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PAPER

Laser stripping of hydrogen atoms by direct ionization

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Abstract

Direct ionization of hydrogen atoms by laser irradiation is investigated as a potential new scheme to generate proton beams without stripping foils. The time-dependent Schrödinger equation describing the atom-radiation interaction is numerically solved obtaining accurate ionization cross-sections for a broad range of laser wavelengths, durations and energies. Parameters are identified where the Doppler frequency up-shift of radiation colliding with relativistic particles can lead to efficient ionization over large volumes and broad bandwidths using currently available lasers.

1. Introduction

The photo-ionization of atoms exposed to strong electromagnetic fields has been the subject of extensive research over many decades [1–5]. Following the invention of lasers, comprehensive theoretical and experimental studies of laser-matter interaction at high intensities have been carried out in the infrared spectral region, uncovering a highly nonlinear regime where ionization occurs through the absorption of a large number of low-energy photons. Recently, ionization by intense UV radiation has also become a reality [6, 7] thanks to the development of new sources based on free-electron lasers, high harmonic generation and plasmas. New interest in this topic has also been sparked by the rapidly developing field of laser stripping of particle beams, a process where electrons are removed from a relativistic beam by irradiation with intense lasers [8–11]. Proton beams can be obtained by passing a H or H^- beam through a thin foil, but often with the drawbacks of significant beam losses, quality degradation and frequent maintenance of foils that can become activated. Uncontrolled beam loss at injection is one of the main challenges encountered in the development of high-intensity, high-energy proton accelerators [12]. As an alternative, multi-step stripping schemes have been proposed [10, 13–15], where lasers excite transitions between bound states of hydrogen, resulting in weakly bound electrons that are easily removed by magnetic fields. Here we show that direct ionization of hydrogen can also be a viable single-step stripping method, since in the rest frame of an atom moving at relativistic speed a colliding infrared laser can be Doppler shifted to UV frequencies where photo-ionization is highly efficient at intensities achievable with commercially available lasers. Since accurate ionization cross-sections are not known in the UV region for a wide range of frequencies and pulse durations, we have conducted an extensive numerical study by solving the time-dependent Schrödinger equation (TDSE) governing the atom-radiation interaction, exploring the properties of one-photon to five-photon ionization and discussing possible applications to laser stripping, using as a test case the beam parameters of Project X, a proton accelerator which was proposed at Fermilab [16].

2. Ionization probabilities

The probability of ionizing hydrogen atoms through exposure to intense laser radiation has been calculated by numerically solving the TDSE using the publicly available code QPROP [17]. This is a package developed to study the non-relativistic atom-field interaction in the dipole approximation, which is applicable when the energy gained by electrons is smaller than their rest energy, a condition met when the dimensionless normalized vector

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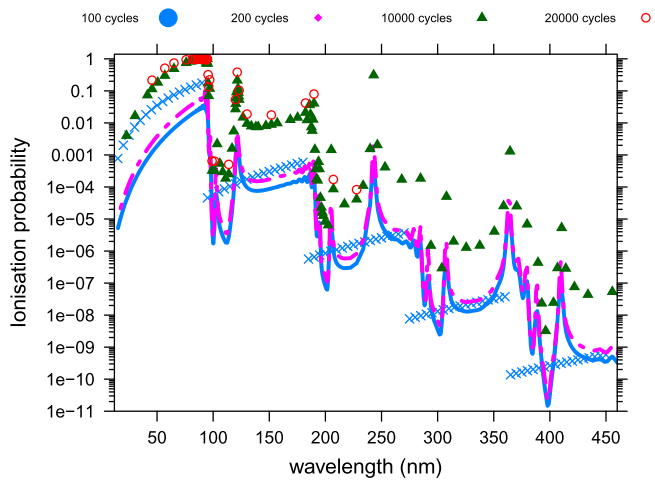

