Non-destructive terahertz imaging of illicit drugs using spectral fingerprints

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Abstract: The absence of non-destructive inspection techniques for illicit drugs hidden in mail envelopes has resulted in such drugs being smuggled across international borders freely. We have developed a novel basic technology for terahertz imaging, which allows detection and identification of drugs concealed in envelopes, by introducing the component spatial pattern analysis. The spatial distributions of the targets are obtained from terahertz multispectral transillumination images, using absorption spectra measured with a tunable terahertz-wave source. The samples we used were methamphetamine and MDMA, two of the most widely consumed illegal drugs in Japan, and aspirin as a reference.

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References and links

- D. M. Mittleman, G. Gupta, B. Neelamani, R. G. Baraniuk, J. V. Rudd, and M. Koch, "Recent advantages in terahertz imaging," Appl. Phys. B 68, 1085-1094 (1999).
- B. Ferguson and X. C. Zhang, "Materials for terahertz science and technology," Nature Materials 1, 26-33
 (2002)
- 3. P. H. Siegel, "Terahertz technology," IEEE T. Microw. Theory Tech. 50, 910-928 (2002).
- J. E. Parmeter, D. W. Murray, D. W. Hannum, Guide for the selection of drug detectors for law enforcement applications, NIJ Guide 601-00, (National Institute of Justice, Washington, 2000).
- A. Fitzgerald, and J. Chamberlain, "An introduction to medical imaging with coherent terahertz frequency radiation," Phys. Med. Biol. 47, R67-R84 (2002).
- T. Löffler, T. Bauer, K. Siebert, H. G. Roskos, A. Fitzgerald and S. Czasch, "Terahertz dark-field imaging of biomedical tissue," Opt. Express 9, 616-621 (2001), http://www.opticsexpress.org/abstract.cfm?URI=OPEX-9-12-616
- S. Wang, B. Ferguson, C. Mannella, D. Gray, D. Abbott and X.C Zhang, "Powder Detection Using THz Imaging," in OSA Trends in Optics and Photonics (TOPS) vol. 73, Conference on Lasers and Electro-Optics, OSA Technical Digest, Postconference Edition (Optical Society of America, Washington DC, 2002), pp. 132-133.
- 8. Y. Watanabe, K. Kawase, T. Ikari, H. Ito, Y. Ishikawa and H.Minamide, "Component spatial pattern analysis of chemicals using terahertz spectroscopic imaging," Appl. Phys. Lett., 83, 800-802 (2003).
- K. Kawase, J. Shikata and H. Ito, "Terahertz wave parametric source," J. Phys. D: Appl. Phys. 35, R1-R14 (2002)
- S. Kawata, K. Sasaki, and S. Minami, "Component analysis of spatial and spectral patterns in multispectral images. I. Basis," J. Opt. Soc. Am. A 4, 2101-2106 (1987).

1. Introduction

The terahertz (THz) waves, categorized between millimeter radio waves and far infrared light waves, exhibit properties of both sides of the electromagnetic spectrum. Like radio waves, they can be transmitted through a wide variety of substances such as paper, cloth, ceramics, plastics, wood, bone, fat, various powders, dried food, and so on. In addition, like light waves, they can easily be propagated through space, reflected, focused and refracted using THz-optics. Furthermore, the short wavelength (several hundred μm), much shorter than that of usual radio waves, allows for a spatial resolution which is sufficient in many imaging applications [1-3]. The range of potential applications is likely to expand even further with the increased availability of many absorption spectra (i.e., fingerprint spectra) peculiar to specific chemicals, including vitamins, sugars, pharmaceuticals, agricultural chemicals, discovered since last year in the THz-wave region.

The absence of non-destructive inspection technique for illicit drugs hidden in mail envelopes has resulted in such drugs being not only smuggled across international borders but also transported from one jurisdiction to another within a country with surprising ease [4]. The situation must also be attributed to the inconvenience of having to obtain a search warrant to examine the contents every time the need arises. A majority of the legal systems in the world prohibit private letters, whether they be suspected or otherwise, from being examined without a search warrant. There exist several inspection techniques such as passing the mail through an x-ray scanner, having it sniffed by a trained dog, or swiping its outside with a trace detection system. However, the ability of x-ray scanners is limited to identifying the shape of a vinyl plastic bag or a tablet, and not the type of the drug, providing insufficient grounds for opening the envelope for examination. Trace detection and canine detection, on the other hand, can only be effective if there are detectable signs outside the envelope, such as a scent or trace amounts of the concealed drug. Development is also underway in the field of millimeter-wave imaging, but the lack of fingerprint spectra in this region makes the identification of the drug type difficult. Another limitation is the low spatial resolution, of several millimeters. As for infrared imaging, where chemical fingerprint spectra do exist, the high degree of absorption and scattering in paper prevents an accurate measurement.

