Electronic Supplementary Information (ESI) for Nanoscale

Plasmonic nanoparticles embedded in single crystals synthesized by gold ion implantation for enhanced optical nonlinearity and efficient Q-switched lasing

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Nonlinear refraction properties

Figure 2S demonstrates the closed-aperture (CA) Z-scan results of both samples and pure Nd:YAG crystal under different pulse energies, which display valley-peak configurations. These results appearing the self-focusing effect, which mainly result from the distortion of electron cloud distribution about an atom or molecule under femtosecond laser irradiation. The nonlinear refractive index, n₂, of these samples can be obtained by fitting CA Z-scan data with the following analytic formula¹

$$T_{Norm}(z) = 1 / \{ [1 - 4x\Delta\phi_0 / (1 + x^2)^2] + [4\Delta\phi_0^2 / (1 + x^2)^3] \}$$

where $\Delta \phi_0 = k \cdot \Delta n \cdot L_{eff}$, $z_0 = k \omega_0^2 / 2$, $x = z / z_0$. Table 1 lists the deduced n₂ values of all samples with different pulse energies. It should be noted that the nonlinear refractive indexes of embedded NPs crystals (i.e., samples A and B) are enhanced by nearly 4 orders of magnitude compared to that of original crystal, which is due to the SPR effect of Au NPs. In addition, as the pulse energies increase, n₂ for different samples have a tendency of rising, respectively.

Pulse energies	Sample A	Sample B	Pure Nd:YAG
50 nJ	$1.28 \times 10^{-12} \text{ cm}^2/\text{W}$	$1.72 \times 10^{-12} \text{ cm}^2/\text{W}$	$2.95 \times 10^{-12} \text{ cm}^2/\text{W}$
100 nJ	$1.31 \times 10^{-12} \text{ cm}^2/\text{W}$	$1.74 \times 10^{-12} \text{ cm}^2/\text{W}$	$3.16 \times 10^{-12} \text{ cm}^2/\text{W}$
150 nJ	$1.43 \times 10^{-12} \text{ cm}^2/\text{W}$	$1.75 \times 10^{-12} \text{ cm}^2/\text{W}$	$3.44 \times 10^{-12} \text{ cm}^2/\text{W}$

Table 1 n₂ of all samples under different pulse energies



Fig 1S The closed-aperture Z-scan results of (a) sample A, (b) sample B, (c) pure Nd:YAG crystal excited by femtosecond laser at the wavelength of 515 nm.

1. M. Sheikbahae, A. A. Said, T. H. Wei, D. J. Hagan and E. W. Vanstryland, *IEEE J. Quantum Electron.*, 1990, **26**, 760-769.