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Detection of Ionizing Radiation using Solar Blind Air Fluorescence

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ABSTRACT

This report looks at the feasibility of detecting radiation sources by using the photons produced by the ionization of the air surrounding the source. Some of these fluorescence photons are emitted in the UV solar blind spectral region which offers the possibility of detection in full daylight. A review of the basic technique is given as well as detailed radiation source simulations using the Geant4 simulation package and some preliminary experimental studies. Some conclusions on the practicality of this technique for radiation detection, as well as unresolved issues, are discussed.

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Detection of Ionizing Radiation using Solar Blind Air Fluorescence

Executive Summary

This report looks at the feasibility of detecting ionizing radiation by using the photons produced by the ionization of the surrounding air. Some of these fluorescence photons are emitted in the UV solar blind spectral region which offers the possibility of detection in full daylight.

If a practical detector could be realized utilizing this detection technique it would offer the following benefits over existing radiation detection methods:

- Standoff imaging detection of ionizing radiation. Such a detector would provide information on source location and morphology while keeping the detector and operator out of the radiation field.
- Standoff detection of alpha and beta radiation. Both beta and particularly alpha particles are strongly absorbed by air. A standoff detection capability would allow for large areas to be efficiently surveyed for alpha and beta contamination.
- Detailed characterisation of radiation distributions from sources with complex morphology, such as from nuclear accidents or from radiological dispersal devices. Such an incident would require a lengthy and complex series of measurements with non-imaging detectors to characterize the radiation hazard. Even gamma-ray imagers, which can image point sources well, struggle to measure complex source scenarios. The fluorescence signal discussed in this paper is comprised of photons, which could be imaged to characterize any complexity of source distribution.

This report summarizes the basic physics involved in fluorescence photon production and includes a preliminary laboratory measurement of solar blind photons from an alpha source. A number of issues are identified that will need to be addressed before an assessment can made on the practicality of this detection technique as part of a radiological defence capability:

- Are the radiological scenarios where this detection technique would outperform other detection methods realistic threat scenarios?
- Are the available solar blind photon detection methods sufficiently sensitive and physically robust enough to provide a field-able instrument capable of detecting realistic threats?
- Is the solar blind photon background, from electrical discharges and other sources, sufficiently low that it will not interfere with the operation of a solar blind radiological detector?

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1 Introduction

When ionizing radiation passes through the atmosphere some of the energy is transferred into the excitation of diatomic gasses, in particular N_2. This effect is responsible for phenomena such as auroral airglow and the light emitted from electrical discharges. Air fluorescence has been used for decades as a way to detect ultra high energy cosmic rays that interact with the Earth's atmosphere to generate large cascades of ionizing particles. This effect is also used for daytime imaging of corona discharge from faulty high tension electrical lines.

The absolute efficiency of fluorescence light production is critical in determining whether this technique can be used for practical radiation monitoring. The physics involved is quite complex, with initial excitation processes, the decay of excited states to metastable energy states, collisional de-excitation with atmospheric gasses and final decay to produce the fluorescence emission. The fluorescence yield is pressure and temperature dependent and atmospheric contaminants such as water vapour also have a small effect on the rate of collisional de-excitation. The effect of de-excitation varies with wavelength, as each excited state has a different lifetime, so the relative intensity of the spectral lines will vary somewhat with atmospheric conditions.

The most accurate measurement of absolute fluorescence yield has been undertaken by the cosmic ray field in the wavelength range 300-420 nm [1, 2, 3]. This range corresponds to the peak of the N_2 emission and allows for the detection, on cloudless moonless nights, of ionization tracks from individual cosmic rays of sufficiently high energy ($> 10^{18}$ eV). The yield measurements are made in carefully calibrated laboratory experiments and give a yield at standard sea-level temperature and pressure of 16 ± 2 photons (300-420 nm) per MeV of ionization deposit [3]. This would mean that a hypothetical source depositing 1 MeV of ionization energy per particle with a source strength of 10 GBq would produce 1.6×10^{11} photons per second in the 300-420 nm range. This would give more than 10^4 photons cm^{-2} s^{-1} at a distance of 10 m, which would be easily detectable with a sensitive UV detection system.

The difficulty with using the 300-420 nm band is that the background rates will be very high. Figure 1.1 shows the solar photon flux at sea level as a function of wavelength. In the range 300-420 nm the surface solar flux would be on the order of 2×10^{20} photons m^{-2} s^{-1}, which would swamp any possible air fluorescence signal even when not looking directly at the sun. This was the conclusion of a previous DSTO report on this detection technique [5]. This report also discussed the possibility of air fluorescence detection in the solar blind region of the UV spectrum, but flux calculations were not made. Figure 1.1 shows that for wavelengths shorter than 280 nm essentially no solar radiation reaches the Earth's surface, primarily because of atmospheric ozone absorption. The only background photons in this spectral region should be from radiation backgrounds or man-made air ionization such as from flames or electrical discharges. Unlike in the 300-420 nm range there do not seem to be accurate measurements of the absolute air fluorescence yield in the solar blind range. Relative fluorescence yields for a variety of gasses, including air, have been published for example in [6] in which the relative fluorescence yield from air is given over the range 200-500 nm. In the 300-420 nm range 5 principle emission lines are identified via [7] as belonging to the second positive to first positive transition of N_2 (N_2^* $C^3\Pi_u \rightarrow B^3\Pi_g$).

