Crater Formation and Deuterium Production in Laser Irradiation of Polymers with Implanted Nano-antennas

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Recent validation experiments on laser irradiation of polymer foils with and without implanted golden nano-particles are discussed. First we analyze characteristics of craters, formed in the target after its interaction with laser beam. Preliminary experimental results show significant production of deuterons when both the energy of laser pulse and concentration of nano-particles are high enough. We consider the deuteron production via the nuclear transmutation reactions

\[ p + C \rightarrow d + X \]

where protons are accelerated by Coulomb field, generated in the target plasma. We argue that maximal proton energy can be above threshold values for these reactions and the deuteron yield may noticeably increase due to presence of nano-particles.

I. INTRODUCTION

Relativistic heavy-ion collisions have shown that the hadronization (or burning) of created matter is a very fast, nearly simultaneous process in proper-time. This is in contrast to the classical Rayleigh-Hugoniot-Taub detonation theory. The possibility of relativistic detonation on a time-like hyper-surface was theoretically discovered in 1987 [1], and was later applied to simulate pellet fusion in Ref. [2]. It was found that it is not possible to obtain a better result than was reached in NIF experiments, where Rayleigh-Taylor instabilities prevented the full burning of the target fuel.

To remedy this problem, we have suggested [3] to implant golden nano-particles (nano-antennas) into the target. In this way one can achieve “time-like” ignition, which occurs nearly simultaneously in the whole target volume. Below we describe the validation experiments within the NAnoPlasmonic Laser Induced Fusion Energy (NAPLIFE) project [4]. They are planned in two steps: (i) verification of the amplified absorption of the laser energy in the target with nano-particles, and (ii) testing a simultaneous detonation in the whole target volume, induced by two short (\( \tau \lesssim 1 \) fs) laser pulses radiating opposite sides of a flat target.

For the validation tests we have chosen the polymer UDMA (see below), which is hard enough to make thin (3 µm) layers of it and construct an altogether 21 µm multi-layer target. By adjusting the nano-particle (NP) density profile one can achieve [7] simultaneous light absorption in the whole volume.

Recently we started validation experiments where a thicker (~160 µm) target was irradiated from one side leading to the formation of a crater [8, 9].

II. CRATER FORMATION

a) Composition of the target

The Urethane Di-MethAcrylate (UDMA) polymer contains 470 nucleons and 254 electrons in 71 atoms. Its molecular formula is \( \text{C}_{23}\text{H}_{38}\text{N}_{2}\text{O}_{8} \). The calculated mass of this molecule equals 470.2628 amu [10]. The reference atom, \( ^{12}\text{C} \) has in vacuum the binding energy of 7.68 MeV/nucleon. Compared to this value the average binding energy per nucleon in the UDMA is equal 0.2628/470 amu \( \approx 0.5207 \text{ MeV} \).

1 The two-sided irradiation was applied earlier in Refs. [5, 6].
After the irradiation by an energetic laser pulse the UDMA molecule may fragment and break up to pieces. In this process numerous chemical configurations, e.g., 2(NO₂), 2(CO₂), CH₄, C₁₂H₃₄, 4C, or even single atoms may appear in the final state. At large enough laser energies, recombination of nucleons becomes also possible, and new atomic species can be formed. For example, a neutron from ¹³C may be transferred to a Hydrogen, producing a Deuterium. This is an endothermic reaction, which requires more energy than needed for the molecular fragmentation.

As far as we know, the Deuterium formation in laser irradiated targets was observed in these validation experiments for the first time. It is interesting that its amount strongly increases with raising density of implanted golden nano-antennas.

In our validation experiments, we have irradiated three types of thick targets: pure UDMA without nano-antennas (Au0), as well as UDMA with golden nano-rod antennas of the size 25x85 nm and volume \( V_{Au} \approx 41724 \text{n}^3 \). We have used targets with two different mass density ratios of NPs: \( m_{Au}/m_U = 0.126\% \) (Au1) and 0.182\% (Au2). It was observed that laser shots brake out craters in the UDMA target. Parameters of these craters were analyzed by microscope. It was found that they depend significantly on the concentration of NPs.

b) Craters with nano-plasmonics

Geometrical parameters of craters were measured in Refs. [11–13]. The laser beam with a Gaussian intensity profile was focused to a spot with diameter of 24.5±3.2 \text{µm} up to 85\% norm (FWHM \( \approx 14.4 \text{µm} \), the shape was not circularly symmetric) [14].

