Supplementary Information for

New up-conversion charging concept for effectively charging persistent phosphors using low-energy visible-light laser diodes

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Brief summary − This file contains: (1) the NIR imaging in $LiGa_5O_8:Cr^{3+}$ nanoparticles colloidal solution; (2) the calculation of safe exposure limits of a 635 nm laser diode to the skins; (3) one supplemental table; (4) seven supplemental figures.

1. NIR imaging in $LiGa_5O_8:Cr^{3+}$ nanoparticles colloidal solution

The $LiGa_5O_8:Cr^{3+}$ nanoparticles were dispersed in deionized water to form a colloidal solution. After the colloid was irradiated by a 635 nm laser diode at 600 mW for 3 s (Fig. S5a), the UCC-induced persistent luminescence from the $LiGa_5O_8:Cr^{3+}$ nanoparticles in the colloid was imaged using a Pentax digital SLR camera that was connected to the eyepiece of an ITT PVS-14 Generation III night-vision monocular (Fig. S5b). The imaging experiment was conducted in a dark room.

2. Calculation of safe exposure limits of a 635 nm laser diode to the skins

For using lasers or laser diodes in medical research and applications, limits of exposure to laser radiation were established by the International Commission on Non-Ionizing Radiation Protection (ICNIRP) to avoid causing adverse biological effects to the eyes and the skins[S1]. The laser exposure limits depend on the laser output power and the exposure duration. According to the method provided in the ICNIRP guideline, we calculated the safety exposure limits of a power tunable (0–1000 mW) 635 nm laser diode to the skin.

The 635 nm laser diode falls in the visible to short wavelength infrared radiation region (400–400 nm); therefore, the following formulas are used:

For 100 ns $\leq t \leq$ 10 s, the exposure limit is $11C_A\, t^{0.25}$ kJ·m^{-2}.

For 10 s $\leq t \leq$ 30,000 s, the exposure limit is $2.0C_A$ kW·m^{-2}.

Here, t is the exposure duration; C_A is the correction factor, which is related to the wavelength dependence of the pigment epithelium absorption in skins. For a 635 nm laser diode, $C_A = 1.0$.

Using these two formulas, the safe exposure limits of a 635 nm laser diode to the skins were calculated as shown in Supplementary Table S1. The calculation shows that for a 635 nm laser diode the safe laser output powers to the skins are ≤100 mW; when the powers are ≤20 mW, the irradiations are safe for any long exposure durations, while in the range of 20–100 mW, the irradiations are safe only for limited exposure durations (~1–10 s). We then used the thus obtained exposure limits to excite the $LiGa_5O_8:Cr^{3+}$ persistent phosphor, measured the persistent luminescence intensities, and drew the 3-D graph of persistent luminescence intensities versus safe exposure limits (excitation powers and excitation durations) in Fig. 5a.

Reference

S1. International Commission on Non-Ionizing Radiation Protection. ICNIRP Guidelines on Limits of Exposure to Laser Radiation of Wavelengths between 180 nm and 1,000 μm. *Heal. Phys.* **105**, 271-295 (2013).

Table S1. Safe exposure limits of a 635 nm laser diode to the skin

Exposure Duration (s)	Power density[(a)] (W/cm^2 or J/cm^2)	Output Power (mW)
1	1.1	110
2	0.65	65
3	0.49	49
4	0.39	39
5	0.33	33
6	0.29	29
7	0.26	26
8	0.23	23
9	0.21	21
10 to 30,000	0.2	20

(a) The beam size of the laser diode is about 10 mm^2.