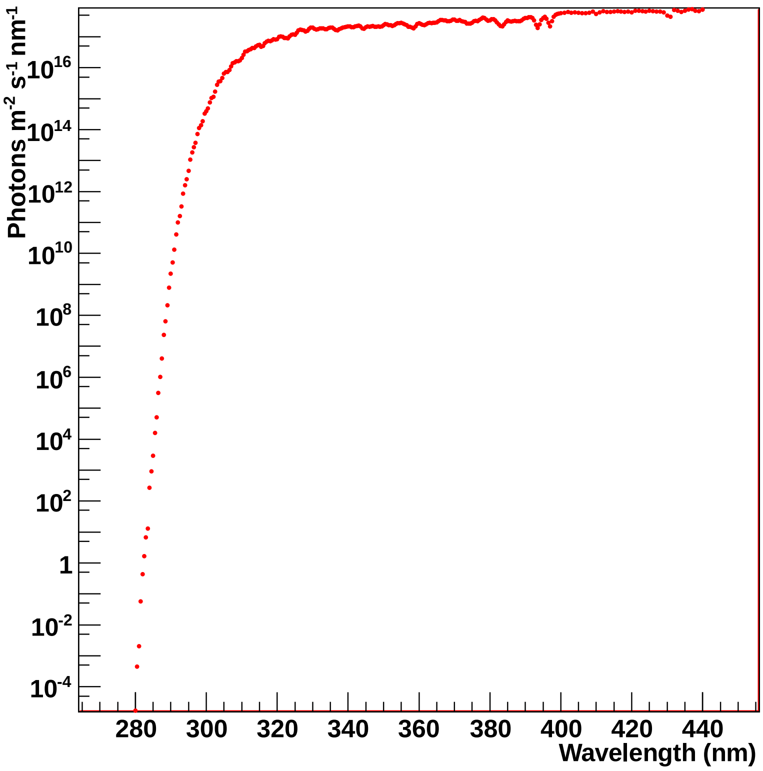

Figure 1.1: *Solar photon flux at the surface of the Earth as a function of photon wavelength [4].*

In the range 200-280 nm 10 emission lines are identified as belonging to the $h^1\Sigma_u^+ \rightarrow x$ transition of N_2.

These data show that photon emission is around 200 times weaker in the solar blind region compared to the 300-420 nm region. Scaling by the known yield in the 300-420 nm range gives an estimated absolute solar blind (200-280 nm) yield of around 0.08 photons per MeV of ionization energy deposit. It should be noted that the measurements in [6] were made at an air pressure of 3 atm, which may affect the relative yield between the solar blind and 300-420 nm regions.

The previous example of a radiation source (10 GBq and 1 MeV ionization deposit per particle) would give a around 50 solar blind photons cm^{-2} s^{-1} at a distance of 10 m. This is clearly a weak signal and the usability of any detector would depend on the collecting area and efficiency of the detector, the dark count rate and any sources of background.

1.1 Geant4 simulation

The details of the radioactive source will also determine the efficacy of the air fluorescence detection method. The type of radiation will affect the physical volume of air which will

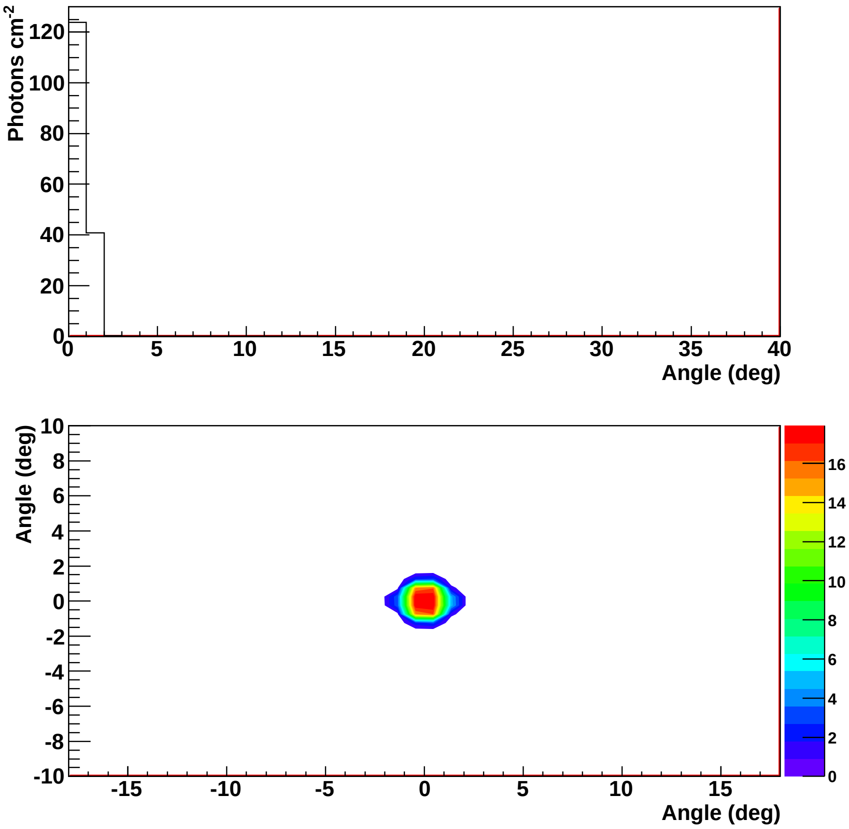

Figure 1.2: *Geant4 simulation of the solar blind photon flux from* 10^{10} *decays of* Am_{241} *viewed by a 40 cm diameter detector at a distance of 10 m from the source. The top plot shows the angular distribution of photon arrival directions with respect to the source position. The bottom plot shows the expected 2D photon image with the arrival angles of photons expressed in degrees. The contour colour in the bottom plot indicates the signal strength (photons* cm^{-2}*). The photon arrival directions include depth of field blurring from the detector aperture.*

be the source of fluorescence photons. The physical distribution of the source and the interaction of radiation and fluorescence photons with nearby materials will also have an impact on detectability using this technique.

