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# Dynamic Nonlinear Effect on Lasing in Random Media

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**Abstract:** We investigate the dynamic effect of nonlinearity on lasing in disordered medium. The third-order nonlinearity not only changes frequency and size of lasing mode, but also modifies laser emission intensity and laser pulse width.

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Recent studies illustrate that adding gain to a disordered medium leads to lasing in the long-lived eigenmodes. [1, 2, 3] Despite the modes with long lifetime are preferably amplified, their wavefunctions are not modified by the presence of gain. However, nonlinearity can change the eigenmodes of a disordered system. In random lasers nonlinearity is large due to high intensity and resonance enhancement.

We first present the experimental evidence of the change of lasing modes by nonlinearity. As the pump intensity increases, the frequencies of lasing modes shift in the opposite direction of the gain spectrum. This phenomenon indicates the spectral shift of lasing modes is caused not by cavity pulling but by nonlinearity. The time-resolved measurement of lasing spectra reveals the temporal shift of lasing frequencies caused by dynamic change of the refractive index.

To understand the nonlinear effect on random lasing, we carry out numerical simulation of a model system. The algorithm is based on the finite-difference time-domain (FDTD) solution to Maxwell equations coupled with the rate equations of electronic populations. Third-order nonlinearity with finite-time response is incorporated into the Maxwell equations. Our calculation results demonstrate that nonlinearity shifts the frequency and modify the size of the eigenmode of a disordered system. Figure 1(a) plots the spatial size of a random lasing mode in both linear and nonlinear cases under pulsed pumping.

The laser emission intensity and laser pulse width of a random laser are also changed by third-order nonlinearity  $\chi^{(3)}$ . Such changes are sensitive to the nonlinear response time  $\tau_0$ . Figure 1(b) plots the total laser emission energy for the same  $\chi^{(3)}$  but different  $\tau_0$ . The effect of nonlinearity on laser pulse intensity and width depends on the relative magnitude of the nonlinear response time  $\tau_0$  and the mode lifetime  $\tau_c$ . When  $\tau_0$  is longer than  $\tau_c$ , the change of laser pulse intensity and width is related to the size change of the lasing mode. If the size of the lasing mode decreases (or increases) in the presence of nonlinearity, light confinement gets better (or worse). The decrease (or increase) of light leakage is equivalent to an increase (or decrease) of the quality factor of the random cavity. Hence, lasing lasts longer (or shorter), and laser emission is stronger (or weaker). When  $\tau_0$  is shorter than  $\tau_c$ , the change of laser output depends only on the sign of  $\chi^{(3)}$ , i.e., positive (or negative) nonlinearity always extract more (or less) laser emission from the random medium at the same pumping rate. When the nonlinear response is much faster than the buildup of the lasing mode, the lasing mode cannot response fast enough to the nonlinear refractive index change. The phase of scattered light changes quickly due to rapid change of refractive index with intensity. The absence of constant phase relations among light waves scattered by different particles undermines the interference effect. Hence, the effect of single particle scattering becomes dominant over the collective effect of many particle scattering. For  $\chi^{(3)} > 0$  (or  $\chi^{(3)} < 0$ ), the refractive index contrast of the binary layers increases (or decreases) as the laser intensity increases. Light scattering of a single particle becomes stronger (or weaker). The increase (or decrease) of scattering strength results in more (or less) efficient lasing, i.e., higher (or lower) laser intensity and longer (or shorter) lasing period.

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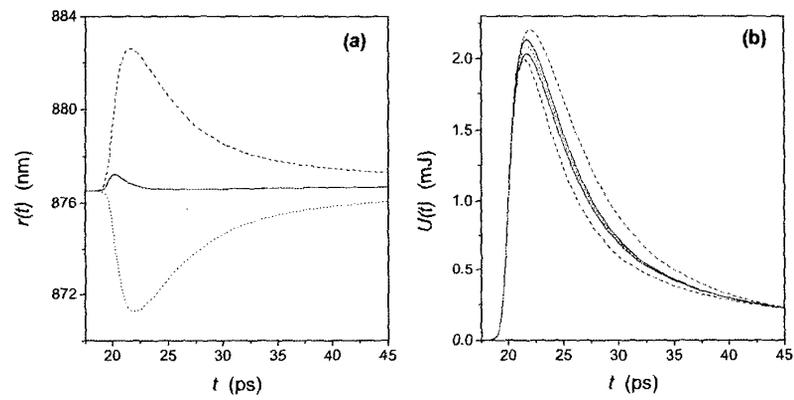


Fig. 1. (a) Spatial size  $r(t)$  of a lasing mode when (from top to bottom)  $\chi^{(3)} = 1.2 \times 10^{-16} \text{ m}^2/\text{V}^2$ ,  $\chi^{(3)} = 0$ ,  $\chi^{(3)} = -1.2 \times 10^{-16} \text{ m}^2/\text{V}^2$ . (b) Laser emission energy  $U(t)$  when (from top to bottom)  $\chi^{(3)} = 1.2 \times 10^{-16} \text{ m}^2/\text{V}^2$  and  $\tau_0 = 6.5 \text{ fs}$ ;  $\chi^{(3)} = -1.2 \times 10^{-16} \text{ m}^2/\text{V}^2$  and  $\tau_0 = 160 \text{ fs}$ ;  $\chi^{(3)} = 0$ ;  $\chi^{(3)} = 1.2 \times 10^{-16} \text{ m}^2/\text{V}^2$  and  $\tau_0 = 160 \text{ fs}$ ;  $\chi^{(3)} = -1.2 \times 10^{-16} \text{ m}^2/\text{V}^2$  and  $\tau_0 = 6.5 \text{ fs}$ .