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1 **Infrared light-emitting devices from antenna-coupled**
2 **Luttinger liquid plasmons in carbon nanotubes**

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13
14 Electrically driven light-emitting devices provide highly energy-efficient lighting at visible
15 wavelengths, and they have transformed photonic and electronic lighting applications. Efficient
16 infrared light-emitting devices, however, have been challenging because bandgap emission
17 from semiconductors becomes inefficient in the mid- to far-infrared spectral range. Here we
18 investigate infrared light-emitting devices (IRLEDs) based on Luttinger liquid (LL) plasmons
19 in one-dimensional (1D) metallic carbon nanotubes. Elementary excitations in LL are
20 characterized by collective charge and spin excitations, *i.e.* plasmons and spinons.
21 Consequently, electrons injected into the nanotubes transform efficiently into LL plasmons, a
22 hybrid excitation of electromagnetic fields and electrons. We design nanoantennas coupled to
23 the carbon nanotube to radiate LL plasmons into the far-field. LL-based IRLEDs can be
24 designed to selectively emit at wavelengths across the far- and mid-infrared spectra. An
25 electrical-to-optical power conversion efficiency up to 3.2% may be achieved. Such efficient
26 and narrowband LL-based IRLEDs can enable novel infrared nanophotonic applications.

27
28 Infrared light sources are important for a wide range of applications ranging from
29 spectroscopy and molecular sensing to material processing because the infrared radiation
30 interacts strongly with molecular vibration modes [1,2]. Efficient solid-state lighting, however,
31 has been limited in the infrared spectral range, unlike its visible wavelength counterparts [1].
32 Solid-state infrared light-emitting devices (IRLEDs) have been explored using narrow-gap
33 semiconducting materials such as III-V, II-VI, and lead-salt semiconductor alloys [1,3,4], but
34 their bandgap emissions suffer from low quantum efficiency (<1%) because the non-radiative
35 Auger process dominates the electron-hole recombination in narrow-gap semiconductors [5–
36 10].

37 Rather than using narrow-gap semiconductors, we explore here IRLEDs based on a Luttinger
38 liquid (LL), a strongly correlated electronic matter confined in one-dimensional (1D) metals
39 with peculiar properties such as the spin-charge separation and the power-law decay of the

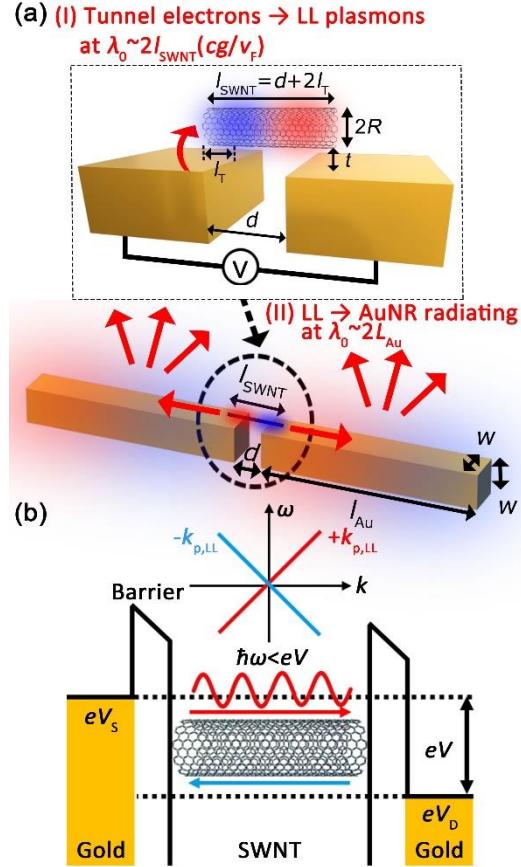


FIG. 1. (a) IRLED structure composed of SWNT and AuNR (l_{Au} : a length of AuNR, w : width of AuNR, d : AuNR gap size, t : a tunneling gap between SWNT and AuNR, l_T : an overlap length of SWNT and AuNR defining $l_{\text{SWNT}}=d+2l_T$). SWNT supports the Luttinger liquid (LL). Inset shows the dashed circle area near the AuNR gap. (b) A two-terminal geometry with the bias voltage $V=V_S-V_D$ and the linear LL dispersion (inset). The injected electrons are converted into the LL plasmons at the frequencies $\omega=(v_F/g)k_{p,LL}=2\pi c/\lambda_0<eV/\hbar$.

