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Thermal and nonthermal exciton light emissions from carbon nanotubes above 1000 K

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High-temperature light emission from nanomaterials has been extensively studied for light-emitting device applications. However, thermodynamic properties of their light emission under high-temperature conditions, which are critical for energy applications, have not been fully elucidated. Herein, we demonstrate that carbon nanotubes emit nonthermal light, specifically exciton luminescence in the near-infrared (NIR) region, even at very high temperatures. By investigating the chemical potential of photons, which indicates an extent of nonthermal excitation beyond thermal equilibrium, we show that exciton light emission from a semiconducting nanotube maintains a nonthermal condition above 1000 K, enabling intense radiation that exceeds thermal radiation at the same temperature. Furthermore, the chemical potential of photons emitted by an individual metallic nanotube under electric-current injection far surpasses that of semiconducting nanotubes at ≈ 1000 K. These findings facilitate applications of carbon nanotubes as high-intensity, narrow-band NIR emitters operable even at very high temperatures for advanced energy-conversion devices.

Thermal and nonthermal light-emission phenomena of nanomaterials such as carbon nanotubes¹⁻¹³, graphene^{14,15}, and atomically thin semiconductors¹⁶ at high temperatures have recently attracted attention as they demonstrate promising potential in various light-emitting device applications. In particular, semiconducting single-walled carbon nanotubes (SWCNTs; Fig. 1a)¹⁷ have exhibited narrow-band thermal radiation in the near-infrared (NIR) region^{4,5} with a bandwidth on the order of $k_{\rm B}T$ (where $k_{\rm B}$ and T denote the Boltzmann constant and temperature, respectively), which is important in wavelength-selective thermal emitters necessary for efficient thermophotovoltaic energy conversion^{18,19}. This unique property of SWCNTs is attributed to thermally stable excitons with substantial binding energies (Fig. 1b; 0.3–0.5 eV), which originate from strong quantum confinement and weak dielectric screening for Coulomb interactions²⁰⁻²². In addition to semiconducting SWCNTs, metallic SWCNTs, wherein the binding energy of excitons in the massive one-dimensional (1D) subband is ~50 meV²³, also exhibit sharp high-temperature light-emission spectra that differ from conventional thermal radiation incandescence¹⁻⁴, demonstrating application potential as NIR light emitters.

Unlike conventional materials, low-dimensional nanomaterials readily attain thermal nonequilibrium among their electronic and lattice (phonon) systems under optical or electronic pumping even at high temperatures^{1,14,15,24,25} owing to limited phase space for quantum scatterings satisfying energy and momentum conservations. To describe the hightemperature light-emission (luminescence) phenomena under the nonequilibrium steady-state conditions in nanomaterials such as SWCNTs and graphene, previous studies^{1,14,15,24,25} have relied on the effective temperatures of the electron and lattice systems. However, the effective temperature $T_{\rm eff}$ is the corresponding temperature of black-body radiation having the same intensity within a given photon energy range, not the real temperature of the system emitting the photons²⁶. Therefore, it cannot properly describe the intraband thermal distribution of carriers or excitons at the real temperature lower than $T_{\rm eff}$ nor the entropy carried away from the material with the emitted photons. This limitation is particularly problematic when trying to apply high-temperature luminescence to energy conversion applications, such as thermophotovoltaic power generation²⁷⁻²⁹. When the intraband thermalization of carriers or excitons is much faster than their interband relaxation, more comprehensively and more compatible with thermodynamics, the thermal and nonthermal radiation of a material can be described by a generalization of Planck's law using the chemical potential of photons μ , which corresponds to the free energy per photon²⁶. The generalized Planck's law has been used to describe the steady-state lightemission phenomena of rare-earth-doped ceramics28, conventional semiconductors³⁰⁻³², and photovoltaic devices^{33,34} under photo-irradiation or electric-current injection. When excited species in a material have a chemical potential μ_{ex} under photo-absorption or electric current injection, the radiation in thermodynamic equilibrium with the material acquires a

