

Article

Ionization Dynamics in Matter with Gold Nanoparticles upon Laser Irradiation of Various Intensities, Numerical Analysis [†]

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Abstract: We perform the numerical study of the response of the media with golden nanoantennas upon irradiation by intense $\sim 10^{17}$ – 10^{18} W/cm² short 0.1 ps laser pulses. We study the influence of resonant nanoantennas on the ionization process and on the ions' energy evolution at various intensities of laser pulses. Numerical modeling is performed with the help of EPOCH software using the “particle-in-cell” numeral method. The response of resonating nanoantennas of dipole and crossed shapes, embedded in dense media, is studied. The dynamics of ionization and the energies of ions acquired during the passage of the laser pulse are studied. The differences in the ionization energies for nanoantennas of dipole and crossed shapes are explored. The ionization dynamics in the matter doped with nanoantennas is examined; crossed-shaped antennas are identified for the best energy absorption in high-intensity fields.

Keywords: nanoantenna; laser pulse; kinetic model; energy; momentum; particle-in-cell simulation



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1. Introduction

Recent advancements in nanotechnology have occurred alongside the rapid development of laser technology and this allows for significant progress in many research fields. The study of the interaction of radiation with matter has multiple applications. A significant emphasis is placed on energy absorption processes in materials subjected to short laser pulses and how doping with resonant nanoparticles can improve this absorption efficiency. Incorporating resonant nanoparticles in a medium has been studied for several years (see, for example, [1–5]). It was shown that resonant nanoparticles in a medium can enhance the energy absorption from radiation and boost the energies of charges after ionization. Nanoantennas are often made of gold or silver; resonating in media, they offer potential applications in energy transfer and controlled heat absorption within materials [3,4], and are capable of reliably withstanding radiation pulses at intensities of 10^{15} W/cm² [5] and higher. It should be noted that the heat transfer from short intense pulses goes beyond the scope of the common Fourier law [6–9]; it is addressed in extended thermodynamic models with heat equations like the Guyer–Krumhansl heat equation and other types. They describe a variety of physical phenomena, including ballistic and wave heat propagation and heat propagation in matter with internal inhomogeneities. They are of great interest from the viewpoint of both mathematics [10–14] and applications. Heat transfer in a media with nanoantennas will be addressed in a dedicated forthcoming publication in the field of

thermodynamics; current research, viewed in the above context, gives a good insight into the energy distribution in an inhomogeneous matter heated by a short laser pulse.

An interesting, relevant-to-laser-matter-interactions field of science, where nanotechnology offers a possible breakthrough, is the Inertial Confinement Fusion (ICF) driven by lasers, which has been supported by novel laser technologies, for example, at the Extreme Light Infrastructure (ELI) Laboratories in the European Union. One of the central challenges in laser-driven ICF is ensuring the effective absorption of laser energy. The use of nanoantenna doping improves laser light absorption, which could significantly enhance the performance of laser-driven ICF. Recently, a novel concept known as NAnoPlasmonic Laser Inertial Fusion Experiments (NAPLIFE) has been introduced, offering a promising nonthermal approach to laser-driven fusion, characterized by a collider configuration. Traditional ICF methodologies, including both indirect- and direct-drive approaches, rely on nanosecond-scale laser pulses. Newer strategies such as fast ignition are moving toward the use of picosecond or even shorter laser pulses. These methods rely on relativistic collisions between target slabs, which are generated using the Laser Wake Field Acceleration (LWFA) mechanism. This process employs femtosecond laser pulses to drive high-velocity, beam-directed collisions, resulting in rapid ignition. Departing from earlier approaches, the NAPLIFE project incorporates innovative concepts, such as nanoplasmonic amplification within a layered, flat target featuring adjustable absorptivity. These novel ideas are being explored with the goal of achieving simultaneous ignition more effectively.

The aim of the current research, which is part of a bigger experimental and theoretical work sketched above, is to find the best and adequate resonant dopes for the maximum energy absorption from a radiation pulse and to examine how the inclusion of nanoantennas can boost the energy and momentum of the charges (electrons, protons, and heavy ions) produced during the ionization process upon the irradiation of matter by external laser fields in a wide range of intensities. The study is based on combining knowledge from recent discoveries in heavy-ion collisions and optics. The resonant properties of nanoantennas can be described by a dynamic model, using the Laser Wake Field Acceleration (LWFA) framework [15]. Relevant laser pulses last for fractions of a picosecond, comparable with those generated by free-electron lasers (FELs) [16–21] in the X-ray band. However, coherent radiation in the visible and infrared bands is much easier to produce in a laboratory or in an experimental plant, where kilometric X-FELs cannot be placed. For example, in the National Ignition Facility-Advanced Radiographic Capability (NIF-ARC) project [22] and other studies [23,24], infrared laser radiation at high intensities $\sim 10^{18}$ W/cm was employed to accelerate protons. The detonations with timelike normal on space-time hyper-surfaces combined with absorption adjustment using nanoantennas allows for the possibility of heating the target in an opposing laser beam setup [4]; a dedicated study of heat transfer in this process will be conducted elsewhere. As a first approach to this thermodynamic problem, we will explore in what follows the role of resonant nanoantennas for the energy absorption from laser radiation. Incorporating resonant nanoparticles in a medium involves new laser technology together with nanotechnology [4,25], and given the right nanoparticle parameters, this could be easily tuned to meet needs in various applied fields and even for practical medical purposes [25].