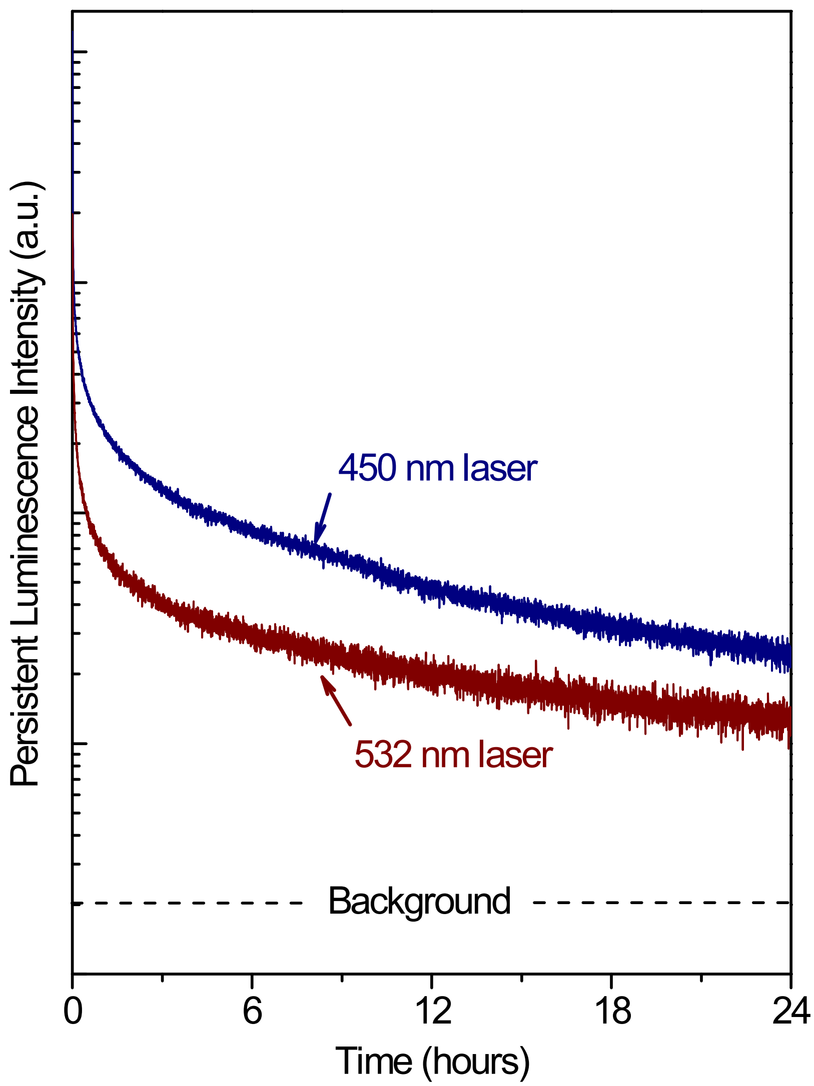

Fig. S1. UCC-induced persistent luminescence decay curves of $LiGa_5O_8:Cr^{3+}$ persistent phosphor after charged by 450 nm and 532 nm laser diodes. The excitation condition for 450 nm laser diode is 1000 mW for 10 s. The excitation condition for 532 nm laser diode is 200 mW for 10 s. The decay curves were monitored at Cr^{3+} 716 nm emission.