The following values of crater diameters have been obtained: 110–130 \text{µm} at the laser pulse energy \( E_L = 5 \text{mJ} \), 120–130 \text{µm} at \( E_L = 10 \text{mJ} \), 130–150 \text{µm} at \( E_L = 15 \text{mJ} \), 155–165 \text{µm} at \( E_L = 20 \text{mJ} \), and 190–200 \text{µm} at \( E_L = 25 \text{mJ} \). The average crater depths, calculated from observed crater volumes and diameters (assuming its cone shape), are as follows: 5.0, 5.2, 5.7 \text{µm} at \( E_L = 5 \text{mJ} \), 7.7, 9.6, 15.6 \text{µm} at \( E_L = 15 \text{mJ} \), and 5.6, 6.7, 14.3 \text{µm} at \( E_L = 25 \text{mJ} \), for the targets Au0, Au1, Au2, respectively. As the effective focus of the laser beam was fixed to about 12.6 \text{µm} , the observed increase of the crater diameter could be attributed to the transverse expansion of hot plasma created by the laser beam. One can see that the crater diameters do not depend much on the concentration of nano-antennas. On the other hand, the crater volumes and depths clearly depend on it, see Figs. 1, 2.

One can see in Fig. 1 that the crater volume \( V_{cr} \) increases with the laser pulse energy and with the density of nano-antennas. As compared to the pure UDMA target (Au0), the crater volume increases with \( E_L \) more rapidly for targets with nano-antennas (Au1 and Au2). Changing the nano-rod antenna density from zero (Au0) to 0.182 \% (Au2) increases the crater volume from 50000 to 135000 \text{µm}³. Thus, the nano-antennas increase the laser light absorption. The largest crater volume \( V_{cr} = 135000 \text{µm}³ \) was observed in the experiment Au2 at \( E_L = 25 \text{mJ} \).

![Crater volume (1000 µm³)](image)

**FIG. 1.** (color online) The crater volume \( V_{cr} \) in the thick UDMA target (in units of \( 1000 \text{µm}^3 \)) as the function of laser pulse energy \( E_L \). Several types of targets are considered: Au1 and Au2 with implanted nano-rod antennas, and Au0 without implantation. The mass concentrations of implanted particles in UDMA are 0.126\% and 0.182\% for targets Au1 and Au2, respectively. For each target type five pulse energies are considered. At the highest pulse energy the crater volume increases rapidly with increasing implantation density. For more details, see Refs. [8, 11].

It is interesting to note that the crater volume increases faster than the deposited laser energy. This indicates that the energy of excavating the crater is appreciably larger than the energy directly coming from the laser. An additional energy may come from exothermic chemical reactions during the fragmentation of UDMA, which are neglected in the present analysis.

The average distance between nano-antennas in the unperturbed target can be estimated as

\[
\langle L_{AuX} \rangle = [V_{Au}(\rho_{Au}/\rho_U)(m_U/m_{AuX})]^{1/3},
\]

where \( V_{Au} \) and \( \rho_{Au} \approx 19.32 \text{g/cm}^3 \) are, respectively, the volume and mass density of a single NP, and \( \rho_U \approx 1.14 \text{g/cm}^3 \) is the mass density of a pure UDMA material. Using Eq. (1), one gets the values \( \langle L_{Au1} \rangle = 0.82 \text{µm} \) and \( \langle L_{Au2} \rangle = 0.73 \text{µm} \). This relatively sparse implantation is well visible on the image of the irradiated Au1 and Au2 targets [12]. In the crater image one can clearly see irregular "cracks" associated with nano-rod positions [13]. We think that these cracks are formed after 'explosion' of nano-rods located near the bottom of the crater.

c) Energy densities in target

One can roughly estimate the average energy density deposited in the target as 

\[
\varepsilon_{cr} = E_L/V_{cr}.
\]
FIG. 2. (color online) Average crater depth assuming a circular cone shape as a function of the energy of the laser irradiation \( E_L \) and the type of target material. The depth is minimal (3.7–7.7 \( \mu m \)) without nano-rod antennas (Au0), while for Au2 target it increases from 5.7 to 15.6 \( \mu m \).

the results shown in Fig. 3. One can see that \( \varepsilon_{cr} \sim 1 \text{ TeV/\( \mu m^3 \)} \) for the top laser energy and NP density. The energy density from the laser beam at higher \( E_L \) and implantation density decreases, because the crater volume increases more rapidly\(^2\). This effect indicates that the nano-rods cause an additional absorption of laser energy, this is why we call them antennas.

In case of \( E_L = 25 \text{ mJ} \) and the Au2 target one can estimate the energy absorbed by a single NP as \( \varepsilon_{cr} V_{Au} \sim 50 \text{ MeV} \), where \( V_{Au} \) is the volume of a single nano-rod. In this case the initial number of nano-antennas in the crater volume is about \( V_{cr}/(L_{Au})^3 \approx 347000 \).

