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Citation: Applied Physics Letters **102**, 254101 (2013); doi: 10.1063/1.4812338
View online: http://dx.doi.org/10.1063/1.4812338
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/102/25?ver=pdfcov
Published by the AIP Publishing

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(Received 30 April 2013; accepted 6 June 2013; published online 24 June 2013)

A standoff alpha radiation detection technique based on the physical mechanism of excited state absorption of air molecules was explored and is presented in this paper. Instead of directly detecting the radiation via measuring the intensity of radiation induced air fluorescence, the radiation is detected via the excited state absorption of alpha radiation excited/ionized air molecules. Both theoretical analyses and experimental verifications were conducted. The experimental results confirmed that the radiation could be detected via excited state absorption of radiation excited/ionized air molecules at a 10 m standoff distance, which was consistent with the theoretical analyses.
 [http://dx.doi.org/10.1063/1.4812338]

Due to the wide spread usage of radioactive materials in today's world, such as in nuclear power plants, radiation detection is critically needed. To minimize the risk of human involvement and radioactive contamination in the environment, it is preferred to detect the nuclear radiation at the standoff distance. Different types of materials and techniques were proposed and developed for detecting different types of radiations. For example, solid-state materials such as scintillation and semiconductor materials have been investigated for gamma radiation detection.[1,2] The physical mechanism of detection was based on the fact that both electrical and optical properties of the materials experience changes upon the exposure to gamma radiation.[3] For instance, gamma rays can produce a change in the density of charge carriers in semiconducting material, which can be used for detection.[4,5]

Although standoff detection is preferred, it is a challenging job. This is particularly true for alpha radiation detection due to the short emitting range in air. The conventional detection techniques require bringing a probe within a few centimeters of the suspect area to operate effectively. Recently, a remote detection technique based on the radiation induced air fluorescence was reported, in which the radiation detection was achieved by detecting the faint light emitted by radiation excited and/or ionized molecules.[6] The air fluorescence is a spontaneous emission which radiates uniformly within an entire 4π solid angle, so its intensity drops rapidly according to $1/r^2$ law, where r is the standoff distance. This intrinsic problem of propagation loss still limits the distance of the standoff detection.

To overcome the above fundamental limitation of $1/r^2$ propagation loss, in this paper, we report a standoff alpha radiation detection technique by exploring the physical mechanism of excited state absorption of radiation excited/ionized air molecules. Instead of directly detecting the faint light emitted by ionized/excited air molecules, a collimated probe beam with a wavelength matching the emitting wavelength of air fluorescence is used to detect the radiation via excited state absorption of air molecules. Since the probe beam is a collimated beam, its propagation does not suffer the fundamental limitation of $1/r^2$ propagation loss. This technology has a potential to realize the long range standoff detection for a stationary environmental monitoring.

It is well known that radioactive materials can generate nuclear radiation including γ–rays, β-particles, and α-particles. These radiations from radioactive materials can excite and/or ionize the air. Depending on the types and energies of the radiation, it may create excited state air molecules, ionized molecular ions, excited state atoms, and ionized atomic ions. The de-excitation from the higher energy excited/ionized states to lower energy states generates air fluorescence, which may cover a broad spectral range from ultraviolet to infrared.[7,8] Among these different types of emission spectra, the emissions from excited state nitrogen molecules can be employed to detect radiation. First, the main emission lines of nitrogen molecules are within the UV spectral region, which is useful for minimizing the influence of sun light for daylight detection operation.[9–11] For instance, one of the emission lines from C^3 II band to B^3 II band of nitrogen molecules is at the wavelength of 337 nm. Second, the lifetime of C^3 II band is on the order of nanoseconds, which is three orders of magnitude shorter than that of B^3 II band,[12] and thus excited state absorption can be easily achieved. This makes it effective to conduct the experiment of excited state absorption. Third, since 337 nm is also the output wavelength of nitrogen laser, it can be conveniently employed as the source of collimated probe beam.