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Fig. 1 | **Excitonic light emission of carbon nanotubes. a** Schematic of a singlewalled carbon nanotube (generated using VESTA 3⁵⁷). **b** Schematic of excitonic Rydberg series. Excitons are exotic atoms with well-defined energy consisting of mutually bound electron–hole pairs. The luminescence of SWCNTs arise from radiative recombination of the first subband exciton state (wavy arrow).

chemical potential μ equal to μ_{ex} . Therefore, the μ is zero for pure thermal radiation but is finite for luminescence²⁶. At a given photon energy, the nonthermal (luminescent) radiation from an object at a given temperature *T* is brighter than the thermal radiation by a factor of $\approx e^{\mu/k_{\rm B}T^{26}}$. Hence, a material that realizes a high μ NIR emission even at very high temperatures is greatly desired as a high-power wavelength-selective NIR light emitter exceeding the blackbody limit of thermal radiation. However, the μ of steady-state light emissions in SWCNTs under various excitation conditions at various temperatures remains unclear.

Herein, we examine the chemical potential μ of high-temperature steady-state light emission from semiconducting and metallic SWCNTs under photoexcitation and electronic excitation at up to temperatures exceeding 1000 K. High-temperature exciton light emission from a semiconducting SWCNT under photoexcitation discernibly transitioned from luminescence to thermal radiation as the temperature increased to 1400 K. From this result, we could determine the chemical potential of photons μ , allowing comprehensive, thermodynamic-compatible treatments of the thermal and nonthermal light emissions of semiconducting SWCNTs. Our observations conclusively demonstrate exciton luminescence from SWCNTs even at temperatures exceeding 1000 K. In addition, we determined the μ of light emission from an individual metallic SWCNT under electric-current injection. The resulting μ far surpassed that of semiconducting SWCNTs at temperatures around 1000 K, suggesting that the maximum number of excitons (electron-hole pairs) that can be accommodated at once is much higher in metallic SWCNTs under electric current injection than in semiconducting SWCNTs under optical excitation. This difference may be understood as a consequence of the different Coulomb interaction strengths between the two types of SWCNTs. Besides quantitatively elucidating the thermodynamic aspects of exciton steady-state luminescence in SWCNTs at high temperatures, our results highlight the promising application potential of SWCNTs as high-intensity, narrowband emitters of light in the NIR region operable even at elevated temperatures.

Results and Discussion

Single nanotube spectroscopy under laser irradiation

Structure-defined, isolated, and pristine SWCNTs were suspended in vacuum and irradiated using a continuous-wave (CW) laser to study high-temperature steady-state light emission (Fig. 2a). Minimizing energy exchange with the surroundings was achieved by suspending individual SWCNTs over open slits in a vacuum chamber. These SWCNTs were

grown directly on a substrate equipped with Pt electrodes (Fig. 2b), enabling electrostatic gating and/or current injection³⁵. Under weak-intensity CW laser irradiation near room temperature (RT), photoluminescence (PL) was induced, with the excitation intensity gradually increasing to provide localized heating while preserving the charge balance of the SWCNTs⁴. Figure 2c shows an image of a semiconducting SWCNT exhibiting PL stemming from the lowest-energy S_{11} excitons, characterized by chiral indices defining the SWCNT structure as (12,5) with a diameter of 1.18 nm. The Rayleigh scattering spectra revealed distinct S_{11} and S_{22} exciton resonances at 0.86 and 1.57 eV, respectively (see Methods for experimental details), along with PL (only for S_{11}) at RT (Fig. 2d, e). The CW laser, operating at 1.534 eV and nearly resonant with the S_{22} exciton state, was focused onto vacuum-suspended SWCNTs using an objective lens, facilitating the detection of exciton light emission via dark-field measurements.