Note, however, that these energy estimates are very crude. In fact, interaction of a short laser pulse with a dense target is a very complicated, space-time dependent process. Presumably, at an early stage the laser beam excites a target in a focal cylindrical channel, which is much more narrow compared to a crater region formed at late times. A part of laser energy goes to ionization of target’s atoms, leading to formation of electron-ion plasma. Under influence of electric fields and microscopic collisions the target particles accelerate and some of them may escape from the crater region. The plasmon excitations in nano-antennas may create ‘hot spots’ in surrounding matter with enhanced energy density [15].

\(^2\) It is clear that the energy of the laser beam should not be equal to the energy absorbed by the target matter, since a part of the beam is reflected.

III. LASER-INDUCED DEUTERIUM PRODUCTION

a) Experimental results

As a result of laser irradiation, many electrons as well as molecular and atomic ions are formed in the target. To our surprise, in the experiments with implanted NPs we have observed [8, 16] a significant fraction of Deuterium atoms, which are emitted on the level of up to 10\% of the observed Hydrogen numbers, \( \frac{D}{2D+H} \). Here D and H are the observed yields of Deuterium and Hydrogen atoms, both determined from the LIBS spectra [9], measured at 0.5 microseconds delay time after the laser pulse [16]. These atoms were detected by presence of Balmer-\( \alpha \) lines in the radiation of the H and D, at \( \lambda = 656.27 \text{ nm} \) and 656.11 nm, \( (E_{B\alpha} = 0.944 \text{ 623 eV and 0.944 \text{ 843 eV})} \), respectively. The absolute amounts of observed H and D were not measured. The relative amount of Deuterium atoms increases linearly with the laser beam energy, reaching 7\% of Hydrogen at \( E_L = 25 \text{ mJ} \) with a target including nano-rod antennas with mass density of 0.126\% (Au1).

We have checked that originally the polymer foils used in our experiments did not contain Deuterium. Below we analyze possible mechanisms of D production.

b) Nuclear transmutation reactions

Let us first assume that D atoms appear due to electron capture (inverse \( \beta \) decay)

\[ p + e \rightarrow n + \nu, \]  \hspace{1cm} (2)

followed by the \( n+p \rightarrow d \) fusion [17]. The reaction (2) is endothermic, and may occur if the kinetic energy of the
electron exceeds 783 keV in the lab frame. The fusion of \( n \) and \( p \) is exothermic process,
\[
n + p \rightarrow d + 2.225 \text{ MeV}.
\]
(3)

Note, that this reaction requires an additional (spectator) particle. However, the reaction (2) is a weak process with a very small cross section. Therefore, this mechanism cannot explain the observed yield of Deuterium \([18]\).

Below we focus on one important aspect of the UDMA fragmentation process, which may lead to nuclear transmutation reactions. Especially interesting are the reactions associated with carbon atoms in the UDMA molecule. Note that, a natural carbon is the mixture of two stable isotopes: 98.4\% of \(^{12}\text{C} \) and 1.66\% of \(^{13}\text{C} \). Fast protons accelerated by laser irradiation may lead to production of deuterons via the stripping reactions
\[
p + ^{13}\text{C} \rightarrow d + ^{12}\text{C}, \quad \text{(4)}
\]
and
\[
p + ^{12}\text{C} \rightarrow d + ^{11}\text{C}. \quad \text{(5)}
\]

These reactions are endothermic and may occur if the proton energy exceeds some minimal (threshold) value. The last neutron in the ground state of the isotope \(^{13}\text{C} \) is relatively weakly bound, with the binding energy of about 4.95 MeV. On the other hand, the neutron separation energy of \(^{12}\text{C} \) equals approximately 15.65 MeV. Due to this reason, the threshold proton energy required for the reaction (5) is larger than for the reaction (4) (see below).

Let us first consider reaction (4). It has a negative \( Q \) value\(^3\)
\[
Q = m(^{13}\text{C}) + m_p - m(^{12}\text{C}) - m_d \\
\simeq -2.7216 \text{ MeV}.
\]
(6)

Therefore, it can only take place if protons created during the laser-target interaction have sufficiently high kinetic energies (see below).

The inverse 'pick-up' reaction
\[
d + ^{12}\text{C} \rightarrow p + ^{13}\text{C}. \quad \text{(7)}
\]
is exothermic. It was extensively studied experimentally in the past (see, e.g., Refs. \([20–22]\)).

The ratio of the cross sections for the pick-up and stripping reactions can be calculated by using the detailed balance relation \([23]\):
\[
\frac{\sigma_s}{\sigma_p} = \frac{3}{4} \left( \frac{p_{cm}^{cm}}{p_{cm}^{cm}} \right)^2,
\]
(8)

where \(\sigma_s\) and \(\sigma_p\) are, respectively, the cross sections of the reactions (4) and (7), \(p_{cm}^{cm} (p_{cm}^{cm})\) is the c.m. momentum of the proton (deuteron). All these quantities should be taken at the same c.m. total energy \(\sqrt{s}\). The numerical coefficient in the right hand side of Eq. (8) is the ratio of spin degeneracy weights of particles in the final and initial states of the reaction (4).

Let us further apply the relations of relativistic kinematics
\[
s = 2m(^{13}\text{C})E_{p}^{lab} + [m_p + m(^{13}\text{C})]^2  \\
= 2m(^{12}\text{C})E_{d}^{lab} + [m_d + m(^{12}\text{C})]^2, \quad \text{(9)}
\]
where \(E_{i}^{lab}\) is the kinetic energy of particles \(i = p, d\) in the lab frame. The threshold proton energy for the reaction (4) is obtained from Eq. (9) by substituting \(E_{p}^{lab} = 0\). Then we obtain that \(\sigma_s \neq 0\) at \(E_{p}^{lab}\) above 2.934 MeV, which is slightly larger than \(|Q|\)-value determined in Eq. (6). Analogous calculation for the reaction (5) shows that the latter has the threshold proton energy \(14.568\) MeV.

To calculate the ratio \(\sigma_s/\sigma_p\) one can use the kinematic relation
\[
\frac{p_{d}^{cm}}{p_{p}^{cm}} = \frac{p_{d}^{lab}}{p_{p}^{lab}} \cdot \frac{m(^{12}\text{C})}{m(^{13}\text{C})} \simeq \sqrt{\frac{m_dE_{d}^{lab}}{m_pE_{p}^{lab}}} \frac{m(^{12}\text{C})}{m(^{13}\text{C})}. \quad \text{(10)}
\]

Here the last equality is written in the non-relativistic approximation. As an example, let us consider the deuteron production with energy \(E_{p}^{lab} = 1\) MeV in the stripping reaction (4). According to Ref. \([20]\) the inverse reaction (7) has the cross section \(\sigma_p \simeq 6\) mb at this bombarding energy. From Eq. (9) one can calculate the corresponding energy of protons in the reaction (4), which equals \(E_{p}^{lab} = 3.857\) MeV. Using further Eq. (10), one obtains the value \(\sigma_s \simeq 0.330\sigma_p \simeq 2\) mb.

Note, that many C-containing groups are present in the UDMA molecules. E.g. the Methyl group \(\text{CH}_3\) is frequent at the sides and ends of the UDMA molecule. The Methyl group is bounded to the rest of the molecule by a single covalent bond (\(-\text{CH}_3\)). In these groups the transition of a neutron from \(C\) to the \(H\) is especially easy, occurring via the reaction
\[
^{13}\text{CH}_3 \rightarrow (^{12}\text{C} + n)(\text{H}_2 + p) \rightarrow 
^{12}\text{CH}_2 + (p+n) \rightarrow ^{12}\text{CH}_2 + D. \quad \text{(11)}
\]

In vacuum this reaction is also endothermic requiring 2.85 MeV additional energy. (If doubled it is \(2 \cdot (\text{CH}_3 + H) \rightarrow C_2\text{H}_4 + 2D\), where \(C_2\text{H}_4\) is Ethylene). Similar transmutation is possible with the Methylene group \(\text{CH}_2\). This group is connected to the remainder of the molecule by two single bonds \(-\text{CH}_2\) (Methylene Bridge) or \(\text{CH}_2 = \). The same reaction with the Methylene group is producing \(D\) and \(C_2\text{H}_2\) (Acetylene). Observing Ethylene or Acetylene as byproducts after the Laser pulse irradiation, would confirm this mechanism of the intra-molecular nucleon exchange.

\(^3\) Here we use numerical values of atomic masses from Ref. \([19]\).
c) Laser-induced proton acceleration

It is well known that energetic laser beams may strip electrons, which then pull protons from the target and accelerate them to multi-MeV energy [24–26]. In this section we argue that parameters of the NAPLIFE laser pulse may be sufficient to drive the \(^{13}\)C\((p, d)^{12}\)C reaction in the UDMA target and, therefore, to contribute to the observed formation of Deuterium. Below we use simple electrostatic arguments to estimate the maximal kinetic energy of accelerated protons.

The important laser parameters are related by the formula for the laser intensity

\[
I_L = 4E_L/(\pi d_L^2\tau_L),
\]

where \(E_L\) is the total beam energy, \(\tau_L\) is the duration of the laser pulse, and \(d_L\) is the diameter of the focal spot.

For our estimates we use the following values (see Refs. [9, 11])\(^4\):

\[I_L = 5\times 10^{17} \text{ W/cm}^2, \quad E_L = 25 \text{ mJ}, \quad d_L = 12.6 \mu\text{m}, \quad \lambda_L = 0.795 \mu\text{m}, \quad \text{and} \quad \tau_L = 40 \text{ fs}.
\]

With these laser parameters one can expect that only a small fraction of laser energy, \(\eta\), goes to stripping of electrons from the target’s atoms. The reason is that laser photons have relatively low energies, only of about 1.56 eV. Therefore, a single hydrogen atom can be ionized if its electrons absorb at least 9 photons.

