Review article

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Terahertz wave interaction with metallic nanostructures

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Abstract: Understanding light interaction with metallic structures provides opportunities of manipulation of light, and is at the core of various research areas including terahertz (THz) optics from which diverse applications are now emerging. For instance, THz waves take full advantage of the interaction to have strong field enhancement that compensates their relatively low photon energy. As the THz field enhancement have boosted THz nonlinear studies and relevant applications, further understanding of light interaction with metallic structures is essential for advanced manipulation of light that will bring about subsequent development of THz optics. In this review, we discuss THz wave interaction with deep sub-wavelength nano structures. With focusing on the THz field enhancement by nano structures, we review fundamentals of giant field enhancement that emerges from non-resonant and resonant interactions of THz waves with nano structures in both sub- and super- skin-depth thicknesses. From that, we introduce surprisingly simple description of the field enhancement valid over many orders of magnitudes of conductivity of metal as well as many orders of magnitudes of the metal thickness. We also discuss THz interaction with structures in angstrom scale, by reviewing plasmonic quantum effect and electron tunneling with consequent nonlinear behaviors. Finally, as applications of THz interaction with nano structures, we introduce new types of THz molecule sensors, exhibiting ultrasensitive and highly selective functionalities.

Keywords: terahertz spectroscopy; optical properties; nanostructures; metamaterials.

1 Introduction

The recent development in terahertz (THz) technology enables a wide range of applications in electronics [1–6] and photonics [7–9]; for medical [10–12], military [13–15], and security purposes [16–18]; in microdisplay [19]; and even in the investigation and conservation of cultural heritages [20–22], which is based on its excellent sensitivity and selectivity. Such advances in THz applications have been stimulated by the improvement in functional THz devices such as filters [23–28], switches [29–34], and sensors [35–40]. Artificially structured metamaterials in subwavelength scale, especially, provide us with the ability to control THz responses over the broad bandwidths [41–46], determined by key parameters: length scale [47, 48], geometry [49], and surrounding materials [50, 51]. The dielectric properties of these metamaterials, changeable by optical [24, 52–54], electrical [55, 56], thermal [57–59], or other mechanical [60–62] means, with or without lateral patterns, are in turn used to control transmission or reflection behavior over entire THz frequencies, further extending device performance. THz wave interaction with deep subwavelength nanostructures with extremely high cross-sections will be discussed in this review as introduced with several milestones regarding the structures from micrometer [39, 42, 63–67] to nanometer [68–78] scale and, finally, the Angstrom scales [79, 80] (Figure 1). In particular, the enhanced THz nonlinear phenomena in Angstrom-sized infinite gaps will find unprecedented resonant and nonresonant applications in metamaterials [81–86]. Finally, several novel types of THz sensing technology based on the field enhancement properties using subwavelength structures for variety of chemistry, biology, and medical applications will be introduced.

2 Fundamentals of giant terahertz field enhancement at an infinitely long nanogap

Using the vector Babinet principle, concentrating terahertz (THz) electric field in a small aperture in metal [69,

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Figure 1: Achieved milestones in THz wave interaction with metallic structures down to the Angstrom scale.

87–92] is equivalent to enhancing the magnetic field of light around rod and patch antennas [93–100]. Although a metal film in THz frequency is neither perfect conductor nor infinitely flat, various areas of THz research [2, 99, 101–115], including THz plasmonics [116–123] and metamaterials [124–130], implicitly use this principle, which remains true in its spirit if not in its rigor. One of the earliest theoretical accounts on the light-aperture interaction had been made by Sommerfeld’s half-plane problem [131–133] that was revisited by Bethe and Bouwkamp to deal with small apertures in infinitely thin perfect conducting plates [134–137]. These earlier studies instigated further investigations on the light interaction with periodic aperture systems [138], which were later extended to studies on enhanced light transmission through the apertures in visible [139–141], infrared (IR) [142], and THz [67, 143–146] regimes. To resolve the enhanced transmission and consequent strong field enhancement near the apertures, numerous theoretical schemes such as coupled-mode theory [90, 147–154], microscopic models [89, 155, 156], transfer matrix method [157, 158], and capacitor model [88], incorporated with various numerical methods [40, 159–161], were developed.

Basically, the field enhancement can be explained in terms of macroscopic accumulation of surface charges near the apertures’ edges, driven by incident light. The accumulated charges give rise to capacitive enhancement of electric field in the gap [69, 88]. This simple picture paved a way to the gigantic field enhancement supported by deep subwavelength apertures in metal (Figure 2), which is, in particular, essential for THz nonlinear optics [163–169] that requires intense electromagnetic field.

In this section, we discuss THz field enhancement in deep subwavelength apertures. Specifically, we focus on nonresonant THz field enhancement by an infinitely long gap with sub-skin-depth thickness and provide a simple model to explain that the field enhancement can be simply given by a ratio of photon wavelength and thickness. We also discuss a Kirchhoff integral formalism that allows quantitative estimation of field enhancement in the near-field from the far-field measurement. After that, we discuss resonant field enhancement supported by a rectangle slot, exhibiting different behavior of field enhancement compared to the non-resonant case.

2.1 THz field enhancement and skin-depth physics

We first concentrate on metal films of thickness 100 nm or less, satisfying the sub-skin-depth condition at THz regime [67]. Consider, for a good conductor, that the ultimate field enhancement (FE) in a high-aspect ratio nanogap of air sandwiched between two sub-skin-depth metallic planes, irradiated by an electromagnetic wave with an incident electric field of E_{inc} , has a simple analytical expression when the gap width w is much smaller than the film thickness h :

$$FE = \frac{E_{\text{gap}}}{E_{\text{inc}}} = \frac{\lambda}{\pi h}; w \ll h, \quad (1)$$

where E_{gap} is the electric field at the gap, λ is the vacuum wavelength, and h the film thickness [76, 88, 170]. When

Figure 2: THz field enhancements in various gap structures.

(A) Nanogap in metal plate, (B) nanogap in rod antennas, and (C) split-ring resonators. A, B, and C are reproduced from Refs. [69], [101], and [162], respectively.

the gap is filled with a dielectric of permittivity, ε_{gap} , the formula is modified to, somewhat trivially,

$$FE = \frac{E_{\text{gap}}}{E_{\text{inc}}} = \frac{\lambda}{\pi h} \frac{\varepsilon_0}{\varepsilon_{\text{gap}}}, \quad (2)$$

where ε_0 is the vacuum permittivity [76]. What is surprising is that the gap FE is seemingly independent of how good the metal is.

To derive Eq. (1), we first consider direct transmission of electromagnetic wave through a thin film of free-standing metal; for the sake of simplicity, we assume a normal incidence. For this instance, by a thin film. We mean films whose thickness h is smaller than the skin depth $\delta = \sqrt{\frac{2}{\mu_0 \sigma_m \omega}}$ but larger than a characteristic thickness of

$$h_0 \equiv \frac{2\varepsilon_0 c}{\sigma_m}; \quad h_0 \ll h \leq \delta, \quad (3)$$

where σ_m is the conductivity of metal, μ_0 is the vacuum magnetic permeability, and $\omega = 2\pi f$ is the angular frequency of the electromagnetic wave. The characteristic thickness, h_0 , at which absorption loss by the metal is 50% is only 0.53 nm for a reasonably good metal of a conductivity 10^7 Siemens per meter ($\Omega^{-1}\text{m}^{-1}$), so that any transition metal films of today's technology fall above this thickness. Skin depth at 1 THz is typically 100 nm or more even for good metals so that transition metal films in the range of 5–100 nm thickness satisfy Eq. (3) (Figure 3). For these thin metal films, the amplitude transmission and reflection coefficients, t and r , respectively, are given as

$$t = \frac{E_t}{E_{\text{inc}}} \approx \frac{2\varepsilon_0 c}{\sigma_m h} = \frac{h_0}{h} \ll 1; \quad r \approx 1 - \frac{h_0}{h} \approx 1, \quad (4)$$

where E_{inc} is the incident electric field and E_t is the transmitted electric field as described in Figure 4.