Light emission spectra in vacuum and nitrogen atmosphere

In Fig. 3a, b, the light emission spectra of the (12,5) semiconducting SWCNT suspended in vacuum are compared at various excitation intensities, alongside spectra obtained in a nitrogen atmosphere to compare the photothermal effect (all spectra from the experiment are shown in Supplementary Figs. 1 and 2). As shown in Fig. 3a, the exciton emission spectra exhibited minimal broadening in a nitrogen atmosphere with increasing excitation laser power, accompanied by negligible peak shifts. Since the peak energy is temperature-dependent³⁶, this result indicates that the temperature of the SWCNT remained near room temperature. In contrast, the spectra recorded in vacuum drastically changed with variations in the excitation intensity (Fig. 3b). Initially, a blueshift of the peak was observed up to an excitation intensity of 1.5 MW cm⁻², followed by a redshift accompanied by remarkable linewidth broadening at higher excitation intensities. The blueshift was potentially attributed to the desorption of gas molecules from the SWCNT³⁷, and the redshift was attributed to an increase in temperature³⁶. The inset of Fig. 3c illustrates the excitation intensity (I_{ex}) dependence of the peak energies (left axis), which enabled the determination of the temperature (right axis; refer to Methods for temperature estimation)^{36,37}. The temperature of the SWCNT in vacuum increased to \approx 1400 K, in contrast to the case of the nitrogen atmosphere.

Figure 3c shows the integrated radiation intensities plotted against the excitation intensity. In a nitrogen atmosphere, the radiation intensity of the SWCNT exhibited a saturation trend attributed to exciton-exciton annihilation (EEA)-the collision of two excitons resulting in the generation of one exciton and heat³⁸. This EEA phenomenon limited the maximum number of photogenerated excitons in the SWCNT. Thus, in excitation intensity above $\approx 2 \text{ MW cm}^{-2}$, the photoexcitation was used only for heat generation without an increase in the number of photogenerated excitons. Conversely, in vacuum, the saturation behavior owing to EEA was evident for I_{ex} in the range of 0.1–1.6 MW cm⁻²; however, the radiation intensity curve qualitatively changed beyond this range. The integrated intensity suddenly decreased at $I_{ex} \approx 1.5$ MW cm⁻², and exponentially increased after $I_{\rm ex} \approx 2 \ {\rm MW} \ {\rm cm}^{-2}$, while the temperature increased monotonically the inset of Fig. 3c. The intensity in vacuum became comparable to and eventually surpassed that in nitrogen gas at excitation intensities roughly corresponding to ≈1100 K. The inset of Fig. 3d shows the temperature dependence of the integrated radiation intensities, where the intensities and temperature were the data shown in Fig. 3c and the inset, respectively. Above ≈ 1100 K, a clear exponential behavior emerged, following $e^{-E/k_{\rm B}T}$ with a fitting result yielding $E = 0.80 \pm 0.02 \text{ eV}$ (red line). This value nearly coincides with the exciton radiation peak energy, as seen in the spectra corresponding to excitation intensities above 3.0 MW cm⁻² (Fig. 3b), suggesting that the light emission is attributed to thermal radiation with its chemical potential $\mu = 0$. Therefore, the red line indicates the temperature dependence of the intensity expected for thermal radiation from the SWCNT. In contrast, the radiation intensities in the temperature range below ≈ 1100 K deviates from and surpasses the thermal radiation line, indicating that the light emission below $\approx 1100 \text{ K}$ is attributed to luminescence.



Fig. 2 | **Dark-field spectroscopy of a carbon nanotube suspended in vacuum. a** Schematic of the experimental setup. **b** Image of the substrate with an open slit featuring electrodes at both ends. Scale bar: 100 μm. **c** PL image of a semiconducting single-walled carbon nanotube (SWCNT). Scale bar: 5 μm. **d**, **e** Near-infrared (**d**)

and visible (**e**) range Rayleigh scattering spectra of (12,5) semiconducting SWCNT for the first (S_{11}) and second (S_{22}) sub-band excitons, alongside the PL stemming from radiative S_{11} exciton recombination. The black arrow in **e** denotes the photon energy of the semiconducting continuous-wave (CW) laser.

