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Loss-Driven Topological Transitions in Lasing

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We experimentally observe lasing in a hexamer plasmonic lattice and find that, when tuning the scale of the unit cell, the polarization properties of the emission change. By a theoretical analysis, we identify the lasing modes as quasi-bound-states in continuum of topological charges of zero, one, or two. A T-matrix simulation of the structure reveals that the mode quality \( Q \) factors depend on the scale of the unit cell, with highest \( Q \) modes favored by lasing. The system thus shows a loss-driven transition between lasing in modes of trivial and high-order topological charge.

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Bound states in the continuum (BICs) are peculiar eigenstates of quantum or classical wave systems: their energies lie within a continuum of other states, but they are nevertheless completely uncoupled from those states. For optical structures such as gratings, cavities, and photonic crystals, this means that their BIC modes are decoupled from free-space radiation and appear as dark states: light cannot couple with the far field at the energy and momentum of the BIC [1,2]. From the fundamental perspective, photonic BICs can be viewed as vortex centers of light polarization. When one measures the polarization of light radiated by the structure from different angles, corresponding to different momentum, the polarization winds around the BIC momentum. Because of this winding, it is impossible to define what would be the polarization exactly at the BIC momentum, which therefore must appear dark, similar to as when the core of a vortex is empty. This description has naturally connected the BICs to the realm of more general topological phenomena, including topological robustness and topological transitions [3–6]. The polarization vortex is associated with the existence of a protected and quantized topological charge that tells how many times the polarization winds in a path circulating the BIC momentum [7,8]. This charge is conserved and cannot be removed with small perturbations. In order to observe BICs, they can be weakly coupled to the radiative continuum via an intentionally designed leakage mechanism or due to material loss [9–11]; the presence of an edge can also act as a leakage channel [12,13]. Such leaky or lossy BICs are called quasi-BICs. Quasi-BICs can have extremely large \( Q \) factors [14–22], which is promising for numerous applications in lasing, filtering, or sensing [10,11,23–26].

BICs of topological charge \( |q| = 1 \) have been observed in many systems, from photonic crystals to nanoplasmonic structures [7,8,10,12,13,27–29]. Switching between non-degenerate BICs with different topological charges of \( |q| \leq 1 \) was recently achieved [29]. The interest in BICs with large topological charges (\( |q| > 1 \)) and their generation, evolution, or annihilation is only just starting. BICs of high-order charge are predicted to exist in systems with a sixfold rotational symmetry [8,30] and it has been theoretically proposed that a BIC of charge \( |q| = 2 \) can undergo a topological transition and break into two BICs of charge \( |q| = 1 \) [31]. The first experimental studies demonstrating vortex beams generated by BIC of higher topological charge in photonic crystals have recently emerged [16,32], while observation of lasing in BICs of higher topological charges still lacks in plasmonic lattices.

In this Letter, we observe lasing in a plasmonic lattice from a BIC of topological charge \( q = -2 \) and demonstrate several topological transitions driven by losses as a structural parameter of the unit cell is changed. We use plasmonic nanoparticle arrays due to their strong dipole moments in the nanoparticles [33–38]. The lasing mode is identified by using theory of coupled dipoles together with group theory and \( T \)-matrix calculations [39,40]. We show that the energy of the modes as well as their loss properties change as the structural parameter is varied. The \( Q \) factor of the modes changes, and the mode with the highest \( Q \) factor lases. By comparing the theory with the experimental measurements, we characterize the polarization vorticity of the lasing mode and confirm topological transitions between a bright mode and quasi-BIC modes of topological charges \( q = -2, q = -1 \), and \( q = +1 \).