40 correlation functions [11–15]. 1D LL does not have quasiparticle excitations, *e.g.* quasi-free
 41 electrons [11]. Instead, the elementary excitations of LL become plasmons and spinons, which
 42 are collective oscillations of charge and spin, respectively [16]. Once electrons are injected into
 43 LL, a substantial fraction of energy is directly converted to plasmons at finite frequencies. It
 44 enables efficient infrared lighting with electrically excited LL plasmons if the near-field
 45 plasmons can be coupled to the far-field. Many 1D metallic systems supports LL [12,17–21],
 46 but LL approximation in the 1D systems often breaks down for high energy excitations in the
 47 1D systems due to the nonlinear dispersion. Contrastingly, single-walled carbon nanotubes
 48 (SWNTs) provide the linear dispersion up to ~ 1 eV, making them an ideal platform for the
 49 infrared LL plasmons [20–31], exhibiting the power-law scaling of the tunneling
 50 conductance [30,31], the shot noise currents [25,34,35], and high quality factor infrared
 51 plasmons [24,32,33].

52 We investigate theoretically high-efficiency IRLEDs using antenna-coupled LL plasmons in
 53 single-walled carbon nanotubes (SWNTs). We estimate a 14% injection efficiency to excite

54 infrared LL plasmons in SWNTs. We design a gold nanorod (AuNR) antenna to couple the
 55 deep-subwavelength LL plasmons to the far-field. It significantly mitigates the large
 56 wavelength mismatch between the LL plasmons and infrared photons and boosts the far-field
 57 radiation yield. Our calculation shows that an electrical-to-optical power conversion efficiency,
 58 *i.e.* the quantum efficiency, over 3.2% can be achieved. Such efficient IRLEDs based on 1D
 59 LL in SWNTs can enable a new type of infrared nanophotonic applications.

60 Figure 1a illustrates the AuNR-coupled SWNT IRLED (SWNT/AuNR IRLED). The metallic
 61 SWNT has l_{SWNT} and $R=1$ nm, which bridges the AuNR antennas with a gap of d . Each AuNR
 62 has $l_{\text{Au}}=3.8$ μm and $W=60$ nm, yielding an antenna resonance around $\lambda_0=11$ μm . This
 63 wavelength is in the middle of the infrared atmospheric window ($\lambda_0=8\sim14$ μm). Electrons can
 64 be injected into LL in SWNT via tunneling from AuNRs [30,31,36–38]. We assume air gap as
 65 a tunneling junction with $t=1$ nm and $l_{\text{T}}=5$ nm (Fig. 1a).

66 Light emission in our IRLED is achieved via two-step process (Fig. 1a). In step I, we inject
 67 electrons into SWNT by tunneling from biased AuNRs, resulting in the efficient generation of
 68 the LL plasmons in a two-terminal geometry [39,40]. The SWNT length l_{SWNT} in our IRLED
 69 is comparable to the LL plasmon wavelength $\lambda_{\text{p},\text{LL}}$ to support the Fabry-Perot resonances
 70 (FPRs). In step II, AuNRs act as a nanoantenna converting the deep-subwavelength LL
 71 plasmons to the photons. We investigate the efficiency of each step and examine the quantum
 72 efficiency of our LL-based IRLEDs, characterizing the electrical-to-optical power conversion.

