Prediction of a New Sequence of Peaks in Above-Threshold Detachment Spectrum in an Intense Laser Field

F. H. M. Faisal and P. Scanzano

Fakultät für Physik, Universität Bielefeld, 4800 Bielefeld 1, Federal Republic of Germany

(Received 12 November 1991)

We predict the presence of a new, equally spaced sequence of peaks in the above-threshold detachment (ATD) spectra in a very intense laser of a fixed frequency. Using a general theoretical argument, originally made by Wigner in the context of nuclear reaction theory, we identify the new comblike peak sequence as due to Wigner cusps at the thresholds of stimulated multiphoton bremsstrahlung channels in the intermediate continuum states. Our prediction is supported by solving the Schrödinger equation of a 3D model of H^- in an intense infrared laser ($\hbar\omega=0.35$ eV). A way of experimentally distinguishing the new sequence from the usual ATD peaks is pointed out.

PACS numbers: 33.80.Rv

When neutral atoms are made to interact with intense laser radiation, electrons appear in the continuum with a characteristic distribution in energy [1,2]. These energy distributions are marked by a sequence of peaks separated by the photon energy and are due to the absorption of many more photons than is necessary to overcome the threshold energy for ionization. Such distributions are now known as the above-threshold ionization (ATI) spectra. Similar spectra for negative ions are also expected to occur due to the analogous process of above-threshold detachment (ATD). The first multiphoton detachment experiment was performed by Hall, Robinson, and Branscomb [3] more than two decades ago. Theoretical studies of photodetachment in strong fields began during the same period in connection with the early efforts to investigate multiphoton processes by nonperturbative models [4–7]. Considerable theoretical work has been undertaken recently examining the ATD process in 1D and 3D systems [8–19]. Moreover, there is now a strong revival of interest in experiments [20–22] for photodetachment in strong fields. Both the ATI and the ATD spectra are "universal" in character in the sense that the overall structure does not depend on the target atom or ion chosen. An important problem in this regard lies in understanding the differences between the two types of spectra. One of the fundamental differences between atoms and the negative ions arises from the short-range nature of the potential which loosely bounds the optical electron in a negative ion as opposed to the long-range Coulomb potential experienced by the ionized electron of a neutral atom. It is now well known that the presence of the Coulomb potential introduces a complex substructure to the ATI peaks, discovered and interpreted first by Freeman *et al.* [23]. This is understood to be determined by the dynamic Stark shift of the Rydberg levels of the target atom into intermediate resonances in the presence of the field. In comparison, the one-electron ATD spectrum is expected to have a much simpler structure with single peaks separated by the photon energy. This is due to the presence only of a single bound state of the optical electron in a negative ion which precludes the occurrence of intermediate resonances. The purpose of this Letter is to report on a new universal sequence of equally spaced peaks in the ATD spectrum, in addition to the usual ATD peak sequence, which can arise despite the lack of intermediate resonances in negative ion systems.

In a classic paper on the general scattering theory, Wigner [24] investigated the relation between the threshold behavior of reaction cross sections as a function of energy and the asymptotic behavior of the interaction potential. It has been known since then that the photoionization process governed by the long-range Coulomb potential starts off abruptly with a finite nonzero cross section at the ionization threshold [25]. In contrast, the usual photodetachment process, governed by short-range potentials, starts off with a vanishingly small cross section at the detachment threshold $\epsilon=0$, with $\epsilon^{l+1/2}$ energy dependence for the lth partial wave contribution. In fact, Bryant and collaborators [21,22] have recently provided data evidencing the square-root dependence on the excess energy of the detached electrons in their ingenious multiphoton detachment experiments with relativistic H^- beams in a CO_2 laser, which permitted Doppler tuning of the laser frequency over a wide range. Here we show that a new structure in ATD spectrum will arise with a fixed-frequency laser at high intensities.

