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LETTER TO THE EDITOR

Near-threshold behaviour of multiphoton ionisation probabilities

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Abstract. The near-threshold behaviour of four-photon ionisation is studied in an energy region where the third laser photon crosses the Rydberg threshold. Particular emphasis is given to the role played by the Stark shift (raising of the ionisation potential) in a Gaussian laser pulse. As a function of the laser frequency the ionisation probability shows (i) a region where individual Rydberg resonances are well resolved, followed by (ii) a smoothly decreasing ionisation probability in the bound-state region and (iii) an almost energy-independent above-threshold ionisation signal.

Motivated by recent experiments (van Linden van den Heuvell *et al* 1987, Noordam *et al* 1988, Freeman 1989) we report in this letter a theoretical study of N-photon resonant $N+1$-photon ionisation of atoms where the Nth photon is tuned close to the ionisation threshold. We address the questions of intensity and frequency dependence of transition amplitudes close to the threshold when both an infinite number of densely spaced Rydberg levels and the continuum states participate in the laser induced dynamics. Our emphasis is on the role played by the AC Stark shifts and the time and space dependence of the ionising laser field.

In our theoretical model we consider four-photon ionisation with the third laser photon tuned close to threshold. For the electric field of the laser pulse we write $E(\rho, t) = \mathscr{E}(\rho, t)\varepsilon\, \mathrm{e}^{-\mathrm{i}\omega t} + \mathrm{CC}$ with $\mathscr{E}(\rho, t)$ the time- and space-dependent amplitude, ω the frequency of the light and ε the polarisation vector. The pulse amplitude is assumed to be a Gaussian with duration τ_{p} and focal waist ρ_{foc}, $\mathscr{E}(\rho, t) = \mathscr{E}_0 \exp[-2\ln 2(t/\tau_{\mathrm{p}})^2 - (\rho/\rho_{\mathrm{foc}})^2]$. The quantity of interest in the present work is the four-photon ionisation probability averaged over the laser interaction volume, $P = \int\int P(\varepsilon, \rho)\rho\, \mathrm{d}\rho\, \mathrm{d}\varepsilon / \int \rho\, \mathrm{d}\rho$, where ε denotes the energy of the continuum electrons, and we assume the atoms to be equally distributed over the laser focus. When the first excitation step of the atom is treated in lowest order perturbation theory we find (all expressions below will be expressed in atomic units)

$$P(\varepsilon, \rho) = 2\pi \int_{-\infty}^{\infty} \mathrm{d}t |\langle\varepsilon|D(\rho, t)|\Psi^a(t)\rangle|^2 \qquad (1)$$

with the transition amplitude from the ground state $|\mathrm{g}\rangle$ to the continuum state $|\varepsilon\rangle$

$$\langle\varepsilon|D(\rho, t)|\Psi^a(t)\rangle$$
$$= -\mathrm{i} \sum\!\!\!\!\!\!\int \mathrm{d}n \langle\varepsilon|D(\rho, t)|n\rangle \exp[-\mathrm{i}(\varepsilon_n t + a(\rho, t))]$$
$$\times \int_{-\infty}^{t} \mathrm{d}t' \exp\{-\mathrm{i}[(\bar{\varepsilon} - \varepsilon_n)t' - a(\rho, t')]\}\langle n|D_{\mathrm{eff}}(\rho, t')|\mathrm{g}\rangle. \qquad (2)$$

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Here $\bar{\varepsilon} = \varepsilon_g + 3\omega$ is the energy of the mean excited Rydberg state, $\langle n|D_{\text{eff}}(\rho, t)|g\rangle$ denotes the effective three-photon matrix element from the ground state to the Rydberg state; it scales as $n^{-3/2}$ close to the Rydberg threshold. The quantity $a(\rho, t)$ is defined as

$$a(\rho, t) = \int_{-\infty}^{t} \mathrm{d}t'[\delta\omega_n(\rho, t') - \delta\omega_g(\rho, t') - \tfrac{1}{2}\mathrm{i}\gamma_n(\rho, t')] \tag{3}$$

with $\delta\omega_n$ the AC Stark shift and γ_n the ionisation rate of the state $|n\rangle$ to the continuum in agreement with Fermi's golden rule. $\delta\omega_g$ is the shift of the ground state. In the energy region much less than a photon energy away from threshold the n dependence of the AC Stark shift $\delta\omega_n$ has the form (Giusti-Suzor and Zoller 1987) $\delta\omega_n = \mathscr{E}^2/\omega^2 + 1/n^3\mu\mathscr{E}^2$, where the first term is the familiar shift of the ionisation threshold and the second term drops off with n^3 (the quantity μ is almost constant as a function of energy and is related to a laser-induced quantum defect). With similar arguments we can show that γ_n scales as n^{-3}.