73 We first estimate the injection efficiency, characterizing the electrical-to-plasmonic power
 74 conversion, in step I. Using a LL theory [22], we calculate the tunneling density of states
 75 (TDOS) for electron tunneling at bias energy $\varepsilon=eV$ from a metallic electrode to LL. TDOS
 76 spectrum is composed of peaks of the quantum numbers of plasmons (n_{p}) and spinons (n_0), *i.e.*

$$77 A(\varepsilon)=\sum_{n_p,n_0\geq 0}A_{n_pn_0}(\varepsilon)=\sum_{n_p,n_0\geq 0}C_{n_pn_0}\delta\left\{E_C+n_p\varepsilon_p+(n_0+1/2)\varepsilon_0-\varepsilon\right\} \text{ with the spinon energy}$$

78 $\varepsilon_0=\pi\hbar v_F/L_{\text{SWNT}}$, the plasmon energy $\varepsilon_p=\varepsilon_0/g$, and the Coulomb energy

$$79 E_C=(\varepsilon_p^2-\varepsilon_0^2)/8\varepsilon_0 \text{ [22]. } C_{n_pn_0}/C_{00}=c_{3/4}^{n_0}\sum_{0\leq i\leq j\leq n_p}c_{2g^{(+)}}^{n_p-1}c_g^{j-i}c_g^i, \text{ determines a height of each}$$

80 TDOS peak, where $g^{(\pm)}\equiv(g^{-1}\pm g)/16$ and $c_g^n\equiv\Gamma(g+n)/\Gamma(g)\Gamma(1+n)$ with the Euler
 81 gamma function Γ . TDOS spectrum (Fig. 2a) shows multipole plasmon resonances; dipole,
 82 quadrupole, and octupole ($n_{\text{p}}=1, 2$, and 3 , respectively), while peaks of $n_{\text{p}}=0$ are the ground
 83 state. Each plasmon accompany spinon resonances of $n_0\geq 0$. TDOS determines the current ($I(\varepsilon)$,

84 Fig. S1a) and the power ($P(\varepsilon)$, Fig. S1b) of the tunnel electrons by

$$85 A(\varepsilon)\propto dI(\varepsilon)/d\varepsilon=d^2P(\varepsilon)/d\varepsilon^2, \text{ where } I(\varepsilon)=\sum_{n_p,n_0\geq 0}\int_0^\varepsilon A_{n_pn_0}(\varepsilon')d\varepsilon'\equiv\sum_{n_p,n_0\geq 0}I_{n_pn_0}(\varepsilon) \text{ and}$$

$$86 P(\varepsilon)=\sum_{n_p,n_0\geq 0}\int_0^\varepsilon I_{n_pn_0}(\varepsilon')d\varepsilon'\equiv\sum_{n_p,n_0\geq 0}P_{n_pn_0}(\varepsilon).$$

87 In the LL-based IRLEDs, the dipolar plasmon ($n_{\text{p}}=1$) alone contributes to the IR emission,
 88 while the higher plasmons ($n_{\text{p}}\neq 1$) and spinons ($n_{\text{p}}\neq 0$) are loss channels. The power of the

