DETECTING SPECIAL NUCLEAR MATERIALS AND EXPLOSIVES USING NUCLEAR RESONANCE FLUORESCENCE AND EFFECTIVE-Z TECHNOLOGY

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A comprehensive approach to the detection of weapons of mass destruction containing special nuclear material (SNM) includes the ability to rapidly and efficiently determine a region of interest in three dimensions where atomic number, Z, is high and subsequently identify and quantify the nuclear isotopes in that region of space. Nuclear Resonance Fluorescence (NRF) combined with effective Z determination (EZ-3D™) provides for rapid scanning of seagoing containers, trucks and other vehicles in short times while providing high probabilities of detection for SNM and other contraband with low false alarms. Finding SNM with high probability of detection is associated with finding regions of high Z using EZ-3D™. Using NRF false positive events are eliminated in order to not restrict the flow of commerce. EZ-3D™ algorithms have been developed that use the non-resonant spectrum of backscattered photons and yield a signal with very high contrast between materials of moderately different Z. NRF provides a signal that is unique for most nuclei with Z>2. Different NRF signatures enable, for example, the discrimination between ^{235}U from ^{238}U. The NRF and EZ-3D™ techniques provide three dimensional displays of the contents of a voxel; respectively the isotopic concentrations and mass, and the effective Z.

I. Introduction

Non-intrusive detection and identification of highly enriched ^{235}U (HEU), ^{239}Pu, explosives and other contraband in sea-going containers, trucks, air cargo and luggage continues to be crucial to the safety of the nation and the global economy. The detection and screening process must be highly effective and efficient, requiring a technology capable of high probability of detection and low false alarms at high throughput rates that can be implemented in a variety of scenarios. Direct elemental detection of explosives and contraband using nuclear inspection technologies would be a significant step towards solving the cargo inspection problem. Nuclear Resonance Fluorescence (NRF) is a technique in which each nuclear species resonantly absorbs and re-emits highly penetrating photons according to specific nuclear excitations.[1, 2] NRF provides a unique signature for each nuclear isotope and exists for most nuclei with Z > 2. Using a continuous distribution of photons, such as is available with electron bremsstrahlung, allows NRF to be a tool that encompasses the individual energetic structure of all nuclei simultaneously. NRF based inspections can be automated and their performance is not dependent on the threat shape and density as with conventional X-ray imaging technologies. NRF can be used to generate a three dimensional map of the isotopic content of a container. It can also be used in transmission using the absorption spectrum to generate a two-dimensional map of the isotopic contents of a container similar to conventional two-dimensional radiographs which only map the average density along a column of material.

NRF uses the photons in narrow regions of the electromagnetic spectrum, generally those within a few eV of a specific energy of a nuclear excitation. Even with several such excitations, the photons involved in the process of exciting a nuclear species only encompass a dozen or so eV of the electromagnetic spectrum. Very little of the bremsstrahlung spectrum is used. On the other hand, almost all the photons in the bremsstrahlung spectrum are used with the EZ-3D™ technology. This technology makes use of the many interactions that a photon can undergo in scattering from a material that culminate in photons produced at back angles. The back scattered photon spectrum carries the information about these processes and their Z-dependence. This is a generally continuous spectrum from which the effective-Z of the scattering material is determined with sharp contrast for modest resolution in Z.

The combination of EZ-3D™ and NRF present a formidable combination. The EZ-3D™ involves many photons and allows the rapid detection of regions of interest (ROI) with specific ranges of Z in highly shielded situations. For example, high Z would indicate SNM or heavy shielding. Low Z in anomalous situations could indicate explosives. Modest photon energy resolution is required compared to NRF and large arrays of economical detection counters such as NaI allow derivation of the signals that are used.

II. Overview of Nuclear Resonance Fluorescence Technology

The process of NRF corresponds to the excitation of a nuclear state by photons and having that state decay to the ground state or an excited state by the emission of a photon.[3] This process does not result in residual long-lived radioactivity. A schematic of the physical processes which are important to NRF are shown in Figure 1.