To derive Eq. (4), we consider transverse magnetic polarized incident light (E_x, H_y) and focus on the magnetic field of light near the thin film. At the incident surface, reflection makes the magnetic field of light approximately twice that of the incident field, whereas on the transmission side, the magnetic field is much smaller than

Figure 3: Thickness of metallic thin films in terms of conductivity is plotted.

In particular, Si (p+), VO_2 , lead, and gold cases are denoted in blue arrows. Given conductivities of materials are at 1 THz [171–176].

Figure 4: Metallic thin film with a thickness less than the THz skin depth is irradiated by THz field.

the incident field. We assume a constant electric field/current density inside and apply Ampere's law, $\nabla \times H = J$, which gives rise to the current density $J \approx \frac{2H_{\text{inc}}}{h}$, ignoring the vacuum displacement-current term. Then, the continuity of tangential component of electric fields at the air-metal interface leads to $E_t = E_m = \frac{J}{\sigma_m} = \frac{2H_{\text{inc}}}{\sigma_m h} = \frac{2\varepsilon_0 c}{\sigma_m h} E_{\text{inc}}$, resulting in Eq. (4) (Figure 5A). Here, E_m is electric field just inside the metal surface of the transmitting side. Since the normal component of the displacement current is the same across the air-metal boundary of the gap, we obtain $E_{\text{gap}} = \left| \frac{\varepsilon_m}{\varepsilon_0} \right| E_m \approx \frac{\sigma_m}{\varepsilon_0 \omega} \frac{2\varepsilon_0 c}{\sigma_m h} E_{\text{inc}} = \frac{\lambda}{\pi h} E_{\text{inc}}$ having taken advantage of the metal dielectric constant in the terahertz regime

$$\varepsilon_m = \varepsilon_\infty + i \frac{\sigma_m}{\omega \left(1 - i \frac{\omega}{\gamma} \right)} \approx i \frac{\sigma_m}{\omega}, \quad (5)$$

Figure 5: Field enhancement in thin and thick conducting films. (A) Magnetic field of light and current density near the thin film is represented in red, where THz impinges to the plane. The magnetic field is approximately twice at the incident surface (up) and much smaller at the transmission side (bottom). (B) Obtained displacement current and E_{gap} around the narrow gap. (C) Displacement current and E_{gap} at films much thicker than the skin depth.

where ε_∞ is the high-frequency dielectric constant of the metal and γ is the Drude damping constant (Figure 5B). The physics of the conductivity independence of the FE is then clear: better conductivity makes the field inside the metal weaker, which is compensated by the higher dielectric constant of the metal when applying the displacement current boundary condition. Equation (1) can be analytically extended into a completely different regime, for samples much thicker than the skin depth (Figure 5C); for this regime, we assume that current flows only at the surface with surface current density $K = 2H_{\text{inc}}$, and terminates at the exit side of the gap, dumping charges along the way, as shown in Figure 6. Interestingly, J_x component at the metal surface in the gap, shown in Figure 6C, is kept nearly constant, while J_z (Figure 6B) is not. This implies that the accumulated surface charge is evenly distributed over the metal surface, giving rise to constant E_x field in the gap. The continuity equation demands the surface charge density being $\sigma = \frac{K}{\omega h}$, from which $E_{\text{gap}} = \frac{\lambda}{\pi h} E_{\text{inc}}$ follows.

Aforementioned FE with thin ($w \sim h < \delta$) and thick ($w < \delta < h$) narrow gaps, together with wide gaps ($\delta, h < w$), are discussed in a recent paper [76]. Shown in Figure 7A are numerically calculated current distributions with narrow (1.5 nm) and wide (200 nm) gaps in 150-nm-thick gold films, and corresponding schematics for charge distributions are shown in Figure 7B. For the wider gap, charges are not mostly accumulated at the metal edges in the gap but are spread over the surface outside the gap. This consequently reduces the FE . For the narrower gap, most charges are accumulated at the metal edge in the gap, giving rise to stronger FE . However, we stress that when the aspect ratio w/h is sufficiently small ($w/h \ll 1$), the charge distribution becomes insensitive to the gap size, so that the FE will exhibit saturating behavior with decreasing w . Also, $1/h$ dependence of FE in narrow gap is well demonstrated (Figure 7C).

Having established a simple FE formulae for thin and thick samples limits, we proceed to include the very thin-regime, where t can no longer assumed to be $\ll 1$. Applying Ampere's law, we obtain $1 + r - t = \frac{\sigma_m t h}{\varepsilon_0 c}$. Applying energy conservation including Ohmic loss, we arrive at $1 = r^2 + t^2 + \frac{\sigma_m t^2 h}{\varepsilon_0 c}$. Solving these two, we get

$$FE = \frac{\lambda}{1 + \frac{h_0}{h}}, \quad (6)$$

with h_0 given by Eq. (3); Eq. (6) can now be applied to all three regimes of thicknesses.

Figure 6: Field enhancement with thick slit.

(A) Numerically calculated map, using the COMSOL software, of the current density with thick slit in freestanding gold, showing “dumping the charge”. 0.5 THz light is incident from the top side onto the gap of $w=10$ nm and $h=10$ μm . Metal surfaces are denoted by white dashed lines. Cross-cut profiles of (B) J_z and (C) J_x along the left metal edge in the gap [from (0, 0) to (0, $-h$)] are present.

Figure 7: w/h -dependent field enhancement.

(A) Numerically calculated maps of induced current J in thin gold films. Here, $h=150$ nm and $w=1.5$ nm (left) and 200 nm (right). 0.3 THz incident light is considered. (B) Schematics of induced current and accumulated charges for narrow ($w \ll h$) and wide ($w \sim h$) gaps. (C) Experimentally measured and analytically calculated FE of 0.3 THz field in a 5 nm gap with varying thickness h . Inset figures illustrate higher surface charge density in a thinner gap, while total accumulated charge is independent of the thickness. Note that FE is modified from Eq. (1) due to the substrate effect. This figure is reproduced from Ref. [76].

To confirm that Eq. (6) gives a reasonably good description of the FE over many orders of magnitudes of conductivity, we plot, in Figure 8A, finite-difference-time-domain (FDTD) calculations (solid red line) for a 50-nm-thick film with 1 nm air gap varying conductivity over five orders of magnitudes. We also plot the FE of Eq. (6) (dash-dot line), showing good agreements. While the metal film

in Figure 8A belongs to a very thin to thin regime, the $h=1000$ nm case shown in Figure 8B certainly belongs to the thick regime for $\sigma_m \geq 10^6 \Omega^{-1}\text{m}^{-1}$; skin depth at 1 THz is 500 nm for a conductivity of $\sigma_m = 10^6 \Omega^{-1}\text{m}^{-1}$.

Next, we consider the effects of a substrate. Assuming an infinitely thick substrate and applying energy conservation and Ampere’s law, it is straightforward to show that

Figure 8: Field enhancement with varying conductivity of metal.

(A) Field enhancement for a 1-nm-wide gap in 50-nm-thick film at 1 THz is plotted in terms of conductivity, calculated by FDTD and analytical formula. In particular, typical conductivities of a heavily doped Si (p+), VO₂, lead, and gold cases are denoted in blue arrows. (B) Enhancement factors for 5-nm-wide gaps in several films with thickness of 100, 200, 500, and 1000 nm at 1 THz are plotted in terms of conductivity.

$$FE = \frac{\frac{\lambda}{\pi h}}{1 + \frac{h_0 n_s + 1}{h \cdot 2}}, \quad (7)$$

where n_s is the index of refraction of the substrate and the electromagnetic field is incident from the air side. In obtaining Eq. (7), as before, it has been assumed that the current flows straight into the gap. However, even for the high aspect ratio gaps, the asymmetry caused by the substrate would bend the current flow, resulting in loss of FE .

