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Review

Application of Laser Compton Scattered gamma-ray beams to nondestructive detection and assay of nuclear material

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Abstract. Generation of energy-tunable gamma-rays via Laser Compton Scattering is of great interest for scientific studies and applications of "MeV" photons which interact with nuclei. One of the promising applications of such energy-tunable gamma-rays is the nondestructive detection and assay of nuclides which are necessary for nuclear security and safeguards. We are developing technologies relevant to gamma-ray nondestructive detection and assay, which include a high-brightness gamma-ray source based on modern laser and accelerator technologies, and gamma-ray measurement methods optimized for highly radioactive samples.

1 Introduction

Laser Compton Scattered (LCS) gamma-ray sources have been used for scientific and industrial applications for many years [1,2]. Recent progress of laser and electron accelerator technologies provides a path to improve the flux, brightness and energy purity of gamma-ray beams from LCS sources [3–5]. In the present paper, we summarize our research and development status of a LCS gamma-ray source and its application to nondestructive detection and assay of nuclear material such as uranium and plutonium for purposes of nuclear security and safeguards.

Figure 1 shows a schematic view of gamma-ray beam generation by LCS, where the gamma-ray energy E_γ, a function of electron energy E_e, laser photon energy E_L, electron speed in units of light speed $\beta = v/c$, and collision geometry, is given by

$$E_\gamma = \frac{E_L(1-\beta \cos \theta_1)}{1-\beta \cos \theta + (E_L/E_e)(1-\cos \theta_2)} \,. \tag{1}$$

We can produce a gamma-ray beam of arbitrary energy by changing the electron energy, laser wavelength or collision angle. Furthermore, the energy width of the

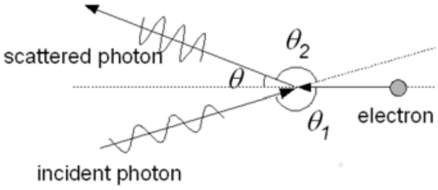

Fig. 1. Principle of gamma-ray beam generation via laser Compton scattering.

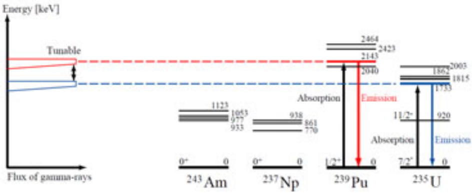

Fig. 2. Principle of nondestructive assay using nuclear resonance fluorescence with mono-energetic gamma-ray beams.

gamma-ray beam can be made narrower by putting a small diameter collimator in the path of the beam to restrict the scattering angle. The LCS gamma-ray beam is distinct from other conventional gamma-ray sources in its energy tunability, narrow energy width and small divergence.

Utilizing this LCS gamma-ray beam in combination with nuclear resonant fluorescence (NRF), we can make nondestructive measurement of arbitrary nuclides. The principle of LCS-NRF is shown in Fig. 2. When a nucleus is irradiated by a gamma-ray and the energy of the gamma-ray is identical to the transition energy from the ground state of the nucleus, the incident gamma-ray is absorbed by the nucleus, and subsequently the nucleus de-excites by gamma-ray emission. Since the energies of the states excited by NRF are inherent in the atomic number and mass of the nucleus of interest, detection and assay of nuclides are possible by a NRF measurement [6,7].

A nondestructive measurement based on LCS-NRF has the following advantages: we can measure both stable and unstable nuclides, the measurement is independent of the shape and chemical state of the material, and the ability of gamma-ray beams to penetrate a sample enable one to detect or assay specific nuclides even inside a sample.

Such nondestructive measurement can be applied to nuclear security and safeguards. Nondestructive inspection for screening special nuclear materials (SNM) is of growing importance in view of nuclear security concerns. One of the aims of nuclear security is to prevent the detonation by terrorists of a yield-producing nuclear bomb containing fissile material by detecting the material in transit at the port of entry. These materials, such as ^{235}U or ^{239}Pu with the weight of several kilograms, may be hidden in a radiation-shielded box and brought into a country using cargo containers. Finding a highly radioactive object hidden in a cargo container is possible with a conventional radiation detector. However, some kinds of nuclear material, ^{235}U for example, cannot be detected by self-radiation. LCS-NRF is a promising method for cargo inspection systems because of its selectivity and high penetration [8,9].