NRF cross sections typically have very large peak values that correspond to hundreds of barns for NRF

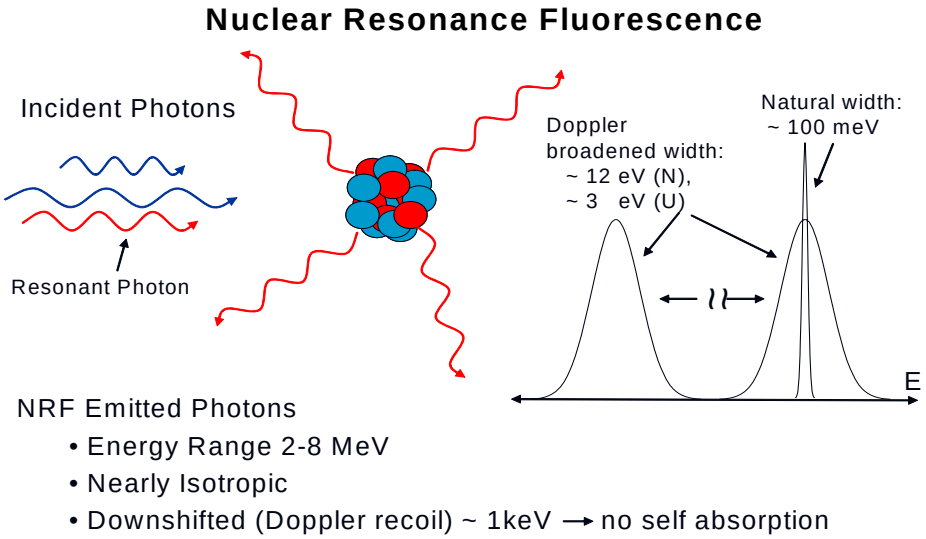

Figure 1: Overview of NRF Physical Processes

states with energies in the range of a few MeV. The states are broadened by the zero-point motion of the atom and thermal motion. For light nuclei, such as nitrogen and oxygen, the broadening can be approximately 20 eV resulting in reduced peak cross sections. However, NRF cross sections for useful states are still considerably larger than the usual electromagnetic processes: photoelectric, Compton and pair production in that energy region.

The elements carbon, nitrogen and oxygen have strong NRF states as shown in Figure 2. Oxygen has NRF states at 6.9 and 7.1 MeV, nitrogen at 7.029, 5.691, 4.915 and 2.313 MeV and carbon (^{12}C) at 4.43 MeV. Examples of NRF spectra for explosive simulants, a common nitrogenous material (melamine) and water are shown in Figure 3. These photon spectra were obtained using an 8.3 MeV electron beam and high-purity germanium detectors at 130 degree viewing angle from the incident beam.[4] The NRF states are clearly visible over the low background. The lines labeled as (SE) correspond to single escape peaks. These states enable automated extraction of the intensity of these lines which, along with the known strength of the NRF states, determine the mass of each element. The scanner would automatically and in real time reduce this information to a spatial map of elements and automatically alarm on regions which contain a threat.

An explosive would be labeled by its elemental composition. Peroxide explosives would have no nitrogen and they would display the approximately characteristic 3/2 ratio of carbon/oxygen. Explosives formed by using nitrogen fertilizer would have mostly nitrogen and oxygen in the characteristic 1/3 ratio of NO_3. Al and Mg powders used to enhance explosive power would also be detected. The mass and volume of the interrogated region would provide the density which can also be part of the characterization of an explosive.

For the detection and identification of materials it is useful to think of NRF in two modes. In one mode the scattered photons are detected allowing for a three dimensional map of all elements. The second mode makes use of the dominance of the NRF process in the energy region of the state and examines the transmitted photon spectrum for absorption lines. The two detection modes can be used simultaneously and they are displayed schematically in Figure 4 as they might be employed in a scanner searching for SNM, explosives and other contraband. The EZ-3D™ detectors could be part of the system examining the continuous portion of the scattered spectra at backward angles for rapid ROI determination.

Commercial high duty factor electron accelerators exist which can produce suitable beams energies (approximately 3 – 10 MeV) with sufficient intensity to enable scan times that are consistent with the requirements of cargo inspection. The bremsstrahlung beam is collimated and viewed by an array of collimated

