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November 22, 2011

Institute of Nuclear Materials Management 52nd Annual Meeting Palm Desert, CA, United States July 17, 2011 through July 21, 2011

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Overview of MEGa-ray-based Nuclear Materials Management Activities at the Lawrence Livermore National Laboratory

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Abstract:

Mono-energetic gamma-ray (MEGa-ray) sources can be readily produced via the optimized interaction of pulsed lasers with relativistic electron beams (inverse laser-Compton scattering). Such sources provide unrivaled photon source monochromaticity, pulse brightness and flux. In the MeV spectral range, MEGa-ray sources can be greater than 15 orders of magnitude higher peak brilliance than the world's largest synchrotrons. Optimized MEGa-ray sources can efficiently excite the isotope-specific resonant structure of the nucleus, i.e. nuclear resonance fluorescence (NRF) and can in turn be used in unique ways to detect the presence, location and amount of specific isotopes within complex objects. A review of the design, optimization and development of past and future MEGa-ray machines as well as a survey of nuclear applications being pursued with them at the Lawrence Livermore National Laboratory (LLNL) is presented.

Introduction:

LLNL has more than a decade of experience in the development of laser-based Compton light sources. Over the past 7 years it has concentrated on the optimization of these sources for the production of mono-energetic gamma-rays (MEGa-rays) which can in turn be used to excite electromagnetic resonances of the nucleus, i.e. nuclear resonance fluorescence (NRF). NRF signatures depend upon the number of protons and neutrons in the nucleus and are unique fingerprints of each isotope. By monitoring the attenuation of resonant MEGa-ray photons one may determine with isotopic specificity the presence, amount and distribution of materials in arbitrary objects. Furthermore, since most NRF transitions of interest occur at photon energies that are highly penetrating, e.g. in the 1 MeV and 4 MeV spectral range, it is possible to assay and detect materials inside of thick objects with MEGa-ray beams.

At LLNL, efforts are currently underway to create a first-of-its-kind, compact, tunable (0.1 MeV to 4 MeV) MEGa-ray source whose spectral flux will be up to 5 orders of magnitude higher and bandwidth 2 orders of magnitude narrower than any existing MEGa-ray capability. This new source will be the centerpiece of a dedicated Nuclear Photonics Facility at LLNL whose aim is to enable NRF-based studies and demonstrations of relevance to a wide variety of unresolved nuclear problems and issues. Examples include: rapid (fractions of a second) detection of concealed nuclear material, high precision (better than 100 parts per million) non-destructive assay of spent nuclear fuel assemblies, isotope-specific, high-resolution (less than 10 micron

spatial resolution) 3D imaging of nuclear materials in existing waste containers and waste processing streams.

Compton Sources:

Compton scattering of laser photons from relativistic electrons was first demonstrated in 1965. [1] In that experiment a giant pulse ruby laser interacted with 6 GeV electrons and created approximately 8 upshifted photons per laser pulse. In the years following this demonstration, laser Compton scattering was used as a diagnostic of electron beam quality in advanced accelerators. In its simplest configuration, laser light is incident head with the electron beam and the on axis, upshifted photons have an energy equal to $4\gamma^2 E_1$, where γ is the normalized energy of the electron and E_1 is the incident photon energy. By monitoring the spectrum of the upshifted photons, one may learn about the energy spread the electron beam. In the 1990's a renaissance in laser Compton scattering arose from the ultrafast materials community which used the process to produce short duration bursts of x-rays, typically of a few 100 fs to few ps in duration.[2,3] In order to reduce the duration of the resulting x-ray pulse in these systems often the laser was incident at right angles to the electron beam direction. While these sources produced short duration x-rays, they also produced relatively broadband x-rays (>10% $\Delta E/E$), were relatively inefficient and the up-scattered photon energy was only half of that from a head on collision. Fundamentally the efficiency of laser Compton scattering is limited by the small magnitude of the Thomson cross section (~0.6 barns) and the inability of electron beams to be focused to spots on par with minimum laser spot dimensions. In 1994, LLNL scientists recognized [4] that the Compton scattering brilliance should increase rapidly as a function of electron beam energy and beam quality. To first order this occurs because at higher electron beam energy it is possible to overcome electrostatic repulsion and focus the electron to smaller spot dimensions. Roughly, the electron spot dimension is proportional to its beam energy and thus the peak brilliance (photons/ sec/0.1%BW/mrad²/mm²) of the laser Compton source increases as function of electron beam energy somewhere between 2nd and 4th power. This rapid increase in peak brilliance is illustrated in Figure 1 and is in stark contrast to the trends of alternative sources, such as large scale synchrotrons. In the nuclear excitation region above 100 keV, the peak brilliance of 3rd generation synchrotrons decreases faster than exponentially. Above 2 MeV, the peak brilliance of a mono-energetic gamma-ray (MEGa-ray) source produced via laser-Compton scattering can exceed that of the largest synchrotrons by more than 15 orders of magnitude. It is important to note that for many nuclear applications and especially for those related to nuclear materials management, it is the bandwidth of the Compton source and not the pulse duration that is of foremost importance.

