Research article

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Fourier-plane investigation of plasmonic bound states in the continuum and molecular emission coupling

<https://doi.org/10.1515/nanoph-2020-0343>

Received June 22, 2020; accepted August 17, 2020; published online September 2, 2020

Abstract: Bound states in the continuum (BICs) or trapped modes can provide an important new avenue for strong light confinement via destructive interference. Dielectric photonic structures have been extensively studied for optical BICs. However, BICs in plasmonic nanostructures have not been explored much yet. Herein, we present a thorough experimental study of plasmonic BICs via Fourier-plane spectroscopy and imaging. Optical mode dispersion in a metal grating covered by a dielectric layer is directly measured in an angle-resolved white light reflection spectrum. Two dielectric layer thicknesses are considered. Both plasmonic and photonic modes are supported in the visible range using a thicker dielectric film; hence, either hybrid or purely plasmonic BICs can be formed. With a thinner dielectric layer, only plasmonic modes are strongly excited and purely plasmonic BICs appear. Our measurements exhibit all features expected for BICs, including a substantial increase in the Q factor. We also demonstrate that the BIC position can be switched from one optical mode branch to the other by tuning a metal grating parameter. Moreover, by mixing luminescent dyes in a dielectric layer, light emission coupling into BICs is investigated. We find that the photoluminescence peak disappears at the BIC condition, which is attributed to the trapping of molecular emission at plasmonic BICs. Therefore, both white light reflection and dye emission

measurements in the Fourier plane clearly indicate the formation of trapped modes in plasmonic nanostructures. Our observation implies that plasmonic BICs can enable a highly effective light trapping device despite the simple structure of the device geometry. Plasmonic supercavity design based on the BIC concept may provide many interesting future opportunities for nanolasers, optical sensing, and nonlinear enhancement.

Keywords: bound states in the continuum; Fourier-plane spectroscopy and imaging; molecular emission; plasmonic nanostructures.

1 Introduction

Optical resonators are fundamental to many optical devices and elements [1, 2]. In particular, strong light confinement in optical resonators is of great importance to the enhancement of various light-matter interactions. High- Q resonances in most optical resonators are based on the constructive interference of confined light. However, bound states in the continuum (BICs) present a novel approach for optical resonator design. BICs or trapped modes are eigenmodes embedded in a radiative continuum [3, 4]. Despite the existence of radiation leakage channels, they can have infinite lifetimes. Studies on BICs initially started from quantum mechanics with pure theoretical curiosities [5]; however, photonic structures, such as dielectric photonic crystals, have been extensively studied for optical BICs recently [6–14]. Both symmetry-protected and nonsymmetry-protected BICs can be formed. Symmetry-protected BICs are often found in a system with a reflection or rotational symmetry. On the other hand, nonsymmetry-protected BICs can be formed by the destructive interference of interacting optical resonances. These accidental BICs provide a new avenue for strong light confinement in a simpler device geometry via destructive interference. They are found to provide new means for high- Q cavity design, which is important for

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lasers, nonlinear optical enhancement, optical sensing, as well as for the enhancement of general light-matter interactions.

Plasmonic structures have unique merits for nanoscale light confinement and manipulation [15, 16]. Furthermore, the strong near-field and large local density of optical states (LDOSs) in plasmonic nanostructures enable the efficient coupling of light emission from nearby emitters [17–20]. Metal absorption in plasmonic structures may pose some limitations to light confinement and manipulation; however, most plasmonic nanostructures still suffer from significant radiative losses. Therefore, there have been continuous interests on the suppression of radiative damping in plasmonic structures in various contexts [21–23]. For example, we notice that high Q factors in plasmonic nanostructures were achieved via surface lattice resonances [24–30]. More recently, the strong interaction between plasmonic and photonic modes have been theoretically investigated to form a hybrid plasmonic BIC in metallic nanostructures [31, 32]. In the strong coupling regime, such a hybrid BIC can appear near the anti-crossing point via destructive interference, following the Friedrich-Wintgen condition [33]. It is noteworthy that the suppression of radiative damping in the strong coupling condition is also known as resonance trapping in nuclear physics [34, 35] and external coupling in coupled microcavities [36, 37]. However, BICs in plasmonic nanostructures have not been extensively investigated in experiments [38].

