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Remote Optical Detection of Alpha Radiation

J. Sand[1], V. Hannuksela[1], S. Ihantola[2], K. Peräjärvi[2], H. Toivonen[2], and J. Toivonen[1]

[1]*Tampere University of Technology, Optics Laboratory, P.O. Box 692, FIN-33101 Tampere, Finland*

[2]*STUK – Radiation and Nuclear Safety Authority, P.O. Box 14, FIN-00881 Helsinki, Finland*

johan.sand@tut.fi

Abstract. Alpha emitting radiation sources are typically hard to detect with conventional detectors due to the short range of alpha particles in the air. However, previous studies have shown that remote detection of alpha radiation is possible by measuring the ionization-induced fluorescence of air molecules [1,2]. The alpha-induced ultraviolet (UV) light is mainly emitted by molecular nitrogen and its fluorescence properties are well known. The benefit of this method is the long range of UV photons in the air. Secondly, the detection is possible also under a strong beta and gamma radiation backgrounds as they do not cause localized molecular excitation. In this work, the optical detection was studied using two different detection schemes; spectral separation of fluorescence from the background lighting and coincidence detection of UV photons originating from a single radiative decay event. Our spectrally integrated measurements have shown that one alpha decay event yields up to 400 fluorescence photons in the air and all these UV photons are induced in a 5 ns time-window. On the other hand, the probability of a background coincidence event in 5 ns scale is very rare compared to the number of background photons. This information can be applied in fluorescence coincidence filtering to discriminate the alpha radiation initiated fluorescence signal from much more intense background lighting. A device called HAUVA (**H**andheld **A**lpha **UV A**pplication) was built during this work for demonstration purposes. HAUVA utilizes spectral filtering and it is designed to detect alpha emitters from a distance of about 40 cm. Using specially selected room lighting, the device is able to separate 1 kBq alpha emitter from the background lighting with 1 second integration time.

1. Introduction

Conventional detectors require direct interaction with the alpha particle which makes the localization of contamination a laborious task. Due to the stealth nature of pure alpha sources, new detection techniques are needed in safety and security applications. The challenge of alpha detection is especially evident in metal recycling and decommissioning but novel real-time monitoring technology is also needed for nonproliferation purposes.

Previous studies have shown that remote detection of alpha radiation is possible by measuring the ionization-induced fluorescence of air molecules [1,2]. In these studies, the alpha-induced fluorescence is detected photographically either by a film or a CCD-camera. This is based on the fact that alpha particles deposit their energy in a small sphere that has a radius equal to the range of alpha particles in the air. Thus the fluorescence is emitted from a small, localized volume [2]. If the level of ambient UV light is low, these hot spots can be observed from a long distance since UV photons have a very long range in the air. Furthermore, the detection is also possible under a strong beta and gamma background because they do not induce as localized fluorescence as alpha radiation [1]. While photographical methods can reveal the spatial distribution of alpha contamination in a single measurement, their time response is somewhat limited due to the small collection angle required for imaging.

In this work, photomultiplier tubes (PMTs) are used to measure the alpha induced fluorescence. The main benefits of photomultiplier tubes are their single photon sensitivity and low dark count rate per detector area. Furthermore, the sensitivity and time response of a PMT is good enough for the remote observation of individual alpha decay events as nearly all of the photons are emitted in a short 5 ns time window [3]. The aim of this work is to demonstrate a real-time alpha monitoring system that could detect surface contamination in a short one second integration time at a distance of 40 cm.

2. Physical properties of alpha induced UV fluorescence

The excitation mechanism of the alpha-induced ultraviolet emission is similar to that in northern lights. In cosmic emissions, charged high energy particles collide with atmospheric molecules and this creates secondary particles such as electrons which produce even more secondary particles [4]. The cascaded process leads to the excitation of air molecules and the de-excitation can be observed as greenish and reddish light. However, at standard atmospheric conditions the fluorescence light is emitted mainly in the near UV region between 300-400 nm and cannot be seen by eye. This UV light is emitted by excited nitrogen molecules whereas a weak visible portion is originated from oxygen. The alpha-induced molecular excitations are supposed to be generated by the secondary electrons, as is the case with northern lights.