The topological charge of a BIC is defined as [8]

\[
q = \frac{1}{2\pi} \oint_C dk \cdot \nabla_k \Phi(k),
\]

where \( \Phi(k) = \arg[p(k) \cdot \hat{x} + ip(k) \cdot \hat{y}] \) is the phase of the polarization vector projected onto the \( xy \) plane in momentum space \( p(k) = (\hat{x} \cdot \langle u_k(r,z) \rangle)\hat{x} + (\hat{y} \cdot \langle u_k(r,z) \rangle)\hat{y} \), and \( \langle u_k \rangle \) indicates the spatial average of the periodic part of the electric field \( E_k(r,z) = e^{ik_zz}u_k(r,z) \) at constant vertical distance \( z \) from the lattice plane much larger than the unit cell size. The charge \( q \) counts the number of times
FIG. 1. (a) The hexamer is composed of six particles (black disks) arranged in a regular hexagon of side length $b$. Purple and orange arrows represent the longitudinal $e^L_i$ and transverse $e^T_i$ unit vectors used in the simple hexamer model, having indicated only nearest neighbor. The hexamer constitutes the unit cell of a triangular lattice of periodicity $a$ (red points). Because of the finite size of the particles, whose diameter is $d$, the edge-to-edge distance between nanoparticles is $l = b - d$. (b) Modes of an isolated hexamer, where the arrows correspond to the electric dipole moment. The modes are labeled according to the corresponding IRs of the symmetry group $C_6$. (c)–(f) Amplitude and phase of the four IRs, filtered with a polarizer oriented according to the arrow in each panel, while the phase in the last column is unfiltered. The modes and their respective topological charges are (c) $A'$: $q = +1$, (d) $E'_1$: $q = 0$, (e) $E'_2$: $q = -1$, and (f) $B'$: $q = -2$. (d) and (e) Balanced superpositions of the degenerate modes with zero relative phase.

the polarization vector $p(k)$ goes around a specific $k$ point, e.g., the $\Gamma$ point. The sign of the topological charge gives the orientation of the winding.

We start by theoretically analyzing the modes of an isolated hexamer [Fig. 1(a)]. Each particle has an in-plane dipole moment $p_i = (p^L_i, p^T_i)$, coupled to all other dipoles in a polarization-dependent way, as in Ref. [12]. We set $\Omega_L$ ($\Omega_T$) to be the coupling between dipoles oriented longitudinally (transversely) to the link $e^L_i = (r_i - r_{i+j})/|r_i - r_{i+j}|$ (such that $e^L_i \cdot e^L_j = 0$) connecting particles $i$ and $i + j$ (typically for dipoles $\Omega_T \gg \Omega_L$). The bare dipole oscillation frequency is $\omega_0 \gg \Omega_{L,T}$. The modes are found from

$$\tilde{\mathbf{p}}_i = \omega_0 \mathbf{p}_i + \sum_{j \neq i} \frac{\Omega_L}{R_j} (\mathbf{p}_{i+j} \cdot e^L_i) e^L_j + \frac{\Omega_T}{R_j} (\mathbf{p}_{i+j} \cdot e^T_i) e^T_j,$$

where $R_j = |r_i - r_{i+j}|$. In the basis $(p^L_i, p^T_i)$, we find 12 modes centered around $\omega_0$. The spatial structure of the lower energy set of eigenmodes is shown in Fig. 1(b), where arrows correspond to the electric dipole moment orientation. The modes belong to the symmetry class of the $C_6$ group, the singlets to the $A$ and $B$ irreducible representations (IRs), and the doublets to the $E_1 = (E'_1, E'_2)$ and the $E_2 = (E''_1, E''_2)$ IRs. The ordering of these modes does not depend on the interparticle distance, in the isolated hexamer case, as losses are not included. From finite element method simulations on arrays with periodic boundary conditions, we find the same modes, and also another set of six modes [41] that have a larger out-of-plane character. Those are not excited in our setup and therefore we will focus on the $A'$, $E'_1$, $E'_2$, and $B'$ modes.

