Lasing Emission from Plasmonic Nanodome Arrays

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Abstract: We demonstrate a new feedback mechanism to achieve nano-lasing using plasmonic nano-dome array (PANDA) substrates. The unique off-normal lasing phenomenon in the PANDA lasers provides useful applications which cannot be implemented by other plasmonic lasers.

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Surface plasmon-polariton (SPP) and localized surface plasmon (LSP) modes supported by metallic nanostructures offer remarkable opportunities for exploration of light-matter interactions at a scale size well below the diffraction limit and for the development of nanoscale lasers. Plasmonic lasing from periodic arrays of metallic nanostructures occurs at a band-edge plasmon mode with directional emission normal to the device surface [1-2]. However, further detailed study of the lasing action resulting from non-band-edge plasmon modes with directional but nonvertical emission has not been extensively reported. We recently developed a plasmonic nano-dome array (PANDA), which supports both LSP and SPP Bloch wave (SPP-BW) modes [3-4]. The investigation of how interactions between LSP and SPP-BW modes with the gain material enables lasing action would advance our fundamental knowledge of plasmonic lasers. In this work, we experimentally and numerically demonstrate a new feedback mechanism to achieve nano-lasing using a PANDA laser comprised of a dye-doped solution as the gain medium and the PANDA structure as the oscillator for lasing in the near infrared region. The key emission features, including lasing wavelength, linewidth, threshold, beam divergence and radiation angle, are experimentally characterized.

Fig. 1(a) shows a schematic illustration of the PANDA laser comprised of a 2D square lattice of gold-coated nanodome structures, a liquid gain material, and a cover glass window. The organic dye solution was excited by ns-laser pulses at an incident angle (θl) while emission from the PANDA laser is measured at a detection angle (θd). Fig. 1(b) shows a cross-sectional diagram for one period of the PANDA laser, where photons emitted by the photoexcited dye molecules residing in the vicinity of PANDA surface couple to the hybridized plasmon (HP) mode. Above the lasing threshold, narrowband directional emission emerges when the plasmonic losses are overcome by the optical gain. Scanning electron microscopy (SEM) images in Fig. 1(c) and (d) show the top-view surface profiles of the fabricated PANDA structures with period of A = 450 and A = 400 nm, respectively.

The extinction spectrum measured at normal incidence for p-polarized illumination is shown as the red curve in Fig. 2(a), which matches well with the FDTD-computed spectrum. The modes associated with the indicated plasmon resonances are assigned to the SPP-BW mode with a narrow linewidth at λ_{SPP-BW} = 625 nm and the dipole LSP mode with a broad feature at λ_{LSP} = 931 nm. Fig. 2(b) and (c) show the spatial distribution of the electric-field intensity enhancement for the SPP-BW and the LSP modes, respectively. The calculated dispersion of SPP-BW modes is depicted as the orange/yellow curves of Fig. 2(d). As the normalized wave vector increases, the (-1,0) SPP-BW mode spectrally overlaps with the broad LSP band. The dispersion relation of two strongly coupled modes shows an anti-crossing behavior (black dashed curves) where new hybridized plasmon (HP) modes form as a result of hybridization of the LSP and SPP-BW modes. The photoluminescence (PL) band of the laser dye spectrally overlaps the upper energy branch of the HP modes, represented by the upper black dashed curve in Fig. 2(d), with a decreased group velocity as the in-plane wave vector k|| increases, allowing photons emitted by the photoexcited dye molecules to couple into the upper HP mode via the near-field interaction. Laser oscillation occurs once the optical gain provided by the gain medium can balance the material and radiation loss of the HP mode.

Fig. 3(a) shows typical emission spectra of the PANDA laser (A = 450 nm; LDS 867) recorded at θd = 50° for emission parallel (red curve) and perpendicular (blue curve) to the plane of incidence. When the PANDA laser was optically pumped with a fluence of 0.89 mJ/cm², a narrow and intense laser oscillation occurred at λ_{lasing} = 907 nm with a FWHM of 2.73 nm, compared with the PL spectrum of LDS 867 (green curve). Fig. 3(b) shows the recorded emission spectra as a function of pump fluence. The inset of Fig. 3(b) summarizes peak intensity at λ_{lasing} = 907 nm as a function of pump fluence. Fig. 4(a) shows a typical emission spectrum of the PANDA laser (A = 400 nm; LDS 765) measured under a pump fluence of 0.63 mJ/cm² at θd = 50°. The lasing emission peak locates at λ_{lasing} = 807 nm with a FWHM of 2.53 nm, compared with the PL spectrum of LDS 765. The evolution of the emission spectra as a function of pump fluence is shown in Fig. 4(b).

