Highly sensitive surface plasmon terahertz imaging with planar plasmonic crystals

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Abstract: We report on the operation of a highly sensitive terahertz imaging system relying on a planar metallic plasmonic crystal as a terahertz surface plasmon resonant (THz-SPR) sensor. The terahertz imaging is based on the resonantly enhanced transmission phenomenon of a periodically perforated metal film. The detection sensitivity and the imaging contrast for small amounts of substance are considerably better than those of the conventional terahertz transmission imaging without a THz-SPR sensor. As a demonstration, a high contrast image of a fingerprint recorded on a thin film can be achieved by using this system.

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1. Introduction
Terahertz (THz) technologies have developed remarkably in the past decade. Since many materials have distinguishing optical properties in the THz region, several practical...
applications were proposed and demonstrated so far, such as medical diagnostics [1,2], security applications [3-5], drug inspection [6], etc. Especially the development of imaging techniques [7,8] is one of most valuable achievements for the research field in the THz region. However, since the wavelength of the THz region is several hundred microns, (about three orders of magnitude larger than that of the visible and near-infrared regions), it is very difficult to investigate samples much thinner than the THz wavelength. Artificial resonant structures offered one solution to this problem [9]. The sensitivity of the sample detection could be increased significantly around the resonant frequency, realizing the detection of very small amounts of sample. For those resonant structures, however, the implementation of an imaging configuration is still challenging because of the difficulty to fabricate large-area arrayed devices.

By using metallic plasmonic crystals (MPC), which are realized as arrays of circular holes in a metallic plate, we can obtain both the high sensitivity and the large-area detection of small sample amounts, which cannot be detected by conventional THz spectroscopic systems. By illuminating the MPC with a THz wave, the surface plasmon polariton (SPP) [10] can be excited in the vicinity of the metal surface as a result of the interference of scattered light from the aperture array [11,12]. The SPP excitation occurs only at a certain resonant frequency, determined by the geometrical structure of the MPC, and thus, a resonant narrow band-pass characteristic appears in the transmission spectrum. Due to the strong localization of the electric field of SPPs near the MPC surface, the resonant transmission characteristic depends strongly on the dielectric distribution in the vicinity of the MPC surface [13]. This characteristic allows us to achieve a highly sensitive detection of small amounts of sample placed at a subwavelength distance from the MPC surface [14]. Furthermore, this sensing technique also allows us to realize large-area imaging with significant high sensitivity owing to the relatively simple design of the MPC.

In this paper, we demonstrate, for the first time, the highly sensitive THz imaging of samples that are very thin and only available in small amounts, by using the characteristics of the MPC surface plasmon resonance (SPR). Such a SPR sensing and/or imaging are possible also in other frequency regions, such as the visible or the near-infrared, with other optical configuration [10]. In the THz region, however, there is a particular interest due to the rich optical properties in biologically-relevant molecules, medical agents or other materials that are closely relevant to the human life.

2. Experimental

The terahertz imaging system is depicted in Fig. 1. The terahertz wave pulses are emitted from a dipole-type photoconductive antenna illuminated by a 100-fs laser pump pulses at 800 nm. The emitted terahertz wave pulses are collimated and then focused with a pair of off-axis paraboloidal mirrors onto the sample attached on the MPC, which has the role of a THz SPR-sensing imaging plate, at normal incidence. A zeroth-order transmitted terahertz wave was again collimated and focused by another pair of paraboloidal mirrors onto the detector. The detection was achieved by a dipole-type photoconductive antenna gated with time-delayed probe laser pulses which were divided from the pump pulses. By varying the difference between the optical path lengths of the pump and probe pulses, the waveform of the terahertz wave is measured in the time domain. The sample was attached on the MPC and was x-y scanned by a pair of linear motor stages for acquiring two-dimensional images.
Our MPC was a metal plate perforated in a triangular lattice with circular holes. The transmission characteristic of the MPC is mainly determined by its geometrical parameters, such as the hole diameter, the spacing between holes and the film thickness. The geometrical parameters of these MPC’s are shown in Table 1. The resonant frequency \( f_{SPP} \) of the SPP on the periodically structured surface is expressed as

\[
\frac{k_{in}}{} + \frac{G}{2\pi} \left( \frac{\varepsilon_m + \varepsilon_d}{\varepsilon_m \varepsilon_d} \right) \right]^{1/2}
\]

where \( k_{in} \) is the in-plane wave-vector component of the incident THz wave, \( G \) is the reciprocal lattice vector of the periodic structure, and \( \varepsilon_m \) and \( \varepsilon_d \) are the dielectric constants of the metal and the interface medium, respectively. In this experiment, we fabricated three MPC’s for different resonant peak frequencies.