As far as the dynamics of the laser excitation process is concerned we may distinguish between two limiting cases. Sufficiently far below threshold the spectral width of the laser pulse $1/\tau_p$ is much less than the energy separation between adjacent Rydberg states and the four-photon ionisation process involves one isolated Rydberg state as an intermediate three-photon resonance. The lineshape of these resonances strongly depends on the space and time dependence of the Stark shift $\delta\omega(r, t) = \mathscr{E}(r, t)^2/\omega^2$, if $\delta\omega_{\max}\tau_p > 1$. Very close to threshold the spectral width of the laser pulse is much larger than the energy level separation $\Delta E_{\bar{n}} = 1/\bar{n}^3$ with $\bar{n}$ the mean excited principal quantum number. In the time domain this corresponds to the limit where the laser pulse duration is much shorter than the classical orbit time $T_{\bar{\varepsilon}} = 2\pi\bar{n}^3$ of the Kepler orbit. In this case a large number of Rydberg levels contribute to the ionisation probability. Physically, this means that the ionisation of the created radial wavepacket takes place before the wavepacket leaves the core (direct four-photon ionisation). Here it is more appropriate to write the transition amplitudes in terms of a multiple scattering expression,

$$\begin{aligned}
&\langle\varepsilon|D(\rho, t)|\Psi^a(t)\rangle \\
&\quad = -\mathrm{i}\sum_{m=1}^{\infty}\int_{n_0}^{\infty} \mathrm{d}n\langle\varepsilon|D(\rho, t)|n\rangle \exp[-\mathrm{i}(\varepsilon_n t + a(\rho, t))]\exp(2\mathrm{i}\pi mn) \\
&\qquad \times \int_{-\infty}^{t} \mathrm{d}t' \exp\{-\mathrm{i}[(\bar{\varepsilon} - \varepsilon_n)t' - a(\rho, t')]\}\langle n|D_{\text{eff}}(\rho, t')|g\rangle \\
&\qquad -\mathrm{i}\int_{\varepsilon_0}^{\infty} \mathrm{d}\tilde{\varepsilon}\langle\varepsilon|D(\rho, t)|\tilde{\varepsilon}\rangle \exp[-\mathrm{i}(\tilde{\varepsilon}t + a(\rho, t))] \\
&\qquad \times \int_{-\infty}^{t} \mathrm{d}t' \exp\{-\mathrm{i}[(\bar{\varepsilon} - \tilde{\varepsilon})t' - a(\rho, t')]\}\langle\tilde{\varepsilon}|D_{\text{eff}}(\rho, t')|g\rangle
\end{aligned} \tag{4}$$

which expresses the transition amplitude as a sum over contributions $m = 1, 2, \ldots$ of periodic classical trajectories (Alber and Zoller 1988). For $\tau_p \ll T_{\bar{\varepsilon}}$ we can perform the energy integration in (4) in a stationary phase approximation with the result that only the $m = 0$ term contributes. This term is identical to what one finds for above-threshold ionisation, and gives rise to an approximately energy-independent four-photon ionisation signal. Note, however, that there is an ambiguity in defining a Rydberg energy spacing $\Delta E_{\bar{n}}$ (or equivalently, a classical orbit time T) because the effective ionisation threshold varies according to the time and space dependence of the laser field. At the beginning (and at the end) of the pulse (when the Stark shift is negligible) Rydberg states around $\bar{\varepsilon} \simeq \varepsilon_n$ are excited. Thus the condition $T_{\varepsilon_n} = \tau_p$ defines a principal quantum number n_{max}. In the course of the laser pulse the ionisation threshold shifts upwards with increasing light intensity and the laser is tuned into resonance with lower-lying states. At maximum intensity Rydberg levels around $\bar{\varepsilon} \simeq \varepsilon_n + \delta\omega_{max}$ are excited. Again the condition $T_{\varepsilon_n} = \tau_p$ defines a principal quantum number n_{min} (which is smaller than n_{max}). Excitation of states above n_{min} corresponds to formation of a wavepacket. In the frequency interval between n_{min} and n_{max} we expect a transition between a region with well resolved bound-state resonances and a flat structureless quasicontinuum part.

We have numerically evaluated the ionisation probability as given by (1) and (2) for a Gaussian laser pulse. In figures 1–3 we plot the ionisation probability (in arbitrary units) as a function of the excitation energy $\bar{\varepsilon}$ (measured relative to the zero-field

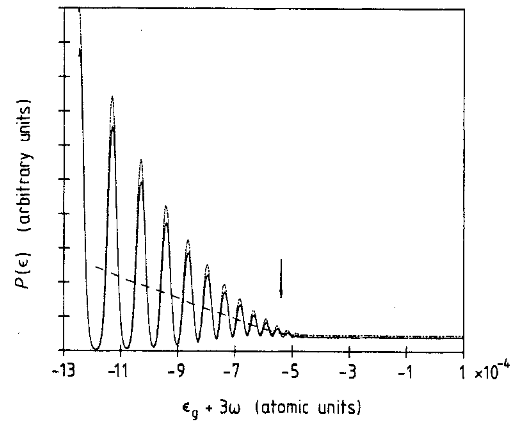

Figure 1. Ionisation probability for a spatially constant (chain curve) and a Gaussian (full curve) laser pulse. The broken curve denotes the ionisation probability averaged between adjacent Rydberg resonances. The parameters are $\tau_p = 4.1$ ps, $I = 1.6 \times 10^{10}$ W cm^{-2}, $\delta\omega_{max} = 0.14$ meV (5.14×10^{-6} au), $\delta\omega_{max}\tau_p = 0.8$.

