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ABSTRACT

In this work, we present the model of plasmonic chiral nanolasers composed of aluminum-coated gallium-nitride (GaN) gammadions, which may lase with a high degree of circular polarization at room temperatures. Using the finite-element method, we examine resonant modes of the four-fold rotationally symmetric cavities of gammadions whose resonant frequencies lie in the gain spectrum of GaN. We find a degenerate doublet of resonant modes which can couple to plane waves in the far-field zone above gammadions. Their near-field profiles exhibit localized distribution in the arms of gammadions and a Fabry-Perot standing-wave pattern along the post. In practice, fabrication imperfections would inevitably spoil the four-fold rotation symmetry of gammadions. Typical perturbation could lift the degeneracy of doublet and leads to mixing of the two degenerate modes which may still output signals with observable handedness above gammadions. Considering a gammadion cavity with a single elongated arm, we show that the magnitude of dissymmetry factor of its resonant mode can be larger than unity. Our calculations are consistent with the experimental results, indicating that the right-handed gammadion cavities lase with a magnitude of dissymmetry factors near 1 at a wavelength of 364 nm. The dimensionless effective mode volume scaled by the cube of effective wavelength is 2.62, reflecting a modal distribution remarkably confined in the plasmonic structures and the capability of enhancing the spontaneous-emission rate noticeably. These chiral nanolasers with an ultrasmall footprint could be potentially utilized as future circularly-polarized photon source at the chip level.

Keywords: Nanolaser, Plasmonics, Circular Dichroism

1. INTRODUCTION

The coherent circularly-polarized (CP) light emission is of great potential in various applications including optical communication and detection, quantum computations, spintronics/valleytronics, and biomolecule characterizations. Coherent CP light sources can be employed for transmitting optical signals if the significant anisotropy of photon detection based on linearly-polarized (LP) waves is present. The insensitivity to anisotropy detection makes the delivery of optical signal more robust. In addition, since the polarization states of photon absorption and emissions corresponding to quantum-mechanical (QM) transitions in most materials are circular polarizations in nature [1-3], the manipulations of QM states in materials would be more precise using coherent light sources with a high degree of circular polarization. This functionality would be very useful for preparations of pure states in advanced usages of optical quantum communication [4, 5] and advanced applications of spintronics/valleytronics [6-15]. The differentiations and assessments of many chiral biochemical compounds through circular dichroism [16-20], which is very important in terms of health and illness, could be magnificently simplified if stable CP light sources are accessible.

The aforementioned usages would require coherent CP optical fields. It is known that typical semiconductor lasers with achiral photonic structures tend to output LP photons rather than CP ones. Hence, compact semiconductor laser sources that directly emit coherent CP photons are nontrivial devices. To incorporate the feature into systems constrained by the reciprocity, the chirality needs to be introduced into active photonic devices in some way. This can be implemented at the material level like, for example, the incorporation of cholesteric liquid crystal as host materials [21-24], or at the structural level such as the fabrication of chiral photonic crystals [25-29] and three-dimensional (3D)
helical structures [30-33]. Among various chiral nanostructures, gammadions are widely utilized in plasmonics or metastructures which play a key role in CP light-emitting devices [34-36]. In fact, with the inherent chirality and compactness resulted from the field confinement of metals, plasmonic gammadion nanocavities are promising candidates for the future development of CP light sources with both nanoscale footprints and high degrees of circular polarization.

The schematic diagram of proposed aluminum-coated (Al-coated) gallium-nitride (GaN) gammadion structures is shown in Fig. 1. The chiral structures are covered with metallic layer to confine the energy inside the cavity. In addition, aluminum is chosen for metal deposition due to its prominent plasmonic characteristics at UV wavelengths. The feature sizes of a single gammadion including the linewidth, length, arm length, and height are 50, 300, 200, and 500 nm, respectively. The gammadions are arranged in square lattice with a period of 500 nm. The metallic layer has a thickness of 50 nm. The gammadions whose arms bend counterclockwise are defined as the right-hand (R-) chiral structure, as indicated in the inset of Fig. 1, and the counterpart with arms bending in the opposite way is defined as the left-hand (L-) chiral structure. We note that the upper and lower ends of gammadion structures are in contact with different materials. This asymmetry makes gammadion metallic cavities intrinsically chiral. In this study, we theoretically evaluate the dissymmetry factors of emitted light from these chiral structures and compare them with experimentally data.