Evaluation of the light emission state

Here, we quantitatively assess the extent of excitation beyond thermal equilibrium in the observed high-temperature steady-state (HTSS) luminescence using the chemical potential of photons μ at each temperature *T*. The μ values are determined from the integrated emission intensities (the inset of Fig. 3d) as follows. The spectrally integrated thermal and nonthermal light emission intensities from excitons, $I_{\rm em}(T, \mu)$, are expressed as^{26,39}

$$I_{\rm em}(T,\mu) = \int_{0}^{\infty} d\omega \frac{\hbar\omega^{3}}{4\pi^{2}c^{2}} e(\omega) f(T,\omega,\mu)$$
(1)

where ω is the optical frequency, $e(\omega)$ corresponds to the spectral emissivity, and $f(T, \omega, \mu) = [e^{(\hbar\omega-\mu)/k_{\rm B}T} - 1]^{-1}$ is the Bose–Einstein distribution function. According to the theory of thermal radiation in the framework of fluctuational electrodynamics³⁹, the asymptotic form of $e(\omega)$ for SWCNTs in the exciton resonance vicinity is obtained as $e(\omega) \simeq \omega d(3c)^{-1} \text{Im}[\varepsilon(\omega)]$, where *d* is the SWCNT diameter and $\varepsilon(\omega)$ is the complex dielectric function along the SWCNT axis for the longitudinal excitons⁵. At the narrow exciton resonance band, the emission intensity was approximately proportional to the Bose–Einstein term, $I_{\text{em}}(T, \mu) \propto A_e f(T, \omega, \mu) \simeq A_e e^{-(\hbar\omega-\mu)/k_{\rm B}T}$ for $\hbar\omega - \mu \gg k_{\rm B}T$, where $A_e \propto \int \omega^3 e(\omega) d\omega$ is the frequency-integrated emissivity. Because the integrated emission intensity of pure thermal radiation ($\mu = 0$) simply follows $I_{\text{em}}(T, 0) \propto A_e e^{-\hbar\omega/k_{\rm B}T}$, the chemical potential μ at each temperature is given by

$$\mu \simeq k_{\rm B} T \ln \left(I_{\rm em}^{\rm exp} \left(T, \mu \right) / I_{\rm em}(T, 0) \right) \tag{2}$$

Thus, the μ value can be determined using the ratio of the measured integrated emission intensity $I_{\rm em}^{\rm exp}(T,\mu)$ to the pure thermal radiation intensity $I_{\rm em}(T,0)$ indicated by the red line shown in the inset of Fig. 3d, through Eq. (2). Figure 3d shows the chemical potential of photons as a function of temperature. A crossover between thermal radiation ($\mu = 0$) and luminescence ($\mu > 0$) was observed with temperature variation. Even at 1000 K, the nonzero μ value of 0.04 eV was maintained. This value is only about 5% of the emission photon energy (0.82 eV), but corresponds to the emission intensity approximately 1.6 times stronger than thermal radiation at the same temperature.

The HTSS luminescence under electrostatic carrier doping

The HTSS luminescence of the semiconducting SWCNT exhibited characteristics similar to those of thermal radiation under electrostatic carrier doping, rather than conventional luminescence at relatively low temperatures. We examined the gate bias dependence of the HTSS luminescence intensity at 900 K, at which the crossover between thermal radiation and luminescence was observed. We used the field effect to dope carriers into the (12,5) SWCNT via electrostatic gating (Fig. 3e). The intensity of the HTSS luminescence at 900 K gradually diminished with increasing gate bias $|V_G|$ (Fig. 3f). Previous studies have reported that carrier doping modulates excitonic absorption and PL at RT due to various factors, including reduced exciton oscillator strength from the state filling effect⁴⁰, exciton screening and carrier-exciton scattering³⁵, charged exciton formation⁴¹⁻⁴³, and/or an increase in the nonradiative decay rate owing to phonon-assisted indirect exciton ionization⁴⁴. Modulations in the intensity of the Rayleigh scattering at RT (Fig. 3g) and PL at 450 K (Fig. 3h; moderate temperature) were also observed in our FET device. Figure 3i summarizes the gate voltage dependencies of the integrated light emission intensities of the exciton spectra shown in Fig. 3f, g, with changes in the amplitude of the integrated emissivity Ae derived from the Rayleigh spectra results (see Methods for details). The HTSS luminescence at 900 K exhibited a gatedependent intensity change closely following the A_e variation. The inset in Fig. 3i illustrates the change in the chemical potential of photons using the same procedure as that in Fig. 3d, plotted as a function of $|V_G|$. μ for PL at 450 K significantly decreased from 0.47 to 0.36 eV owing to the abovementioned mechanism. In contrast, $\mu \approx 0.13 \text{ eV}$ for the HTSS luminescence at 900 K remained nearly unchanged until $|V_G| = 2$ V, indicating that the light variation was primarily governed by the change in emissivity A_e , resembling the behavior that should be observed in pure thermal radiation (refer to Supplementary Note 1 for a detailed discussion on the mechanism).