Nondestructive assay of fissionable nuclei is a key technology in nuclear material management which concerns both nuclear security and safeguards. In the next generation safeguards initiative (NGSI) program of the United States Department of Energy (DOE), nondestructive assay of plutonium in spent nuclear fuel is the top priority in technology development [10]. LCS-NRF is a possible solution for satisfying this demand. We evaluated the performance of LCS-NRF for the measurement of

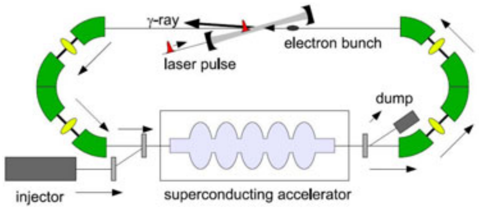

Fig. 3. A schematic view of Compton gamma-ray source utilizing an energy-recovery linac (ERL).

plutonium in spent fuel and found that the ^{239}Pu in spent fuel can be measured with a small statistical error, 2–3% during 4000 s measurement time, if we can utilize the high-flux gamma-ray source described in the following section [11,12].

2 Design of high-flux and high-brightness gamma-ray sources

For the generation of high-flux and high-brightness gamma-ray beams via laser Compton scattering, we need to increase the collision density of the laser and electron beams. Therefore, a small emittance and high-current electron beam and a high-power laser are necessary. Since average flux is essential rather than peak flux in the application of gamma-rays to the nondestructive measurement of nuclides, the electron beam current and the laser power should be evaluated in a sense of average values.

An energy-recovery linac (ERL) is the optimum apparatus to accelerate electron beams of small emittance and high-average current [13]. The principle of the ERL is as follows: an electron beam from an injector is accelerated by a time-varying radiofrequency (RF) field stored in a superconducting linear accelerator and subsequently transported to a recirculation loop. After the recirculation, the electron beam is injected again to the superconducting accelerator with a deceleration RF phase. The recirculated electrons are decelerated and feed back the energy to the superconducting RF cavity. This recycled RF energy is again used to accelerate subsequent electrons. The ERL is thus composed of an injector, a superconducting linac and a recirculation loop. Figure 3 shows a schematic view of the LCS gamma-ray source based on an ERL, where a laser cavity is installed in the recirculation loop for laser Compton scattering.

In a LCS gamma-ray source, because the cross section of the Compton scattering is not large, only a small portion of the electrons and photons collide with each other to generate a gamma-ray beam. Recycling the electrons and photons is, therefore, important to realizing high-flux and high-brightness gamma-ray sources. The ERL technology enables one to recycle an electron beam as described above. Recycling of laser photons, on the other hand, is achieved by a laser enhancement cavity. The laser enhancement cavity is a high-finesse Fabry-Perot optical cavity, which stores optical pulses injected from an external mode-locked laser. The laser power stored in the cavity depends on the injected laser power, mirror reflectivity and accuracy of mirror position control. Recently, an intra-cavity power of over 400 kW was demonstrated with a mode-locked laser of 450 W and an optical cavity optimized for thermal effects [14].

Utilizing a laser enhancement cavity for LCS gamma-ray sources, we can make laser photons in the cavity interact with electrons many times to generate gamma-rays. For future applications to the nondestructive measurement of plutonium in spent nuclear fuel, we have proposed a conceptual design of a gamma-ray source to produce a gamma-ray beam at a flux of 1×10^{13} ph/s. The gamma-ray source consists of a

350-MeV, 13-mA ERL and a laser enhancement cavity with a 700 kW intra cavity power [15].

3 R&D status for LCS-gamma ray

In order to realize a high-flux and high-brightness gamma-ray source based on the ERL, we are developing key technologies which include the generation and acceleration of small-emittance and high-average current electron beams in an ERL, storage of high-power laser pulses in a laser enhancement cavity, collision of electrons and laser photons at a small spot size, stabilization of electron and laser beams, and so on. Following is the development status of these key technologies.

3.1 Electron gun

An electron gun optimized for the ERL has been designed and fabricated at JAEA and consists of a DC electron gun equipped with a semiconductor photocathode to generate small-emittance electron beams at high-average current. We have demonstrated the operation of the electron gun at the world's highest voltage, 500 kV, and a high-average current up to 10 mA [16]. In high-voltage DC photoemission guns, the operational voltage has been restricted to 350 kV or lower owing to the field emission problem, which causes electrical breakdown or punch-through on the ceramic insulator surface. In the JAEA gun, we have employed a segmented insulator with rings to keep the insulator safe from the field emission generated from a central stem electrode to solve the field emission problem [17].

After the successful demonstration of 500-keV beam generation, the gun was shipped to the High Energy Accelerator Research Organization, KEK, and installed at the Compact ERL (cERL), a test accelerator at KEK. We started beam operation of cERL from April, 2013. Now, the gun provides an electron beam for daily operation of cERL.

3.2 Superconducting accelerators

A superconducting accelerator (SCA) for high-average current electron beams is another key component for the ERL. We have a collaboration with KEK for the SCA development. So far, two types of superconducting accelerators for cERL have been developed at KEK, one for the injector and the other for the main linac; both of them are operated at 1.3 GHz RF.

