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Gamma-ray mirrors for direct measurement of spent nuclear fuel

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ABSTRACT

Direct measurement of the amount of Pu and U in spent nuclear fuel represents a challenge for the safeguards community. Ideally, the characteristic gamma-ray emission lines from different isotopes provide an observable suitable for this task. However, these lines are generally lost in the fierce flux of radiation emitted by the fuel. The rates are so high that detector dead times limit measurements to only very small solid angles of the fuel. Only through the use of carefully designed view ports and long dwell times are such measurements possible. Recent advances in multilayer grazing-incidence gamma-ray optics provide a possible means of overcoming this difficulty. With a proper optical and coating design, such optics can serve as a notch filter, passing only narrow regions of the overall spectrum to a fully shielded detector that does not view the spent fuel directly. We report on the design of a mirror system and a number of experimental measurements.

1. Introduction and motivation

The nondestructive assay (NDA) of spent fuel is of high priority for the international safeguards community. In the United States, the Department of Energy's (DOE's) National Nuclear Security Administration has started the Next Generation Safeguards Initiative (NGSI), and one of its pillars is technology development. The NGSI spent fuel NDA project has its primary goal "to enable direct and independent quantification of Pu mass in spent fuel with an uncertainty of more than 5%" [1].

Direct measurement of Pu and U gamma rays is a highly desirable NDA method that could be used in a reprocessing environment, since it does not require inference of the fissile content from measurements of gamma rays from other fission products. However, the high background and intense radiation from directly viewed spent fuel make direct measurements difficult to implement since the relatively low activities of U and Pu are masked by the high activity from fission products. To overcome this problem, we propose the use of grazing-incidence multilayer mirrors to selectively divert hard X-ray and soft gamma rays in the 90 – 420 keV energy band emitted by U and Pu into a high-purity germanium (HPGe) detector shielded from the line-of-sight radiation from spent fuel. This energy range encompasses the element-specific K-shell fluorescence emission lines from U and Pu, isotope-specific lines from ^{235}U and ^{239}Pu, and lines from other Pu isotopes of interest, including ^{238}Pu, ^{240}Pu, and ^{241}Pu. Table 1 lists the primary K-shell fluorescence lines and their relative intensities, while Table 2 lists the brightest nuclear emission lines and their specific activities for individual isotopes.

Robust detection of these lines could provide key information to improve the shipper–receiver difference or input accountability at the start of Pu reprocessing [4,5]. Previous work by several groups [5–8] has shown that it is possible to detect the K-shell fluorescence lines of U and Pu from spent fuel using Ge detectors. Our goal is to extend this spectroscopic technique to the isotope-specific lines of U and Pu by filtering out intense, unwanted out-of-band emissions from fission products that overwhelm the relatively weak lines.

In Section 2, we discuss the basic properties of the reflective multilayer mirrors and recent developments in their fabrication that we have leveraged to produce mirrors at the DOE's Lawrence Livermore National Laboratory (LLNL) appropriate for nuclear safeguards. We then summarize the results of an extensive measurement campaign used to validate the performance of the multilayer elements.

In Section 3, we discuss a proof-of-concept experiment conducted at the DOE's Oak Ridge National Laboratory (ORNL) using sealed radioactive sources to show the efficacy of multilayer mirrors as a pass-band filter for reflecting specific emission lines into the HPGe detector. Finally, in Section 4 we briefly describe our plans for the future, which include using gamma-ray mirrors to measure spent nuclear fuel at the Irradiated Fuels Examination Laboratory (IFEL) at ORNL.

2. Reflective X-ray optics

2.1. Background

It was Compton [9] who first realized that X-rays incident on a surface at a shallow or grazing angle (on the order of a few degrees

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Table 1
Primary K-shell fluorescence lines of U and Pu[a].

Emission line (designation)		**Energy (keV)**		**Relative intensity (%)**	
Siegbahn	**IUPAC**	**U**	**Pu**	**U**	**Pu**
$K\alpha_2$	$K\text{-}L_2$	94.67	99.55	61.9	62.5
$K\alpha_1$	$K\text{-}L_3$	98.44	103.76	100	100
$K\beta_3$	$K\text{-}M_2$	110.41	116.27	11.6	11.7
Kβ1	$K\text{-}M_3$	111.30	117.26	22.0	22.2
$K\beta_2$	$K\text{-}N_{2-5}$	114.50	120.60	12.3	12.5

Table 2
Brightest isotopic emission lines from U and Pu in the 90–420 keV band[a].

Isotope	Energy (keV)	Specific activity (γ/g s)	Isotope	Energy (keV)	Specific activity (γ/g s)
^{235}U	143.8	8.76×10^3	^{239}Pu	98.44	1.55×10^5
^{235}U	166.3	4.06×10^3	^{239}Pu	111.3	3.56×10^4
^{235}U	185.7	4.57×10^4	^{239}Pu	129.3	1.44×10^5
^{235}U	205.3	4.01×10^3	^{239}Pu	146.1	2.73×10^4
^{238}Pu	99.85	4.49×10^7	^{239}Pu	375.0	3.60×10^4
^{238}Pu	152.7	5.90×10^6	^{239}Pu	413.7	3.42×10^4
^{241}Pu	103.7	2.26×10^6	^{240}Pu	160.3	3.37×10^4
^{241}Pu	148.6	7.15×10^6	^{240}Pu	104.2	5.85×10^5
^{241}Pu	208.0	2.04×10^7			

[a] Data on Pu isotopes obtained from [2]; data on ^{235}U isotopes obtained from [3].

or less) can undergo total external reflection from the surface of a material because the index of refraction n is less than unity at X-ray wavelengths. The index of refraction is commonly written as $n=1-\delta-i\beta$, where the real part, δ is the refractive index decrement and the imaginary part, β is the absorption index and is proportional to the atomic photoabsorption cross-section. The transition between total external reflection and absorption occurs at the critical angle θ_c, where $\theta_c=\sqrt{2\delta}$, and depends strongly on the material and the photon energy E.