In what follows, we study the dynamics of electron density and behavior near and within gold nanoantennas of dipole and crossed geometries; the dimensions of nanoantennas are optimized for resonant interaction with laser wavelengths based on the prior research [15,26–28]. Embedded in a UDMA-TEGDMA copolymer medium with a refractive index of $n \approx 1.5$, nanoantennas exhibit optimal resonant lengths for energy absorption at specific wavelengths. Numerical simulations were conducted using EPOCH 4, a particle-in-cell (PIC) computational framework. This software allows the study of localized

surface phenomenon of resonant oscillations of conducting electrons on the surface of metal nanoparticles in plasmonic forms (LSPs). The formation of LSPs enhances the local electromagnetic field and increases the optical absorption in the medium at the designated resonance frequencies [29]. The simulations reveal how nanoantenna shape and orientation influence energy absorption and ionization efficiency. Dipole nanoantennas exhibit strong resonance properties, and quadrupole-shaped nanoantennas also resonate well; their resonance was analyzed in fields of various intensities in [30,31]. In the dielectric function $\epsilon(\lambda)$ the real part of the permittivity counts in the calculation of the effective resonant wavelength; its limit in an infinite frequency is ϵ_∞ [24]. For a golden nanoantenna, the value of $\epsilon_\infty = 11$, the plasma wavelength $\lambda_p = 138$ nm. The speed of light in media is $c_s = 1/\sqrt{\epsilon_s}$ and the refractivity index $n = \sqrt{\epsilon_s}$ employs the effective speed c_s expressed in terms of the dielectric permittivity of media ϵ_s . A resonating length L of a dipole antenna is a half wavelength of the radiation, $L = \lambda/2$; the effective resonating wavelength λ_{eff} of a dipole in a medium was estimated in [32] as

$$\lambda_{eff} = 2\pi R \left(13.7 - 0.12 \left(\frac{\epsilon_\infty}{\epsilon_s} + 141 \right) - \frac{2}{\pi} + \frac{\lambda}{\lambda_p} 0.12 \frac{\sqrt{\epsilon_\infty + 141\epsilon_s}}{\epsilon_s} \right) \quad (1)$$

Gold dipoles are embedded in media with $n = 1.53$, and their optimal antenna length is $L = \lambda_{eff}/2 \approx 85$ nm for a 25 nm diameter rod. The effective resonant wavelength tuning was studied in the preceding publications, where the optimal length of the rod of 85 nm was worked out based on the technical facility 25 nm diameter of the nanorod. The antenna is positioned in the Y-Z plane normally to the incident radiation and is subject to vertically polarized laser radiation propagating along X (see Figure 1); the dipole nanoantenna is a vertical part (along Y axis) of the cross shown in Figure 1.

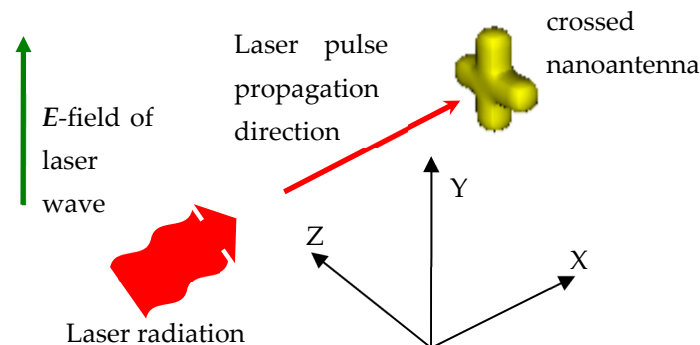


Figure 1. Schematics of the nanoantenna of a crossed shape subject to laser wave irradiation.

A large volume of data amounting to ~15 TB was obtained during simulations. We use the particle-in-cell (PIC) method [33], where the marker particles represent a conglomerate of real particles in dynamics; charge densities and currents are computed within stationary mesh cells. The EPOCH program [34] takes the ion–electron collisions into account [35,36] for the analysis of electron and plasmon dynamics; electron kick out [37] is also accounted for in our simulations. The laser field causes the resonant motion of conducting electrons in gold, the formation of a plasmon on the surface of golden nanoantenna and the consequent kick out of the conducting electrons from gold. We consider linearly polarized laser radiation at the wavelength of 795 nm, propagating towards the dipole or crossed nanoantenna (see Figure 1) in pulses of ~0.1 picosecond durations. The laser field intensities vary between $\sim 10^{15}$ W/cm² and 10^{18} W/cm².

2. Resonating Dynamics with Nanoantennas

In what follows, we consider and compare with each other the ionization dynamics in matter doped with dipole and crossed nanoantennas. The geometric dimensions of the antennas are based on the estimate (1) for a dipole, which defines the characteristic scale of the inclusion but does not determine the resonant sizes for the investigated shapes. Previously [28], we examined the energy absorption by dipoles of various thicknesses and lengths, deviating $\pm 30\%$ from the estimated in (1) length of 85 nm for the 25 nm diameter rod. In cases where the antenna size deviated from the estimate (1), the energies of the protons after ionization by electromagnetic impulse were lower, so we keep the size $85 \text{ nm} \times 25 \text{ nm}$.