Figure 1. Ionization probability of atomic hydrogen as a function of laser wavelength at an intensity of 10^{12} W cm^{-2} and for pulse durations of 100 (continuous line), 200 (dashed line), 10 000 (triangles) and 20 000 (circles) cycles. (Fewer points have been calculated for long pulse durations because of the long computational time.) The crossed line shows the ionization probability for 100 cycles predicted by the analytical formula derived in [23].

potential of the laser-field $a_0 = |eA|/mc \approx 8.5 \times 10^{-10}\,\lambda\,(\mu\text{m})\sqrt{I_0\,(\text{W cm}^{-2})}$ is less than 1. Here e is the electron charge magnitude, A the laser-field vector potential, m the electron mass and c the speed of light. This approach restricts the laser intensity I_0 to values below $\sim 10^{18}$ W cm^{-2} for wavelengths $\lambda \approx 1\ \mu$m, or $\sim 10^{20}$ W cm^{-2} for $\lambda \approx 100$ nm. Non-dipole features can however be induced by the laser magnetic field and by retardation effects at intensities about one order of magnitude lower [18, 19]. In this work only intensities up to 10^{16} W cm^{-2} have been considered, a range appropriate for laser stripping, where high irradiances over large volumes would be difficult to achieve and would potentially lead to degradation of laser and particle beam quality.

TDSE simulations have been performed using a grid with 500 points in the radial direction and a step size of 0.2 au. Electrons reaching the boundaries are absorbed, causing the norm $\mathcal{N}(t)$ of the wave function to decrease with time. The ionization probability is calculated as $\mathcal{P} = 1 - \mathcal{N}(t)$. The number of angular momenta, which corresponds to the maximum number of absorbed photons, has been set to 5 for wavelength ranges where ionization is dominated by one or two photon absorption, and 6–8 in the three–five photon regimes. These parameters offer a good compromise between accuracy and computational speed and their validity has been verified by repeating some calculations for 10 angular momenta and by computing the photoelectron spectra. All simulations have been performed for a linearly polarized $\sin^2$ pulse where the electric field is described by the time profile $E(t) = E_0 \sin^2\left(\frac{\omega t}{2N_c}\right)\cos(\omega t)$, with ω the radiation angular frequency and N_c the number of cycles, corresponding to a pulse duration $T_{\text{FWHM}} = 2(\pi - 2\arcsin(2^{-1/4}))\,N_c/\omega \approx 2.29\,N_c/\omega$ (FWHM of the intensity). Our results have been compared with the TDSE simulations of [20, 21], as well as with the experimental measurements reported in [22], finding an excellent agreement.

The dependence of the ionization probability on laser wavelength is shown in figure 1 for pulses with intensity of 10^{12} W cm^{-2} and durations of 100, 200, 10 000 and 20 000 cycles. Five different regions can be identified. For wavelengths shorter than 91nm the photon energy is larger than the 13.6eV electron-proton binding and ionization is driven predominantly by the absorption of a single photon. Here the ionization probability is highest, reaching 1 in the 80–90 nm window for intensities around 10^{11}–10^{12} W cm^{-2} and picosecond-scale pulse durations. When saturation does not occur, the ionization probability proportionally decreases with frequency as $\omega^{-4.6}$, a behaviour in agreement with the $\omega^{-9/2}$ dependence expected at high frequencies from calculations of the one-photon ionization of hydrogen in the first-order Born approximation [24]. Past the 91.1 nm resonance, a sharp dip at about 95–100 nm leads to a new region dominated by two-photon ionization and characterized by a plateau of roughly constant amplitude up to about 190 nm. Peaks corresponding to the 102.5 nm (1s–3p) and 121.5 nm (1s–2p) transitions are prominent. Dips around list 190, 270 and 370 nm signal the passage to the three, four and five photon regimes. Calculations have been repeated for different laser intensities observing the same general behaviour, with differences only in the saturation threshold and in the amplitude of the oscillations close to resonance. A comparison with the results predicted by the analytical model presented in [23] (plotted in crosses in figure 1 for 100 cycle pulses) reveals similar general features, although the analytical formula cannot reproduce the resonances and also appears to overestimate the ionization probability at short wavelengths, probably because it employs a perturbative expansion for the Coulomb correction which requires $\omega \ll 1$ au.