Wigner [24] and especially Baz [26] have investigated the important consequence of the unitarity constraint on the probability amplitude in a given channel at the opening of a new channel. It is known from these investigations that the reaction cross section of a given process can exhibit a singular behavior as the energy passes through the threshold of a new channel. This behavior is characterized by a discontinuous derivative of the probability density at the threshold. Furthermore, the singularity can take one of the four different shapes shown in Fig. 1. The actual shape taken in a given situation depends on the details of the strengths of the couplings involved. Such a threshold structure with a derivative discontinuity has been called a *Wigner cusp*. In fact, due to the Floquet periodicity for a given frequency of the laser, we expect a whole sequence of cusps (Wigner's comb), separat-

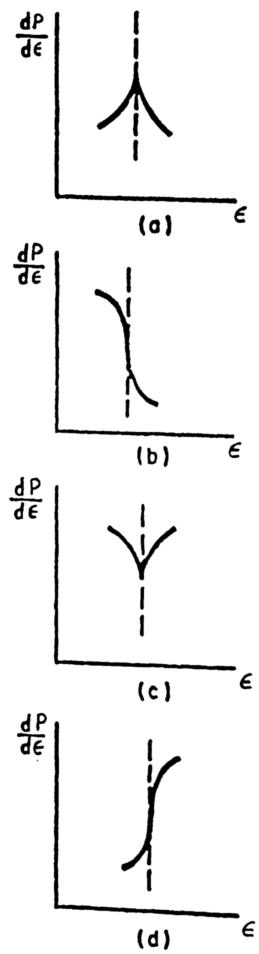

FIG. 1. The four types of Wigner cusps.

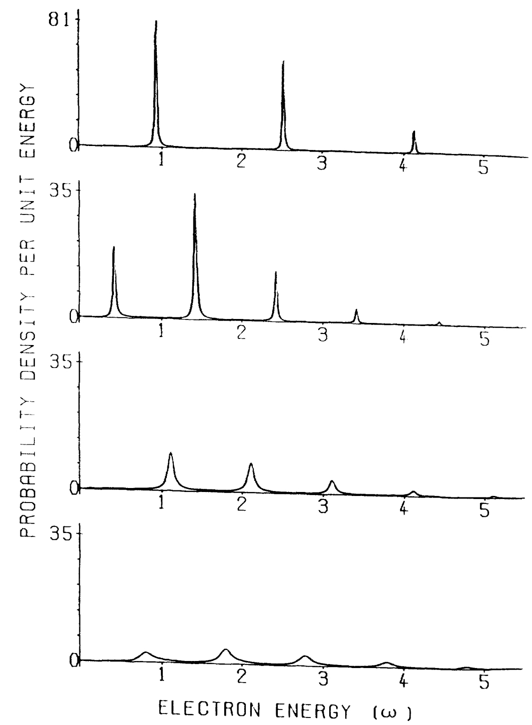

FIG. 2. The ATD spectra of H^- for four different intensities at $\omega = 0.35$ eV. From the top: $I = 1.7\times10^{11}$ W/cm^2, spectrum is "regular"; $I = 2.6\times10^{11}$ W/cm^2, exhibits "peak switching"; $I = 4.3\times10^{11}$ W/cm^2, exhibits "peak suppression"; $I = 6.0\times10^{11}$ W/cm^2, exhibits "peak disappearance."

ed exactly by the photon energy to appear in the spectrum.

A very general argument for the existence of Wigner's comb structure in the ATD spectrum is the recognition of a sequence of thresholds for physical channels, which become available for the ATD electrons. For a fixed frequency of the laser field such a sequence is provided by the stimulated multiphoton bremsstrahlung channels which open to ATD electrons in the intermediate continuum states in the presence of the field. For electrons which are ejected with kinetic energy ϵ between $\hbar\omega \le \epsilon < 2\hbar\omega$ the one-photon stimulated bremsstrahlung channel becomes open at the threshold value $\epsilon = \hbar\omega$. Similarly for electrons ejected with $2\hbar\omega \le \epsilon < 3\hbar\omega$, the two-photon stimulated bremsstrahlung channel also becomes open. In general, for ATD electrons ejected within $\hbar\omega \le \epsilon < (n+1)\hbar\omega$, all the n-photon stimulated bremsstrahlung channels will be open. Hence, while monitoring the ATD spectrum with a fixed-frequency laser we expect from the Wigner-Baz reaction theory the appearence of a comb structure (Wigner's comb) in the spectrum.

