

Article

# Extreme Energy Density Confined inside a Transparent Crystal: Status and Perspectives of Solid-Plasma-Solid Transformations

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**Abstract:** It was demonstrated during the past decade that ultra-short intense laser pulse tightly focused deep inside a transparent dielectric generates the energy density in excess of several MJ/cm<sup>3</sup>. Such energy concentration with extremely high heating and fast quenching rates leads to unusual solid-plasma-solid transformation paths overcoming kinetic barriers to formation of previously unknown high-pressure material phases, which are preserved in the surrounding pristine crystal. These results were obtained with the pulse of Gaussian shape in space and in time. Recently it was shown that the Bessel-shaped pulse could transform much larger amount of material and allegedly create even higher energy density than that was achieved with the Gaussian beam (GB) pulses. Here we present a succinct review of previous results and discuss the possible routes for achieving higher energy density employing the Bessel beam (BB) pulses and take advantage of its unique properties.

**Keywords:** light-matter interaction, ultra-short laser pulses, high-pressure/density conditions, phase transitions

## 1. Micro-explosion studies with Gauss-shaped beam

The studies of confined micro-explosion during the last decade revealed the major features of this complicated phenomenon where the processes of electro-magnetic field/dielectric interaction, plasma formation and high-pressure hydrodynamics are intertwined. The concise description of these processes is as the following. The tight focusing of the laser beam deep inside a transparent crystal allows achieving the absorbed energy density in excess the strength of any material in a sub-micron volume surrounded by the pristine solid. After energy transfer from hot electrons to ions the expanding strong shock wave accompanied by the rarefaction wave starts propagating outside of this volume. After the shock decelerating and stopping the void, surrounded by shell of compressed and pressure modified material converted to the novel phases, is formed. All transformed material remains confined inside the bulk of undamaged material ready for the further studies. These studies employed the short intense laser beam with the Gaussian spatial and temporal intensity profile [1–4].

The short intense laser beam with the Gaussian spatial and temporal intensity profile tightly focussed inside a transparent crystal generates the energy density of several MJ/cm<sup>3</sup>. The pressure produced is in excess of a few TPa which is higher than the strength of any existing material (diamond has the highest Young modulus of 1 TPa = 1 MJ/cm<sup>3</sup>). The laser pulse, 150 fs, 100–200 nJ, 800 nm, tightly focussed inside sapphire with microscope lens ( $NA = 1.4$ ) creates the solid density plasma at the temperature of a few tens electron Volts ( $\sim 5 \times 10^5$  K) with the record-high heating rate of  $10^{18}$  K/s [1,2]. It was found that the novel (previously unobserved) high-pressure phases of Aluminium and Silicon were formed [3,4] following the ultrashort laser-induced confined microexplosion. Pressure/temperature conditions created in the micro-explosion are similar to those

in hot cores of stars and planets (“primeval soup” or Warm Dense Matter). The material converted to high pressure/temperature solid density plasma is then transformed into the novel solid phase during the ultra-fast cooling and re-structuring. The major difference from the core-star conditions is the record-fast cooling ( $\sim 10^{16}$  K/s) from plasma state to solid state. In the previous experiments the study of the pressure-affected materials was produced postmortem, well after the end of the pulse when transformed material was cooled down to the ambient conditions. The structure of laser-transformed material was determined by the synchrotron x-rays diffraction [3] and with the electron diffraction [4] in transmission electron microscopy (TEM) [4].

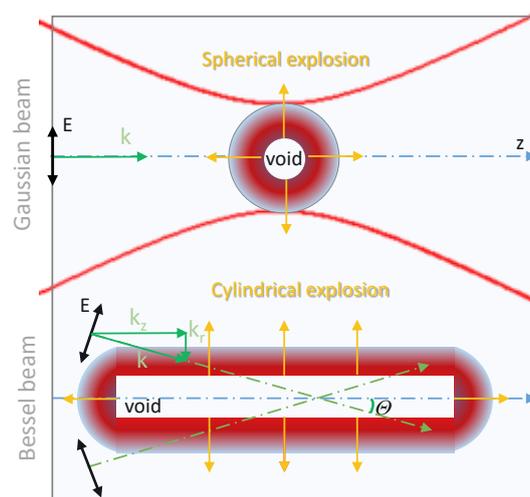
### 1.1. Novelty of the phase transformation path during and after confined micro-explosion

