Table-top generation and spectroscopic study of ~10 TPa high-energy density materials with C_{60}^{+} hypervelocity (~100 km/s) impact

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Abstract. The use of nanoparticles as flyers to create shock pressures exceeding 10 TPa and to investigate the matters in planetary or stellar interiors has been pursued by the author for two decades. Previous studies led by the author at Brookhaven National Lab (BNL) in 1994 proved that such ultra-strong shocks can be generated with charged bio and water nanoparticles by accelerating them with an electrostatic accelerator and impacting them on solids at ~ 100 km/s. The author in 2008 showed that the BNL nanoplasmas produced intense bursts of soft x-rays (hν ~ 100 eV) from optical decay of excimer-like Metastable Innershell Molecular State, MIMS, formed by inner-shell electron excitation. The conversion efficiency from the nano-flyer kinetic energy to the radiation energy was unexpectedly high, ~38%, which was attributed to high efficiency pressure ionization conversion of impact energy to MIMS excitation energy and MIMS collective optical decay in tens of fs via Dicke Superradiance. Now, this paper reports an experimental study performed with C_{60} as a nano-flyer that permitted reduction of the size and complexity of the apparatus by orders of magnitude compared with the BNL one. The present results confirm the BNL results unambiguously, demonstrate a pathway to scaling up of soft x-ray intensity, and open doors to a wide range of applications from lithography to inertial fusion.

1. Introduction
Investigations of matters under extreme conditions as in planetary or stellar interiors are crucial for advancing a wide range of scientific and technological fields, such as astrophysics [1,2], inertial confinement fusion [1,2], x-ray lasers [3], and material science [4-7] and biological science [7]. Traditionally, such matters have been created and investigated with the use of ultra-large scale experimental setups, such as underground nuclear explosions [8,9], laser fusion facilities [10], z-machines [11] and X-ray Free-Electron Lasers (XFEL) [3,6,7].

Recent theoretical and experimental studies revealed matters under such extreme conditions have highly rich quantum characteristics [4,5,6,12-16], including that metals may become insulators at high pressures [4,5,12]. The Hugoniot curves of many materials, such as aluminium, silicon, and iron, show atomic quantum effects of inner-shell excitation and ionization near or above pressures of 10 TPa [13,14]. The theoretical data were compared with experimental data, but large uncertainties in the experimental data prevented definitive confirmation of the existence of the inner-shell quantum effect.
The effects of crystalline or molecular states, of which transition energies are predicted to range from hundreds of eV to tens of keV, in such matters have also been theoretically investigated [4,5,12,15,16]. In particular, Winterberg [16] predicted the existence of quasimolecular states in the pressures on the order of 10 TPa and super-intense x-ray radiations during their formation. On the other hand, experimental studies that could illuminate the theoretical predictions have been very scarce.

The large scale approaches including laser fusion [1,2] show that producing and investigating matters with pressure exceeding 10 TPa are extremely complex and challenging. Our approach to overcome these challenges has been to shrink the flyer size to atomic dimension [17,18]. Once the size of flyers becomes smaller than 10 nm, they can be charged and readily accelerated to velocities exceeding 100 km/s with the use of established accelerator technologies. However, the fundamental question is what the minimum number of atoms in the flyers is required for inducing hydrodynamic shock behaviour. The author and his colleagues at BNL demonstrated that the minimum number is about 50 [17,18]. We further demonstrated that pressures exceeding 10 TPa can be generated with nano-flyer impact and discovered the nano-flyer generated anomalous signals [17,18]. The author in 2008 showed [19] that the anomalous signals were extremely intense narrow bandwidth (FWHM ~ 10%) soft x-ray photons that are generated during the impact time scale of 10 fs that is orders of magnitude shorter than the optical decay time of typical excited states of atoms. Another surprising aspect of the discovery was that the conversion efficiency of the nano-flyer impact energy to photon energy was as high as ~38%, which indicates virtually 100% of atoms are ionized during the impact [19]. However, these atoms are expected to decay predominantly via non-radiative channels, therefore, such high intensity radiation could not be explained with conventional models.