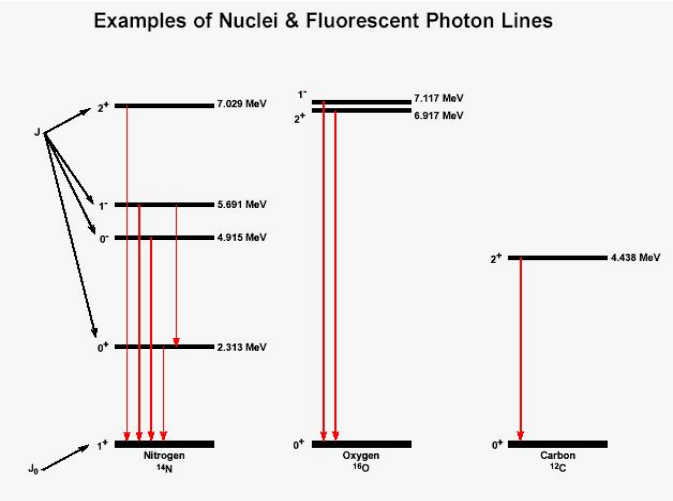

Figure 2: Nuclear Fluorescent Photon Lines

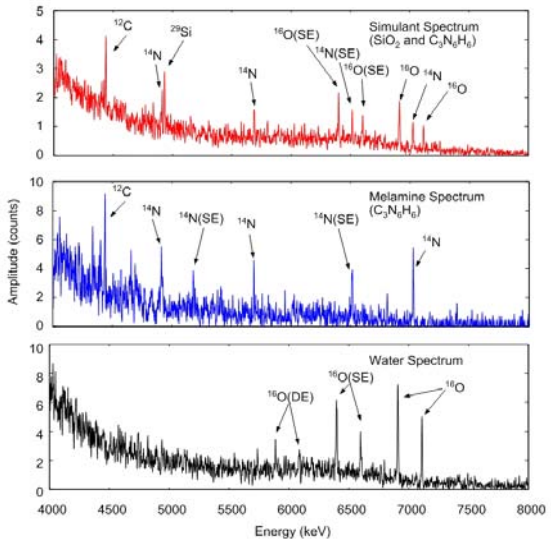

Figure 3: Water, Melamine and Simulant

detectors at back angles to the beam (3-D, NRF and EZ).

The intersection of the view of the detectors and the photon beam defines a voxel. Material in the voxel will scatter some of the photons in the beam via NRF in the direction of the detectors. The detectors for the 3-D NRF Imager are at back angles since there is no single electromagnetic process that produces a photon above 0.5 MeV at this angle and therefore the background for NRF states in the MeV range is minimized. The relative composition of almost all elements (Z>2) can be determined accurately from known integrated cross sections for the various NRF states, limited for the most part by statistical precision.

The 2-D NRF transmission imager is placed after the inspection volume. In Figure 4 there could be three reference scatterers, carbon, nitrogen and oxygen, the primary elemental constituents of explosive devices. If a specific material is present in the path of the photon beam, the spectrum is attenuated by standard non-resonant electromagnetic processes discussed earlier. Also, in the energy regions where the NRF states of the material exist, the attenuation will be dominated by the nuclear absorption and "holes" that are a few eV wide will be generated in the photon spectrum. Therefore, a decrease in the NRF signature for a reference scatterer (relative to non-resonant absorption) in the 2-D NRF Image is directly related to the amount of that reference scatterer along the beam path through the cargo.

Both the NRF 3-D and 2-D techniques have their advantages and special engineering challenges. The 3-D technique generates a three dimensional image and also simultaneously detects all elements, both of which are highly advantageous properties for a screening device.

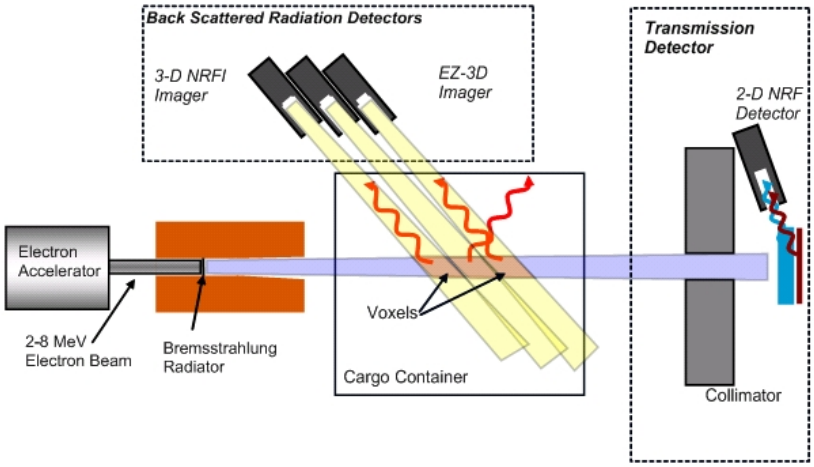

Figure 4: NRF and EZ-3D™ Scattering Showing 3D Voxels and 2-D NRF Transmission Detection

In order to obtain the highest sensitivity, detectors with high energy resolution are needed. The ability to detect all elements without change of hardware further allows changes in threat selection and threat algorithms with only changes in software. The detector of choice remains a semiconductor germanium detector (Ge) with approximately 0.1% energy resolution for MeV photons. However, these detectors need to be cooled and are also relatively expensive. The 2-D NRF transmission technique requires the selection of specific reference scatterers for the elements to be measured. However, in many practical situations the 2-D NRF transmission technique is more amenable to finer spatial resolution. This may be important for detecting certain threats configured as sheets. It is likely that an NRF-based cargo scanner would use both of these NRF detection modalities to optimize performance for explosives detection.

The detection of SNM requires knowledge of the NRF lines from ^{235}U and ^{239}Pu as well as common shielding materials such as lead. A portion of the spectrum of NRF lines from natural lead is shown in Figure 5.[5] Identification of lead as a shield is very clear with very strong lines above 4 MeV. In Figures 6 and 7 the NRF spectra from ^{235}U and ^{239}Pu are displayed, respectively.[6] The data for ^{235}U and ^{239}Pu were taken using a 2.1 and 2.8 MeV electron beam respectively, and approximately 200 g of HEU and 7 g of ^{239}Pu.[6, 7] Measurements on ^{235}U at higher energies are underway and also planned for ^{239}Pu. The presence of other materials (e.g. Mn, Cr, Al) is due to either the SNM container or normalizing materials. These SNM results have not been fully analyzed for cross section strengths and branching ratios and are still preliminary. They will be the subject for a further publication when these details are complete. However, several features are clear from these data. Lines exist that correspond to the decay of some of the excited states to the first rotational state built on the ground state rotational band. These are indicated on the figures and correspond to excitation energies of 46.2 keV (J=9/2) in ^{235}U and 7.86 keV (J=3/2) in ^{239}Pu. Also, the lines are sufficiently strong and unique that unambiguous identification of these isotopes is practical in a cargo scanning scenario.

The use of transmission technology is illustrated in Figure 8. A photon beam is incident on a "threat" simulated by melamine. Two detectors are shown, one viewing a carbon target and the other viewing a boron target. Other detectors could be used, in particular, one viewing a target of nitrogen and one viewing a target of oxygen. As discussed earlier, the photons resonant with the carbon energy at 4.43 MeV are resonantly absorbed in the melamine and the absorption of the photons is reflected in the strength of the NRF signal from the carbon reference target following the melamine. There is also electronic absorption in the melamine which is monitored by the boron target. Boron does not have resonant lines that overlap with any of the materials in the melamine. The ratio of counts for the resonant line in carbon with and without the melamine absorber compared to the ratio of counts for the resonant line in boron with

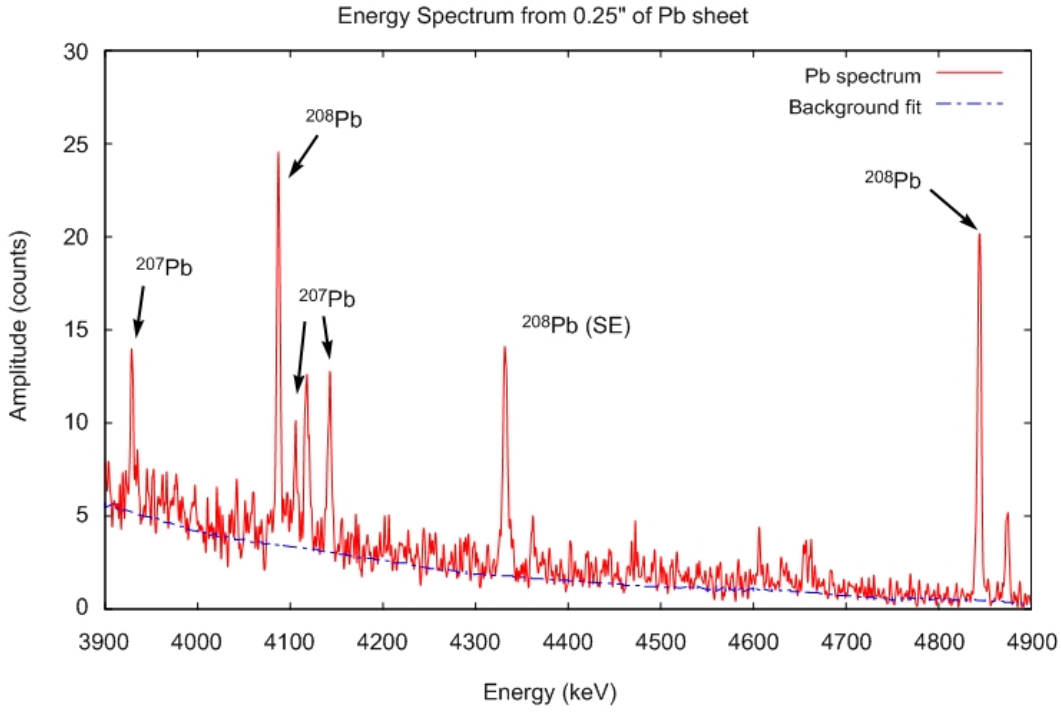

Figure 5: Natural Pb NRF Spectrum

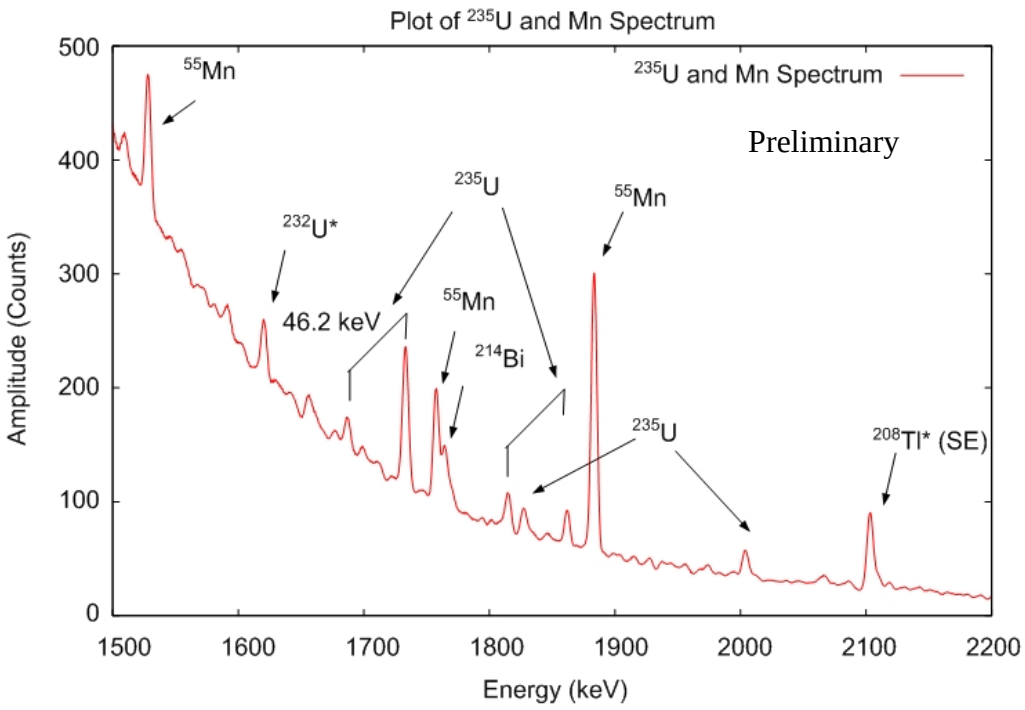

Figure 6: ^{235}U NRF Spectrum with Mn Normalization (preliminary)

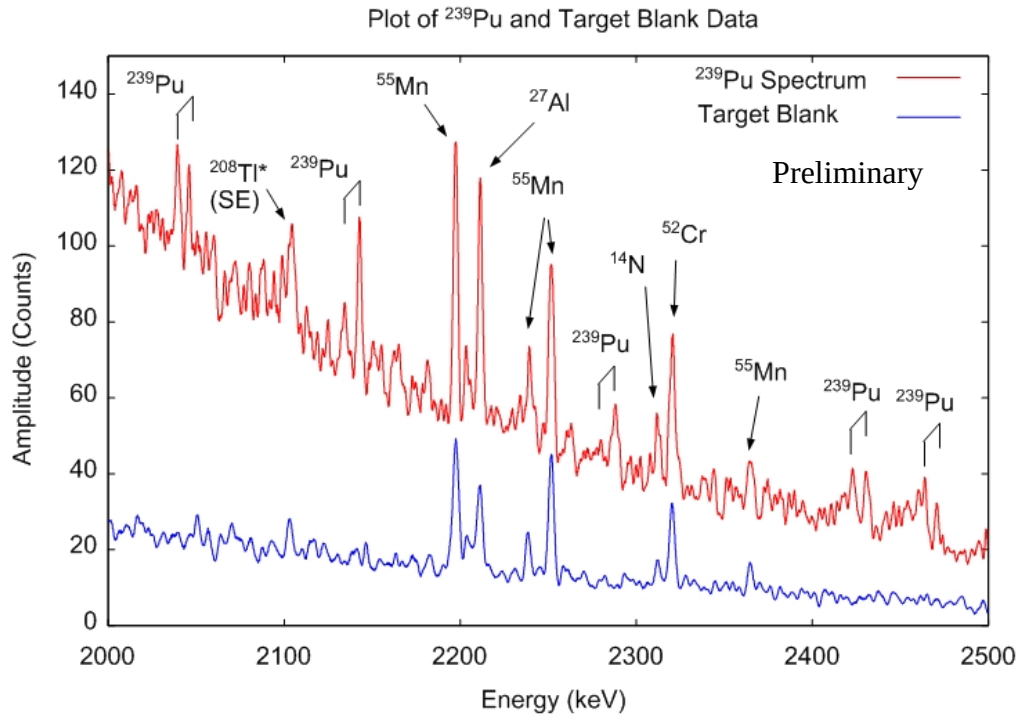

Figure 7: ^{239}Pu NRF Spectrum (Embedded in Nitronic-40 Steel) (preliminary)

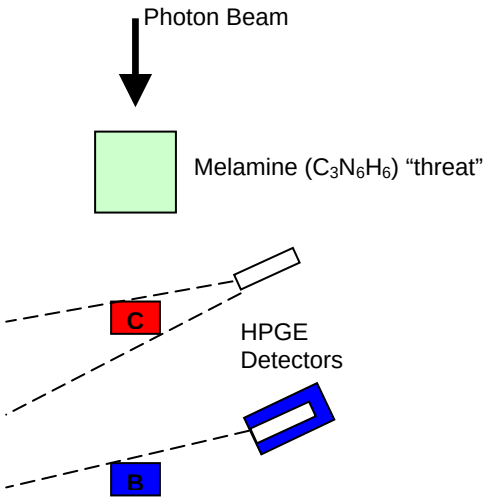

Figure 8: Transmission Geometry Using a Melamine Simulant and Detecting Carbon

and without the melamine displays the absorption of the resonant process in carbon over and above the electronic absorption. Corrections are necessary for the energy difference between the reference boron resonance and the resonance in carbon. Also, corrections are required for differential processes such as Delbruck and Raleigh scattering. These latter processes are well understood and present small differential effects when the monitoring target is close in Z and the resonant lines are close in energy.

The ratio of the counts in the boron line with one and two feet of melamine are in agreement with the calculated non-resonant absorption that is expected. The ratios of the carbon line are consistently lower than those of the boron line showing the significant effect of the nuclear absorption, approximately 30% with two feet of melamine.

An interesting feature of the spectrum from one element is the fact that many of the events originate from the NRF lines from the target. This means that the Compton edge, and for some distance below that edge, all events arise from the NRF line. The implication is that it is possible to amplify the efficiency of detecting an NRF line when only one target is involved by using the events in a portion of the continuous spectrum below the resonance line. This is particularly evident for light nuclei which play an important role in explosives and other contraband. Only one reference target can be used for each isotope in the transmission technique, making it a candidate to use the integration of events as suggested above and possibly using lower resolution detectors of less expense.

III. Overview of Effective-Z (EZ-3D™) Technology

The effective-Z technology discovered by Passport Systems, Inc. depends on the analysis of the entire spectrum of photons scattered from a target when irradiated by energetic photons. The nature of the spectrum depends on the complex interaction of multiple processes and depends sensitively on the Z of the target. The results of this analysis can be summarized by a set of parameters that display the sensitivity to Z and one such datum normalized to Z=82 is displayed in Figure 9 for the bremsstrahlung endpoint energy of 5.3 MeV.

The EZ-3D™ effect depends dramatically on Z. The effect can be used to delineate the effective-Z of a region of space along with an estimate of the mass associated with that effective-Z. Because the entire photon spectrum is used, the counting rates for the EZ-3D™ technology are much higher than for NRF allowing a rapid determination of a region of interest for further inspection by NRF for isotopic determination. The EZ-3D™ measurement establishes the ROI with very high probability of detection and low false positives. A ROI could be a region with a high value of the effective-Z if the interest is SNM such as ^{235}U or ^{239}Pu and associated shielding. On the other hand, a ROI could be a region where an anomalously low value of the effective-Z is detected compared to that expected based on surrounding values or a manifest.

In Figure 10 an example of high-Z is demonstrated wherein a different sequence of examples is scanned in an environment of 1.2 g/cc of steel. The first shows a scan when 2 inch thick iron plates are used. The second shows a scan when the iron plates are replaced by a one inch thick plate of lead. The third shows a scan when the one inch thick lead plate is replaced by a sandwich made of ¼

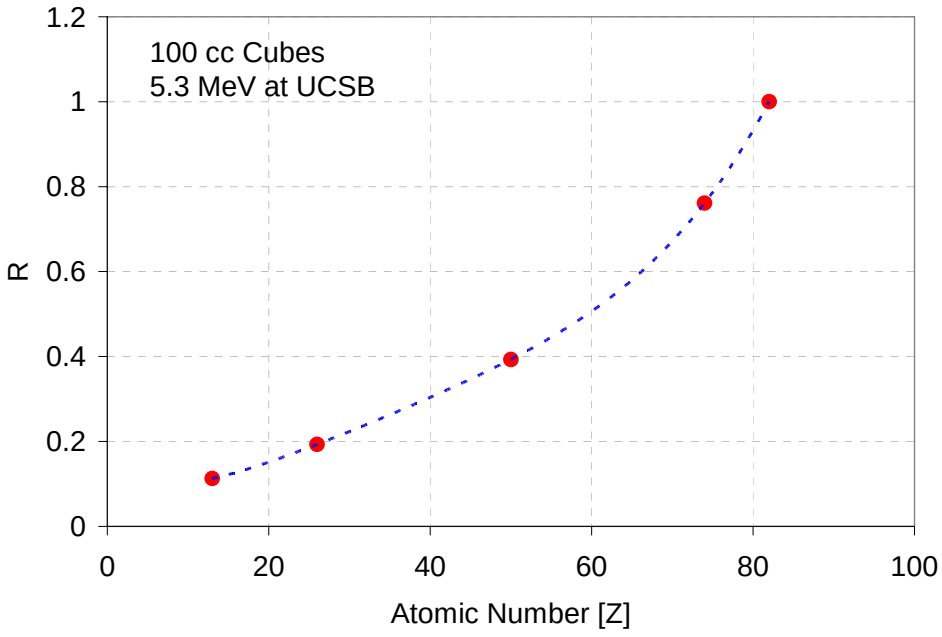

Figure 9: Dependence of an Effective-Z Parameter on Z

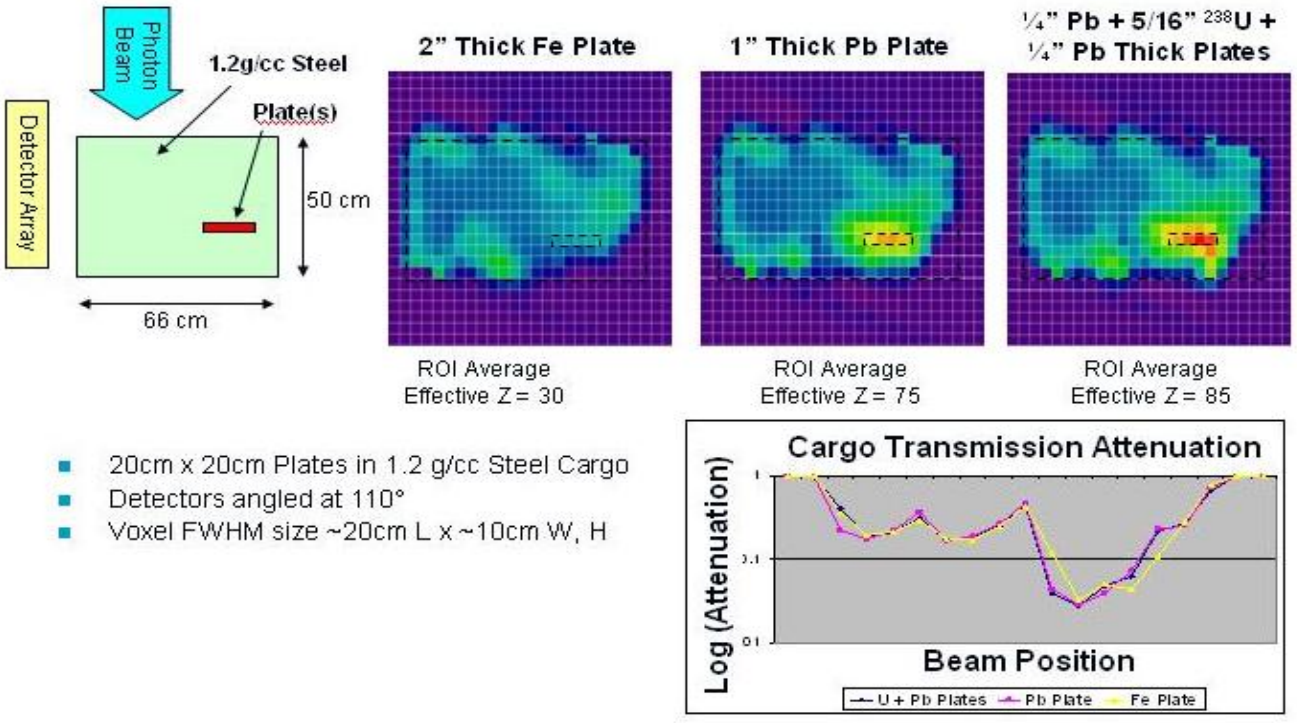

Figure 10: Effective Z Detection of ROI with 238U and Pb

inch lead plate, 5/16 inch DU plate and a ¼ inch lead plate. In all three cases the attenuation is approximately the same as shown by the x-ray transmission data in the insert on the lower right of Figure 10. The iron plate, the lead and the uranium/lead all show the same pattern and are indistinguishable. However, the effective-Z of the region with the plates shows the dramatic change due to the introduction of the DU. In a similar fashion, different regions of low-Z can be distinguished.

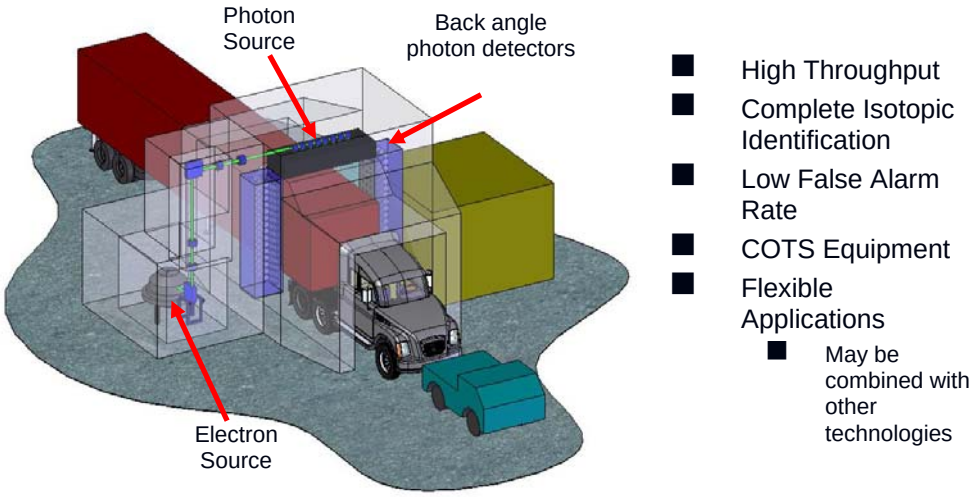

Figure 11: Schematic of a Possible Deployable Scanner

IV. Conclusions

The combination of EZ-3D™ and NRF presents a formidable capability to detect SNM and also explosives. One possible configuration is shown in Figure 11 for a large container system. Other configurations are possible as well that emphasize different types of