To investigate some of these effects detailed simulations have been conducted using the Geant4 simulation package [8]. The code was modified from the standard WLS example code distributed with Geant4. The particle source was changed to the general particle source model (G4GeneralParticleSource) which allows for the source to be a specified radioactive isotope and provides a full decay chain for the isotope. The world volume in the simulation was defined as a box of air and the fluorescent characteristics of the air, including the wavelength dependence of the air fluorescence, were defined for the solar blind spectral region as shown in Table 1.1.

Other optical characteristics of the air, such as refractive index, Rayleigh scattering length and absorption length were also defined. For the simulations presented here it was

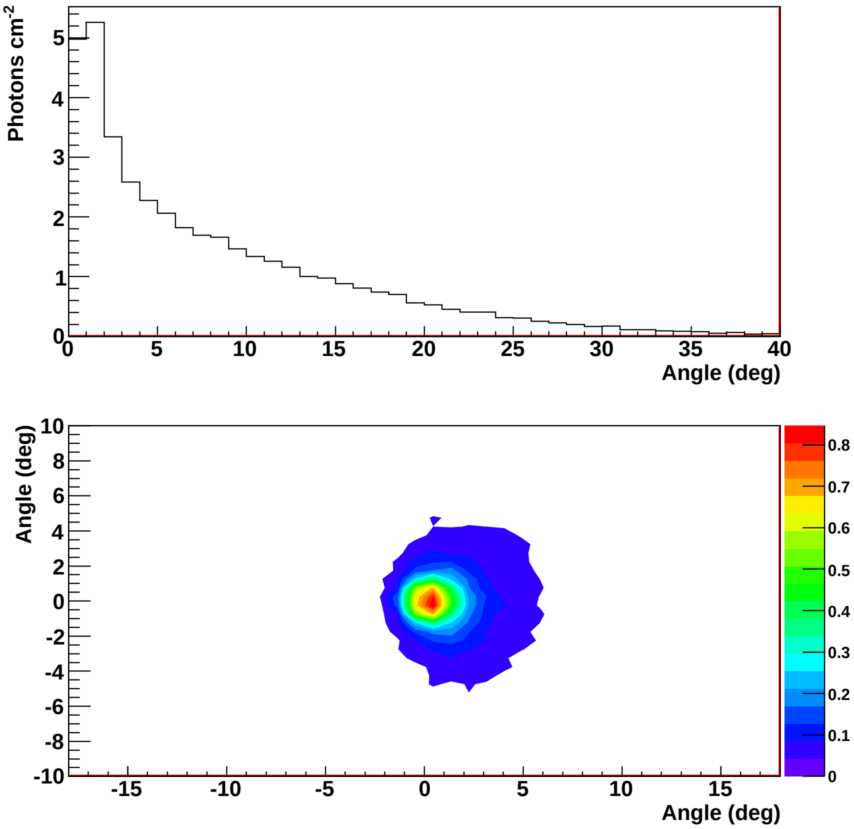

Figure 1.3: *Same as for Figure 1.2 but with the source being* 10^{10} *decays of* Sr_{90}*. The asymmetry of the image is a simple parallax effect with the signal nearer the detector being collected more efficiently.*

assumed that the radioactive source was located on top of a concrete slab. The concrete material definition did not include a fluorescence term, but the slab will provide a target

Wavelength range (nm)	Relative yield
200-210	0.0
210-220	0.0
220-230	0.06
230-240	0.21
240-250	0.55
250-260	1.0
260-270	1.0
270-280	0.85

Table 1.1: *Relative fluorescent yield defined in the Geant4 simulation based on spectrum in [6].*

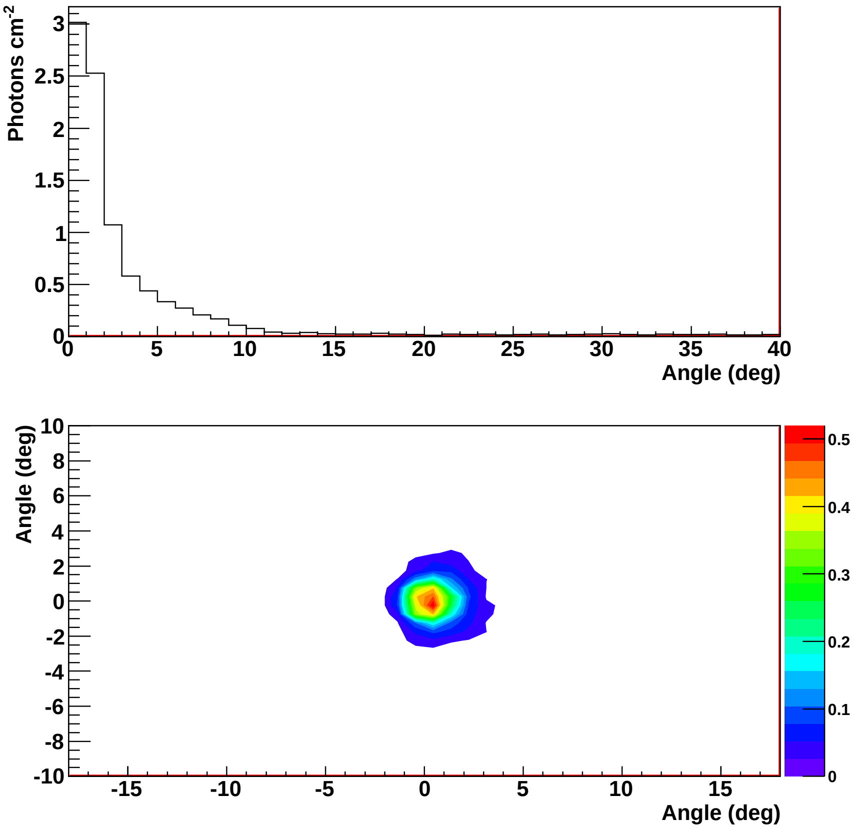

Figure 1.4: Same as for Figure 1.2 but with the source being 10^{10} decays of Cs_{137}. Most of the fluorescence signal is produced by low energy beta particles from the source. Deeper exposures show a faint extended halo around the source from gamma interactions with the air and floor materials.

for generating secondary particles, such as delta rays, that might generate air fluorescence photons. In these simulations the albedo of the concrete surface is set to zero, but this could be changed to simulate optical reflections from the surface. To investigate the effects of source shielding, the simulation also included an option to contain the source in a lead box.