Chemical potential of photons in emission of a metallic type

Having established a method for evaluating the chemical potential of photons in SWCNT light emission in the previous section, we extended this approach beyond semiconducting SWCNTs to examine the peaked light emission of metallic SWCNTs under electric current injection^{1–3}. This phenomenon has long been debated, hindering a comprehensive understanding of SWCNT light emission. Although this light emission has been attributed to a thermal equilibrium between the electronic system with nonthermal optical phonons excited under electric current injection, the extent of excitation beyond thermal equilibrium has remained elusive. In metallic SWCNTs, the optical transition between massless 1D metallic bands is optically forbidden⁴⁵, resulting in the observation of a peak originating from the exciton (electron-hole) recombination in the first massive gapped sub-band (M_{11}) under current injection (inset in Fig. 4a). While one might initially attribute this to incandescence similar to a light bulb, the



Fig. 3 | Light emission of a semiconducting carbon nanotube. a, b Light emission spectra of the (12,5) semiconducting single-walled carbon nanotubes (SWCNTs) in nitrogen (a) and vacuum (b) at various excitation intensities, normalized at maximum values. c Excitation intensity dependence of the integrated intensities of the light emission spectra in (a, b). The inset illustrates the excitation intensity (I_{ex}) dependence of the peak energy (left) and the corresponding temperature (right). d Temperature (T) dependence of photon chemical potential (μ). The inset shows the integrated intensities of the light emission (I_{int}) as a function of the inverse ($1/k_BT$; bottom) and corresponding temperatures (top). The red solid line is the

fitting result based on the Boltzmann statistics. **e** Field-effect transistor device of the SWCNT. $V_{\rm DS}$ and $V_{\rm G}$ represent the drain–source and gate voltages, respectively. **f-h** Gate voltage dependence of high-temperature steady-state luminescence (HTSSL) spectra at 900 K (**f**), Rayleigh scattering spectra at room temperature (RT) (**g**), and photoluminescence (PL) at 450 K (**h**). $E_{\rm P}$ denotes the photon energy. **i** Gate voltage dependence of integrated intensities of spectra shown in (**f-h**). The frequency-integrated emissivity A_e was estimated from the Rayleigh scattering spectra. The inset shows the photon chemical potential (μ) of the HTSSL (red circles) and the PL at 450 K (blue squares).

situation differs for SWCNTs owing to their unique nonequilibrium optical phonon state under current injection^{1,24,46} (see Supplementary Note 2 and Supplementary Fig. 3 for details). Although the effective temperatures of the optical phonons were evaluated within the framework of the none-quilibrium optical phonon model^{24,46}, the fundamental thermodynamic aspects of this light emission type, including its photon chemical potential and its relation to the actual temperature, remain unclear.

