Nuclear Instruments and Methods in Physics Research B 285 (2012) 72–85

Contents lists available at SciVerse ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Searching for illicit materials using nuclear resonance fluorescence stimulated by narrow-band photon sources

M.S. Johnson [a,b,*], C.A. Hagmann [a], J.M. Hall [a], D.P. McNabb [a], J.H. Kelley [c], C. Huibregtse [c], E. Kwan [d,1], G. Rusev [d], A.P. Tonchev [d,1]

[a] Lawrence Livermore National Laboratory, Livermore, CA 94550, United States
[b] San Jose State University, San Jose, CA 95192, United States
[c] North Carolina State University, Raleigh, NC 27695, United States
[d] Duke University, Durham, NC 27708, United States

ARTICLE INFO

Article history:
Received 29 January 2011
Received in revised form 11 May 2012
Available online 5 June 2012

Keywords:
Nuclear resonance fluorescence
Gamma-ray detection methods
Gamma-rays
NRF

ABSTRACT

We report the results of an experimental study of the sensitivity of two distinct classes of systems that exploit nuclear resonance fluorescence (NRF) to search for illicit materials in containers. One class of systems is based on the direct detection of NRF photons emitted from isotopes of interest. The other class infers the presence of a particular isotope by observing the preferential attenuation of resonant photons in the incident beam. We developed a detailed analytical model for both approaches. We performed experiments to test the model using depleted uranium as a surrogate for illicit material and used tungsten as a random choice for shielding. We performed the experiments at Duke University's High Intensity Gamma Source (HIGS). Using the methodology we detail in this paper one can use this model to estimate the performance of potential inspection systems in certifying containers as free of illicit materials and for detecting the presence of those same materials.

1. Introduction

Systems to interdict the transport of contraband such as special nuclear materials (SNM) are currently being developed to improve the throughput of choke point screening procedures as part of a multi-layered approach to enhancing national and international security [1,2]. There are several challenges that need to be met in order to make these technologies broadly useful. Systems need to be flexible enough to search for a broad range of illicit materials, reliable enough to limit false alarms to manageable levels and be capable of scanning large containers and vehicles in time scales of seconds to minutes, all while maintaining reasonably safe radiological dose levels. Nuclear resonance fluorescence (NRF) has been proposed as a physical process that might prove useful in this regard, specifically for the detection of fissile materials [3,4]. NRF (analogous to normal atomic fluorescence) is a process in which a photon at a resonant frequency for a given nucleus interacts with and excites the nucleus. The excited nucleus then decays to its ground state via either direct or multiple transitions. For actinides, these excitations are believed to be either magnetic dipole oscillations such as scissors mode (*cf.* [5]), or electric dipole oscillations assumed to be from octupole-quadrupole vibrations or some other mode. NRF frequencies vary with isotope, thereby providing unique signatures for many materials, and the natural line-widths of the resonances are quite narrow (~few eV) compared with nuclear level spacings (~100 keV), so accidental interferences between different materials are unlikely, especially when high-resolution detectors are being used. Morover, empirical evidence indicates that overlapping is unlikely. Since NRF states are typically stimulated with MeV-scale photons and the integrated cross sections tend to be quite large (~1–100 eV barns) compared with normal atomic attenuation cross sections, it should be possible to penetrate ~100–300 g/cm^2 of typical benign material while maintaining a high sensitivity to illicit materials. With narrow-band gamma-ray sources such as those based on Compton backscattering, the radiological dose delivered to an object under inspection could be substantially smaller than that associated with conventional Bremsstrahlung sources and signal-to-noise levels in detectors could also be improved [6].

There are many potential ways to interrogate cargo and vehicles for contraband such as explosives and special nuclear materials (*cf.* [7]). Systems that combine multiple approaches will likely be used to cover a broad range of possible scenarios, but the natural questions that arise are how to best optimize these systems and how to

* Corresponding author at: San Jose State University, San Jose, CA 95192, United States.
E-mail address: johnson329@llnl.gov (M.S. Johnson).
[1] Present address: Lawrence Livermore National Laboratory, Livermore, CA 94550, United States.

0168-583X/$ - see front matter © 2012 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.nimb.2012.05.029

compare the effectiveness of different detection schemes. These can be complex issues to address, particularly given the expectation that actual inspection systems in the field will detect nothing (*i.e.* obtain null results) in the vast majority of cases.

The particular case that we will address here is that of quasi-monoenergetic photon sources stimulating NRF. Data for a variety of scenarios will be used to benchmark an analytical model of performance for two types of systems over a broad range of different scenarios. One class of systems is based on the direct detection of NRF photons emitted from isotopes of interest. The other class infers the presence of a particular isotope by observing the preferential attenuation of resonant frequency photons in the incident beam. For each class of systems we will use the model to estimate the statistical accuracy of a decision metric for a broad set of hypothetical cargo scenarios and the time that it would take to decide on the disposition of the cargo (*i.e.* pass vs. alarm).

The paper begins by reviewing the basic detection protocols for both classes of NRF systems in the next section. Section 3 highlights our approach to describing the phase space of possible threats to be detected or excluded. Our approach to modeling system performance is described Section 4, followed by sections on the model validation experiments, results and our conclusions.

