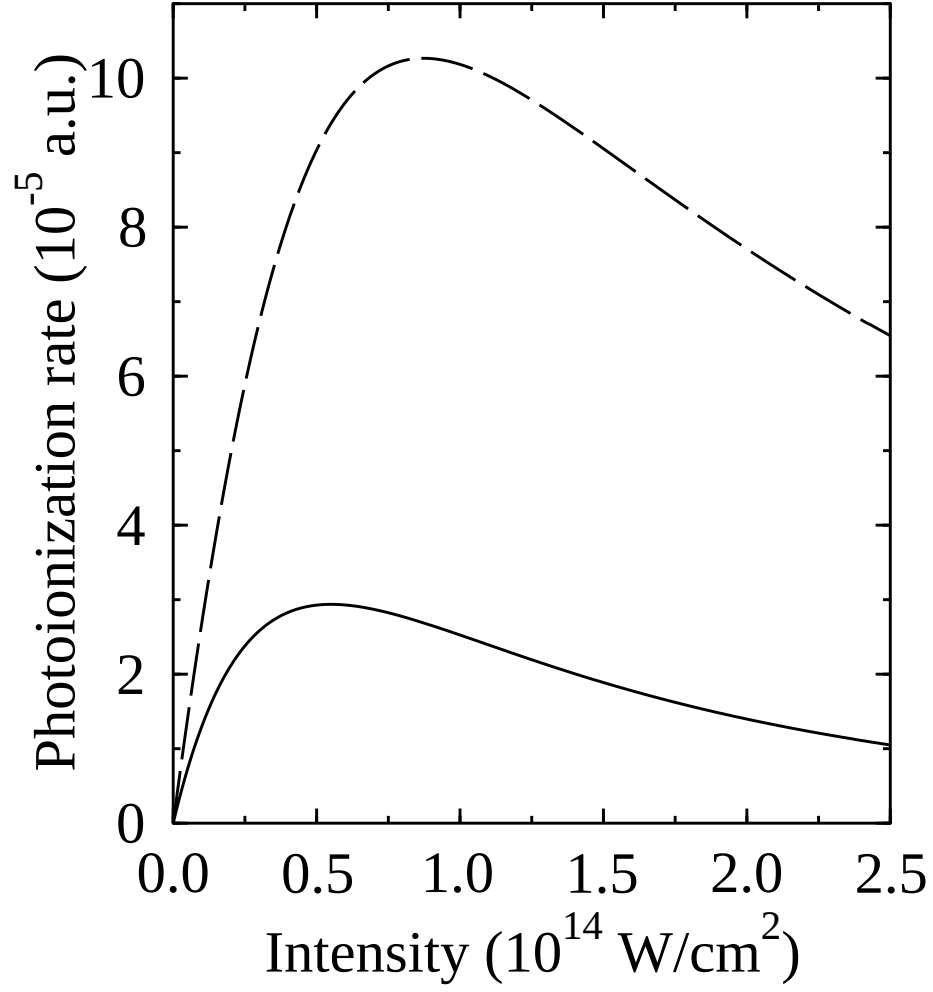


FIG. 1. Variation with intensity of the rate of photoionization of atomic hydrogen from the $5g(m=3)$ state (dashed line) or the $5g(m=4)$ state (solid line) at 620 nm wavelength. The field is linearly polarized along the axis of quantization of the angular momentum. The rate is expressed in atomic units.