Following the outlined approach, we conducted a direct comparison between the M_{11} light emission from a metallic SWCNT under current injection (Fig. 4a) and pure thermal radiation of the same state obtained through laser heating (Fig. 4b). As depicted in Fig. 4c, the emission intensity under current injection surpassed that of the thermal radiation line (red line) by over a hundredfold. Notably, the light emission intensity from current injection at 1000 K corresponds to the intensity of pure thermal radiation at \approx 2000 K. The apparent temperature much higher than the real temperature is consistent with the effective temperatures of specific optical phonons under current injection in the negative differential conductance regime²⁴. However, the light emission spectra maintain the relatively sharp peak feature, which is quite different from the broad thermal radiation spectra of individual metallic SWCNTs observed at \approx 2000 K in our previous study⁴. The photon chemical potential of $\approx 0.4 \text{ eV}$ was evaluated using Eq. (2) (inset in Fig. 4c). This contrasts with the semiconducting SWCNTs under CW-laser irradiation for which the chemical potential value was only 5% of the emission photon energy at 1000 K. In metallic SWCNTs under the current injection conditions, a process similar to thermal exciton generation, induced by multiphonon absorption, as observed in semiconducting SWCNTs⁵, is expected to occur in the massive 1D sub-bands of metallic SWCNTs by absorbing nonequilibrium optical phonons. Therefore, adhering to the tradition in luminescence nomenclature, this distinct HTSS luminescence may aptly be termed "phono-luminescence" rather than simply being categorized as electroluminescence.

Comparison of the light emission states

In Fig. 5, we summarize the observed light emission conditions of semiconducting and metallic SWCNTs as a function of temperature and the chemical potential of photons normalized to the peak (approximately mean) photon energy $\mu/\hbar\omega$. The $\mu/\hbar\omega$ value corresponds to the fraction of free energy per photon, and the remaining $(1 - \mu/\hbar\omega)$ corresponds to the fraction of the energy accompanying entropy carried away from the SWCNTs by radiation. Regarding the semiconducting SWCNT under CW



Fig. 4 | Light emission of high-temperature metallic carbon nanotubes. a, b Light emission spectra of the field-effect transistor device featuring the (24,15) metallic single-walled carbon nanotube (SWCNT) with a different drain-source voltage (a) and under CW laser irradiation (b). The inset in (a) shows the one-dimensional band structure of a metallic species, comprising the first (M_{11}) massive sub-bands alongside a linear band (E, energy; k, wavenumber vector). c Integrated light emission intensities as a function of the inverse ($1/k_B T$; bottom) and corresponding temperatures (top). The inset shows the temperature (T) dependence of the photon chemical potential (μ) emitted from the current-injected SWCNT.

laser irradiation (shown as circles), the luminescence observed at the temperature below 500 K is that commonly referred to as "cold" light because there is minimal thermal radiation in the same photon energy range at that temperature. The cold and the HTSS luminescence for the semiconducting SWCNT are almost aligned along a single line (orange-shaded line). This is because the number of excitons, which is approximately proportional to the number of emitted photons, remains nearly constant owing to efficient exciton-exciton annihilation³⁸ during luminescence conditions. Conversely, the HTSS luminescence of the metallic species under current injection deviates considerably from the orange-shaded line for the semiconducting SWCNT. This deviation suggests that metallic SWCNTs may accommodate more excitons (or electron-hole pairs) than their semiconducting counterparts. This difference could be attributed to the weaker Coulomb interaction strength in the metallic SWCNTs that could cause inefficient exciton-exciton annihilation. Furthermore, it was observed that the chemical potential marginally increased with temperature, presumably owing to the presence of highly nonequilibrium phonons under high-bias conditions. Even at ≈1000 K, the chemical potential is more than that for the luminescence of the semiconducting one below 600 K, which indicate their potential for applications requiring intense NIR light at high temperatures. Thus, classifying light emission conditions in SWCNTs based on their chemical potential provides a comprehensive understanding of their high temperature luminescence that cannot be regarded as cold light anymore, but rather can be called as "noncold" luminescence.