Nitrogen fluorescence was chosen for the remote alpha detection since the most of the alpha-induced fluorescence is originated from nitrogen molecules. In addition, the fluorescence properties of molecular nitrogen are well known and studied by several authors. Table 1 shows the wavelengths of the most intense fluorescence band heads of nitrogen [4]. The peaks have a full-width of half-maximum of about 1 nm which enables the spectral separation of fluorescence from background lighting.

Table 1. Most intensive fluorescence bands of neutral (2P) and ionized (1N) nitrogen molecules [4]. Peaks have a full-width half-maximum of about 1 nm. The upper vibrational state is marked with v' and the lower with v''. The intensity of a transition is proportional to the Einstein coefficient $A_{v',v''}$.

λ [nm]	System	v'	v''	$A_{v',v''}$ [s^{-1}]
311.67	2P	3	2	$5.94\ 10^{6}$
313.60	2P	2	1	$1.01 \cdot 10^{7}$
315.93	2P	1	0	$1.19 \cdot 10^{7}$
337.13	2P	0	0	$1.31 \cdot 10^{7}$
353.67	2P	1	2	$5.54 \cdot 10^{6}$
357.69	2P	0	1	$8.84 \cdot 10^{6}$
375.54	2P	1	3	$4.93 \cdot 10^{6}$
380.49	2P	0	2	$3.56 \cdot 10^{6}$
391.44	1N	0	0	$1.14 \cdot 10^{7}$

In order to illustrate the alpha decay induced UV emission, a point like alpha emitter on a surface was modelled with the FRED Optical Engineering Software (Photon Engineering LLC). It was assumed that the alpha particles travel 4.1 cm in the air at maximum. During the alpha particle trajectory, it excites atmospheric molecules through secondary effects. Figure 1 (a) shows how the excitation sites are distributed around the point like alpha emitter in our model. From these sites, the photons are emitted in random directions, as shown in fig. 1 (b). Thus, the point like alpha emitter forms a fluorescence emitting volume around itself. The ongoing work suggests that the number of emitted photons is around 400 per one 5 MeV alpha decay event in the air.

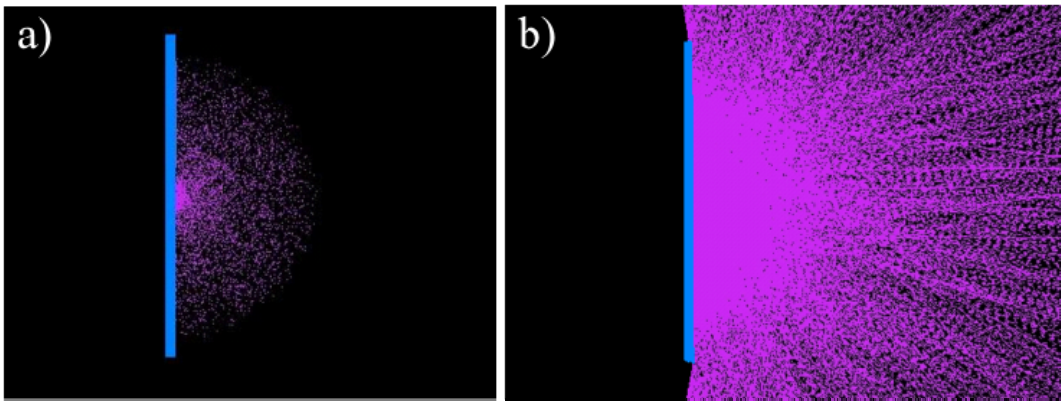

Fig. 1. Model of point like alpha emitter on surface acting as light source. ***a)*** *Alpha particles are inducing fluorescence photons in volume inside of the hemisphere having radius of 4.1 cm.* ***b)*** *Fluorescence photons are emitted in random directions.*

3. Detection techniques

The main challenge of the optical detection is to discriminate the weak fluorescence signal from the background lighting. In order to overcome this challenge, two different detection schemes were studied. The first method is based on separate detection of fluorescence and background light via spectral filtering. This technique was chosen for the basis of a HAUVA demonstration device due to its easy implementation. The second method utilizes coincident detection of two photons generated by a single decay event. It turns out that this is also a very attractive method for alpha monitoring.