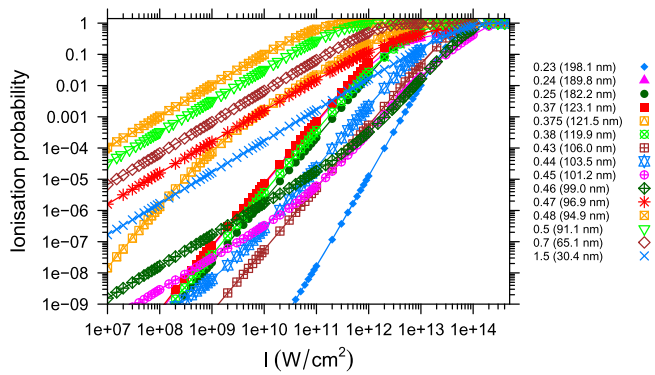

Figure 2. Ionization probability of atomic hydrogen as a function of laser intensity for 10 000 cycle pulses of varying frequency. For $\omega > 0.5$ au ($\lambda < 91.1$ nm) the ionization probability depends linearly on intensity. For 0.25 au $< \omega <$ 0.5 au (91.1 nm $< \lambda <$ 182.2 nm) the typical dependence is quadratic. For 0.125 au $< \omega <$ 0.25 au (182.2 nm $< \lambda <$ 273.4 nm) the dependence is cubic. Mixed behaviour is observed at the transition between different regimes.

Table 1. Effective n-photon ionization cross-sections σ_n for 10 000 cycle laser pulses of varying frequency interacting with atomic hydrogen.

n	λ (nm)	ω (au)	σ_n (cm^{2n} s^{n-1})
1	30.4	1.5	2.88×10^{-19}
1	50.6	0.9	1.25×10^{-18}
1	65.1	0.7	2.52×10^{-18}
1	86	0.53	5.28×10^{-18}
1	91.1	0.5	6.21×10^{-18}
1	93	0.49	1.12×10^{-17}
2	106	0.43	2.00×10^{-51}
2	123.1	0.37	1.62×10^{-49}
2	151.9	0.3	1.10×10^{-50}
2	182.2	0.25	1.55×10^{-50}
3	198.1	0.23	9.67×10^{-84}

The ionization probability versus the laser intensity I is shown in figure 2 for 10 000 cycle pulses and a range of wavelengths corresponding to the one–three photon regimes. Most curves follow the expected I^n dependence [2], where n is the number of absorbed photons, although a mixed behaviour is observed at the transition between different ionization regimes, in the wavelength regions close to the sharp dips visible in figure 1. Here more photons are absorbed as the intensity increases, as noticeable for example in the curves at 101.2 nm ($\omega = 0.45$ au) and 99 nm ($\omega = 0.46$ au). The curve at the 121.5 nm ($\omega = 0.375$ au) resonance is also atypical, since it is characterised by a quadratic growth at low intensities followed by a linear growth at high intensities. Similar features are observed for femtosecond and picosecond durations, although shorter pulses exhibit small-scale variations that are smoothed away for longer durations.