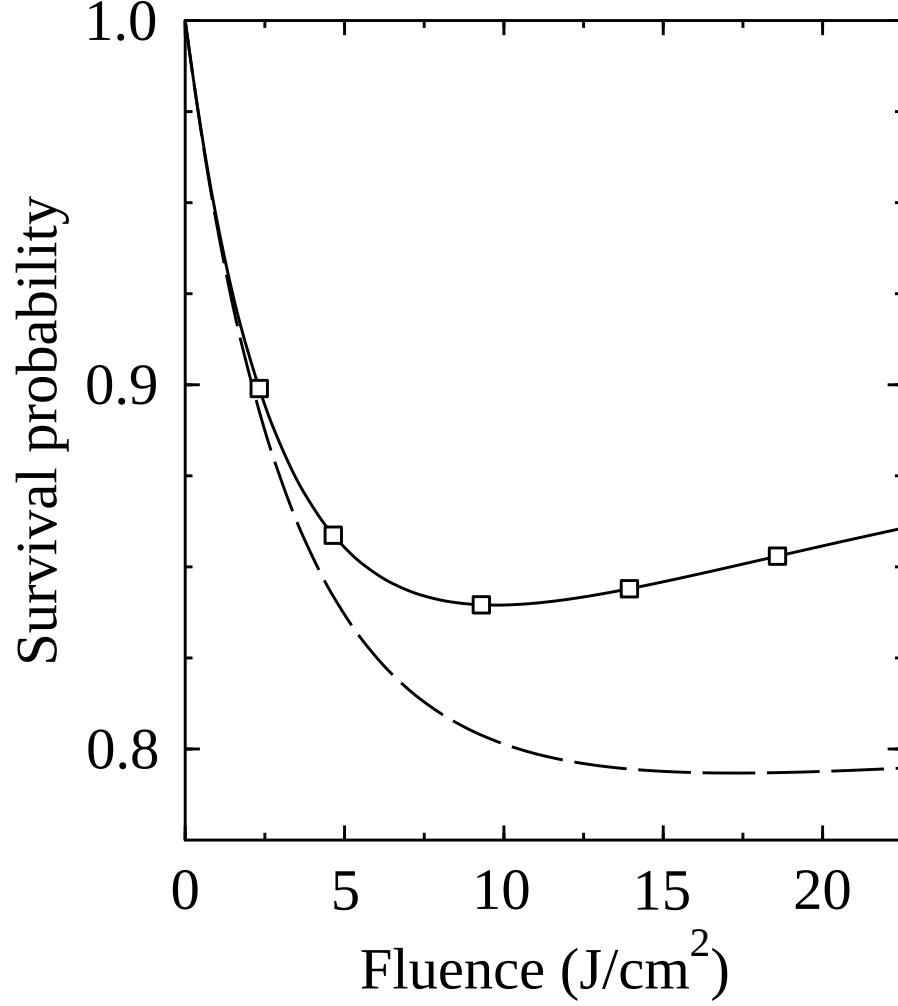


FIG. 2. Probability that an atom of hydrogen initially in the $5g(m = 4)$ state of hydrogen is in the same state at the end of a pulse of 620 nm wavelength and 90 fs duration (FWHM in intensity), as a function the pulse fluence. The field is linearly polarized along the axis of quantization of the angular momentum. Solid line: results of the Floquet calculation for the finite-duration pulse defined in the text. Squares: results of the time-dependent calculation for the finite-duration pulse. Dashed line: Floquet results for a pulse with a sech^2 -profile in intensity.

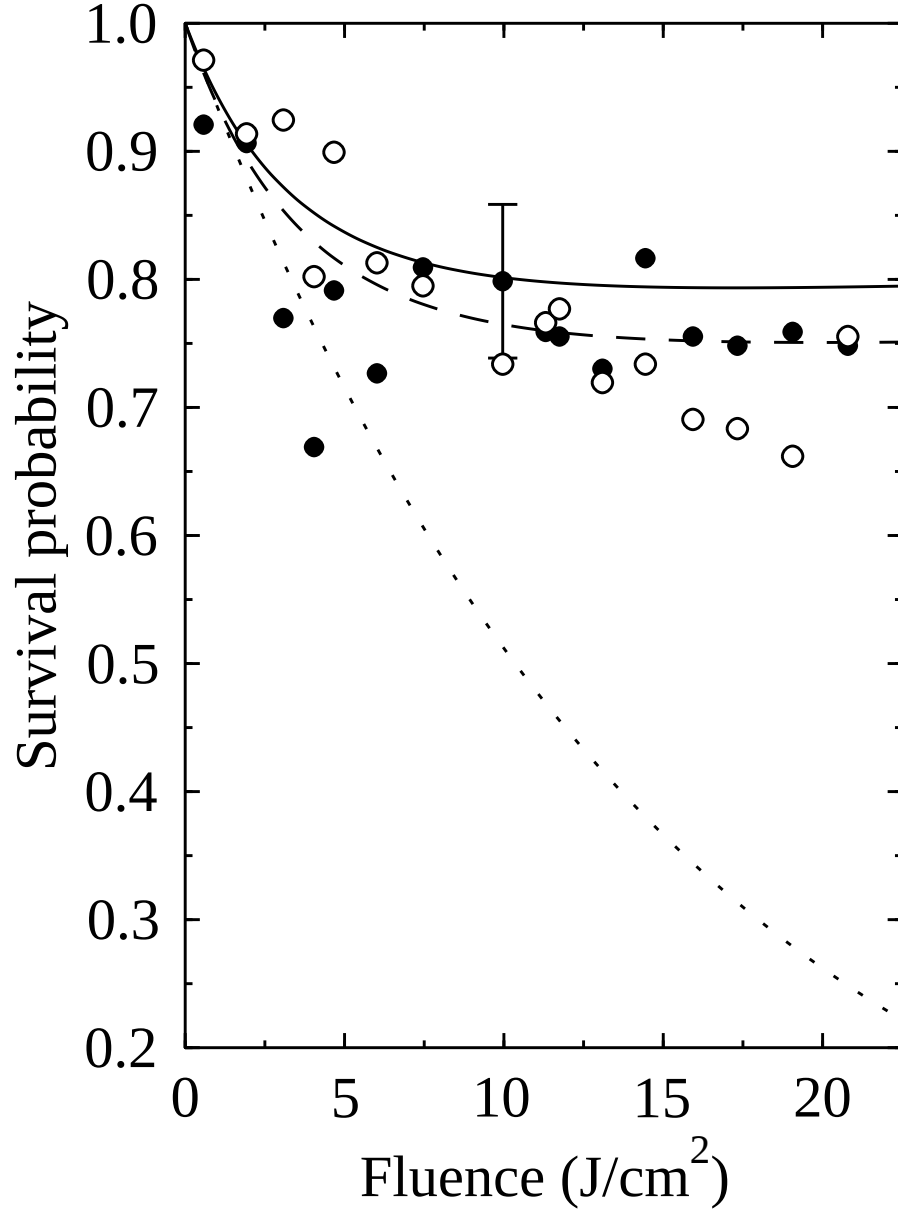


FIG. 3. Surviving fraction of the $2p^55g$ population. Open circles: surviving fraction measured by van Druten *et al.* [7]. Solid circles: complement to unity of the ionized fraction [7]. Solid curve: probability calculated non-perturbatively for a pure $5g(m=4)$ initial state. Dashed curve: same as the solid curve, but for an incoherent superposition of $5g(m=4)$ state (87 %) and $5g(m=3)$ state (13 %). Dotted curve: prediction of first-order perturbation theory for a pure $5g(m=4)$ initial state.