ADVANCED OPTICAL MATERIALS

Supporting Information

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Efficient White Light Emission from Ga/Ga_2O_3 Hybrid Nanoparticles

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Supplementary Note 1: Derivation of quantum efficiency

The method used to estimate the quantum efficiency of the hot-electron intraband lumindescence from Ga nanoparticles (NPs) was the same as that described in our previous work (Ref. 27). In Figure N1a,b, we show the optical paths of the excitation laser light and the generated PL in our experimental setups. Ga NPs are located on an Au/SiO_2 substrate. The femtosecond laser light is reflected by a dichroic mirror and focused on the Ga NPs by the objective lens, as shown in Figure N1a. The reflected laser light from the Ga NPs on the Au/SiO_2 substrate is attenuated by the dichroic mirror and a stop-band filter with an optical density of $OD \sim 2.3 \times 10^{-6}$. If we denote the attenuation coefficient of the whole optical system (except the dichroic mirror + filter) as α and the gain and quantum effiency of the charge coupled device (CCD) as g_{ex} and η_{ex} (~0.75 at 760 nm), then the total number of photons detected by the CCD, i.e., N_2^{ex}, can be expressd as follows:

$$N_1^{ex} * r * OD * \alpha * g_{ex} * \eta_{ex} = N_2^{ex}, \quad (1)$$

where N_1^{ex} represents the total number of photons irradiating on the Ga NPs and r demotes the corresponding reflectivity (see Figure N1a).

When we examined the dependence of the reflected laser light intensity on the excitation irradiance, a linear relationship is observed at small excitation irradiances, as shown in the main text Figures 3c-d. When the excitation irradiance exceeds a critical value, a deviation from the linear relationship occurs, implying that the nonlinear absorption of the Ga NPs becomes effective (see main text Figures 3c-d). The number of photons absorbed by the Ga NPs (N_1^{abs}) due to nonlinear processes can be deduced by the number of reduced photons observed in the reflected laser light (N_2^{abs}) by using the following relationship:

$$N_1^{abs} * r * OD * \alpha * g_{ex} * \eta_{ex} = N_2^{abs}. \quad (2)$$

Referring to Figure N1b, we can get the relationship between the number of detected photons (N_2^{pl}) in the luminescence and the number of emitted photons from the Ga NPs (N_1^{pl}), which is given in the following:

$$N_1^{pl} * \beta * \alpha * g_{pl} * \eta_{pl} = N_2^{pl}. \quad (3)$$

Here, the collection efficiency of the objective lens (β) is taken into account. For Ga NPs located on the Au/SiO_2 substrate, the average collection efficiency was found to be $\beta \sim 0.50$.

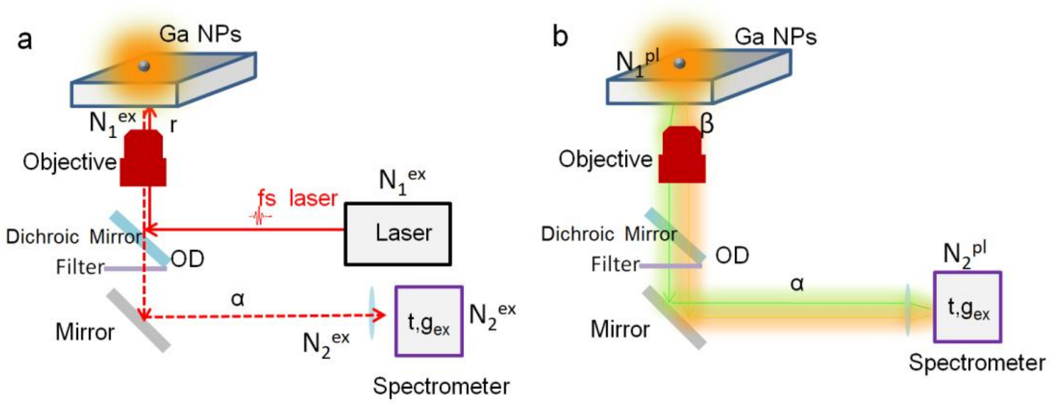
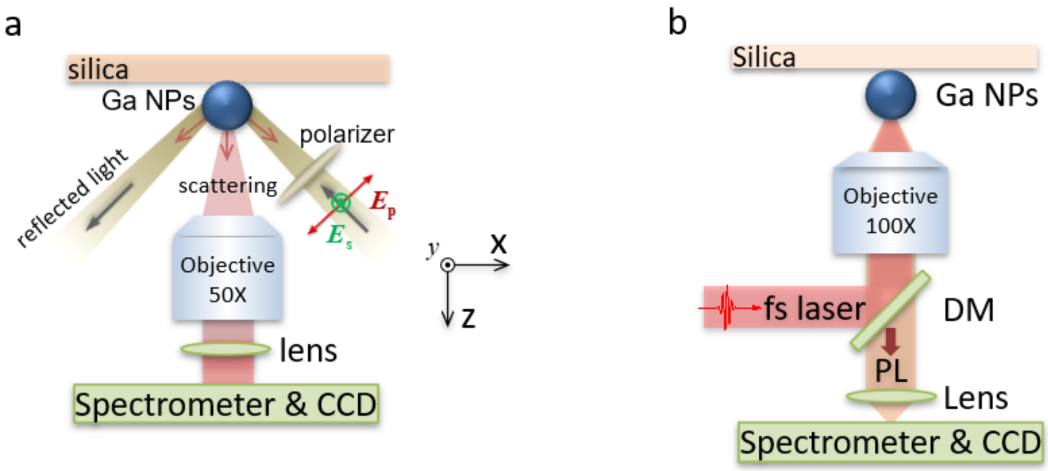