Figure 1 illustrates the system configuration of the proposed standoff alpha radiation detection technique based on the excited state absorption of excited/ionized air.[13] In the operation, a collimated UV probe beam with a wavelength of 337 nm from a nitrogen laser is used to pass through an area affected by the radioactive materials. The radiations from the

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radioactive materials can first excite and/or ionize the nitrogen molecules in the air and pump them from the ground state to the excited states of the B^3 II band and the C^3 II band to generate air fluorescence. Since the radiation induced ionization is a continuous process, the population of carriers in the upper and lower excited states should be directly proportional to their respective carrier lifetimes, while the population related to the thermal energy are negligible because the energy levels of both B^3 II band and C^3 II band are much higher than the thermal energy at room temperature. Because of the big difference in their carrier lifetimes, the carrier population density in the B^3 II band should be much higher than that in the C^3 II band. This implies that the population density in the B^3 II band is an indication of the strength of ionization of nitrogen molecules and proportional to the intensity of radiation. As a result, when the UV probe beam passes through the air fluorescence induced by radiation, excited state absorption occurs as the carriers are excited from the lower excited state level to the upper excited state level. Note that the carriers raised to the upper excited state level also go through the spontaneous emission back into the lower excited state level. Since the spontaneously emitted photons are incoherent and radiate to the entire 4π solid angle in free space, the intensity quickly drops to an undetectable low level at long range standoff distance. This should have negligible influence on measuring the intensity of excited state absorption since the collimated probe beam does not suffer the inverse-square law of propagation loss and should be able to maintain almost the same intensity over the distance. Thus, the amount of excited state absorption can be used to quantitatively detect the radiation intensity at standoff distance by measuring the drop of the intensity of the collimated probe beam. It is important to point out that this proposed detection technique is suitable to monitor the radiation level at the known location. If the location is unknown, a scanning is required. Or, one can detect the backscattered light. However, it will suffer the $1/r^2$ propagation loss for the backscattered returning signal.

To quantitatively analyze the excited state absorption of radiation induced air fluorescence, the rate equations are employed to describe the population densities of the B^3 II and C^3 II bands, as given by

$$\begin{aligned} \frac{dN_C}{dt} &= \chi_{OC} - \frac{N_C}{\tau_C} - P_{em} \\ \frac{dN_B}{dt} &= \chi_{OB} + \frac{N_C}{\tau_C} - \frac{N_B}{\tau_B} + P_{em}, \end{aligned} \tag{1}$$

where N_C and N_B are the carrier densities on the C^3 II and B^3 II bands, respectively, χ_{OC} and χ_{OB} are the carrier excitation rates from the ground energy level to the C^3 II and B^3 II bands, respectively, τ_C and τ_B are the carrier lifetimes of these two energy bands, and P_{em} is the photon emission rate caused by radioactive transition from C^3 II band to B^3 II band. Under the quasi-steady-state mode, N_C and N_B are found by setting $\frac{dN_C}{dt} = \frac{dN_B}{dt} = 0$ in Eq. (1). Under this circumstance, one can obtain

$$\begin{aligned} N_C &= (\chi_{OC} - P_{em})\tau_C \\ N_B &= (\chi_{OC} + \chi_{OB})\tau_B. \end{aligned} \tag{2}$$

Notice that there is a much higher population density expected in the B^3 II band than that in the C^3 II band, $N_B \gg N_C$, primarily due to the fact that carrier lifetime of the B^3 II band is much longer than that of C^3 II band, $\tau_B \gg \tau_C$.

It is worth noting that according to exponential growth function, the power absorption of a probe beam at the transition wavelength between B^3 II and C^3 II bands can be expressed as

$$\frac{P_{out}}{P_{in}} = e^{[-(\sigma_a N_B - \sigma_e N_C)L]}, \tag{3}$$

where σ_a and σ_e are the absorption cross section of the molecules on the B^3 II band and the emission cross section of the molecules on the C^3 II band, respectively. N_B and N_C are the

