Highly efficient laser action from free-shape dye-doped soft matter systems

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Experimental investigations of random laser action in a partially ordered, dye doped nematic liquid crystals with long-range dielectric tensor fluctuations are reported. Above a given pump energy value, a randomly distributed series of bright tiny spots appear giving rise to a strongly fluctuating spatio-temporal emission pattern. The spectral analysis reveals discrete sharp peaks of about 0.5 nm (FWHM). The unexpected surviving of interference effects in recurrent multiple scattering of the emitted photons provide the required optical feedback for lasing in nematics. Coherent backscattering of light waves in orientationally ordered nematic liquid crystals manifests a weak localization of light which strongly supports diffusive laser action in presence of gain medium. Unlike distributed feedback mirror-less lasers, this system can be considered as a cavity-less microlaser where the disorder unexpectedly plays the most important role, behaving as randomly distributed feedback laser. In particular, was studied the role of the thermally modulated order parameter in the diffusive laser action observed in systems having various sizes and different confining geometrical constraints. Important experimental evidence reveal a strong temperature dependence of the random lasering characteristics in the nematic phase and in close proximity of the nematic-isotropic (N-I) phase transition. A lowering of the laser emission intensity as the temperature increases is strictly related to the shift of the lasing threshold as function of the temperature, even though the pump energy is kept fixed. The optical losses increase, owing to the thermal fluctuation enhanced scattering, and drive the input-output smoother behavior until the system stops to lase, because situated below threshold. The unexpected reoccurrence of random lasing at higher temperatures, in proximity of N-I transition is found to be related to a different scattering mechanism, the micro-droplets nucleation and critical opalescence. These scientific aspects overlook features of great interest characteristic of laser physics and material science.

The thermal analysis of the emission properties of nematic liquid crystals doped with fluorescent guest molecules and confined in diverse symmetry boundaries emphasizes a striking temperature behaviour. Remarkable is the change of the input-output curve found experimentally as the temperature is varied within the nematic phase range. The lasing threshold usually extrapolated by the input-output curve overcomes an important shifting towards higher values as the temperature is increased. This is directly responsible for the monotonic decreasing of the random lasing intensity even though the pump energy is kept fixed. The laser emission generated by the feedback provided by thermal fluctuations scattering shows clearly the characteristics of random emission by varying stochastically in time, space and frequency. Even more interesting is the reoccurrence of random lasing as the nematic-isotropic phase transition is approached. The already reported critical opalescence effects manifested as a colossal increase of the scattering regime at the N-I transition suggested that a new feedback mechanism is behind the observed lasing effect. Here, the mechanism at the basis of the amplification process is due to the multiple scattering yielding weak localization of light waves, but now the scattering is generated by poly-dispersed nematic micro-droplets immersed in an isotropic environment which nucleated as the N-I transition is approached. This mechanism is referable to that responsible for random lasing in aggregated nanoparticles dye solutions. The scientific aspects presented herein overlie features of great interest characteristic of laser physics and material science.

References: