

Development of Fluorescence Spectral Imaging for Location of Uranium Deposited on Surfaces – 9163

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ABSTRACT

Since the 1980's, depleted uranium (DU) has been the primary material used by the US military in armor-piercing rounds. Domestic firing ranges that have been used for DU munitions training purposes are located around the country and have varying extents of contamination by other types of projectiles. A project is underway to develop a set of sensors to locate expended DU rounds and to process soil and debris to recover the material. In the environment, metallic DU readily oxidizes to form uranium compounds that contain the uranyl (UO_2^{+2}) moiety. For more than a hundred and fifty years, it has been known that when illuminated with ultraviolet (UV) light, uranyl compounds exhibit characteristic fluorescence in the visible region (450 – 650 nm). We report our efforts to develop a transportable, quantitative Fluorescence Spectral Imaging (FSI) system to locate and quantify uranyl compounds dispersed in soils and on other surfaces on domestic firing ranges; this system can also be utilized to monitor excavation of DU munitions and separation of uranyl compounds from soils. FSI images are acquired by illuminating a surface with a UV light and using a narrow bandpass filter on a camera, recording an image of the resulting fluorescence. FSI images provide both spatial and spectral information. The FSI system is described and its performance characterized in the field and also by using field samples. The development and characterization of an improved transportable FSI system is presented. The applicability of this system for detection of uranium compounds deposited on surfaces for Decontaminating and Decommissioning (D&D) activities is discussed.

INTRODUCTION

The US military has employed a variety of dense metal alloys as armor piercing projectiles. These have included machine hardened steel, tungsten carbide, an improved tungsten alloy, a four-component alloy containing depleted uranium, and a depleted uranium-titanium alloy (DU). Use of DU for penetrating enemy armor (tanks) has become the standard since the 1980s and is currently the superior material for that application.

At least fourteen different locations in the US have been used by the military for testing and training purposes employing DU ammunition. The contaminated areas at each of these test sites vary in size, based upon how targets were arranged. Catch boxes with a footprint of approximately one acre (0.4 hectares) have been constructed for certain applications. These are also open firing ranges that have been used by both artillery and aircraft and are as large as thousands of acres (hundreds of hectares).

The ability to defeat armor using kinetic energy penetrator rounds constructed using depleted uranium represents a tactical advantage to the US military on the battlefield. It is critical to maintain the capability to train with currently developed DU munitions and to test DU rounds undergoing improvement or development. The location, removal and/or containment of DU residues generated during test and training operations is an integral part of range management.

Range management practices often call for locating and recovering fired penetrators either to prevent metal on metal impacts or to comply with Nuclear Regulatory Commission (NRC) license requirements. The recovery process can be complicated by the fact that metallic uranium is reactive, oxidizing once it

has been deposited in soil. The recovery process needs a variety of measurement systems for precisely locating the DU material, facilitating recovery of DU with a minimal quantity of contaminated soil, and controlling process equipment used to separate DU from soil and associated debris.

The Army Range Technology Program (ARTP) initiative was established for the purpose of (1) developing technologies for the sensing of DU munitions residues in soils and water and (2) for the physical separation of DU from soils/water at testing and training facilities. The ability to locate, remove, and/or contain DU following use on training ranges without generating large quantities of waste is critical to the sustainable operation of testing and training facilities.

US fighting forces benefit from having access to the most effective weapon systems that developers, testers, and military personnel can assemble. Range management benefits from the tools developed under this program for maintaining sustainable testing and training facilities for munitions that contain depleted uranium. In addition, the ARTP program will reduce human and environmental risks by developing and applying scientifically defensible low cost range management practices.

Mississippi State University's contribution [1,2] to the ARTP effort has been focused on developing the best detection and measurement technologies that can rapidly and/or accurately detect and locate fired DU penetrators. It should be pointed out that much of the previous fieldwork identifying locations of radiological contamination has been conducted for the purpose of designing remedial actions [3-5]. However, there is a significant difference between conducting environmental sampling for the purpose of remediation of a site as compared to surveying a site for the purpose of recovering fired DU projectiles. The latter case requires a much more precise determination of the vertical and horizontal location of DU material so that the smallest amount of soil possible will be removed during excavation. The potential for DU corrosion products to migrate away from the metallic DU project must be taken into account.