2. THEORETICAL MODEL FOR MODES OF GAMMADIONS

We first investigate the generic modes in these plasmonic chiral cavities. When we observe these ideal gammadions arranged in square lattice along the direction of material growth (denoted as z-axis as shown in Fig. 1), the symmetry group of whole structural contains the 4-fold rotation symmetry. Since the metal coating magnificently reduce the overlap between the modal fields associated with each gammadion, the role of primitive translations (lattice) could be excluded from the investigation tentatively. A single gammadion holds the symmetry of point group C4. We could therefore use the group theory to classify the cavity modes through the modal profiles \( \mathbf{E}(\mathbf{r}) \) according to the irreducible representations of the group. The rotation \( \mathbf{R}_{\pi/2} \) by \( \pi/2 \) around the z axis is one of the four group elements in the point group C4. The characters \( \chi \) of this rotation in the irreducible matrix representations of group C4 may take values \( i^n \) (\( n = 0 \) to 3). Eigenmodes with character \( \chi = \pm 1 \) appear as the combination of spherical-like waves whose fields exhibits azimuthal dependencies \( \exp(\text{i}m\phi) \) with \( m = 4n' + 4n'' + 2 \) (\( n' \in \mathbb{Z}' \)). These modes exhibit a vanishing field for an observer right above the cavity in the far-field region \( (k_e \rightarrow \infty) \). On the other hand, CP waves in the far-field zone with polarizations \( \mathbf{e}_x = (\hat{x} \mp i\hat{y})/\sqrt{2} \) originate from the eigenmodes with \( \chi = \pm i \) (\( n = 0 \) and 3). In other words, for the detection of robust and coherent CP radiation, the lasing modes shall be those with imaginary characters.

The experimental dissymmetry factor \( g_e \) which quantifies the degree of chiral dichroism of radiation from the plasmonic gammadion cavities can be expressed as

\[
g_e = 2 \frac{I_L - I_R}{I_L + I_R}. \tag{1}\n\]
The radiation with perfect circular polarizations would show a dissymmetry factor with maximal modulus $|g_{\text{e}}| = 2$. The positive (negative) dissymmetry factor $g_{\text{e}}$ of the optical output from an active photonic device corresponds to the LCP-like (RCP-like) radiation. The two degenerate CP-like modes may have an identical probability to be selected out during the lasing action, but in terms of far fields, they still show distinct amplitudes and leads to observable circular dichroism.

To clear the ambiguity, we start with the modal profiles of the LCP-like and RCP-like modes $E_{\text{L}}(r)$ and $E_{\text{R}}(r)$, respectively, whose electric energies in the active region (posts of GaN gammadions) are set equal for fair comparisons (proper normalization of modal profiles). Under such circumstances, the far fields of $[E_{\text{L}}(r), E_{\text{R}}(r)]$ asymptotically behave as $[a_L \hat{e}_-, a_R \hat{e}_+] \exp(ik_0 z)/(k_0 z)$ at the top of gammadions, where $a_L$ and $a_R$ are amplitudes of the LCP- and RCP-like spherical waves. The important characteristic introduced by the chirality is that the two far-field magnitudes $|a_L|$ and $|a_R|$ may be distinct even though the two CP-like modes have identical electric energies stored in the active region. Therefore, even if both of the modes start to lase with identical strengths in an ideal gammadion, the overall far field still show a nonvanishing dissymmetry factor $g_{\text{e}}^{(4f)}$ as follows:

$$g_{\text{e}}^{(4f)} = 2 \frac{|a_L|^2 - |a_R|^2}{|a_L|^2 + |a_R|^2},$$  

where the superscript “4f” represents an ideal “4-fold” rotationally symmetric cavity. In practice, a typical gammadion in experiment is hardly 4-fold rotationally symmetric. Any variations of the relative permittivity tensor $\Delta \tilde{\varepsilon}(r, \omega)$ that break the 4-fold rotation symmetry at a frequency $\omega$ would lift the degeneracy of two CP-like modes. However, let us assume that the reciprocity is unbroken $[\Delta \tilde{\varepsilon}(r, \omega) = \Delta \tilde{\varepsilon}(r, \omega)]$, and the permittivity variation $\Delta \tilde{\varepsilon}(r, \omega)$ influences the two CP-like modes in a similar fashion, which is typically what happens for uncontrolled perturbations. In this way, to the lowest order, the two modal fields $E_{\text{L}}(r)$ and $E_{\text{R}}(r)$ would be linearly combined into those $E_1(r)$ and $E_2(r)$ of perturbed modes with close weights:

$$E_{(1,2)}(r) \approx \frac{1}{\sqrt{2}} [E_{\text{R}}(r) \pm e^{i\Theta} E_{\text{L}}(r)] \frac{\exp(ik_0 z \pm \infty)}{\sqrt{2} k_0 z} \left[a_R \hat{e}_+ \pm e^{i\Theta} a_L \hat{e}_-\right],$$

where $\Theta$ is a phase which depends on the perturbation; and $k_{0,1}$ and $k_{0,2}$ are propagation constants of the two perturbed modes in vacuum. Since the two new modes turn nondegenerate, one of them would become more favorable than the other does through the amplification in the cavity. Still, regardless of which mode lases, the far-field dissymmetry factor in the would be close to $g_{\text{e}}^{(4f)}$ in the regime of mild perturbations, as manifested by the far-field weights of LCP and RCP radiations of $E_1(r)$ and $E_2(r)$ in Eq. (3).

Through the finite-element method (FEM), we obtain the modal electric field $E(r)$ corresponding to our target mods of the gammadion. At the Brillouin zone center of square lattice, the field $E(r)$ satisfies periodic boundary conditions along the two directions (denoted as $x$ and $y$), and therefore it may be decomposed into a two dimensional Fourier series. Since we are interested in the far-field on the top of gammadions, we may also expand it as with the three-dimensional propagating waves and evanescent waves which are compatible to the two-dimensional Fourier series. More specifically, the modal field $E(r)$ at a vertical position $z$ above the gammadion surface is written as

$$E(r) = \sum_{u,v=-\infty}^{\infty} E_{uv} e^{i(k_{x,uv} x + k_{y,uv} y + k_{z,uv} z)} e^{i \Theta},$$

$$k_{x,uv} = \frac{2\pi u}{p}, \quad k_{y,uv} = \frac{2\pi v}{p}, \quad k_{z,uv} = \sqrt{k_0^2 - k_x^2 - k_y^2},$$

where $E_{uv}$ is the Fourier field components labeled by integer indices $u$ and $v$; $k_{x,uv}$, $k_{y,uv}$, and $k_{z,uv}$ are the Fourier wavenumbers corresponding to $u$ and $v$ in the $x$, $y$, and $z$ directions, respectively; $p$ is the period of the square lattice;
and $k_0 = 2\pi/\lambda$ is the free-space propagation constant ($\lambda$ is the resonant wavelength of the mode). In Eq. (4), if the wavenumber $k_{zuv}$ is real, the related Fourier component represents a plane wave propagating away into the free space on the top of gammadions. In contrast, an imaginary $k_{zuv}$ corresponds to an evanescent wave. In the experiment, the period $p$ is 500 nm while the wavelength $\lambda$ is around 364 nm. Since the lens in experiment only collected optical rays within a receiving angle of $30^\circ$ (NA = 0.5), the off-axis radiation corresponding to Fourier components other than that at $(u, v) = (0, 0)$ need not be considered.

In views of the aforementioned arguments, we only need the Fourier component $E_{00}$ at $(u, v) = (0, 0)$ from the numerical results of modal field $E(r)$. To this end, the orthogonality of Fourier components along the $xy$ plane could be applied. At a certain position $z$ above the gammadions, we may perform the numerical surface integral to obtain:

$$
\frac{1}{p^2} \int_{-p/2}^{p/2} \int_{-p/2}^{p/2} \, dx \, dy \, E(r) = E_{00} e^{ik_0 z} = e^{ik_0 z}(E_{00,R} \hat{e}_+ + E_{00,L} \hat{e}_-),
$$

where $E_{00,(RL)}$ are the components corresponding to $\hat{e}_\pm$. The theoretical dissymmetry factor $g_e$ is then estimated as