These numerical results can be used to calculate generalized multi-photon cross-sections, which include the small modifications to the ionization mechanism caused by atomic-level resonances induced by the rapidly evolving laser field [25]. Effective cross-sections can be estimated by fitting the curves derived from TDSE simulations to the equation $\mathcal{P} = 1 - \exp\left(-\sigma_n t_{\rm eff}\,[I/(\hbar\omega)]^n\right)$, where $\mathcal{P}$ is the ionization probability, σ_n is the n-photon effective cross-section and $t_{\rm eff} = \int_{-\infty}^{+\infty} F(t')^n {\rm d}t'$ is the effective pulse duration tailored to an n-photon process [26], with $F(t) = I(t)/I$ the normalized laser-pulse intensity profile and $\hbar$ the reduced Planck constant. For the $\sin^2$ pulse considered here $F(t) = \sin^4\left(\frac{\omega t}{2N_c}\right)$ and $t_{\rm eff} = 2\sqrt{\pi}\,\Gamma(2n + 1/2)N_c/(\omega\Gamma(2n + 1))$, with Γ the gamma function. Effective cross-sections for a range of wavelengths are presented in table 1 for 10000 cycle pulses and in table 2 for 100 cycle pulses.

Below saturation the ionization probability depends linearly on the pulse duration, as shown in figure 3 for $\lambda = 91.1$ nm and $\lambda = 182.2$ nm. Close to the 91.1 nm resonance saturation is already appreciable at 10^{11} W cm^{-2}, and the dependence on pulse duration becomes nonlinear. For intensities and durations where the linear scaling holds, the ionization rate can be obtained by dividing the probability by the pulse duration.

Table 2. Effective n-photon ionization cross-sections σ_n for 100 cycle laser pulses of varying frequency interacting with atomic hydrogen.

n	λ (nm)	ω (au)	σ_n (cm^{2n} s^{n-1})
1	30.4	1.5	2.88×10^{-19}
1	50.6	0.9	1.25×10^{-18}
1	65.1	0.7	2.52×10^{-18}
1	86	0.53	5.22×10^{-18}
1	91.1	0.5	5.98×10^{-18}
1	93	0.49	6.56×10^{-18}
2	106	0.43	2.04×10^{-51}
2	123.1	0.37	2.33×10^{-49}
2	151.9	0.3	1.10×10^{-50}
2	182.2	0.25	1.40×10^{-50}
3	198.1	0.23	1.13×10^{-83}

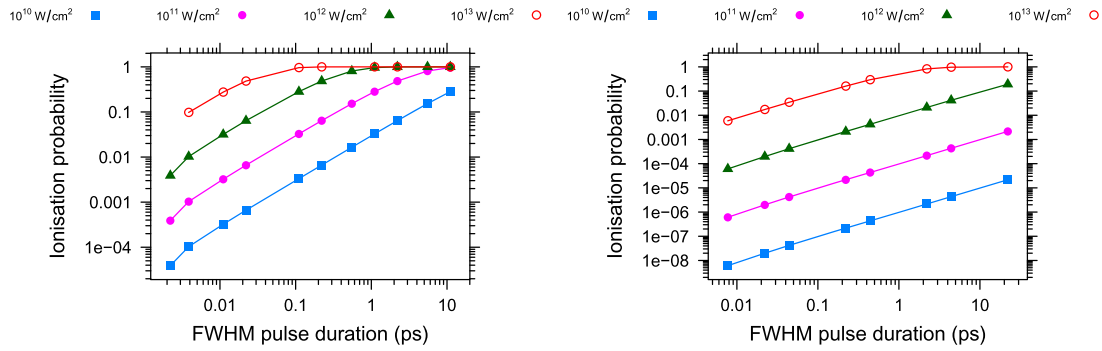

Figure 3. Ionization probability of atomic hydrogen versus laser pulse duration for $\lambda = 91.1nm$ ($\omega = 0.5$ au) (left) and $\lambda = 182.2$ nm ($\omega = 0.25$ au) (right) at intensities of 10^{10} W cm^{-2} (squares), 10^{11} W cm^{-2} (filled circles), 10^{12} W cm^{-2} (triangles) and 10^{13} W cm^{-2} (empty circles).