In what follows, we analyzed the electric field around nanoantennas. In Figure 2, the electric field around a crossed nanoantenna is shown at about the half-time of the laser pulse of $4 \times 10^{17} \text{ W/cm}^2$, whose duration was 120 fs. The field around the nanoantenna at the half-time of the pulse (see Figure 2a) is stronger than that at the end of the pulse in Figure 3a. This behavior is different from that of the radiation pulse of the low intensity $4 \times 10^{15} \text{ W/cm}^2$, where the field around the antenna continuously increased towards the end of the pulse as explored in [28]. The electron density also decreases towards the end of the pulse (see Figures 2b and 3b). In Figures 2–4, a view is given from the side in direction Z, so the crossed antenna is seen as a vertical rod from the side (refer to Figure 1 for clarity on the presented view).

We computed and analyzed the electric field around a dipole nanoantenna and we found a significant dependence of the field intensity around the dipole on its orientation, respectively, to the vector of the laser electric field E . The electric field around the resonating dipole is stronger when the dipole is directed along vector E of the radiation wave as compared with the case where the dipole is perpendicular vector E . Contrary to the dipole, a crossed nanoantenna is not sensitive to the polarization of the incident laser radiation; rotation in the plane Y-Z perpendicular propagation of the laser pulse on X (see Figure 1) did not change results. This gives the advantage to the crossed vs. the dipole antenna in moderate and strong fields.

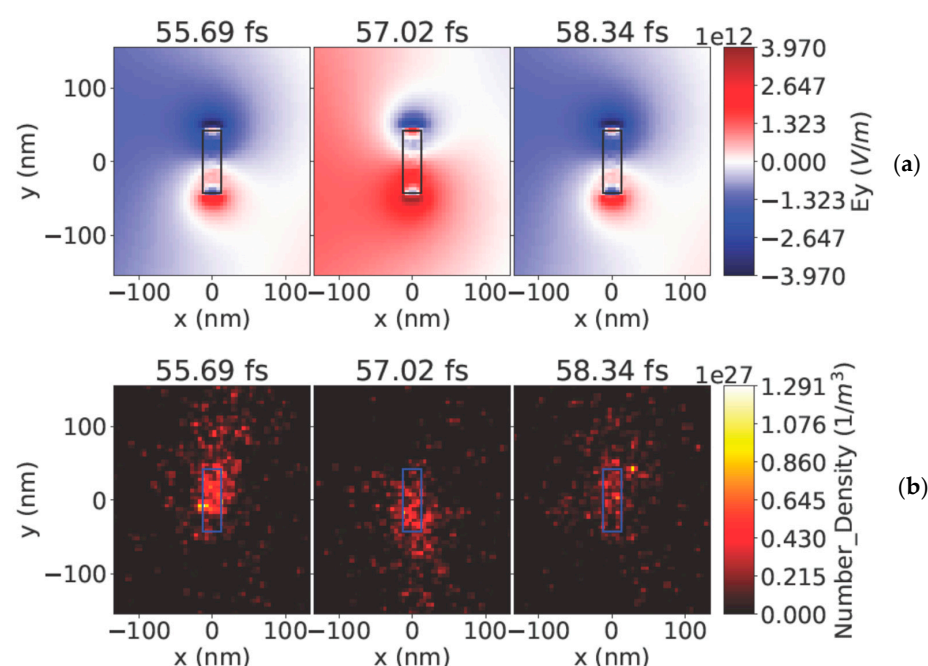


Figure 2. Electric field E around the crossed antenna (a) and the electron number density (b) at the half-time of the laser pulse at $\approx 57 \pm 2 \text{ fs}$ for the moderate laser intensity of $4 \times 10^{17} \text{ W/cm}^2$.