We assume that free electrons are accelerated by the laser field via the ponderomotive force proportional to \(j \times B\). Although this process is rapid and non-equilibrium, one can introduce an effective specific energy of an accelerated electron, \(\Theta\). It is estimated by using the scaling relation suggested in [27], assuming thermalization of absorbed laser energy:

\[
\Theta \simeq 0.42 m_e c^2 \left(\frac{I_L \lambda_L^2}{I_0 \lambda_0^2}\right)^{1/3},
\]

where \(m_e\) is the electron mass, \(c^2 = 1 \mu\text{m}\).

\[
\eta \approx 0.025 \left(\frac{I_L}{I_0}\right)^{0.74}.
\]

We further assume that ionization electrons occupy initially the ‘irradiation region’ (IR), which has approximately a cylindrical shape with the axis along the beam direction, as schematically shown in Fig. 4. Its diameter is approximately equal to the laser focal spot and its depth does not exceed the crater depth. The initial IR is smaller than the finally measured crater dimensions, since part of the absorbed laser energy is spent for heating and evaporating the surrounding matter.

Neglecting the 3D expansion effects, one can estimate the length of IR as

\[
L_{ir} \simeq c\tau_L = 12 \mu\text{m}.
\]

The radius of this cylinder is estimated as

\[
R_{ir} \simeq \sqrt{\frac{E_L}{\pi I_L \tau_L}} \simeq 6.3 \mu\text{m},
\]

which agrees with the observed diameter of the laser focus, \(d_L = 12.6 \mu\text{m}\) [11]. The volume of the IR is

\[
V_{ir} = \pi R_{ir}^2 L_{ir} \simeq 1496 \mu\text{m}^3.
\]

In this volume the original number of H atoms in UDMA is

\[
N^H_{ir} = V_{ir} \cdot n_U \cdot 38 = 1.36 \cdot 10^{14},
\]

where \(n_U = 1.5 \cdot 10^{21} \text{ cm}^{-3}\) is the UDMA number density in the target.

By using the energy conservation, one can find the total number of stripped (ionization) electrons in the IR, \(N^e_{ir}\), as

\[
N^e_{ir} = \eta E_L/\Theta.
\]

We assume that free electrons leave quickly the IR before protons and heavier ions start to accelerate. Then using the electro-neutrality, one can estimate the net positive charge of this region as \(\epsilon N^e_{ir}\) where \(\epsilon\) is the proton charge. From Eqs. (18), (19), (13), one obtains the total positive charge number of IR (in units of \(\epsilon\)) as

\[
N_0 \simeq N^e_{ir} \simeq 1.60 \cdot 10^{10}.
\]

This leads to the following estimate for the Coulomb potential acting on protons

\[
U \sim \frac{\epsilon N_0}{R_{ir}}.
\]

Under influence of the Coulomb field, protons (located initially inside the IR) acquire an additional energy of the

\[
\epsilon U \sim 100 \text{ keV}.
\]
order of $eU$. This leads to the maximal kinetic energy of protons

$$E_p \sim eU \approx 3.65 \text{ MeV.} \quad (22)$$

By substituting the above energy into Eq. (9) one obtains that the maximal deuteron energy in the reaction (4) is about 1 MeV.

At later stages of the target evolution one can expect smaller values of acceleration due to expansion of the initially produced plasma. It will be demonstrated in the next section that the local charge density will significantly increase in the vicinity of NPs, if they are imbedded in the target material. Then one can get larger proton energies as compared to that estimated in Eq. (22). Moreover, proton energies may even exceed the threshold values for the reaction (5).

IV. THE ROLE OF NANO-ROD ANTENNAS

The NAPLIFE experiments investigated the effect of golden NPs implanted in UDMA targets. At 25 mJ pulse energy in the case Au1, about 7-9 % Deuterium abundance was detected [16] compared to the observed amount of Hydrogen atoms in the emitted plum created after the laser shot.

The EPOCH PIC estimates [15] concluded that a single nano-rod antenna with dimensions 85 nm $\times$ 25 nm increases the electromagnetic field intensity (in the NP’s vicinity) by a factor of 25.9 (see Eq. (3) in Ref. [15]). Note however, that this result has been obtained for relatively low intensity $I_L = 4 \cdot 10^{15}$ W/cm$^2$.

According to these calculations, initially about $10^3$ electrons fluctuate between the ends of the resonant nano-antenna, with transverse peak momentum of about 0.15 MeV/c. The estimated maximal potential gradient is approximately 2.9 kV/nm.