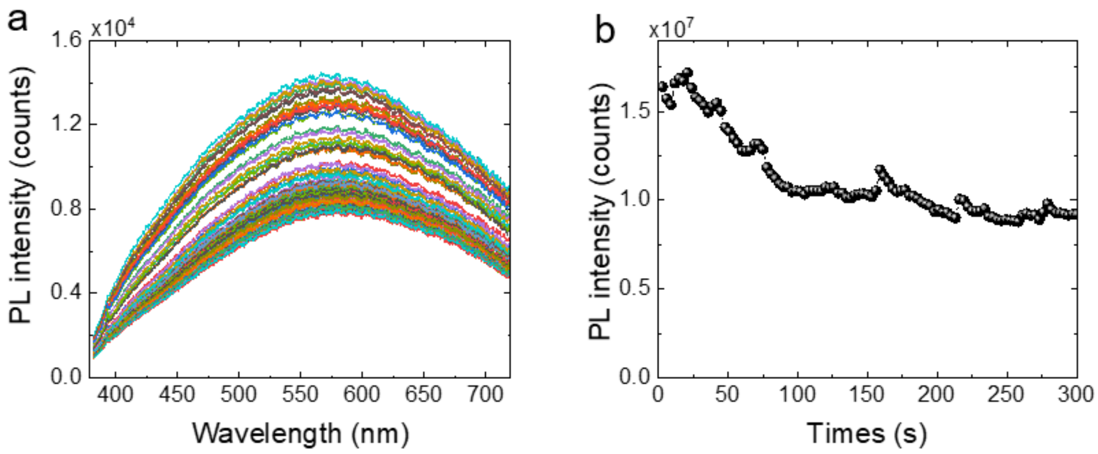
Figure N1. Schematics showing the optical paths of the excitation laser light (a) and the PL emitted from a Ga NP (b) in the experiment setup used to estimate the quantum efficiency of the Ga NP.

With the information on the number of photons absorbed by the Ga NPs (N_1^{abs}) and that emitted from the Ga NPs (N_1^{pl}), one can derive the quantum efficiency of the hot-electron intraband luminescence (i.e., Eq(3) divided by Eq(2)), which is expressed as follows:

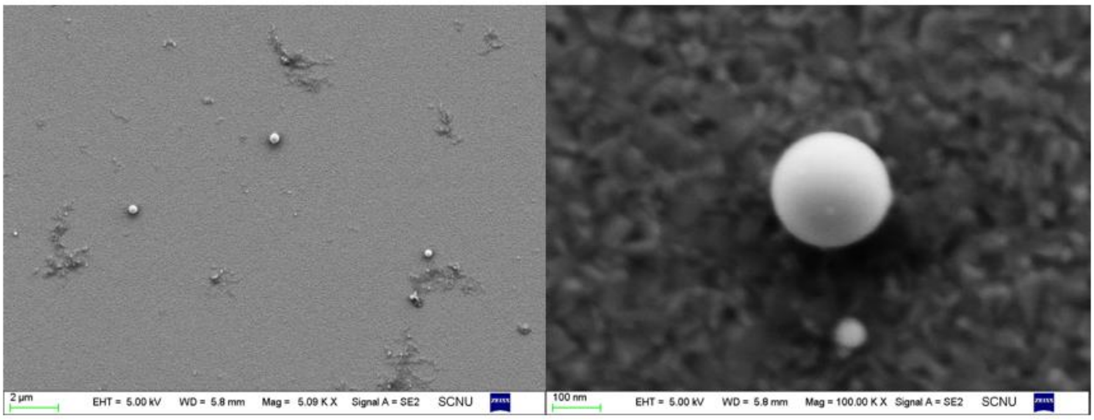
$$Q = N_1^{pl}/N_1^{abs} = N_2^{pl} * r * OD * \eta_{ex}/(N_2^{abs} * \beta * \eta_{pl}). \quad (4)$$



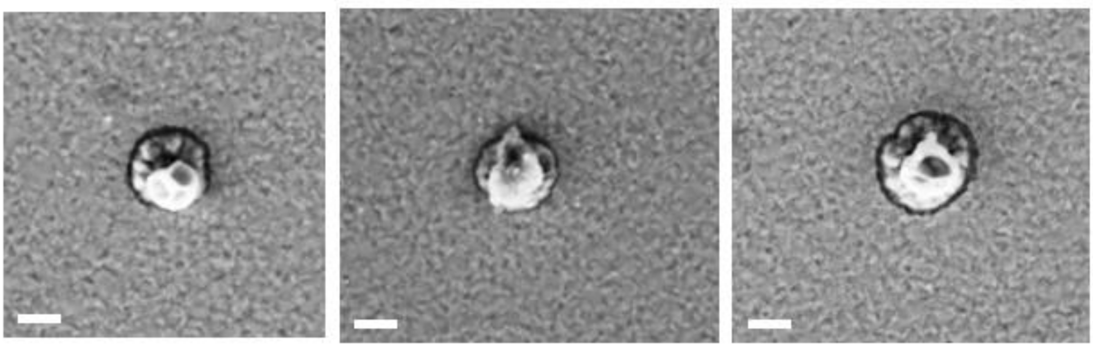
Supplementary Figure S1. Experimental setups used to characterize the (a) linear and (b) nonlinear optical responses of Ga and Ga/Ga_2O_3 NPs.



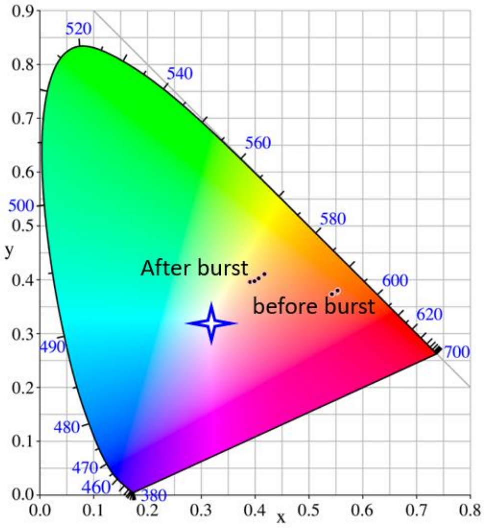
Supplementary Figure S2. (a) PL spectra of a Ga/Ga_2O_3 NP measured at different excitation times. (b) Evolution of the integrated PL intensity of the Ga/Ga_2O_3 NP with excitation time. The excitation irradiance was fixed at 0.22 mJ/cm^2.