The author recently proposed excimer-like Metastable Innershell Molecular State (MIMS) in the 10 TPa matters to explain the extremely high population of the radiating excited state [19]. Furthermore, since impact created nanoplasmas are typically smaller than or on the order of the wavelength of radiation involved, Dicke Superradiance [19,20] mechanism speeds up the radiative process by orders of magnitude, thus the optical decay process becomes more predominant than the non-radiative processes. Thus, the nano-flyer approach permits efficient optical probing of the transient states in the time scale of tens of fs, in which the electron temperature is much smaller than the ion temperature. Therefore, the approach can probe into non-equilibrium states of matter, which can provide highly favourable environments for forming delicate molecular states that cannot be easily generated otherwise.

2. Experimental

The use of bio and water nanoparticles as flyers as in BNL experiments requires sophisticated ion sources, extensive pumping systems, mass spectrometric systems and large accelerator systems [17,18]. The author recently realized that the size and the complexity of the apparatus can be drastically reduced if C_{60} is used as a flyer. The reason for the choice of C_{60} lies on the fact that the BNL data [18] showed that the onset of the hydrodynamic shock behaviour begins near the size of ~50 molecules (or heavy atoms). Because C_{60} has more than 50 atoms, and because the science and technology of C_{60} have been well established, C_{60} is believed to be one of the best nano-flyer candidates. Moreover, recent molecular dynamics simulations [21] show that at the impact kinetic energy of 15 keV C_{60} impact (v=94 km/s) results in a strong shock that creates craters while that of a Ga atom does not. Therefore, the author built a tabletop apparatus that can generate, ionize and accelerate C_{60} to velocities exceeding 100 km/s, and extensively investigated the soft x-ray generated by the nano-flyer impact.

The exact shock pressure that can be generated by C_{60} impact is difficult to estimate. Our BNL experiments [17,18] were able to measuring the energy deposition (dE / dx) in thin films when bio-nanoparticles penetrate through, and the data resulted in the energy densities on the order of 10^{13} J/m^3. Because the pressure is approximately equal to energy density, the impact pressures could be roughly
estimated from the energy deposition to be 10 TPa. It was found [17,18] that the estimated pressure from the energy deposition was on the order of the shock pressure, $P_s$, estimated by [22]

$$P_s = \frac{4}{3} \rho_p v^2,$$

$$\rho_s = \frac{(\gamma+1)}{(\gamma-1)} \rho_T,$$

where $v$ is the nano-flyer impact velocity, $\rho_p$ and $\rho_T$ are normal densities of projectiles and targets, $\rho_S$ is the shocked material density, and $\gamma$ is the adiabatic index. Based on this theory, the C$_{60}$ impact energy for generating 10 TPa shock compression would require at least 17 keV. However, because C$_{60}$ is small, there should be considerable sharing of its energy with target atoms in very short time, equation 1 should result in only the theoretical upper bound of the shock pressure that can be generated by C$_{60}$ ion impact. The details of such behaviours are subjects of future investigation.

Briefly, C$_{60}$ ions were generated by photoionizing C$_{60}$ clusters that were in turn sublimated from commercial C$_{60}$ powder in a Knudsen cell in a tabletop apparatus schematically illustrated in figure 1. Here all the required components were chosen such that they can increase apparatus simplicity and soft x-ray production scalability. The Knudsen cell temperature was kept below 600 C to minimize thermal fragmentation of C$_{60}$. Photoionization with photon energies of ~10 eV, i.e. close to the photonionization threshold energy of 7.6 eV, was used to minimize ionization-induced fragmentation [23]. The photons were generated by a Krypton-line lamp manufactured by Resonance Ltd, which was capable of generating $1\times10^15$ photons/sec. The generated C$_{60}^+$ ions were accelerated towards a cathode at voltages up to -100 kV. The soft x-ray photons generated by the C$_{60}^+$ impact were observed with three thin film coated Si-photodiodes by Opto Diode Corp. for detecting photons with energies in the ranges of 40-73 eV (Detector A), 60-120 eV (Detector B), and 80-170 eV (Detector C), respectively. According the manufacturer, their responses are absolute and linear over 12 decades. The main noise contribution to the detector resulted from the secondary electrons from C$_{60}^+$ impacting on the cathode, which were accelerated to the detectors with the same kinetic energy as C$_{60}^+$. These electrons can penetrate through the thin films coated on the detectors, thus are registered in the detectors along with photons. Arrays of rare-earth permanent magnets were used to discriminate against the accelerated secondary electrons. The average magnetic fields in front of the detectors are on the order of 1 kG. The C$_{60}^+$ impact also generate secondary negative ions, but they were efficiently blocked by the thin films coated on the detectors.