Light absorption by embedded NPs was modelled earlier at 1000-fold larger laser energy in Ref. [28]. However, the obtained electric field (about 11.3 V/nm) is by more than two orders of magnitude less than in our estimate. The main reason is that non-resonant antenna/frequency combination was used! This indicates the importance of the use of resonant antennas. Such features are well known for classical antennas in radio communication.

Of course, the orientation of antennas with respect to the beam direction is rather important. Unfortunately, at present, target manufacturing does not produce oriented nano-antenna implantation.

Recently, the proton acceleration up to 93 MeV energy has been obtained with the PHelix laser at GSI [24]. However, proton energies exceeding 20 MeV were achieved in these experiments only for $I_L > 10^{20}$ W/cm$^2$. We expect that employing targets with properly oriented, resonant nano-antennas may produce 10-20 MeV protons even at lower intensities $I_L \lesssim 10^{17}$ W/cm$^2$.

The increased energy and momentum absorption, as well as the enhanced pulse intensity may lead to a significant increase of the proton acceleration in the vicinity of a nano-rod.

Under the influence of electromagnetic field, a single NP will be ionized, emitting some amount of electrons $\Delta N_s$. Let us express the number of ionization electrons as

$$\Delta N_s = \xi N_{Au}, \quad (23)$$

where $N_{Au}$ is the number of atoms in a single NP, and $\xi$ is the ratio of the ionized electrons per golden atom$^5$. According to calculations of Ref. [31], made for laser-irradiated golden foils, one can expect the values $\xi \approx 30$ at laser intensities $I_L \gtrsim 10^{18}$ W/cm$^2$. We consider $\xi$ as a parameter, although it may depend on characteristics of the laser, orientation and sizes of NPs, etc.

A proton located near the surface of a charged nano-rod gets an additional Coulomb energy

$$\Delta E \sim \frac{2 \varepsilon^2 \Delta N_s}{l_{\text{min}}} \approx 290 \xi \text{ keV,} \quad (24)$$

where $l_{\text{min}} = 25$ nm is the diameter of the nano-rod. In the numerical estimate we have taken $N_{Au} \approx 2.46 \cdot 10^6$.

We can also estimate the Coulomb energy per atom of the ionized NP:

$$\frac{E_C}{N_{Au}} \sim \frac{2 (\varepsilon \Delta N_s)^2}{N_{Au} l_{\text{min}}} \approx 290 \xi^2 \text{ keV.} \quad (25)$$

If $\xi$ is not too small, this energy will exceed the typical ‘binding’ energy $\approx 10$ eV per atom in a non-excited NP. According to Eq. (25), this occurs at $\xi \gtrsim 6 \cdot 10^{-3}$. In this case such NPs will be destroyed due to Coulomb-induced explosion.

It is interesting to estimate the additional (positive) charge provided by nano-particles in the IR. It is equal to $e \Delta N_s N_{ir}^{(NP)}$, where

$$N_{ir}^{(NP)} \approx \frac{V_{ir}}{L_{Au2}^2} \approx 3850 \quad (26)$$

is the number of NPs in the IR. In the second equality we substituted the volume of the IR, $V_{ir}$, from Eq. (16) and the average distance between nano-rods, $< L_{Au2} >$, obtained in Sec. IIb. Again we give numerical estimates for the case Au2.

Due to appearance of the additional charge, protons get larger acceleration in targets with NPs. Instead of Eqs. (21) and (22) we obtain the following estimate for the proton energy

$$E_p \sim \frac{e^2 (N_0 + \Delta N)}{R_{ir}} \approx (3.65 + 2.15 \xi) \text{ MeV,} \quad (27)$$

$^5$ At $\xi \lesssim 1$ ionization electrons come mainly from the conductivity zone of the Au metal.
where

$$\Delta N = \Delta N_s N^{(NP)}_{ir} \approx 9.5 \cdot 10^9 \xi$$

is the additional charge (in units of e) of the IR due to the presence of ionized nano-rods.

At large enough $\xi$ many deuteron-production processes with threshold energies larger than for the reaction (4) may be open. For example, at $\xi \sim 10$ one gets almost 7-fold increase of proton energy, i.e. $E_p \simeq 22$ MeV, which is above the threshold energy of the reaction (5). As compared to the case without NPs, where the number of ionized protons $N_p = N_0 \lesssim 10^{-4} N_{ir}$ is relatively small, at $\xi \gtrsim 10$ one can expect much larger $N_p$-values for target with nano-rods.