The simulated fluorescence photon detector was defined as a simple spherical surface with 100% detection efficiency. The detector writes the arrival direction and position of photons at its surface to a file which can then be used to simulate a detailed detector response. The detector was 40 cm in diameter and was placed at a distance of 10 m from the source (at position x=5.7 m, y=5.7 m, z=5.7 m). Figures 1.2 through 1.4 show simulated solar blind photon air fluorescence emission from common radiological sources (Am_{240},Cs_{137} and Sr_{90}). In each case 10^{10} decays have been simulated from a point source at the origin. All sources produce a fluorescence signal that is consistent with expectation based on arguments made earlier. The alpha emitter produces the strongest signal, with a near point-like source of fluorescent photons.

A detailed comparison between Geant4 simulation and measurement would require an absolute fluorescent yield measurement and a wavelength dependent absolute calibration of the photon detector which is beyond the scope of this work.

1.1.1 Effects of source shielding

The effects of source obscuration or shielding on detectability are of interest for many radiation detection applications. Clearly even a very thin opaque layer will absorb fluorescence photons, but energetic particles can penetrate shielding either directly or via secondary cascades. These particles may then generate a fluorescence signal in the air surrounding the shielding container.

This report will not include a detailed assessment of the effects of shielding on fluorescence detection, but Figures A1 through A3 in Appendix A show some example Geant4 simulations of shielded sources. As expected the signal is greatly reduced, but a faint glow around the exterior of the shielding container is present.

2 Measurement of fluorescence signal from a Po_{210} alpha source

Laboratory measurements were conducted to provide a basic validation of the modelling work discussed in this report. The experimental capabilities developed will be used in the future to measure the yield efficiency of solar blind air fluorescence and to test the feasibility of radiation detection with this technique.

The work was undertaken with a 185 MBq (5 mCi) Po_{210} source purchased in April 2012. Po_{210} is a nearly pure alpha emitter which is widely used for industrial processes such as de-ionization. The photon detector consisted of a Hamamatsu H10682-09 photon counting module with an Acton Research 254-N bandpass filter. The efficiency curves, based on manufacturer supplied data, are shown in Figure 2.1.

The measurements were made in the dark box shown in Figure 2.2. Although the detector is nominally solar blind, it still has some small detection efficiency for wavelengths above 280 nm and the dark box is used to eliminate noise from background photons. Measurements were taken in on-off pairs, with a stepper motor actuated plastic cup covering the source to provide off source data (Figure 2.2). Each measurement consisted of 200 photon detection counts, with the acquisition time recorded to provide a count rate.

Results from a set of 20 on-off pairs of measurements are shown in Figure 2.3 when the Po_{210} source was present (blue plot) and replaced by a dummy source (red plot). The results show the difference in count rate between shutter open and shutter closed conditions for each on-off pair. A clear excess in the count rate is seen in the presence of the source. No excess in the on-off count rate difference is seen when the source is not present. The off source count rates ($\approx$5 Hz) are consistent with manufacturer measured dark count rates for the PMT.

Figure 2.4 shows the same experiment as for Figure 2.3, but with an opaque but very thin plastic film (thickness $<$ 0.1 mm) covering the detector. This is to exclude the

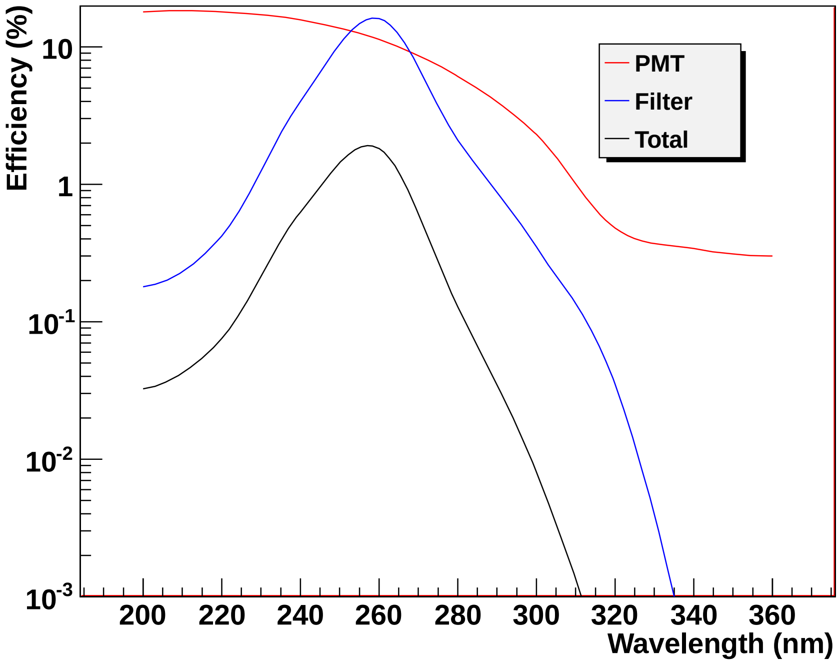

Figure 2.1: *Wavelength dependent efficiency of the photon detection system. PMT efficiency is only the manufacturer claimed photocathode production efficiency and does not include first dynode collection or threshold trigger efficiency.*

possibility that the on-source signal is due to gamma-ray triggers, either directly from the source or from the interaction of the alpha particles with nearby matter. No excess was seen with the Po_{210} source present when the plastic film was in place.

