Electrically controlled plasmonic lasing resonances with silver nanoparticles embedded in amplifying nematic liquid crystals

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Received 15 May 2014, revised 23 August 2014  
Accepted for publication 1 September 2014  
Published 25 September 2014

Abstract

We demonstrated an electrical control of coherent plasmonic random lasing with very diluted Ag nanoparticles dispersed in a dye-doped nematic liquid crystal (NLC), in which the external electric field dependent emission intensity and frequency-splitting were recorded. A modified rate equation model is proposed to interpret the observed coherent lasing, which is a manifestation of the double enhancements caused by the plasmon–polariton near-fields of Ag particles on the population inversion of laser dye molecules and on the optical energy density of lasing modes. The featured laser quenching as weakening the applied field indicates that the present lasing resonances are very sensitive to the fluctuant dielectric perturbations in the NLC host, and are thus most likely associated with some coupled plasmonic oscillations among the metal nanoparticles.

Keywords: plasmon–polariton, Ag nanoparticles, liquid crystals, random lasers, electrically controlled lasing

(Some figures may appear in colour only in the online journal)

1. Introduction

Since the concept of random lasers was proposed by Letokhov in his seminal papers [1, 2], interest in these new kinds of mirrorless laser has grown very rapidly because of their extensive application prospects in the fields of display, target identification, biomedicine, and photonic devices. The necessary conditions for achieving a typical random lasing are where the disordered laser material is required to support coherent or incoherent feedback, and in the meantime provide a sufficient optical gain for the seed photons to be lased. In the original experimental investigations, some dielectric particles with high positive permittivities were chosen to fabricate random lasers [3–6], where the multiple scattering mechanism was essential in order to establish optical feedback. Until recently, one of the most inspiring breakthroughs was made by Meng et al when they observed coherent random lasing from dye-doped polymer films embedded with very tiny Ag nanoparticles [7, 8]. In these extremely transparent disordered samples, the scattering mean free paths of light were far larger than the wavelengths as well as the sample thicknesses, which ruled out any evident multiple scattering mechanism of confining light. These efforts suggest that the localized plasmon–polariton fields of Ag nanoparticles exhibit very competitive capabilities for improving and tailoring the stimulated emission characteristics of the surrounding gain media. Moreover, the plasmon–polariton particles can be easily controlled by engineering the particle sizes and shapes, changing the dielectric and chemical environments, and modulating the electromagnetic coupling with other adjacent particle partners. However, the full exploitation of the merits of metal-nanoparticle-based random lasers in designing optical devices
is sometimes hindered by a lack of effective dynamic control over their emission. In view of the rich optical properties of nematic liquid crystals on birefringence and light scattering due to the orientations and fluctuations of their directors \cite{9}, and in particular those which can be controlled by the external field, the environmental temperature, and the anchoring force at the interfaces, we hope to incorporate some tunable NLC environments into a plasmon-type random laser as well as tailoring the optical properties of such a device. This desire is also driven by another unknown question as to whether the intrinsic scattering properties of the NLC itself can be directly utilized to offer effective optical-feedback for random lasing or, at least, can provide an enhancement effect for plasmon-type random lasers.

In this article, we report on an experimental and theoretical investigation into the random lasing behavior of a dye-doped NLC embedded with diluted silver nanoparticles under optical pumping excitation. We found that over the associated thresholds, profound lasing resonances emerged in this random laser system if the director orientational fluctuation of the NLC was suppressed by an applied electric field. However, no random lasing signal was detected in the presence of sufficient director fluctuation, i.e., introducing dynamic light scattering due to NLC director fluctuation destroyed any coherent feedback already provided by the included Ag nanoparticles, and then extinguished the random lasing. By modulating the director fluctuation via a tunable, external electric field, we achieved switching of this Ag-nanoparticle-based random lasing. Furthermore, we recorded an intriguing frequency-splitting (~2.5 nm) and slight shrinkage of the lasing resonances as the strength of the external electric field increased. Based on the laser rate equation theory, we found that the plasmon-polariton near-fields of the Ag nanoparticles contributed to an overlapped local-field enhancement on both the population inversion of the laser dye (excited at pumping frequency) and the energy density of the laser field (oscillating at lasing frequencies), thus accounting for the coherent lasing spikes on the emission spectra. As expected, the measured dependence of random laser intensity on the external electric field can be qualitatively interpreted according to the physics of the dynamic light scattering of NLCs.