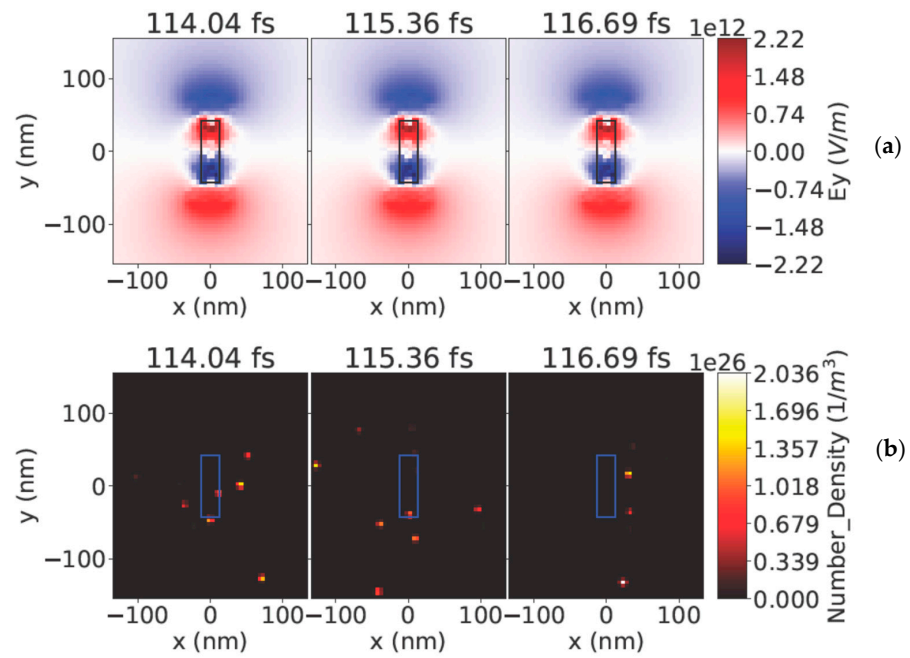


Figure 3. Electric field E around the crossed antenna (a) and the electron density (b) at the end of the laser pulse at 115 fs for the moderate laser intensity of $4 \times 10^{17} \text{ W/cm}^2$.

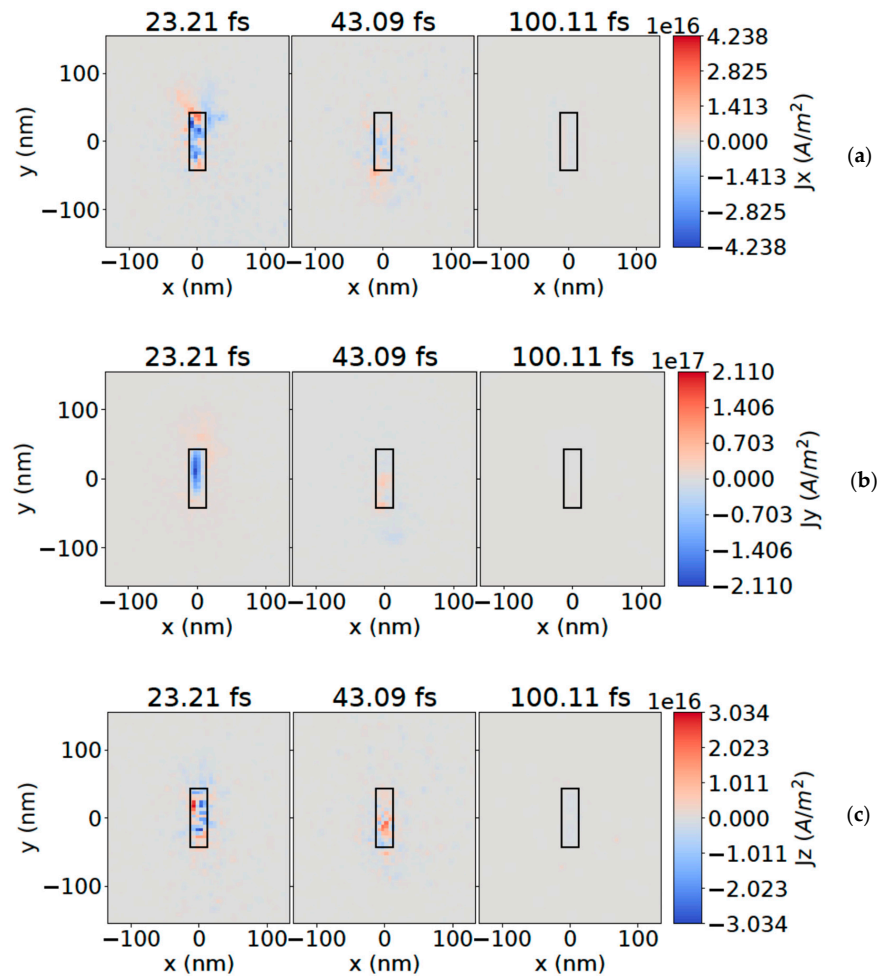


Figure 4. Electric current (a) X-, (b) Y-, and (c) Z-components around the crossed antenna at the beginning of the laser pulse at 23 fs, at 44 fs and at the end of the pulse at 100 fs for the moderate laser intensity of $4 \times 10^{17} \text{ W/cm}^2$.

The laser pulse of the $4 \times 10^{17} \text{ W/cm}^2$ intensity generates a strong electric current; it spreads out from the resonating nanoantenna due to the kick out of conducting electrons from gold and the ionization of hydrogen matter to proton–electron pairs at the beginning of the pulse; examples for the current at 23 fs, 43 fs, and 100 fs are shown in Figure 4a–c, for the X-,Y-, and Z-components respectively. The current then decreases towards the end of the pulse (see Figure 4 for 100 fs) and nearly vanishes because most of the conducting electrons are kicked out of the gold after ≈ 100 fs. For the vertical dipole that is parallel the electric field E of the incident radiation wave, the current behaves similarly to that for a crossed antenna.