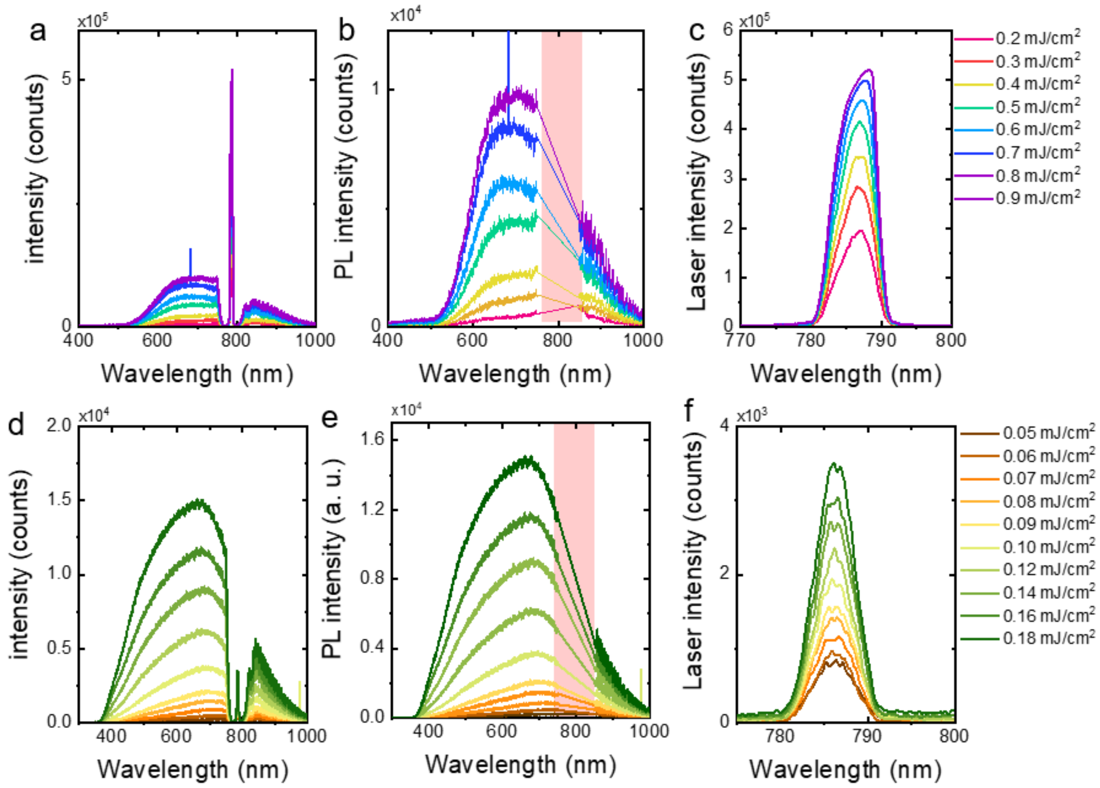
Supplementary Figure S3. (a) SEM image of Ga NPs fabricated by using femtosecond laser ablation. (b) SEM image of a spherical Ga NP with a diameters of ~200 nm.



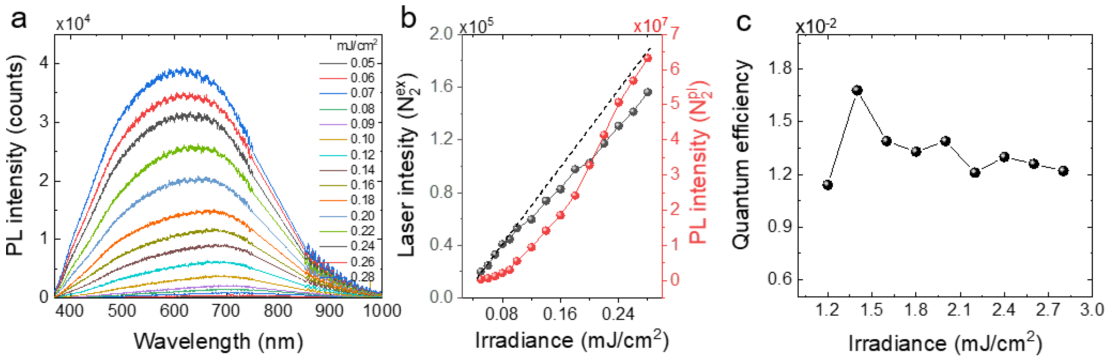
Supplementary Figure S4. SEM images of three Ga NPs after the luminescence burst (i.e., Ga/Ga_2O_3 NPs). The length of scale bars in all cases is 200 nm.