The optimization of laser-Compton scattering to produce narrowband MEGa-rays involves a different approach to machine design than that pursued for short duration x-ray sources. Schematically the differences are illustrated in Figure 2. The bandwidth of the Compton source is driven by three effects; the energy spread of the electron bunch, the bandwidth of the laser photons and the spread due to the angle correlation in the interaction region. Bandwidth can be minimized with high quality (low emittance) electron beams, 10 ps or longer laser pulses and near collimated laser-electron interaction geometries. Fractional bandwidths of $10^{-3} \Delta E/E$ or ~ 2

orders of magnitude less than that demonstrated from short-duration, laser-Compton x-ray sources are possible with careful design.



Figure 1. Peak brilliance of a laser-Compton MEGa-ray source relative to that of a state-of-the-art 3rd generation synchrotron



Figure 2. Schematic of laser-Compton optimization for different applications. Left panel: femtosecond x-ray configuration. Middle panel: high efficiency x-ray configuration. Right panel: narrowband MEGa-ray configuration.

Nuclear Photonics:

MEGa-ray sources based on laser Compton scattering can enable "Nuclear Photonics", that is the photon-based manipulation and study of the nucleus. In particular MEGa-ray sources are well suited to excitation of nuclear resonance fluorescence (NRF). NRF is a unique signature of the isotope as opposed to element. While NRF transitions at room temperature are very narrow (10⁻⁵ to 10⁻⁶ Δ E/E), selective excitation is possible with an optimized, 10⁻³ Δ E/E bandwidth MEGa-ray source. Furthermore, NRF cross sections of interest are large compared to background and often occur within the max-transparency window for most materials (see Figure 3).



Figure 3. Scatter plot of relative NRF cross sections as a function of mass number and photon energy. Dot size is proportional to cross section magnitude. Red dots are larger than the atomic background at the particular energy.

MEGa-ray excitation of NRF can be used in "reflection" or in "transmission" to determine the presence or absence of a particular isotope. In reflection, one illuminates the object in question and looks for the characteristic NRF relaxation radiation which is emitted into 4π . In transmission one looks for the absence of resonant photons in the transmitted beam. Besides being intrinsically less susceptible to clandestine attempts to obscure signals, transmission based systems can also provide quantitative assay and high resolution spatial information regarding the isotopic content of the object. In transmission the primary issue is low angle Compton scattering which can create new photons at the resonance energy and degrade measurement accuracy. This problem is alleviated for sufficiently narrowband and collimated MEGa-ray sources. [5]

In order to illustrate the assay and imaging potential of MEGa-ray sources, we have added selected NRF cross sections to an existing LLNL Monte Carlo, COG [6] and used this for a series of isotopic-specific material detection, assay and imaging simulations.

In one example, we consider a test object consisting of 2.2 cm diameter tungsten spherical shell in which a LiH test object is placed (see Figure 4). The test object has had material removed to provide an imaging challenge.



Figure 4. Test object for imaging simulation. Left is front view of the 2.2 cm W sphere containing LiH. The dark bars are "infinite" absorbing numerical resolution grid. Right is the 3D rendering of the test object .

Two images are constructed using the LLNL COG code; one simulating illumination by a 9 MeV Bremsstrahlung source and one simulating illumination by a MEGa-ray source tuned to the 478 keV NRF transition of ⁷Li. The bandwidth, beam source size and beam divergence of the MEGa-ray source are obtained from other LLNL laser-Compton simulation codes that have been benchmarked as part of previous LLNL laser-Compton source projects. The Bremsstrahlung source characteristics are taken from standard models. High flux MEGa-ray detection is assumed to be accomplished using LLNL's novel, Dual Isotope Notch Observation (DINO) detector configuration [6]. As expected the Bremsstrahlung image, which is basically a map of the electron density seen by the beam, provides no spatial information, i.e. the 3 electrons around Li provide negligible contrast relative to the 74 electrons per atom of tungsten. On the other hand, contrast in the MEGa-ray image is provided by the strength of the ⁷Li NRF transition. The MEGa-ray image reveals not only the structure of the material contained within the sphere but also illustrates the significantly higher spatial resolution possible with a MEGa-ray source.



Figure 5. Monte Carlo simulation of MEGa-ray imaging capability of W/Li test object. Left is a 9 MeV endpoint Bremsstrahlung image and right is a 478 keV (⁷Li NRF resonance) MEGa-ray image.

In a second example, we consider a standard nuclear fuel rod containing isotopic defects which including variations in density and enrichment (see Figure 6).



Figure 6. UO₂ fuel rod model.