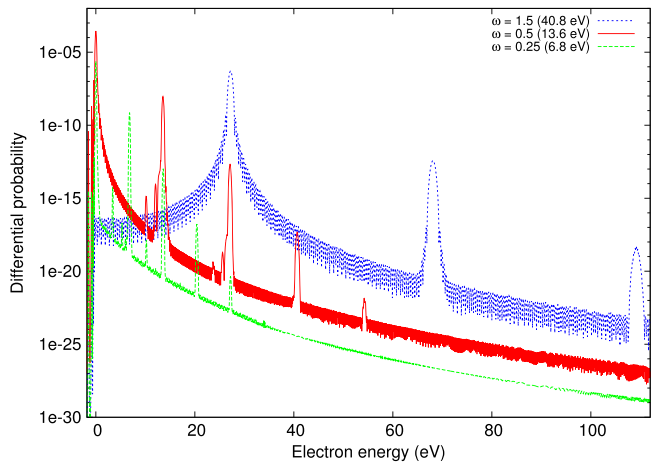

Figure 4. Photoelectron spectra generated in atomic hydrogen by a laser with intensity 10^{12} W cm^{-2}, pulse duration of 100 cycles and frequency $\omega = 1.5$ au ($\lambda = 30.4$ nm, $E = 40.8$ eV, dotted line), $\omega = 0.5$ au ($\lambda = 91.1$ nm, $E = 13.6$ eV, continuous line) and $\omega = 0.25$ au ($\lambda = 182.2$ nm, $E = 6.8$ eV, dashed line).

Figure 4 shows photoelectron spectra calculated for 100 cycle pulses with intensity 10^{12} W cm^{-2} for $\lambda = 30.4$ nm ($\omega = 1.5$ au), $\lambda = 91.1$ nm ($\omega = 0.5$ au) and $\lambda = 182.2$ nm ($\omega = 0.25$ au). Above-threshold ionization peaks [27] at energies $\mathcal{E}_n = -|\mathcal{E}_{1s}| + n\hbar\omega$ are present, showing that more photons than required may be absorbed. For the high frequencies considered here, the ponderomotive shift by $-E_0^2/(4\omega^2)$ normally observed with infrared lasers is very small.

3. Laser stripping

The cross-sections obtained from the numerical solutions of the TDSE have been used to explore the feasibility of laser stripping of H beams by direct ionization. Calculations of the ionization fraction in a volume of hydrogen atoms have been performed for a reference particle beam with parameters matching the proposed Project X at Fermilab [16], which if realized would have delivered a beam with 8 GeV ($\gamma = 9.526$) energy, transverse size $\sigma_r = 1.5$ mm and temporal profile consisting of 15 ps micropulses at 325 MHz emitted in bursts of 1.25 ms macropulses duration and 5 Hz repetition rate.

For moderate laser intensities and high-energy particles, the interaction is not expected to significantly alter the properties of the two beams. Therefore the efficiency of laser stripping over an extended volume is obtained by directly mapping the laser-intensity spatial distribution into ionization fraction using the cross-sections derived in the previous section. In general, high laser intensities not only induce non-dipole and relativistic effects in the atomic response to the radiation, as discussed in the previous section, but they also affect the laser propagation by stimulating nonlinear processes such as self-focusing and self-phase modulation, which can alter the laser-pulse spatial and spectral properties, potentially leading to beam filamentation or defocusing [28]. At relativistic intensities refractive-index variations caused by electron-mass changes can introduce additional nonlinear processes such as laser-pulse self-steepening and relativistic self-focusing [29]. For applications to laser stripping, where high efficiencies should be achieved over large volumes, it is however preferable to work with loosely focused laser beams of modest intensities and in the following discussion ionisation induced effects on the laser propagation are not included.