FLUORESCENCE SPECTRAL IMAGING

Most of the oxidized uranium compounds formed when uranium metal is deposited in soil contain the uranyl (UO_2^{+2}) moiety. Uranyl compounds have a characteristic yellow color and have been long known to emit characteristic fluorescence in the 450-650 nm spectral region when excited in the ultraviolet or short-wavelength visible region [6,7]. Hence uranyl fluorescence provides a means of locating DU penetrator residues during site screening, excavation, and during soil processing. The fluorescence can be excited either with a laser or with a high-intensity lamp. Lasers have the advantage of higher intensities and hence the ability to detect lower concentrations; high-intensity lamps (such as mercury (xenon) Hg(Xe) lamps) have the advantages of being able to illuminate a larger area and easier on-the-spot repairs. When the fluorescence is detected by using a narrow bandpass filter on a camera, the resulting spectral image provides not only spectral information about the emitting species, but also information about the spatial distribution and location of the emitting species.

Because of the characteristic uranyl fluorescence, Fluorescence Spectral Imaging (FSI) has the potential to sensitively monitor the presence and amount of uranyl present. However, it is limited to uranyl that is present on the surface and cannot detect buried DU. In addition, it cannot on the basis of fluorescence distinguish metallic DU from other objects present (such as stones); however, application of image pattern recognition would enable the detection of spent metallic DU penetrators.

PROOF-OF-CONCEPT DEMONSTRATION AT YUMA PROVING GROUND

A prototype uranyl FSI detection system was assembled from available components. A 150-W mercury (xenon) [Hg(Xe)] arc lamp (Oriel) was chosen as the illumination source for proof-of-concept demonstration at Yuma Proving Ground (YPG), AZ in June 2007. This selection was made by recording

fluorescence spectra at ICET produced by a 150-W Hg(Xe) arc lamp, a 1000-W Hg(Xe) arc lamp, and a 1000-W quartz tungsten halogen (QTH) incandescent lamp. These experiments were performed using a known uranyl compound (uranyl acetate). The resulting fluorescence was acquired by a UV-grade fiber optic cable with a pick-up lens. The fluorescence signal was wavelength dispersed by a 0.5-m imaging triple-grating monochromator and detected by a blue-sensitive, thermoelectrically cooled, data acquisition intensified charged-coupled device (ICCD) camera. The spectrum of Hg(Xe) arc lamps in the ultraviolet (UV) contain both broadband, structureless “white light” emission and atomic emission, primarily from atomic mercury transitions. The QTH incandescent lamp is a broadband, structureless “white light” source. Since both types of lamps produce light over a very broad spectral range, it was necessary to restrict the wavelength region used for illumination to the UV region where uranyl compounds absorb so that the spectral images detected would contain only uranyl fluorescence and not light from the lamp (which is likely to be more intense and hence mask the uranyl fluorescence). This was accomplished by using a small monochromator as a tunable bandpass filter. The light was directed onto the sample using a 10 cm x 10 cm (4”x4”) aluminum first-surface mirror. In order to allow the light beam exiting the monochromator to be as large as possible in order to illuminate the largest possible area, the exit slits removed from the monochromator; the illuminated area was approximately 2.5 cm x 10 cm (1” x 4”) at a distance of approximately 1.2 m (4’) from the exit of the monochromator. A blocking plate was mounted on the exit of this monochromator so as to selectively block the long-wavelength portion of transmitted spectral region, thus allowing the maximum illumination bandwidth without extending into the uranyl fluorescence spectral region (450 – 650 nm). Uranyl compounds absorb light at wavelengths shorter than 500 nm. Wavelength regions “centered” on 400 nm and on 443 nm were chosen for investigation. Longer wavelength regions were not used since they would overlap the uranyl fluorescence region (450 – 650 nm) and hence make detection of uranyl fluorescence difficult. Wavelengths shorter than 400 nm were not used since they are not visible to the human eye and hence it was not possible to visually determine the location of the illuminated area.