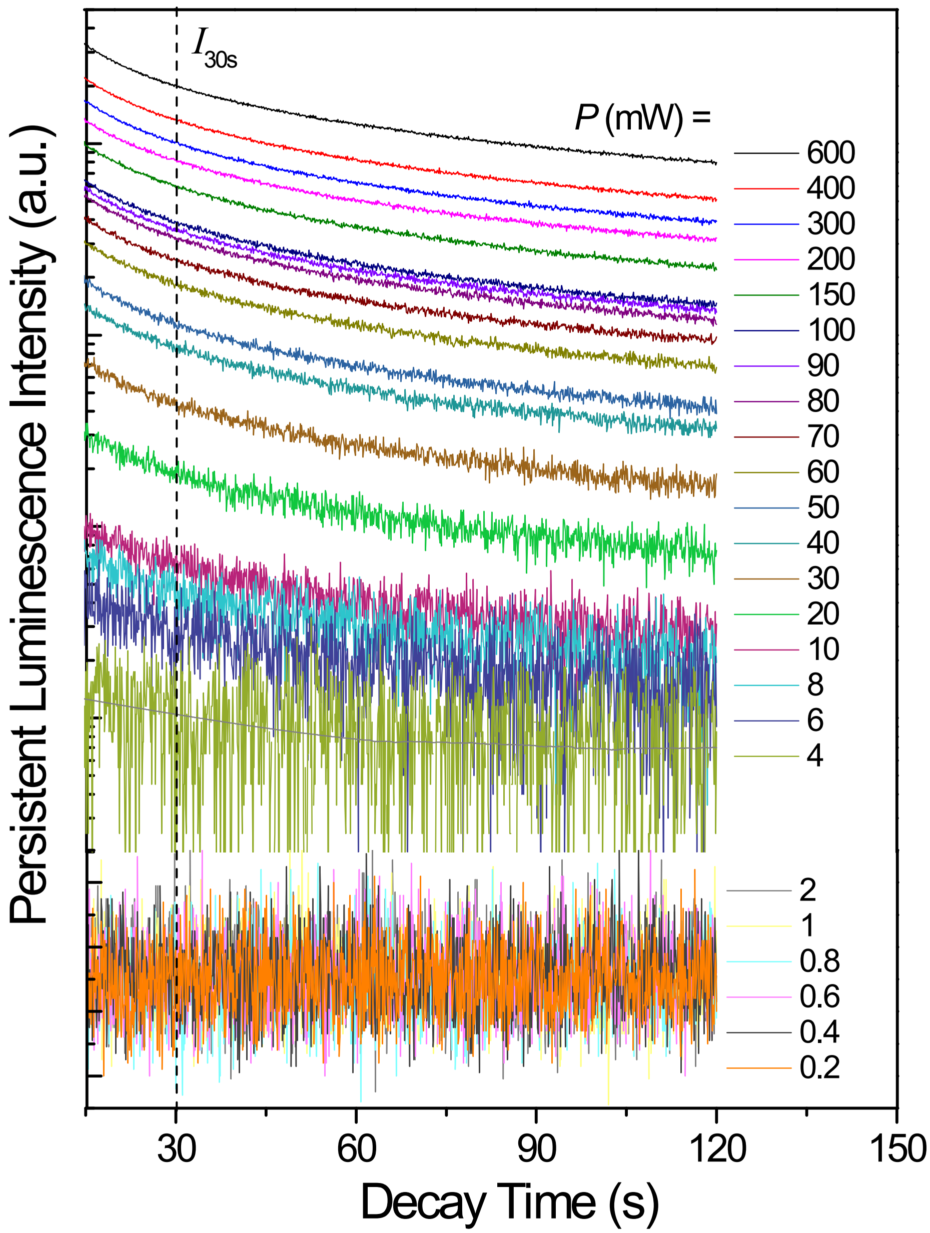

Fig. S2. UCC-induced persistent luminescence decay curves of $LiGa_5O_8:Cr^{3+}$ persistent phosphor after charged by a 635 nm laser diode. The laser output power varies from 0.2 mW to 600 mW. The excitation duration for each excitation is 10 s. The monitoring wavelength is Cr^{3+} 716 nm emission. UCC-induced persistent luminescence is detected when the laser output power is higher than 2 mW (corresponding to a power density of ~20 mW/cm^2), and the persistent luminescence intensity increases as the laser power increases from 2 mW to 600 mW. The persistent luminescence intensity at time of 30 s after the stoppage of each irradiation (I_{30s}; indicated by the vertical dash line) is used to plot the persistent luminescence intensity as a function of laser output power shown in Fig. 3a of the main text.