When laser radiation with wavelength λ_l collides with particles moving at speed $v = \beta c$, the wavelength in the particle rest frame is Doppler-shifted to $\lambda_{\mathrm{PF}} = \lambda_l/[\gamma(1 + \beta\cos\theta)]$, with $\gamma = 1\big/\sqrt{1-\beta^2}$ the relativistic factor, θ the angle in the lab frame between laser and particle trajectory and c the speed of light. This frequency up-shift is accompanied by an intensity boost, since in the particle frame the photon energy increases and the pulse duration shortens. An intensity I_0 in the lab frame transforms into $I_{\mathrm{PF}} = (1 + \beta\cos\theta)^2\gamma^2 I_0$ in the particle frame. For a head-on collision ($\theta = 0$) $I_{\mathrm{PF}} = (1+\beta)^2\gamma^2 I_0 \approx 4\gamma^2 I_0$, with an intensity boost by ~360 times for $\gamma = 9.526$. Such a counter-propagating geometry is advantageous for laser stripping applications, since a single laser pulse can interact with a large portion of a particle beam train. Furthermore, an infrared laser will be Doppler-shifted towards shorter wavelength where ionization is more efficient. For example, a 1–1.7 μm laser colliding with an 8 GeV beam will be shifted to 50–90 nm, an optimum spectral region as shown by the ionization probabilities presented in figure 1.

The ionization fraction in a volume of hydrogen atoms has been estimated for a laser beam described by a Gaussian intensity distribution $I(r, z) = I_0 \frac{w_0^2}{w(z)^2}\exp\left[-2\frac{r^2}{w(z)^2}\right]$, where w_0 is the beam waist at the focus, $w(z) = w_0\sqrt{1 + \left(\frac{\lambda_l z}{\pi w_0^2}\right)^2}$ and r and z are the transverse and longitudinal coordinates respectively. For a head-on collision the ionization probability depends weakly on the longitudinal properties of the particle beam and a uniform distribution is used with full width in the rest frame $2\sigma_t\gamma c = 90$ mm for $\sigma_t = 15$ ps. The transverse distribution is assumed Gaussian, $\rho(r) = \rho_0\exp\left(-\frac{r^2}{\sigma_r^2}\right)$, with $\sigma_r = 1.5$ mm.

Figure 5 shows the radiation energy versus wavelength (both in the particle rest frame) required to achieve 99% ionization fraction in a $6 \times 6 \times 90$ mm^3 volume of hydrogen atoms after interaction with a counter-propagating laser beam focused at the center of the region with waist $w_0 = 3$ mm and pulse duration 100, 200, 10 000 and 20 000 cycles. The highest efficiency is achieved in a wavelength window corresponding to the single-photon regime of ionization, around 50–100 nm, where the required energy is lowest and largely independent of pulse duration. At wavelengths shorter than ~40nm, however, 99% ionization cannot be reached for femtosecond-scale pulses, because the ionization probability is reduced by the onset of stabilization [30]. On the contrary, the ionization probability for two or more photon absorption is higher for short pulses, but the energy required is at least one order of magnitude larger. Figures 5 and 6 show that in the optimum wavelength window the required laser energy depends weakly on beam waist and looser focusing can be employed, allowing for longer interaction lengths with trains of particle pulses. Since the laser energy is boosted by the photon-frequency up-shift in the particle frame, for example by ~19 times for $\gamma = 9.526$ and head-on collision, a 100 mJ laser can provide efficient ionization over a large bandwidth, accommodating also for a few percent energy spread in the particle beam.

The long longitudinal extent of particle beams is one of the main challenges for effective laser stripping. The Rayleigh length $z_r = \pi w_0^2/\lambda$ of an infrared laser with waist 3–4 mm is of the order of 25–50 m, therefore over a length $L = 2z_r$ a single laser pulse can interact with a slice $2\,L/c \approx$ 300–600 ns of a particle beam, where the factor 2 is due to the counter-propagating geometry. If beam quality can be maintained, the interaction length can be further extended using a cavity. The intensity of a laser beam bouncing between two mirrors decreases in time according to the formula $I(t) = I_0\exp(-t/\tau_c)$, with $\tau_c = \frac{2L}{c(1 - R_1R_2)}$ and $R_{1,2}$ the mirror reflectivity. Assuming