Solid transforms to a solid-density plasma state ( $T_e \sim 50$  eV) during the pulse time shorter than all energy relaxation times. Strong shock wave (SW) starts propagating from the energy deposition region several picoseconds after the pulse due to energy transfer from electrons to the ions. The shock wave decelerates and converts into the sound wave in the surrounding cold pristine crystal. The phenomenon is similar but not identical to the underground nuclear explosion: the massless energy carriers (photons) deliver the energy inside a transparent crystal without changing the atomic and mass content of a material. All laser-affected material is expelled from the energy deposition area by the combined action of shock and rarefaction waves, forming a void surrounded by the shell of material compressed against the surrounding cold pristine crystal. The material returns from the high-pressure plasma state (high entropy, chaotic) to the ambient conditions at room temperature/pressure, however attaining the phase state different from the initial solid state. In all known methods of high pressure phase formation the initial crystalline structure is re-structured, i.e. the atoms are moved from the initial arrangement to the new positions under the action of high pressure. During the transformation path under confined micro-explosion the initial state of a crystal is completely destroyed and forgotten. The irradiated material converted into the chaotic mixture of ions and electrons at high temperature. Therefore relaxation to the ambient conditions occurs along the unknown paths going through the metastable intermediate equilibrium potential minima. The theoretical (computational, modified DFT-studies) during the last decade searched for the possible paths of material transformations under high pressure from the initially chaotic (stochastic) state [5]. These studies uncovered many physically allowed paths for formation of multiple novel phases (including incommensurable phases) from the initially chaotic state. The confined micro-explosion method now is the only practically realised way for formation novel material phases from the plasma state preserving the transformed material confined inside the pristine crystal for the further structural studies.

### 1.2. Limitations of the confined micro-explosion method with the GB

There are limitations on the energy density and amount of laser-affected material in confined micro-explosion generated by tightly focused Gauss beam. The main limitation imposed by diffraction: the radius of the diffraction-limited focal spot is [2,6]:  $r_{Airy} = 0.61\lambda/NA$ ; defines the central Airy disk at the focus. For  $\lambda = 800$  nm and  $NA = 1.4$  one gets  $r_{foc} = 0.35$   $\mu\text{m}$  and focal area of  $0.38$   $\mu\text{m}^2$ . The absorption length in dense plasma equals to  $\sim 30$  nm giving the energy deposition volume  $\sim 10^{-14}$   $\text{cm}^3$ . With absorbed energy around 100 nJ the absorbed energy density amounts to  $10^7$   $\text{J}/\text{cm}^3 = 10$  TPa. The number of laser-affected atoms constitutes around  $10^{11}$  atoms (a few picograms) making structural studies extremely difficult. Therefore, the questions arise: is it possible to increase the absorbed energy density and/or increase the amount of the laser-affected material and thus the amount of the novel phase? Preliminary studies have shown that it is very difficult to overcome the energy density of several  $\text{MJ}/\text{cm}^3$  (several TPa of pressure) and increase the amount of laser-affected material using tightly focused Gauss beam. First, the ionisation wave moving towards the laser pulse with increasing intensity increases the absorbing volume and limits the energy density [7]. Moreover, the experiments with increasing laser pulse energy demonstrated

that at the energy per 150 fs pulse of 200 nJ the cracks surrounding the focal area destroyed the regular void formation [2]. Diffraction free Bessel beams (BB) arisen a hope to achieve higher energy density and larger amounts of the material affected. Below we describe the recent progress made with these studies. Then we describe some effects (and unresolved problems), which solutions may lead to further increase of the absorbed energy density.



**Figure 1.** Schematic presentation of radial micro-explosion driven inside transparent material by a focused linearly polarised (E-field) Gaussian beam (GB) and Bessel beam (BB) with projection  $E_z$  along the optical axis (z-axis);  $\theta$  is angle with the optical axis (wavevector,  $k = \sqrt{k_r^2 + k_z^2}$  is shown on the upper half of the conical wave). Resonant absorption of the  $E_r$  component (along the radial direction  $k_r$ ) allows a higher energy density deposition for the cylindrical micro-explosion. With the central void diameter in sapphire comparable for the GB [1] and BB [8] pulses the volume was 180 times larger in the case of BB.