Although the result of FE from our model is surprisingly simple, we would like to emphasize that the same FE rule can be obtained from more rigorous analytic theory. Coupled mode theory with single-mode approximation, dealing with boundary conditions of Maxwell's theory with single quantized waveguide mode in the gap, provides that the FE by a gap in very good conductor on the substrate can be written as [177]

$$FE = \left| \frac{4}{e^{-ik_0 h}(1+W_s)(1+W_0) - e^{ik_0 h}(1-W_s)(1-W_0)} \right|, \quad (8)$$

where W_0 and W_s are the light-gap coupling factors defined by

$$W_0 \equiv k_0 \frac{1}{2w_0} \int_0^w dx' H_0^{(1)}(k_0 \sqrt{(x-x')^2})$$

and

$$W_s \equiv n_s^2 k_0 \frac{1}{2w_0} \int_0^w dx' H_0^{(1)}(n_s k_0 \sqrt{(x-x')^2}), \quad (9)$$

with $H_0^{(1)}$ being the Hankel function of the first kind. For a narrow gap ($w \ll \lambda$), the coupling factors can be approximated as

$$W_{0,s} \approx n_{0,s}^2 k_0 w (2i \ln(n_{0,s} w k_0 / 2) + 2i\gamma_E - 3i + \pi) / 4\pi, \quad (10)$$

where $n_0 = 1$ and γ_E is the Euler constant. The explicit form of FE is somewhat complicated, but clearly, a further simplification can be made by the fact that the coupling factor approaches zero as the gap size approaches a deep subwavelength regime. Interestingly, one can readily find from Eqs. (8) and (10) that, in the limit of $w \ll h \ll \lambda$, regardless of the substrate, Eq. (8) can be reduced to

$$FE \approx \frac{\lambda}{\pi h}, \quad (11)$$

which perfectly coincides with our main result of FE in Eqs. (1) and (7) with $h_0 \ll h$. Even though the FE in the ultimate limit is the same with the free-standing case, however, we point out that the FE with a substrate will be generally lower. This is related to the nature of logarithmic dependency of the coupling factor in Eq. (10), which yields, in the presence of the substrate, slower vanishing of the coupling factor as w approaches zero. We point out that the physical account of the reduced FE can be found from the impedance mismatch at the air-substrate interface in the gap that naturally leads to reduction of the tangential component of the electric field near the surface [147].

It should be also noted that, for periodic arrays of slit, FE is limited by the periodicity especially when the

Figure 9: Field enhancement in periodic arrays of slit, clearly exhibiting that the enhancement factor is strongly limited by the subwavelength period P .

This figure is reproduced from Ref. [89].

period is in subwavelength scale, as shown in Figure 9 [89]. This is an intuitive result in that the enhancement is basically related to the funneling of electromagnetic energy and that periodic slits share available energy from the incident wave, giving rise to reduced FE in each slit.

Another relevant physics that should be addressed is the enhancement of the magnetic field with metal nanowire, a Babinet complementary structure of nanogap (shown in Figure 10). Although the strict Babinet principle is applicable only to structures of an infinitesimally thin perfect electric conductor, Koo et al. [68] theoretically demonstrated that a qualitative prediction of FE in a complementary structure can be doable from a metal of finite thickness and conductivity.

2.2 Estimation of the field enhancement from far-field measurements: Kirchhoff integral method

Experimental verification of the huge FE by the direct measurement of near-field is a technically challenging issue because of the limited resolution of the near-field imaging, restricted by the spot size of optical probe beam, which is usually in a few micrometers [178–185]. To improve this issue, a more elaborately complemented scheme is introduced, of which a main idea is based on the Kirchhoff diffraction theory [186, 187] that provides approximated relationship between the near-field information and the signal in sufficiently far distance [69, 188]. A typical experimental setup utilizing the Kirchhoff method to extract the THz FE in the gap is illustrated in Figure 11. This utilizes the transmitted signal through the “normalizing aperture” as reference signal.

The scheme starts from the Kirchhoff integral expression of far-field electric field diffracted by an aperture,

$$\vec{E}(\vec{r}) = \frac{ie^{ikr}}{2\pi r} \vec{k} \times \int_A \hat{n} \times \vec{E}(\vec{r}') e^{-ik\vec{r} \cdot \vec{r}'} da', \quad (12)$$

where r is the distance from the origin, i.e. center of the aperture; A is the aperture area; and $\hat{n}$ is the normal vector to surface A [188]. We note that Eq. (12) can be significantly simplified if the measurement is performed at the diffraction center, $(0, 0, z)$ with $z \gg \lambda$. For a simple instance, assume that we measure a far-field signal from a $w \times L$ -sized slit together with the normalizing aperture of same length L but different width w_a . The far-field signal E_{far}^s from the slit at $(0, 0, z)$ can be written as

Figure 10: FDTD-calculated electric (left) and magnetic (right) field enhancements with nanogap and nanorod, respectively (gold, $h=100$ nm, $w=100$ nm).

This figure is reproduced from Ref. [68].

Figure 11: Schematic for far-field measurement of THz field passing through a gap in metal. Near-field enhancement can be estimated quantitatively by using the Kirchhoff integral method.

$$E_{\text{far}}^s = \frac{e^{ikz}}{i\lambda z} \iint_0^L E_{\text{near}}^s dx dy = \frac{e^{ikz}}{i\lambda z} \langle E_{\text{near}}^s \rangle w L, \quad (13)$$

where $E_{\text{near}}^s(x, y)$ and $\langle E_{\text{near}}^s \rangle$ are the electric field at the aperture-air boundary ($z=0$) and its averaging over the aperture area A , respectively. In the same manner, the far-field signal from the normalizing aperture can be expressed as

$$E_{\text{far}}^a = \frac{e^{ikz}}{i\lambda z} \iint_0^L E_{\text{near}}^a dx dy = \frac{e^{ikz}}{i\lambda z} \langle E_{\text{near}}^a \rangle w_a L. \quad (14)$$

Equations (13) and (14) give rise to

$$\frac{|E_{\text{far}}^s|}{|E_{\text{far}}^a|} = \frac{|\langle E_{\text{near}}^s \rangle| w}{|\langle E_{\text{near}}^a \rangle| w_a} = \frac{|\langle E_{\text{near}}^s \rangle|}{|\langle E_{\text{near}}^a \rangle|} \beta, \quad \beta \equiv \frac{w}{w_a}. \quad (15)$$

This reveals that the ratio of far-field signals, directly measurable quantities, is related to that of the FE s, introducing the scale factor β given by the ratio of w and w_a . Here, in the limit of $w_a > \lambda$, one can find that FE by the reference aperture is close to 1, giving rise to $|\langle E_{\text{near}}^a \rangle| \approx |E_{\text{inc}}|$, where E_{inc} is the amplitude of incident field. This enables a further approximation of Eq. (15), implying that FE can be determined exclusively by quantities measurable in far-field:

$$FE \equiv \frac{|\langle E_{\text{near}}^s \rangle|}{|E_{\text{inc}}|} \approx \frac{1}{\beta} \frac{|E_{\text{far}}^s|}{|E_{\text{far}}^a|} \quad (16)$$

Despite its simple expression, we stress that Eq. (16) was successfully used in various THz experimental studies to estimate FE with reasonable accuracy in comparison with the theoretical predictions [69].

In the presence of the substrate, the FE can be also estimated by the same scheme, taking into account of the

transmission of waves at the interfaces of substrate. At first, as shown in Figure 12, consider the far-field measurement at the substrate side with incident THz wave impinging upon the metal from the top side. If the thickness of the substrate is sufficiently thick, the transmission amplitude of the incident THz waves through the substrate-only sample, E_{sub} , can be approximated by the direct transmission with ignoring the multiple reflection effect. That is, $E_{\text{sub}} \approx \frac{4n_s}{(n_s+1)^2} E_{\text{inc}}$. Then, the far-field transmission amplitude normalized by E_{sub} can be written as

$$|t| \equiv \frac{|E_{\text{far}}^s|}{|E_{\text{sub}}|} \approx \frac{|E_{\text{far}}^a|}{|E_{\text{inc}}|} \frac{\beta \frac{2n_s}{1+n_s} |\langle E_{\text{near}}^s \rangle|}{\frac{4n_s}{(n_s+1)^2} |E_{\text{inc}}|} = \beta \frac{|E_{\text{far}}^a|}{|E_{\text{inc}}|} \frac{2}{1+n_s} FE. \quad (17)$$

Here, we used the result of Eq. (16). Since t is an experimentally measurable quantity, Eq. (17) says that FE with substrate sample can be also quantitatively estimated from the far-field signal by using the Kirchhoff integral method: $FE \approx \frac{1+n_s}{2\beta} \frac{|E_{\text{inc}}|}{|E_{\text{far}}^a|} |t|$.

For the reversed case, i.e. far-field measurement at the top side with incident THz wave impinging from the substrate side, the same scheme can be applied. What is different is that, in that case, the near-field signal radiates to the far-field freely without disturbance by the substrate. Therefore, Eq. (17) for the reversed measurement can be rewritten as

$$|t| \equiv \frac{|E_{\text{far}}^s|}{|E_{\text{sub}}|} \approx \frac{|E_{\text{far}}^a|}{|E_{\text{inc}}|} \frac{\beta |\langle E_{\text{near}}^s \rangle|}{\frac{4n_s}{(n_s+1)^2} |E_{\text{inc}}|} = \beta \frac{|E_{\text{far}}^a|}{|E_{\text{inc}}|} \frac{(n_s+1)^2}{4n_s} FE. \quad (18)$$

Figure 12: Schematics for the Kirchhoff method with substrate.

Interestingly, for the reversed measurement, it is better to just get the FE without using the substrate normalization. Then, Eq. (18) will be simply be $FE \approx \frac{1}{\beta} \frac{|E_{inc}|}{|E_{far}^a|} |t'|$, where t' is the transmission amplitude without going through the substrate normalization process.

2.3 Resonant versus nonresonant field enhancement

What we have discussed so far is basically nonresonant field enabled by the funneling process of THz waves through the gap, which is also related to the “capacitive enhancement”. In contrast to such nonresonant FE , it is also extensively discussed that two-dimensional apertures such as circular or rectangular holes can support resonant FE when certain resonance conditions are met. In terms of strong FE , a rectangular hole, also known as slot antenna, is practically more suitable than other types of holes. This is because of the nature of resonance condition that usually requires a geometric size of the hole to be in wavelength-scale. A slot antenna having an ultrahigh aspect ratio of width w and length l (i.e. $w/l \ll 1$) can utilize resonant light-slot coupling together with capacitive enhancement, maximizing the FE .

There have been extensive investigations to obtain quantitatively accurate FE by single slots in metal [69, 90, 91, 189, 190]. The description of FE is shown to be also quite simple. However, compared to the single-slit case, its behavior is completely different. Analytic calculation based on the coupled-mode theory predicts that the resonant FE by a single slot of width w and length l (shown in Figure 13) is [90, 177]

$$FE_{res} \approx \frac{3l}{2w} \sin\left(\frac{\pi x}{l}\right), \quad (19)$$

where the resonance condition is simply given by

$$\lambda_{res} \approx 2l. \quad (20)$$

In Eq. (19), x varies from 0 to w . By taking average of the FE over the slot area, we have

$$\langle FE_{res} \rangle \equiv \frac{1}{wl} \int_0^w \int_0^l FE_{res} dx dy = \frac{3l}{\pi w} = \frac{3}{2\pi} \frac{\lambda_{res}}{w}. \quad (21)$$

It should be noted that retrieval of FE from the far-field measurement by using the discussed Kirchhoff integral method is related to Eq. (21), not Eq. (19). It is interesting to see that the resonant FE by single slot is given by the aspect ratio of width and length. Clearly, compared to the nonresonant $FE = \frac{\lambda}{\pi h}$, Eq. (21) shows distinctive behaviors.

This simplified FE can be broadly applied to the range of micron gaps to nanogaps. Enhanced electromagnetic field through subwavelength gaps have been intensively explored both theoretically and experimentally over ultra-broadband wavelength regime covering microwave [191, 192], THz [98, 193–195], IR [196, 197], and visible regime [198, 199]. Especially subwavelength photonics has been focused on THz frequencies (0.1–10 THz, 0.03–3 mm), having taken advantage of their relatively large aspect ratio between the wavelengths and structure scales, in turn providing a colossal FE effect. The studies, unavoidably, have strongly relied on the development of the state-of-the-art fabrication technologies for engineering small structures, which have been greatly advanced in the past decade [200–202]. As an earlier work on THz metallic gap structures, micrometer scale rectangular hole arrays were prepared using femtosecond laser machining technique, which guarantees wavelength length scale

Figure 13: Field enhancement in a rectangle slot.

(A) Schematic of single rectangular hole punctured onto a metallic thin film with a length l and a width w and a thickness of h is irradiated by THz incidence. Transmitted THz field distributions for wide gap (B) and narrow gap (C) calculated by FDTD [184].

perforation onto metal thin film (typically several tens to hundreds μm) and somewhat less [203, 204], as shown in Figure 13A.

The resonance can be tuned by changing the length of the rectangular holes, l . Then, as expected, the confined THz field near the gap and FE value increase with decreasing of the hole width (Figure 13B, C and Figure 14). The FE value is independent of the thickness h . Although we discussed the resonant FE only with a single slot case mostly so far, we note that a single slit can also support two different types of resonances: Fabry-Perot-type resonance that appears when the thickness of the slit is comparable to the half-wavelength [64, 205] and the so-called fractional resonance from an ultrathin single slit filled with negative permittivity media [191].

3 THz nonlinearity in few- and subnanometer gaps by quantum electron tunneling

Classical theory of electromagnetism, describing microscopic light-matter interactions in terms of effective dielectric constant, predicts the THz FE by nanogaps in metals, in the form of electric field distribution obtained as a solution of the required boundary conditions at interfaces of metal and free-space [206–210]. The predicted result is quantitatively accurate especially when the gap size is a few nanometers or larger [88, 89, 156], and the enhancement by an infinitely-long nanogap is shown to

increase with decreasing gap size up to the enhancement factor, $\frac{\lambda}{\pi h}$. It is also confirmed experimentally that this behavior is still valid even when the gap approaches the regime of the sub-skin-depth [69].

One immediate question that arises here is whether this increasing behavior continues to the subnanometer regime. Considering the zero-gap limit in which there is no FE and that gap-size-dependent FE changes continuously with varying gap, one can presume that there should be a critical turning point of the gap size at which the near-field strength eventually starts decreasing with even narrower gap. Classical theory predicts that, in the perfect electric conductor approximation widely used in the study of THz wave interaction with metals, there is no such decrease in the FE . Exceptionally, the decreased FE with finite conducting metal is mainly due to the evanescent decay of the fundamental slit waveguide mode that becomes more pronounced with narrower gap in lossy material [91, 148]. Recently, experimental and theoretical studies demonstrated that when the gap size approaches a few nanometers or Angstrom regimes, FE can be strongly modified and the classical theory cannot provide a quantitative understanding without taking into account quantum effects [86, 167, 211–215]. In this regime, numerical schemes such as conventional finite-element method (FEM) and FDTD based on far-field dielectric function of the material also should have an implementation of the quantum effect for accurate calculations of FE .

There are two major quantum effects that come into play when the gap size approaches the nanometer or Angstrom regime. Nonlocal plasmon response [216–230] is an extensively discussed, quantum-mechanical counterpart of the classical local response theory, giving rise to a suppression of the local FE through a modification of effective boundaries of materials within the Fermi-wavelength scale (nm to Angstrom). We note that the nonlocal effect can be directly applied to the FEM and FDTD schemes by adapting quantum-corrected dielectric functions of the materials. It should be also noted that, in some extent, the nonlocal effect is related to the Landau plasmon damping [231–233] that results in additional suppression of the plasmonic electric field in the proximity of metals [234].

In this section, we discuss the other important quantum effect, the electron tunneling [235–238], through a potential barrier of nanogap at THz frequencies [76, 80, 86], incorporating recent studies. In the point of view of electrons, a gap in a metal plate can be assumed as a tunnel junction [239, 240], as shown in Figure 15A. Usually, the gap size for an efficient electron tunneling needs to be subnanometer for typical potential barriers of the gap, a

Figure 14: Normalize-to-area light transmission through single rectangular slot in perfect electric conductor plate, implying that the total transmission is almost independent of the width w . Here, $\lambda_c = 2l$ is the resonance condition. This figure is reproduced from Ref. [48].

Figure 15: Electron tunneling through a nanogap. Tunnel junction (A) without and (B) with strong electric field in the gap, E_{gap} . Here, η is the Fermi level and ϕ is the barrier height.

few eV. As shown in Figure 15B, an intense electric field is required to pull down the barrier for tunneling. We will see that, thanks to the ability of the nanogap in metal to capture the intensive THz field, the THz quantum tunneling and consequent strong nonlinear behavior result both in subnanometer [79, 80, 85] and even in super-nanometer gaps [76, 86].

3.1 Fabrication of wafer-scale nanogap arrays

A challenging issue for the experimental studies on THz plasmonics with few- or subnanometer gap is sample fabrication because of extremely high ratio of photon wavelength and gap size. To have a reliable THz field interaction with nanogap and consequent plasmonic behaviors such as giant local FE and nonlinearity, the length of the gap should be at least in the scale of photon wavelength. Therefore, in pursuit of sufficiently intense far-field signal, the sample should cover several mm^2 area with nanogap

Figure 16: High-resolution and high-throughput technique for nanogap array fabrication.

arrays of ultrahigh aspect ratio. To realize such large-area sample with high-resolution patterning, various recipes have been introduced, e.g. fs-laser machining for micro-scale punctured structures [64, 241, 242], focused ion beam [243, 244], e-beam lithography [245, 246], and photolithography techniques [125, 247, 248] for metamaterial structures and, finally, atomic layer deposition (ALD) for atomic-scale gap structures [75, 193, 249, 250]. For nanoscale gap structures, photolithography can be considered as a promising fabrication method for mass production at the same time, as shown in Figure 16. The stepping method using patterned mask guarantees several hundreds of nanometers in gap width with hundreds of microns in gap length, required for resonant enhancement of THz field (Figures 13 and 14). Also, by varying the length of the gap, l , one can obtain frequency tunable THz resonators (Figure 17A), showing a clear relationship between l and the fundamental resonance frequency, f_{res} (Figure 17B).

Figure 17: Transmittance with varying slot length.

(A) Measured THz transmittances for various slot arrays. (B) The resonance frequencies for different slot arrays are plotted in terms of the slot length.

For the ultimately small subnanogaps, on the one hand, a spacer deposition method [75, 251] provided strong reliability in terms of preventing collapsing of the gap and controllability of precise thickness, enabling a 1-nm gap uniformly formed with several 100 microns length.

Figure 18 illustrates the sample fabrication scheme introduced by Jeong et al. [102], an improved version of the previously introduced scheme by Chen et al. [75] (Figure 19). Firstly, on the 3 nm chromium (Cr)/100 nm Au/sapphire substrate, 30 nm Cr and 150 nm aluminum Al layers are patterned by using a standard photolithography and liftoff process. This double layer is a sacrificial layer that will be used for removing excess metals and making the structure planar. Then, because Al is

resistant to ion beam, ion milling is applied to remove exposed Au area while Au underneath the Al/Cr layer is sustained. After that, the ALD of aluminum oxide (Al_2O_3) is applied to form a uniform clad on the whole structure with nanometer-scale thickness. After the deposition of a second Au layer with adhesive Cr layer, Al/Cr wet etching is applied to remove the overhanging Au and Al layers.

In the above recipe, the nanogap is defined by the ALD of Al_2O_3 , providing controllability in nanometer accuracy. Based on spacer deposition and filling schemes, and by adding a step of sacrificial layer, this method provides high-throughput fabrication of nanogaps with an ultrahigh aspect ratio.

Figure 18: High-throughput fabrication of arrays of nanoslits with ultrahigh aspect ratio introduced by Jeong et al. [102].

Figure 19: Wafer-scale atomic layer lithography.

(A) A patterned substrate coated with a thin Al_2O_3 by using ALD. (B) Additional metal evaporation to plug the second layer. (C, D) Removal of the excess metal by using an adhesive. (E) Optical micrograph of 5-nm gaps in silver. White scale bar indicates 0.2 mm. (F) Half of a Si wafer planarized by using an adhesive. This figure is reproduced from Ref. [75].

Figure 20: Fabrication of vertically oriented graphene spacer in Cu-single layer graphene (SLG)-Cu composite.

(A) Schematic of the fabrication procedure. (B) Cross-sectional image of Cu-SLG-Cu array. (C) Top views of SEM (left), reflection type (center), and dark field scattering (right) optical microscope images. This figure is reproduced from Ref. [80].

Shown in Figure 20 is fabrication of Angstrom gaps introduced by Bahk et al. [80]. First, on a patterned 300-nm-thick Cu film on a quartz substrate, a single-layer graphene (SLG) is seamlessly grown to cover the sidewalls. Then, an additional thinner Cu layer is deposited on the same sample by thermal evaporation. Finally, the second Cu layer is selectively peeled off by using an adhesive tape: the final result is Cu-SLG-Cu composite as shown in Figure 20C.

3.2 Nonlinear THz response by light-induced electron tunneling through nano- and Angstrom-sized gaps

A basic strategy to realize the THz nonlinearity is utilizing deep subwavelength gap to induce huge electric field that consequently pulls down the barrier in the tunnel junction by the potential energy $V = eE_{\text{gap}}w$, where e is the charge of electron, E_{gap} is the induced electric field in the gap, and w is the gap size. Since the electric field normal to the metal surface is related to the surface charge, E_{gap} can be quantitatively determined by the accumulated surface charges at the two metal walls in the gap. This implies that, once electron tunneling occurs, a further charge accumulation by the incident THz field is limited and the FE will be reduced. Also, because the pulling down of the potential barrier increases with stronger E_{gap} , naturally more intense THz field leads to more electron tunneling through

the potential barrier, giving rise to more pronounced nonlinear responses in local electric FE and transmission of THz waves through the gap.

As discussed above, THz nonlinear behaviors through the nanogaps have been introduced by recent experimental studies [86, 252, 253]. In Ref. [86], arrays of nanogap are prepared based on the previously discussed ALD method with the sacrificial layer [75, 86]. An electron microscope image of the nanogap is shown in Figure 21A. Four different samples with selected gap sizes (1.5 nm, 2 nm, 5 nm, and 10 nm) are prepared to examine the THz nonlinearity depending on the tunneling probability. Shown in Figure 21B is the cross-section images of a 1.5-nm nanogap taken by a scanning electron microscope (top) and a scanning transmission electron micrograph (bottom). Note that the Al_2O_3 layer, presented in Figure 21B as a white area, defines the gap size, and its thickness, i.e. gap size, is controllable by the number of ALD cycles. The main result of this study is shown in Figure 21C. The THz transmission dependent on both E_{gap} and gap width clearly shows that the tunneling process works to reduce the THz transmission, corresponding to the reduced local electric field. Also, we note that reduced transmission in a nanogap with smaller width is more sensitive to an increase in E_{gap} , which is an intuitive result from that the smaller gap allows electron tunneling with higher probability. Nevertheless, it is clear that for the super-nano-gaps, what is most important is not the transient voltage applied to the gap so much as the electric field on the gap.

Figure 21: Nonlinearity in THz transmission through nanogaps.

(A) Optical micrograph image of arrays of nanoslot of 10 nm gap size. (B) Cross-section image of a 1.5-nm-wide gap from an SEM (top) and an STEM of the Al₂O₃ layer between gold (bottom). (C) THz transmittance with varying electric field amplitudes inside the gap. The dashed region (right) indicates where the gap is damaged by the induced electric field. This figure is reproduced from Ref. [86].

A nonlinear THz transmission can be quantitatively studied based on the dielectric response of the gap associated with the modal expansion method. This is an effective medium model in which the contribution of tunneling is interpreted as the transient modification of an effective dielectric function in the gap. As shown in Figure 21C, tunneling yields the tunneling current density, J , redistributing the induced surface charges near the gap. For a time-harmonic THz wave with angular frequency, ω , the polarization $P = i(J/\omega)\exp(-i\omega t)$ can be obtained, and consequently, the effective dielectric function ε can be described as

$$\text{Im}\left(\frac{\varepsilon}{\varepsilon_0}\right) = \frac{J}{\varepsilon_0 \omega E_{\text{gap}}}. \quad (22)$$

The explicit form of the current density, J , dependent on both the gap size and the dielectric function of filling medium, i.e. Al₂O₃ in this case, can be evaluated by

the image force model [254]. By applying Eq. (22) to the nanogap with a filling medium possessing effective dielectric function, the FE can be obtained in analytic ways that we discussed in Section 2. As shown in Figure 21C, the calculated results are in very good agreement with experimental results.

Recently, squeezing THz waves into much narrower gaps, Angstrom-size gaps, and consequent tunneling effect resulting in significant reduction of light transmission were demonstrated with the help of two-dimensional (2D) van der Waals (vdW) materials [80]. A heterostructure of 2D SLG and an effective vertically aligned 3 Å gap in copper layer, formed by Cu/1.5 Å gap/Cu/1.5 Å gap/Cu double vdW gaps of a 5 mm length, has revealed extreme THz nonlinearity with unprecedented 97% reduction of the transmittance through the gap (Figure 22). This self-limiting, gigantic optical nonlinearity is achieved by a massive THz funneling through Angstrom gaps, strongly pulling down the gap barrier and boosting the electron tunneling.

Figure 22: Nonlinearity in THz transmission through Angstrom-sized vDW gap.

(A) Cross-sectional TEM image of Cu-SLG-Cu composite (top). The green curve is vertically averaged line profile of the composite. The red curve (middle) denotes the plane-averaged inverse valence electron charge density. (B) Maximum normalized transmittance with varying incident THz field E_0 . Inset represents retrieved FE calculated by using the Kirchhoff method. (C) Measured (dots) and calculated (curve) nonlinear optical response where T_0 is the transmittance without nonlinear response. This figure is reproduced from Ref. [80].

We note that the discussed THz quantum tunneling spans the boundaries of the study on quantum effects in microscopic light-matter interaction: down to the low frequency of 0.1 THz, up to strong field amplitudes of 5 V/nm, with barrier width from Angstrom to super-nanometer regimes.

4 Nanostructure-based absorber integrated terahertz sensor applications

Since many intramolecular and intermolecular vibration modes of molecules, including nucleobase [255–257], amino acid [258–261], and protein [262–264], exist at THz frequency range, THz technology has been considered a promising mean for detection and spectroscopy of such small biomaterials. Even bigger and more complex biological structures such as cell [265–268] and tissues [269, 270] can be explored with THz spectroscopic tools as well.

Unlike ultraviolet light or X-rays, the low photon energy of THz (1 THz = 4 meV) is an advantage to use because of noninvasiveness and nonionizing manner [271, 272]. Nevertheless, an extremely small absorption cross-section has limited widespread use in the THz regime. In most spectroscopic cases, chemical compound samples were prepared in closely packed pellet form to get reasonably big signal. Furthermore, bio or chemical samples in solution state have huge water absorption at the THz frequency, and thermal fluctuation at room temperature can hinder high-throughput applications. Sometimes, the experiments have been performed at a low temperature to rule out thermal fluctuation issues [273]. In this respect, recently advanced THz detection technologies assisted by micro-to-nano patterned (gap) structures can provide an excellent solution to these fundamental [103, 274–285]. Giant absorption cross-section enhancement of molecules inside nanogap structures will be discussed in this section showing a completely new type of label-free detection methods in room temperature. Examples of recent works will show that various type of samples are detectable even

in very low concentration, including chemicals, protein, microorganism, and even viruses.

4.1 Increased molecular absorption cross-section by THz nanogaps

Metal nanogap structures can enormously enhance and strongly localize the THz electric field at a hot spot, increasing the THz absorption coefficients of molecules [274]. Using a single nanogap structure in gold, it has been demonstrated that the molecular cross-section and absorption coefficient are enhanced by a factor of $E_{\text{gap}}/Z_0 H_{\text{gap}} \sim 10^3$, where H_{gap} is the magnetic field inside the gap. The origin of increased absorption coefficient is a strong asymmetry between THz electric and magnetic FE s: over a thousand times for the electric field, but in order of one for the magnetic field inside the gap. By considering the cross-section $\frac{dS}{dz} = -\sigma NS$ where dS is the Poynting vector absorbed between the points z and $z + dz$ along the path of a THz beam, N is the number of absorptive molecules per unit volume, and σ is the molecular absorption cross-section, molecular absorption can be calculated as follows: the Fermi Golden rule says that one molecule absorbs $\frac{2\pi}{\hbar} \mu^2 E^2 \rho(\hbar\omega_0) \hbar\omega_0$ (Joule per second), where μ is the electric dipole moment of the molecule, $\rho(\hbar\omega_0)$ is the density of states, and ω_0 and E are the resonant angular frequency and the local electric field of light, respectively. A volume, Adz (A is a surface area and dz is a differential thickness), can be defined, inside which the molecular absorption is occurring. By energy conservation, we have

$$\begin{aligned} [S(z) - S(z + dz)]A &= (NAdz) \times \frac{2\pi}{\hbar} \mu^2 E^2 \rho(\hbar\omega_0) \hbar\omega_0 \\ \frac{dS}{dz} &= -N \times \frac{2\pi}{\hbar} \mu^2 E^2 \rho(\hbar\omega_0) \hbar\omega_0 \equiv -\sigma NS \\ \sigma &= \frac{2\pi}{\hbar} \mu^2 E^2 \rho(\hbar\omega_0) \frac{\hbar\omega_0}{S} = \frac{dS}{dz} = \frac{2\pi}{\hbar} \mu^2 \rho(\hbar\omega_0) \hbar\omega_0 \frac{E^2}{S} \end{aligned} \quad (23)$$

The third line of Eq. (23) implies that the molecular absorption cross-section, σ , is sensitive to the electromagnetic environment of E^2/S , which greatly increases in the nanogap relative to the vacuum, owing to the asymmetric electromagnetic enhancements. A single nanogap with varying gap width from 50 nm to 5000 nm was used as a launching pad for strongly enhanced and localized THz field and a sensing hot spot at the same time. The first tested sample was an RDX (1,3,5-trinitroperhydro-1,3,5-triazine)

powder that has an absorption feature around 0.8 THz, attributed to a molecular conformation or a weak hydrogen bond between RDX molecules. Therefore, the sensing chip with a fundamental resonance at 0.87 THz was designed to selectively enhance the absorption frequency and used to detect RDX in a very low amount as shown in Figure 23.

The strongly increased molecular cross-section by $>10^3$ was translated into a colossal absorption coefficient of $\sim 170,000 \text{ cm}^{-1}$. Thereby, extremely small quantities about 40 ng (even 22 fg inside the gap) could be detected [260]. A theoretical model was also introduced to explain this and is in good agreement with a reference [286]. At this point, it should be noted that there are several critical parameters in designing the THz nanoslot antenna in terms of the target samples. First, the fundamental resonance frequency shift for the nanogap structures should be accounted for molecule detection. When bio and chemical compounds are filled inside the gap, their effective refractive index can significantly affect the measured THz spectrum. The resonance frequency can be determined

by the following relation [90]: $f_{\text{res}} = \frac{c}{\lambda_{\text{res}}} = \frac{c}{\sqrt{2(n^2 + 1)l}}$, where l is the length of the gap and n is the real part of the complex refractive index. This means that, because the refractive index is dependent on the covered (drop-casted or filled) dielectric sample material, so is the shifting behavior of the resonance frequency [103, 275, 280]. Also, the gap width is a very important parameter related to the amount of the THz FE [188]. As discussed in the second section, a narrower gap yields a stronger THz FE , resulting in the increase in detection sensitivity. The gap-size-dependent detection sensitivity can be directly observed in the measurement of an extremely small amount of pesticides [287]. According to the measurements in Ref. [287], the observable minimum concentration level of targeted methomyl solution is determined by the gap size, and surprisingly, using 500 nm and 100 nm width gap antennas, 8 ppm and 10 ppb of methomyl at the measureable limit were detected, respectively. This observable limit value is essential for the label-free sensing [288], especially for extremely low concentration of chemical and biological residual substances.

4.2 THz nanogap sensor applications for chemical compound identification

As previously discussed, enhanced THz field by metallic nano structures can provide highly advanced sensing performance basically in label-free [289], noncontact,

Figure 23: Interaction of molecules with THz nano antennas.

(A) Schematic of the enhanced absorption cross-section of RDX molecules inside a THz nanogap. (B) Absorption spectrum for 1 mg of RDX molecules onto the bare substrate. (C) Transmission spectra for RDX onto the substrate without nanogap. (D) Transmission spectra for a single THz nanoslot antenna without and with RDX ranging from 40 ng to 2 μg. This figure is reproduced from Ref. [274].

and noninvasive manners. THz optical characteristics of various biomaterials can be represented in terms of the dielectric response in the THz range. For example, the refractive indices for various saccharides can be extracted from the transmitted THz spectra in the range of 0.5–2.5 THz [290, 291]. Using the fast Fourier transform, the THz signal in time domain is converted to the amplitude and phase spectra in the frequency domain. Then the complex optical constants can be calculated with the waveforms for reference (typically void aperture) and target sample as a film (or pellet) form as follows:

$$E_s(\omega) = E_r(\omega) \cdot \exp\left(-\frac{d \cdot \alpha(\omega)}{2}\right) \cdot \exp\left(i \frac{2\pi}{\lambda} n(\omega) d\right), \quad (24)$$

where $E_s(\omega)$ is the amplitude of the transmitted signal through the sample and $E_r(\omega)$ is the amplitude of the input signal through empty space occupied with the film. $n(\omega)$ and $\alpha(\omega)$ are the real parts of the refractive index and absorption coefficient, respectively, and d is the thickness of the film. The power absorption extracted from the difference between the spectral amplitudes passing through the sample and reference is attributed to the imaginary

part of the refractive index, $\kappa(\omega)$. The real part of the refractive index, $n(\omega)$, is obtained from the phase difference between two signals as

$$n(\omega) = 1 + \frac{\varphi_r - \varphi_s}{2\pi d} \lambda, \quad (25)$$

and the absorption coefficient,

$$\alpha(\omega) = -\frac{2}{d} \ln(T) = -\frac{2}{d} \ln\left(\left(\frac{E_s(\omega)}{E_r(\omega)}\right)^2\right) = \frac{4\pi k}{\lambda}, \quad (26)$$

where φ_r is the phase of the reference waveform, φ_s is the phase of the signal that passed through the sample, and λ is the wavelength. So, in conclusion, the extracted optical parameters from the time-domain THz signals can be used in identifying the substances. On the other hand, the optical characteristics including absorption and refractive index should be further modified in the nanogap antenna system.

To closely investigate how the enhanced electric field can contribute to the detection situation, FDTD numerical simulation was implemented. We adapted a nonuniform meshing tool with a 10-nm grid at minimum to describe

a deep subwavelength thickness of the film. The detectable target samples were assumed as homogeneously dielectric and somewhat absorptive clads with a certain thickness (much less than the wavelength), having the complex refractive index as $A \cdot n + iB \cdot \kappa$, where A and B are constant values over the broadband frequency regime in the range of 1.0–3.0. Using the auxiliary differential equation method, the absorptive media were implanted in the gold nanogap antenna.

Because the directly transmitted THz field passes the absorptive medium with a trend of exponential decay, the reduction of the transmission can be described as $\frac{T_s(\omega)}{T_r(\omega)} = C \cdot e^{-\kappa k(\omega)h}$, where $T_r(\omega) = (E_r(\omega))^2$ and $T_s(\omega) = (E_s(\omega))^2$ are the transmittances through the nanoantenna without and with the clads, C is the transmittance ratio at the air to clad interface, $k = 2\pi/\lambda$ is the incidence momentum, and h is the thickness of the clad, proportional to the molecular concentration (Figure 24A). As the clad increases from 0.5 to 5.0 μm , the transmittance decreases at the resonance frequency, an indication of increased absorption (Figure 24B).

On the one hand, the transmittance can be affected by the complex refractive indices as plotted in Figure 25A. While absorption is accompanied by the change in the imaginary part of the refractive index, κ , the change in the real part of the refractive index, n , the resonance frequency is mainly affected by n . The various changes in transmission spectra, therefore, can be clear evidence to discriminate the species of the target sensing materials. As plotted in Figure 25B, the peak of transmittance clearly decreases in terms of the imaginary part of the refractive index, κ ; meanwhile, the resonance frequency mainly related to the real part of the refractive index, n , shows a small change. However, it should be noted that

Figure 24: An interpretation of molecule interaction with nano antennas.

(A) Schematic of the cross-section view of the absorptive clad covered onto the gold nanogap antenna. (B) Normalized transmittance to the incidence was calculated for different thickness of clads from 0.5 to 5.0 μm in thickness.

a stronger absorption can give an appreciable change in the resonance frequency.

In Ref. [276], different types of saccharide molecules possessing different molecular vibration modes (e.g. glucose at 1.4 THz and sucrose and fructose at 1.7 THz) were clearly distinguished using different length of gap antenna based sensing chips with different fundamental

Figure 25: Light transmission with varying imaginary part of the refractive index of the clad.

(A) Normalized transmittance to the incidence was simulated for various complex refractive indices. (B) The peak of the transmittance value and the resonance frequency were plotted in terms of the refractive index.