During measurements the Po_{210} source rests on top of a wooden pedestal that is painted matt black. Against the possibility that it is this surface (rather than the air) that is fluorescing, measurements were also taken with black tape covering the pedestal surface (see Figure 2.5). Changing the pedestal surface does not affect the yield of fluorescence photons.

3 Discussion

Simple calculations and detailed simulations show that radioactive sources will produce air fluorescence solar blind photons that could be used for source detection. The signal is very weak, with 10s or 100s of photons cm^{-2} for 10^{10} decays at a detection distance of 10 m for typical radioactive isotopes. A simple experiment has been conducted using a solar blind photon detector and a Po_{210} alpha source to confirm the production of solar blind photons.

The practical usefulness of this signal will depend on the details of the radioactive

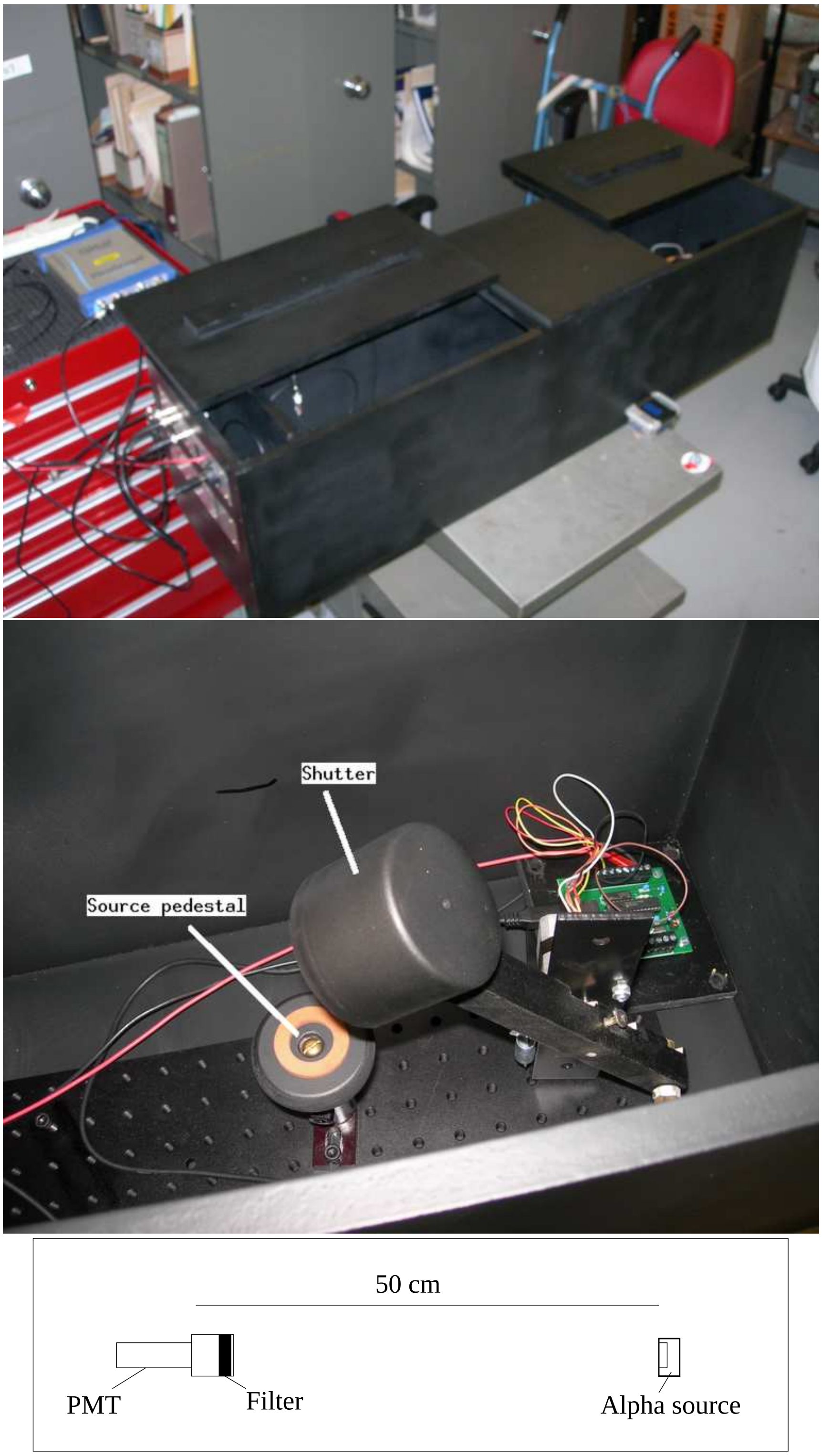

Figure 2.2: *Dark box (top), shutter mechanism (middle) and top view schematic of experimental layout (bottom).*