3. Ionization Dynamics with Crossed Nanoantennas

In Figure 5, we show the plasmon formation in the vertically polarized laser field. For the radiation field with angled linear polarization, the plasmon moves on the antenna following the E field direction, and not vertically as it does in the case of vertical polarization. We have studied vertical, horizontal and angled polarization vector orientations of the laser field to test the sensitivity of the crossed and dipole antennas to the direction of polarization. We found that the crossed antenna was not sensitive to polarization and this positively distinguishes it against the dipole antenna, whose performance depends on the dipole orientation. In the field of a moderate intensity of $4 \times 10^{17} \text{ W/cm}^2$, the formation of a plasmon on the nanoantenna can be observed at the beginning of the pulse after ~ 10 – 20 fs (see Figure 5a,b; conducting gold electrons are yellow, gold ions are orange). The conducting electrons in gold oscillate following the oscillations of laser field E . The common motion of the conducting electrons yields the formation of the plasmon on the antenna (see Figure 5b,c). At a certain point, the plasmon detaches from antenna; see Figure 5d where the plasmon detaches from the cross at ~ 55 fs. This is different from the behavior in the weak field of $4 \times 10^{15} \text{ W/cm}^2$ (see [28]), where the plasmon detachment did not occur through the whole duration of the pulse. After some time, the conducting gold electrons are dispersed around the antenna as shown in Figure 5e for the moment of time at ~ 80 fs. Note that in a stronger field of $4 \times 10^{18} \text{ W/cm}^2$, the plasmon's life is shorter compared to that in a moderate $4 \times 10^{17} \text{ W/cm}^2$ laser field.

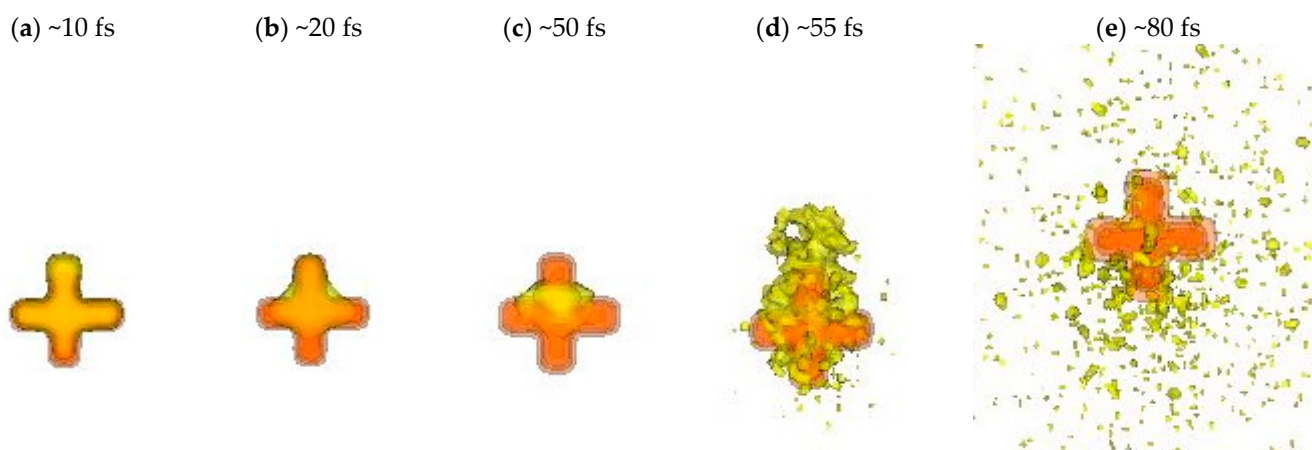


Figure 5. Electron and gold ion number density upon irradiation by the laser pulse of a moderate intensity of $4 \times 10^{17} \text{ W/cm}^2$ at (a) ~ 10 fs, (b) ~ 20 fs, (c) ~ 50 fs, the (d) detachment of the plasmon from the antenna at ~ 55 fs, and (e) electrons dispersed around the nanoantenna at ~ 80 fs.

4. Evolution of Ion Energies in Moderate and Intense Laser Fields

In what follows, we study the evolution of the energies of protons and gold electrons, which appear during ionization in the field of a moderate intensity of $4 \times 10^{17} \text{ W/cm}^2$ and

in a stronger field of $2 \times 10^{18} \text{ W/cm}^2$ intensity. Upon irradiation at a moderate intensity of $4 \times 10^{17} \text{ W/cm}^2$, the dipole antenna in its best orientation along vector E of the laser field yields $\approx 0.11\text{--}0.12 \text{ MeV}$ energies for protons (magenta solid line in Figure 6). If the dipole is in the X-Z plane (see Figure 1), it has an unfavorable orientation across vector E , and then it does not resonate well with the field and this yields a lower energy for protons around the antenna of $\approx 0.06 \text{ MeV}$ (black and blue solid lines in Figure 6). The crossed nanoantenna in the field of a moderate intensity of $4 \times 10^{17} \text{ W/cm}^2$ in the Y-Z plane, as shown in Figure 1, yields the highest amount of proton energy up to 0.15 MeV , as shown by the green dashed line in Figure 6. Note that as the gold electrons' energies reach $\approx 0.15\text{--}0.17 \text{ MeV}$, the marginally lower value of $\approx 0.12 \text{ MeV}$ is reached for the gold electrons' energies of the dipole in the unfavorable horizontal position in the X-Z plane (compare black and blue lines vs. the magenta line for dipoles and vs. the green dashed line for the cross in Figure 6b). Note in Figure 6b that the electron energy decreases after 100 fs because the laser pulse ends and the motion of the charges is damped by the collisions; moreover, the marker particles leave the simulation box.