Here, we present a thorough experimental study of plasmonic BICs via Fourier-plane spectroscopy and imaging [39–43]. We consider a metal grating covered by a polymer layer and investigate the mode dispersion by directly measuring the spectrum and image in the Fourier plane. By placing a pinhole in the Fourier plane, we selectively measure the reflection spectrum at a specific angle (i.e., with a specific wavevector component). By moving the pinhole and combining the measured reflection spectra, we obtain a reflection map that shows optical mode dispersion clearly. In this study, we consider two thicknesses for the dielectric film. The sample with a thicker dielectric film ($t \sim 500$ nm) supports both plasmonic and photonic modes in the visible range. Either hybrid BICs (originating from the interaction between plasmonic and photonic modes) or purely plasmonic BICs can be formed in this sample. However, in a thinner film ($t \sim 100$ nm), only plasmonic modes are strongly excited in the considered wavelength region. In this case, purely plasmonic BICs can appear from the interaction between two plasmonic modes. We investigate and carefully characterize hybrid plasmonic and purely plasmonic BICs in

our samples, and our measurements clearly show all features expected for BICs, including a substantial increase in the Q factor. Additionally, we experimentally demonstrate that the BIC position can be flipped from one optical mode branch to the other by tuning a metal grating parameter.

Moreover, by mixing luminescent dyes in a polymer matrix, we study light emission coupling in metal nanostructures too. Photoluminescence (PL) measurements in the Fourier plane clearly indicate the optical mode coupling of dye emission. Especially, we observe the trapping of molecular emission at the BIC condition in both the Fourier-plane PL spectrum and image. The eigenmode at the BIC can be excited by emitters; however, we find that dye emission is trapped and the PL peak disappears at the BIC condition. Therefore, both reflection and PL measurements in the Fourier plane clearly indicate the formation of trapped modes in plasmonic nanostructures. This implies that a plasmonic structure with BICs might be a highly effective light trapping structure despite the structural simplicity. Our experimental observations suggest that plasmonic BICs may be promising for various nanocavity designs as they provide sharper optical resonances, as well as strong near-fields and large LDOSs. Optical devices based on plasmonic BICs may further improve plasmonic nanolasers [44, 45], nonlinear optical enhancement [46, 47], and optical sensing [48].

2 General theory for BICs from coupled resonances

In this section, we describe the general theory of BICs that originate from two interacting resonances via a radiative continuum. When two optical resonances are coupled to the same radiation channel, the optical interaction between two resonances is described by the following non-Hermitian Hamiltonian, neglecting nonradiative damping terms [3, 4]:

$$\begin{aligned}\hat{H} &= \begin{pmatrix} E_1 & \kappa \\ \kappa & E_2 \end{pmatrix} - i \begin{pmatrix} \gamma_1 & e^{i\psi} \sqrt{\gamma_1 \gamma_2} \\ e^{i\psi} \sqrt{\gamma_1 \gamma_2} & \gamma_2 \end{pmatrix} \\ &= \begin{pmatrix} E_1 - i\gamma_1 & \kappa - ie^{i\psi} \sqrt{\gamma_1 \gamma_2} \\ \kappa - ie^{i\psi} \sqrt{\gamma_1 \gamma_2} & E_2 - i\gamma_2 \end{pmatrix} \quad (1)\end{aligned}$$

Here, E_1 and E_2 are the resonance energy levels of two optical resonances, γ_1 and γ_2 are the corresponding radiative damping rates, and κ is the internal coupling strength. $\sqrt{\gamma_1 \gamma_2}$ is the radiative coupling term, while ψ is the phase difference between two modes.

The eigenvalue of the corresponding Hamiltonian is determined by the following secular equation:

$$|\hat{H} - \tilde{E}\hat{I}| = \begin{vmatrix} E_1 - i\gamma_1 - \tilde{E} & \kappa - ie^{i\psi}\sqrt{\gamma_1\gamma_2} \\ \kappa - ie^{i\psi}\sqrt{\gamma_1\gamma_2} & E_2 - i\gamma_2 - \tilde{E} \end{vmatrix} = 0 \quad (2)$$

Then, we obtain the following complex eigen energy $\tilde{E}$:

$$\tilde{E} = \frac{E_1 + E_2}{2} - i\frac{\gamma_1 + \gamma_2}{2} \pm \sqrt{\left(\frac{E_1 - E_2}{2} - i\frac{\gamma_1 - \gamma_2}{2}\right)^2 + (\kappa - ie^{i\psi}\sqrt{\gamma_1\gamma_2})^2} \quad (3)$$

Eq. (3) describes the general solution for energy levels and radiative damping rates resulting from two coupled optical resonances in an *open* photonic system.