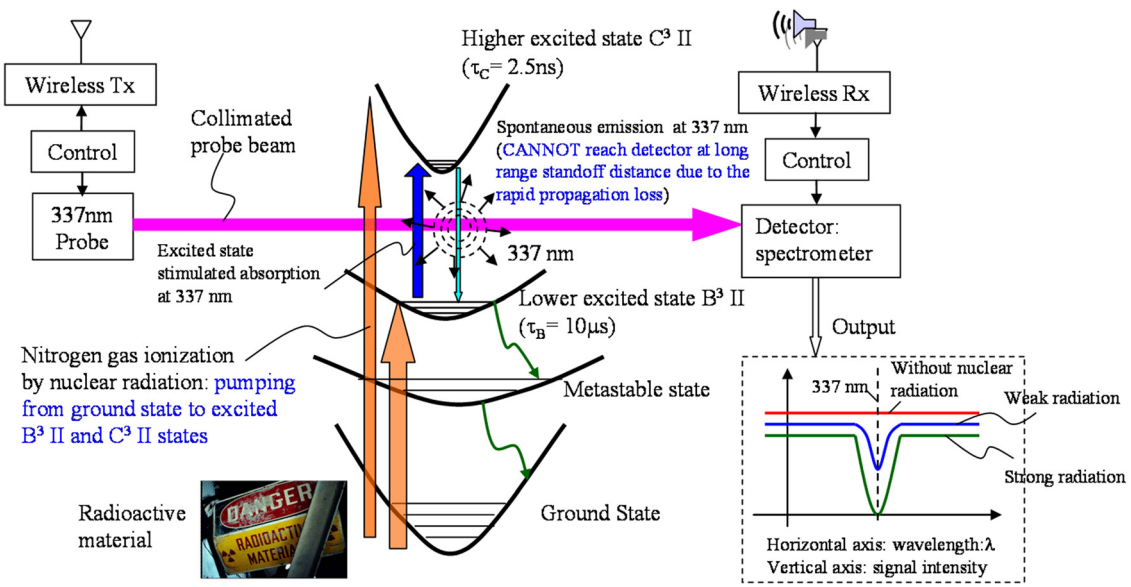

FIG. 1. A conceptual illustration of standoff detection of radiation via excited state absorption of radiation excited/ionized air (see Ref. 13).

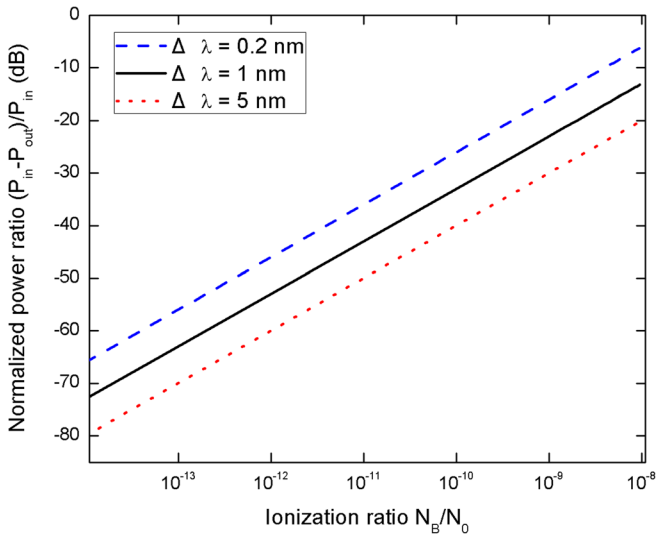

FIG. 2. The simulation results of optical power transmission ratio for a UV laser at wavelength of 337 nm as a function of ionization ratio with different optical signal spectral linewidths; blue dashed line: $\Delta\lambda = 0.2$ nm, black solid line: $\Delta\lambda = 1$ nm, and red dotted line: $\Delta\lambda = 5$ nm.

