TECHNICAL NOTE

Norimitsu Akiba,¹ Ph.D.; Naoki Saitoh,¹ B.S.; and Kenro Kuroki,¹ Ph.D.

Fluorescence Spectra and Images of Latent Fingerprints Excited with a Tunable Laser in the Ultraviolet Region

ABSTRACT: Fluorescence spectra of sebum-rich latent fingerprints were studied with a tunable laser for non-destructive fingerprint detection without chemical treatment. The tunable laser consists of a nanosecond pulsed Nd-YAG laser and an optical parametric oscillator (OPO) crystal. The fluorescence spectra and images were measured at various excitation wavelengths in the ultraviolet region by the time-resolved fluorescence method. We have previously reported that a typical fluorescence spectrum of fingerprints consists of two peaks located at *c*. 330 and 440 nm. In order to determine the wavelength of optimal excitation, excitation at 280 nm. The images of latent fingerprints on white papers were also measured at the clearest image was obtained with excitation at 280 nm. The influence of continuous irradiation on the fluorescence of fingerprints was measured at the optimal excitation, whereas the 440 nm peak, which was weak at first, increased gradually.

KEYWORDS: forensic science, fingerprints, fluorescence spectroscopy, ultraviolet rays, time-resolved spectroscopy, tunable laser

Fingerprints provide important information during criminal investigations. Many reports on fingerprint detection have been published. Most have focused on experiments using chemicals, such as ninhydrin; in addition, many studies using a laser have also been reported. Dalrymple et al. first demonstrated the detection of fingerprints using inherent fluorescence with a laser, in 1976 (1). Subsequently, many reports have been published on fingerprint detection using lasers (2,3). Most of the earlier studies on the detection of fingerprints with a laser were done using pre-processing procedures with various chemicals before detection to enhance sensitivity because the fluorescence of fingerprints was very weak. Bramble et al. performed nondestructive inherent fluorescence visualization of latent fingerprints on white paper without chemical treatment using a Nd-YAG laser in 1993 (4,5). They took photographs of fingerprints using a bandpass filter in the ultraviolet region. The forensic detection of untreated body fluid using short wavelength UV luminescence was also performed by Springer et al. in 1994 (6).

Time-resolved spectroscopy is a method which utilizes the difference in fluorescence lifetime between a substrate and a sample. Fluorescence from a sample was selectively observed by timeresolved spectroscopy. This method was useful for laser fingerprint development on strongly luminescent substrates. A time-resolved luminescence imaging system was suggested by Murdock and Menzel (7). However, time resolution in their experiment was millisecond or microsecond with a mechanical chopper. Seah et al. demonstrated the nano-second time-resolved imaging technique for latent fingerprints in 2004 (8). However, they used fluorescent powders to enhance sensitivity, and observed fingerprints using an Ar-ion laser in the visible region. We previously reported the

fluorescence spectra of fingerprints using nano-second time-resolved spectroscopy with 266 nm UV irradiation using a Nd-YAG laser without chemical treatment in 2000 (9). However, the excitation wavelength was fixed to 266 nm in 9. In this paper, we studied the spectra of fingerprints and the dependence of fluorescence on the excitation wavelength (excitation spectrum) by using a tunable laser. The aim of this study was to clarify the optimal excitation wavelength for detecting fluorescence of fingerprints in the ultraviolet region and to obtain fluorescence images of fingerprint with the optimal excitation wavelength. We also studied the influence of continuous irradiation on the fluorescence of fingerprints at the optimal excitation.

Methods

Experiments were done using the following system. The tunable laser system consists of a pulsed Nd-YAG laser (Continuum, Powerlite Precision II 8010), OPO (Optical Parametric Oscillator) with a Doubler (Continuum, Panther EXST-1), a cooled CCD camera with an image intensifier (ICCD, Roper Scientific, PI-MAX 1K-RB-FG-43) and a spectrometer (Acton Research Corporation, Spectra-Pro-300i). Details of the experimental setup are described in 8. Nano-second time-resolved fluorescence spectra can be measured with this system. The gating control of the exposure time of the ICCD camera and the delay time of observation were done by a programmable timing generator (PTG) through a personal computer. Fluorescence was collected with a UV lens (UV-Nikkor 105 mm) and put into the spectrometer with a grating monochrometer. Spectra were measured by the ICCD camera attached to the spectrometer with the gating control. The spectra obtained were digitized in 16 bit. In order to suppress noise, the ICCD was cooled to -20°C. In addition, the background spectrum was subtracted at each measurement. In case of observation of fingerprint fluorescence, it was necessary to accumulate many spectra as the fluorescence intensity is weak. As shortwave UV is hazardous to the eyes and skin, and as shortware

¹Second Forensic Science Division, National Research Institute of Police Science, 6-3-1 Kashiwanoha, Kashiwa, Chiba 277-0882, Japan.