$$
g_e = \frac{|E_{00,L}|^2 - |E_{00,R}|^2}{|E_{00,L}|^2 + |E_{00,R}|^2}.
$$

### 3. RESULTS AND COMPARISONS

To understand the effect of chirality on the coherent radiation from gammadion metal cavities, we use FEM to find the potential CP-like lasing modes with characters $\chi = \pm i$ around the wavelength of 364 nm. Figures 2(a) and 2(b) show the intensity profiles $|E(r)|^2$ of the candidate RCP- and LCP-like cavity modes on a horizontal cross-section in an R-gammadion cavity. These cavity modes are Fabry-Perot resonances of specific guided modes in the metal-covered gammadion waveguides along the $z$ direction. The horizontal profiles in Fig. 2(a) and (b) are just taken at the antinode near one-quarter of the cavity height. Under the protection of 4-fold rotation symmetry, the CP-like cavity modes are degenerate. The LCP-like and RCP-like cavity modes show similar intensity profiles. Both modes exhibit bright spots in each arm and the dimmer one in the center. Additionally, the 4-fold rotation symmetry only ensures that the transverse fields of the CP-like modes to be circularly-polarized at the center of each horizontal cross-section in the gammadion.
The perfect CP radiation of these modes is due to the far-field interference of local fields on the nearby arms of gammadion, whose polarizations and amplitudes are rotated consecutively by \( \pi/2 \) in the real space and shifted by \( \pm \pi/2 \) in phase, respectively.

The standing-wave patterns of the CP-like modes which pass through the yellow dashed line in the inset of Fig.1 are shown in Figs. 3(a) and 3(b), respectively. The cavity modes have two antinodes inside the arms of cavity, and their intensities are relatively weak near the top and bottom of gammadion structures. At the upper ends, modal fields encounter strong out-of-phase reflections due to the metal, while at the lower ends, the mismatch of transverse modal profile also leads to significant reflection. In addition to the CP-like modes shown in Figs. 2 and Figs. 3, other cavity modes might be present in the gain window of GaN spectrum as well. The target lasing cavity mode should not only be able to output field in the CP state (CP-like modes) but also overcome its threshold at the smaller material gain in the presence of optical pumping. Keeping those points in mind, we evaluate the complex eigen-frequencies \( \omega_r \) of cavity modes at various gain levels. In the convention of temporal dependency \( \exp(-i\omega_r t) \), as we introduce the more negative imaginary part \( \kappa_o \) of refractive index of GaN inside the gammadion structure, the more gain is brought into the active region, and the imaginary parts \(-\text{Im}[\omega_r] \) of resonance frequencies with a sign flip should become lower. At \( \text{Im}[\omega_r] = 0 \), the magnitude \( \kappa_o \) is proportional to the threshold gain of the cavity mode. The corresponding real parts \( \text{Re}[\omega_r] \) of resonance frequency are then the resonant frequencies.

As shown in Fig. 4, we show the ratios \( \text{Im}[\omega_r]/\text{Re}[\omega_r] \) of the five modes around the lasing wavelength of 364 nm versus their resonance wavelengths under different gains [(\( \kappa_o \))]. For each mode at threshold (\( \text{Im}[\omega_r] = 0 \)), respectively from the shorter to longer wavelengths, \( |\kappa_o| \) is \( 8.21 \times 10^{-2}, 7.91 \times 10^{-2}, 8.06 \times 10^{-2}, 8.09 \times 10^{-2}, \) and \( 1.03 \times 10^{-1} \). Experimentally, the material gain of GaN dropped rapidly at the short-wavelength side of 364 nm, leading to a narrow gain window. The modes presented in Fig. 4 and 5 are the only CP-like ones in the window, and they reach the threshold at \( |\kappa_o| = 8.09 \times 10^{-2} \) around a wavelength of 362.8 nm. All other modes have real characters \( c = \pm 1 \) and are not circularly-polarized on the top of gammadion array. The ones with resonance wavelengths shorter than 362.8 nm could have the lower thresholds than those of CP-like modes, but they are not in the gain window. On the other hand, the mode at the long-wavelength side (366 nm) may lie in the gain window, but its threshold \( |\kappa_o| = 0.103 \) is much higher than that of CP-like modes, rendering it less competitive. With these analyses, the lasing signal in the experiment should originate from the CP-like modes (or their linear combinations) in gammadion metal cavities.

The experimental circular dichroism was measured from emission of the gammadions which did not have the perfect 4-fold rotation symmetry. Knowing little about the details of perturbation in experiment, we could still estimate the dissymmetry factors since typical perturbations should affect the two degenerate CP-like modes evenly and mix them with similar weights. In views of this, we intentionally lengthen one of the arms of ideal gammadions by 3% to break the 4-fold rotation symmetry. Shown in Fig. 5 are the horizontal field distributions of two perturbed modes (at the antinode) which have close connections to the two CP-like modes in ideal 4-fold rotationally symmetric gammadion metal cavities.
Figure 4. The ratios $-\text{Im}[\omega_x]/\text{Re}[\omega_x]$ versus the resonance wavelengths for various modes under different imaginary parts $\kappa_x$ of the refractive index of GaN in the gammadiion structure. The corresponding $|\kappa_x|$ at thresholds, from the short- to long-wavelength sides, are $8.21 \times 10^{-2}$, $7.91 \times 10^{-2}$, $8.06 \times 10^{-2}$, $8.09 \times 10^{-2}$, and $1.03 \times 10^{-1}$, respectively. The ratios of potential CP-like lasing mode (362.8 nm) are marked with red circles. All other modes marked with blue hollow circles have characters $\chi = \pm 1$.

Figure 5. The horizontal field profiles $|E(r)|^2$ of the (a) $y$- and (b) $x$-like modes at the antinode corresponding to a perturbed R-gammadion metal cavity. The outer arm indicated by white dashed lines is prolonged by 3% for breaking the 4-fold rotation symmetry. While the $y$-like mode is mostly an equal-weight combination of two CP-like modes shown in Figs. 2 and 3, the $x$-like one is significantly mixed with other modes with $\chi = \pm 1$.

One of the perturbed modes may be simply regarded as the linear combination of two CP-like modes, as indicated by the profile of $y$-like mode in Fig. 5(a). The profile of another perturbed mode exhibits a bright spot in one of the 4 outer arms, as shown in Fig. 5(b). The $x$-like mode could be a result of the high-order perturbation which mixes it with other modes carrying real characters ($\chi = \pm 1$). Using Eq. (6), we further calculated the dissymmetry factors from the far fields of the $y$-like and $x$-like modes, which are 0.92 and 0.8, respectively. The $y$-like mode shows the more significantly CP-like feature than $x$-like mode does because it is less affected by modes with $\chi = \pm 1$. Experimental dissymmetry factor of the lasing signal form R-gammadion is 1, close to the counterpart from the $y$-like mode of the imperfect gammadiion considered here.

The local photon density of states in the GaN gammadiion could be modified remarkably by the plasmonic structure, which would increase the spontaneous emission rate of electron-hole pairs therein. The metal-coated gammadiion would
also alter the experimentally measurable carrier lifetime. The enhanced transition rate partly originated from the small modal volumes $V$ of resonant modes in the plasmonic cavity. We may take the LCP-like mode in an ideal R-gammadion metal cavity as an example and calculate its dimensionless effective volume $V_{\text{eff}}$ as follows:

$$V = V_{\text{eff}} \left( \frac{\lambda}{n} \right)^3 = \frac{\int d\mathbf{r} \varepsilon_{r,g}(\mathbf{r}, \omega_r) |\mathbf{E}^{(r)}|^2}{\max[\varepsilon_{r,g}(\mathbf{r}, \omega_r) |\mathbf{E}^{(r)}|^2]}$$

where $\lambda$ is the wavelength of the mode; $n$ is the real part of refractive index of GaN; $\mathbf{E}^{(r)}$ is the corresponding electric field; and $\varepsilon_{r,g}(\mathbf{r}, \omega_r) = \partial \Re[\omega \mathbf{e}_r(\mathbf{r}, \omega)]/\partial \omega |_{\omega=\omega_r}$ is the relative group permittivity at position $\mathbf{r}$ and (real) resonance frequency $\omega_r$ of the mode at threshold. At resonance wavelength $\lambda = 362.8$ nm and a refractive index $n = 2.53$, the dimensionless effective volume $V_{\text{eff}}$ of this mode is approximately 2.62. This effective volume should be sufficiently small to enhance the spontaneous-emission rate. This could explain the shortening of the carrier lifetime from 300 to 130 ps due to the presence of metal-coated gammadion in the experiment.

4. SUMMARY
We present the model of ultraviolet plasmonic chiral nanolasers composed of Al-coated GaN gammadions which lase with a high degree of circular polarization at room temperature. We investigate the resonant cavity modes and show that the lasing mode can output far fields with significant degrees of circular polarization even if the perturbation may break the 4-fold rotation symmetry of gammadions. Our result is consistent with the experimental fact that the gammadion cavities can lase with a high dissymmetry factor around unity at room temperature. These chiral nanolasers have ultrasmall footprint and could be used as a potential platform for advanced applications of CP photons or utilized in the future photonic integrated systems at the chip level.

REFERENCES