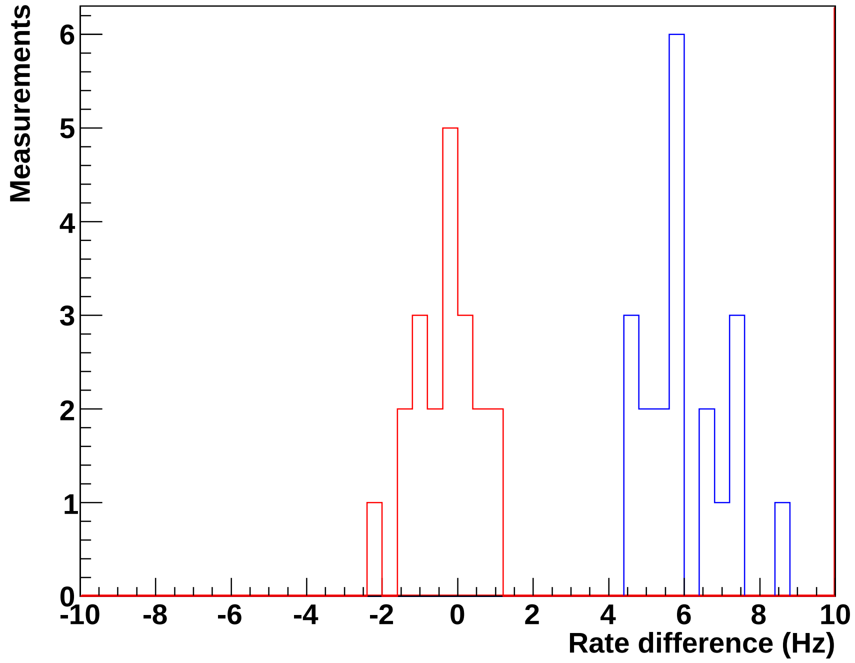

Figure 2.3: *Counting rate difference (shutter open - shutter closed) when the source is present (blue plot) and when the source is absent (red plot).*

source scenario: source strength and shielding, detection timescale, detector efficiency and dark noise rate and background sources of solar blind photons.

3.1 Advantages of fluorescence detection

Signal is ubiquitous
Ultimately any radiation source will produce some level of ionization, either directly from primary decay particles or from their interaction with the environment. Even with significant source shielding there is often punch-through of particles into the surrounding air. Most other detection techniques are specific to the type of radiation emitted.

Signal provides an image
The signal consists of photons which can be imaged to provide information on source location and morphology.

Standoff capability
In the solar blind UV range the typical atmospheric scattering length at sea level will vary from around 1 km at 200 nm to around 10 km at 280 nm. This will vary particularly with atmospheric aerosol loading, but indicates that at least for very strong sources this technique could give a standoff detection capability.

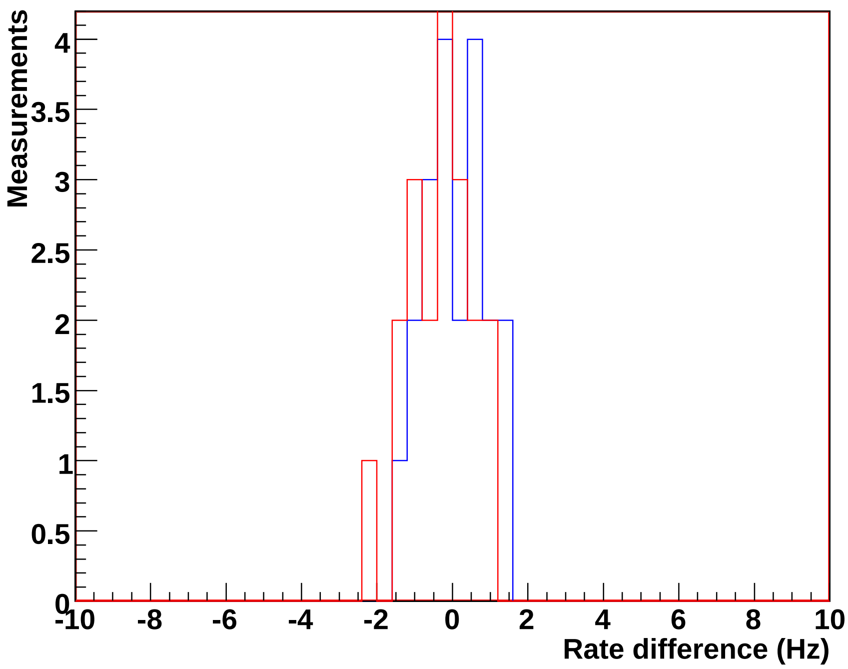

Figure 2.4: *Same as for Figure 2.3 but with a very thin sheet of opaque plastic in front of the detector. This demonstrates that the observed count rate excess when the source is present is not due to directly detected gamma-rays from the source or secondary interactions.*

Non-line of sight detection

The scattering of the fluorescence photons allows for the possibility of detection even when direct line of sight to the source is obscured. The fluorescence photons will reflect off local surfaces and also scatter in the air to produce a halo around the source. This could mean that a radiation source could be detected even when no direct radiation from the source reaches an observer.

3.2 Disadvantages and unknowns

Signal strength

The obvious disadvantage of this method is that the signal is extremely weak due to the low yield efficiency in the solar blind spectral range. Only very strong sources would be detectable and would require detectors with large collecting area, high efficiency and low dark count rate. There are commercially available solar blind photon detectors. The most obvious choice would be photomultiplier tubes (PMT) or microchannel plates (MCP). For example, Hamamatsu make PMT modules with CsTe photocathodes that have sensitivity primarily in the 200 - 300 nm range and a peak quantum efficiency of around 18%. These PMTs have quoted dark count rates of around 5 counts s^{-1} per cm^2 of photocathode area which could be reduced by active cooling. Various manufacturers make MCP imaging