Figure 1. Schematic diagram of the experimental setup.
3. Results and Discussions
The author previously proposed that the BNL soft x-ray yields as a function of nano-flyer impact energy could be described with the Arrhenius equation that suggests an existence of a barrier to forming MIMS [19]. Therefore, in this work the signals from the C$_{60}^+$ impact measured with the three detectors are presented as a function of inverse kinetic energy (1/KE) in figure 2 (Detector A, B) and figure 3 (Detector C). The data follow Arrhenius behaviour as in the BNL data [17,18]. With 1/KE < 0.04 (E>25 keV), the data from all the detectors show identical behaviours, which were determined to result from the high energy secondary electron infiltration through the magnetic field, because their trajectories have larger radii of curvature. With 1/KE > 0.04 (E<25 keV), the detector A and B data (figure 2) show responses from soft X-ray photons, which are greater than that of the secondary electron noise, while the detector C data (figure 3) do not.

The analyses of the current data with C$_{60}^+$ nano-flyers resulted in the activation energies of 1.08 keV/atom and 1.44 keV/atom for detectors A and B, respectively. The difference is interpreted to reflect the wide bandwidth spectra of MIMS radiative decay, presumably as in bound-free radiative decay of excimers. Figure 3 (Right) shows the quantum efficiencies of the three detectors. The analysis of the detector A signals (figure 2a) resulted in the conversion efficiency of the C$_{60}^+$ kinetic
energy to photon energy ≥35%, which is similar to the result (~38%) by Bae et al. [17-19], indicating the existence of an underlying unifying theory.

The quantum mechanical details of physical characteristics and the genesis of MIMS are still being developed. The author propose here that MIMS would be an analogue to excimer states that are typically formed by a ground state and an excited state of closed electron shell atoms. One of more common ways of excimer state decay is bound-free radiation, which involves a radiative decay from a metastable bound state to a repulsive free state. If the proposal on the excimer like bound-free radiative decay of MIMS is valid, the MIMS radiation should have photon energies similar to but lower than the energies of the parent atomic inner-shell excited states. For the present aluminium targets, the radiative recombination of electrons from the valence band to the LIII or LII levels [25] results in emission that ranges in energy from approximately 62 eV to ~73 eV. Vinko et al. measured the emission spectra from the solid density plasma generated by intense L-shell photoionization with XFEL [7,24]. Figure 3 (Right) also shows a graphical representation of their data [24], which can be fitted with the theoretical curve with 10 holes per 32 atoms (~30% ionization). Because binding energy of MIMS is expected to be much smaller than the inner-shell excitation energy of the constituent atoms, the spectrum of aluminium MIMS is expected to be similar to the aluminium L-shell radiation. In this case, the aluminium MIMS can only be detectable on the detectors A and B, and the responses of the two detectors would be roughly equal, which are in agreement with the data shown in figures 2 and 3.

It is proposed here that the Al L-shell holes are generated by pressure ionization [26] and they subsequently form excimer-like MIMS during the inertial confinement stage of the impact generated nanoplasmas. In this picture, the barrier to forming MIMS results from the pressure ionization process [26]. For bulk aluminium plasma, the total decay of such an L-shell vacancy is dominated by the non-radiative Auger decay, and is estimated to be on the order of 40 fs [27]. Typically, the Auger decay probability is two orders of magnitude larger than the radiative decay probability for lighter atoms, thus only ~ 1% of the excited energy will be converted to radiative energy [27]. However, in compressed materials, other non-radiative decay processes, such as collisional decay, become competitive with the above processes. Therefore, the radiative decay probability in the compressed nanoplasmas is expected to be considerably smaller than 1%. However, in the nanoplasmas, because the wavelength of the soft x-ray photon (~20 nm) is larger than the nanoplasma size (a few nm), the entire community of MIMS decay collectively in a time scale of τ/N [19,20], where τ is the radiative lifetime of MIMS (a few ps), and N is the number of MIMS in the impact generated nanoplasma. Thus, the nanoplasma with N~100 is estimated to have a lifetime on the order of a few tens of fs, which is shorter than the L-shell Auger decay time [27]. This shortening of the radiative lifetime results in predominant Dicke Superradiance [19,20]. More detailed MIMS and its radiation characteristics that will illuminate MIMS quantum structures and formation mechanisms are underway with a soft x-ray spectrometer in the author’s lab. In addition, various methods of scaling up of soft x-ray generation are currently investigated. For example, the author and colleagues at Los Alamos National Laboratory are currently investigating the use of explosive driven compressors for the scaling up [28].