As illustrated in Figure 7, the Bremsstrahlung image can identify the density defects but misses the enrichment problems while image obtained with ²³⁵U-resonant, 1733 keV MEGa-rays is able to identify the location and magnitude of the enrichment variations. Separate analysis [6] suggests that MEGa-ray systems currently being constructed will be able to assay nuclear fuel assemblies with better than 100 ppm accuracy per isotope of interest.





LLNL MEGa-ray Sources:

In 2008, LLNL produced MEGa-ray output from a machine called T-REX (Thomson-Radiated Extreme X-rays). Shown in Figure 8, T-REX utilized an existing 120 MeV, 40 year old, S-band

linear accelerator, a state-of-the-art, high brightness photo-gun and modern solid state laser technology to produce tunable photon beams from 0.1 MeV to 0.9 MeV. T-REX was used in transmission to validate the ability of a MEGa-ray system to excite NRF transitions and to utilize these signals to detect the presence of low density material (⁷Li) behind high density (Pb and Al) shielding. [7].



Figure 8. LLNL's T-REX MEGa-ray Source. Foreground is the photo-gun of the 120 MeV accelerator.

The output from the T-REX source was also used to validate the LLNL-developed, laser-Compton performance codes. Other efforts at Duke University in the US [8] and Japan [9] have also created similar MEGa-ray output from Compton scattering and used this light for NRFbased experiments.

While useful for proof-of-principle experiments, the output spectral flux from T-REX and other existing Compton-based gamma-ray sources [8,9] has been of the order 10's of photons/sec/eV. The bandwidth of these machines has also been relatively broad, of order 10% $\Delta E/E$. For many applications a much higher spectral flux (of order 1,000,000 photons/sec/eV) and narrower bandwidth (or order 0.1% $\Delta E/E$) are desired if not critical. Furthermore, smaller machines and/ or compact machines that can be transported to remote locations are often desired. To this end, LLNL has undertaken the development of a new class of high-flux, ultra-narrowband, compact

MEGa-ray light sources based on x-band accelerator technology, high-average-power, diodepumped lasers and highly-optimized, laser-electron interaction geometries.

X-band RF frequency is nominally 4x higher than S-band (12 GHz vs 3 GHz). This results in a significant reduction in size of the RF power supplies which to first order are proportional in volume to the cube of the RF wavelength. At x-band it has also been shown that it is possible to create significantly higher acceleration gradients without electrical breakdown. More than a decade of R&D at the SLAC National Accelerator Laboratory [10] has now resulted in x-band accelerator structures capable of reliable operation at gradients approaching 100 MV/m. For comparison, the T-REX S-band accelerator operated at an effective acceleration of \sim 10 MeV/m.

In partnership with the SLAC National Accelerator Laboratory, LLNL is now designing and constructing hardware for a 350 MeV, high brightness x-band accelerator designed specifically for MEGa-ray applications. [11] This accelerator will be combined with a state-of-the-art, 120-Hz, 1-Joule laser system to create a new MEGa-ray source capable of producing 10⁶ photons/ sec/eV with 0.1% bandwidth and tunable from 0.1 MeV to 4 MeV. The new MEGa-ray machine is the centerpiece of the new Nuclear Photonics Facility (NPF) at LLNL (see Figure 9).



Figure 9. LLNL's Building 391 Nuclear Photonics Facility. Currently under construction.

The NPF is dedicated not only to the development of NRF-based, isotope-specific material detection, assay and imaging technologies but also to exploration of fundamental nuclear physics with MEGa-ray sources.

In support of the development of high-flux, MEGa-ray sources, the US Department of Homeland Security through its Domestic Nuclear Detection Organization has sponsored the construction of an independent, high-brightness x-band accelerator test stand at LLNL. This machine, shown in the bottom of Figure 9, enables the rapid evaluation of next-generation, high-gradient, x-band accelerator structures and high-brightness photoguns, exploration of multi-bunch, x-band accelerator operation and development of techniques for the reduction of dark current-induced, gamma-ray noise.

While the LLNL Nuclear Photonics Facility's MEGa-ray source will be the first of its kind in the world and significant leap in MEGa-ray capability relative to existing machines, there are now similar scale MEGa-ray sources planned and facilities on the drawing boards both in Europe and in Japan. In particular in Europe the 850M euro Extreme Light Infrastructure project [12] plans to construct by 2016 in Romania a dedicated nuclear physics facility that will house a 600 MeV accelerator and MEGa-ray source with similar output characteristics to the LLNL machine [13].

Conclusions:

Next generation MEGa-ray sources will enable isotope-specific capabilities to rapidly and precisely detect, assay and image nuclear materials in a non-destructive and non-activating manner. MEGa-ray sources being constructed today are based on high-gradient compact accelerator and efficient, diode-pumped laser technologies that are geared toward eventual creation of mobile, re-locatable sources. These technologies have the potential to significantly alter the methods and procedures by which all nuclear materials are managed.

Acknowledgements

This work was performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

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