Electronic Supporting Information

Plasmon-exciton Coupling Dynamics and Plasmonic Lasing in Core-shell Nanocavity

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The characteristics of modes

A_m is the effective mode area, L_p is the propagation length, and Γ_{wg} is the waveguide confinement factor[1-5]

$$A_m = \iint W(r) d^2 r / \max\{W_m(r)\} \quad (1)$$

$$L_p = 1/\left[2\,\mathrm{Im}(k_z)\right] \quad (2)$$

$$\Gamma_{wg} = (n_a / 2\eta_0) \int_{A_a} \left|E(\rho)\right|^2 d\rho / P_z \quad (3)$$

Where the denominator and numerator of A_m are the ratio of the total mode energy $W(r)$ and its peak energy density $\max\{W_m(r)\}$, respectively. k_z is the imaginary part of the mode propagation constant; $E(\rho)$ is the electric field expressed in cylindrical coordinates; P_z is the power flow in the propagation direction; n_a is the refractive index of the gain medium; A_a is the region of the gain medium; and η_0 is the intrinsic impedance. Figure. S1(a),(b) show the characteristics (propagation distance, waveguide confinement factor) of modes in Fig.1(d)-(g) as functions of the diameter of ZnO nanorod.

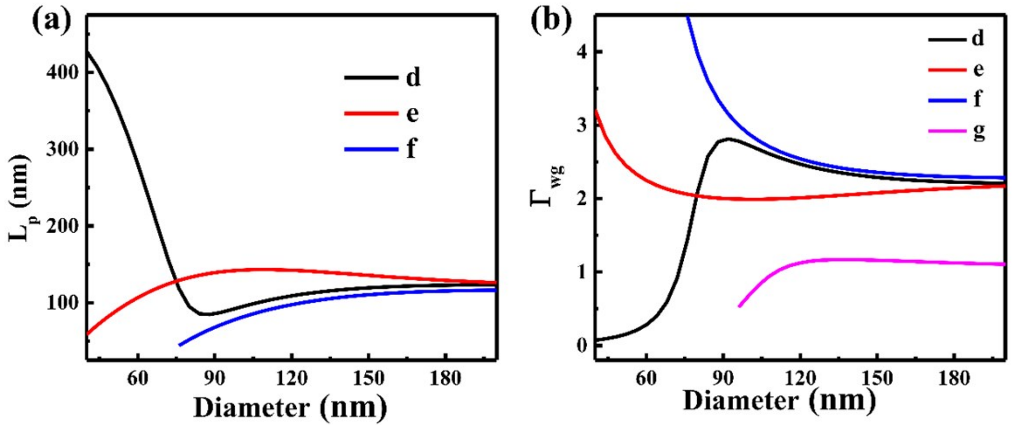

Figure S1 When the Al thickness is 15 nm, the characteristics of modes as functions of the diameter of ZnO nanorod. (a)The propagation length. (b) The waveguide confinement factor.