3.1 Spectral filtering

The aim of the optical design is to collect as much of the emitted photons as possible. This leads to a large numerical aperture of optics i.e. a large collection angle for the optics. With the large collection angle and the volume emitter source, the spectral wavelength separation cannot be made by conventional spectrographs without losing most of the photons. To collect more photons from the source area, the wavelength separation in this work is done by interference filters. The fluorescence is collected with a 40 nm bandpass filter (Semrock, Inc.) having the centre wavelength at 320 nm. Furthermore, the level of background lighting is detected by combining the first fluorescence filter with a 15 nm bandpass filter having the centre wavelength at 295 nm. This effectively limits the background detection to a narrow wavelength range of 298 nm – 303 nm.

The interference filters used in this work have the transmittance of about 10^{-6} to 10^{-7} in the visible wavelengths. The visible light blocking efficiency of the demonstration device is enhanced by introducing three similar fluorescence filters in a row. The transmittance also drops at the bandpass wavelength, but as the transmittance of a single filter in bandpass is about 90 %, the effect is not pronounced. A small decrease in the fluorescence signal is necessary to get rid of the visible background. Artificial lighting that does not contain UV light can then be used with such a filtering. With this method, the visible lighting is effectively filtered out, and the fluorescence is only marginally decreased.

Figure 2 shows the optical design of HAUVA device. The design is optimized for the maximum collection efficiency for a detector having a diameter of 15 mm. The total collection efficiency was calculated to be 0.12 % when the point like alpha emitter is located at 40 cm distance from the first lens having a diameter of 75 mm. The interference filters are marked in the figure with F1 and F2.

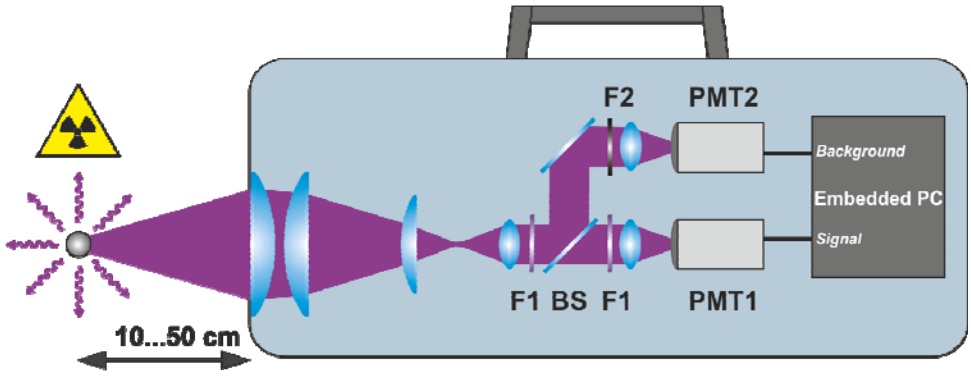

Fig. 2. Schematic model of HAUVA device. The collected light is divided with a beam splitter (BS) to the lower channel and the upper channel. The spectral selection of wavelengths is done with interference filters F1 and F2. PMT1 detects the fluorescence signal at wavelength range of 300 nm – 340 nm and the PMT2 detects background lighting at 300 nm. The data are collected with an on-board computer.

The optics are mounted inside of a tube, so that all the lenses have the same optical axis. The tube is sealed in a way that no light will leak to the detector from the side. The only optical access to the detectors is through to the first lens. Channel multipliers MP 1982 (PerkinElmer Optoelectronics) were used as detectors. The optics were packaged with detectors, electronics and a battery to a box having the total weight of 6 kg. HAUVA is portable or can be mounted on top of a tripod. The current detector readings are printed on a small display in the back of the device. The data can also be streamed via a Bluetooth connection to a mobile phone for graphic representation. Figure 3 shows the demonstration device in a laboratory environment.