Methods

Synthesis and characterization of carbon nanotubes

SWCNTs were synthesized and suspended over open slits measuring 20-30 µm in width and cut into substrates (Fig. 2b) using an ambient chemical vapor deposition technique involving a modified fast-heating process47. The determination of SWCNTs' semiconducting or metallic properties relied on their chiral structures, which were identified using an empirical table correlating chiral structures with optical exciton resonances⁴⁸. These resonances were investigated via elastic scattering (Rayleigh) spectroscopy⁴⁸⁻⁵¹, where scattering was particularly pronounced at exciton resonances. Our study utilized Rayleigh spectroscopy across a wide photon energy range of 0.8 - 2.8 eV, encompassing many exciton resonance peaks necessary for chiral structure determination^{4,52,53}. The supercontinuum source (Fianium, WL-SC-400-PP-4) emitted broadband light, which was directed onto the SWCNTs through an objective lens with a numerical aperture (N.A.) of 0.42. The power integrated into the system was approximately 2 mW (0.56-2.8 eV). The scattered light was then captured by another objective lens with a 0.42 N.A. and detected using either a with a thermoelectrically monochromator equipped cooled indium-gallium-arsenide two-dimensional photodiode array (Princeton Instruments, NIRvana) or one featuring a thermoelectrically cooled chargecoupled device camera (Princeton Instruments, ProEM). The Rayleigh scattering cross-section was proportional to $\omega^3 |\chi(\omega)|^2$, where ω and $\chi(\omega)$ are the optical frequency and susceptibility, respectively⁵⁰. All Rayleigh spectra in Figs. 2d, e, and 3e, and Supplementary Fig. 3c, d were corrected for the ω^3 scattering efficiency factor to show the optical susceptibility.

Field-effect transistor device of SWCNT

The silicon substrate contained an open slit with Pt electrodes positioned at both the ends (Fig. 2b). Electrostatic doping was facilitated by directly applying gate bias to the silicon substrate (Fig. 3e). For current injection into a metallic SWCNT, the drain–source voltage was applied to the Pt electrodes, while a gate voltage of -5 V was maintained. The detailed device specifications can be found in ref. 35. After device wiring, surplus SWCNTs were selectively removed using a laser, leaving only the desired SWCNT.

Integrated radiation intensities of light emission of semiconducting SWCNT

As illustrated in Supplementary Fig. 1, all spectra were successfully modeled using the Lorentz function. The integrated radiation intensities were subsequently derived from the area under the Lorentz function obtained during fitting.



Fig. 5 | Summary of the light emission of carbon nanotubes. Temperature dependence of the photon chemical potential (μ) of the light emission of the semiconducting (circles) and metallic (squares) carbon nanotube normalized by the photon energy ($\hbar\omega$). HTSS, high-temperature steady-state. The orange-shaded area illustrates the temperature dependence of the $\mu/\hbar\omega$ under light irradiation of the semiconducting species, where the number of excitons is constrained to remain constant owing to exciton–exciton annihilation.

Temperature estimation

The suspended SWCNTs' temperature was estimated by integrating techniques from previous studies. The PL of semiconducting SWCNTs, exhibited a universal temperature-dependent peak shift (ΔE), regardless of the chiral structure³⁶. The peak energy shift adhered to Varshni's empirical formula, similar to that of other semiconductors⁵⁴. The following empirical law for the temperature (*T*)-dependent PL peak energy (*E*) was proposed³⁶:

$$E(T) = E_0 + \Delta E(T),$$

$$\Delta E(T) = -\frac{aT^2}{T+T_e},$$

where $a = 0.177 \text{ meV K}^{-1}$, $T_0 = 1800 \text{ K}$, and E_0 is the PL peak energy at 0 K that depends on the chiral structure. This equation was applied only to suspended SWCNTs without adsorbed molecules. E_0 was determined as 0.886 eV using the room-temperature PL peak energy of 0.878 eV for the (12,5) SWCNT without adsorbed molecules (taken from ref. 37).