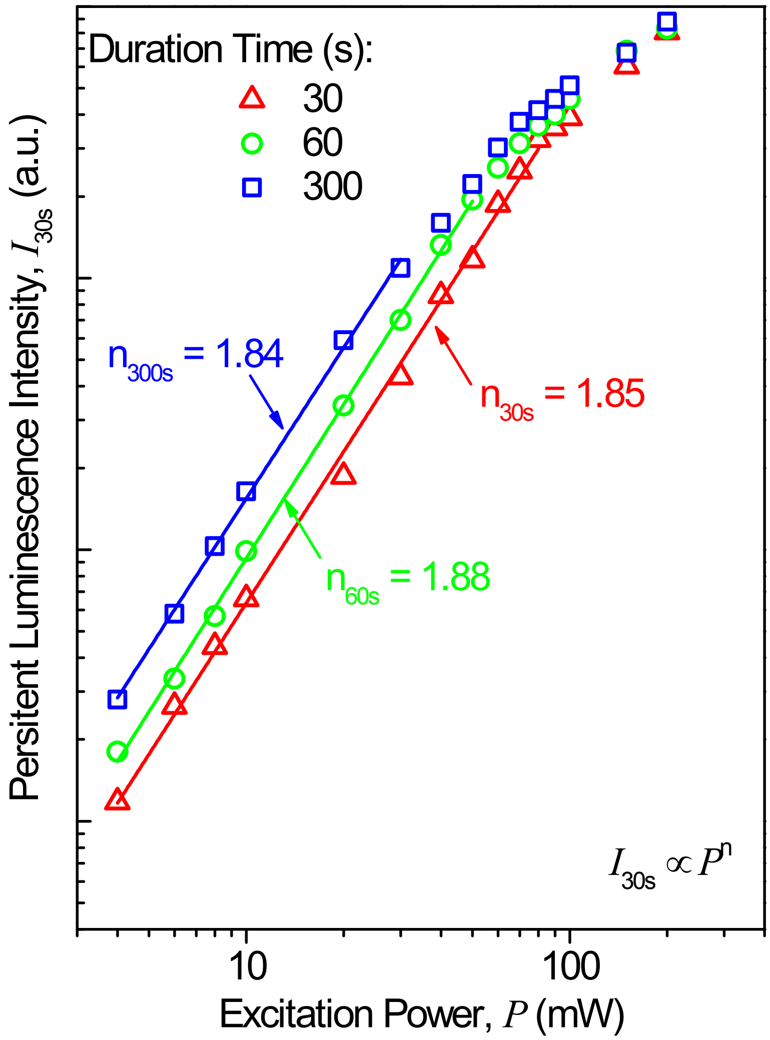

Fig. S3. Excitation power dependent UCC persistent luminescence intensity in $LiGa_5O_8:Cr^{3+}$ persistent phosphor. The phosphor was charged by a 635 nm laser diode at varied output power (0.2–300 mW) for 30 s, 60 s, and 300 s. The plots were obtained using the same method as that used in Fig. 3a of the main text. That is, for each excitation duration (30 s, 60 s, or 300 s), the phosphor was charged with laser output power tuning from 0.2 mW to 300 mW. UCC-induced persistent luminescence decay curve monitoring at 716 nm was acquired after each excitation, and the persistent luminescence intensity at time of 30 s after the stoppage of each irradiation (I_{30s}) is used to plot the persistent luminescence intensity as a function of laser output power (P). The straight lines are the quadratic fittings of the plots. The slopes of the fitting lines are 1.85, 1.88, and 1.84 for 30 s, 60 s, and 300 s excitations, respectively, which are close to the value of 1.82 for 10 s excitation in Fig. 3(a) of the main text. This quadratic I–P relationship clearly reveals that two 635 nm photons are absorbed in order to pump the Cr^{3+} system to the high-energy delocalization state.