To avoid any ambiguity it should be clearly noted that the stimulated bremsstrahlung channels available to the ATD electron in the *intermediate* continuum states cannot be directly measured in the ATD spectrum, but only their "signature" in the form of threshold cusps are expected to be seen here, in accordance with the general multichannel Wigner-Baz reaction theory. For the related theory of stimulated bremsstrahlung *per se* and its inverse we direct the reader to Refs. [27] and [28] and the related works cited therein.

In order to test our prediction, based on the general arguments made above, we have carried out a series of numerical calculations for ATD spectra for a 3D-model negative ion [29], which is subjected to intense laser fields at a given frequency. The nonperturbative variational method used to calculate the ATD spectra has been described in detail by us recently [30] and will not be repeated here. We note only that for the present numerical experiments the short-range model potential is chosen with the H^- negative ion in mind and is required to yield a single s-wave bound state with a binding energy $\epsilon_b = 0.754$ eV, corresponding to the only bound state of H^-. The laser photon energy is chosen to be $\hbar\omega = 0.35$ eV, corresponding to the third harmonic of an intense CO_2 laser, and the field is chosen to be circularly polarized.

In Fig. 2 we present a sequence of ATD spectra calculated at four different laser intensities ranging (from the top) from $I=1.7\times10^{11}$ to 6.0×10^{11} W/cm^2. These spectra show typical dynamical phenomena which are expected in analogy with the ATI spectra, namely, peak shifting due to ac Stark shift, and peak reversal, suppression, and disappearance as a function of intensity. Thus at the lowest intensity $I=1.7\times10^{11}$ W/cm^2 we see a so-called regular spectrum in which the lower-energy peaks are higher than the higher-energy peaks, in accordance with the expectation based on the perturbation theory. Note that the position of the first peak, as well as of the others, is redshifted from the unperturbed position for the nominal three-photon detachment [30]. At $I=2.6\times10^{11}$ W/cm^2 one finds the first sign of the highly nonperturbative behavior known as the peak reversal, in which a peak at a lower energy becomes higher than a peak at a higher energy. At $I=4.3\times10^{11}$ W/cm^2 one observes the phenomenon of peak suppression, in which the first peak is reduced to a negligible height so that it may be considered totally suppressed in comparison with the next higher peak. In the last spectrum in this series, at $I=6.0\times10^{11}$ W/cm^2, we note the phenomenon of peak disappearance due to the ac redshift, which is large enough to displace the previously visible first peak below the detachment threshold at $\epsilon=0$ into the negative energy region. In fact, the first visible peak in this panel is due to the redshift of what was previously the second peak in the spectrum at lower intensities. One observes that throughout the sequence of spectra, the ATD peaks are broadened systematically, implying an increasing rate of decay of the initial bound state with increasing intensity. In this intensity range, however, there are as yet no observable signs of the predicted Wigner comb structure. In Fig. 3 we extend the sequence shown in Fig. 2 to higher intensities. Examination of the spectrum at the top in Fig. 3 at $I=8.5\times10^{11}$ W/cm^2 shows the first evidence of the predicted equally spaced Wigner cusp structure at the stimulated bremsstrahlung channel thresholds $\epsilon=\hbar\omega, 2\hbar\omega, 3\hbar\omega, \ldots$. Raising the intensity to $I=1.1\times10^{12}$ W/cm^2 (middle panel) clearly reveals their presence at the same energy points. They are also easily identified to have the form of the type (a) cusps of Fig. 1. The last spectrum is calculated at a still higher [31] intensity, $I=1.2\times10^{12}$ W/cm^2. The Wigner comb structure at this high intensity appears even more prominently on the shoulder of the greatly broadened ATD peaks and their wings. Finally, we emphasize the important structural property of the Wigner comb: It remains attached to the thresholds of multiphoton stimulated bremsstrahlung channels, independent of the intensities considered. This positional rigidity of the Wigner comb structure, in contradistinction to the intensity-dependent positions of the ATD peaks, therefore can be used to distinguish the former unambiguously from the latter by experiments performed at a fixed frequency but different laser intensities.