The ZnO/Al core-shell structure of different Al thickness

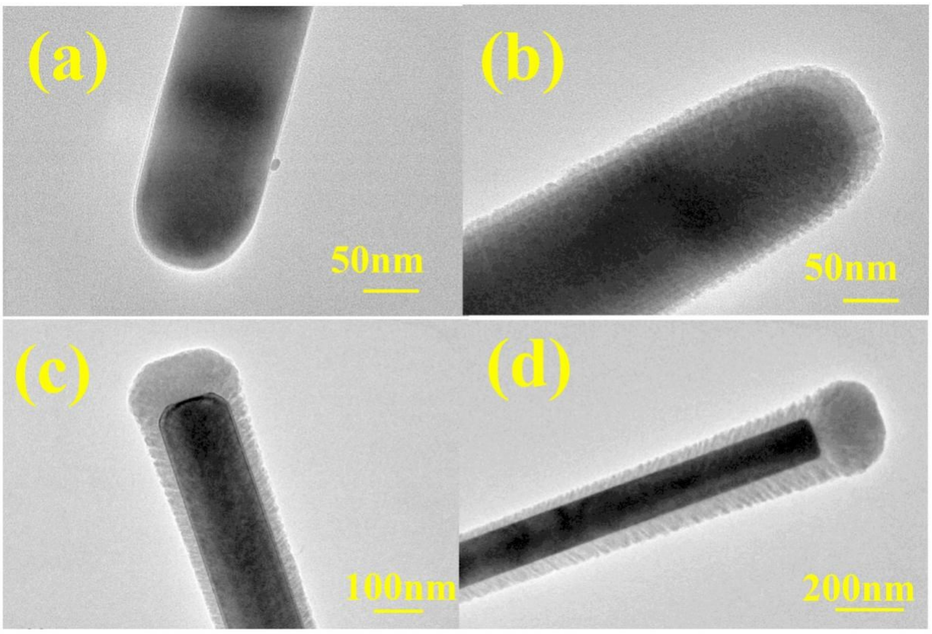

Figure S2 TEM image of ZnO/Al core-shell plasmonic nanolaser with sputtering Al layer time of ~5 min, ~10 min, ~15min, ~20 min. (a)-(d)ZnO/Al core-shell plasmonic nanolaser for different Al layer thickness of ~5 nm, ~15 nm, ~30 nm, ~50 nm, respectively.

The threshold material gain and the Purcell factor

The threshold material gain g_{th} can be related to the photon lifetime τ_p by Formula (4). The Q of the cavity can be theoretically calculated by the following Formula[1,3-5].

$$\frac{1}{\tau_p} = v(\omega)\Gamma_{wg} g_{th} = v(\omega)\left[\alpha + \frac{1}{2L}\ln\left(\frac{1}{R_1R_2}\right)\right] \tag{4}$$

$$\frac{1}{Q} = \frac{1}{\tau_p\omega} = \frac{v(\omega)}{\omega}\left[\alpha + \frac{1}{L}\ln\left(\frac{1}{R_1R_2}\right)\right] = \frac{1}{Q_{abs}} + \frac{1}{Q_{mir}} \tag{5}$$

$$\frac{1}{Q_{abs}} = \frac{v(\omega)\alpha}{\omega} \tag{6}$$

$$\frac{1}{Q_{mir}} = \frac{v(\omega)}{\omega}\frac{1}{2L}\ln\left(\frac{1}{R_1R_2}\right) \tag{7}$$

Where v(ω) is close to the material group velocity of the semiconductor material, which is related to frequency ω. L is c-axial length of nanorods and R_1, R_2 are two end face reflectivity of the ZnO/Al core-shell structure, respectively. Q_{abs} is an inverse relationship with modal loss α from the Formula (6). So Q_{abs} increases with the increase of Al thickness in Figure. S3(b) black curve. Q_{mir} is an inverse relationship with mirror loss in the Formula (7). As shown in Figure. S3(b) blue curve, the Q_{mir} increase with the increase of the Al thickness. Figure. S3(a) is effective mode area in different Al thickness. The effective mode area decrease corresponding to the increase of Al thickness. it presents that the thicker Al layer can enhance optical confinement. Thus, the Purcell factor F_p and exciton recombination rate increases with the increase of the Al thickness.

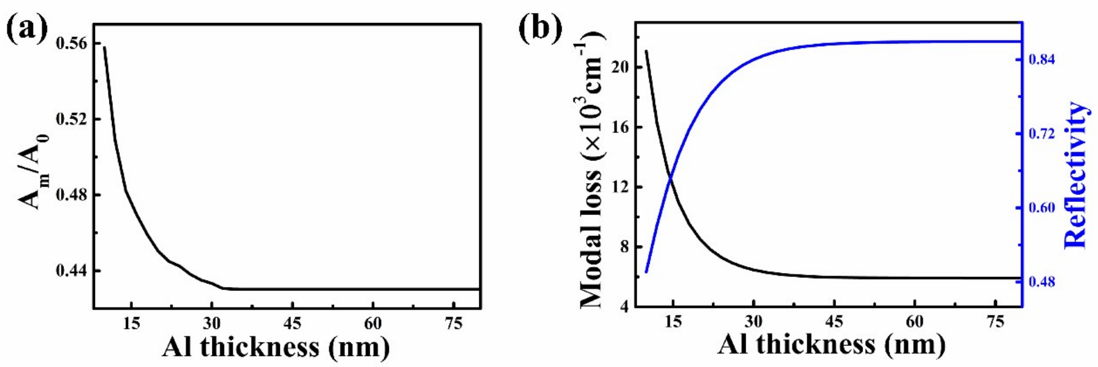

Figure S3 When the diameter of ZnO nanorod is 200 nm, the dependence of mode characteristics and different Al thickness. (a) The effective mode area. (b) The modal loss (blue line), the reflectivity of 378nm emitting light (black line).