2. Experimental details

The semi-closed sample cell consisting of a pair of ITO-conductive glass separated by a 40 \( \mu \)m spacer and glued on three sides was prepared in advance. The disordered material used to create a random laser was an ultrasonically-dispersed mixture consisting of a commercial NLC (\( n_\rho = 1.52 \) and \( n_e = 1.72 \) at 610 nm and 20 °C), dodecanethiol-passivated Ag nanoparticles with nearly monodisperse dimension, and a 0.5 mMol L\(^{-1}\) laser dye DCM. The Ag nanoparticles were synthesized by wet chemical synthesis \cite{10} and characterized using a transmission electron microscope (JEM-1011, JEOL) operating at a 100 kV accelerating voltage. Figure 1(c) shows the TEM image of the Ag nanoparticles, whose average particle size was about 10 nm in diameter. The number density of the Ag nanoparticles was arranged to be \( \sim 8.85 \times 10^{17} \) m\(^{-3}\). This disordered mixture was infiltrated into the prepared sample cell to form the random laser sample, which we hereafter refer to as the NLC-Ag-DCM sample. As shown in figures 1(a) and (b), this sample was optically pumped by the second harmonics of a Q-switched neodymium-doped yttrium aluminum garnet (YAG) laser (\( \lambda = 532 \) nm, 10 Hz repetition rate, 10 ns pulse duration) in order to perform random laser experiments. The pump beam polarized along the \( x \)-axis was focused on the sample through a cylindrical lens to form an excitation stripe 1.8 mm in length with a variable width of 0.20–0.28 mm. The emission along the stripe was measured using a fiber optic spectrometer (AvaSpec-2048) with a spectral resolution of 0.2 nm.

3. Result discussions and analyses

To study the emission behavior, we initially exerted an external electrical field of 0.75 V \( \mu \)m\(^{-1}\) across the sample and adjusted the pump energy. The emission spectra exhibited broad spontaneous emission features at weak pumping, with a narrow peak appearing at 608 nm once the pumping energy exceeded a critical threshold value, as seen in figure 2, in which the insert gives the intensity of this peak as a function of the pumping energy, thus yielding an input–output curve with the pronounced threshold characteristic of lasers. However, the low ratio of the laser peak to the enhanced background of spontaneous emission suggests that there was still a minority of excited DCM molecules contributing to the stimulated emission in the laser mode. When increasing the pumping energy, more distinguishable spectral lines with full widths of \( \sim 0.2 \) nm at half maximum (FWHM) emerged on the backgrounds of the enhanced spontaneous emission spectra. Although this was a clear signal of coherent lasing resonances, the physics behind these laser spikes was by no means an obvious mechanism. As we will see in the following analysis, the observed coherent lasing resonances are at least associated with two enhancement effects caused by the particle plasmon–polariton near-fields of the Ag nanoparticles: One is the enhanced excitation rate of the dye molecules at pumping frequencies, and the other is their enhanced stimulated transition probability at lasing frequencies.

In all the possible optical feedback mechanisms we first evaluated the contribution of the multiple scattering of light by the randomly distributed Ag nanoparticles. It was expected that, when the fluctuation of NLC director was suppressed by a strong applied electrical field, the NLC-Ag-DCM sample would become almost transparent and its residual scattering property would be dominated by the buried silver nanoparticles. The extinction cross section of a 10 nm diameter silver nanosphere embedded in the NLC host (average refractive index \( \sim 1.59 \)) was calculated by Mie theory to be \( \sigma_e \sim 6.91 \times 10^{-19} \) m\(^2\) for \( \sim 610 \) nm emission light. Together with the information on particle density, \( \rho \sim 8.85 \times 10^{17} \) m\(^{-3}\), the transport mean free path \( l_t \) of the emission light in the NLC-Ag-NPs sample could be estimated via the formula \( l_t = 1/(\rho \sigma_e) \sim 1.64 \) m, which was much greater than the wavelength and the sample size, indicating that this NLC-Ag-DCM
sample was actually operating in a ballistic regime as it was exposed to a considerable external electric field. This consideration for a transport mean free path also excluded an Anderson localization mechanism [11, 12] for the optical feedback to establish random lasing.

Since there was no apparent multiple-scattering-like optical feedback to account for the observed random lasing from this NLC-Ag-DCM sample, we turned to the possible influences resulting from the plasmonic oscillations of the Ag nanoparticles. A UV-visible extinction spectrum for an extremely dilute suspension of these Ag nanoparticles (embedded in a toluene solvent with a refractive index very close to that of the NLC host at the random lasing frequencies) was obtained from a spectrophotometric measurement, and compared with the absorption and fluorescence spectra of the DCM molecules (in the absence of any Ag nanoparticles), as well as one typical emission spectrum of the random lasing, as shown in figure 3. The extinction spectrum of the Ag-NPs exhibited a surface plasmon resonance band centered at 425 nm, corresponding to the collective oscillations of the 5s electrons of silver atoms. As in all conventional dye lasers, the observed random lasing spikes emerged at wavelengths where the dye absorption was practically absent, but where the fluorescence emission was still strong. Furthermore, this lasing behavior actually survived in a region far beyond the plasmon resonance peak of individual Ag nanoparticles, thus avoiding a notable absorption loss caused by this. However, because both the natural fluorescence emission of the dye molecules and the plasmon-excitation of the individual metal particles exhibited none of discrete frequency-dependences, the essential coherent feedback mechanism responsible for the sharp lasing resonances was still hidden from us. One potential interpretation is that the coherent optical feedback in our NLC-Ag-DCM sample

Figure 1. (a) Schematic illustration of the configuration of the NLC-Ag-DCM sample and the arrangement of the excitation-detection. (b) Schematic diagram of the plasmonic random lasing in the abovementioned NLC-Ag-DCM sample. (c) The TEM image showing the morphology and size of the silver nanoparticles. (d) The energy density distribution of a particle plasmon–polariton mode excited by incident photons at 610 nm. The dashed black circle indicates the particle surface.

Figure 2. The main panel indicates the evolution of emission spectra as a function of pump energy per pulse, and the insert gives the dependence of emission intensity on pumping energy. The emission spectra were obtained from a pumped strip ~1.8 mm in length and ~0.28 mm in width, under a fixed electric field of 0.75 V µm⁻¹.
Anomalous of the NLC-Ag-DCM sample (4). Fluctuation of the NLC director washed out any coherent interresonances did indeed disappear as the disordered dynamic field to release the direct fluctuation without changing the condition into the NLC host by removing the pre-applied electrical perturbation to validate this hypothesis, we introduced a dielectric perturbation to the single-particle plasmon resonance [13–17]. To rise to a large red-shift in the coupled plasmon resonance with each other across the gap between them, thus weakening the nanosized metal particles is excited by external irradiation, the primary part of the light field outside the metal sphere can be obtained directly according to Mie scattering theory, which shows that the total outer field can be well-approximated by the driving field overlapping an infinite series of spherical vector harmonics:

\[ \mathbf{E} = E_0 e^{i k r} \cos \theta \mathbf{e}_p + \sum_{n=1}^{\infty} E_n (\mathbf{a}_n \mathbf{N}_{n1}(h) - b_n \mathbf{M}_{n1}(h)) + o(\epsilon_d), \] (3)

in which \( E_n = i^n E_0 (2n + 1)/(n^2 + n) \), and \( E_0 \) is the amplitude of the incident driving field; the vector spherical harmonis \( \mathbf{M}_{n1}(h) \) and \( \mathbf{N}_{n1}(h) \) (superscript ‘H’ relating to the first spherical Hankel function), and the corresponding Mie scattering coefficients \( b_n \) and \( a_n \) take the standard definitions that can be found in Bohren’s monograph [18].