The injector SCA is a 2-cell, 3-cavity type to capture the 500-keV electron beam from the gun and accelerate it to $\sim$5 MeV. The amplitude and phase of the RF stored in the cavities are adjustable individually for the best acceleration of the beam [18]. Commissioning of the injector SCA started from May, 2013. We have confirmed beam acceleration to 5.7 MeV at the exit of the injector SCA [19].

The main linac SCA, a 9-cell, 2-cavity type, was assembled and installed at cERL. A high-power test was conducted to demonstrate an accelerating voltage of 14.2 MV and 13.5 MV in each of the two cavities [18]. Acceleration tests of an electron beam were started from December 2013.

3.3 High-power laser

A high-power mode-locked laser for the laser Compton scattering is under development at Kansai Photon Science Institute, JAEA [21]. The laser consists of a mode-locked oscillator and 4-stages of amplifiers; all of them utilize Yb-doped fibers as laser

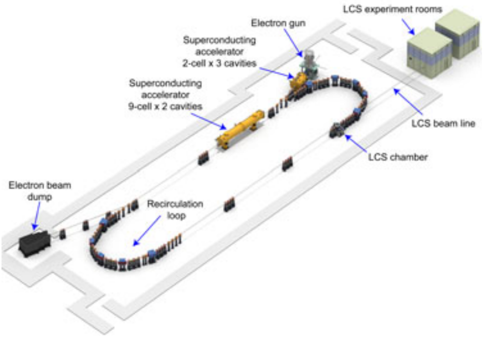

Fig. 4. A schematic view of Compact ERL.

gain media. Since the LCS gamma-ray bandwidth is affected by laser bandwidth, we are developing the high-power laser with particular priority on narrow-bandwidth laser pulses. The laser system is, thus, equipped with two pulse stretchers and one compressor to avoid nonlinear spectral broadening during amplification. Bandpass filters are also inserted between the amplification stages. The oscillator and 3 stages of the amplifiers have been completed to produce laser pulses with the following properties: wavelength 1030 nm, average power 20 W, bandwidth 1.4 nm (FWHM) and repetition rate ∼80 MHz. The final amplification stage to 100 W of average power is under commissioning.

A laser enhancement cavity to generate linearly polarized gamma-ray beams is of benefit to improving the signal-to-noise ratio in a nuclear resonance fluorescence measurement, because gamma-ray emission from nuclear dipole transitions has an anisotropic angular distribution with respect to the polarization plane of the incident gamma-ray beam. An optical cavity with a three-mirror image inverter has been proposed for this purpose. This cavity enables one to control the polarization of the stored laser pulses in an arbitrary direction, horizontal or vertical, to produce linearly polarized gamma-ray beams [22].

4 Demonstration experiment planned at the Compact ERL

We plan to demonstrate high-flux LCS photon generation at the Compact ERL. We will install a laser enhancement cavity at the recirculation loop of cERL to generate a LCS beam which is then transported to an experimental room for evaluation. Figure 4 shows a floor layout of the LCS experiment at cERL.

The electron gun and superconducting accelerators have already been installed at cERL as described above. We completed the recirculation loop in November 2013 and started commissioning of cERL from December 2013. The LCS experiment is scheduled in March 2015.

Since the design energy of electron beams at cERL is 35 MeV, LCS photons will have an energy of 22 keV. However, we consider the experiment as a demonstration of a high-flux laser Compton light source for future application to nuclear material detection and assay. Increasing the LCS photon energy is achievable simply by adding more superconducting cavities for higher electron energy. We have reserved space for additional superconducting cavities and a second recirculation loop in cERL for a future upgrade to higher energies [20]. After the full upgrade of cERL, an electron beam of 245 MeV will be available and will produce gamma-ray beams of 1 MeV.

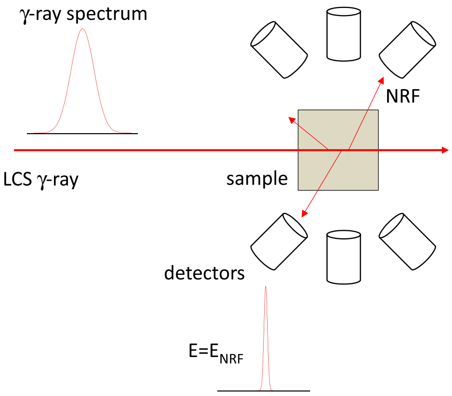

Fig. 5. Nondestructive measurement of a specific nuclide in a sample by collecting nuclear resonance fluorescence gamma-ray scattered directly from the sample.