In contrast, the THz wave is suitable for drug detection purposes, being able to screen the contents of envelopes and our measurement results having proven the existence of fingerprint spectra peculiar to illicit drugs in the THz region. Having spent the past ten years in developing a widely tunable THz-wave parametric oscillator (TPO) that is both compact and easy to use, we have demonstrated a THz spectroscopic imaging system by introducing the component pattern analysis method. The spatial distribution of the drugs inside the envelope was extracted from the multispectral images using the absorption spectra. This procedure takes advantage of two key elements, namely the TPO's wide tunability and the fingerprint spectra. Although there have been several reports on multispectral THz-imaging [5,6], and powder detection in the envelope [7], they did not utilize spectral fingerprints of the target. Therefore, it has been difficult for them to determine the specific kinds of drugs in the envelope.

2. Experimental methods

The THz spectroscopic imaging system [8] consists of a Q-switched Nd:YAG laser, a TPO [9], imaging optics, an xy scanning stage, a detector, a lock-in amplifier, and a personal computer, as shown in Fig. 1. The Nd:YAG laser (wavelength 1.064 µm) pumps a nonlinear optical crystal MgO:LiNbO₃ (length 65mm), simultaneously generating a THz-wave (wavelength 120 - 300 µm, frequency 1 - 2.5 THz) and a idler wave (wavelength 1.068 - 1.074 µm) by non-collinear phase matched parametric oscillation. The frequency tuning is achieved by rotating the oscillator slightly so that the phase-matching angle is changed. The generated THz-wave is focused on the target by a polyolefin plastic lens (focal length 50 mm) producing a focal spot of about 0.5 mm in diameter. The target is raster scanned by the xz stage over a 20×38 mm² area, which corresponds to $40 \times 76 = 3040$ pixels. The measurement

time was approximately ten minutes. The transmitted THz wave is projected onto a pyroelectric or a Si-bolometer detector. The signal is separated from noise with a lock-in amplifier synchronized on the laser pulse frequency. The stability of this imaging system was RMS = 2.3%. In this condition, the required minimum absorption per path was approximately 6.9% (3σ level).

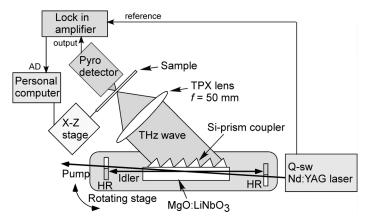


Fig. 1. Schematic of THz spectroscopic imaging system using THz wave parametric oscillator (TPO).

The calculation procedure, component spatial pattern analysis [10], is based on the following principle: Consider that a target, which is composed of M substances having different spectral characteristics, is imaged at N frequencies. Each image is composed of L pixels, which are thought of as being rearranged one-dimensionally, for ease of calculation. The transmitted intensity can be described by the following linear matrix equation:

$$[I] = [S] [P] \tag{1}$$

where [I] is a $N \times L$ matrix of the N recorded images whose row vectors represent each an L-pixel image taken at an individual frequency, [S] is a $N \times M$ matrix of the measured spectra of M drugs whose column vectors represent the spectrum data set of each substance at N frequencies, and [P] is a $M \times L$ matrix of the spatial distribution of the M drugs whose row vectors represent the spatial pattern of each drug with L pixels. For the case when N = M, [P] is simply given by $[P] = [S]^{-1}$ [I]. For the case when N > M, [P] can be solved using a least-squares method as

$$[P] = ([S]^{t} [S])^{-1} [S]^{t} [I]$$
(2)

where *t* denotes transpose. By this means, the spatial distribution of a specific component in a sample made up of several chemicals can be imaged. As the incident THz waves are mainly attenuated by absorption in the sample drugs, the transmitted intensity satisfies the Lambert-Beer law. Therefore, the detected intensity is first divided by the standard intensity of the THz wave and then the logarithm of this ratio is taken for the elements of matrix [*I*] in order to satisfy the linearity required by Eq. (1).

3. Experimental results

As samples we chose for this experiment three drugs that were: methamphetamine (*d*-methamphetamine hydrochloride, more than 98% purity), currently the most widely consumed drug of abuse in Japan, MDMA (*dl*-3,4-methylenedioxymethamphetamine hydrochloride, 67% purity), another drug of abuse becoming widespread on a global scale, and aspirin (100% purity) as a reference. As shown in Fig. 2, ~20 mg of each substance were

placed in a small 10×10 mm polyethylene bag. The three bags were then placed inside a usual airmail-type envelope. THz images of the rectangular area indicated by the yellow line in Fig. 2 were captured.

