Electronic Supplementary Information

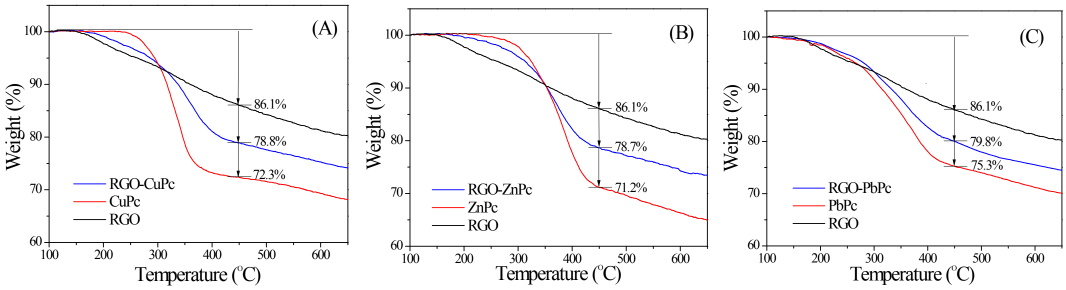

Fig. S1 TGA curves of (a) RGO-CuPc, RGO and CuPc, (b) RGO-ZnPc, RGO and ZnPc, and (c) RGO-PbPc, RGO and PbPc.

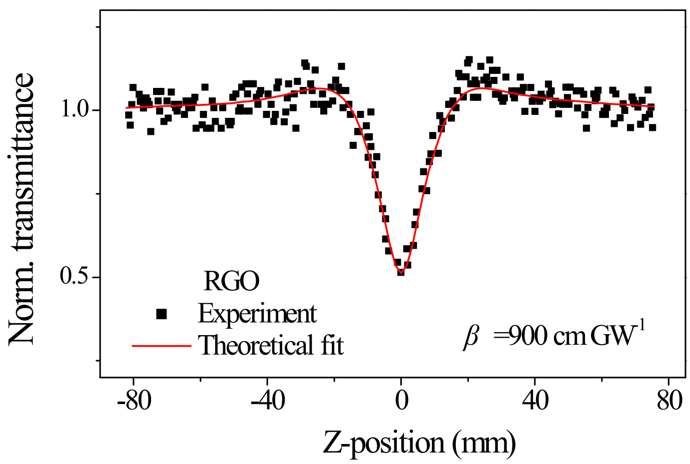

Fig. S2 Open aperture Z-scan curves of the reference RGO.

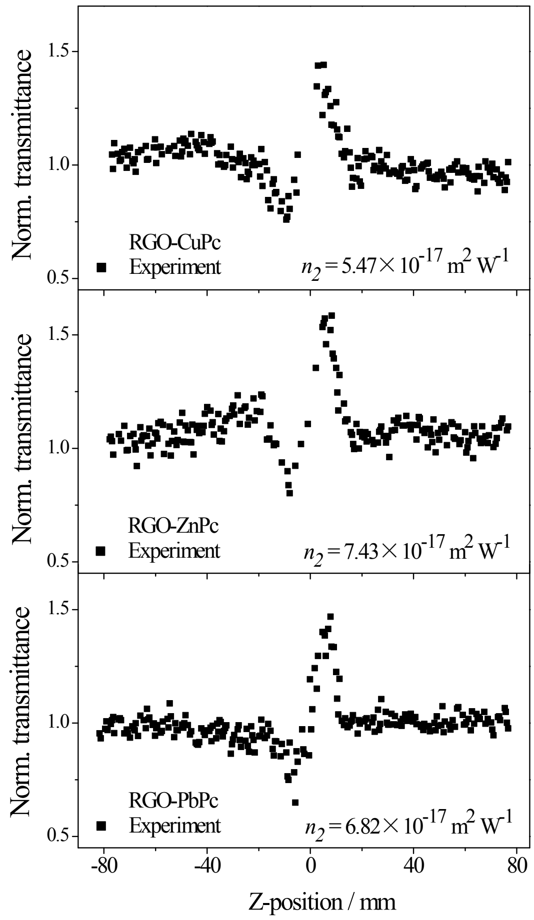

Fig. S3 Closed aperture Z-scan curves of RGO-CuPc, RGO-ZnPc and RGO-PbPc.