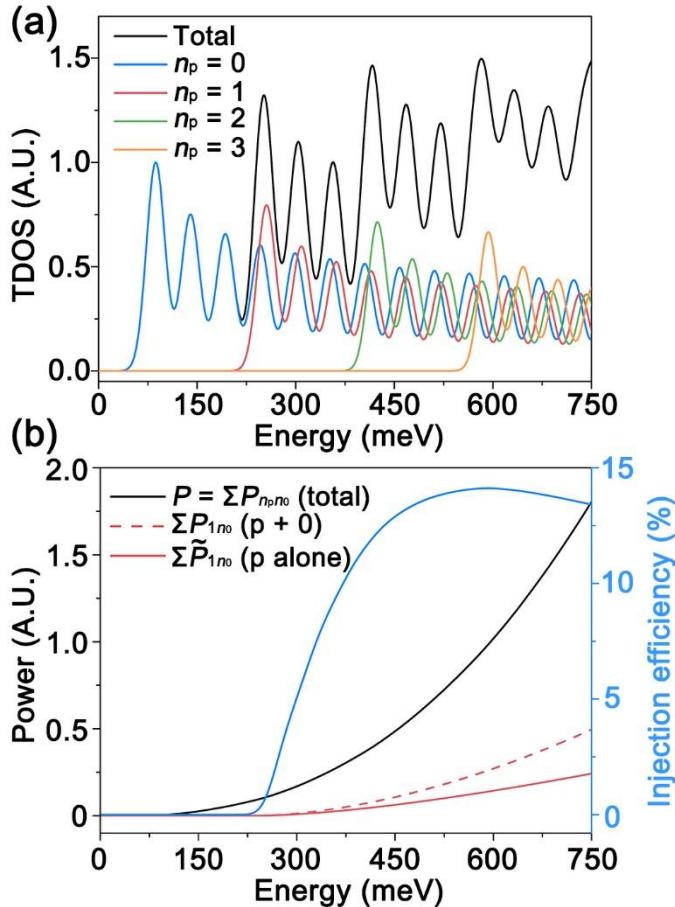


FIG. 2. (a) The tunneling density of states (TDOS) at the end of SWNT with $g=0.31$, $R=1$ nm, and $l_{\text{SWNT}}=31.2$ nm (black line). TDOS is decomposed into the plasmon (n_p) and spinon (n_0) modes (colored lines with $\varepsilon_0=53.0$ meV, $\varepsilon_p=169$ meV, $E_C=60.5$ meV, and the broadening of 20.0 meV). (b) Power spectra of the total LL ($P(\varepsilon) \equiv \sum_{n_p \geq 0, n_0 \geq 0} P_{n_p n_0}(\varepsilon)$; black line), the dipolar plasmons with the spinons ($\sum_{n_0 \geq 0} P_{1 n_0}(\varepsilon)$; dashed red line), and the pure dipolar plasmons ($\sum_{n_0 \geq 0} \tilde{P}_{1 n_0}(\varepsilon)$; red line). The injection efficiency of the dipolar plasmons ($\eta_{\text{inj}}(\varepsilon) \equiv \sum_{n_0 \geq 0} \tilde{P}_{1 n_0}(\varepsilon) / P(\varepsilon)$; blue line).

89 dipolar plasmons can be calculated as
90 $\sum_{n_0 \geq 0} \tilde{P}_{1 n_0}(\varepsilon) \equiv \sum_{n_0 \geq 0} P_{1 n_0}(\varepsilon) [\varepsilon_p / \{\varepsilon_p + (n_0 + 1/2)\varepsilon_0 + E_C\}]$. We can define the injection efficiency
91 as $\eta_{\text{inj}} \equiv \sum_{n_0 \geq 0} \tilde{P}_{1 n_0}(\varepsilon) / P(\varepsilon)$. Fig. 2b shows η_{inj} can be achieved up to 14% at 590 meV, and it is
92 significantly higher than the injection efficiency of narrow-gap LEDs (<1%) [5–10] and the
93 inelastic tunneling efficiency through a metallic gap ($\sim 10^{-6}$ /eV) [41].