Figure 26: Detection of the sugar molecule by a THz metamaterial.

(A) Schematic of the sugar molecule detection using a nano-antenna array-based sensing chip. (B) Normalized THz spectra measured for a bare substrate used as a substrate and 250 mg/dl of glucose on the same Si substrate. (C) Normalized THz spectra measured with the glucose antenna for D-glucose and (D) sucrose molecules. (E) The changes in the maximum values of the normalized transmittances are plotted for D-glucose, sucrose, and cellulose as a function of the molecular concentration level. (F) Frequency shifts at the maximum transmittance for three samples are plotted. This figure is reproduced from Ref. [276].

resonance frequency as shown in Figure 26. Using glucose-antenna ($f_{\text{res}} = 1.4$ THz), only the glucose sample shows the sensitively changed transmission spectrum; meanwhile, other saccharide samples do not. In the imaging data performed with the fructose-antenna ($f_{\text{res}} = 1.7$ THz), a strongly changed image contrast was observed at the fructose sample dropped area.

Applying this concept to various biomolecules and chemical molecules with specific vibration modes at

broadband THz frequency regime, target samples in unprecedentedly low concentration level can be detected. This is valid even in case of more complex biological systems including protein, cell, and tissues with no specific spectral features due to their superposition and broadening of the spectrum. Such relatively large biomaterials can be considered a combination of many proteins, with vibration modes with inhomogeneous broadening such that no recognizable absorption features exist in the

Figure 27: Detection of complex biological system by using THz metamaterial.

(A) THz transmission amplitudes for bare substrate with (red solid line) and without (black dashed line) deposition of penicillia. (B) Microscope images of metamaterials with the deposition of penicillia (left) and after fungicide treatments (right). (C) THz transmission amplitudes measured before and after the fungicide treatment. This figure is reproduced from Ref. [275].

THz spectrum [292]. In those cases, especially, the spectral shift can be treated as a key parameter. As shown in Figure 27, the resonance frequency shift of the metamaterial by the fungicide treatments is interpreted in terms of the number density and the dielectric constants of the microorganisms inside a gap area.

Besides single resonance gap structures, ultra-broadband resonance filters composed of several gap antennas with log-periodically varied lengths and periods in their arrangement at the same time can be an excellent alternative, utilizing their field-enhancement advantage but freely applicable to any target frequency [57, 189]. Such ultra-broadband sensing chip has a decided merit for unknown targets, in other word, samples without any obvious spectral features in THz fingerprinting. This is very useful to identify and quantify such virus samples as shown in Figure 28 [282].

THz optical characteristics based on the transmission reduction and resonance frequency shift by the covered virus samples can be mapped for different subtypes of the viruses and their quantifications. The suggested sensitive and selective THz detection in the reference provides abundant optical information of measured viruses, suggesting quick and accurate monitoring and rapid diagnosis of viruses.

5 Discussions and outlook

Maxwell's theory provided descriptions of light interaction with metal in terms of microscopic coupling between incident electromagnetic waves and charges residing in metal. This immediately allowed manipulation of the moving of charges by the external light, which is now developed to colossal field enhancement by accumulating charges near a narrow gap in metal. This huge field enhancement is in particular important in the THz spectral regime as it can partly compensate the relatively low average power of THz sources and small cross-sections [293]. Nonlinear THz response [294–299] is a prime example utilizing intense THz field that boosts light-matter interaction [300, 301]. Furthermore, it is very clear that integration of the huge field enhancement in a gap with other novel optical/electrical/plasmonic properties of materials [302–304] or devices [9, 305–313] can expand the practical use of the THz field. Graphene-integrated plasmonic system [34, 129, 314–318] is one immediate example, enabled by atomically thin and electrically tunable nature of graphene (Figure 29). We also expect that, considering the reciprocity of electromagnetic radiation [319], the light-capturing ability of nanogap could be a key principle to extract and radiate THz waves from the integrated devices.

Although we have mainly discussed resonant and nonresonant field enhancements in fundamental types of gaps, i.e. slit and rectangle slot, there are many other opportunities to realize even higher field enhancement. For instance, coupling of two or more symmetric/asymmetric gaps can be considered to obtain field enhancement by symmetric and antisymmetric resonance modes [71, 320, 321]. Also, localizations of incident/transmitted light before/after the gap can be incorporated to increase the field enhancement in the gap [322–325] (Figure 30). The other example is to make use of hybridization of a gap with plasmonic metastructures or metaelements [71, 114, 191, 320–329] that results in intensive localization of electric field in the form of surface waves such as vortex-plasmon and spoof surface plasmon modes.

Figure 28: THz metamaterials for versatile detection of molecules.

(A) Absorption spectra for pallet types of virus included a protein sample (H9N2) and a control without virus. (B) THz detection of virus samples in liquid state using a nano slot-antenna array-based sensing chip. (C) Optical images of dropped virus solutions onto the multiresonance nano-antenna array before (top) and after (down) THz excitation. (D) Transmittance spectra through multiresonance nano-antenna with and without H9N2 virus. (E) The difference in transmitted intensity (ΔT) and shifted resonance frequency from each fundamental resonance peak (Δf) for H9N2 virus. This figure is reproduced from Ref. [282].

6 Conclusions

In this review, we discussed both the fundamentals and applications of THz wave interaction with deep sub-wavelength nanostructures. The enormous THz field enhancement can be explained as a result of surface charge accumulation near the nanogaps, with narrower gaps supporting stronger electric field enhancement. The

fundamentals of giant THz field enhancement at an infinitely long nanogap were introduced in Section 2. Specifically, nonresonant THz field enhancements by nanogaps with both sub- and super-skin-depth thicknesses were discussed with a simple model based on the boundary conditions of Maxwell's equations. From this model, we obtained a surprisingly simple description of the field enhancement valid over many orders of magnitudes

Figure 29: Graphene-integrated plasmonic metal structures for THz modulation, utilizing tunability of graphene and strong local field enhancement.

A hybrid structures of (A) graphene and ring apertures, and (B) graphene and an array of slits. A and B are reproduced from Refs. [34] and [129], respectively.

Figure 30: Boosted field enhancements by localization of incident/transmitted light by using additional plasmonic couplers.

(A) Coupling with split-ring-resonators, and (B) plasmonic islands. A and B are reproduced from Refs. [322] and [323], respectively.

of conductivity of metal as well as many orders of magnitudes of the film thickness. In terms of the real-world measurement, we revisited the Kirchhoff integral formalism that enables quantitative estimation of near-field enhancement by typical far-field measurements. At the end of Section 2, resonant THz field enhancement by critically defined boundaries such as rectangular slot shapes was also discussed. In Section 3, more extreme cases with deep subwavelength structures from nanoscale to Angstrom scale were discussed with some example of

plasmonic quantum effect and electron tunneling with consequent nonlinear behaviors. Enhanced THz nonlinear phenomena through nano- to Angstrom-sized gaps show a new pathway for the observation of unprecedented resonant and nonresonant changes in various. In Section 4, finally, a new type of THz molecule sensor based on the field enhancement via subwavelength structures, a promising sensing tool for chemistry, biology, and medical applications was introduced. As one of representative applications, ultrasensitive and highly selective

THz molecule sensor has been suggested. The ultrasensitive THz molecule sensing mechanism follows the huge THz electric field enhancement via nanogap structures, leaving the magnetic field magnitudes almost intact but with very small spatial curvatures introduced, resulting in greatly increased absorption cross-section of the covered substances. Since many intramolecular and intermolecular vibration modes of molecules exist in the THz range, frequency controllable THz metamaterials can be used as special molecule targeted sensors. As a further step, it can be applied for real-time capturing of some of the physical dynamics prominent in biological systems.

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