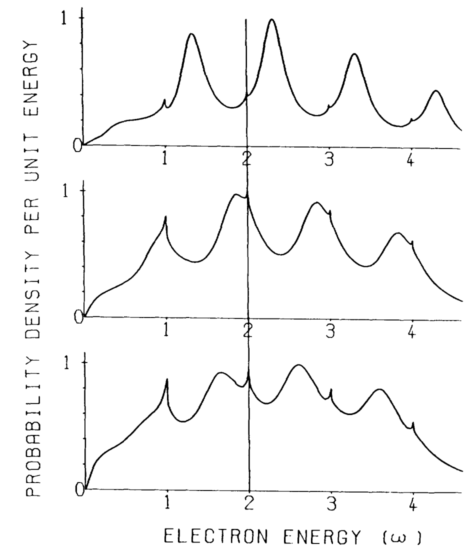

FIG. 3. Evidence for the Wigner comb structure in ATD spectra of H^- for high intensities at $\omega=0.35$ eV. From the top: $I=8.5\times10^{11}$ W/cm^2, $I=1.1\times10^{12}$ W/cm^2, and $I=1.2\times10^{12}$ W/cm^2. The Wigner comb remains attached to the thresholds of multiphoton stimulated bremsstrahlung channels, independent of the intensity of the laser field (shown at the threshold for the two-photon process).

To summarize, based on the Wigner-Baz reaction theory we predict the occurrence of a new sequence of equally spaced structure (Wigner's comb) in the above-threshold detachment (ATD) spectra obtained with a very intense fixed-frequency laser. Numerical experiments are carried out to obtain ATD spectra of a 3D model of H^- subjected to the third harmonic of CO_2 laser frequency in the intensity range $I\approx1.7\times10^{11}$ to 1.2×10^{12} W/cm^2, which confirm our general argument. The positional rigidity of the Wigner comb structure against the intensity can be used to distinguish it experimentally from the usual peaks in an ATD spectrum.

This work has been supported by Deutsche Forschungsgemeinschaft under Project No. SFB-216-M2. A preliminary version of the results reported here has been presented at the Big Sky Workshop on Super-Intense Laser-Atom Physics.

[1] P. Agostini, F. Fabre, G. Mainfray, G. Petite, and N. K. Rahman, Phys. Rev. Lett. **42**, 1127 (1979).

[2] P. Kruit, J. Kimman, and M. J. Van der Wiel, J. Phys. B **14**, L597 (1981).

[3] J. L. Hall, E. J. Robinson, and L. M. Branscomb, Phys. Rev. Lett. **14**, 1013 (1965).

[4] L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1965) [Sov. Phys. JETP **20**, 1307 (1965)].