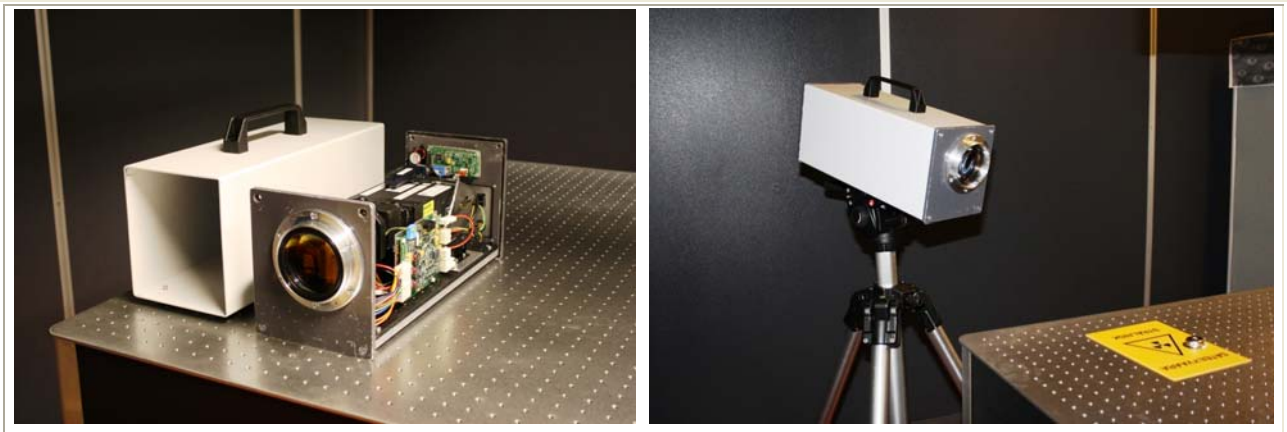

Fig. 3. HAUVA device for optical remote detection of alpha radiation.

3.2 Performance of HAUVA device

The performance of HAUVA was studied using a 1.2 kBq source that had equal activities of ^{239}Pu, ^{241}Am and ^{244}Cm. The surface emission rate of 620 alphas per second was verified by Deutscher Kalibrierdienst. This source yielded on average of 8 counts per second (cps) to the measurement channel at a distance of 40 cm. This is enough to overcome the noise level in darkness as both PMTs exhibit a dark count rate of less than 3 cps. However, in operative use the reliability of the detection is of utmost importance and therefore alarms should be made only on a strong basis.

In the field, the ratio of fluorescence and background signal is compared with Currie's critical level using the on-board computer. The critical level is set so that the result is a false positive only once in a million measurements with no real activity present. Given this confidence level, it is possible detect 100 kBq point sources at a distance of 40 cm using one second integration time in the presence of bright yellow fluorescent illumination or white LED illumination.

The spatial sensitivity of the demonstration device was studied by modelling and measurements. Figure 4a shows that the fluorescence signal is quite constant in the longitudinal direction at distances less than 30 cm. The almost constant signal strength in this range is beneficial to operational work, as the operator does not need to tune the distance to the surface very accurately to get reliable readings. At longer distances, the collection efficiency drops rapidly which is also predicted by the ray tracing model. The agreement between the model and the measurement is very good also in the transverse direction. This was studied by moving a point like alpha emitter transversally to the optical axis at the distance of 40 cm from the device. Figure 4b shows the measured and predicted curves. It can be concluded that the spatial resolution in the transverse direction is almost too good, as the 2 cm offset at the distance of 40 cm already halves the fluorescence signal. However, it means that point like alpha emitters can be localized very accurately.

