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Terahertz Localized Surface Plasmon Resonances in Coaxial Microcavities

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W.W. dedicates this work to the memory of Boonchai Withayachumnankul.

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Coaxial microcavities etched into the surface of a doped silicon substrate are shown to support localized surface plasmon resonances (LSPRs) at terahertz frequencies. The underlying mechanism involves coupling of freely propagating terahertz waves with surface plasmon polaritons (SPPs), which propagate in a coaxial mode along the cavity walls in the axial direction. A Fabry-Pérot resonance is built up when the SPP wavenumber appropriately relates to the cavity depth. Owing to the Ohmic loss of the silicon at terahertz frequencies, the energy of the resonating SPPs is largely dissipated, leading to a modified reflection spectrum. Strong field enhancement is observed inside the cavities at resonance. The theoretical analysis

is supported by numerical and experimental results. This study is a promising pathway for development of terahertz devices with applications in the areas of photonic integrated circuits, molecular sensing, and subwavelength imaging.

1. Introduction

The last twenty years have seen significant progress in terahertz research, driven by the introduction of coherent sources and detectors such as photoconductive antennas, electrooptic crystals, and quantum cascade lasers. In recent years, terahertz technology has seen another large leap with the advent of solid-state terahertz sources operating at room temperature.^[1–3] This latest development may potentially propel some fundamental concepts, including terahertz sensing, imaging, and communications, towards commercial products. Along the path of terahertz technology development, increasing the wave confinement beyond the diffraction limit is a major challenge with significant implications. Enhanced wave confinement promises increased spatial resolution that could potentially lead to single-molecule sensing in the terahertz region, where numerous substances exhibit rich spectroscopic features.^[4] Integrated optics essential for terahertz communication applications is another area that could benefit from this breakthrough through component miniaturization. This challenge is extremely relevant due to the relatively long wavelengths of terahertz waves. The spatial confinement of a propagating wave is limited by diffraction, and is thus a function of the wavelength. This limitation can be circumvented by using surface plasmon polaritons (SPPs),^[5] the bound electron oscillations propagating along the interface between a dielectric and a conductor. For Drude conductors, SPPs can be sustained at frequencies right below the plasma frequency, where the real permittivity is slightly negative and the imaginary part is small. This region lies in the visible and UV ranges for most metals. At terahertz frequencies, the properties of metals approach a perfect electric conductor (PEC), leading to field

delocalization into grazing waves or Sommerfeld-Zenneck waves.^[6–8] Periodically-structured metal surfaces can help to increase the confinement of surface waves at these frequencies to achieve so-called designer or spoof SPPs.^[9–12] Additionally, moderately-doped semiconductors with a plasma frequency dependent on the carrier concentration are shown to support terahertz SPPs.^[13–16]

A localized surface plasmon resonance (LSPR) is a manifestation of surface plasmons that can exist in subwavelength conductive elements. Typically, the resonance takes place at equilibrium between the charge displacement force, *via* an external excitation, and the ionic restoration force. The size and shape of elements, together with the properties of materials, determine the resonance characteristics. Recently, a few different types of elements made of semiconductors have been shown to support LSPRs at terahertz frequencies, including dipoles,^[17] bowties,^[17,18] and coupled disks.^[19] Owing to strong field localization in a subwavelength volume, terahertz LSPRs are very attractive for many applications, including sensing and imaging.^[20] As an alternative to resonant plasmonic elements, complementary elements in the form of resonant cavities have a potential to sustain terahertz LSPRs, but have yet to be realized. Nonradiative modes of these resonant cavities promise minimal inter-cell coupling, whilst the self-supporting structure of the cavities reduces the fabrication complexity.