V. ESTIMATION OF DEUTERON TO PROTON RATIO

a) Rate equations for deuteron production

As discussed above, initially, ionization electrons and protons are produced in the cylindrical channel, which is much more narrow than the crater. We approximate the crater as a cone with radius $R_C$ and the depth $L_C$ (see Fig. 4). According to NAPLIFE data the volume of the crater for case Au2 can be estimated as

$$V_C = \frac{1}{3}\pi R_C^2 L_C \simeq 452 400 \mu m^3,$$

where we have substituted the values $R_C = 120 \mu m$ and $L_C = 30 \mu m$. One can see that $V_C$ is about 300 times larger than the volume of the cylindrical irradiation channel (16).

The ionized protons are produced in the cylindrical channel in a short interval $t \lesssim \tau_L \sim 40$ fs. At later times these protons propagate through the UDMA material and may induce transmutation reactions in the whole crater volume. Propagation of the produced protons with the energy $E_p = m_p v^2/2 \simeq 4$ MeV through the crater volume takes time

$$t_C \simeq \frac{R_C}{v} = \frac{120 \mu m}{0.092 \cdot 35 \mu m} \simeq 4.35 \text{ ps},$$

where $m_p$ and $v$ are, respectively, the mass and velocity of the proton.

The number of deuterons, which can be produced by the stripping reaction (4) can be estimated by using a simple rate equation

$$\frac{dN_d}{dt} = \langle \sigma_s v \rangle n_{13C} \cdot n_p(t) V(t),$$

where $\sigma_s$ is the cross section of the reaction (4), $n_{13C} \simeq \text{const}$ is the number density of $^{13}$C in the UDMA matter, and $V(t)$ is the volume occupied by the ionization protons.

Obviously, after a short initial stage, $t > \tau_L$, the total number of protons remains the same, but their density drops, such that $N_p = n_p V(t) = \text{const}$, where $N_p$ is given by Eq. (20). Therefore, all terms in the right hand side of Eq. (31) are approximately time-independent and one may write its solution at $t > \tau_L$ as

$$\frac{N_d(t)}{N_p} = \langle \sigma_s v \rangle n_{13C} \cdot (t - \tau_L)$$

Now we can estimate the $d/p$ ratio taking $t - \tau_L \sim t_C$, choosing the input parameters: $\sigma_s \simeq 2 \text{ mb}$, $v \simeq 0.092 c \simeq 2.76 \cdot 10^9 \text{ cm/s}$ (at $E_p \simeq 4$ MeV) and substituting $n_{13C} = 3.66 \cdot 10^{20} \text{ cm}^{-3}$. Finally we get the following ratio (for the case Au2)

$$\frac{N_d}{N_p} = 5.52 \cdot 10^{-18} \text{ cm}^{-3} \times 3.66 \cdot 10^{-20} \frac{1}{\text{ cm}^3} \times 4.35 \cdot 10^{-12} \simeq 8.8 \cdot 10^{-9}.$$ 

Nov let us estimate the $d/p$ radio for the reaction (5). One can do this using the Eqs. (31) and (32), but replacing $n_{13C}$ by the two-order of magnitude larger density of $^{12}$C nuclei $n_{12C} \lesssim 3.41 \cdot 10^{22} \text{ cm}^{-3}$, and substituting the cross section $\sigma_s$ of the reaction (5). For a rough estimate we choose the proton energy $E_p = 20$ MeV and the cross section $\sigma_s \simeq 25 \text{ mb}$ from Ref. [32]. Then, instead of (33) one obtains the estimate

$$\frac{N_d}{N_p} = 1.55 \cdot 10^{-16} \text{ cm}^{-3} \times 3.41 \cdot 10^{-22} \frac{1}{\text{ cm}^3} \times 1.94 \cdot 10^{-12} \simeq 1.02 \cdot 10^{-5},$$

which is by a factor 1160 larger than the $d/p$ ratio for the reaction (4).

Nevertheless, both $d/p$ values are too small to explain the D/H ratios extracted from the LIBS spectra [9, 16]. Note that both ratios are below the background ratio $N_d/N_p = 1.6 \cdot 10^{-4}$ in the Earth atmosphere under normal conditions.

However, one should bear in mind that to be visible in LIBS spectra, protons and deuterons should form neutral H and D atoms by recombination with electrons in the UDMA target. Therefore, the LIBS measurements give only indirect information about the abundances of ionized deuterons and protons, produced by the irradiation of UDMA targets. It is clear that the recombination process will shift the D/H ratio to larger values. The reason is that deuterons produced in reactions (4), (5) have significantly lower velocities as compared to protons. But, the recombination cross section increases with decreasing particle velocity. Therefore, the recombination probability for deuterons will be larger then for protons.

b) Role of electron-ion recombination

To estimate the numbers of the atomic hydrogen (H) and Deuterium (D) one can use the rate equations similar to (31), but replacing $\sigma_s$ by the recombination cross
section $\sigma_r$ and $n_{13C}$ by the electron density $n_e$. Then the ratio D/H will be given by the equation similar to (32) with the replacements $n_{13C} \rightarrow n_e$ and $\sigma_n \rightarrow \sigma_r$.