From the electric dipole moment orientation in each mode, we calculate the real-space diffraction pattern generated by the nanoparticle array, together with the far field in momentum space. BICs have distinct momentum-space features, as visible in Figs. 1(c)–1(f), where the far field is obtained by Fourier transforming the real-space field with a polarization filter oriented according to the arrow in each panel; these calculations are done as in Refs. [12,42]. The last column in Fig. 1(c)–1(f) shows the phase of each mode’s polarization calculated from the Stokes parameters $[\Phi = \frac{1}{2}\arctan(S_1/S_2)]$ without any polarization filter. Figure 1(d) shows that the $E'_1$ doublet modes are bright modes, since their emission in momentum space is featureless along all polarization directions while the phase is uniform. These modes have no topological charge ($q = 0$) and correspond to dipoles oscillating in phase along the same direction. All other modes are quasi-BICs, and the dipole oscillate in such a way that the emission from $k = 0$ is dark. With a polarization filter, we see a nonzero emission in the vicinity of the $\Gamma$ point, on two
or four opposite lobes of a donut-shaped amplitude pattern [14,20,29,43]. As the polarization filter is rotated from vertical polarization in a clockwise direction, the lobes rotate as well, following the nontrivial phase polarization (see last column). In particular, the polarized amplitude of mode \(A'\) in Fig. 1(c) has two lobes that rotate in the clockwise direction, which correspond to \(q = 1\). Accordingly, the phase in the last column of Fig. 1(c) shows a \(2\pi\) rotation, corresponding to a unit winding. Similarly, the doublet modes \(E'_2\) have \(q = -1\) in Fig. 1(e), since the two lobes rotate counterclockwise; this is also visible in the different phase winding. Finally, the charge \(q = -2\) of mode \(B'\) in Fig. 1(f) is evidenced by a four-lobe rotation in a counterclockwise direction. Consequently, the phase in the last column has an overall \(4\pi\) winding. The topological charge \(q_{\text{IR}}\) of a mode corresponding to an IR can be derived [41] from the \(n\)-fold rotational symmetry \(C_n\),

\[
q_{\text{IR}} = 1 - \frac{n}{2\pi} \arg(\epsilon_{\text{IR}}),
\]

where \(\epsilon_{\text{IR}}\) can be found in character tables. Values given by Eq. (2) for \(C_n\) equal those given by our analysis of coupled dipoles in Fig. 1.

We now consider an extended array composed of hexamers arranged in a finite-size triangular lattice. The edge-to-edge distance between nanoparticles of the same hexamer is \(t\). The geometry of the system is depicted in Fig. 1(a). We fabricated arrays of cylindrical gold nanoparticles with an edge-to-edge distance that is varied as \(45 < t < 200\) nm. The system hosts dispersive plasmonic-poitonic modes [33–35,38]. The dispersion of such an array for \(t = 50\) nm is shown in Fig. 2(a). The arrays were combined with a solution of fluorescent dye molecules (IR-140) with a concentration of 10 mM. Under optical pumping with a left-circularly polarized (LCP) 100 fs laser pulse [center wavelength 800 nm (1.55 eV)], we observe a nonlinear increase in the emission at \(k_t = 0\) with increasing pump fluence as shown in Fig. 2(b): arrays with different \(t\) show different behavior. Furthermore, the peak intensity differs as well: the lasing peak of the \(t = 120\) nm array is about 1 order of magnitude larger than the one for the \(t = 200\) nm array. More details of the experiments are given in the Supplemental Material [41], where we also show the real-space emission from the arrays in the lasing regime.

The different thresholds, peak intensities, and lasing energies in Fig. 2(b) and 2(c) hint toward having a qualitatively different mode lasing for different array geometries. To understand this, we recorded angle- and polarization-resolved measurements of the lasing emission from arrays with a wide range of different hexamer sizes. In Figs. 2(g)–2(j), we show angle-resolved images of arrays with \(t = 50, 120, 170,\) and 200 nm with different polarization filters. For \(t = 50\) nm, we see that the momentum-space emission in Fig. 2(g) mainly concentrates in four lobes that rotate in a counterclockwise direction, compatibly with the \(B'\) mode of \(q = -2\). For \(t = 120\) nm in Fig. 2(h), there is only a single lobe, with no visible change as the polarization filter is rotated; this mode is the doublet mode \(E'_1\), which has no topological charge. As we further increase the size of the unit cell, for \(t = 170\) nm, in Fig. 2(i) we see two lobes rotating in a counterclockwise direction, compatibly with the doublet \(E'_2\) of \(q = -1\). For even higher \(t = 200\) nm in Fig. 2(j), the momentum-space emission comes from two lobes that rotate in a clockwise direction, as for mode \(A'\) with \(q = +1\). These observations directly correspond to the amplitudes and phases theoretically calculated in Fig. 1(c).

We have employed \(T\)-matrix calculations [39,40] (see also [41]) to explain why there are four regimes in which the lasing mode corresponds to a different IR. We obtain the energy of the modes at the \(\Gamma\) point, by varying \(t\) in the experimentally relevant parameter range, and plot their real and imaginary parts in the complex plane, in Fig. 2(d). We see that the mode energies mostly lie on a linear trajectory. The imaginary part of the energy accounts for the losses, which in our system are radiative and/or Ohmic losses. The modes \(A', B',\) and \(E'_2\) are dark modes that do not radiate to the continuum: for this reason, they only experience Ohmic losses, and the complex energy curve has the same slope for all three. The bright mode \(E'_1\) is subjected to both Ohmic and radiative losses; thus its energy slope for \(t < 150\) nm is steeper than for the three dark modes. However, for \(t > 150\) nm the slope of \(E'_1\) gets smaller, indicating that the mode experiences less radiative losses. In general, all modes’ losses behave differently for small \((t < 100\) nm) and large \((t > 210\) nm) hexamers; for example, at small \(t\), mode \(B'\) seems to have a very small imaginary part, and the same is true for mode \(A'\) for large \(t\) instead. As the modes have different radiative properties when the hexamer size \(t\) is varied, a loss-induced transition between the modes could occur in lasing (see [41] for results also on a quadrumer array). The change of losses for the various modes with different \(t\) is supported by finite element method simulations, where we see that the modes’ electric field intensity distribution in the unit cell changes significantly with changing \(t\), despite the symmetry of the modes being the same [41]. The field hot spots away from the nanoparticles can contribute to how lossy a given mode and hence its quality factor \(Q\), and also its coupling to the gain medium.

From the complex energies of the modes in Fig. 2(d), we calculated the \(Q\) factor of each IR as a function of the hexamer size, see Fig. 2(f). For small hexamers (light green region \(t < 100\) nm), the singlet \(B'\) with \(q = -2\) has the largest \(Q\) factor; the experiment shows lasing from a quasi-BIC of charge \(q = -2\) in the dark green region. For intermediate hexamers (light pink region \(t > 150\) nm), the other doublet \(E'_2\) with \(q = -1\) has the
The largest $Q$ factor in $T$-matrix simulations and the experiment evidences lasing with this topological charge in the dark pink region. Finally, the experiment shows lasing in the $A'$ IR with $q = +1$ for $t \approx 200$ nm, although this mode has a lower $Q$ factor than $E'_2$. We attribute this inconsistency to the different spatial overlap of the mode with the gain medium mentioned above, causing mode competition between $A'$ and $E'_2$ [41].
In an equilibrium topological transition, the topological character of the ground state changes. In our nonequilibrium lasing system, modes of different topological charge coexist with nearly identical energies, but the mutual ordering of their $Q$ factors changes with a structural parameter. Therefore, lower losses (effectively higher gain) can drive a transition of lasing between modes of different topological charge. We have demonstrated lasing in a topologically trivial bright mode and quasi-BIC states of topological charge. We have demonstrated lasing in a topologically trivial bright mode and quasi-BIC states of topological charge. We have demonstrated lasing in a topologically trivial bright mode and quasi-BIC states of topological charge.

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[41] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.129.173901 for a derivation of the topological charges from symmetry arguments; details on the $T$-matrix and finite element simulations; details about the sample fabrication, experimental setup and lasing experiments; Fig. 2(d) without the energy shifts; real space images; topological regions of a quadrumer plasmonic lattice.