The QTH lamp did not produce detectable fluorescence using our standard experimental conditions for the detection system. Both the 150-W Hg(Xe) arc lamp and the 1000-W Hg(Xe) arc lamp produced readily detectable fluorescence with our experimental setup. The 1000-W Hg(Xe) could not be used at full power since it could and did thermally damage nearby optical components; the maximum power used was approximately 650 W and most of the tests were performed with about 400 W power. Since the 150-W Hg(Xe) arc lamp system is more compact than the 1000-W Hg(Xe) arc lamp system and since the 150-W Hg(Xe) arc lamp can easily produce detected uranyl fluorescence, the 150-W Hg(Xe) arc lamp was chosen for field demonstration at YPG.

Fluorescence spectral images (FSI) were acquired with a variety of cameras, viewing the objects of interest through bandpass filters mounted in a motorized filter wheel. The 10-nm full-width-at-half-maximum (FWHM) bandpass filters used for this effort had center wavelengths of 550 nm, 589 nm, 600 nm, and 800 nm; these filters were chosen because of their availability. Uranyl compounds do not fluoresce at 800 nm so images recorded at 800 nm were utilized for background subtraction image processing. Initially images were recorded in our ICET laboratory using a monochrome CCD camera. The prototype FSI system was extensively tested at ICET using uranyl acetate and also DU penetrators provided by YPG. For demonstration at YPG, a more sensitive, blue-enhanced ICCD camera was utilized.

For the YPG demonstration, the prototype FSI system was deployed using a wheeled cart. The image acquisition camera was either deployed mounted to the cart, facing vertically downward or from a tripod. A canopy was placed over the cart to reduce glare on the computer monitor. The images were stored on a desktop computer. [A desktop computer rather than a laptop computer was utilized because a laptop could not internally contain the required image acquisition board; future efforts will use a ruggedized portable computer.] The image acquisition software was developed in house at ICET based upon an ANSI-C development platform (LabWindows/CVI, National Instruments). The entire prototype FSI system was

mounted on a movable cart. The photograph on the left in Figure 1 shows the prototype FSI system and two other ICET imaging-based systems at YPG. The photograph on the right in Figure 1 shows two spent DU penetrators (as can be seen by the characteristic yellow of uranyl compounds) on top of the soil at YPG. A total of 124 FSI images were recorded at YPG.

Figure 2 shows typical FSI images. Of the filters used, the 550-nm filter produces images with higher intensity/brightness for FSI detection; this is consistent with uranyl fluorescence where the uranyl emission typically occurs near 488 nm, 509 nm, 533 nm, 560 nm, and 587 nm [6,7]. Analysis of the FSI images acquired at YPG were post-analyzed at ICET. A variety of image processing approaches and representation formats were investigated using MATLAB software (with imaging toolkit, MathWorks) (as illustrated in Figure 3).

The observed intensity is greater for 443-nm excitation than for 400-nm excitation, as shown in Figure 4. This difference in recorded intensity may be due to (1) differences in light intensity produced by the Hg(Xe) lamp in these two wavelength regions and/or (2) to the “443-nm” excitation interval partially overlapping the camera filter bandpass spectral region. Analysis of other FSI images indicates that “400-nm” excitation provides better discrimination than does “443-nm” excitation. Thus choice of excitation wavelength depends upon whether image intensity or image discrimination is of greater concern.



Figure 1. Left photograph shows image acquisition of fired DU penetrator by FSI and by other ICET imaging techniques near DU catch box at Yuma Proving Ground, AZ. Photograph on right shows two DU penetrators *in situ*; a red plastic marker flag is included for identification.