## 2. Status of the BB-transparent crystal interactions

It was demonstrated recently that the BB (150 fs, 2  $\mu$ J) focused inside sapphire produced the cylindrical void of 30  $\mu$ m length and 300 nm diameter [8]. The void volume,  $V = 30 \mu\text{m} \times \pi r^2 = 2.12 \times 10^{-12} \text{ cm}^3$ , appears to be two orders of magnitude larger than that generated by the GB. The conclusions based solely on the void size measurements and on the energy and mass conservation laws without any ad hoc assumptions about the interaction process are the following [9]. The material initially filled the void was expelled and compressed into a shell by the high-pressure shock wave. The work necessary to remove material with the Young modulus  $Y$  from volume  $V$  equals at least to  $Y \times V = 0.848 \mu\text{J}$  ( $Y = 4 \times 10^5 \text{ J/cm}^3$  – the Young modulus of sapphire). This is the evidence of strong (> 40%) absorption of the pulse energy. In order to generate a strong shock wave capable of expelling such amount of material the absorbed energy should be concentrated in the central spike with the much smaller diameter than that of the void (the absorbed energy density is still unknown either theoretically nor experimentally).

The unique features of the diffraction-free Bessel beam spatial distribution of intensity in the focal area allow to understand some of the experimental findings and indicate to new problems and opportunities. The spatial distribution of intensity across the cylindrical focal volume in a transparent medium unaffected by light and observed experimentally is close to the Durnin's

solution,  $J_0^2(k_r r)$  [10]: the central spike surrounded by circular bands with the maximum of intensity on the axis approximately five times higher than in the next band.

Parameters of the quasi non-diffracting BB created by any device from incoming cw-laser pulse in air (axicon, circular slit, spatial light modulator (SLM), etc.) with the cone angle  $\theta$  are the following (Fig. 1): radius of incoming beam before BB-creating device,  $R$ ;  $z_{max} = R / \tan(\theta)$ ;  $k_r = k \times \sin(\theta)$ ;  $k_z = k \times \cos(\theta)$  [10]. Building the intensity distribution in the low intensity short pulse BB occurs in a similar way to that as with cw-laser as it was demonstrated experimentally [8]. It is worth noting that in these experiments during the pulse time the beam propagates a distance comparable with the length of elongated cylindrical focus ( $Z_{max}$ ),  $L_{pulse} = t_{pulse} \times c/n$  ( $n$  - is refractive index in a transparent crystal unaffected by laser). For example, 150 fs (800 nm) low intensity pulse in sapphire propagates  $\sim 30$  microns that is close to  $t_{pulse} \times c/n = 30 \mu\text{m}$  [8].

In the non-absorbing media the length of the focus (the distance where diffraction is strongly suppressed) apparently is independent on the pulse duration. On the axis of the BB (in the focal area) the time of the interaction of electromagnetic wave with matter could be shorter than the pulse duration. Therefore a beam of any duration allegedly propagates the same distance  $Z_{max}$  allowed by the focusing device. This seemingly obvious statement should be confirmed experimentally.