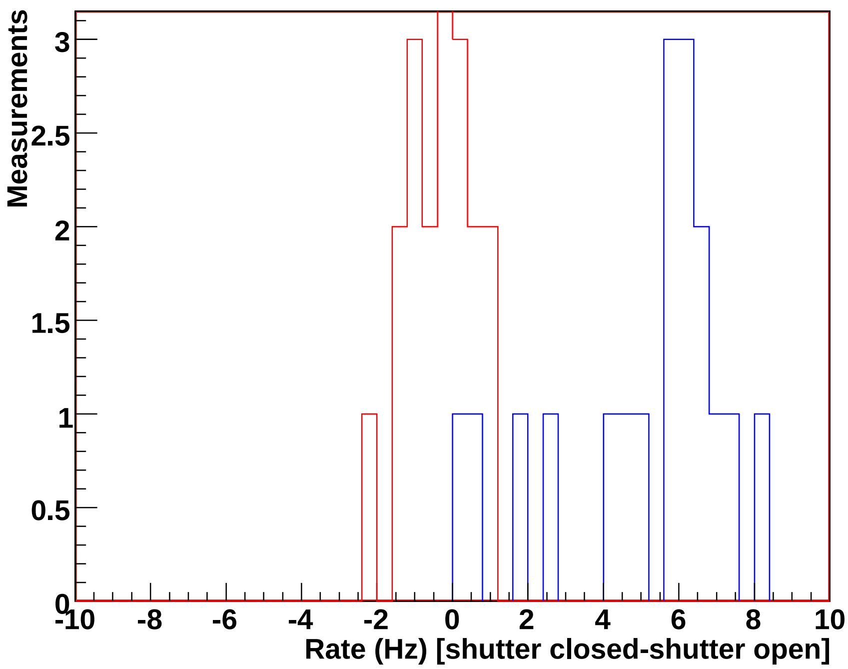

Figure 2.5: *Same as for figure 2.3 but with plastic tape covering the top of the pedestal. The yield of photons is unchanged compared to Figure 2.3 indicating that the pedestal surface is not the origin of the fluorescence signal.*

versions of these detectors, including Hamamatsu who produce a photon counting imaging version (model R10110U-03). The CsTe photocathode does have some sensitivity to wavelengths above 280 nm so solar blind filtering would be required for daylight operation. Ofil Systems, for example, produce a filter (model SB-AF) which gives good solar blind performance when combined with a CsTe photocathode detector. The combined detection efficiency of filter and detector is however rather low (around 1%).

There is considerable research activity in the area of solar blind detection which has applications in flame sensing, missile detection, communications, electrical breakdown detection and so on. It may be that Gas Electron Multiplication (GEM) detectors commonly used for flame detection [9] would provide a better detection platform for this application than either PMTs or MCPs. These detectors are intrinsically solar blind and may provide much higher detection efficiency (10-25%) than CsTe/Filter PMT or MCP detectors.

Exact measurement of yield efficiency
The yield value of 0.08 solar blind photons per MeV of energy deposit used here is an estimate based on measurements under non-STP conditions. A more accurate measurement of absolute yield efficiency in this wavelength range would be necessary for a detailed study on the practicality of using this technique.

Insensitivity to isotope type
The spectrum of fluorescence light is determined by the excitation states of N_2 and does

not contain information about the type of source that is producing the ionization. The morphology of the emission region may give some information about the type of emission (alpha, beta, gamma) but this could easily be obscured by, for example, the physical distribution of the radioactive material.

Fluorescence of background surfaces
Surfaces near the radioactive source should reflect some fluorescence light which may enhance the detected signal. Some materials may also fluoresce under radiation.

Backgrounds
Figure 1.1 shows that below a wavelength of 280 nm no solar radiation will be present at ground level. Solar blind photons will, however, be produced by processes such as flames, natural radiation, electrical discharge etc. The expected signal from a radioactive source will be weak, and detectability will depend on low background rates.

Timescales for detection
If an instrument were developed it might work best in a survey mode where it could accumulate data for long periods.This would clearly only be useful for monitoring a stationary source.

4 Conclusions

Using solar blind air fluorescence photons has some potential advantages over other radiation detection methods. It could possibly provide a source imaging standoff capability that would be sensitive to any ionizing radioactive source. It is also clear that the signal will be very weak and will require detector sensitivity and background rates that may not be achievable in practice. The viability of this technique also depends strongly on the radiological source scenario: source type, strength, containment and the time permissible for detection. Further studies would be required to resolve these issues.

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Appendix A

Some examples of Geant4 simulation of shielded sources.

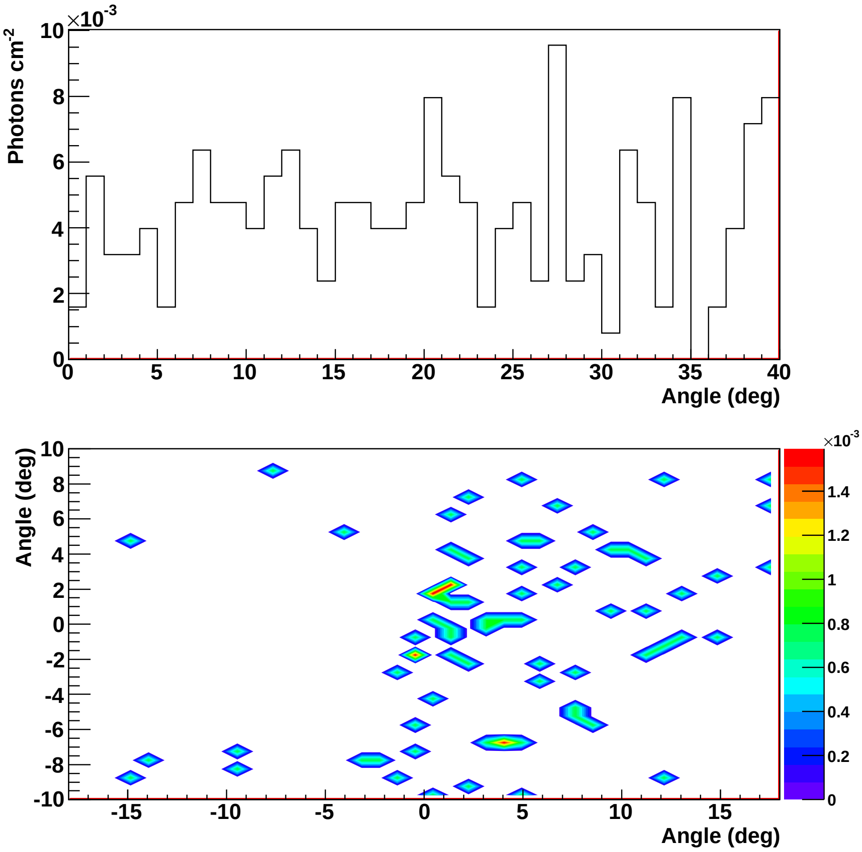

Figure A1: *Geant4 simulation the solar blind photon flux from of* 10^{11} *decays of* Am_{241} *in a 20 X 20 X 20 cm box with 2 cm thick lead walls. The detector location and size is the same as previous simulations.*