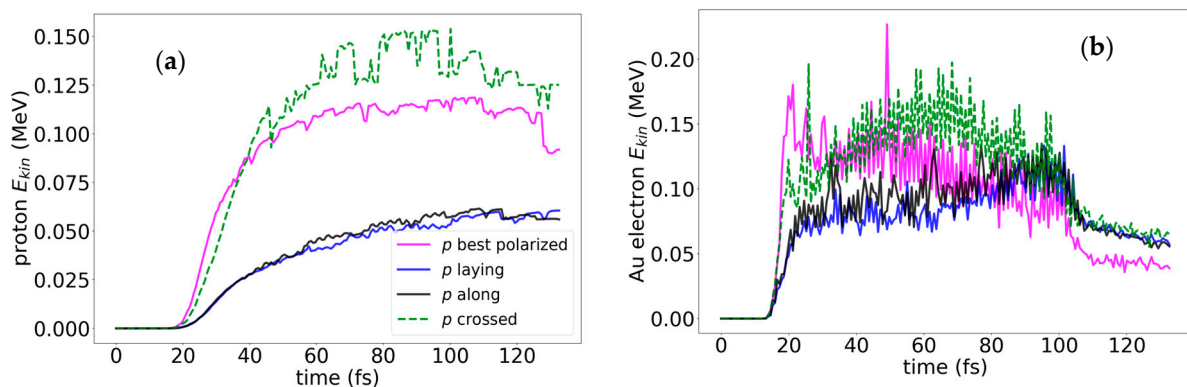


Figure 6. Evolution of energies of (a) protons and (b) gold electrons around the dipole and crossed nanoantennas (see legends) subject to a vertically polarized laser pulse of a moderate intensity of $4 \times 10^{17} \text{ W/cm}^2$.

Consider a stronger laser field of $2 \times 10^{18} \text{ W/cm}^2$. For the dipole antenna, even in its best vertical position $\parallel E$, the increase in the laser field intensity from $4 \times 10^{17} \text{ W/cm}^2$ (magenta solid line in Figure 7) to $2 \times 10^{18} \text{ W/cm}^2$ (green dashed line in Figure 7) gives little additive energy to protons that is $\approx 0.12 \text{ MeV}$ (see Figure 7a). The ionization of the golden dipole in the field of $2 \times 10^{18} \text{ W/cm}^2$ essentially ends at $\sim 60 \text{ fs}$ (green dashed line in Figure 7b). The increase in the laser field intensity expectedly shortens the time of ionization for the antenna; but for the dipole, the increase from $4 \times 10^{17} \text{ W/cm}^2$ to $2 \times 10^{18} \text{ W/cm}^2$ is not worth the minimal gain in the proton energy from $\approx 0.11 \text{ MeV}$ to $\approx 0.12 \text{ MeV}$. For the crossed nanoantenna, contrary to the dipole, this increase in the field intensity yields a noticeable increase in the proton energy from $\approx 0.15 \text{ MeV}$ to $\approx 0.2 \text{ MeV}$ (see Figure 8a) and this is almost twice as high as the energy of protons around the dipole in the same field, $\approx 0.12 \text{ MeV}$ (see Figure 7a). In Figure 8b, note that the gold electron energies around a crossed antenna reach $\approx 0.45\text{--}0.5 \text{ MeV}$ in the field of $2 \times 10^{18} \text{ W/cm}^2$ vs. $\approx 0.15 \text{ MeV}$ in the field of $4 \times 10^{17} \text{ W/cm}^2$ (Figure 7b). The gold electrons around the cross in the field of $2 \times 10^{18} \text{ W/cm}^2$ stop gaining energy after 30 fs and the process for electrons ends at 40 fs as shown in Figure 8b. Thus, the time interval during which ionization occurs is shorter for the cross than for the dipole in the strong field, but the energies of protons are higher for the cross.

