Terahertz field enhancement in asymmetric and tapered nano-gaps

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Abstract: We investigate field enhancement inside metal-insulator-metal gaps with asymmetric thicknesses and tapered shapes in the terahertz regime. Finite-difference time-domain simulations were conducted for calculation of field enhancement factor. The calculation indicates that for asymmetric sample, field enhancement increases proportionally with the decrease of the thinner of the two metal film thicknesses surrounding the gap. Concomitantly, angle variation has little effect on the field enhancement if the thickness of the narrowest gap region is fixed. A model based on the capacitor concept is proposed for intuitive understanding of the phenomena.

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References and links

1. Introduction

Electric field can be enhanced near the edge of a metal structure due to free charge accumulation at the apex. When two sharp edges are brought together forming a small gap, the field is even more enhanced because opposite charges further concentrate on either edges. This enhanced electric field can be utilized to detect nonlinear phenomena such as surface enhanced Raman scattering (SERS) [1–4], harmonic generation [5–7], optical rectification [8–10], etc., in which high field intensity is needed. The field enhancement becomes higher as the gap width decreases, or as the wavelength increases [11]. For achieving high field enhancement in the gap with relatively long wavelength of THz frequency, metal planes with nanometer-scale gaps, the length of which is as long as a few hundred μm, are needed. These gaps also need to be aligned vertically for optical experiments. Conventional electron-beam lithography and focused ion beam technique are adequate for making nm-scale gap, but it is hard to form sub-10 nm gaps by these techniques. Recently a technique which forms 1-10 nm vertical gap between the metals was demonstrated by atomic layer deposition (ALD) and exfoliation technique [12,13]. In the exfoliation process, discontinuity between the layer to be removed, which is a secondly deposited metal on top of the previously deposited metal, and that to be left, which is the secondly deposited metal directly on substrate, is important for successful fabrication. The gap is formed between the firstly deposited and the secondly deposited metal layers. Giving different thicknesses between these layers increases the throughput of sample fabrication. In addition, tapered gap shape often appears intentionally or unintentionally, which makes it difficult to predict field enhancement amongst other characteristics. Figure 1(a) shows cross-section scanning electron microscopy (SEM) images of two samples fabricated by deposition methods. The samples are rectangular ring arrays in which a unit cell consists of inner gold metal and outer gold metal separated by a thin insulating alumina spacer. Inplane dimension of a rectangular ring is 50 μm x 50 μm, and insulating gap between metals is about 5 nm. Although these two samples have the same inplane dimension with the same gap width, they show different transmission characteristics. Shown in Fig. 1(b) are field enhancement spectra estimated from the THz transmission measurement using Kirchhoff integral formalism [14]. Both the peak positions and the field enhancement values differ significantly between the two samples. This means that metal thickness and the shape of the gap affect the transmission characteristics and also field enhancement inside the gap [15]. Considering applications of such a gap including nonlinearities and sensitive detection [16,17], it is important to estimate the field enhancements correctly.

In this paper we investigate tapered gap structures with asymmetric thicknesses at THz regime. We used 2-dimensional finite-difference time-domain (FDTD) method to simulate THz transmission through the structures. By calculating field enhancement of the gap with changing tapered angle and thickness symmetrically and asymmetrically, the effects of the gap shape and the symmetry on field concentration between metals are examined. A model based on the capacitor concept is also proposed for an intuitive explanation.
Fig. 1. (a) Cross section SEM images of Au-Al$_2$O$_3$-Au gap samples. Inplane dimensions of two samples are the same, and insulating gap between metals is about 5 nm. (b) Field enhancement spectra estimated from THz transmission measurement of the gap samples. The spectra show different resonance positions and field enhancement values, which means that characteristic of the structure is determined by metal thickness and gap shape.