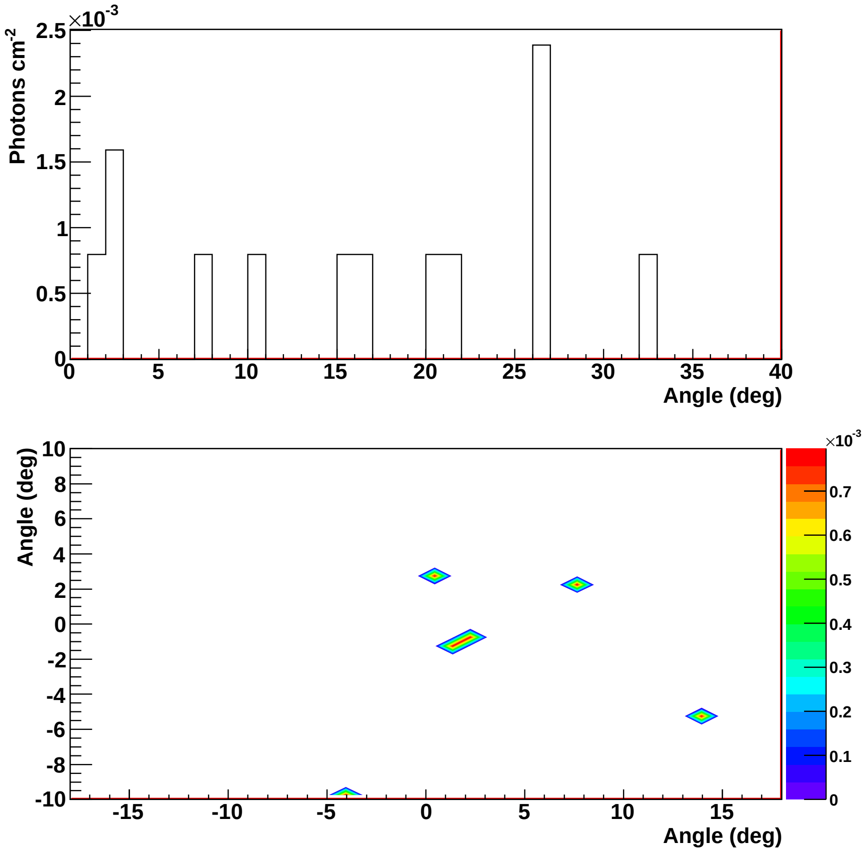

Figure A2: *Same as for Figure A1 but with the source being* 10^{11} *decays of* Sr_{90}.

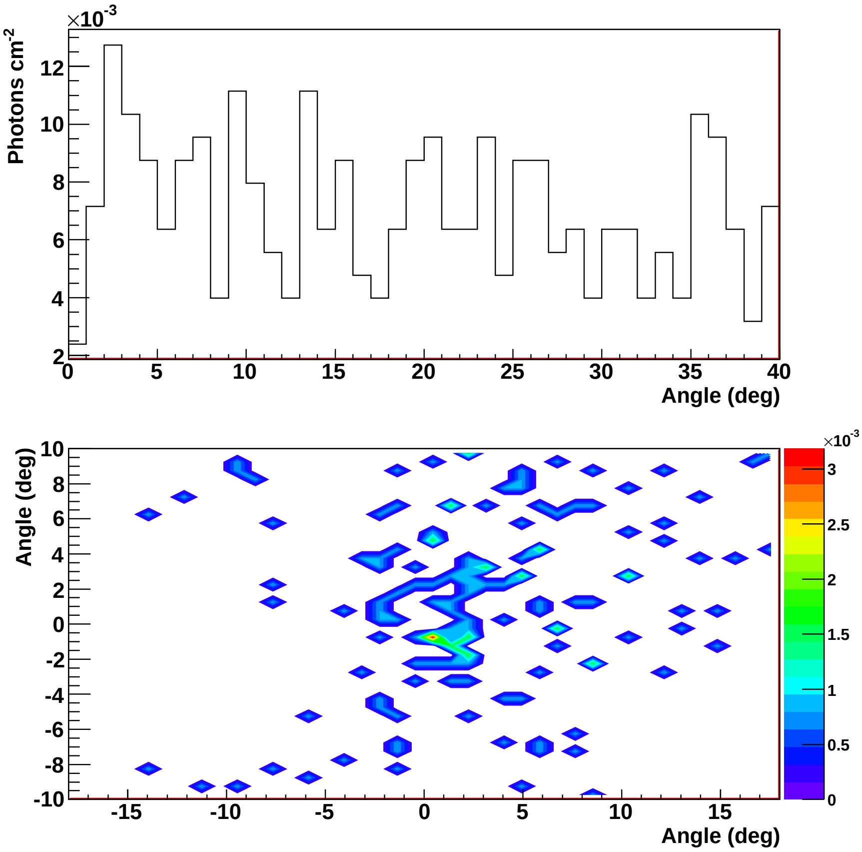

Figure A3: *Same as for Figure A1 but with the source being* 10^{11} *decays of* Cs_{137}.

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19. ABSTRACT

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