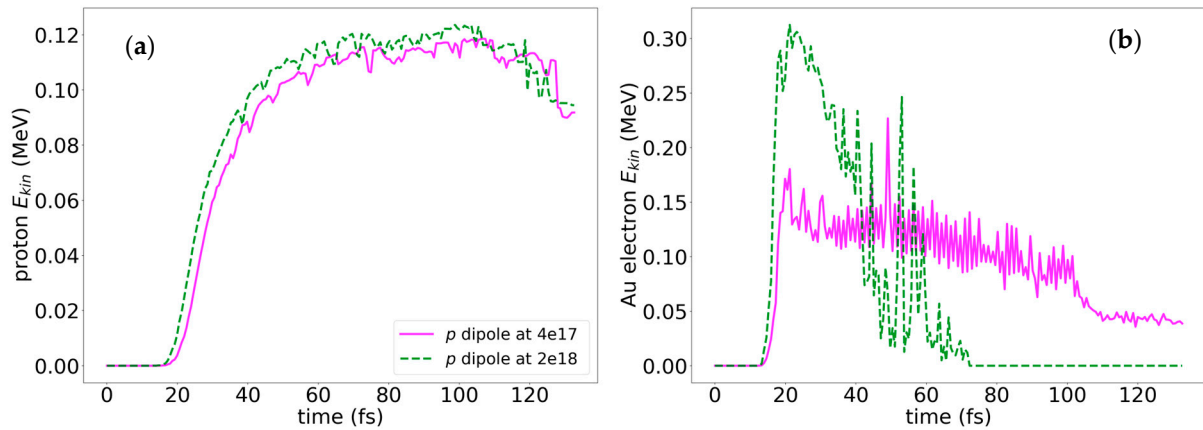


Figure 7. Evolution of energies of (a) protons and (b) gold electrons around the dipole nanoantenna that is subject to vertically polarized laser pulses of moderate $4 \times 10^{17} \text{ W/cm}^2$ (magenta solid line) and high $2 \times 10^{18} \text{ W/cm}^2$ (green dashed line) intensities.

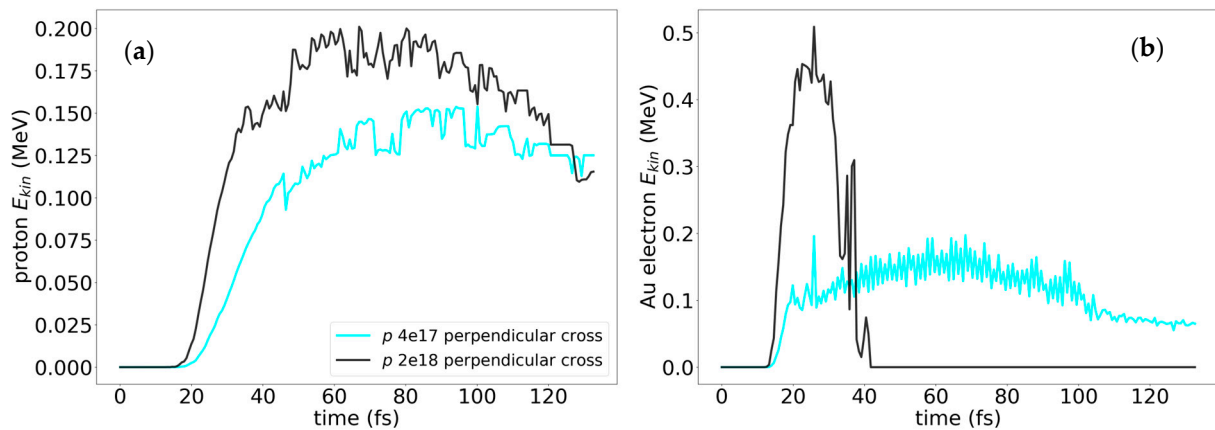


Figure 8. Evolution of energies of (a) protons and (b) gold electrons around the crossed nanoantenna that is subject to vertically polarized laser pulses of moderate $4 \times 10^{17} \text{ W/cm}^2$ (light blue solid line) and high $2 \times 10^{18} \text{ W/cm}^2$ (black solid line) intensities.

The higher energy provided by the crossed antenna in the strong field compared to that of the dipole can be explained by the higher number of conducting electrons in the crossed antenna compared to the dipole. Thus, in a strong field, the knock out of the conducting electrons from a cross is less critical and the cross maintains resonant properties whereas the dipole runs short on electrons and loses resonant properties.

Eventually, we demonstrate that the cross antenna in the plane normal to the direction of the laser wave propagation (see Figure 1) is not sensitive to the polarization of the field E . In Figure 9, we present the evolution of the energies of protons (a) and gold electrons (b) around the crossed antenna in the diagonally polarized laser fields of $4 \times 10^{17} \text{ W/cm}^2$ and $2 \times 10^{18} \text{ W/cm}^2$ intensities. Upon comparison of the respective plots in Figure 8 vs. Figure 9, we see that the maximum energies of protons in the diagonally or vertically polarized radiation fields are the same, $\approx 0.15 \text{ MeV}$ for $4 \times 10^{17} \text{ W/cm}^2$ and $\approx 0.2 \text{ MeV}$ for $2 \times 10^{18} \text{ W/cm}^2$. The energies of electrons kicked out of the gold do not depend on the polarization of the incident radiation; the time domain for the gold electrons and their energies are also the same for vertically and diagonally polarized radiation: $\approx 105 \text{ fs}$ (compare Figure 8b vs. Figure 9b).