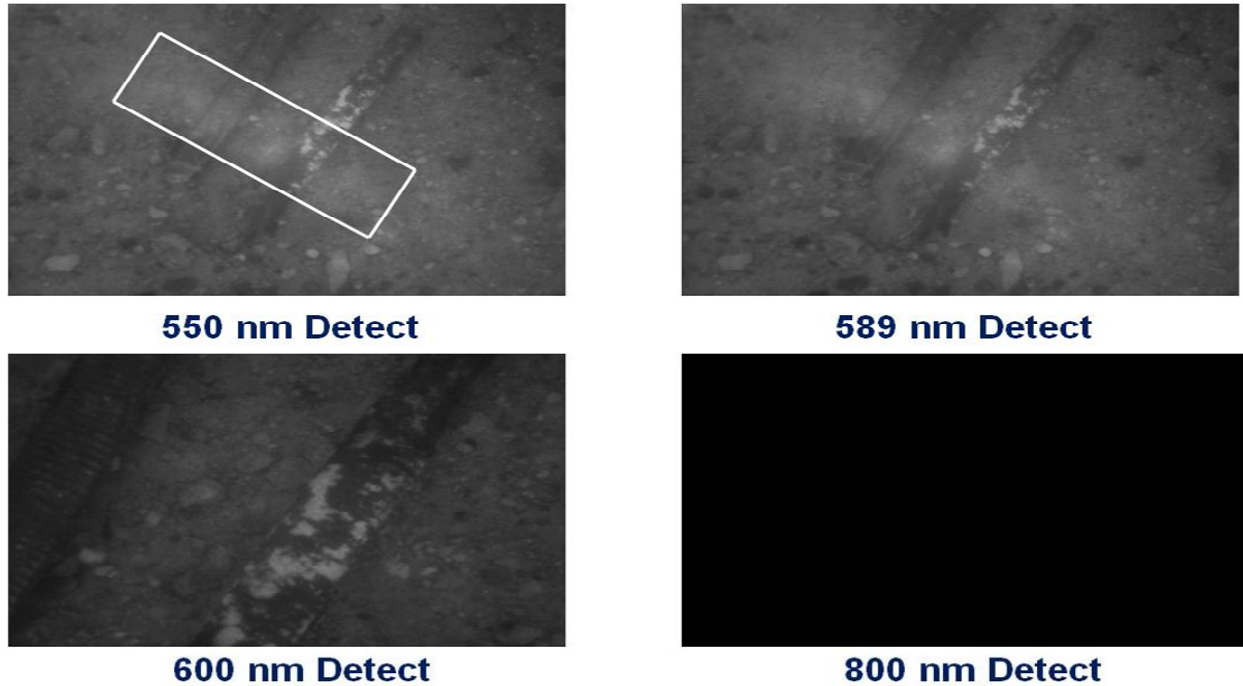


Figure 2. FSI images recorded of YPG DU penetrators on June 8, 2007. The white box in the upper left photograph indicates the location of the 400-nm light beam from the Hg(Xe) arc lamp. Two DU penetrators are present: one with a significant amount of uranyl compound and the other with little.

