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Title: Forward/backward switching of plasmonic wave propagation using sign-reversal coupling

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Abstract: Backward waves with wave-front propagation opposite in direction to that of energy flow have attracted considerable interest in the context of photonic metamaterials. However, switching between forward and backward waves in the same frequency range has remained a challenge. Here, on a platform of coupled designer surface plasmon resonators in the microwave regime, we demonstrate multi-band forward/backward switching of plasmonic wave propagation. Our approach makes use of sign-reversal coupling that occurs when switching the coupling configuration between tightly confined photonic modes. Direct experimental measurements of plasmon dispersion curves confirm the forward/backward propagation of plasmonic

waves in the same frequency range. Our study provides a solution to forward/backward switching of subwavelength plasmonic wave propagation, and may find potential applications in photonic integrated systems.

In negative refractive index materials, the phase velocity of electromagnetic (EM) waves is opposite to the direction of the power flow. This phenomenon is widely known as the backward wave propagation. This unusual class of EM waves forms the foundation of negative refraction^[1] and has attracted considerable interests in the past decade in the context of photonic metamaterials.^[2,3] Recently, the concept of backward waves has been further extended to surface plasmon polaritons (SPPs)^[4,5]. However, in most previous approaches, the backward waves only exist in a single frequency band, and lack the capability of switching between forward and backward waves in the same frequency band. Moreover, in previous approaches of achieving backward waves for SPPs, the backward propagation generally occurs at the anomalous dispersion region within a single narrow band, being unable to cover multiple frequency bands.

Apart from typical SPPs that propagate on a continuous metal surface,^[4-5] plasmonic waves can also propagate through near-field coupling between plasmonic particles supporting tightly localized photonic modes.^[6] In fact, this kind of near-field coupling between tightly localized photonic modes, as an analogue of the tight-binding atomic wave coupling in solid-state systems, is of fundamental importance in many modern photonic applications ranging from slow-light engineering,^[7-12] quantum simulation,^[13,14] to topological photonics^[15-18] and $\mathcal{PT}$ -symmetric photonics.^[19-21] The wave propagation direction along a chain of plasmonic particles with tightly localized photonic modes is determined by the sign of the coupling factor,^[6,22] which stems from the overlap of the evanescent tails of tight-binding photonic modes. Although the coupling strength can be easily tuned by adjusting the inter-particle distance, it remains difficult to flip the sign of coupling factor^[23-28] and consequently switch between forward and backward wave propagations.

Here, by directly measuring the tight-binding Bloch waves on a periodic array of coupled designer SPP resonators (“meta-particles”) ^[29-32] in the microwave regime, we demonstrate that the coupling factor between tightly localized photonic modes can reverse its sign by switching between two different coupling configurations. As a consequence, two surface modes with positive and negative group velocities arise in the same frequency range, forming a Dirac-point-like crossing of dispersions.^[33] We experimentally measured the Bloch wavevectors of these modes as a function of frequency and achieved the functionality of switching between forward and backward surface Bloch waves using selective excitation. We also show that this sign-reversal coupling approach to realize forward/backward switching is applicable to any multipolar mode that is achievable in multiple frequency bands.

We first consider two identical two-dimensional (2D) resonators hosting tightly localized photonic modes, as shown in **Figure 1a**. Without loss of generality, we assume that the 2D resonators are made of a homogeneous and isotropic material with a constant permittivity. In a typical coupling configuration,^[22] the two resonators couple through facing two “poles,” or intensity maxima, of resonant wave patterns that are tightly confined around the two resonators. Defining the x -axis as in **Figure 1a** and setting the radii of two identical resonators as R , the multipolar resonance mode profile of each resonator can be expressed as $E_z = B_m(kr)\cos(m\varphi)$, where m is the order of multipolar resonance mode, and (r, φ) are the polar coordinates with the center of each

resonator as the origin. $B_m(kr) = \begin{cases} J_m(k_d r), & r < R \\ \frac{J_m(k_d R)}{H_m^1(ik_0 R)} H_m^1(ik_0 r), & r > R \end{cases}$, where $J_m(\cdot)$ is

Bessel function, $H_m^1(\cdot)$ is first-kind Hankel function, and k_d and k_0 are wavevectors in resonators and the environment, respectively. The coupling factor (κ) is a mathematical expression^[22] that depends on the integration of overlapped resonance wave fields of

the two resonators (Figure S1, Supporting Information). We can easily understand that this kind of “pole-pole” coupling is associated with a strong interaction between the two resonators.