Once the necessary parameters are determined as follows: \( \epsilon_1 = \epsilon_0 \mu_0^2 = 2.31 \epsilon_0 \), \( \epsilon_{Ag} = \epsilon_0 \mu_{Ag}^2 \), and \( n_{Ag} = 0.06 + 4.152 i \) for \( \sim 610 \text{ nm wavelength} \) extracted from the experimental data [19], a straightforward calculation of the Mie scattering coefficients shows that, for the investigated 10 nm-diameter Ag nanoparticle, in expression (3) only the terms characterizing the incident driving field and the electric dipole scattering field \( \mathbf{N}_{n1}(H) \) are relatively significant, and thus we have:

\[ \mathbf{E} \approx E_0 \left( e^{i k r} \cos \theta \mathbf{e}_p - \frac{3}{2} a_1 \mathbf{N}_{11}(H) \right), \] (4)

A time-harmonic lightwave with a frequency \( \omega \) propagating in the NLC host is described by the associated Maxwell wave equation:

\[ \nabla \times (\nabla \times \mathbf{E}) - \omega^2 \mu_0 \epsilon_0 \mathbf{E} = \omega^2 \mu_0 \epsilon_0 \mathbf{n} \cdot \mathbf{E}. \] (1)

Here, \( \epsilon_d = \epsilon_1 - \epsilon_2 \) denotes the difference between the two principal dielectric constants \( \epsilon_1 \), for the polarizations parallel to the optical axis of the NLC medium, and \( \epsilon_2 \), perpendicular to this axis; the unit vector \( \mathbf{n} \) dictates the average director orientation of NLC and coincides with oriented field, the \( \epsilon \), direction in the previously mentioned experimental configuration (figure 1(a)). By contrast, inside the Ag nanoparticle the matched time-harmonic light wave only obeys the vector wave equation:

\[ \nabla \times (\nabla \times \mathbf{E}) - \omega^2 \mu_{Ag} \epsilon_0 \mathbf{E} = 0 \] (2)

with an isotropic and metallic dielectric function \( \epsilon_{Ag} \).

As a simple model we assumed that the Ag nanoparticl had a spherical shape characterized by its radius \( R \), and the total laser field around the particle was driven by some incoming photons emitted from the DCM molecules doped in the NLC host. Specifically, we now arranged an approximate local spherical-polar-coordinate system \((r, \theta, \phi)\), symmetrically fixed on the Ag nanosphere, so that these driving photons performed as a linearly polarized plane wave incident along the polar axis, with the polarization direction dictated by radial unit vector \( \mathbf{e}_p \equiv \mathbf{e}_r |_{\phi = 0, \theta = \pi/2} \). The scattered field together with the incident driving field determined the details of the enhanced near-field of this Ag nanosphere. In the following discussion we introduce a wave number parameter \( k = \omega \sqrt{\epsilon_0} \mu_0 \). By treating the \( \epsilon_1 \ll \epsilon_2 \) as a small perturbation parameter and under the zero-order approximation, the primary part of the light field outside the metal sphere can be obtained directly according to Mie scattering theory, which shows that the total outer field can be well-approximated by the driving field overlapping an infinite series of spherical vector harmonics:

\[ \mathbf{E} = E_0 e^{i k r} \cos \theta \mathbf{e}_p + \sum_{n=1}^{\infty} E_n (\mathbf{a}_n \mathbf{N}_{n1}(h) - b_n \mathbf{M}_{n1}(h)) + o(\epsilon_d), \] (3)

in which \( E_n = i^n E_0 (2n + 1)/(n^2 + n) \), and \( E_0 \) is the amplitude of the incident driving field; the vector spherical harmonis \( \mathbf{M}_{n1}(h) \) and \( \mathbf{N}_{n1}(h) \) (superscript ‘H’ relating to the first spherical Hankel function), and the corresponding Mie scattering coefficients \( b_n \) and \( a_n \) take the standard definitions that can be found in Bohren’s monograph [18].

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in which \( E_n = i^n E_0 (2n + 1)/(n^2 + n) \), and \( E_0 \) is the amplitude of the incident driving field; the vector spherical harmonis \( \mathbf{M}_{n1}(h) \) and \( \mathbf{N}_{n1}(h) \) (superscript ‘H’ relating to the first spherical Hankel function), and the corresponding Mie scattering coefficients \( b_n \) and \( a_n \) take the standard definitions that can be found in Bohren’s monograph [18].