2. NRF detection protocols

Protocols for detecting threat materials via their NRF signatures using both reflection- and transmission-based NRF detection schemes have already been discussed in the literature [4]; however, we will review the fundamentals of a relatively simple, yet robust, set of "operational" protocols here for completeness.

2.1. Reflection

In reflection-based detection schemes, the high-energy photon spectrum emerging from the cargo is recorded by an array of detectors located upstream from the cargo and oriented at backward angles to the incident photon beam, where backgrounds from beam-related Compton scattering and other processes tend to be at lower energies than the NRF emission lines from materials of interest. Several millimeters of attenuating material are usually placed in front of these detectors in order to reduce the background count rates to acceptable levels. The rate of beam-induced NRF counts from the cargo is then compared to the rate of transmitted (unattenuated) photons recorded by a beam flux monitor located downstream from the cargo (although perhaps not intuitive, the *transmitted* photon rate is typically used here rather than the *incident* photon rate in order to avoid the possibility of inadvertently failing to detect a threat simply because the surrounding (nominally benign) cargo was too thick for the incident beam to penetrate).

The decision metric (r) for these schemes will be defined as

$$r = \frac{R_{NRF}}{R_T} = \frac{R_S - R_B}{R_T}, \tag{1}$$

where R_S, R_B and R_T are the total signal, background and transmitted count rates, respectively. In contrast to the conventional approach of using "Receiver-Operator Characteristic" (ROC) curves to define the probability of detecting a threat versus the probability of false alarm for *specific* threat scenarios, we will use key values of the decision metric, r, to calculate the "times to decide" (*i.e.* pass vs. alarm) for *arbitrary* cargos at fixed probabilities of false negative (PFN) and false positive (PFP).

The first of the key decision metric values required for the analysis will be r_0, the expected value of r for a completely benign cargo (no trace of threat material). For reflection-based detection schemes, r_0 will be zero since $R_{NRF} = 0$. The second key metric value will be r_2, the expected value of r for a threat cargo. The value of r_2 will depend on both the "minimum credible threat" (MCT) of interest *and* its shielding in this case; however, it will always be $>r_0$. The third key metric value will be r_D, a user-defined boundary between relatively "clean" and relatively "dirty" cargos (*i.e.* between benign cargos which contain relatively small vs. relatively large trace quantities of the threat material, respectively) that will allow us to avoid discontinuities in the "time to decide" estimates. For convenience, the value of r_D will be defined here to be midway between r_0 and r_2. Note that the value of r_D has no effect on the "time to pass" with a given PFN. Finally, r_2 and r_D will combine with the given values for PFN and PFP to define a fourth key metric value, r_1, which will serve as the effective (operational) decision boundary (pass vs. alarm). The relationship between these key r values is illustrated in Fig. 1, where we have used a Gaussian-like distribution to describe the measured decision metric, r_m, and its underlying statistical uncertainty and have indicated the range of r_m values which we will chose to define as "false negative" or "false positive" results.

By using a Gaussian distribution in Fig. 1, there will always be a non-zero integral that extends below r_0. We have exaggerated the distribution here so as to not confuse the reader and to be as precise as possible. In practice the distribution will become much narrower as the integration time increases, such that it will approach a delta-function limit. Therefore the integral below r_0 is negligible. The true distribution will not be a Gaussian but some truncated version that has a sharp cutoff at $r_0 \sim 0$. However, it has been shown by Feldman and Cousins (see Ref. [8]) that the confidence interval for a positively constrained Gaussian is similar to a non-constrained Gaussian, whose mean is sufficiently distinct from zero. This would be the case for typical scenarios implied by this paper and sufficient counting time. Therefore, without loss of generality we use a non-constrained Gaussian distribution to make our model.

If presented with an unknown cargo to inspect using a reflection-based detection scheme, we will decide on its disposition (pass vs. alarm) based on the final value of r_m relative to r_1. During the course of the scan, the statistical uncertainty in r_m will decrease (*i.e.* the distribution will become more narrow). Referring to Fig. 1, it is apparent that, if r_m eventually stabilizes at a value $<r_1$, then the criteria for passing even a somewhat "dirty" (but, by definition, still benign) cargo with a given PFN will be met more quickly than the criteria for sounding an alarm with a given PFP. Conversely, if r_m settles into a value $> r_1$ (indicating either an exceptionally "dirty" cargo or an actual threat), then the reverse will be true. Given this observation, the operational "time to decide" will be defined here as the minimum of the time required to pass the cargo with a given PFN and the time required to sound an alarm with a given PFP.

The time required to pass an unknown cargo will depend on the PFN value used. For the Gaussian-like distribution for r_m shown in Fig. 1, the "critical z value" associated with the upper-tail PFN region will be

$$z_{PFN} = \frac{r_2 - r_m}{\sigma_{r_m}(t)}, \tag{2}$$

where $r_m < r_2$ and, neglecting the relatively small uncertainty in R_T (which will always be much less than the uncertainties in either R_S or R_B),

$$\sigma_{r_m}(t) = \frac{\sqrt{(R_S + R_B)}}{R_T\sqrt{t}} = \frac{\sqrt{(r_m R_T + 2R_B)}}{R_T\sqrt{t}} = \sqrt{\frac{(r_m + 2R_B/R_T)}{R_T t}}. \tag{3}$$

In reflection-based detection schemes, the ratio R_B/R_T will depend on both the MCT of interest *and* its shielding (both intentional