94 To evaluate the radiation efficiency in step II, we perform electromagnetic calculations for

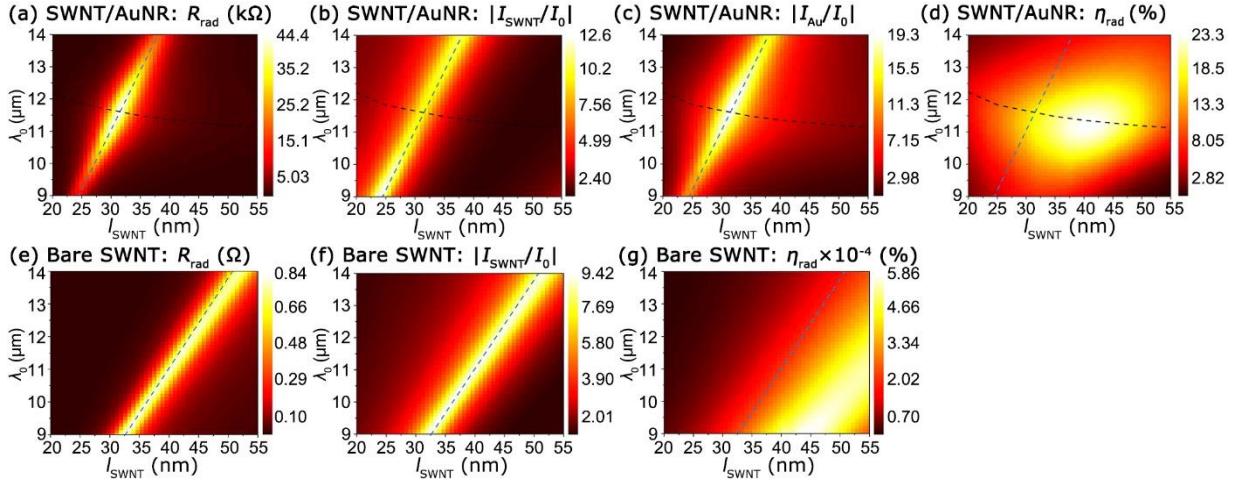


FIG. 3. (a) The radiation resistance $R_{\text{rad}} \equiv 2P_{\text{rad}}/I_0^2$, (b) the current $|I_{\text{SWNT}}/I_0|$ of SWNT, (c) the current $|I_{\text{Au}}/I_0|$ of AuNRs, and (d) the radiation efficiency $\eta_{\text{rad}} \equiv P_{\text{rad}}/(P_{\text{rad}} + P_{\Omega,\text{Au}} + P_{\Omega,\text{SWNT}})$ of the SWNT/AuNR IRLEDs with the radiation power P_{rad} , the gold absorption power $P_{\Omega,\text{Au}}$, and the SWNT absorption power $P_{\Omega,\text{SWNT}}$ upon the current source I_0 in SWNT. Black and blue dashed lines are AuNR and SWNT FPRs, respectively. (e) The radiation resistance R_{rad} , (f) the current $|I_{\text{SWNT}}/I_0|$, and (g) the radiation efficiency $\eta_{\text{rad}} = P_{\text{rad}}/(P_{\text{rad}} + P_{\Omega,\text{SWNT}})$ of bare SWNTs.

95 our IRLEDs. We model SWNT as a metallic cylinder with a radius of R and the permittivity of
96 ϵ_{SWNT} . By correlating a LL theory with an electromagnetic theory, we can express ϵ_{SWNT} as (see
97 Supplemental Material for the derivation):

$$98 \quad \text{Re}\{\epsilon_{\text{SWNT}}(\omega)\} = -\frac{16}{1-g^2} \frac{k_e e^2}{\epsilon_{\text{eff}} \hbar v_F} \frac{v_F^2}{\pi R^2} \frac{1}{\omega^2}, \quad (1)$$

99 with $k_e = 1/4\pi\epsilon_0$ and the reduced Plank constant \hbar . We find ϵ_{SWNT} can be written in terms of g
100 and R . $\text{Im}(\epsilon_{\text{SWNT}})$ can be obtained by $Q = \{\omega \text{Re}(d\epsilon_{\text{SWNT}}/d\omega)\}/\{2\text{Im}(\epsilon_{\text{SWNT}})\}$ [42].
101 Hereinafter, we use experimental values [24,32], $g=0.3$ and $Q=20$.

102 In Fig. 3 we compare the radiation properties of a bare SWNT and our SWNT/AuNR IRLEDs.
103 As shown in Fig. 3a, the radiation resistance $R_{\text{rad}} \equiv 2P_{\text{rad}}/I_0^2$ is determined by the spectral
104 proximity of SWNT FPR (blue line) and AuNR FPR (black line). Inheriting the linearity of LL
105 plasmon dispersion (Fig. 1b), the SWNT FPR follows a linear FPR dispersion,
106 $\lambda_0 = 2l_{\text{SWNT}}(cg/v_F)/(1-\varphi_{\text{SWNT}}/\pi)$ with $\varphi_{\text{SWNT}}/\pi \sim 0.36$ [43,44]. The AuNR FPR is almost
107 constant over l_{SWNT} changes due to the AuNR FPR condition $\lambda_0 = 2l_{\text{Au}}/(1-\varphi_{\text{Au}}/\pi)$. The
108 maximum radiation resistance $R_{\text{rad}} \sim 44.4$ kΩ appears at the crossing point of two modes, namely,
109 the maximum radiation point, *i.e.* $(l_{\text{SWNT}}, \lambda_0) = (31 \text{ nm}, 12 \mu\text{m})$. Contrastingly, the bare SWNT
110 in Fig. 3e shows poor R_{rad} , originating from the wavelength mismatch.