To get the quantitative estimate, we assume that recombination lengths for protons and deuterons are longer than the crater size $\sim 100 \mu m$. Then the ratio of recombination probabilities will be approximately equal to the ratio of recombination cross sections. According to Ref. [33], they are inversely proportional to the fifth power of the ion velocity [34]. Finally, we obtain the approximate relation

$$\frac{D}{H} \sim \left( \frac{m_d E_p}{m_p E_d} \right)^{5/2} \frac{d}{p}. \quad (35)$$

Note that we have assumed that propagation times of proton and deuterons in the target are inversely proportional to ion velocities. Now we can calculate the D/H ratios for two stripping reactions (4) and (5) considered before.

For the reaction (4), taking $E_p = 4$ MeV and $E_d = 1$ MeV, one has from Eqs. (35), (36)

$$\frac{D}{H} \sim 181 \times \frac{d}{p} \simeq 1.6 \times 10^{-6}. \quad (36)$$

One can see that this reaction still gives too small D/H ratio even if one takes into account the recombination corrections.

In the case of the reaction (5), substituting $E_p = 20$ MeV, $E_d = 5.92$ MeV (this value follows from Eq. (9)), and using Eqs. (34), (35), one gets the estimate

$$\frac{D}{H} \sim 118 \times \frac{d}{p} \simeq 1.2 \times 10^{-3}. \quad (37)$$

This value is still below experimental ratios for the Au2 target, but it is much closer to them. We plan to check these results by using more accurate calculations in the future. In particular, we are going to take into account feeding of the Balmer-$\alpha$ states from higher atomic levels.

VI. CONCLUSION AND OUTLOOK

In the present article we have analysed the results of NAPLIFE experiments on laser irradiation of polymer targets. They show a significant formation of deuteron atoms at high enough laser pulse energies. The deuteron’s yield increases with the concentration of implanted nano-particles.

In our analysis we have focused on the possibility of nuclear transmutation mechanisms, namely, due to the stripping reactions $p + C \rightarrow d + X$ with the carbon isotopes $^{12}$C and $^{13}$C. It is argued that proton energies achievable in the laser–target interaction are sufficient for deuteron formation in such reactions, especially for targets with implanted golden NPs.

Additional investigations are necessary for verifying these results. First, it would be desirable to reproduce our observations at higher laser energies e.g. at GSI/FAIR PHELIX and future ELI-ALPS facilities. Additional diagnostics tools would be desirable, including measurements of X-rays, neutrons, $\alpha$-particles, etc. We plan to perform new experiments with different pulse widths, target thickness and profiles of NPs.

On the theoretical side, the numerical modeling of target plasma evolution is necessary to estimate yields of secondary particles and their momentum distributions. Special investigation should be made to study properties of hot-spots created by nano-particles, and their role in the nuclear transmutation.

ACKNOWLEDGMENTS

Enlightening discussions with Miklós Kedves, Márk Aladi, and Oliver A. Fekete are gratefully acknowledged. T.S. Biró, M. Csete, N. Kroó, I. Papp, A. Szenes, and D. Vass acknowledges support by the National Research, Development and Innovation Office (NKFIH) of Hungary. Horst Stöcker acknowledges the Judah M. Eisenberg Professor Laureatus chair at Fachbereich Physik of Goethe Universität Frankfurt. Dénes Molnár acknowledges support by the US Department of Energy, Office of Science, under Award No. DE-SC0016524. We would like to thank the Wigner GPU Laboratory at the Wigner Research Center for Physics for providing support in computational resources. This work is supported in part by the Frankfurt Institute for Advanced Studies, Germany, the Eötvös, Loránd Research Network of Hungary, the Research Council of Norway, grant no. 255253, and the National Research, Development and Innovation Office of Hungary, for projects: Nanoplasmonic Laser Fusion Research Laboratory under project Nr-s NKFIH-874-2/2020 and NKFIH-468-3/2021, Optimized nanoplasmonics (K116362), and Ultrafast physical processes in atoms, molecules, nanostructures and biological systems (EFOP-3.6.2-16-2017-00005).

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Note, that $p \rightarrow n$ conversion may also occur by capturing electrons from bound atomic states. However, such processes are slow and may occur only for sufficiently heavy atoms.

One can also consider the deuteron production in exothermic reactions $p + p \rightarrow d + e^+ + \nu_e$. But this weak interaction has nonzero Coulomb barrier and the corresponding cross section is relatively small.

For a rough estimate we assume that target's electrons recombination with a hydrogen-like ion, can also be produced by capturing combination may also occur by capturing electrons from bound atomic states. However, such processes are slow and may occur only for sufficiently heavy atoms.

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