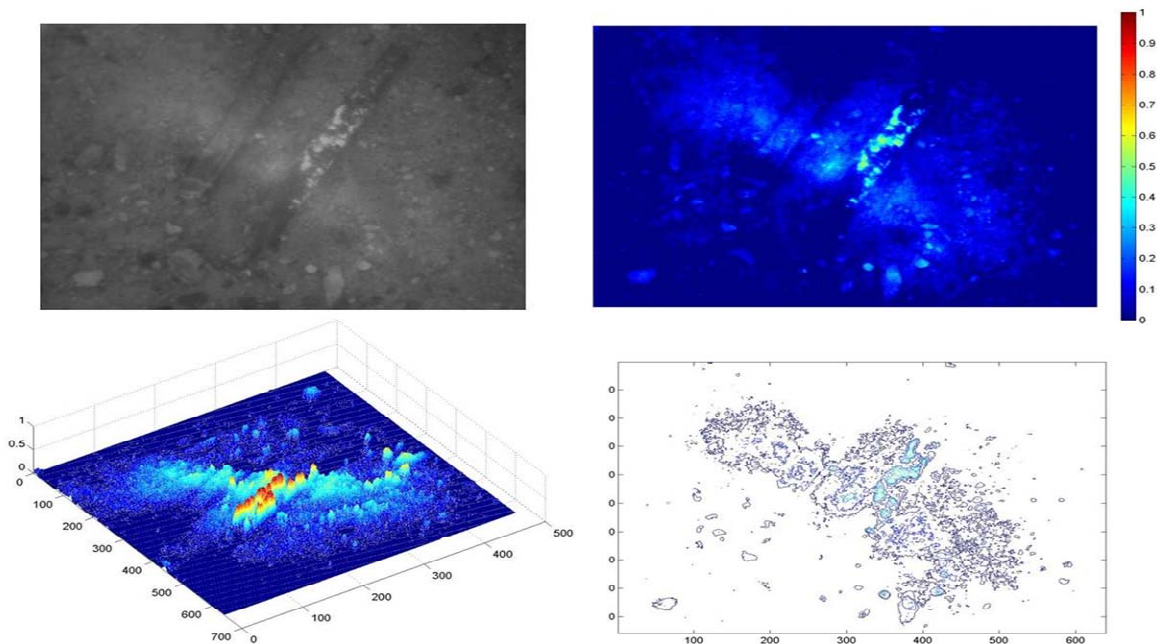
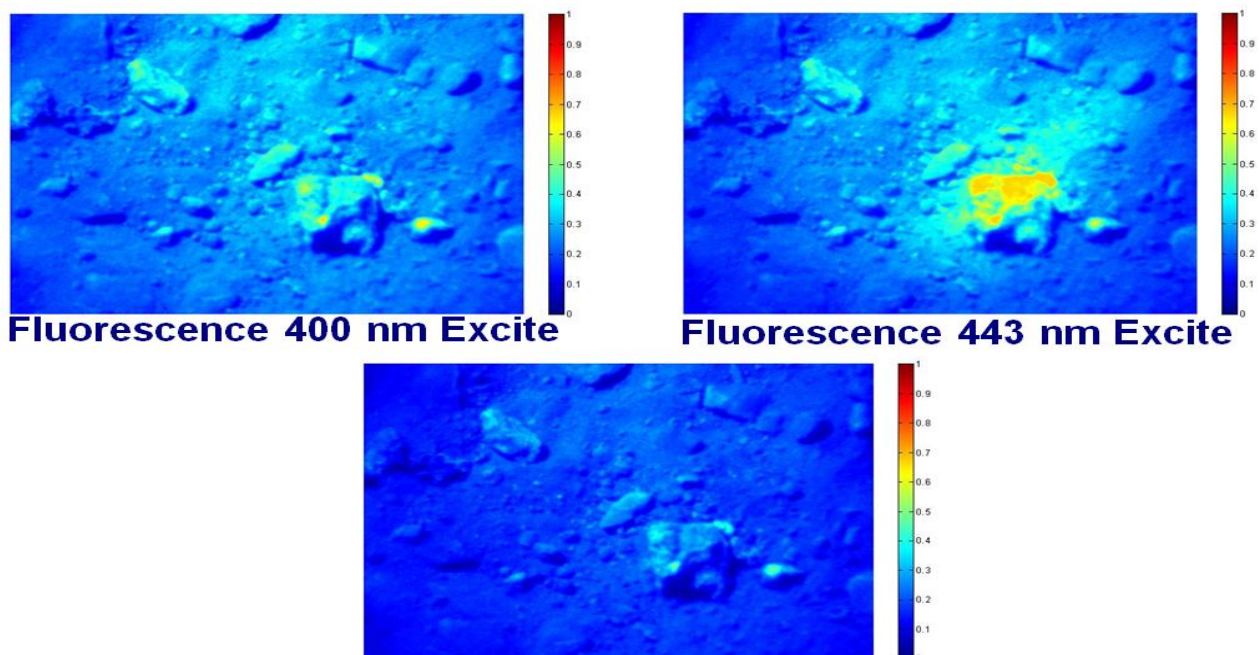


Figure 3. Different presentations of same FSI image (550-nm detection) presented in Figure 2. The monochrome image on the upper left is the original “raw” image.



Spectral Imaging

Figure 4. False-color images (550-nm detection) of oxidized remains of spent YPG penetrator. The top two images were FSI images recorded using a 150-W Hg(Xe) lamp. The bottom image is a spectral imaging (SI) image recorded using an incandescent light bulb for illumination.

Figure 4 also compares FSI images with those obtained without fluorescence excitation using traditional Spectral Imaging (SI) detection. As is shown by Figure 4, fluorescence spectral imaging (FSI) is more sensitive for detecting uranyl compounds than is spectral imaging (SI). Thus, FSI can provide a more comprehensive examination for the presence of uranyl compounds, enabling uranyl compounds to be detected at lower concentrations.

SUBSEQUENT FSI DEVELOPMENT EFFORTS

Based upon the successful demonstration of FSI and other techniques at YPG, ICET has obtained additional funding to develop improved versions of our DU detection and location systems. For the FSI effort, new equipment has been purchased, including a modified diverging “flood” illumination system (Newport) for illuminating a much larger area than we were able to do so previously; also the use of a monochromator for selecting the excitation wavelength band is avoided. The flood illumination system uses a 1-kW Hg(Xe) lamp for light, a dichroic mirror for preferentially reflecting light in the 280-400 nm region, and then a tempered glass filter (Schott UG5) to preferentially block light in the 450-600 nm region where uranyl compounds fluoresce. The light from the flood illumination system is directed vertically downward. The size of the illuminated area as a function of distance from the exit window has been characterized. At a distance of 117.5 cm (46¼”) from the exit window, the flood system illuminates a rectangular region 65 cm x 70 cm (25½” x 27½”).