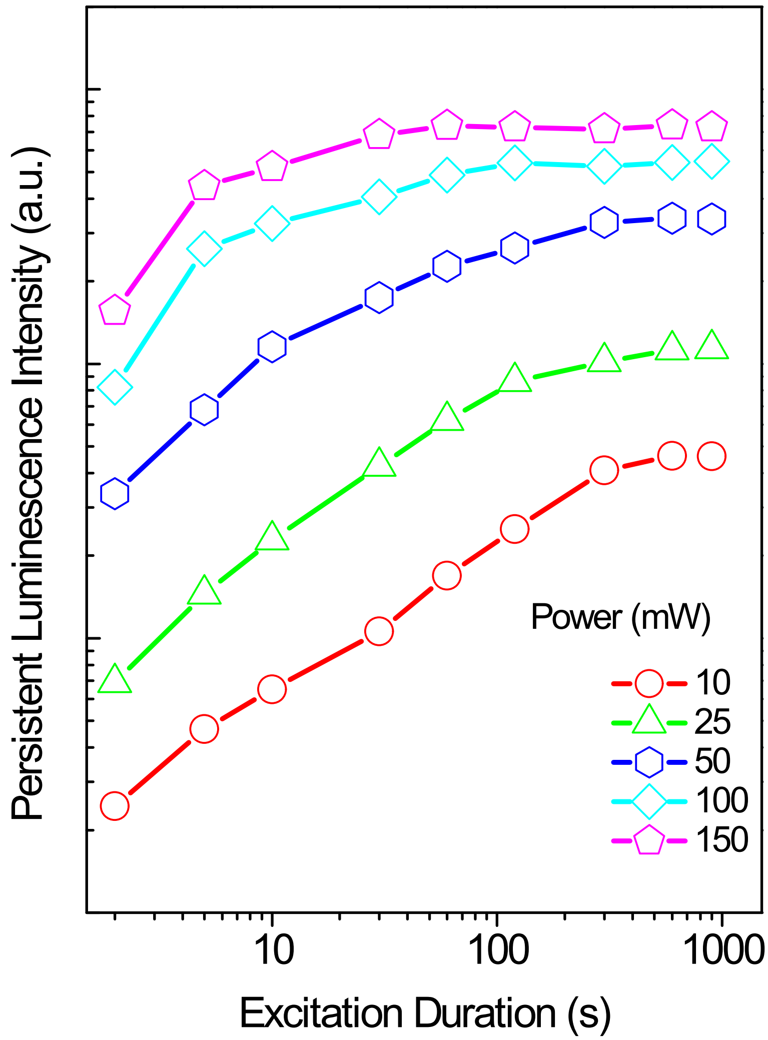

Fig. S4. Excitation duration dependent UCC persistent luminescence intensity in $LiGa_5O_8:Cr^{3+}$ persistent phosphor. The phosphor was charged by a 635 nm laser diode for 2–1000 s with laser output powers at 10 mW, 25 mW, 50 mW, 100 mW and 150 mW. After each excitation, UCC-induced persistent luminescence decay curve monitoring at 716 nm was acquired, and the persistent luminescence intensity at time of 30 s after ceasing the irradiation (I_{30s}) was used to plot the persistent luminescence intensity as a function of excitation duration.