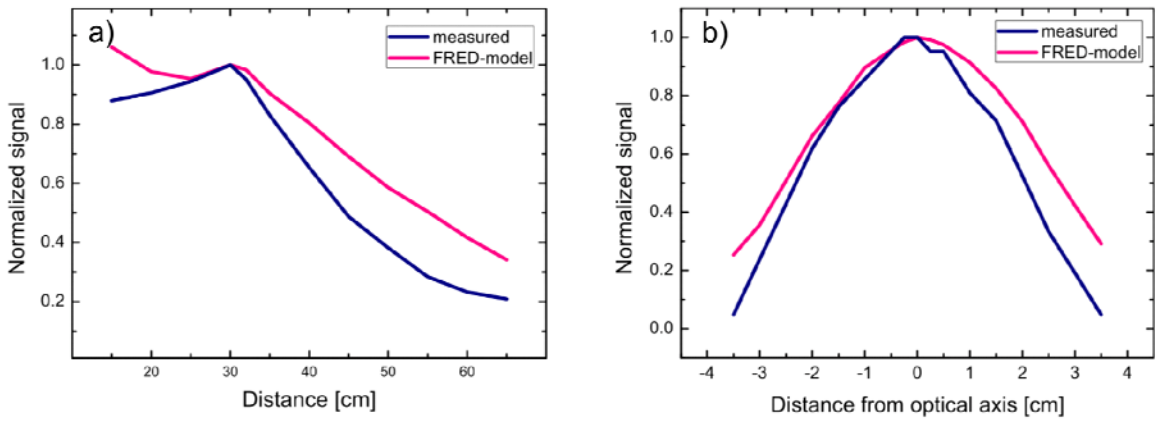

Fig. 4. ***a)*** *Normalized fluorescence signal of HAUVA device as function of distance from alpha emitter.* ***b)*** *Spatial distribution of normalized fluorescence signal of demonstration device as the point like alpha emitter is moved away from the optical axis. The result of optical ray tracing model (FRED) is shown as a reference in both graphs.*

3.3 Coincidence filtering

The coincidence detection was studied by measuring the time distribution of two photons induced by the same decay-event. A 13.0 kBq ^{241}Am source that had been extracted from an old smoke detector was used as an alpha emitter. The time distribution of fluorescence photon pairs was detected with fast photomultiplier tubes and a time correlated single photon counting (TCSPC) unit (Becker & Hickl GmbH). A distance of 20 cm was used between the alpha emitter and a collecting lens, and the average UV photon count rate at both channels was about 60 cps. Figure 5 shows a schematic representation of the measurement setup.

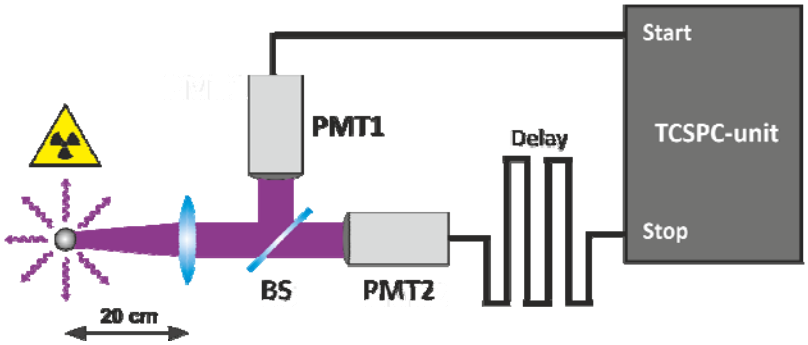

Fig. 5. Coincidence measurement with time correlated single photon counting unit. The fluorescence signal was gathered with a lens and then divided to both channels with a 50/50 beam splitter.

The time-to-amplitude conversion of the TCSPC-unit is triggered by a pulse from PMT1 and halted by the PMT2 pulse. The delay line is used to fine tune the centre of the distribution in the middle of the measurement window. The data was acquired with a 1024 channel multichannel analyzer and the time resolution of the measurement setup was verified to be better than 1 ns. The results are shown in Figure 6. The vertical axis shows the number of detected pulse pairs and in the top right corner is shown the background photon count rate of a single pmt.