population densities of B^3 II and C^3 II bands, respectively, and L is the optical interaction length. Since a weak excitation of the probe beam does not drastically change the population densities, the values of N_B and N_C are mainly determined by the ionization rate and the carrier lifetimes of the energy bands. Also, as mentioned earlier, the lifetime of the B^3 II band is much longer than the C^3 II band, $\tau_B \gg \tau_C$, hence, the Eq. (4) can be simplified as

$$\frac{P_{out}}{P_{in}} = e^{[-N_e \sigma_a \tau_B L]}. \tag{4}$$

Further solving the Eq. (4), one can obtain the following power absorption approximation:

$$\frac{P_{out}}{P_{in}} = \left(1 - \eta N_0 \frac{e^2 \lambda^2 f_{ik} L}{4\varepsilon_0 m_e c^2 \Delta\lambda}\right), \tag{5}$$

where $\eta = \frac{N_B}{N_0}$ represents the ionization ratio at the B^3 II band that is directly proportional to the intensity of radiation, e is the electron charge, λ is the optical wavelength, f_{ik} is the line oscillation strength, ε_0 is the dielectric constant, m_e is the electron mass, c is the speed of light, and $\Delta\lambda$ is the signal spectral linewidth.

Based on Eq. (5), we calculated the relationship between the power absorption and ionization ratio. Figure 2 shows the normalized power absorption calculated as a function of ionization ratio, η, with different optical signal spectral linewidths. The calculation was based on a 337 nm wavelength probe beam over a 10 m optical path length with the assumption of the initial total density of nitrogen molecules, N_0 of $10^{19}/cm^3$. One can see that the normalized power ratio increases when the ionization ratio increases. Thus, from the quantitative relationship between normalized power absorption and ionization ratio, one can quantitatively measure the intensity of radiation by measuring the power absorption. The theoretical detection limit of the presented technology depends on the sensitivity and the signal to noise ratio of the detector as well as detection method. Since a −60 dB detection can be achieved by using a lock-in amplifier, one can detect an ionization ratio 10^{-13} with a spectral linewidth of 0.2 nm, as illustrated in Fig. 2. Furthermore, since such kind of level ionization ratio can be achieved by alpha source, the effort of this paper is focused on the alpha radiation detection.

In the experiment, two tube shape polonium-210 alpha sources (Model P-2035, NRD LLC) were used as the radiation sources. Each source had a maximum activity of 40 mCi, a decay energy of 5.3 MeV, and a half-life of 138.4 days. The reasons we chose the alpha source were due to its high ionization ratio, low penetration, and very short stopping distance in dense media. The alpha radiation usually travels about 3.8 cm in air which minimized its influence on the photomultiplier tube (PMT) photodetector. To determine the excited state absorption of air, a UV nitrogen laser manufactured by Spectra-Physics with an emission wavelength at 337 nm, a spectral bandwidth of 0.1 nm, and a pulse width of 4 ns for full width at half maximum was used as the probe beam light source. In order to measure the excited state