Temperature-dependent PL spectra

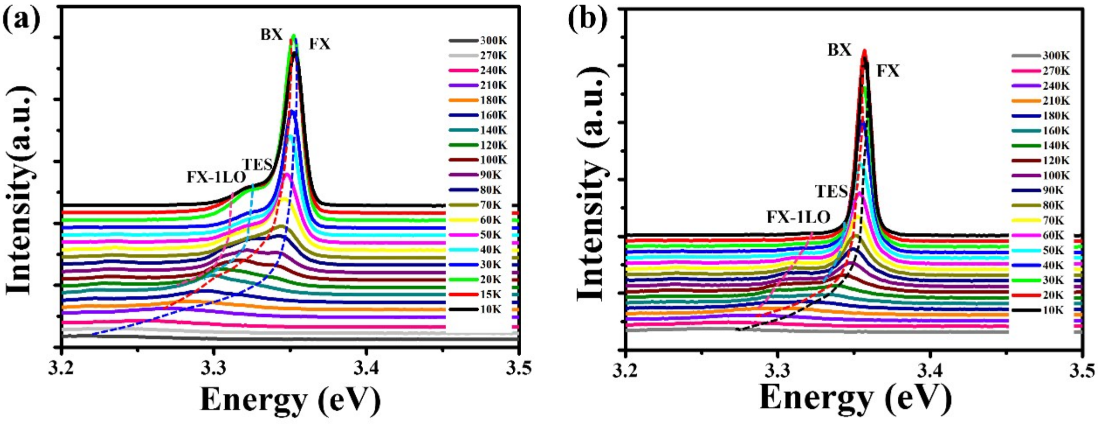

Figure S4 Temperature-dependent PL spectra. (a) ZnO photonic structure and (b) the ZnO/Al core-shell plasmonic structure.

The lasing spectra

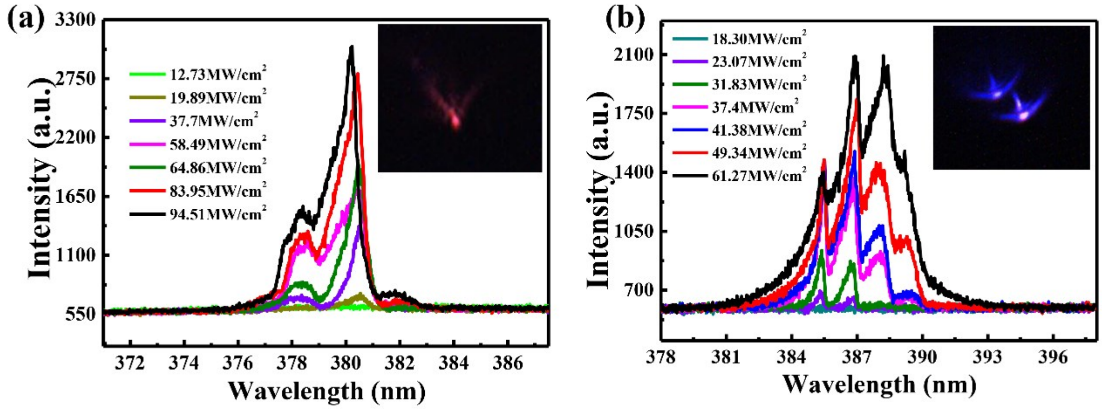

Figure S5 The lasing spectra under different pumping power density. (a) the ZnO/Al core-shell plasmonic nanolaser. (b) ZnO photonic nanolaser. The inset on the right correspond to microscope images of nanolasers, where the scattered light output is from the end facets.

Rate equation analysis

The rate equations are[5]:

$$\frac{dn}{dt} = p - An - \beta\Gamma As(n - n_0) \tag{8}$$

$$\frac{ds}{dt} = \beta An + \beta\Gamma As\left(n - n_0\right) - \gamma s \tag{9}$$

Here, p is a pump rate, s is photon number of emitted laser mode, n is excited electronic state population, n_0 is the excited state population at transparency. β is the spontaneous emission factor. γ is the total cavity mode loss rate and Γ is the overlap factor quantifying the spatial distribution of gain relative to the laser mode. $\gamma_g = \beta\Gamma A s n_0$ is the absorption rate due to the gain medium, A is spontaneous emission rate.

From the rate equations in steady state, the photon number is described by the following equation.

$$\gamma s = \frac{1}{2}\left(p - p_{ph}^{(n)}\right) + \frac{1}{2}\left(p + p_{ph}^{(n)}\right)\left[1 - \frac{4}{p}\frac{(1-\beta)An_\infty}{\left(1 + p_{ph}^{(n)}/p\right)^2}\right]^{1/2} \tag{10}$$

Here, $p_{th}^{(n)} = (\gamma + (1-\beta)\gamma_g)/\beta\Gamma$, $n_\infty = (\gamma + \gamma_g)/\beta\Gamma A$.

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