2. Results and discussions

A schematic of the structure used for FDTD simulation is shown in Fig. 2(a). In all simulations, the gap width $w$ and overall thickness $t$ were fixed at 5 nm and 200 nm, respectively. Gap material was assumed to be alumina with refractive index, $n_{\text{gap}}$, of 2.12 and substrate was set to be sapphire with $n_{\text{sub}}$ of 3.4. Although gap material and substrate had the same chemical formula as Al$_2$O$_3$, thickness dependent refractive index was considered [18]. Au metal was implemented using Drude model, in which plasma frequency and damping frequency are $1.37 \times 10^4$ THz and 40.7 THz [11]. For efficient calculations non-uniform grid was adopted [19,20], where the minimum grid size was set at 0.25 nm in the vicinity of the gap. Incident source was plane wave of THz pulse with center frequency of 0.5 THz. The simulations were conducted under transverse magnetic mode. We calculated field enhancement factor as average electric field, normalized by transmitted electric field through substrate-only without the metallic gap structure, along the output part of the smallest gap which is depicted by red dashed line in Fig. 2(a). As can be seen in Fig. 2(b) the structure can be classified into four types according to tapered angles, $\theta_L$ and $\theta_R$ where subscript L means left and R means right: $\theta_L = \theta_R = 90^\circ$ for symmetric and non-tapered; $\theta_L = 90^\circ$, $\theta_R = 0^\circ$ for asymmetric and non-tapered; $\theta_L = \theta_R = \theta$ for symmetric and tapered; $\theta_L = 90^\circ$, $\theta_R = \theta$ for asymmetric and tapered structures.

Fig. 2. (a) A schematic of the structure used for FDTD simulation. $w$ and $t$ are fixed at 5 nm and 200 nm, respectively. To calculate field enhancement factor, electric field at output side of 5 nm gap (depicted by red dashed line) is normalized by transmitted electric field through substrate-only without the metallic gap structure. (b) Four classified structures according to tapered angle ($\theta_L$, $\theta_R$): symmetric, non-tapered ($\theta_L = \theta_R = 0^\circ$); asymmetric, non-tapered ($\theta_L = 90^\circ$, $\theta_R = 0^\circ$); symmetric, tapered ($\theta_L = \theta_R = \theta$); asymmetric, tapered ($\theta_L = 90^\circ$, $\theta_R = \theta$).
Fig. 3. (a) x-component of electric field distributions and (b) x-component of current density distributions for each type of the gap structure when gap thickness $t_{gap}$ is 50 nm and $w$ is 5 nm. For the tapered structures, tapered angle is $60^\circ$. In (a) the electric fields with similar intensities are concentrated inside the smallest gap in all cases. In (b) current densities along the metal edges forming the smallest gap are very similar to each other, although they differ significantly once we move away from the smallest nanogap region.
FDTD results for each type of the structure are plotted in Fig. 3 when the gap thickness $t_{\text{gap}}$ is 50 nm and $\theta$ is set at 60° for all the said four structures: x-component of the electric field distributions [Fig. 3(a)] and x component of the current density distributions [Fig. 3(b)]. Each point of these plots represents the 0.5 THz frequency component of the Fourier transform. It is clearly shown in Fig. 3(a) that electric fields are concentrated inside the smallest gap. These enhanced electric fields have similar intensities in all four cases even though radiation patterns are different. The field enhancement is related to a current flow inside the metal which determines the edge surface charge density. When electromagnetic wave impinges on the metal structure, the incident field induces a current on the metal surface. Because $t$ of the structure is thinner than the skin depth, multiple reflection occurs in the metal and the current flows inside the overall metal. Unlike flat metal, in our structures the induced current is concentrated on the vicinity of the smallest gap as shown in Fig. 3(b). It should be noted that current densities along the metal edges forming the smallest gap are very similar to each other in all four cases, although they differ significantly once we move away from the smallest nanogap region. This is consistent with the same level of the enhanced electric field in all cases as will be shown in Fig. 4.

Field enhancement factors as a function of $t_{\text{gap}}$ are plotted in Fig. 4(a) where $\theta$ is set at 60°. It goes higher as $t_{\text{gap}}$ becomes smaller. We start with $t_{\text{gap}} = t = 200$ nm, where all four types of the structures become the same. The field enhancement factor of 380 with $t_{\text{gap}} = 200$ nm increases to $> 870$ with $t_{\text{gap}} = 50$ nm for all types of the structures but the differences are within 10%. In particular, field enhancements differ only by 3% or less amongst the two symmetric and two asymmetric structures respectively, with the symmetric structures having larger field enhancements by about 8% in case of $t_{\text{gap}} = 50$ nm. Clearly, despite these details, field enhancement is predominantly determined by $t_{\text{gap}}$. Dependence of field enhancement on the tapered angle is represented in Fig. 4(b) when $t_{\text{gap}}$ is fixed at 100 nm. When $\theta$ is 0° the structure is non-tapered. At angles larger than zero, the structure is tapered eventually becoming non-tapered with larger thickness when $\theta$ reaches 90°. It is worth noting that field enhancements are almost the same with the tapered angle being smaller than 70° for each symmetric and asymmetric cases. Field enhancement factor with $\theta$ of 80° even shows a difference by only about 5% with that of $\theta = 0°$ case. We now provide an intuitive picture for tapered cases using the concept of capacitance.
Fig. 5. A schematics for modeling using capacitor concepts. Overall structure can be regarded as parallel connected capacitors. And capacitor of tapered part is approximately divided several capacitors in parallel with increasing capacitor width $d_m$.