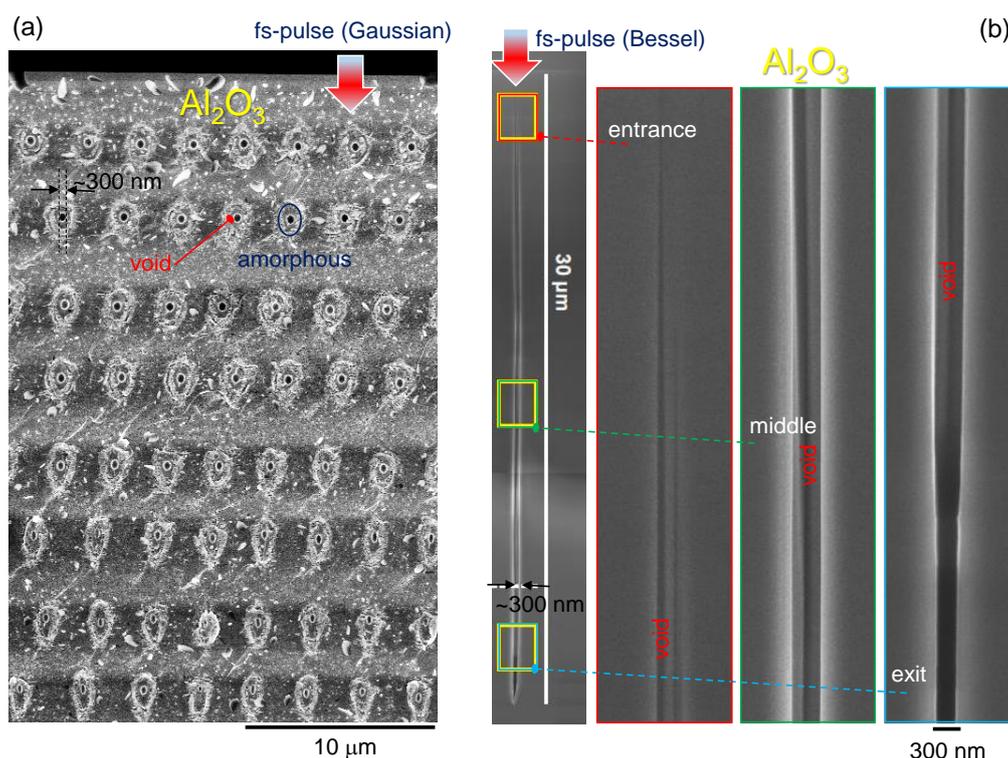
Under the action of intense pulse the ionization breakdown occurs early in the pulse time near the central spike where intensity has maximum. The studies of interaction process of intense BB at intensity above the ionization threshold are absent for the best of our knowledge. Estimates, suggestions and problems relating to the formation of the intensity distribution and interaction process based on the studies of confined micro-explosion and intense short pulse interactions with dielectrics are presented below. Experimental observation of the void formation by GB and BB pulses is shown in Fig. 2. The BB pulses are used to dice transparent materials [11] and to inscribe high efficiency optical gratings in silica [12].

### 2.1. Control of energy deposition by BB pulses

In short intense BB interaction with the initially transparent medium the ionization threshold is reached at the axis of the focal volume where intensity attains the maximum value. It occurs early in the pulse time close to the beginning of the elongated focal region. Narrow cylindrical plasma region is created along the axis. Incident light starts absorbing in plasma. Let's take the incident field structure near the axis as the following  $E (E_r, E_\phi = 0; E_z)$ ;  $H (0; H_\phi, 0)$ . Then the Poynting vector reads,  $\mathbf{S} = \frac{4\pi}{c} (\mathbf{E} \times \mathbf{H})$ . Therefore the energy flows are generated inward along the radius and along the  $z$ -axis in direction of the beam propagation:  $S_r = \frac{4\pi}{c} (H_\phi \cdot E_z)$  and  $S_z = \frac{4\pi}{c} (H_\phi \cdot E_r)$  [ $\text{J}/(\text{cm}^2\text{s})$ ].

Thus, by changing the cone angle one can control the radial and axial energy flows. The interaction mode of intense BB with transparent crystal dramatically changes after the ionization threshold is achieved. The surface, where the real part of the permittivity is zero,  $\epsilon_{re} = 0$ , separates the dielectric ( $\epsilon_{re} > 0$ ) and plasma ( $\epsilon_{re} < 0$ ) regions. The gradient of the permittivity is directed along the radius of a cylinder. The energy flow goes inward in the radial direction. Thus the incident wave splits into the evanescent and reflection waves. The resonance absorption occurs in the vicinity of the zero-epsilon surface creating a plasma wave (plasmon) propagating along the radius in the direction to the axis of cylindrical focal region. The evanescent wave decays along the radius in the same direction. Thus zero-permittivity surface generates simultaneously coherent plasmons and evanescent waves coming together (focusing) to the axis of cylindrical focal region. One may expect that coupling of evanescent waves and plasmons also contribute into increase of the intensity and energy density near the axis in a way similar to that discovered in the studies of extraordinary optical transmission (EOT) through sub-wavelength hole arrays [14].

The ideal diffraction free beam is the monochromatic Bessel beam [10], created via superposition of plane waves whose wave vectors are evenly distributed over the surface of a cone. The Bessel function of the first kind zero order,  $J_0$ , is a sum of the Hankel functions of the first and second kind [15] where the inward energy flow is balanced by the outward flow.