Received 18 Aug. 2006; and in revised form 24 Feb. 2007; accepted 1 April 2007; published 7 Sept. 2007.

UV has been reported to damage DNA evidence (10), care must be taken when using shortwave UV systems.

In our experiments we used excitation wavelengths from 220 to 310 nm, as the laser power is very weak in the wavelength ranges of 220 nm, or less, and from 310 to 400 nm. The repetition rate of the laser pulse was 10 Hz and the time width of the individual pulses was *c*. 5 nsec. The intensity of each fluorescence spectrum was normalized by an averaged power value acquired from a laser power meter. The delay time was from 4 to 10 nsec. Samples were prepared as follows. A fingerprint was pressed on a nonfluorescent quartz slide glass or on high-grade white paper of 64 mg/cm² in weight after wiping the forehead or nose with fingers. Before impression, the finger was washed with soap and dried in the air.

Results and Discussion

A typical fluorescence spectrum of a fingerprint has two main peaks, when the excitation wavelength is 266 nm (11). One is located at c. 320-340 nm (peak A) and the other at c. 430-460 nm (peak B). We first measured the fluorescence of fingerprint residue on quartz glass, changing the excitation wavelength to find the optimal excitation wavelength. The resultant fluorescence spectra of peak A are shown in Fig. 1. The delay time was 4 nsec and five spectra are shown out of ten spectra measured. The intensity of fluorescence in a vertical axis was normalized for each laser power value. The intensity of peak A becomes maximal with excitation at 280 nm. The excitation spectra of peak A are shown in Fig. 2. Results for the delay time of 6 and 10 nsec are also shown in this figure. As shown in Fig. 2, there are two peaks, at 230 and 280 nm, in the excitation spectra for all delay times. The peak intensity at 280 nm is higher than that of the peak at 230 nm. This means that the optimal excitation wavelength is c. 280 nm. The intensity of fluorescence of peak A becomes minimal at 250 nm. Fluorescence excited at 230 nm was slightly weaker than at 280 nm, but this does not mean that the excitation at 230 nm is inappropriate. The excitation spectra of peak B were measured but reproducible results were not obtained, because the laser power at each excitation fluctuated more than that of peak B. Therefore, the excitation spectra of peak B did not show reproducibility in this experiment.

Next, fluorescence images of fingerprint on high quality white paper were measured. As white paper has strong fluorescence from 400 to 500 nm, which overlaps with peak B of fingerprints, it is necessary to suppress fluorescence of paper to obtain



FIG. 1—Fluorescence spectra of peak A. The intensity of peak A is the maximum at 280 nm.



FIG. 2—Excitation spectra of peak A.

fluorescence images of fingerprints, which leads to the reduced intensity of peak B. Imaging was performed using peak A, as peak A does not overlap with the fluorescence of paper. For imaging, this is effective with the use of a band-pass filter. The center wavelength of the band-pass filter is 360 nm, which has a maximum transmittance of 70% and full width at half maximum (FWHM) of 60 nm. Moreover, remnant fluorescence of paper is suppressed by the time-resolved spectroscopy which utilizes the difference in fluorescence lifetime. Figures 3a,b,c show fluorescence images with excitation at 230, 280 and 300 nm, respectively. The clearest fluorescence image of a fingerprint was obtained with excitation at 280 nm, which agrees with results obtained from the excitation spectra. For peak B, the same experiments were performed with the time-resolved fluorescence method using a band-pass filter using a 440 nm center wavelength, at 40% of the maximum transmittance and 70 nm FWHM. However, in this case, no clear fluorescence images of fingerprints were obtained. This is because fluorescence of paper is stronger by three or four orders of magnitude than that of fingerprints and, therefore, fluorescence of fingerprints was masked by the fluorescence of the white paper.