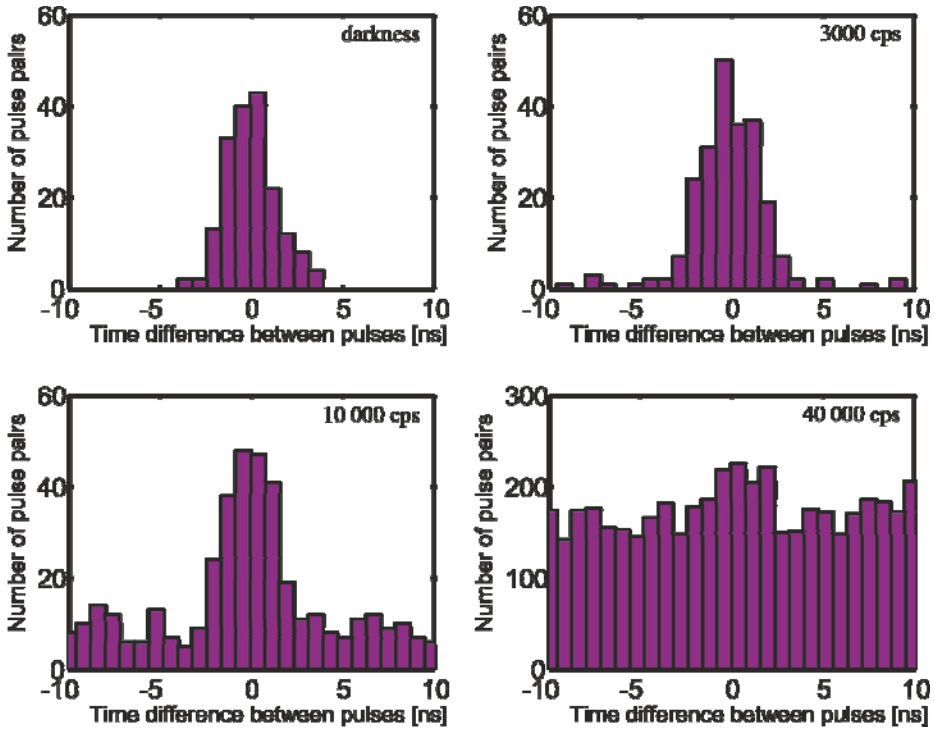

Fig. 6. *Time distribution of fluorescence photons induced by the same decay event. The background count rate of a single pmt is indicated in top right corner of each box. The first picture shows that most of alpha induced UV photons are emitted in short time window. The acquisition time for each measurement was 100 s.*

Figure 6 shows that all of the detected photon pairs arrive at the detectors in a short time window of 5 ns. The narrow distribution of photon pairs means that the fluorescence lifetime of a nitrogen molecule is well shorter than 5 ns, which is mainly due to fluorescence quenching by oxygen. The narrow distribution of the photon pairs is especially useful as the fast physical phenomenon enables a short coincidence time window. This enhances the detection significantly since the background photons are randomly distributed in time and therefore the probability of a background coincident event is low in a short time window. The performance of coincidence filtering was verified by measuring the coincidences in presence of three different levels of background lighting. It is shown in Figure 6 that the coincidence peak is still distinguishable when the background photon count rates in both detectors are on the order of 40 000 cps. Thus, the coincidence filtering enables the fluorescence detection even if the background lighting is about 500-times stronger than the nitrogen fluorescence signal.

4. Conclusion

The optical remote detection of alpha radiation was studied by two different methods. During the project, a portable demonstration device called HAUVA was designed and constructed. The device utilizes interference filters and a beam splitter to detect alpha-induced fluorescence signal and background lighting separately. The optics of HAUVA are designed to gather the maximum signal at a distance of 40 cm from the point like alpha emitter. The collected photons are counted with photomultiplier tubes and the data are evaluated using Currie's critical level. In carefully selected lighting, the device is able to separate 1 kBq alpha emitter from the background lighting with one second integration time. In operative use under bright artificial lighting, HAUVA can reliably detect 100 kBq sources in one second. Furthermore, it was shown that the interfering effect of the background lighting can be significantly reduced using coincidence detection. With this method, it was possible to separate the fluorescence signal from about a 500-times stronger background. The method will be studied in the future by combining spectral and coincidence filtering in the same system.

Acknowledgment

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