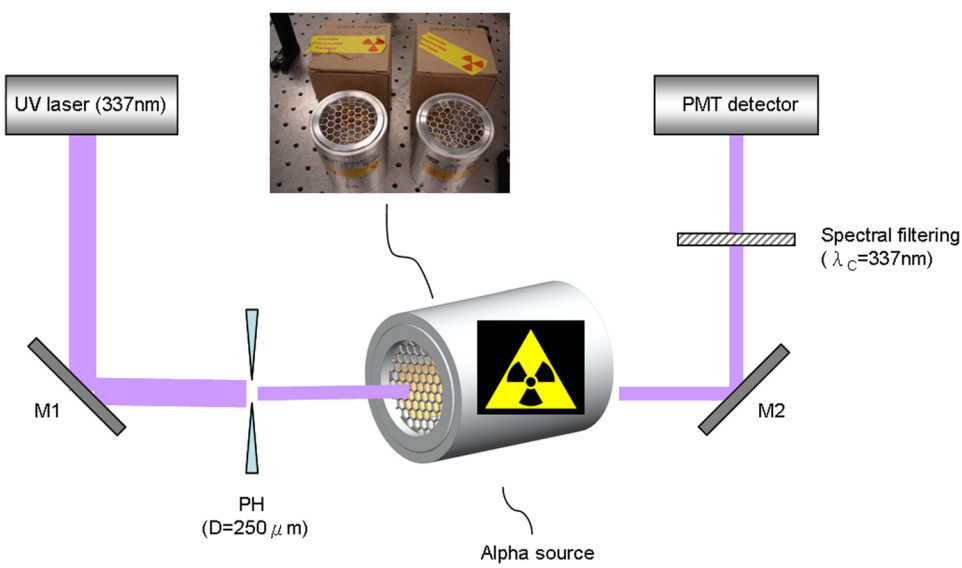

FIG. 3. A schematic illustration of the experimental setup used to conduct the excited state absorption of alpha radiation induced air fluorescence, including a 337 nm UV laser, two mirrors (M1 and M2), a pinhole (PH), a spectral filter centered at 337 nm, and a photodetector (PMT).

absorption, a Hamamatsu H7827 PMT was used as a photodetector due to its single photon sensitivity.

Figure 3 illustrates the experimental setup used to conduct the excited state absorption of alpha radiation induced air fluorescence. The 337 nm UV nitrogen laser probe beam passes through the air fluorescence region induced by the alpha sources. Due to the meshing nature of the stainless steel protection screen of the alpha source, the UV probe beam was first passed through a circular aperture of 250 μm in diameter so that it passed through the empty central region of the tube shape alpha source without blocking by the protection mesh screen. Inside this region, the air was excited and ionized by the radiation of alpha source. After that, the UV probe beam passed through a spectral filter with a central wavelength around 337 nm with a 7 nm pass bandwidth and a transmittance of about 10^{-5} in the visible region to block the background visible light, and then detected by the PMT photodetector. To achieve precise monitoring, the PMT photodetector needs to be well aligned with the laser probe beam.

To prove the concept of using excited state absorption of air for the alpha radiation detection, the intensity of probe beam was measured under different levels of alpha radiation: (1) 0, (2) 40 mCi (with one alpha source), and (3) 80 mCi (by cascading two alpha sources together). Figure 4 depicts the measured light intensity as a function of time under different levels of alpha radiation. The black solid line of Fig. 4 indicates the measured light intensity without any alpha source. On the same figure, the red dotted line indicates the measured light intensity with one Po-210 alpha source (40 mCi) and the blue dashed line indicates the measured light intensity with two cascaded Po-210 alpha sources (80 mCi). One can clearly see that the normalized value of red dotted line is lower than that of black solid line due to the alpha radiation induced excited state absorption. In addition, the normalized value of blue dashed line is even lower than that of red dotted line due to the increased excited state absorption of air by two cascaded alpha sources. This experimental result confirmed that the alpha radiation detection could be detected by the excited state absorption of radiation excited/ionized air. The proposed technique is suitable for measuring the relative intensity of radiation.

It should be noted that the magnitude of response, as shown in Fig. 4, is not linear due to the following two reasons: First, two alpha sources used in our experiment were not identical. The radiation level of one alpha source was slightly stronger than the other one. Second, the radiation follows Beer's law. Therefore, even when two alpha sources are identical, the response is not linear.

To further illustrate the standoff detection capability, the experimental setup, as illustrated in Fig. 3, was modified to a maximum 10 m distance between the probe beam UV source and alpha radiation source by reflecting the probe beam multiple times via mirrors. Figure 5 depicts the measured light intensity as a function of time with only one alpha source (40 mCi) for different standoff distances. The black solid line of Fig. 5 indicates the measured light intensity without any alpha source. Additionally, the red dotted, blue dashed, and green dashed-dotted lines indicate the measured light intensities under 40 mCi alpha radiations for 0 m, 5 m, and 10 m standoff distances, respectively. One can clearly see that the detected signal as a function of time is not sensitive to the separation distance between the light source and alpha radiation source. These experimental results confirmed that the detected signal was not influenced by the separation distance between the probe beam light source and alpha radiation because the collimated probe beam did not suffer $1/r^2$ propagation loss where r is the standoff distance. Similarly, it is also true that the detected signal was not influenced by the separation distance between the alpha source and the photodetector. Future development efforts will focus on the evaluation of the sensitivity of the proposed detection technique and increase it by employing advanced phase-locking system and multiple-path absorptions with a gated PMT photodetector.