In this article, a form of LSPR cavities, namely coaxial microcavities, is investigated and realized at terahertz frequencies. A single coaxial microcavity can be described as a blind annular hole etched into a doped semiconductor wafer, which intrinsically supports terahertz SPPs. The resonance mechanism is closely related to that of optical coaxial (or annular) aperture arrays, which exhibit extraordinary transmission via the interplay between cylindrical SPPs and cavity resonance modes.^[21–25] Another related structure is an array of coaxial nanocavities with a V-shaped groove, which has been demonstrated recently at optical

frequencies on metallic surfaces.^[26,27] As opposed to the straight profile, the tapered profile of the groove gives rise to the depth dependence of the SPP characteristics. These optical apertures and cavities form a basis for the study of straight-profile coaxial microcavities sustaining LSPRs at terahertz frequencies. It is worth noting that the principle of these optical and terahertz coaxial cavities is radically different from the realization of terahertz blind annular holes on a copper surface.^[11,12] In these studies, terahertz spoof SPPs were established over the metallic surface, which was textured to realize an effective plasma frequency. Those holes were operated below the cutoff to allow evanescent fields perpendicular to the apertures, with no surface plasmons and LSPRs in the holes.

2. Design, Fabrication, and Experiments

A unit cell of the coaxial microcavity is illustrated in **Figure 1**, along with the dimensions and the incident field vectors. The cavity is described as an annular air-filled groove with a straight cross-sectional profile that is etched into a homogeneous material, in this case a moderately doped silicon substrate. Each sample is composed of thousands of identical cavities, periodically arranged as a 2D array on the substrate. As shown in **Table 1**, samples with two different cavity depths are investigated to observe variations of the plasmon resonance. The average radius of the annular groove is fixed for all of the designs, resulting in a nearly constant cutoff frequency of the resonance mode. The various wall separations introduce different conditions for the mode confinement. The wafer is thick enough to suppress field transmission to the other side. The choice of silicon as a substrate is based on availability and manufacturability, although InSb has a higher carrier mobility or equivalently lower dissipation. It will be demonstrated later that doped silicon can reasonably support non-propagating terahertz LSPRs, despite a moderate electron mobility and hence a moderate plasmonic absorption.

According to the specification, the silicon wafers are (100) oriented and n-type doped with phosphorus. The material resistivity lies in between 0.02-0.05 Ω cm. In order to experimentally determine the optical properties of these wafers at terahertz frequencies, the measured complex reflectivity of the bare silicon wafer is fitted with the Fresnel equation incorporating the Drude model.^[28] In this procedure, the carrier concentration is the only free parameter, whilst the electron mobility is determined from this concentration through an empirical model.^[29] As a result, the carrier concentration of the silicon at room temperature is estimated as $2.0 \times 10^{17} \text{ cm}^{-3}$, yielding a plasma frequency of 7.87 THz, whilst the electron mobility is estimated as $603 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, and hence the collision frequency equals 1.78 THz. The DC resistivity estimated from the mobility and the concentration equals 0.05 Ω cm, within the range specified by the manufacturer. From these quantities, the complex permittivity at 1.0 THz is determined to be $-3.14-26.45j$. Hence, the SPP propagation length on the silicon-air interface is 1.27 mm, and the decay lengths normal to the interface are 246 μm into the air and 9.24 μm into the silicon substrate.

The coaxial microcavities are fabricated by using conventional micro-fabrication techniques involving photolithography and deep reactive ion etching (DRIE). The fabricated structures are then examined using a Philips XL-30 scanning electron microscope (SEM) to determine the depth and smoothness of the vertical walls obtained from the DRIE process. **Figure 2** shows the SEM micrographs of the coaxial microcavities imaged with a 45° tilt. The SEM micrographs confirm that smooth and straight sidewalls are obtained. One of the wafers is cleaved and loaded orthogonally into the SEM to image the cross-sectional depth of the etched wafers (not shown here), which confirms the data obtained from a profilometric analysis. The total area for each sample is $10 \times 10 \text{ mm}^2$, containing about 2500 identical cavities. To provide a reference in the measurements, the surface of a bare silicon wafer is coated with a 200 nm thick gold layer to create a terahertz mirror.

The measurement is carried out in reflection-mode terahertz time-domain spectroscopy (THz-TDS) with a pair of emitting/detecting photoconductive antennas (PCAs) on LT-GaAs substrates. The generated linearly polarized terahertz beam propagates through a beam splitter and is then focused by an elliptical mirror to a focal beam diameter of 8 mm onto the sample at normal incidence. The beam reflected from the sample is collected by the same mirror back to the beam splitter that partially reflects the energy into the PCA detector. The entire terahertz path is purged with nitrogen to avoid water vapor absorption lines. The total scanning duration is 68 ps and is digitized with a step size of 0.06 ps. For comparison, electromagnetic simulations are performed with a commercial full-wave frequency-domain solver, CST Microwave Studio, based on the finite-element method. In the simulated models, the dielectric properties of the silicon are described via the Drude model with the plasma frequency and scattering rate estimated earlier. Unit-cell boundary conditions are employed for the transverse boundaries to replicate an infinite planar array. A Floquet port excites the array with a normally incident plane wave and also collects the scattered field. The simulation using ANSYS HFSS with a similar setting produces comparable results.

3. Design, Fabrication, and Experiments

The normalized reflection magnitude profiles for the 50 μm depth cavity arrays are shown in **Figure 3**. The measurement and simulation results are in general agreement with slight discrepancies due to fabrication tolerances, system noise, and sample alignment. No transmission through the wafer is detectable within the system bandwidth and dynamic range for all of the samples. The normalized reflection spectrum for the bare silicon illustrates the transition from metal to dielectric, corresponding to the zero permittivity crossing at 1.46 THz. The high reflection towards low frequencies demonstrates the metallic characteristic of the material. The response of all the microcavity samples follows the silicon reflection baseline. For all the cavities with a 50 μm depth, the resonance is evident around 1.0-1.2 THz, where

the arrays dissipate a relatively large amount of incident energy. As the wall separation in the cavities becomes narrower, the resonance frequency undergoes redshift, and the resonance strength becomes weaker. Supplementary simulation reveals that the lattice constant has a negligible effect on the resonance characteristics, except for the resonance strength that decreases with a larger separation. Observed in the simulation results in **Figure 3(b)**, the lattice mode is manifested as a small dip at 1.5 THz, which corresponds to the lattice constant of one wavelength. It is noteworthy that no resonance is observed in simulations of PEC cavities with the same dimensions.

The simulated field distribution on resonance is given in **Figure 4** for a specific sample. The electric field pattern in **Figure 4(a)** resembles the TE_{11} (dipolar) mode of a coaxial waveguide. The fundamental TEM mode, which is radially symmetric, cannot develop inside the cavity because of the plane wave excitation. Associated with this TE_{11} mode is the strong field enhancement close to the cavity walls at the aperture, owing to the plasmonic effect and the singularity at the corners, as shown in **Figures 4(b) and 5**. **Figure 5** further shows that a narrower wall separation leads to stronger field strength in the middle of the aperture. This developed hyperbolic field variation for the component perpendicular to the aperture walls is similar to the behavior of gap-plasmon modes exclusively found in plasmonic waveguides.^[31] In these coaxial cavity the aperture with a subwavelength gap size is excited by a nearly constant field to couple to only the even or symmetric mode.^[32] On closer inspection, the absolute electric field strength in **Figure 4(b)** reveals a pair of a node at the bottom and an anti-node at the top of the 50 μm depth cavity. This node pair indicates the fundamental Fabry-Pérot resonance.

Further insight into the response of these cavities can be gained from the dispersion behavior of plasmonic coaxial waveguides. The nature of the SPP mode in the waveguides is close to that of the planar heterostructure comprising conductor/dielectric/conductor, where the SPP

modes on the two interfaces interact and give rise to the coupled mode.^[33] The approximated dispersion model can be expressed in the transcendental form:^[34]

$$\tanh\left(\sqrt{\beta^2 + \left(\nu/r_{\text{avg}}\right)^2 - k_0^2 \varepsilon_d} \frac{g}{2}\right) = -\frac{\varepsilon_d \sqrt{\beta^2 + \left(\nu/r_{\text{avg}}\right)^2 - k_0^2 \varepsilon_d}}{\varepsilon_c \sqrt{\beta^2 + \left(\nu/r_{\text{avg}}\right)^2 - k_0^2 \varepsilon_c}} \quad (1)$$

where β and k_0 denote the SPP and free-space wavenumbers, respectively; ε_c and ε_d for the relative permittivities of the doped silicon and free space, respectively; ν for the angular momentum, equal to 1 for the TE₁₁ mode; $r_{\text{avg}} = (r_o + r_i)/2$; and $g = r_o - r_i$. **Equation 1** yields the dispersion diagram for the designed cavities, given in **Figure 6** in comparison to the dispersion relation for the PEC waveguide operating in the same mode. It is obvious that the PEC waveguide sustains fast waves, with the exact cutoff frequency at about 0.73 THz. On the other hand, the same mode in the plasmonic coaxial waveguides evolves from fast waves at low frequencies to slow waves at high frequencies. In case of the lossless plasmonic waveguides in **Figure 6(a)**, the propagation constant goes towards infinity, as the frequency approaches the surface plasmon frequency at 1.30 THz where $\varepsilon_c = -1$.^[33] Contrarily, the propagation constant for the lossy waveguides becomes finite at the surface plasmon frequency, as illustrated in **Figure 6(b)**. As the wall separation is reduced, the propagation constant for the plasmonic waveguide increases, equivalent to an increase in the effective refractive index. Correspondingly, the cutoff frequency for the plasmonic waveguide becomes dependent on the wall separation.

The resonance condition has to account for the cavity length, propagation constant, and the phase changes upon reflection at both ends of the cavity. The behavior of the cavity open end is close to that of a perfect magnetic conductor (PMC) that induces a zero phase change to the reflected wave. The bottom of the cavity, made of the doped silicon, imposes about -0.9π reflection phase shift, estimated from a plane-wave simulation. The slight deviation from the PEC reflection phase of $-\pi$ is due to significant field penetration into the material. From this

observation, the condition for the resonance can be approximately given as $\beta_N = (2N + m)\pi/2d$, where β_N is the SPP wavenumber inside the cavity for the N^{th} -order resonance ($N = 0, 1, 2, \dots$), d is the cavity depth, and $m = \{0.9, 1.0\}$ for the plasmonic and PEC boundaries, respectively. From **Figure 6(b)**, the fundamental resonance $N = 0$ of the 50 μm depth PEC cavity operating in the TE_{11} mode would take place at 1.67 THz. For the plasmonic cavities L40, L30, and L20 sustaining the same mode, the resonance frequencies are estimated to be around 1.34, 1.28, and 1.18 THz, compared with the simulation and experimental results of 1.20, 1.13, and 1.04 THz, respectively. A slight difference between the analytical and experimental results is likely from a small phase change introduced by the fringing field above the cavity opening. As the physical length of the cavity becomes shorter, in the case of a lossless plasmonic material the resonance is expected to converge to the surface plasmon frequency where the wavenumber is infinite.^[33] However, this convergence is not observed in the lossy material, since the wavenumber becomes finite. **Figure 7** shows the response of the 25 μm depth cavities, S20, S30, and S40 in the TE_{11} mode. In these cases the reflection dip can be observed around 1.5 THz. From the resonance condition and the dispersion relation, these cavities are expected to exhibit the fundamental resonance at the wavenumber of $\beta_0 = 5.7 \times 10^4 \text{ m}^{-1}$, or equivalently around 2.5 THz. However, as discussed earlier, beyond 1.46 THz this doped silicon becomes a lossy dielectric, and hence, no longer supports the SPP mode. Therefore, the structure cannot sustain the expected resonance at 2.5 THz, and the observed dip in the reflection magnitude is in fact the resonance shoulder. On the other hand, as the physical length of the cavity becomes longer, the fundamental resonance experiences redshift, and at the same time higher-order cavity modes $N > 0$ can be supported. The simulation shows that a first-order mode ($N = 1$) can be observed when the cavity depth is at least 140 μm . For the structure with the same radii as L30 and the depth of 140 μm , the fundamental and first-order

resonances take place at 0.72 THz and 1.35 THz, respectively. Despite that, the fabrication to achieve this level of cavity depth becomes difficult with present capabilities.

The resonance mechanism involving the plasmonic effect can be summarized as follows. A freely propagating wave is incident on the sample and then diffracted by the apertures of the subwavelength cavities. The diffraction allows phase matching and excitation of the bound mode along the cavity walls that support cylindrical SPPs. The characteristics of cylindrical SPPs depend on the mode field distribution, which in this case corresponds to a coaxial TE_{11} mode, as well as the cavity geometry that influences the mode coupling between the two sidewalls. The resonance originates from the standing wave of the guided SPP inside the cavity upon reflection at its open and blind ends. In effect, the resonant cavity appears slightly longer because of the plasmonic confinement along the walls, the fringing fields at the aperture, and the field penetration at the bottom of the cavity. The observable shift in the resonance despite a fixed cavity depth is due to a reduced wall separation that results in a change in the SPP wavenumber. Resonance damping is caused by resistive loss in the silicon. Owing to the non-radiative mode in the cavity, the resonance frequency is virtually independent of the lattice constant.

4. Conclusions

This article has presented the concept and realization of plasmonic coaxial micro-cavities operating at terahertz frequencies. It is experimentally demonstrated that the microcavities built into a doped silicon layer can support LSPRs through the interplay between the cylindrical SPPs and the cavity modes. The interaction between the bound electromagnetic waves and surface electron oscillations effectively leads to the subwavelength dimensions of the cavity. Since the resonance originates inside the cavity, the inter-cell coupling is minimal. The dispersion model for plasmonic coaxial waveguides can be used to quantitatively explain

the influence of the geometry on the resonance behavior. The strong field enhancement in this sub-wavelength plasmonic element promises applications in sensing and imaging,^[36] whilst the large energy dissipation by the electron oscillation on resonance suggests the implementation of perfect terahertz absorbers.^[37] The field confinement inside the cavity is suitable for terahertz integrated optics. Additionally, it is possible to realize negative-index materials from this terahertz coaxial microcavity array.^[38] A higher quality factor for the resonance can be obtained by replacing silicon with InSb.

5. Experimental

The silicon wafers were patterned using photolithography. Before spin coating the wafers with photoresist, the silicon wafers were thoroughly cleaned with acetone and isopropyl alcohol (IPA) followed by a dehydration bake. The cleaned wafer was then spin-coated with positive type photoresist (AZ 5214, MicroChemicals) and soft-baked on a hotplate. Using contact lithography both the mask and the wafer were brought in contact and exposed with UV light. The exposed wafer was developed using developer (AZ400K) to create openings for the etching. After developing the photoresist, the wafers were hard-baked to ensure the photoresist mask layer can withstand the selected etching process.

To obtain the coaxial microcavities, the photolithographically patterned silicon wafers were etched using the Bosch DRIE process. Bosch DRIE is a physical etching process to anisotropically etch silicon wafers and it can produce high aspect ratio vertical side walls. In the Bosch process, a mixture of SF₆ and C₄F₈ gas were alternatively permitted into the high vacuum chamber and ionized by applying RF power. The ionized SF₆ etchant gas was accelerated towards the silicon wafer to create deep trenches in silicon wafer, C₄F₈ gas mixture passivates the side walls to obtain smooth high aspect ratio vertical trenches. The etch rate of the silicon wafer using the Bosch DRIE process was approximately 2.5 $\mu\text{m min}^{-1}$ and

the samples were timed to etch 25 μm and 50 μm deep trenches into the silicon wafer. This was confirmed using a profilometer (Ambios XP-2). On completion of etching, the photoresist was stripped off using acetone and the samples were cleaned using oxygen plasma to remove any polymer residue on the wafers.

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Figure 1. Diagram for a unit cell of the coaxial microcavity in a silicon substrate. (a) Top view, and (b) side view. The square unit cell size a is fixed at $200\ \mu\text{m}$, and the silicon substrate thickness t equals $270\ \mu\text{m}$. The inner radius (r_i), outer radius (r_o), and cavity depth (d) are given in Table 1. The diagram is not to scale.

Figure 2. Scanning electron micrographs of the fabricated microcavities. Partial view of the cavity arrays (a) L40, (b) L30, and (c) L20. (d) Magnified view of a single cavity L20. The micrographs are taken at the tilt angle of 45° . All of the samples shown here have a depth of $50\ \mu\text{m}$.

Figure 3. Normalized reflection spectra for microcavity arrays with $50\ \mu\text{m}$ cavity depth. (a) Experimental and (b) simulation results. The reflection spectrum for the bare silicon is included for comparison. All the spectra are normalized to the reference taken from the gold-coated silicon wafer. The shaded area in (a) marks the limit of the system dynamic range.^[30]

Figure 4. Numerically resolved field distribution in the coaxial microcavity. (a) Instantaneous electric field pattern over the opening of the coaxial cavity, and (b) absolute electric field strength across the center of the cavity. The cavity dimensions are according to sample L40. The field plots are on resonance at 1.2 THz. The maximum field strength is set to 0 dB.

Figure 5. Numerically resolved absolute electric field strength for the 50 μm cavities. The frequency for each field profile is at the resonance of the structure, *i.e.* 1.04, 1.13, and 1.20 THz for samples L20, L30, and L40, respectively. The field profile is located across the opening of the cavity along the incident polarization. The field strength is the component normal to the cavity walls, and is normalized to that on the bare silicon surface.

Figure 6. Dispersion diagram for the TE_{11} mode in the PEC and plasmonic coaxial waveguides. (a) Lossless plasmonic cavities, *i.e.* $\Im(\epsilon_c) = 0$, and (b) lossy plasmonic cavities. The dispersion for the PEC waveguide depends only on the average radius,^[35] set to 65 μm as that for all the plasmonic cavity designs. The dispersion for the plasmonic waveguides is determined from Equation 1. The vertical lines indicate the wavenumber β_0 where the fundamental resonance is expected for cavities with a depth of 50 μm .

Figure 7. Normalized reflection spectra of microcavity arrays with 25 μm cavity depth. (a) Experimental and (b) simulation results. The reflection spectrum for the bare silicon is included for comparison. All the spectra are normalized to the reference taken from the gold-coated silicon wafer. The shaded area in (a) marks the limit of the system dynamic range.

Table 1. Dimensions of the fabricated coaxial cavities. All the units are in micrometers. The average radius r_{avg} equals 65 μm for all of the samples.

Sample	Cavity depth d	Wall gap $g = r_o - r_i$	Outer radius r_o	Inner radius r_i
L40	50	40	85	45
L30	50	30	80	50
L20	50	20	75	55
S40	25	40	85	45
S30	25	30	80	50
S20	25	20	75	55

The table of contents entry should be fifty to sixty words long, written in the present tense, and refer to the chosen figure.

Table of Contents Entry

Coaxial microcavities etched into the surface of a doped silicon substrate are shown to support localized surface plasmon resonances (LSPRs) at terahertz frequencies. This study is a promising pathway for development of terahertz devices with applications in the areas of photonic integrated circuits, molecular sensing, and subwavelength imaging.

Keywords

plasmonics, localized surface plasmon resonance, terahertz, metamaterial, coaxial cavity

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Title

Terahertz Localized Surface Plasmon Resonances in Coaxial Microcavities

ToC Figure