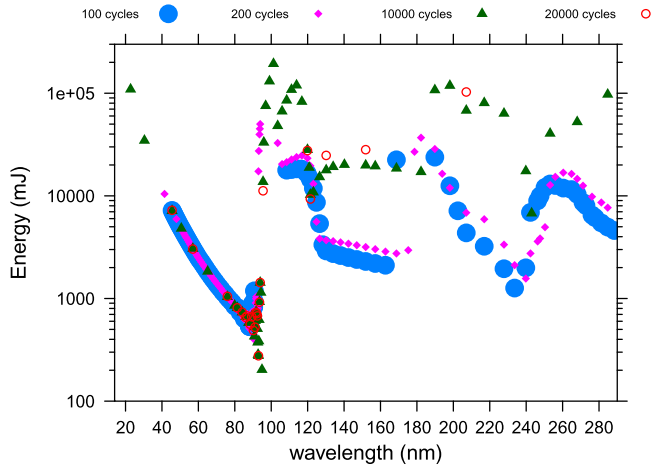

Figure 5. Laser energy in the particle rest frame required to achieve 99% ionization fraction in a $6 \times 6 \times 90$ mm^3 volume of hydrogen atoms characterized by a uniform longitudinal distribution and transverse Gaussian distribution with size $\sigma_r = 1.5$ mm. The laser is modelled as a Gaussian beam with waist $w_0 = 3$ mm and pulse duration of 100 (filled circles), 200 (diamonds), 10000 (triangles) and 20000 (empty circles) cycles.

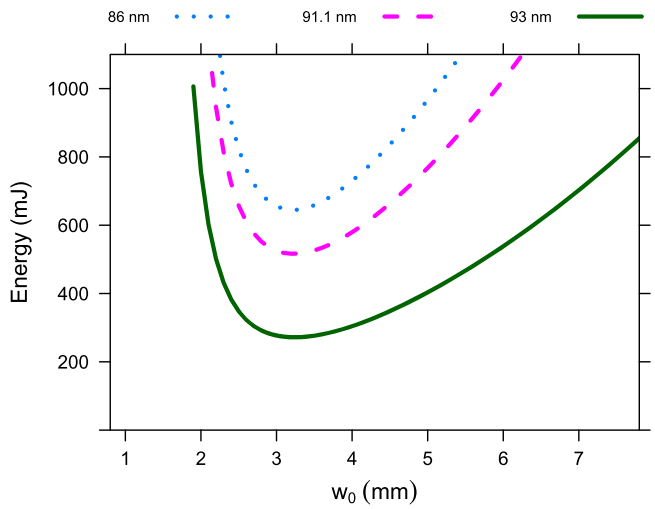

Figure 6. Dependence on laser beam waist w_0 of the energy in the particle frame required to achieve 99% ionization fraction in a $6 \times 6 \times 90$ mm^3 volume of hydrogen atoms characterized by a uniform longitudinal distribution and transverse Gaussian distribution with size $\sigma_r = 1.5$ mm interacting with a 20 000 cycle pulse of wavelength 93 nm (continuous line), 91.1 nm (dashed line) and 86 nm (dotted line).

$L = 100$ m and 99% reflectivity, a single pulse can interact with a particle beam 50 times before its intensity has dropped by $1/e$, corresponding to an interaction time of approximately 33 μs. In general, laser and cavity parameters should be tailored to the specific machine characteristics. Advanced cavity designs have been developed for Compton scattering sources [31–33], which encounter similar challenges. Coupled to the use of multiple laser beams or a new generation of intense high-repetition lasers [34], these technologies make direct ionization a viable candidate for efficient laser stripping.

4. Conclusions

In conclusion, we have investigated the photo-ionization of atomic hydrogen exposed to laser radiation by numerically solving the TDSE, showing that in the 50–90 nm spectral window nearly full ionization can be achieved over large volumes at laser energies as low as 100mJ for picosecond-scale pulse durations. Such irradiations are realizable with technology currently available or at an advanced development stage, making direct ionization a viable method for laser stripping of high-energy H beams.

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