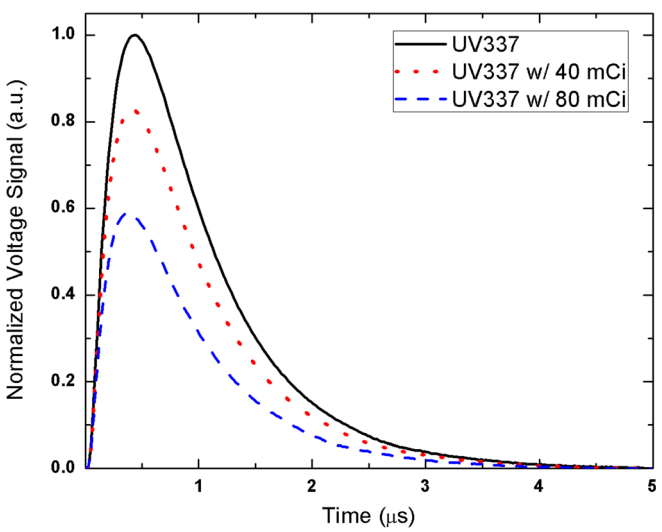

FIG. 4. The experimentally measured excited state absorption of alpha radiation excited air fluorescence as a function of time with different source fluence. Black solid line: without air fluorescence and red dotted and blue dashed lines: with air fluorescence under the alpha source fluence of 40 mCi and 80 mCi, respectively.

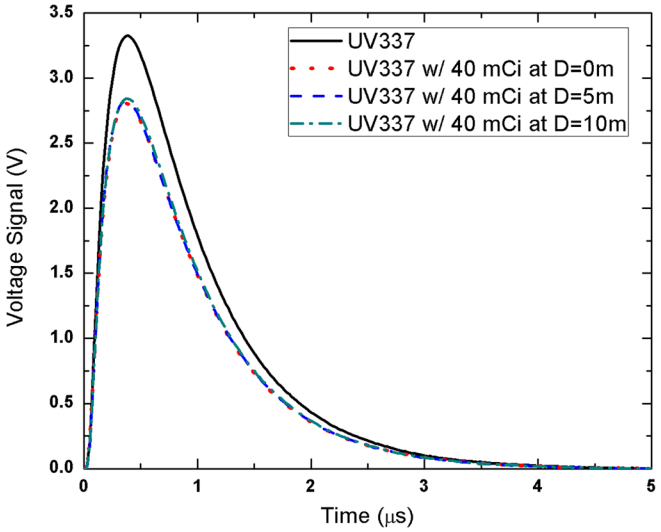

FIG. 5. The experimentally measured alpha radiation as a function of time with one alpha source for different standoff distances. Black solid line: without air fluorescence and red dotted, blue dashed, and green dashed-dotted lines: with air fluorescence under 40 mCi alpha radiations for 0 m, 5 m, and 10 m standoff distances, respectively.

In summary, a physical mechanism of alpha radiation detection based on excited state absorption of radiation excited/ionized air was investigated. The detailed description on technical approach and theoretical modeling were provided. The proposed radiation detection physical mechanism was also confirmed by the experiment. In the experiment, polonium-210 alpha sources were used as the radiation source, a nitrogen laser with an output wavelength of 337 nm was used as the collimated probe beam source, a PMT photodetector was used to measure the excited state absorption of the probe. Indeed, the experimental results validated that the excited state absorption increased as the fluence of radiation increased. The major advantage of the proposed detection mechanism is the capability of long range standoff detection because the collimated probe beam does not suffer the $1/r^2$ propagation loss. Thus, this proposed detection technique not only advanced the technique of radioactive material detection, but also provided an effective tool to safely and remotely monitoring the nuclear facilities such as nuclear power plans.

Authors greatly acknowledge the partial financial support of this work by DTRA Basic Research and Applied Science Program. The views and opinions of its authors do not necessarily state or reflect those of the U.S. Government or any agency thereof. Reference to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the U.S. Government or any agency thereof.

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