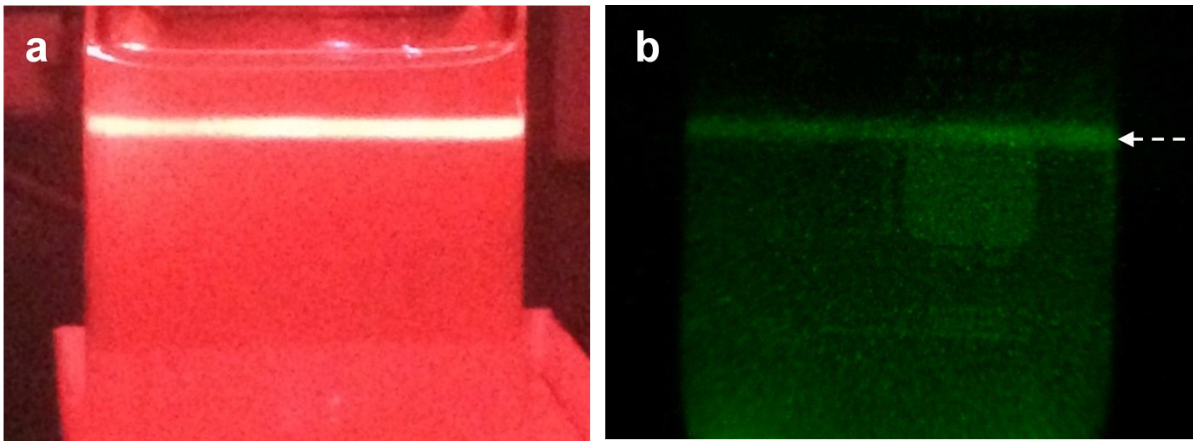

Fig. S5. UCC-induced persistent luminescence in $LiGa_5O_8:Cr^{3+}$ nanoparticles colloidal solution. **a**, Digital image showing a $LiGa_5O_8:Cr^{3+}$ nanoparticles colloidal solution being irradiate by a 635 nm laser diode at 600 mW. The irradiation duration is 3 s. The image was taken using a Pentax digital SLR camera. **b**, NIR persistent luminescence image taken using a Pentax digital SLR camera that is connected to the eyepiece of an ITT PVS-14 Generation III night-vision monocular at 1 min after ceasing the irradiation. The bright line (indicated by a white arrowhead) along the original laser beam line shows the UCC-induced persistent luminescence from the $LiGa_5O_8:Cr^{3+}$ nanoparticles in the colloid. The imaging experiment was conducted in a dark room.