During experiments involving metallic SWCNTs under CW laser irradiation, the CW laser facilitated heating and temperature assessment via Raman spectroscopy of the in-plane carbon stretching mode (*G*-mode; Supplementary Fig. 4)⁴. The *G*-mode frequency of carbon nanotubes exhibited a consistent temperature dependence irrespective of sample type or laser excitation wavelength⁵⁵. Supplementary Fig. 4 illustrates the *G*-mode features in the Raman spectra of the metallic (24,15) SWCNTs under various CW laser intensities. Raman spectra at 300 K (bottom panel) were acquired in a nitrogen atmosphere to suppress laser heating effects through convective heat transfer. Conversely, spectra obtained in vacuum aimed to induce heating. At 300 K, Raman spectra displayed two peak components attributed to longitudinal optical (LO) and transverse optical (TO) modes at lower frequency⁵⁶. This spectral shape was fitted as follows:

$$I(\omega) = I_1 \frac{\left[1 + 2(\omega - \omega_1)/q\Gamma_1\right]^2}{1 + \left[2(\omega - \omega_1)/\Gamma_1\right]^2} + I_2 \frac{1}{1 + \left[2(\omega - \omega_2)/\Gamma_2\right]^2}$$

where ω represents the frequency; $I_{1(2)}$, $\omega_{1(2)}$, and $\Gamma_{1(2)}$ denote the line strength, phonon frequencies, and full widths at half maximum for the two

peaks, respectively; and the parameter q signifies the asymmetry in a Breit–Wigner–Fano line shape. The first and second terms on the right side correspond to the LO and TO modes⁵⁶, respectively, depicted by the dotted curves. An increasing excitation intensity led to a redshift in the *G*-mode frequency, as observed in Supplementary Fig. 4. The temperature was determined from the peak frequency shift of the LO mode at 300 K, employing the empirical law proposed in ref. 55. This empirical law for the temperature (*T*)-dependent *G*-mode frequency, $\omega(T)$, is expressed as follows:

$$\omega(T) = \omega_0 - \frac{A}{\exp(B\hbar\omega_0/k_{\rm B}T) - 1}$$

where ω_0 , \hbar , and k_B are the zero-temperature *G*-mode frequency, reduced Planck constant, and Boltzmann constant, respectively. According to ref. 55, parameters *A* and *B* are defined as $A = 38.4 \text{ cm}^{-1}$ and $B = 0.438 \text{ cm}^{-1}$, respectively. ω_0 was determined as $\omega_0 = 1583.03 \text{ cm}^{-1}$, such that $\omega(300 \text{ K})$ was consistent with the room-temperature result.

For the metallic SWCNTs under current injection, similar to semiconducting SWCNTs, temperature determination relied on the peak energies of emitted light. Optical transition energies between sub-bands exhibited a continuous decrease with temperature, mirroring the monotonic decline in PL energy corresponding to the band gap, following the Varshni formula (e.g., second sub-band exciton of a semiconducting SWCNT⁴). For the metallic (24,15) SWCNT, we obtained the resonance energy of the massive first sub-band (M_{11}) exciton transition at 300 K from the Rayleigh scattering spectrum and those at 1030 K–1240 K using a combination of thermal radiation and the abovementioned Raman spectroscopy. Temperature estimation relied on the nearly linear relationship previously observed between peak energy and temperature³⁶.

Estimation of the integrated emissivity from the Rayleigh scattering spectra in the gate voltage dependence measurement

The Rayleigh scattering cross-section was directly proportional to $\omega^3 |\chi(\omega)|^2$, where ω and $\chi(\omega)$ are the optical frequency and susceptibility, respectively⁵⁰. The relationship between $\chi(\omega)$ and the dielectric function $\varepsilon(\omega)$ is defined as $\varepsilon(\omega) = 1 + \chi(\omega)$. $\varepsilon(\omega)$ provides the emissivity $e(\omega)$ via $e(\omega) \simeq \omega d(3c)^{-1} \text{Im}[\varepsilon(\omega)]$. The integrated emissivity A_e is given by $A_e \propto \int \omega^3 e(\omega) d\omega$.

Data availability

All relevant data are available from the corresponding authors upon reasonable request.

Code availability

The code used in this study is available from the corresponding author upon reasonable request.

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Author contributions

T.N. and Y.M. conceived the concept and Y.M. directed the project. A.T. synthesized the nanotubes. T.N. arranged and carried out all optical experiments. T.N., S.K., and Y.M. considered the mechanism. All the authors contributed to writing the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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