**Figure 2.** SEM side-view images of the voids made with ultra-short Gaussian [1,13] (a) and Bessel [8] (b) single pulses in sapphire. Focusing of the 800 nm/130 fs Gaussian pulses of  $\sim 150$  nJ energy was carried out with an objective lens of numerical aperture  $NA = 1.3$  and was stacked into a vertical plane of the void-structures [13]. This plane was used to split sapphire sample for the side-view SEM observation. The voids made at larger depth were affected by spherical aberration which reduced the void and elongated amorphous region. The 800 nm/140 fs, 2  $\mu$ J energy Bessel pulses were used to make cylindrical voids of diameter  $\sim 300$  nm revealed by focussed ion beam (FIB) milling [8].

It was suggested [16] that the quasi diffractionless BB can be presented as the result of the interference of two conical running Hankel beams, carrying equal amounts of energy towards and outwards the beam axis, and yielding no net transversal energy flux in the BB. Interference of two Hankel beams with different amplitudes creates unbalanced BB where the net radial energy flux appears. Unbalancing creates the inward radial energy flux from the conical tails of the beam. The study of stability in the frame of non-linear Shrodinger equation (NLSE) equation revealed that the Bessel-like solutions in pure Kerr media are unstable [17].

In the interaction of intense short pulse BB with transparent dielectric at the intensity below the ionization threshold the BB apparently retains its balanced structure. After the plasma formation the energy flow directed inward to the axis is created due to absorption leading to destruction of this balance. One may argue that after ionization threshold the Hankel function of the first kind might be considered as an appropriate approximation of the field distribution near the axis of cylindrical focus being the exact solution of the Bessel equation describing the electric field increasing while focusing. One may conjecture that the BB becomes unstable tending to focus onto the cylindrical axis thus creating the energy density higher than tightly focused but diffraction limited Gaussian beam.

Let us now consider the relation between the pulse duration, absorption, focal region length and laser-affected area length. In short intense BB interaction with the initially transparent medium the ionization threshold is reached at the axis of the focal volume where the intensity has maximum. It occurs early in the pulse time close to the beginning of the elongated focal region. Intense pulse converts the initially transparent material into strongly absorbing plasma practically at the

moment of its arrival at some space point. Therefore the plasma region gradually increases along the axis as the pulse proceeds until the end of the pulse. The last portion of light arrives after travelling through the transparent crystal the distance  $t_p \times c/n$ . The laser-affected distance then reads,  $L_{las} = (t_p \times c/n) \cos \theta$  ( $\theta$  is the half-cone angle; Fig. 1). One can see now the difference between the BB-affected area in transparent medium (diffraction-free focus) and laser-affected area in intense short pulse laser/crystal interaction. For sufficiently short pulses laser-affected area might be shorter than the diffraction-free zone,  $L_{las} < Z_{max}$ . Thus laser pulse duration might be another lever (along with the cone angle) to control the energy deposition volume.

Experiments demonstrated that short intense BB could affect much larger amount of material producing solid-plasma-solid transformation (direct measurements) at allegedly pressure of several TPa (conclusions on the basis of the analysis of the experiments) [8,9]. J. Hu [18] measured the average speed of the shock wave,  $v_{sw} \approx 60$  km/s, during the cylindrical micro-explosion, generated by the BB in sapphire, by the pump-probe technique. The estimate of the driving pressure based on this measurement,  $P_{sw} = \rho_0 v_{sw}^2 = 14.4$  TPa ( $\rho_0$  is the initial mass density of sapphire), gives the direct experimental evidence of the extreme energy density created by the BB in the focal volume.

There are indications from the theoretical studies [9] that the originally stable diffraction free BB at high intensity in the presence of strong ionization nonlinearity may become unstable. Now it is difficult to conclude if it may happen in a way similar to the self-focusing instability with Kerr-like non-linearity (rather not because the paraxial approximation is invalid in this case) or similar to the instability of two unbalanced Hankel beams, which seems more relevant to the case (again the ionization non-linearity should be accounted for).