111 Figure 3 also shows the optical currents,

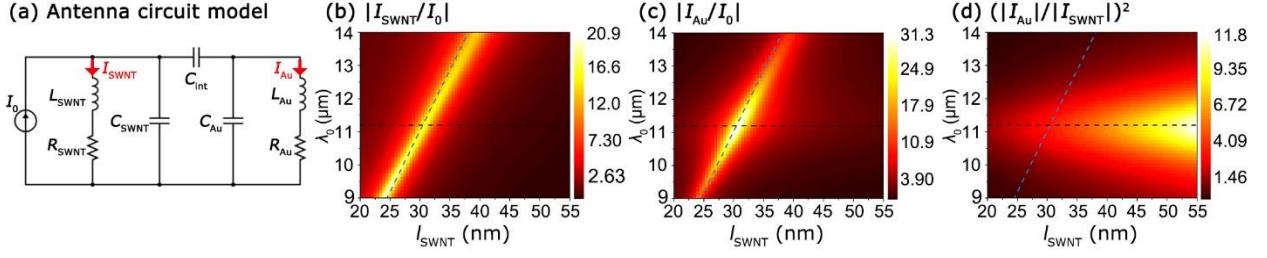


FIG. 4. (a) The antenna circuit model consists of two RLC circuits with the resistance R , inductance L , and capacitance C . C_{int} connects two RLC circuits characterized by the resonant impedances $Z_{SWNT}(\omega)$ and $Z_{Au}(\omega)$, respectively. Model results of (b) $|I_{SWNT}/I_0|$, (c) $|I_{Au}/I_0|$, and (d) $(|I_{Au}|/|I_{SWNT}|)^2$ (Black and blue dashed lines: FPR modes of AuNR and SWNT, respectively.)

112 $|I_{Au(SWNT)}| \equiv (\text{area / length}) \int_{V_{Au(SWNT)}} dV | -i\omega (\epsilon_{Au(SWNT)} - \epsilon_0) \mathbf{E} |$, in SWNT and AuNR [45]. They s

113 how FPR modes of SWNT and AuNR (Figs. 3b, c, and f). In Figs. 3b&c, we find that $|I_{Au}/I_0|$ i
114 s maximized, while $|I_{SWNT}/I_0|$ slightly decreases when two FPRs meet. This implies AuNRs ta
115 ke the current from SWNT to radiate the LL plasmons to the far-
116 field. Comparing R_{rad} (Fig. 3a) and $|I_{Au}/I_0|$ (Fig. 3c), we can find that they exhibit the same pat
117 tern. This shows that AuNRs play a dominant role in the far-field radiation in our IRLEDs.

118 Figures 3d&g compare the radiation efficiency η_{rad} of our SWNT/AuNR IRLED and the bare
119 SWNT. We define η_{rad} as

$$120 \eta_{rad} \equiv P_{rad} / P_{tot} = \left(2P_{rad} / |I_{Au}|^2 \right) / \left\{ \left(2P_{rad} / |I_{Au}|^2 \right) + R_{\Omega,Au} + \left(|I_{SWNT}| / |I_{Au}| \right)^2 R_{\Omega,SWNT} \right\}$$
 with the total