In order to characterize the output of the flood illumination system, a spectrum has been recorded. The spectrum reveals that light in the 410- to 525-nm region is very efficiently blocked. However, the mercury 546-nm, 577-nm, and 615-nm transitions are partially transmitted by the flood light filtering system. Consequently, it is desirable to monitor fluorescence from the uranyl 488-nm and 509-nm emission bands

rather than the 560-nm and 587-nm emission bands in order to avoid detecting light from the flood illumination system.

We are currently engaged in an extensive characterization/development/validation effort. This includes: (i.) characterization of the spatial uniformity of the flood illumination system as a function of flood-to-surface distance; (ii.) characterization of FSI detection of uranyl as a function of position within the illuminated area; (iii.) characterization of factors that may lead to false positives, such as transmission of camera filters and signal from YPG soil; (iv.) determination of “standard” camera operating conditions; (v.) characterization of FSI detection of uranyl as a function of both uranyl concentration and position within the flood-illuminated area; and (vi.) determination of limits of detection. The wheeled cart used for demonstration at YPG during June 2007 will be modified for a more challenging demonstration at YPG during early 2009.

POTENTIAL APPLICATION OF FSI TO DOE D&D APPLICATIONS

When the Fluorescence Spectral Imaging (FSI) system has been fully developed and characterized, it has the potential for application to US Department of Energy (DOE) Decontaminating and Decommissioning (D&D) activities. Because metallic uranium is so chemically reactive, the oxidation of metallic uranium quickly occurs whenever metallic uranium is exposed to the environment. The oxidized uranium compounds produced typically contain the uranyl (UO_2^{+2}) moiety. Because of uranyl’s characteristic fluorescence, it is straightforward to use Fluorescence Spectral Imaging (FSI) to cost effectively and sensitively detect the presence and spatial distribution of uranyl compounds on contaminated surfaces. Because the isotopes of uranium all have very long half-lives (ranging from 10^5 to 10^9 years), long counting times are required for radiological detection of small quantities of uranium. The use of cameras that detect gamma-ray emission suffers from the fact that uranium has a very large number of rather weak gamma emission transitions so relatively large concentrations of uranium are required for detection.

All techniques have their strengths and weaknesses. FSI’s strengths are (i.) its sensitivity, (ii.) its ease of use, and (iii.) that it is less expensive than other techniques. FSI’s weaknesses are (i.) that it is not sensitive to uranium that is not present in the form of a uranyl compound and (ii.) that it can only monitor the surface and is not sensitive to any uranyl present beneath the surface.

SUMMARY

We have successfully demonstrated *in situ* a first-generation, transportable Fluorescence Spectral Imaging (FSI) system for locating uranyl compounds dispersed in soils and on other surfaces of a domestic firing range. FSI images provide both spatial and spectral information. FSI images are acquired by illuminating a surface with a UV light and using a narrow bandpass filter on a camera, recording an image of the resulting fluorescence. This system has the potential to also be utilized to monitor excavation of DU munitions and separation of uranyl compounds from soils. For the first-generation prototype FSI system, images acquired with “443-nm” radiation have greater intensity than those acquired with “400-nm” radiation; however, use of “400-nm” radiation permits better discrimination. FSI images enable smaller amounts of uranyl compounds to be detected than does traditional Spectral Imaging (SI) detection.

A second-generation, transportable, FSI system is currently under development and is being extensively characterized so that quantitative determinations can be made of the amount of uranium present. In order to characterize the output of the new flood illumination system, a spectrum has been recorded that reveals that light in the 410- to 525-nm region is very efficiently blocked. However, the mercury 546-nm, 577-nm, and 615-nm transitions are partially transmitted by the flood light filtering system. Consequently, it is desirable to monitor fluorescence from the uranyl 488-nm and 509-nm emission bands rather than the 560-nm and 587-nm emission bands in order to avoid detecting light from the flood illumination system.

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