5 Nondestructive measurement of nuclear material

We can realize isotope-specific nondestructive detection and assay systems based on LCS-NRF in several ways: resonance scattering, resonance transmission, integral resonance transmission and photo-fission [27].

Figure 5 shows the resonance scattering method, where signals of nuclear resonance scattered gamma-ray are collected with high energy-resolution detectors such as high-purity germanium (HPGe) detectors. The incident gamma-ray beam has a narrow bandwidth and is tuned to a resonance energy in the isotope to be measured.

A series of demonstration experiments based on the scattering method have been conducted at the LCS gamma-ray facility at the National Institute of Advanced Industrial Science and Technology (AIST). These experiments include one-dimensional mapping and two-dimensional mapping of a specific nuclide hidden in an iron box [23,24], one-dimensional mapping of two nuclides [25], and simultaneous detection of two nuclides [26].

When the scattering method is applied to the measurement of plutonium in spent nuclear fuel, the performance of the measurement system, such as statistical uncertainties, is affected by radiation background from the spent fuel as well as beam-induced background due to coherent scattering such as Rayleigh, nuclear Thomson, and Delbruck scattering. We investigated these effects and found that the radiation background from the spent fuel can be significantly reduced by choosing a measurement energy at 3–5 MeV. The coherent scattering can be distinguished from the NRF signal, if we observe the NRF transition to the first excited state instead of the transition to the ground state. For actinide nuclides, the first excited state is about 10 to 50 keV above the ground state, thus the transitions to the first excited state is about 10 to 50 keV lower than that of the coherent scattering [12]. This type of measurement is only possible with an incident photon beam of narrow bandwidth, $\Delta E/E \sim 0.1\%$, which is available at the ERL-LCS.

Reduction of background in the nondestructive assay of radioactive material is also possible with the resonance transmission method as shown in Fig. 6. In this configuration, we measure resonant scattered gamma-rays from a witness plate, a reference material containing the isotope to be measured, to determine the absorption of resonant gamma-rays in a sample. The amount of the specific isotope in the sample is measured from the decrease of resonant scattering from the witness plate. Most of the radiation from the sample can be rejected with appropriate shielding walls between the sample and the witness plate. Mitigation of coherent scattering is available with a witness plate having a high concentration of the isotope to be measured.

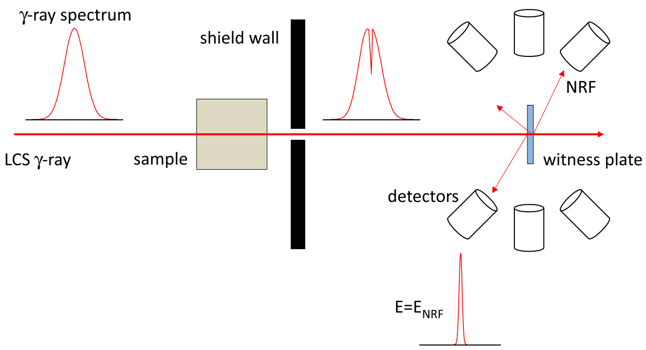

Fig. 6. Nondestructive measurement of a specific nuclide in a sample by collecting nuclear resonance fluorescence gamma-ray scattered from a witness plate placed downstream of the sample.

The witness plate method can be further improved in its efficacy by extending it with the integral resonance transmission (IRT) method [27]. The IRT method uses the integral strength of all resonances excited by the incident gamma-ray beam, including weaker states that are not resolvable with current HPGe detectors. The quasi-monoenergetic nature of the incident beam ensures that the integral signal is due primarily to the NRF signal and not background. The integral signal enables the use of cheaper, more efficient scintillator detectors and uses the additional absorption signature from the many unresolved states, both of which improve assay sensitivity. The first demonstration of the IRT method was successfully conducted at the laser Compton gamma-ray facility at Duke University, HIGS, where we used tantalum as a substitute for fissile material. Another IRT experiment with ^{239}Pu was also carried out at the same facility [28], and is being analyzed.

6 Summary

Energy-tunable gamma-ray beams generated by laser Compton scattering are becoming a common probe to investigate photo-nuclear reactions for scientific and industrial uses. Nondestructive detection and assay of nuclides utilizing nuclear resonance fluorescence, which is an isotope-specific photo reaction, is one of the promising applications of such energy-tunable gamma-ray beams. We have proposed a high-flux and high-brightness gamma-ray source based on an energy-recovery linac and a laser enhancement cavity to produce a gamma-ray beam of 10^{13}ph/s. Critical components for the gamma-ray source are under development at JAEA in collaboration with KEK to demonstrate laser Compton scattered photon generation at the Compact ERL. Gamma-ray measurement methods optimized for highly radioactive samples are also under development.

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