Under perfect conductor approximation, for example, the overall structure of symmetric tapered gap can be regarded as two capacitors in parallel. One is a capacitor of the smallest gap region and the other is that of the tapered region. The latter can be again divided by several capacitors approximately as shown in Fig. 5. Using simple capacitor equations, capacitance per unit length $C$ can be calculated as follows:

$$C_{\text{total}} = C_{\text{gap}} + C_{\text{tapered}} = \varepsilon_0 \varepsilon_{\text{gap}} \frac{t_{\text{gap}}}{w} + \lim_{n \to \infty} \left[ \sum_{m=1}^{n} \varepsilon_0 \frac{\Delta y}{d_m} \right]$$

where $d_m = w + 2(m-1)\Delta y / \tan \theta$, $\Delta y = \frac{t-t_{\text{gap}}}{n}$.

$$C_{\text{total}} = \varepsilon_0 \varepsilon_{\text{gap}} \frac{t_{\text{gap}}}{w} + \int_{0}^{t_{\text{gap}}} \varepsilon_0 \frac{\tan \theta}{2y + w \cdot \tan \theta} \, dy$$

$$= \varepsilon_0 \varepsilon_{\text{gap}} \frac{t_{\text{gap}}}{w} + \varepsilon_0 \frac{\tan \theta}{2} \ln \left[ 1 + \frac{2(t_{\text{gap}})}{w \cdot \tan \theta} \right]$$

$\varepsilon_0$, $\varepsilon_{\text{gap}}$, $d_m$, and $\Delta y$ are permittivity of air, relative permittivity of gap material, width of $m$-th capacitor segment for the tapered part, and thickness of the segment, respectively. In the calculation for the capacitance of the tapered region, a capacitance between $j$-th segment and $k$-th segment ($j \neq k$) was ignored. Capacitance of asymmetric and tapered structures can be also calculated in a similar manner where each capacitor segment of tapered region is regarded as a capacitor with two dielectrics of alumina and air in tandem. If it is assumed that total induced charges accumulated on both the smallest gap and the tapered region do not change with angle, then the electric field is inversely proportional to capacitance ($E \propto 1/C$). With calculated capacitances and this proportionality, we plotted the calculated field enhancement factor for symmetric and asymmetric structures as dashed line in Fig. 4(b) when an enhancement factor of this capacitor model with $\theta$ of 0° was assumed to have the same value as that of the FDTD result. Calculation from the model shows similar tendency with FDTD, which indicates that capacitance of the structure does not change so much until $\theta$ is less than 70°. The enhancement factors of the model have smaller values than those of FDTD despite of underestimation of capacitances by ignoring cross-coupling between different segments in the model. This may be due to the change in total amount of the accumulated charges on both the smallest gap and the tapered region. As $\theta$ increases from 0°, a part of charges from the smallest gap goes to the tapered region and charges on the outer interfaces along x-direction.
of the metals also come to the tapered region, so that the capacitance model underestimate the field enhancements.

3. Conclusion

Field enhancement in asymmetric and tapered gaps between two metals with 5 nm width was examined at THz regime by numerical studies. FDTD results show that the thickness of the smallest gap region is the most important factor in field enhancement. In non-tapered structures, the field enhancement increases proportionally with the decrease of the thinner of the two metal film thicknesses. For the tapered structures, variation of the tapered angle has little effect on the field enhancement if the angle is smaller than 70 degree. A model based on the capacitor concept is in agreement with FDTD simulations. Our results provide an intuitive way of estimating field enhancements in terahertz nanogap transmission, which will have important implications on terahertz nonlinear optics, molecular sensing, and hysteresis control using these extreme nanogap samples.

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