[5] F. H. M. Faisal, J. Phys. B **5**, L89 (1973); Nuovo Cimen-

to **33B**, 775 (1976).
[6] I. J. Berson, J. Phys. B **8**, 3078 (1975).
[7] H. R. Reiss, Phys. Rev. A **22**, 1786 (1980).
[8] H. G. Muller, A. Tip, and M. J. Van der Wiel, J. Phys. B **16**, L679 (1983).
[9] B. Chen, F. H. M. Faisal, S. Jetzke, H. O. Lutz, and P. Scanzano, Phys. Rev. A **36**, 4091 (1987).
[10] C. Cerjan and R. Kosloff, J. Phys. B **20**, 4441 (1987).
[11] J. Zakerzewski and K. Zyczkowski, Phys. Rev. A **36**, 4311 (1987).
[12] L. A. Collins and A. L. Merts, Phys. Rev. A **37**, 2415 (1988).
[13] J. N. Bardsley, A. Szöke, and A. J. Comella, J. Phys. B **21**, 3899 (1988).
[14] Th. Mercouris and C. A. Nicolaides, J. Phys. B **21**, L285 (1988).
[15] W. Becker, J. K. McIver and M. Confer, Phys. Rev. A **40**, 6904 (1989).
[16] M. Dörr and R. Shakeshaft, Phys. Rev. A **38**, 543 (1988); M. Dörr, R. M. Potvliege, D. Proulx, and R. Shakeshaft, Phys. Rev. A **42**, 4138 (1990).
[17] W. G. Greenwood and J. H. Eberly, Phys. Rev. A **43**, 525 (1991).
[18] F. H. M. Faisal, P. Filipowicz, and K. Rzazewski, Phys. Rev. A **41**, 6176 (1990).
[19] M. Crance, J. Phys. B **23**, L285 (1990); **21**, 3559 (1988).
[20] R. Trainham, G. D. Fletcher, N. B. Mansour, and D. J. Larson, Phys. Rev. Lett. **59**, 2291 (1987).
[21] C. Y. Tang, P. G. Harris, A. H. Mohagheghi, H. C. Bryant, C. R. Quick, J. B. Donahue, R. A. Reeder, S. Cohen, W. W. Smith, and J. E. Stewart, Phys. Rev. A **39**, 6068 (1989).
[22] C. Y. Tang, H. C. Bryant, P. G. Harris, A. H. Mohagheghi, R. A. Reeder, H. Sharifian, H. Toutounchi, C. R. Quick, J. B. Donahue, S. Cohen, and W. W. Smith, in *Multiphoton Processes*, edited by G. Mainfray and P. Agostini (SPA, CE Saclay, France, 1990), p. 69: C. Blondel, M. Crance, C. Delsart, and A. Giraud, in *Multiphoton Processes*, *ibid.*, p. 59.
[23] R. R. Freeman, T. J. McIlrath, P. H. Bucksbaum, and M. Bashkanski, Phys. Rev. Lett. **57**, 3156 (1986).
[24] E. P. Wigner, Phys. Rev. **73**, 1002 (1948).
[25] H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Systems* (Springer-Verlag, Berlin, 1957).
[26] A. I. Baz, Zh. Eksp. Teor. Fiz. **33**, 923 (1958) [Sov. Phys. JETP **6**, 709 (1958)]; see also G. Breit, Phys. Rev. **107**, 1612 (1957); L. D. Landau and G. M. Lifschitz, *Quantum Mechanics* (Pergamon, Oxford, 1975), pp. 565–570.
[27] F. H. M. Faisal, Comments At. Mol. Phys. **15**, 119 (1984).
[28] L. Dimou and F. H. M. Faisal, Phys. Rev. Lett. **59**, 872 (1987).
[29] The role of dimensions for threshold behavior and cusp formation is significantly different for different dimensions since the density of states in one and three dimensions changes from $\epsilon^{-1/2}$ to $\epsilon^{1/2}$, respectively.
[30] F. H. M. Faisal and P. Scanzano, Comput. Phys. Commun. **63**, 265 (1991). This method is based on a variational representation of the binding potential combined with a Fourier transformation method to solve the initial value problem and the energy spectrum. The transition amplitudes calculated by this method satisfy Schwinger's variational principle.
[31] At this intensity the quiver radius of the free electron, $\alpha_0 = E_0/\omega^2 \simeq 5.8$ a.u., is significantly greater than unity.