Now, if we apply the Friedrich-Wintgen condition [33]

$$(E_1 - E_2) \cdot p \sqrt{\gamma_1\gamma_2} = \kappa(\gamma_1 - \gamma_2), \quad (4)$$

the term inside the square root in Eq. (3) can be simplified further. Here, p is the parity (+1 or -1) of the phase difference between two modes (i.e., in-phase or out-of-phase), which corresponds to $\psi = \pi l$ where l is an integer number.

$$\begin{aligned} & \left(\frac{E_1 - E_2}{2} - i\frac{\gamma_1 - \gamma_2}{2}\right)^2 + (\kappa - ip\sqrt{\gamma_1\gamma_2})^2 \\ &= \left(\frac{\kappa}{p\sqrt{\gamma_1\gamma_2}} - i\right)^2 \cdot \left(\frac{\gamma_1 + \gamma_2}{2}\right)^2 \end{aligned} \quad (5)$$

Therefore, we have the following two complex energy levels:

$$\tilde{E}_A = \frac{E_1 + E_2}{2} + p\frac{\kappa}{\sqrt{\gamma_1\gamma_2}} \cdot \frac{\gamma_1 + \gamma_2}{2} - i(\gamma_1 + \gamma_2) \quad (6)$$

$$\tilde{E}_B = \frac{E_1 + E_2}{2} - p\frac{\kappa}{\sqrt{\gamma_1\gamma_2}} \cdot \frac{\gamma_1 + \gamma_2}{2} \quad (7)$$

We see that the one eigenmode $\tilde{E}_B$ becomes purely real (i.e., radiationless, $\gamma_B = 0$), corresponding to a BIC. The other eigenmode $\tilde{E}_A$ becomes more radiative, of which the radiative loss is being the sum of two original modes, $\gamma_A = \gamma_1 + \gamma_2$. This can be also mathematically understood from the fact that the trace of a square matrix equals the sum of its eigenvalues. Generally, this can result in super-radiant and sub-radiant eigenmodes in coupled optical resonances. We notice that Eq. (4) is, in fact, the mathematical condition in that one of two eigenmodes has a purely real eigenvalue. In realistic plasmonic structures, the total damping rate is still limited by absorption losses – i.e., E_1 and E_2 in Eqs. (6) and (7) are not purely real but have nonzero imaginary parts owing to the absorption losses. However, the total Q factor can be still enhanced significantly by suppressing radiative damping. When two optical modes interact via radiative coupling, one of two

coupled modes can become radiationless by destructive interference, called a Friedrich-Wintgen BIC, although it is an eigenmode overlapped with the radiative continuum. Therefore, BICs can provide another route to create efficient light trapping structures via destructive interference. From Eqs. (6) and (7), we also find that, depending on the sign of parity p or coupling strength κ , the BIC can be located either in the low- or high-energy branch.

3 Experimental results and discussion

Figure 1A shows a schematic for our sample, where a metal grating is covered by a dielectric film. Figure 1B shows the procedure for the device fabrication. An optically thick silver film was sputtered on Si(111) at a high temperature (350 °C). Through atomic force microscopy (AFM), the surface roughness of the sputtered silver film was measured to be approximately 2.18 nm. Subsequently, silver nanostripes (thickness: $h \sim 30$ nm) were fabricated on top of the film by electron beam lithography and silver evaporation. Figure 1C, D shows the height profile obtained from AFM (grating period: $P = 250$ nm) and Scanning Electron Microscope (SEM) images ($P = 250, 550$ nm) of the fabricated silver grating. They confirmed the high quality of the fabricated metal gratings. Finally, a Poly(methyl methacrylate) (PMMA) layer was spin-coated on top of the silver grating. Two thicknesses for the dielectric film were considered in this study. The sample with a thicker dielectric film ($t \sim 500$ nm) supports both plasmonic and photonics modes. Figure 1E shows the simulated field profiles for plasmonic and photonics modes; the plasmonic mode in the metal grating is more confined near the metal surface, whereas the photonic mode is dispersed throughout the entire dielectric slab. However, in a thinner film ($t \sim 100$ nm), only plasmonic modes are strongly excited in the considered wavelength region.

We investigated the optical mode dispersion in our plasmonic nanostructures via Fourier-plane spectroscopy and imaging. Figure 2A shows the white light reflection setup used for our Fourier-plane measurements. Light reflected from the sample surface at a specific angle (or with a specific wavevector k) is focused in the back focal plane. Therefore, the back focal plane in the microscope objective was imaged instead of the sample surface to conduct Fourier-plane measurements. After passing the lens L_1 , the reflected beam with a specific k component is collimated and then refocused in the Fourier plane (FP in Figure 2A) by another lens L_2 . Subsequently, by placing a pinhole in the

Figure 1: (A) Schematic for the sample, where a metal grating is covered by a dielectric (PMMA) film. Two thicknesses for the dielectric film are considered ($t \sim 500$ and 100 nm). (B) Fabrication procedure for silver gratings. (C) Height profile obtained from atomic force microscopy (AFM) (grating period: $P = 250$ nm). (D) SEM images of the fabricated silver grating ($P = 250, 550$ nm). (C) and (D) confirm the high quality of the fabricated metal gratings. (E) Simulated field profiles for plasmonic and photonic modes.

Fourier plane, we selectively measured the angle-resolved reflection spectrum. The inset in Figure 2A shows a schematic of the Fourier plane. The white light reflection appears as a circle in the Fourier plane (or normalized k_x - k_y plane), the radius of which corresponds to the numerical aperture (NA) of the microscope objective: $NA = \sin \theta_m$, where θ_m is the maximum collection angle of the microscope objective. By moving the pinhole and collecting the measured reflection spectra, we obtained the reflection map as a function of wavevector and wavelength. The measured raw spectrum was normalized to the background reflection spectrum from an unpatterned silver film to correct the natural intensity variation across the Fourier plane. Furthermore, we measured the PL spectrum of dye molecules in the same Fourier setup by using a diode pump laser instead of white light illumination. Additionally, Fourier plane images were obtained in the same setup. In this case, the pinhole was removed and incident light was focused on the monochromator slit entrance. By fully opening the slit and setting the monochromator grating to the zero position, the Fourier plane was directly imaged on a two-dimensional charge-coupled device camera. The

optical mode dispersion appears as a circular arc at a specific wavelength, of which the radius is the effective mode index n_{eff} . The momentum conservation requires

$$\left| \vec{k}_{\text{in-plane}} - m\vec{G}_x \right| = \left| \vec{k}_{\text{mode}} \right| = n_{eff}k_0 \quad (8)$$

where $\vec{k}_{\text{in-plane}} = k_x\hat{x} + k_y\hat{y}$ is the in-plane wavevector of incident light, $\vec{G}_x = 2\pi/P\hat{x}$ is the reciprocal lattice wavevector of the one-dimensional grating (m is an integer number), $\vec{k}_{\text{mode}}$ is the wavevector of an optical mode (either the plasmonic or photonic mode), and $k_0 = 2\pi/\lambda_0$ is the free-space wavenumber. Then, it follows that

$$\left(\frac{k_x}{k_0} - m \frac{G_x}{k_0} \right)^2 + \left(\frac{k_y}{k_0} \right)^2 = n_{eff}^2 \quad (9)$$

This means that an optical mode appears as a circular arc with a radius of n_{eff} in the Fourier plane, and the center of the circle is shifted by grating momentum $m\frac{G_x}{k_0}$. This Fourier-plane spectroscopy and imaging enables the investigation of the optical mode dispersion and angular response without having to rotate a sample for each angle. This Fourier-plane setup is particularly suitable for optical

Figure 2: (A) Fourier-plane measurement setup. By placing a pinhole in the Fourier plane, the reflection spectrum at a specific angle can be measured. By moving the pinhole and combining the measured reflection spectra, a reflection map is obtained. (B) White light reflection spectrum map (k_x/k_0 vs λ) for the plasmonic-photonic hybrid structure ($t \sim 500$ nm) with different silver grating periods ($P = 350, 400$, and 450 nm). Optical modes appear as a clear dip in the reflection spectrum. (C) Reflection map for $k_y = 0$ and $k_y \neq 0$.

measurements over a small area of a nanopatterned sample, where the size of the nanopattern can be limited to the micrometer scale.

Figure 2B shows the transverse magnetic (TM)-polarized white light reflection spectrum (k_x/k_0 vs λ) for the plasmonic-photonic hybrid structure ($t \sim 500$ nm) with different silver grating periods ($P = 350, 400$, and

450 nm). We varied the pinhole position across the k_x axis while maintaining $k_y = 0$ (see the inset in Figure 2A). The dispersion relation in our sample is clearly seen as a dip in the reflection spectrum. Each dispersion line originates from either a plasmonic or photonic mode. We also notice that the overall spectrum moves upward to the longer wavelength as the period increases (i.e., the grating

momentum decreases). Additionally, we performed measurements for nonzero k_y . As shown in Figure 2C, when k_y is varied slightly, the entire spectrum shifts in accordance with Eq. (9). The same behavior can be also observed in the

Fourier plane image (two-dimensional image in the k_x - k_y plane), as will be shown later.

Our measurements clearly show all features expected for both symmetry-protected and nonsymmetry-protected

Figure 3: Nonsymmetry-protected (blue circle) and symmetry-protected (orange circle) bound states in the continuum (BICs) for $P = 400$ nm and $t \sim 500$ nm.

(A) and (B) are experimentally measured and simulated reflection maps. (C) and (D) shows the reflection spectrum and the extracted total Q factors at different incident angles for nonsymmetry-protected and symmetry-protected BICs, respectively. Near the BIC position, the Q factor increases rapidly because of the suppressed radiative loss. The yellow shaded areas in (C) and (D) mean the BIC region, corresponding to the blue and orange circles in (A), respectively.

BICs. Figure 3 shows the TM-polarized white light reflection spectrum for the grating period of $P = 400$ nm. The reflection spectra obtained from experiment (Figure 3A) and simulation (Figure 3B) match well overall. TM-polarized light excites both plasmonic and photonic modes. Therefore, either hybrid plasmonic BICs between the plasmonic and photonic modes or purely plasmonic BICs can be formed in this sample. Meanwhile, transverse electric (TE)-polarized light excites only photonic modes and thus plasmonic BICs do not appear.

The two circles in Figure 3A represent the experimentally measured nonsymmetry-protected and symmetry-protected BICs, respectively. The blue circle at an off- Γ position ($k_x/k_0 = 0.19$, corresponding to the incident angle of 11°) indicates a nonsymmetry-protected BIC formed by the strong coupling between plasmonic and photonic modes. This hybrid BIC is formed near the anticrossing point and is located in the lower energy (longer wavelength) branch. In the white light reflection spectrum, incident light does not couple to this point although there exists an eigenmode; this trapped mode is an energy eigenmode but decoupled from the outside. Incident light reflects strongly at this point (i.e., it appears red at this point in the color map), and the dispersion line is interrupted (or vanished). Note that this is fundamentally different from the photonic bandgap, where optical eigenmodes do not exist. The low energy branch contains a BIC owing to the destructive interference between plasmonic and photonic modes, whereas the high energy branch becomes more radiative, as shown by the blue color in the high energy branch near the anticrossing point. This agrees with the general theory presented in Section 2.

The orange circle at the Γ position shows a symmetry-protected BIC formed by the interaction between surface plasmon modes. Two counter-propagating plasmonic modes form a standing wave at this point, resulting in symmetric and antisymmetric modes at different energy levels (i.e., forming a plasmonic bandgap). Figure 3C, D show the reflection spectrum for different incident angles and the extracted Q factors for the corresponding nonsymmetry-protected (hybrid) and symmetry-protected (purely plasmonic) BICs, respectively. The yellow shaded areas in Figure 3C, D mean the BIC region, corresponding to the blue and orange circles in Figure 3A, respectively. The Q factors were determined from experimental data by curve fitting. Near the BIC position, the Q factor increases rapidly because of the suppressed radiative loss, whereas the reflection dip weakens for both cases, as expected for BICs. The experimentally measured Q factors near plasmonic BICs are significantly higher than those of original, uncoupled plasmonic modes. However, they are still

limited to around 200 in our measurements. This experimental limitation can be caused by the silver surface roughness root mean square (RMS ~ 2.18 nm) and finite grating size ($20 \times 20 \mu\text{m}$) in our sample, as well as the nonradiative losses caused by metal absorption. We also notice the size difference in the plasmonic bandgap between experiment and simulation. In experiment (Figure 3A), the plasmonic bandgap covers the wavelength region of 620–670 nm, while it covers 620–710 nm in simulation (Figure 3B). The smaller bandgap in experiment results from the reduced strength in surface plasmon excitation and the reduced interaction between plasmon modes, which can be attributed to nonidealities in our sample again. We think it is possible to further increase the Q factor around plasmonic BICs by improving sample quality and design. More discussions on experimental spectra are given in the Supplementary information.

We prepared another metal grating sample with a thinner PMMA layer ($t \sim 100$ nm). This sample excites only plasmonic modes strongly in the considered visible range. Figure 4 shows TM-polarized white light reflection maps for the grating periods of $P = 300$ and 480 nm. A strong dip in the reflection map shows plasmonic mode dispersions. In this case, purely plasmonic BICs are formed. Two counter-propagating surface plasmon modes form a standing wave at the Γ position ($k_x = 0$) or at the Brillouin zone boundary ($k_x = G_x/2 = \pi/P$), resulting in symmetric and antisymmetric modes at different energy levels. The upper panel ($P = 300$ nm) in Figure 4 shows the plasmonic BIC at the Γ position, formed by two plasmonic modes $\text{SP}[\pm 1]$, where the integer in the bracket is the grating momentum index m . The lower panel ($P = 480$ nm) in Figure 4 shows the plasmonic BIC at the Brillouin zone boundary, formed by two plasmonic modes ($\text{SP}[+2]$ and $\text{SP}[-1]$, or $\text{SP}[-2]$ and $\text{SP}[+1]$). Figure 4C, D show the reflection spectrum and the extracted Q factor for both cases. Similarly, the Q factor increases substantially around the BIC position. This type of plasmonic BICs occurs only at special points (the Γ position and Brillouin zone boundary), while the hybrid BIC in Figure 3 occurs inside the Brillouin zone. In either case, two optical modes reside in the same region, and the coupling strength between two modes is important for the formation of BICs.

Furthermore, we observed that, by varying the width (w) of the metal stripe, the BIC position in both cases can be switched from one plasmonic mode branch to the other. This is clearly shown in Figure 4 for both $P = 300$ nm (upper panel) and $P = 480$ nm (lower panel). For smaller widths, the BIC is located in the lower energy (longer wavelength) branch. At $w = 175$ nm, two plasmon dispersion lines simply cross and the BIC does not exist. For a larger stripe

Figure 4: Sample with a thinner dielectric layer ($t \sim 100$ nm) that excites only plasmonic modes strongly.

(A) and (B) are TM-polarized white light reflection maps for the grating periods of $P = 300$ and 480 nm, respectively. (C) and (D) are the reflection spectrum and the extracted Q factor for $P = 300$ and 480 nm, respectively. The Q factor increases substantially around the bound states in the continuum (BIC) position.

width, the BIC appears in the higher energy (shorter wavelength) branch. As described in Section 2, a BIC can be located either in a low- or high-energy branch depending on the coupling condition. In the plasmonic grating, a standing wave can exist at the Γ position or at the Brillouin zone boundary, and the coupling between two plasmonic modes results in super-radiant and sub-radiant eigenmodes. And the subradiant mode (or plasmonic BIC) can be located either in the low- or high-energy branch depending on the grating parameter. It is noteworthy that similar phenomena have been considered in the context of plasmonic bandgap formation [49–51]. Here, we show that it can be understood in the general framework of non-Hermitian physics.

Moreover, by mixing luminescent dyes in a polymer matrix (Figure 5A), we study the coupling of light emission into plasmonic BICs. Plasmonic structures exhibit strong near-fields and large LDOSs, which results in strong

emission coupling from nearby emitters. The Fourier-plane PL spectrum and image clearly indicate optical mode coupling from dye emission. Figure 5 compare the TM-polarized white reflection spectrum and PL spectrum for a thicker PMMA layer ($t = 500$ nm) and a grating period of $P = 400$ nm. The PMMA layer was mixed with Rhodamin6G (R6G) dyes, and the sample was optically pumped using a 450 nm diode laser. PL spectra were measured for different incidence angles (shaded region in Figure 5B). The red and blue spectra in Figure 5C represent the PL spectra from the metal grating and flat metal regions, respectively. The R6G dye emission spectrum is strongly modified by optical mode coupling (red curves in Figure 5C); the PL spectrum from the flat metal plane exhibits a broad R6G emission spectrum, while the PL spectrum from the metal grating shows sharp PL peaks on top of the broad background spectrum from the uncoupled dye emission. Prominent PL peaks appear because of optical

Figure 5: Trapping of molecular emission at a hybrid plasmonic bound states in the continuum (BIC).

(A) Sample schematic. The R6G dye in a polymer matrix is spin-coated on a silver grating. (B) TM-polarized white light reflection map for $P = 400$ nm. (C) Photoluminescence (PL) spectra at different emission angles, corresponding to the shaded region of the reflection map in (B). The dye emission spectrum is strongly modified by optical mode coupling (red curve). Prominent PL peaks appear because of optical mode coupling, on top of the broad background spectrum from the uncoupled dye emission. The PL peak disappears at the BIC condition ($k_x/k_0 = 0.20$) because of emission trapping.

mode coupling to plasmonic and photonic modes. The PL peak position and peak intensity also change with the emission angle ($k_x/k_0 = 0.15$ – 0.25), following the mode dispersion. By comparing this angle-resolved PL spectra to the reflection spectrum, we find that the PL peak position follows the mode dispersion in the reflection spectrum.

The low-energy branch containing a hybrid plasmonic BIC is indicated by the yellow arrow in the PL spectra (Figure 5C). Interestingly, we notice that the PL peak disappears at the BIC condition ($k_x/k_0 = 0.20$), and only uncoupled background PL remains. Because optical mode coupling to the BIC should still occur by emitters, we attribute this to the trapping of molecular emission at BICs. Our experiment indicates that the dye emission coupled to a BIC is trapped and the PL peak disappears, although an eigenmode exists above the light line. Therefore, both the Fourier-plane white light reflection and PL spectrum measurements indicate the formation of trapped modes (or

BICs) in plasmonic nanostructures. This trapping of molecular emission at the BIC condition implies that plasmonic BIC structures can be highly effective light trapping structures despite their simple geometry. Plasmonic supercavity design based on the BIC concept can enable sharper resonances together with large LDOSs and strong near-field enhancement.

The trapping of molecular emission at the BIC condition is also confirmed from the Fourier-plane PL images. A Fourier-plane PL image at a specific wavelength was measured using a proper bandpass filter. Figure 6 compares TM- and TE-polarized PL images for three PL wavelengths ($\lambda = 600$, 610 , and 620 nm). These three wavelengths correspond to the three dotted horizontal lines in the reflection map shown in Figure 5B. In the TM-polarized PL image, the PL line is interrupted at the hybrid BIC condition (yellow dotted circles at $\lambda = 610$ nm), indicating the trapping of molecular emission. On the

Figure 6: Fourier-plane photoluminescence (PL) images.

(A) and (B) TM- and TE-polarized PL images for three PL wavelengths ($\lambda = 600, 610$, and 620 nm), corresponding to the three dotted horizontal lines in the reflection map in Figure 5B. The TM-polarized PL image shows the interrupted PL line at the BIC condition (yellow dotted circles at $\lambda = 610$ nm), indicating the trapping of molecular emission. The TE-polarized PL images only show continuous arcs.

contrary, the TE-polarized PL images show just continuous arcs from the TE-polarized photonic modes. Therefore, plasmonic BICs may be ideal for a very efficient light trapping structure, which is highly desirable, e.g., for plasmonic nanolasers.

Our Fourier-plane spectroscopy and imaging measurements clearly demonstrate all characteristic features of BICs and indicate that plasmonic BICs can enable an extremely effective light confinement. We expect the BICs to facilitate the design of various plasmonic supercavities with sharper optical resonances by using either extended or localized plasmonic modes. Such plasmonic supercavities can be applied to nanolasers, optical sensing, and nonlinear enhancement. Furthermore, plasmonic supercavities can be designed with novel functionalities. For example, polarization vortices result from BICs in the far field [14, 38, 52, 53], which can be used for vortex beam generation and topological photonics. Moreover, as discovered recently, circularly polarized states can be derived from BICs by in-plane symmetry breaking [54], and this might be useful for splitting and manipulating valley exciton emission in monolayer transition metal dichalcogenides for future nanophotonic devices [55, 56].

Therefore, we envision that many interesting future opportunities exist regarding plasmonic supercavity design.

We notice that a study by Zhang et al. [38] utilized plasmonic structures for polarization vortex measurements. While this reference focused on the measurement of polarization vortices in single-resonance parametric BICs, we here conducted a thorough study on plasmonic BICs occurring from two coupled resonances (both hybrid plasmonic and purely plasmonic BICs). We also studied molecular emission coupling to BICs and clearly demonstrated the trapping of molecular emission at the plasmonic BIC condition.

4 Conclusions

In conclusion, we experimentally investigated BICs in plasmonic structures via Fourier-plane spectroscopy and imaging. The dispersion relation in plasmonic structures was studied by measuring an angle-resolved white light reflection spectrum. By moving a pinhole in the Fourier plane and combining the measured reflection spectra, we obtained a reflection map as a function of wavevector and

wavelength. Our measurements clearly showed all features expected for both symmetry-protected and nonsymmetry-protected BICs, confirming the strong suppression of radiative damping around a BIC. Furthermore, we demonstrated that the position of a BIC can be switched from one optical mode branch to the other by tuning a metal grating parameter. Moreover, by mixing luminescent dyes in a polymer matrix, we studied the coupling of light emission into BICs. The PL peak disappeared at the BIC condition, and this was attributed to the trapping of molecular emission at plasmonic BICs. Therefore, both white light reflection and dye emission measurements in the Fourier plane clearly indicate the formation of trapped modes in plasmonic nanostructures. This observation implies that plasmonic BICs can enable an effective light trapping structure despite their structural simplicity. Plasmonic supercavity design based on the BIC concept may provide many interesting future opportunities for nanolasers, optical sensing, and nonlinear optical enhancement with novel functionalities.

5 Methods

An optically thick silver film (~200 nm) was sputtered on Si (111) at 350 °C at a deposition rate of 1.56 nm/s to obtain a smooth metal surface [57, 58]. The quality of the sputtered silver film was verified by the X-ray diffraction and AFM measurements. On top of the flat silver film, an array of silver gratings with varied grating parameters (period and width) was fabricated by electron beam lithography and silver evaporation, where the size of each grating was $20 \times 20 \mu\text{m}$. Finally, a PMMA layer was spin-coated on the silver grating.

A collimated white light beam (Ocean Optics, HL-2000-LL) was linearly polarized using a broadband Glan Thompson polarizer (Thorlabs) and was focused onto a sample surface by a microscope objective with an NA of 0.8 or 0.42. A pinhole with a diameter of $20 \mu\text{m}$ was mounted on a motorized stage (Thorlabs, Z812B) and placed in the Fourier plane. When the pinhole size is fixed, the angular resolution ($\Delta\theta$) varies depending on the pinhole position in the Fourier plane (note that $k_x = k_0 \sin\theta$). The maximum angular resolution achieved in our measurements was estimated to be approximately 0.55° for NA 0.42 and around 0.8° for NA 0.8. With the larger NA of 0.8, the angular resolution achieved was smaller but a wider range of angles ($\pm 53^\circ$) can be scanned.

For PL measurements, the R6G dyes were mixed in 9 wt% PMMA/chlorobenzene with a dye concentration of $2.78 \times 10^{-3} \text{ mol/L}$. This PMMA/R6G solution was spin-coated on the silver grating at a spin speed of 4000 rpm for 1 min and baked at 180 °C for 1 min. A 450 nm-diode laser (Thorlabs, CPS450) with an optical power of 0.33 mW was used to excite the R6G dyes. A longpass filter at 500 nm was used to cut off the reflected laser light from the sample surface. The PL spectrum was measured using a pinhole of diameter $50 \mu\text{m}$. For Fourier-plane PL image measurements, bandpass filters with a 10 nm bandwidth (Thorlabs) were used for three center wavelengths of $\lambda = 600, 610, \text{ and } 620 \text{ nm}$.

Acknowledgments: This work was financially supported by the National Research Foundation (NRF) of Korea (NRF-2019R1A2C1008330) and Research fund (1.200095.01) of UNIST (Ulsan National Institute of Science and Technology).

Author contribution: All the authors have accepted responsibility for the entire content of this submitted manuscript and approved submission.

Research funding: This work was financially supported by the National Research Foundation (NRF) of Korea (NRF-2019R1A2C1008330) and Research fund (1.200095.01) of UNIST (Ulsan National Institute of Science and Technology).

Conflict of interest statement: The authors declare no conflicts of interest regarding this article.

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- Supplementary Material:** The online version of this article offers supplementary material (<https://doi.org/10.1515/nanoph-2020-0343>).