Reflective X-ray optics have been developed primarily for two scientific applications: X-ray light sources (such as synchrotrons and free electron lasers) and astronomy and astrophysics. In X-ray synchrotrons or free electron lasers, the optics comprise elements in a beamline and serve to condition, spectrally filter, or focus the X-rays as they propagate to an end-station for use in a scientific investigation. Examples of this include the primary mirror systems used at the Linac Coherent Light Source (LCLS) and the X-ray free electron laser operating at the DOE's SLAC National Accelerator Laboratory [10].

Focusing X-ray optics allow true imaging, and the first practical design was proposed by Kirkpatrick and Baez in 1948 [11]. Now called Kirkpatrick–Baez or KB optics, these optical designs require two mirror elements—the first focusing in one dimension, the second focusing perpendicular to the first—and are widely used at light sources or in other applications where the source flux is high. In 1952, Wolter [12] proposed a new focusing design requiring pairs of mirrors built from surfaces of revolution of conic sections (e.g., hyperboloids and ellipsoids) that function together to achieve excellent focusing properties across a wide field of view (FOV). Fig. 1 illustrates the basic imaging concepts of KB and Wolter X-ray optics.

Although Wolter had envisioned using such optics for biological studies, the first successful implementation of his designs was employed for an X-ray telescope in the late 1960s [13]. Since then, the high-energy astrophysics community has spent billions of dollars building and refining reflective X-ray optics for many satellite-based observatories. An important realization of the community was that sets of co-focal mirrors could be placed inside one another, resulting in a nested optical system to increase the collection efficiency by orders of magnitude above that of a single pair of mirrors. Over the last two decades, space-based X-ray satellites such as *ASCA*, *XMM-Newton*, and *NuSTAR* have used nested telescopes consisting of several tens to more than 100 nested layers to achieve large collecting areas [14–16].

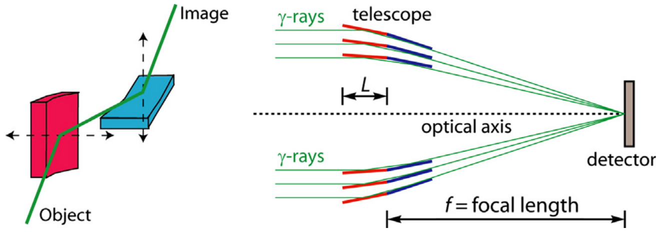

Fig. 1. (Left) Schematic of Kirkpatrick–Baez (KB) optic. The photons are first focused in the horizontal direction by the red mirror, and then focused in the vertical direction by the blue mirror. (Right) Schematic cross-section through a Wolter optic. Three nested shells are shown. The photons are first reflected from parabolic mirrors (red), and then hyperbolic mirrors (blue). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

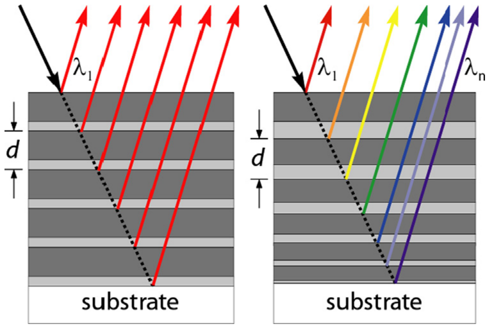

Fig. 2. Illustration of multilayer designs. (Left) In a constant-*d* spacing design, the thickness *d* of the alternating layers of high- and low-density materials is the same throughout the coating. This maximizes reflectivity for a unique combination of wavelength ($\lambda\propto 1/E$) and graze angle θ. (Right) In a graded-*d* spacing (depth-graded) design, the thickness *d* of the bi-layer is varied, allowing Bragg's law to be satisfied for a range of energies and angles.

Even with practical optics designs, until recently, the useful upper limit of the energy of such optics was limited to $E\approx 10$ keV because of the extremely small graze angles required for use at higher energies. To extend the energy band, several groups proposed the use of multilayer coatings in conjunction with Wolter-like optical designs [17,18]. Just as dielectric multilayers can affect the transmission or reflection of visible, alternating layers of material with a differing n can affect the reflection of X-rays. Coatings made of alternating layers of low- and high-density materials cause the radiation to constructively interfere and obey Bragg's law: $m\lambda=2d\sin\theta$, where m is an integer representing the order of the reflection, λ is the wavelength of the photon, d is the period of the multilayer, and θ is the graze angle. Fig. 2 illustrates two broad classes of multilayer recipes: constant-*d* spacing coatings, designed to have the same period throughout the coating to work for a specific angle and photon energy ($E\propto 1/\lambda$), and graded-*d* spacing (depth-graded) coatings, designed to have a range of periods throughout the coating and to work over a range of energies and angles. Whatever the design, fabricating coatings with small period thickness d makes it possible to work at larger graze angles, for a given energy, or at higher energies, for a given angle.

The highest energy reflective X-ray multilayer optics currently used for an operational application are the two telescopes flying onboard *NuSTAR*, a NASA satellite launched in June 2012 [16]. Each