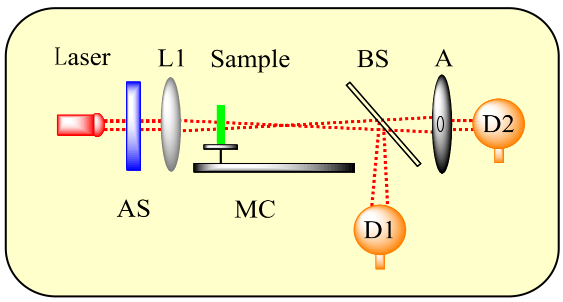

Fig. S4 The experimental apparatus for the Z-scan measurement. AS is adjusted system; L1, lens; MC, motion controller; D1 and D2, detectors; BS, beam splitter; A, aperture.

Z-scan analysis

In Z-scan measurement systems, the input laser beam can be considered as TEM_{00} Guassian beam as given as:

$$I_i = \frac{I_0 \exp\left[-2\left(r/\omega\right)^2 - \left(t/t_0\right)^2\right]}{1+\left(z/z_0\right)^2} \qquad (1)$$

where $z_0 = \pi\omega_0^2/\lambda$, ω_0 is the beam waist (50 μm in this measurement system), r is the distance from the axis of the beam, and ω is the radius of the beam at the z-position. The exit laser intensity from the sample may be obtained as:

$$I_t = I_i(r,t,z)e^{-\alpha L} \qquad (2)$$

where L is the thickness of the sample, z is the sample position and α is the absorption coefficient.

In the determination of the nonlinear absorption coefficient β of the samples taking into account the observed bleaching of the sample transmission occurring at lower pump intensity region, the corresponding Z-scan recordings were fitted by using the intensity variation equation and adopting an intensity-dependent absorption coefficient. The absorption coefficient α had the form $\alpha = \alpha_0/(1 + I/I_s) + \beta I$, where α_0 is the linear absorption coefficient, I_s is the saturation intensity, β is the nonlinear absorption coefficient and I is the laser intensity within the sample.

The phase variation can be expressed as:

$$\Delta\phi = \frac{kn_2 I_0 L_{eff}}{1+\left(z/z_0\right)^2}\exp\left(-2r^2/\omega^2\right) \qquad (3)$$

where $L_{eff} = [1-\exp(-\alpha_0 L)]/\alpha_0$. The electric field $E_s(r,t,z)$ at the exit surface of the sample is completely determined by Eqs. (2) and (3) [i.e. $E_s(r,t,z) \propto I_t^{1/2}\exp(i\Delta\phi)$]. In our experiment, the electric field E_a at the aperture is obtained by applying an H-F propagation integral:

$$E_a(r,t,z) = \frac{2\pi}{i\lambda(d-z)}\exp\left[\frac{i\pi r^2}{\lambda(d-z)}\right]\int_0^\infty r'dr' \\ \times E_s(r',t,z)\exp\left[\frac{i\pi r'^2}{\lambda(d-z)}\right]J_0\left[\frac{2\pi rr'}{\lambda(d-z)}\right] \tag{4}$$

where d is the distance between the aperture and the focal plane. The theoretical fits to the Z-scan data were calculated from the normalized transmittance equation:

$$T(z,S) = \frac{c\varepsilon_0 n_0 \pi \int_{-\infty}^{\infty} dt \int_0^{r_a} \left|E_a(r,t,z)\right|^2 rdr}{S\int_{-\infty}^{\infty} P_i(t)dt} \tag{5}$$

where $P_i(t) = (\pi\omega_0^2/2)I_0\exp[-(t/t_0)^2]$ is the instantaneous input power, $S = 1-\exp(-2r_a^2/\omega_a^2)$ is the aperture linear transmittance with r_a, ω_a donating the aperture radius and the beam radius at the aperture in the linear regime.

An effective NLO refractive index n_2 can be calculated from the difference between normalized transmittance values at valley and peak positions ($\triangle T_{v-p}$) using equation (6).

$$n_2 = \frac{\lambda\alpha_0}{0.812\pi I_0(1-s)^{0.25}(1-e^{-\alpha_0 L})}\Delta T_{v-p} \tag{6}$$

Where $\triangle T_{v-p}$ is the difference between normalized transmittance values at valley and peak positions, I_0 is the peak irradiation intensity at focus (z=0) and λ is the wavelength of the laser.