4. Conclusions

An innovative desktop experimental technique is demonstrated to create 10 TPa HEDM by using the ions of buckyballs (C60) as nano-flyers and to diagnose them by observing the soft X-ray radiation from the impact. The highly compressed nanoplasma created by the impact behaved like a “nanostar” by emitting a burst of superradiant soft X-ray photons at a kinetic energy to photon energy conversion efficiency exceeding 35%. The superradiance is proposed to result from a collective optical decay of excimer-like MIMS [19] in a compressed nanoplasma via Dicke Superradiance [20]. It is anticipated that further higher-resolution spectroscopy of the superradiance as a function of impact velocity, thus pressure, using soft x-ray spectrometers will reveal the evolution dynamics and quantum
characteristics [13-16,19] of the matters under extreme conditions, and the pathway to harnessing the radiation power from them for a wide range of applications from lithography to inertial fusion [28].

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References
[1] Davidson R 2003 Frontiers in High Energy Density Physics: The X-Games of Contemporary Science (Washington, DC: National Academies Press)
[2] Drake R P 2009 Phys. Plasmas 16 055501
[3] Rohringer N et al. 2012 Nature 481 488
[4] Pickard C J and Needs R J 2010 Nature Materials 9 624
[5] Ma Y M et al. 2009 Nature 458 182
[6] Nagler B et al. 2009 Nature Physics 5 693
[7] Vinko S M et al. 2012 Nature 482 59
[8] Lomonosov I V 2007 Laser Particle Beams 25 567
[9] Vladimirov A S et al. 1984 Sov. Phys. JETP Lett. 39 85
[10] Taylor A et al. 2007 Science 315 1092
[11] Haines M G et al. 2006 Phys. Rev. Lett. 96 075003
[12] Gatti M Tokatly I V and Rubio A 2010 Phys. Rev. Lett. 104 216404
[13] Rozsnyai B F et al. 2001 Phys. Lett. A 291 226
[14] Pain J C 2007 Phys. Lett. A 362 120
[15] Younger S M et al. 1988 Phys. Rev. Lett. 61 962
[16] Winterberg F 2008 Jour. Fusion Energy 7 250
[17] Bae Y K, Chu Y Y and Friedman L 1995 Phys. Rev. A 51 R1742
[18] Bae Y K et al. 1996 Nucl. Inst. Meth. Phys. Res. B 114 185
[19] Bae Y K 2008 Phys. Lett. A 372 4865
[20] Dicke R H 1954 Phys. Rev. 93 99
[21] Postawa Z et al. 2004 J. Phys. Chem. B 108 7831
[22] Zel’dovich Y B and Raizer Y P 1996 Physics of Shock Waves and High-Temperature Hydrodynamic Phenomena (Mineola, NY: Dover)
[23] De Vries J et al. 1992 Chem. Phys. Lett. 188 159
[24] Vinko S M et al. 2010 Phys. Rev. Lett. 104 225001
[25] Henke B L, Gullikson E M and Davis J C 1993 X-ray interactions: photoabsorption, scattering, transmission, and reflection at ED50-30,000 eV, ZDI-92. Atomic Data Nucl. Data Tables 54 181-342
[26] Nantel M et al. 1998 Phys. Rev. Lett. 80 4442
[27] Almbladh C, Morales A and Grossmann G 1989 Phys. Rev. B 39 3489
[28] Bae Y K 2013 Phys. Lett. A 377 3304