The influence of continuous irradiation on fluorescence of fingerprints was measured at the optimal excitation wavelengths. Figure 4 shows the time-lapse of peak A with excitation at 280 nm. The time duration range is 0-240 min and the delay time is 10 nsec. The laser power with excitation at 280 nm is c. 3 mJ per shot of laser. The repetition rate of the laser pulse is 10 Hz. The emission wavelength of spectra ranges from 290 to 430 nm. The intensity of peak A is maximal at 0 min and then diminishes with continuous irradiation. After a few hours irradiation, peak A seemed to disappear. Figure 5 shows the time-lapse of fluorescence spectra of peak B with excitation at 280 nm, from 0 to 210 min. The delay time is 10 nsec. The emission wavelength ranges from 370 to 510 nm. When the fingerprint was examined just after being pressed, only a part of the foot of peak A was observed in this wavelength region and peak B was not observed. Peak B appeared with time and its intensity increases slightly with continuous irradiation. The influence of continuous irradiation on fluorescence of fingerprints was also measured at 230 nm, which was another optimal excitation wavelength. The result was similar to that of excitation at 280 nm.

Fingerprint samples in our experiment were sebum-rich prints, which were obtained by wiping the forehead or nose. Ben-Yosef



FIG. 3—Fluorescence images of fingerprints on white paper at peak A with excitation at (a) 230 nm, (b) 280 nm, and (c) 300 nm.



FIG. 4—Time-lapse of peak A. Peak A appears with time and its intensity increases with irradiation.



FIG. 5—Time-lapse of peak B. Peak B diminishes with continuous irradiation.

et al. showed that much weaker results were obtained with eccrine prints (12). We also performed experiment using eccrine prints, but we were not obtained clear images in our experiments.

Summary

In the present study, we determined the fluorescence of latent fingerprints using a tunable laser. Excitation spectra were measured for two main peaks at c. 330 and 440 nm. For the 330 nm peak component, the optimal wavelength for fluorescence spectrum was c. 280 nm in the excitation range of 220–310 nm. The images of latent fingerprints on white paper were observed and the clearest image was obtained with excitation at 280 nm. The influence of continuous irradiation on the fluorescence of fingerprint was also measured at the optimal excitation wavelengths. The 330 nm peak was dominant at first, but its intensity reduced as the total irradiation time increases. Although the 440 nm peak was weak at first, its intensity increased after continuous laser irradiation.

1106 JOURNAL OF FORENSIC SCIENCES

References

- Dalrymple BE, Duff JM, Menzel ER. Inherent fingerprint luminescencedetection by laser. J Forensic Sci 1977;22:106–15.
- Menzel ER. Fingerprint detection with lasers. 2nd ed. New York: Marcel Dekker, 1999.
- 3. Lee HC, Gaensslen RE. Advances in fingerprint technology. 2nd ed. Boca Raton: CRC Press, 2001.
- Bramble SK. Fluorescence spectroscopy as an aid to imaging latent fingermarks in the ultraviolet. J Forensic Sci 1996;41:1038–41.
- Bramble SK, Creer KE, Gui Qiang W, Sheard B. Ultraviolet luminescence from fingerprints. Forensic Sci Int 1999;59:3–14.
- Springer E, Almog J, Frank A, Ziv Z, Bergman P, Qiang WG. Detection of body fluids by inherent short wavelength UV luminescence: preliminary results. Forensic Sci Int 1994;66:89–94.
- Murdock RH, Menzel ER. A computer interfaced time-resolved luminescence imaging system. J Forensic Sci 1993;38:521–9.
- Seah LK, Dinish US, Ong SK, Chao ZX, Murukeshan VM. Timeresolved imaging of latent fingerprints with nanosecond resolution. Opt Laser Tech 2004;36:371–6.
- Kuroki K, Saitoh N, Takeuchi S. Development of the imaging system of time-resolved spectrometry with YAG laser. Proceedings of the 6th Annual Meeting of the Japanese Journal of Science and Technology for

Identification, 118. 2000 Nov 9-11; Tokyo, Japan. Chiba, Japan: Japanese Association of Science and Technology for Identification, 2000.

- Anderson J, Bramble S. The effects of fingermark enhancement light sources on subsequent PCR-STR DNA analysis of fresh bloodstains. J Forensic Sci 1997;42:303–6.
- Saitoh N, Akiba N. Ultraviolet fluorescence spectra of fingerprint. Sci World J 2005;5:355–66.
- Ben-Yosef N, Almog J, Frank A, Springer E, Cantu AA. Short UV luminescence for forensic applications: design of a real-time observation system for detection of latent fingerprints and body fluids. J Forensic Sci 1998;43:299–304.

Additional information and reprint requests: Norimitsu Akiba, Ph.D. Physics Section Second Forensic Science Division National Research Institute of Police Science 6-3-1 Kashiwanoha

Kashiwa Chiba 277-0882

Japan E-mail: akiba@nrips.go.jp