### 3. Result and Discussion

We first measured the transmission characteristics of the MPC’s. Figure 2(a) shows the zeroth order transmission spectra of the three MPC’s at normal incidence. The incident THz beam is a collimated wave limited by a metallic circular aperture 10 mm in diameter and located in front of the metal hole array. Clear peaks are observed at 0.31, 1.57, 1.96 THz for the transmission spectra of MPC1, MPC2 and MPC3, respectively. The slight red shift of the peak frequency with respect to the estimated SPP resonant frequency is due to the Fano-like interference effect between directly transmitted electromagnetic waves and surface plasmon excited in the vicinity of metal surfaces [11, 12]. At the peak frequency, the SPP is excited.

### Table 1. Geometrical parameters of three different MPC’s used in the experiments. The SPP resonant frequencies, transmission peak frequencies and the quality factors are also listed.

<table>
<thead>
<tr>
<th>sample</th>
<th>hole diameter (( \mu m ))</th>
<th>hole spacing (( \mu m ))</th>
<th>thickness (( \mu m ))</th>
<th>( f_{SPP} ) (THz)</th>
<th>( f_{peak} ) (THz)</th>
<th>( Q )</th>
</tr>
</thead>
<tbody>
<tr>
<td>MPC1</td>
<td>500</td>
<td>1000</td>
<td>300</td>
<td>0.34</td>
<td>0.31</td>
<td>18.6</td>
</tr>
<tr>
<td>MPC2</td>
<td>100</td>
<td>200</td>
<td>50</td>
<td>1.73</td>
<td>1.57</td>
<td>14.3</td>
</tr>
<tr>
<td>MPC3</td>
<td>80</td>
<td>160</td>
<td>30</td>
<td>2.16</td>
<td>1.96</td>
<td>14.6</td>
</tr>
</tbody>
</table>

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\[
f_{SPP} = k_{in} + G \left( \frac{\varepsilon_m + \varepsilon_d}{2\pi \varepsilon_m \varepsilon_d} \right)^{1/2}
\]
and the electric field is strongly localized in the vicinity of the metal surface. The distribution of the SPP electric field intensity ($E_z$ component) is calculated by using a finite difference time domain (FDTD) method and is depicted in Fig. 2(b) for the geometrical parameters of MPC1. The attenuation length in the direction of the surface normal is estimated to be 370 μm, which is less than half of the wavelength at 0.31 THz. The transmission peak frequency of the MPC depends strongly on the dielectric constant on the metal surface because the excitation situation of SPPs changes dramatically. Thus, one can detect the difference of the material located in the vicinity of the metal surface, even if the difference is very small, by monitoring the transmission spectrum of the MPC. Especially, since the electric field of SPPs is located within the half of wavelength from the metal surface as shown in Fig. 2(b), the SPP is particularly sensitive to any dielectric changes within a subwavelength region from the metal surface and the highly sensitive detection can be achieved.

When the MPC is used as a SPR sensor, the quality factor of the transmission peak is critically important for sensitivity. Assuming that the transmission peak has a Gaussian profile, the quality factor can be defined as $Q = f_0 / \Delta f$, where $f_0$ is the peak frequency and $\Delta f$ is the FWHM of the transmission peak. In our measurements, the quality factor of MPC1 ($Q = 18.4$) is higher than that of MPC2 ($Q = 14.6$) and MPC3 ($Q = 14.8$). The spatial resolution is also an important specification for an imaging system. In order to achieve a high spatial resolution for our imaging system, the incident terahertz beam has to be focused onto the sample. In this case, however, the quality factor is possibly reduced due to the finite-size effect [15]. Figure 3 shows the transmission spectra of MPC1 where the incident wave is a collimated beam 10 mm in diameter as well as a focused beam only about 2 mm in diameter. Compared to the collimated incidence, in the case of the focused incident THz wave, both the peak transmittance ($T \sim 0.25$) and quality factor ($Q \sim 10.0$) become lower. The coupling of the incident wave to the surface plasmon on the metal hole array relies on the interference between the electromagnetic wave scattered by individual holes [16]. Thus, the coupling efficiency corresponds to the intensity of the Fourier component of the real-space geometrical