121 power $P_{tot} \equiv P_{rad} + P_{\Omega,Au} + P_{\Omega,SWNT}$, the absorption power of SWNT (AuNR)
122 $P_{\Omega,SWNT(Au)} = |I_{SWNT(Au)}|^2 R_{\Omega,SWNT(Au)} / 2$, and the Ohmic resistance of SWNT (Au)
123 $R_{\Omega,SWNT(Au)} = (\text{length/area}) \text{Im}\{1/\omega\epsilon_0(1-\epsilon_{SWNT(Au)})\}$ (Fig. S2c&d). η_{rad} of the bare SWNT (Fig. 3g)
124 is poor because of the wavelength mismatch. SWNT/AuNR IRLED (Fig. 3f) has higher η_{rad}
125 with the enhancement along AuNR FPR. The maximum efficiency point with $\eta_{rad}=23\%$ appears
126 at $(l_{SWNT}, \lambda_0)=(40 \text{ nm}, 11 \mu\text{m})$ away from the maximum radiation point at $(31 \text{ nm}, 12 \mu\text{m})$ with
127 $\eta_{rad}=17\%$. The resultant quantum efficiencies at the two points reach $\eta_Q=\eta_{inj} \times \eta_{rad} \sim 3.2\%$ and
128 2.4%, respectively. These are higher than other light sources using the mid-infrared bandgap
129 emission, whose quantum efficiencies are limited to <1% [1,8–10].

130 To understand the emission properties, we develop a circuit model shown in Fig. 4a (see
131 Supplemental Material for details). It consists of two parallel RLC circuits describing the
132 resonances in SWNT and AuNRs, respectively. Two RLC circuits are capacitively coupled
133 through the interaction capacitance C_{int} . The antenna impedance,

$$134 Z_{SWNT(Au)}(\omega) = i\omega L_{SWNT(Au)} \left(\omega_{0,SWNT(Au)}^2 / \omega^2 \right) \left\{ \left(\omega^2 + i\gamma_{SWNT(Au)}\omega \right) / \left(\omega^2 - \omega_{0,SWNT(Au)}^2 + i\gamma_{SWNT(Au)}\omega \right) \right\}$$

135 characterizes each RLC circuit, and it includes the resonance information of SWNT (AuNR)
136 with $\omega_{0,SWNT(Au)} \equiv \sqrt{1/L_{SWNT(Au)}C_{SWNT(Au)}}$ and $\gamma_{SWNT(Au)} \equiv R_{SWNT(Au)} / L_{SWNT(Au)}$. Solving the
137 antenna circuit in Fig. 4a, we can obtain I_{SWNT} and I_{Au} (Figs. 4b&c) that reproduce Figs. 3b&c.

138 Fig. 4a also explains η_{rad} in Fig 3d. In η_{rad} , the ratio $(|I_{SWNT}|/|I_{Au}|)^2$ (Fig. S2a) is responsible for
139 η_{rad} because $(2P_{rad}/|I_{Au}|^2)$, $R_{\Omega,Au}$, and $R_{\Omega,SWNT}$ (Figs. S2b-d) do not show η_{rad} behavior in Fig. 3d.
140 We obtain

141 $\left(\frac{|I_{Au}|}{|I_{SWNT}|}\right)^2 = \left(\frac{L_{SWNT}}{L_{Au}}\right)^2 \left\{\left(\omega^2 + \gamma_{SWNT}^2\right) / \left(\omega^2 + \gamma_{Au}^2\right)\right\} |Z_{Au}(\omega)|^2 / \left|1/i\omega C_{int} + Z_{Au}(\omega)\right|^2$
142 from Fig. 4a. Fig. 4d shows that $(|I_{Au}|/|I_{SWNT}|)^2$ is enhanced at the AuNR FPR not at the SWNT
143 FPR because it does not have $Z_{SWNT}(\omega)$. It also has $(L_{SWNT})^2 \propto (l_{SWNT})^2$, explaining why the
144 maximum efficiency point (Fig. 3d) differs from the maximum radiation point (Fig. 3a).

145 In conclusion, we suggested IRLED based on antenna-coupled LL plasmons. Our IRLED
146 can operate with the quantum efficiency of more than 3.2% with the combination of the highly
147 efficient LL plasmon excitation and nanoantenna. Further improvement of the IRLED
148 efficiency can be achieved by further optimization of the nanoantenna design and a
149 semiconducting injection electrode. Also, our scheme can be applied to a broad spectral range
150 from terahertz to near-infrared frequencies where the emission wavelength can be tuned by
151 geometry of SWNT and AuNR lengths.

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