threats. The use of EZ-3D™ allows for a high throughput with high probability of detection. The use of NRF allows a complete isotopic identification with a low false alarm rate. The system can be built with commercially available components. The system is modular and each detection modality can be used in combination with other inspection systems as well.

V. Acknowledgment

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References

1. W. BERTOZZI and R.J. LEDOUX, NIM in Phys Res. **B 241** (2005) 820-825
2. W. BERTOZZI, S. KORBLY, R. J. LEDOUX, W. PARK, NIM in Phys. Res. **B 261** (2007) 331-336
3. KNEISSL, H. H. PITZ, A. ZILGES, Prog. Part. Nucl. Phys. **Vol. 37** (1996) 349-433 (and references therein). This reference provides a comprehensive review of theory and data.
4. Data taken at the S-DALINAC of the Technical University of Darmstadt, Darmstadt, Germany
5. Data taken at the Free Electron Laser Facility at the University of California at Santa Barbara
6. Data taken at the Electron van de Graff Facility of the High Voltage Research Laboratory (HVRL) of the Massachusetts Institute of Technology (MIT)
7. These data were part of a collaborative effort using the Passport Systems Test Bed at the HVRL of MIT. The collaboration involved experimenters from Lawrence Livermore National Laboratory that provided the ^{239}Pu target and the Pacific Northwest National laboratory that provided the ^{235}U target and Passport Systems, Inc.