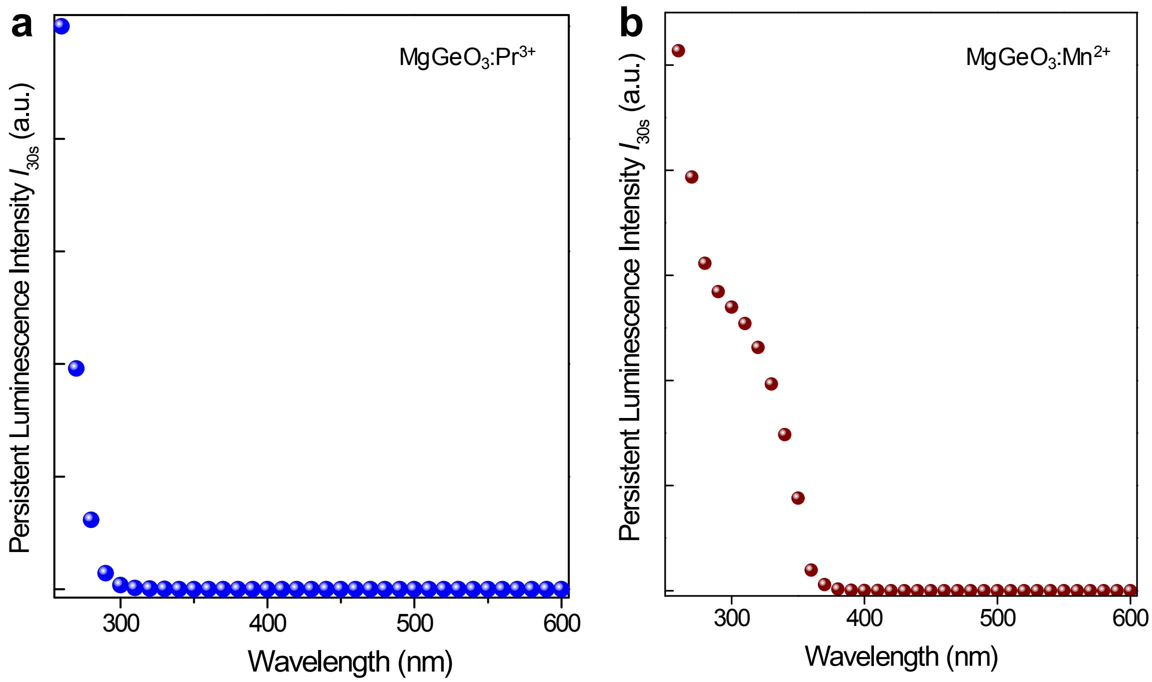

Fig. S6. Persistent luminescence excitation spectra obtained using a 450 W xenon lamp excitation. a, $MgGeO_3:Pr^{3+}$ persistent phosphor. **b**, $MgGeO_3:Mn^{2+}$persistent phosphor. The phosphors were irradiated with monochromatic light between 260–600 nm in 10 nm steps for 5 min. Decay curves were then recorded by monitoring at 624 nm for $MgGeO_3:Pr^{3+}$ or at 678 nm for $MgGeO_3:Mn^{2+}$ after each irradiation. The persistent luminescence intensity at time of 30 s after ceasing the irradiation (I_{30s}) was used to plot the persistent luminescence intensity as a function of the excitation wavelengths, i.e., the persistent luminescence excitation spectrum. The results show that, with the excitation of a xenon lamp, the persistent luminescence of $MgGeO_3:Pr^{3+}$ and $MgGeO_3:Mn^{2+}$ persistent phosphors can only be effectively achieved by <300 nm and <370 nm UV light excitation, respectively.

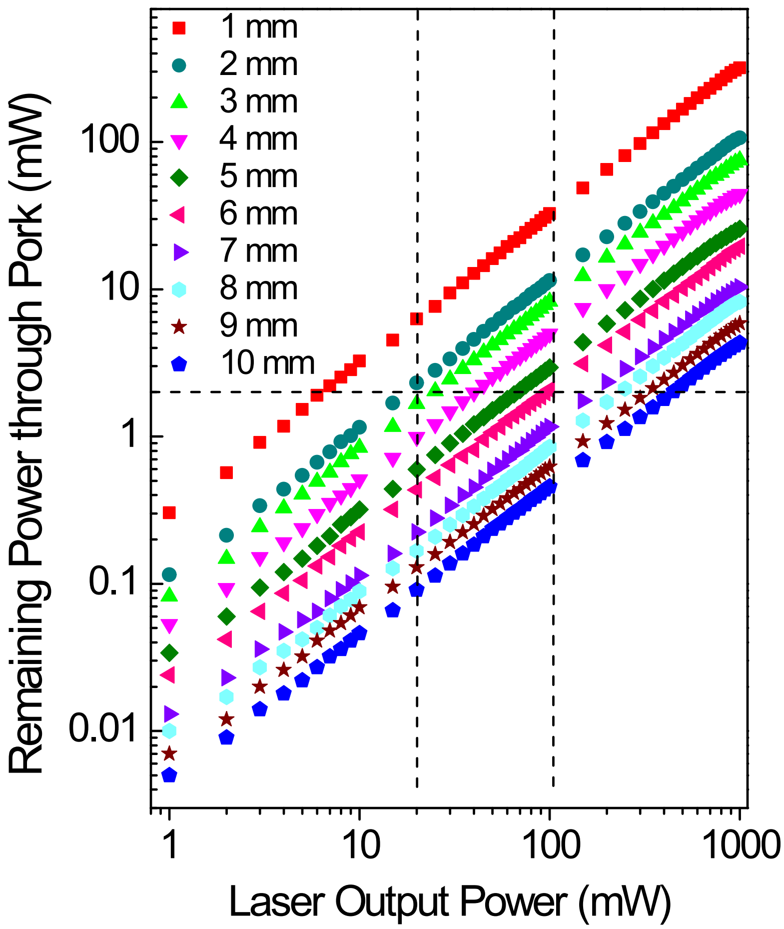

Fig. S7. Remaining laser power of a 635 nm laser diode after the laser beam penetrates pork tenderloin. The laser output power varied from 1 mW to 1000 mW. The thickness of pork tenderloin varied from 1 mm to 10 mm. The remaining power of the laser beam was measured using a Thorlabs PM100D power energy meter. The horizontal dash line represents the minimum 2 mW power for producing detectable UCC persistent luminescence in $LiGa_5O_8:Cr^{3+}$ phosphor, and the two vertical dash lines represent 20 mW and 100 mW laser output powers. The measurement shows that at the output powers of 20 mW and 100 mW, the remaining powers of a 635 nm laser diode are high enough (i.e., >2 mW) to charge $LiGa_5O_8:Cr^{3+}$ phosphor located at up to 3 mm and 6 mm, respectively, deep within pork.