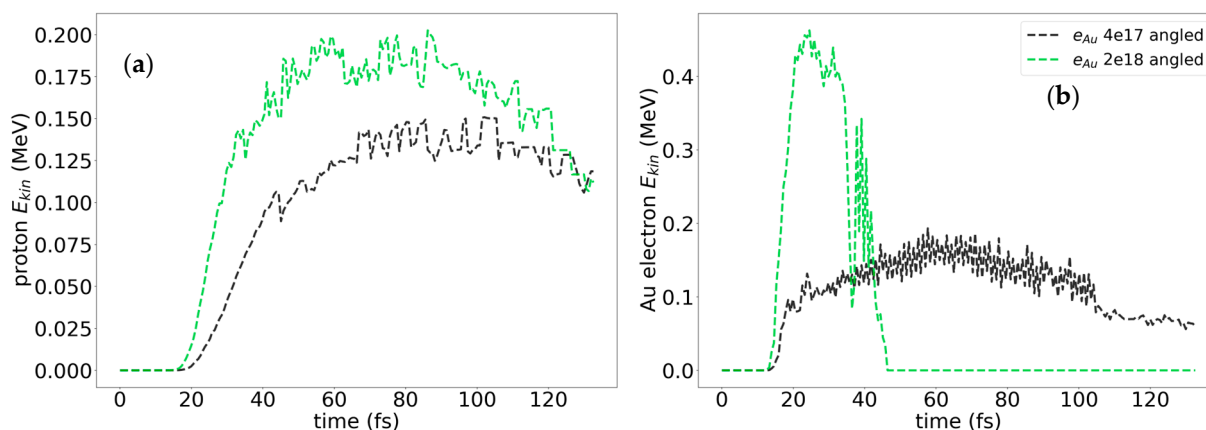


Figure 9. Evolution of energies of (a) protons and (b) gold electrons around the crossed nanoantenna that is subject to diagonally polarized laser pulses of moderate $4 \times 10^{17} \text{ W/cm}^2$ (green dashed line) and high $2 \times 10^{18} \text{ W/cm}^2$ (black dashed line) intensities.

5. Conclusions

In the present study, we have examined the dynamics of the interaction of laser pulses of moderate $4 \times 10^{17} \text{ W/cm}^2$ and high $2 \times 10^{18} \text{ W/cm}^2$ intensities with matter doped with a resonating golden nanoantenna. We studied the performance of dipole and crossed-shaped antennas for the radiation wavelength of $\sim 800 \text{ nm}$ with linear polarization and a duration of $\sim 120 \text{ fs}$. The hydrogen in the model represented a dense dielectric matter. The computations were made with EPOCH 4 software. The analysis of the obtained results was carried out with the help of the custom compiled and ad hoc adopted programs based on the software LLNL VisIt 3.2, and Matplotlib (version 3.10), in Python. The behavior of various ion species and the dynamics of ionization were modeled and demonstrated graphically. The evolution of ion energy acquisition was studied comprehensively and traced separately for all ion species.

The analysis of the results evidenced that crossed nanoantennas were not sensitive to the orientation of the polarization vector of the laser field and thus crossed antennas were advantageous compared to common dipole antennas. For a laser pulse of a $4 \times 10^{17} \text{ W/cm}^2$ intensity, the ionization of the gold antenna develops as the laser pulse progresses to $\approx 100 \text{ fs}$. Gold loses a substantial number of conduction electrons, which are ejected during the electromagnetic pulse propagation. By the midpoint of the laser pulse, the majority of the conduction electrons are “knocked out” of the gold nanoantenna, leaving behind ionized gold. At this stage, the resonance between the antenna and the radiation diminishes noticeably concerning the contribution of the conduction electron gas. By the end of the pulse, after 100 fs , the result is an ionized gold nanoantenna, with the remaining cloud of electrons and protons dispersing outward from the nanoantenna. Analysis of the current data indicates that as the pulse intensity exceeds $4 \times 10^{17} \text{ W/cm}^2$, energy saturation occurs for the ions around the antenna during the ionization process. This is caused by the ejection of conduction electrons from the nanoantenna and the subsequent loss of its resonant properties. Our analysis suggests that saturation occurs earlier in dipole-shaped nanoantennas compared to cross-shaped ones. The latter not only facilitate greater energy acquisition by the ions during the laser pulse but also provide a higher energy ceiling during the ionization process.

Comparison of the ion energies in fields of intensities of $4 \times 10^{17} \text{ W/cm}^2$ and $2 \times 10^{18} \text{ W/cm}^2$ showed that in the field of a $4 \times 10^{17} \text{ W/cm}^2$ intensity, protons gain higher energy, $\sim 0.15 \text{ MeV}$, for the crossed antenna than the $\approx 0.11\text{--}0.12 \text{ MeV}$ for a dipole antenna. Study of the ionization in the intense field of $2 \times 10^{18} \text{ W/cm}^2$ reveals that the increase in intensity of five times yields a just marginal increase in the proton energies up

to ~ 0.2 MeV for the crossed antenna for the pulse of the same duration. Expectedly, the stronger the radiation field is, the earlier the energy is gained. For the dipole antenna, the increase in the laser field from 4×10^{17} W/cm² to 2×10^{18} W/cm² yields an insignificant increase in the proton energy and is unfeasible. The higher energy of the protons of the crossed antenna than for those of the dipole in the strong field perhaps is due to the larger number of conducting electrons in the cross. Thus, the loss of conducting electrons from gold, occurring in the strong field, is less critical for the cross, which maintains resonant properties better than the dipole.

Moreover, crossed nanoantennas are not sensible for the polarization of the incident radiation.

Therefore, resonating crossed nanoantennas are recommended for use as dopants to increase energy absorption in matter that is subject to intense radiation fields of $\sim 10^{17}$ W/cm² or higher.

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