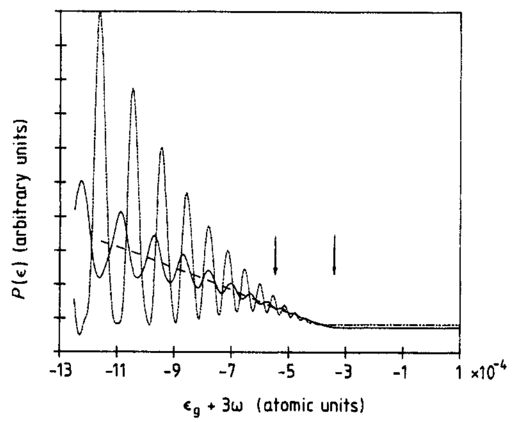

Figure 2. Same as figure 1 with $I = 6.3 \times 10^{11}$ W cm^{-2}, $\delta\omega_{max} = 5.5$ meV (2.04×10^{-4} au), $\delta\omega_{max}\tau_p = 34$.

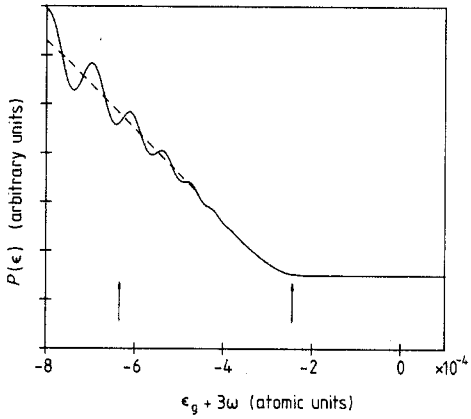

Figure 3. Same as figure 1 with $I = 1.21 \times 10^{12}$ W cm^{-2}, $\delta\omega_{max} = 10.65$ meV, (3.91×10^{-4} au), $\delta\omega_{max}\tau_p = 57$, $\tau_p = 3.5$ ps.

ionisation threshold). The durations of the laser pulses are $\tau_p = 4.1$ ps, 4.1 ps and 3.5 ps and the laser intensities are $I = 1.6 \times 10^{10}$ W cm^{-2}, 6.3×10^{11} W cm^{-2} and $I = 1.21 \times 10^{12}$ W cm^{-2} in figures 1, 2 and 3, respectively. This corresponds to Stark shifts $\delta\omega_{max} = 0.14$ meV (5.14×10^{-6} au), $\delta\omega_{max} = 5.5$ meV (2.04×10^{-4} au) and $\delta\omega_{max} =$ 10.65 meV (3.91×10^{-4} au) or $\delta\omega_{max}\ \tau_p = 0.8$, $\delta\omega_{max}\ \tau_p = 34$ and $\delta\omega_{max}\ \tau_p = 57$ (ignoring the shift of the ground state). For $\delta\omega_{max}\tau_p > 1$ the process is dominated by the effects of the AC Stark shift. The above parameters have been chosen to demonstrate various typical lineshapes. In figures 1-3 the chain and full curves correspond to a spatially constant and Gaussian laser beam, respectively. The broken curves indicate a frequency average over resonance profiles in the spatially inhomogeneous laser beam. In figure 1 we have $\delta\omega_{max} \ll 1/2(\tau_p/2\pi)^{-2/3}$, e.g. the shift of the effective ionisation threshold is negligible. However, as $\delta\omega_{max}\tau_p = 0.8$ the space and time dependence of the threshold shift manifests itself in the lineshape of the resonances. In figures 2 and 3 the laser pulse is so intense that the lineshapes strongly depend on the details of the pulse form and the effective threshold is significantly shifted.

According to these figures we can distinguish three regions of different physical behaviour. Far below threshold the resonance structure is well resolved with the maxima of the resonance profiles falling off as n^{-6} as long as the Stark shift is negligible. This can be understood from the $n^{-3/2}$ scaling of the bound-bound and bound-free transition matrix elements. A Stark shift of the order of the level spacing leads to a slower decay. Closer to threshold we have an energy region where individual Rydberg resonances are no longer resolved. Here the ionisation probability is still a decreasing function of frequency in agreement with the extrapolated curve obtained by averaging over the resonances (broken curves). Finally this is followed by a flat continuum which smoothly joins the region of above-threshold ionisation. The arrows in figures 2-3 indicate the positions of n_{min} and n_{max} defined above, which agree well with the border lines of the three regions mentioned above. From figure 3 we can see that our results are in agreement with experiment (Noordam *et al* 1988).

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