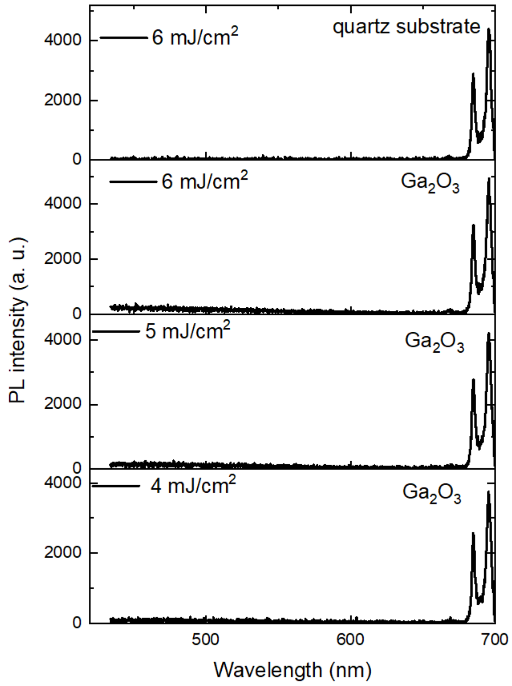
Supplementary Figure S5. Color indices derived from the PL spectra of Ga NPs based on CIE 1931. The star at the center represents an ideal white light source.



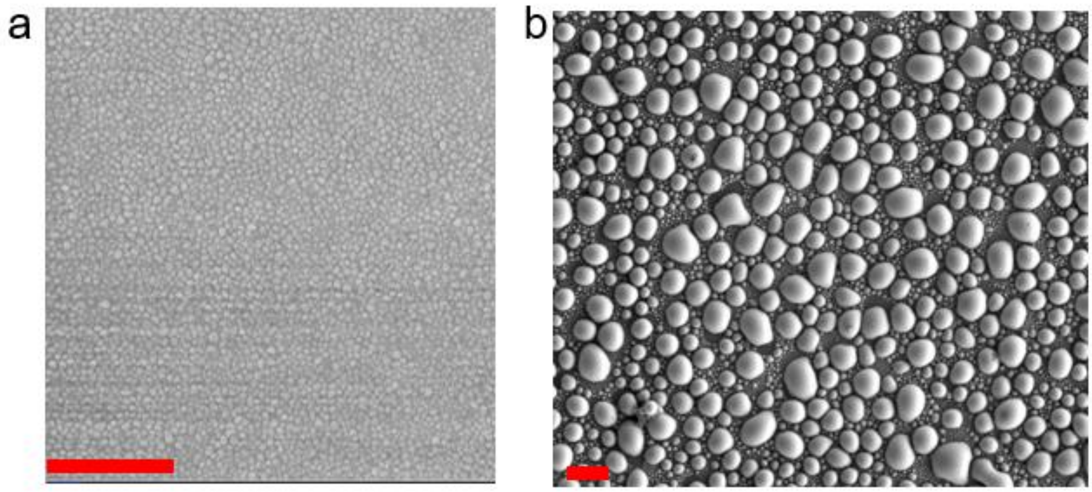
Supplementary Figure S6. PL spectra measured for a Ga NP before (a-c) and after (d-f) the luminescence burst. (a,d) Original PL spectra of the Ga NP obtained at different irradiances. (b,e) Reconstructed PL spectra of the Ga NP used for estimating the quantum efficiency. (c,f) Spectra of the reflected laser light obtained at different excitation irradiances.



Supplementary Figure S7. (a) PL spectra measured for a Ga/Ga_2O_3 NP at different excitation irradiances. (b) Dependences of the integrated intensities of the PL and the reflected laser light on the excitation irradiance. (c) Dependence of the quantum efficiency of the Ga/Ga_2O_3 NP on the excitation irradiance.



Supplementary Figure S8. PL spectra measured for a Ga_2O_3 microparticle placed on a quartz substrate by using femtosecond laser pulses with different excitation irradiances. The excitation wavelength was chosen to be 785 nm.



Supplementary Figure S9. SEM images of closely-packed Ga nanoislands deposited on SiO_2 substrates by using different sputtering times of (a) 15 s and (b) 60 s. The length of the scale bars in both cases is 1.0 µm.