![Fig. 2](image-url)
structure in the illuminated area. For the focused incident wave, the intensity of the Fourier component decreases due to the smaller number of relevant holes taking part in scattering the THz wave, and, consequently, the transmittance and the quality factor of the transmission peak decrease. Since the sensitivity and the spatial resolution are in such a tradeoff relation, it is necessary to make a compromise between these two capabilities.

As demonstrations of the highly sensitive imaging with the MPC, firstly we measured the image of a letter “R” written by soluble ink on a paper, shown in Fig. 4(a). The paper was attached on the MPC and illuminated by the focused THz wave to measure the transmission spectrum. By scanning the illumination point in the plane of the sample surface with the step length of 250 μm, we obtained the two-dimensional transmission image. Figures 4(b) and (c) show the transmission image at 0.40 THz and 1.50 THz by using the MPC1 and MPC2, respectively. For these frequencies, the diameters of the focused beam are about 1.5 mm and 0.4 mm, respectively. In our experiment, the frequency resolution is 25GHz, and therefore, we can choose the single frequency image from the many images of broad band frequencies with 25GHz interval. Although the high contrast image can be obtained around the peak frequency of MPC, the frequency of the highest contrast image is not always the peak frequency. This is because, by attaching the paper or the thin film on the MPC surface, the transmission spectrum changes from that of the free standing MPC. Especially, in the asymmetric structure, where the paper is attached on one side of the MPC, the transmission spectrum becomes more complex. Owing to this characteristic, the spectrum change related to SPPs lies within the relatively broad frequency range, and therefore we can detect such the spectrum change at
0.4 THz. A conventional transmission image, without using the MPC, is also shown in Fig. 4(d) at 1.50 THz for comparison; in this image no contrast is observed. On the other hand, owing to the excitation of the surface plasmon, the sensitivity to detect small amount of solvent ink are dramatically increased and consequently the apparent images of the ink are obtained as shown in Fig. 4(b) and 4(c). For the image with the MPC1 [Fig. 4(b)], the edge of the character is indistinct. Considering from the higher quality factor of MPC1 compared with that of MPC2, this is not due to the lower sensitivity for MPC1 but to the lower spatial resolution for the THz wavelength at 0.40 THz with relatively large beam diameter. For the image with the MPC2, a clearer edge of the character image can be obtained due to the relatively small beam diameter and consequently, higher spatial resolution at 1.50 THz.

As a more practical application of this highly sensitive THz imaging, we demonstrate the detection of a human fingerprint recorded on a polypropylene film [Fig. 5(a)]. We obtained the transmission image using the same method explained above except that MPC3 was chosen; its transmission peak is observed around 1.96 THz. By using the MPC3, we can achieve the spatial resolution required for the purpose of fingerprint imaging. Figure 5(b) shows the transmission image of the fingerprint at 2.09 THz. For comparison, the transmission image of the same fingerprint without any MPC is shown in Fig. 5(c); the contrast is significantly lower. This indicates a dramatic increase of the detection sensitivity for very small amounts of biological tissue in the THz imaging technique.

4. Summary

In conclusion, we demonstrated a highly sensitive surface plasmon sensing and imaging measurement for very small amounts of sample which cannot be detected by the simple THz spectroscopic systems. Owing to the strong localization of the electromagnetic wave in the vicinity of the MPC surface, the detection sensitivity, and therefore the imaging contrast, increases significantly from those of the conventional terahertz transmission imaging without MPC. We believe that this highly sensitive imaging technique has a great potential to be included in various THz applications.

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