The oblique incidence, inherent for the formation of the BB and long focus, implies the possibility of the surface wave (plasmon) formation and propagation along the zero-real-permittivity surface at the same time with the plasmon moving radially due to the resonance absorption. The plasma wave may converge to the axis contributing to increase in the absorbed energy density. One may conjecture if it might be relevant to some kind of the Langmuir collapse.

It would be crucially important to find the electric field distribution up to the central axis in order to determine the absorbed energy density. It requires a solution of the Maxwell equations in cylindrical geometry coupled to material equations accounting for the change in the permittivity (electrons' number density and collision rate) in accord to intensity in any space/time point. It is a formidable task however it can be clearly formulated for the numerical solution. Different approximations may also be discussed.

It was demonstrated experimentally [18] that the Bessel beam induced micro-explosion in sapphire, producing open-ended channel, proceeds as axial-symmetric cylindrical explosion and a mass conservation was experimentally validated [19]. Therefore the direct theoretical modeling of the cylindrical explosion after the energy deposition of the BB beam inside a narrow on-axis cylinder also can be performed in the frame two-temperature plasma hydrodynamics in cylindrical geometry in a way similar to that was done with the Gauss beam in spherical geometry [2].

### 3. Conclusions and Outlook

In conclusion we should state that further progress in achieving and steering the high energy density strongly depends on the future pump-probe experiments, which will register with time/space resolution the history of the BB generated micro-explosion, processes of returning to the ambient state and new phases formation. It is worth to show the time and space scales for the succession of events comprising such a history that might in some approximation be extracted from the previous studies [2,4,9].

Let's suggest the BB,  $2 \mu\text{J}$ , 800 nm, 150 fs, impinges sapphire crystal several tens of microns thick creating a focal region of  $\sim 30 \mu\text{m}$  long at the ten microns depth from the outer surface of a sample. The stages of successive transformations are the following, time count starts at the beginning of the pump pulse:

1. Low intensity stage before ionisation threshold lasts a few fs at the beginning of the pulse;
2. As the ionisation threshold is attained, the cylindrical plasma region is created at the axis of the focal region with the diameter less than a micron. One should note that the full length of the focal region of 30  $\mu\text{m}$  is reached at the end of the pulse, assuming that light propagates as in unaffected sapphire with the speed of  $c/n \sim 2 \times 10^{10}$  cm/s;
3. Cylinder diameter of the energy absorption region to the end of the pulse allegedly might be around the doubled absorption length in a dense plasma  $\sim 60$  nm;
4. The shock wave is created after the energy transfer from electrons to ions in 7-10 ps time span;
5. The shock wave propagates during another 4-6 ps until it is converted into the acoustic wave, effectively stopped by the cold pressure of the crystal ( $\sim$ Young modulus of sapphire). The void surrounded by the shell of compressed material is formed by the rarefaction wave;
6. Thermal wave of conventional heat conduction spreads into the laser-unaffected crystal cooling the laser-affected area down to the ambient conditions during tens of nanoseconds. The material re-structuring occurs most probably during the stages 5 and 6. The whole area affected by the heat from the laser-heated region is a cylinder with length around 32-34 microns with a diameter about 2-4 micrometers.

Thus, the whole area affected by the shock and heat waves from the energy deposition region is a cylinder 30 micron long and a few microns in diameter. Time span for the whole processes of material transformation is around tens of nanoseconds. Recent arrival of X-ray free electron lasers with pulse duration as short as 7-15 fs and the photon energy 8-10 keV currently available at EuroXFEL at DESY in Hamburg and in SACLA XFEL at Spring-8 at Riken Institute in Japan creates new opportunities for uncovering the mechanism of formation of the new states of matter. Up to 17 keV pulses expected in near future at the SLAC National Acceleration Laboratory in Stanford, are the ideal sources to be used as a probe pulses to uncover the process of formation such unusual material states. For such experiments, tailored axial intensity distribution of the optical BB pulses can be prepared using diffraction optical elements [20], which can be made with a central hole for the co-axial fs-optical-pump and fs-X-ray-probe.

To conclude the light (or x-ray) probe with sub-picosecond duration and sub-micron spatial resolution may shed light on the unusual formation of novel high-pressure phases starting from "primeval soup" (Warm Dense Matter) to the solid state at the ambient being preserved on the laboratory table top and confined inside a bulk of pristine crystal ready for the further structural studies [7].

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