Interestingly, there is another configuration to couple these two resonators through facing two “nodes,” or intensity minima, of resonance wave patterns between the two resonators, as shown in **Figure 1b**. In this case, the multipolar resonance mode profile of each resonator can be expressed as $E_z = B_m(kr)\sin(m\varphi)$. Intuitively, the strength of coupling, or the magnitude of coupling factor κ , in this “node-node” coupling configuration should be relatively small as compared with that in the “pole-pole” coupling configuration. However, a rigorous derivation shows that the coupling factor κ keeps its magnitude unchanged, and simply reverses its sign (Figure S2, Supporting Information). In other words, this sign-reversal coupling reverses the phase relation between binding/anti-binding photonic modes of two coupled resonators, as well as the Bloch-wave dispersion slope of a periodic resonator array, as we will demonstrate later.

To facilitate the experimental retrieval of coupling factor κ , we first describe the two coupled resonators with the coupled mode theory^[34] as follows:

$$\begin{cases} -\frac{da_1}{dt} = i\omega_0 a_1 + i\kappa\omega_0 a_2 \\ -\frac{da_2}{dt} = i\omega_0 a_2 + i\kappa\omega_0 a_1 \end{cases} \quad (1)$$

where a_1 and a_2 represent resonance wave fields in the two resonators, and ω_0 denotes the intrinsic resonance frequency of a multipolar resonance mode in a single resonator. By solving this eigenvalue problem in Eq. (1), we obtain two orthogonal eigen solutions of $[a_1 \ a_2]^T$ as $[1 \ -1]^T$ and $[1 \ 1]^T$, in which the former corresponds to the out-of-phase (a_1 and a_2 differ by a phase of π) coupled mode with eigen frequency $\omega_{1,-1} = \omega_0 - \kappa\omega_0$, and the latter to the in-phase (a_1 and a_2 have the same phase) coupled mode with eigen frequency $\omega_{1,1} = \omega_0 + \kappa\omega_0$. This relation indicates that the magnitude of coupling

factor κ determines the frequency shift in mode splitting, and the sign of κ corresponds to the phase relation of binding/anti-binding modes, i.e. whether it is out-of-phase or in-phase.

We then proceed to experimental demonstration. We adopt recently proposed designer SPP resonators^[32] as shown in **Figure 2a**, which are constructed by decorating periodic subwavelength grooves on circular metallic disks. Designer SPPs are tightly confined EM modes on patterned metallic structures with subwavelength corrugations, whose dispersion properties and spatial confinements are akin to those of natural SPPs at optical frequencies.^[31] Recently, designer SPP resonators are shown to support localized designer SPPs,^[31-32] opening an opportunity to image directly the near-field coupling mechanism. Because of the analogy between localized EM resonance modes and atomic wave functions, these designer SPP resonators are termed as a type of “meta-atoms”^[32] in the field of metamaterials. Another benefit of this designer-SPP-resonator metamaterial platform is that it allows selective excitation of multipolar resonance modes with opposite parity, meaning that the analogous “atomic wave functions” can be selectively controlled. We construct a periodic array of designer SPP resonators, as shown in **Figure 2b**. A monopole source is employed at one side of the array to selectively excite a multipolar resonance mode. The Bloch waves on the array is recorded by a near-field scanning probe, connected to a microwave network analyzer.

We first consider coupling between two designer SPP resonators. **Figure 3a** shows the measured near-field transmission spectra through this coupled-resonator dimer. We adopt two excitation configurations. Firstly, we locate the source and the probe in symmetric positions at two ends of the dimer, as indicated by a pair of red dots in the inset of **Figure 3a**. Secondly, the source and the probe are both placed by one side of two resonators, as indicated by a pair of blue dots in the inset of **Figure 3a**. We call the

first configuration as “end excitation” and the second as “side excitation.” Each of these excitation configurations will selectively excite one of the multipolar resonance modes with opposite parity. The transmission spectrum for a single resonator is also measured for comparison by placing the source and probe at opposite sides of the resonator. Three resonance modes can be observed in a single resonator at 5.34 GHz, 6.07 GHz, and 6.44 GHz as shown in **Figure 3a**. They correspond to the quadrupole (labeled as “Q”), hexapole (labeled as “H”), and octopole (labeled as “O”) modes, respectively, as will be demonstrated later.

Let us consider the end excitation. When the coupled-resonator dimer is excited with the end excitation, each of the three resonance modes in a single resonator splits into binding and anti-binding modes: from one mode at $Q=5.34$ GHz to two modes at $Q_\pi=5.19$ GHz (binding mode) and $Q_0=5.49$ GHz (anti-binding mode), from one at $H=6.07$ GHz to two at $H_0=6.03$ GHz (binding mode) and $H_\pi=6.11$ GHz (anti-binding mode), and from one at $O=6.44$ GHz to two at $O_\pi=6.42$ GHz (binding mode) and $O_0=6.47$ GHz (anti-binding mode). Here the subscript “ π ” or “0” denotes out-of-phase or in-phase phase relation for the two resonators after mode splitting. To explicitly show the phase relation, we measure the resonance wave pattern for each resonance mode by scanning the probe in a transverse plane 1 mm above the coupled-resonator dimer, as shown in **Figure 3b**. The source position is indicated by a red arrow. We observe that for the coupled quadrupole and octopole resonance modes, the two resonators are out-of-phase at lower resonance frequencies (Q_π and O_π), and in phase at higher resonance frequencies (Q_0 and O_0). However, the situation is reversed for the hexapole mode: the two resonators are in-phase at the lower resonance frequency (H_0), and out-of-phase at the higher resonance frequency (H_π). The different phase relation of two coupled resonators for the coupled hexapole resonance mode is simply because the hexapole

mode is an odd mode (mode order $m=3$) while both the quadrupole mode (mode order $m=2$) and the octopole mode (mode order $m=4$) are even modes.

Using the eigen-frequency analysis from Eq. (1), we can experimentally retrieve the coupling factor κ from the measured split resonance frequencies and the phase relation between the coupled two resonators. We obtain that with end excitation, $\kappa_Q = 0.0281$, $\kappa_H = -0.0066$, and $\kappa_O = 0.0039$ for the coupled quadrupole, hexapole, and octopole resonance modes, respectively. It is apparent that a higher-order multipolar resonance mode has a smaller magnitude of coupling factor due to its tighter field confinement. The negative sign of κ_H for the hexapole resonance mode arises from the phase relation of binding/anti-binding modes, which is opposite to that in the quadrupole and octopole resonance modes.

Now we consider the side excitation. It can be seen in **Figure 3a** that the split resonance frequencies match well with those in the end excitation. For the quadrupole and octopole modes, because of their four-fold rotational symmetry, the resonance wave patterns for their split modes are almost the same with those in the end excitation. We thus do not discuss them here. However, it is not the same situation when it comes to the hexapole mode. As shown in **Figure 3b**, we measure the resonance wave patterns for the two split hexapole modes ($H_\pi^* = 6.03$ GHz and $H_0^* = 6.11$ GHz) in the coupled-resonator dimer, where the source position is indicated by a red arrow. While all previous resonance wave patterns correspond to the “pole-pole” coupling configuration as in **Figure 1a**, here these two resonance wave patterns correspond to the “node-node” coupling configuration as in **Figure 1b**. Although frequencies of the two split hexapole modes are consistent with those in the “pole-pole” coupling configuration, the phase of these two split modes has been exchanged: the in-phase mode (H_0^*) now has a higher resonant frequency than the out-of-phase mode (H_π^*). Following the process above to

retrieve the coupling factor, we can get $\kappa_H^* = 0.0066$ for the hexapole mode in the “node-node” coupling configuration. This confirms the sign reversal of coupling factor compared to the previously retrieved $\kappa_H = -0.0066$ for the hexapole mode in the “pole-pole” coupling configuration.

We can proceed to construct a coupled-resonator optical waveguide (CROW)^[22] with a periodic array of designer SPP resonators, as shown in **Figure 4a**. Each multipolar resonance mode in a single resonator now spreads continuously to form a transmission band on the CROW. We describe the CROW that consists of an infinite chain of designer SPP resonators with coupled mode theory^[34] as follows:

$$-\frac{da_n}{dt} = i\omega_0 a_n + i\kappa\omega_0 a_{n-1} + i\kappa\omega_0 a_{n+1} \quad (2)$$

where a_n , a_{n-1} and a_{n+1} denote resonance fields in the n -th, $(n-1)$ -th and $(n+1)$ -th resonators, respectively. The periodicity of this structure allows the application of Bloch theorem which gives $a_{n+1} = a_n e^{iK\Lambda}$, where K represents the wavevector of Bloch waves on the CROW, and Λ is the periodicity of designer SPP resonators. Then the intrinsic dispersion relation of the Bloch waves on the CROW can be obtained as $\omega = \omega_0 [1 + 2\kappa \cos(K\Lambda)]$, being consistent with Yariv’s original derivation.^[22] Apparently, the sign of coupling factor κ affects the relation between Bloch wavevectors and frequencies.

Given the experimentally retrieved coupling factors, we plot with solid black lines in **Figure 4b** the dispersion curves for the quadrupole, hexapole, and octopole resonance modes along the CROW in the “pole-pole” coupling configuration. This coupling configuration can be achieved with the end excitation, by locating the source and probe at positions as indicated by a pair of red dots in **Figure 4a**. Working bandwidth gets narrower as the mode order gets higher, because the tighter field confinement in a higher-order mode leads to a smaller magnitude of κ . Because of the

positive sign of κ_Q and κ_O , the dispersion curves of Bloch waves for the quadrupole and octopole resonance modes exhibit properties of “backward waves,” in which the phase velocity and the group velocity are in opposite directions. Regarding the hexapole resonance mode, because of its negative coupling factor κ_H , its Bloch waves are “forward waves,” whose phase velocity and group velocity are in the same direction.

Similar to previous discussions on the coupled-resonator dimer, the side excitation in the CROW will turn on the “node-node” coupling configuration for the hexapole resonance mode with a sign-reversal coupling factor, i.e. $\kappa_H^* = -\kappa_H$. The setup of end excitation is indicated by a pair of blue dots in **Figure 4a**. We plot the dispersion curve for the hexapole resonance mode in the “node-node” coupling configuration in **Figure 4b** with a red solid line. This dispersion curve shares the same bandwidth with that of the hexapole resonance mode in the “pole-pole” coupling configuration. The crossing of the two dispersions gives rise to a Dirac point.^[33] Because of the sign reversal of coupling factor, Bloch waves of this dispersion curve in the “node-node” coupling configuration exhibit properties of “backward waves,” where the phase velocity and group velocity are in opposite directions.

We adopt two steps in experiment to demonstrate the above predictions. In the first step, we measure transmission spectra to verify transmission bandwidth. The results are shown in **Figure 4c**, being consistent with dispersion curves in **Figure 4b**. In the second step, we directly image Bloch waves propagating on the CROW. We select three characteristic Bloch wavevectors ($K\Lambda = 0, \frac{\pi}{2}, \pi$) as indicated by three dots in each dispersion curve in **Figure 4b**. For the quadrupole mode in the “pole-pole” coupling configuration, as shown in **Figure 4d**, the frequency of Bloch waves decreases as the Bloch wavevector increases. A similar situation applies to the octopole mode in the

“pole-pole” coupling configuration, as shown in **Figure 4e**. These results confirm their “backward-wave” properties.

What is more interesting is the hexapole mode that can be excited with both the “pole-pole” and “node-node” coupling configurations, where sign reversal of the coupling factor is expected. **Figure 4f** shows the measured Bloch waves of the hexapole mode in the “pole-pole” coupling configuration. It can be seen that the frequency of Bloch waves increases as the Bloch wavevector increases. This confirms the “forward-wave” properties of these Bloch waves. In **Figure 4g**, the measured Bloch waves are plotted for the hexapole mode in the “node-node” coupling configuration. Instead, the frequency of Bloch waves decreases as the Bloch wavevector increases. Apparently, they are “backward waves.”

Note that the sign-reversal coupling for the quadrupole and octopole modes can also be achieved by locating the excitation position at the middle between the current positions of end excitation and side excitation. We also plot in **Figure 4b** the dispersion relations of quadrupole and octopole mode (black dashed lines) for the “node-node” coupling configuration, when the excitation is relocated at the middle between the current positions of end excitation and side excitation. We can see that the surface modes will also switch from backward waves to forward waves in the same frequency range (experimental data now shown for brevity). Therefore, the sign-reversal coupling approach to realize the switching between forward and backward waves is general, being applicable to any multipolar resonance mode in multiple frequency ranges.

In the following, we will show that the sign-reversal coupling can occur in a single CROW, in which the Bloch waves with both positive and negative coupling factors can co-exist. We construct a right-angle bent CROW as shown in **Figure 5a**. Similar to previous demonstrations, the end (side) excitation is achieved by placing the source and

probe at positions indicated by a pair of red (blue) dots. Because of the four-fold rotational symmetry of mode profiles, the guidance of Bloch waves for the quadrupole and octopole modes will not be affected by the bending corner. In other words, their coupling factors will not change over the bending corner. On the other hand, the Bloch waves of the hexapole mode, when they propagate across the bending corner, will switch from the “pole-pole” coupling configuration to the “node-node” coupling configuration, or vice versa, and thus induce the sign reversal of coupling factor at the bending corner. We first measure the transmission spectra as shown in **Figure 5b**, whose results are consistent with dispersion curves in **Figure 4b** in the main text. We then image the Bloch waves. Both the quadrupole mode (**Figure 5c**) and the octopole mode (**Figure 5d**) are excited in the “pole-pole” coupling configuration. The sign-reversal coupling does not occur at the bending corner. For the hexapole mode excited with end excitation (**Figure 5e**), the “pole-pole” (“node-node”) coupling configuration applies to the horizontal (vertical) arm. The coupling factor changes its sign from negative to positive at the bending corner. The hexapole mode can also be excited with side excitation (**Figure 5f**), where the “node-node” (“pole-pole”) coupling configuration applies to the horizontal (vertical) arm. This time the coupling factor changes its sign from positive to negative at the bending corner.

In summary, we have revealed a new way to flip the propagation directions of plasmonic waves along a chain of plasmonic particles by using the sign-reversal coupling between tightly localized photonic modes. On the platform of designer SPP resonators, we confirm the sign reversal of coupling factors by experimentally monitoring the phase of resonance wave fields in the coupled two photonic modes. By monitoring Bloch waves in a periodic array of designer SPP resonators, we show that the sign-reversal coupling will induce two different waveguiding dispersion curves in

the same frequency range with opposite group velocities. The forward wave and backward wave can be switched within the same frequency range through changing the excitation position. This sign-reversal coupling approach can be applied to any multipolar resonance modes in multiple frequency ranges. In view of the demanding coupling tuning and backward waves, we expect that our study will find novel use in designs of functional photonic systems and metamaterials.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Figures

Figure 1. Coupling configurations between tightly confined multipolar photonic modes.
a) “Pole-pole” coupling configuration by facing intensity maxima between resonators.
b) “Node-node” coupling configuration by facing intensity minima between resonators.

Figure 2. Experimental setup for measuring coupled resonance waves. a) Photo of a designer SPP resonator with radius $R=12$ mm. The depth of grooves is $r=9$ mm. The width and periodicity of grooves are $a=0.625$ mm and $d=1.255$ mm, respectively. b) Schematic of a periodic array of designer-surface-plasmon resonators. A network analyzer records the Bloch waves by scanning the near-field distribution above the resonator array.

Figure 3. Mode splitting of a coupled-resonator dimer. a) The measured near-field transmission spectra through a coupled-resonator dimer with end and side excitations. Inset: photo of the two designer-surface-plasmon resonators with inter-resonator distance $\Lambda = 30$ mm. b) The measured resonance wave patterns. The position of source is indicated by a red arrow.

Figure 4. Bloch waves on a periodic resonator array. a) Photo of the designer SPP array with periodicity $\Lambda = 30$ mm. Positions of the source and probe for the end and side excitations are indicated as a pair of red dots and blue dots, respectively. b) Dispersion curves calculated with experimentally retrieved coupling factors for the quadrupole, hexapole, and octopole modes. Black solid lines correspond to the “pole-pole” coupling configuration. The red solid line corresponds to the “node-node” coupling configuration for the hexapole mode. The black dashed lines correspond to the “node-node” coupling configuration for the quadrupole and octopole modes. c) The measured transmission spectra through the array. The transmission spectrum without the array is also measured for comparison. d-g) Measured Bloch waves of the quadrupole (d), octopole (e) and hexapole (f, g) modes with three characteristic Bloch wavevectors as indicated by three dots in each dispersion curve in (b). The red arrow indicates the position of source.

Figure 5. Sign-reversal coupling in a bent resonator array. a) Photo of the bent designer SPP array. Positions of the source and probe for the end and side excitations are indicated as a pair of red dots and blue dots, respectively. b) Measured transmission spectra through the bent resonator array. The transmission spectrum without the bent resonator array is also measured for comparison. c-f) Measured Bloch waves of the quadrupole (c), octopole (d) and hexapole (e,f) modes. The red arrow indicates the position of source.