The lasing emission is strongly angular-dependent. To investigate the correlation, angle-resolved emission measurements were carried out to study the radiation pattern and the corresponding results are shown in Fig. 5(a) and (b) for the PANDA lasers with A = 450 and 400 nm, respectively. Fig. 5(c) plots the lasing emission intensities as a function of θd. The divergence angle (∆θd) of the lasing beam is 1.46° measured on the plane parallel to the high symmetry direction. Fig. 5(d) shows the simulated angular distribution of far-field emission of the PANDA laser (A = 450 nm) at λ = 905 nm for randomly
placed and oriented dipoles. Strong emission can be clearly seen at θ = 50° with the azimuthal angle of ϕ = 0°, 90°, 180° and 270°, respectively. The simulated result is in good agreement with the measurements, as shown in Fig. 5(c). We believe that the unique off-normal lasing phenomenon in the PANDA lasers provides useful applications which cannot be implemented by other plasmonic lasers.

Fig. 1. PANDA laser. (a) Schematic of the 2D square lattice of the PANDA substrate. Green and red arrows represent the 532 nm pump beam at the incident angle (θi) and lasing emission at the detection angles (θd), respectively. (b) Schematic of the lasing action for one unit cell of the PANDA laser. (c) and (d) Top-view SEM images of the fabricated PANDA surfaces with period of (c) λ = 450 nm and (d) λ = 400 nm on a flexible plastic substrate. Scale bars = 500 nm.

Fig. 2. Optical characteristics of the PANDA laser (λ = 450 nm; LDS 867 doped methanol). (a) Comparison of emission spectra of the PANDA laser and PL of LDS 867 doped methanol. H (red) and V (blue) respectively represent emission polarized parallel and perpendicular to the lattice direction (Γ–X) of the PANDA laser. (b) Evolution of lasing emission spectra for different pump fluences and emission polarizations. Inset: peak intensity as a function of detection angle (θd) for λ = 450 nm. (c) The angular emission profiles for PL band and LSP mode. Detection angle, θd (deg).

Fig. 3. Lasing characteristics of the PANDA laser with λ = 450 nm. (a) Comparison of emission spectra of the PANDA laser and PL of LDS 867 doped methanol. H (red) and V (blue) respectively represent emission polarized parallel and perpendicular to the lattice direction (Γ–X) of the PANDA laser. (b) Evolution of lasing emission spectra for different pump fluences and emission polarizations. Inset: peak intensity as a function of detection angle (θd) for λ = 450 nm. (c) The angular emission profiles for PL band and LSP mode. Detection angle, θd (deg).

Fig. 4. Lasing characteristics of the PANDA laser with λ = 400 nm. (a) Comparison of emission spectrum of the PANDA laser and PL of LDS 765 doped methanol. V (blue) represents emission polarized perpendicular to the lattice direction (Γ–X) of the PANDA laser. (b) Evolution of lasing emission spectra for different pump fluences. Inset: peak intensity as a function of detection angle (θd) for λ = 450 nm.

Fig. 5. Angular dependence of the lasing emission. Measured angular emission spectra of (a) the PANDA laser (λ = 450 nm; LDS 867 doped methanol) and (b) the PANDA laser (λ = 400 nm; LDS 765 doped methanol) as a function of detection angle (θd). Each slice on the vertical axis represents the s-polarized emission spectrum measured at a particular θd by optically pumping the PANDA laser at a fixed incident angle, θi = 0°. (c) The angular emission profiles for the spectral wavelength of λSPP = 907 nm in (a) and λLSP = 807 nm in (b). (d) Simulated angular distribution of far-field emission of the PANDA laser (λ = 450 nm) at θi = 905 nm. The black characters represent the polar angle (θ) and the white the azimuthal angle (ϕ).

References