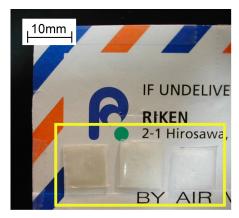


Fig. 2. View of the samples. The small polyethylene bags contain from left to right: MDMA, aspirin, and methamphetamine. The bags were placed inside the envelope during imaging. The area indicated by the yellow line represents the imaging target, 20×38mm in size. Since methamphetamine and aspirin are similar in appearance, we used a slightly longer bag for the latter to avoid confusion.

By changing the frequency emitted by the TPO within the 1.3 to 2.0 THz range, we obtained N=7 multispectral images as shown in Fig. 3, generating a matrix [I] of dimensions $N\times L=7\times 3040$. In Fig. 3, the scale of the image $-\ln(I_t/I_0)$ is the logarithm of the transmitted THz-wave intensity I_t divided by the intensity of the THz wave that was only transmitted through the envelope I_0 . This means that the greater the absorption, the brighter the shades. Aspirin appears darker at this stage due to its relatively low absorption. Subsequent component pattern analysis, however, will cancel the effect of absorption intensity, leaving only the spatial pattern. Therefore, the low absorption will not interfere with its detection.

The absorption spectra of the three drugs were measured with the same TPO system as shown in Fig. 4. The corresponding absorption intensity values at the seven frequencies were extracted to obtain the matrix [S] of dimensions $N \times M = 7 \times 3$. Although the spectra of methamphetamine and MDMA are similar, the difference between them enabled us to distinguish between the two using the component pattern analysis method.

By substituting [I] and [S] thus obtained into Eq. (2), the spatial pattern [P], represented by a matrix of dimensions $M \times L = 3 \times 3040$, was calculated. Figure 5 shows the result of extracting the three components from this matrix, with each image corresponding to each of the sample drugs. As it is evident from these images, the three drugs have been clearly distinguished and the corresponding spatial patterns obtained. A ROI (region of interest) was set in each area of the component patterns in Fig. 5 and then we took the average of tone in each ROI. The ROI was a square with 20×20 pixels, which is similar to the size of a plastic bag. The averages of MDMA, aspirin, and methamphetamine were 122, 119 and 138, respectively. The errors were less than $\pm 10\%$, which is sufficient for the drug detection purposes.

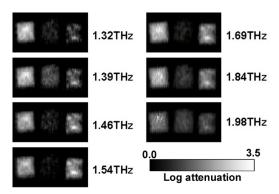


Fig. 3. Seven multispectral images generating a matrix [1].

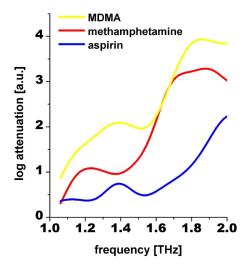


Fig. 4. Absorption spectra of MDMA (yellow line), methamphetamine (red line), and aspirin (blue line). The corresponding absorption intensity values at the seven frequencies were extracted to obtain the matrix [S].

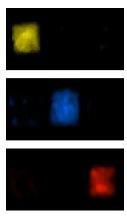


Fig. 5. Extracted spatial patterns of MDMA (yellow), aspirin (blue), and methamphetamine (red), using Eq. (2). The three drugs are clearly distinguished and corresponding spatial patterns obtained.

5. Conclusion

In conclusion, the non-destructive detection of illicit as well as legally available drugs hidden in envelopes was shown to be possible at a concentration of ~20 mg/cm². In addition to the results reported in this article we have also verified experimentally that we can isolate and extract the spatial patterns of each component by using the above method even in cases where the target is a mixture of multiple drugs or arranged in layers. Aside from the three drugs chosen for the experiments reported here, we also confirmed by conducting spectroscopic measurements that the component pattern analysis method can be applied in the same way to other drugs, including *d*-amphetamine (stimulant drug), *l*-ephedrine and *l*-methylephedrine (stimulant raw materials), *l*-methamphetamine (ingredient of Vicks Inhaler), acetaminophen (Tylenol), and caffeine. We plan to conduct non-destructive imaging on these drugs, while exploring the possibility of applying our technology to screening packages, security frisking, quality inspection of pharmaceuticals, and pathologic diagnosis. A joint project is currently underway to develop a high-speed THz-spectroscopic imaging system that uses CCD electrooptic sampling, which is expected to drastically shorten the measurement time of our method.

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