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\[ \mathbf{E} \approx E_0 \left( e^{i k r} \cos \theta \mathbf{e}_p - \frac{3}{2} a_1 \mathbf{N}_{11}(H) \right), \] (4)
from which the enhancement of energy density (time average value) for the near-field can be estimated by:

\[
\frac{\mathbf{E}^* \mathbf{E}}{E_0^2} \approx 1 + \frac{3 \text{Im} \{a_1\} \left(1 - 3 \sin^2 \theta \cos^2 \varphi \right)}{k^3} \frac{r}{\lambda} + \frac{9|a_1|^2 \left(1 + 3 \sin^2 \theta \cos^2 \varphi \right)}{4k^6} \frac{1}{r^5}. \tag{5}
\]

As shown in figure 1(d), this plasmon–polariton field at a lasing resonance exhibited an enhancement of up to 17 times the energy density on the charge-accumulated surface. Although such a plasmon–polariton was excited by the photons assuming a plane wave form, more general excitations should be expanded with plane wave bases, and the average over propagation directions and polarizations do not change the basic picture. This example reveals that a linearly polarized pump beam at 532 nm can excite a surface electric field with its energy density enhanced by more than 25 times, which results in an enhanced pumping rate of dye molecules.

We assume that the dye molecules in the NLC-Ag-DCM sample can be modeled by an open two-level system, with the upper and lower levels being allowed to exchange their populations with a thermal reservoir, independently [20]. In addition to the stimulated emission and absorption channels induced by a coherent laser field, the decay of the upper-level population \(N_2\) is dominated by the processes of spontaneous fluorescence emission and possible energy transfer to silver particles; in contrast, the decay of the lower-level population \(N_1\) is mainly determined by intrinsic non-radiative losses. Therefore, the associated rate equations of laser molecules are given by:

\[
\frac{dN_1}{dt} = -\gamma_{1p}^\text{eq} (N_1 - N_1^\text{eq}) + (N_2 - N_1) B_0 I(\omega), \tag{6a}
\]

\[
\frac{dN_2}{dt} = \lambda_p (\gamma_{2hsp} + A_{sp}) (N_2 - N_2^\text{eq}) - (N_2 - N_1) B_0 I(\omega), \tag{6b}
\]

in which the pumping rate \(\lambda_p\) for dye molecules is proportional to \(|E_p|^2 P_{pump}|^2\), with \(E_p\) being the local pump field and \(P_{pump}\) the related transition dipole moment; \(I(\omega)\) is the partial energy intensity of the local laser field that is ‘seen’ by the dye molecules; \(A_{sp}\) and \(B_0\) are the Einstein coefficients corresponding to the spontaneous and stimulated emissions, respectively; the superscript ‘eq’ labels the thermal equilibrium values of the two populations; \(\gamma_{2hsp}\) presents the absorption rate caused by the Ag nanoparticles; and \(\gamma_{1p}^\text{eq}\) denotes the intrinsic non-radiative decay rate of the lower-level dye molecules. From equations (6) the steady population inversion is given by:

\[
\Delta N = \frac{\lambda_p (\gamma_{2hsp} + A_{sp})^{-1} - (N_1^\text{eq} - N_2^\text{eq})}{1 + [\gamma_{2hsp}^{-1} + (\gamma_{2hsp} + A_{sp})^{-1}] B_0 I(\omega)}, \tag{7}
\]

which in principle determines the threshold condition, \(\Delta N = 0\), for the local net output of stimulated photons and predicts a gain saturation at high laser intensities. As making use of the distance-to-particle-dependent pumping rate, the energy absorption rate, and the spontaneous emission coefficient, as proposed in a large selection of the literature [21–24], for dipole fluorophores outside a metal nanoparticle, we can infer a positive population inversion for the dye molecules if sufficient pumping is allowed.

To clarify this property we need only to focus on the numerator of expression (7) and plot (as shown in figure 4) a radial profile of the term \(\lambda_p (\gamma_{2hsp} + A_{sp})^{-1}\) normalized by dividing its asymptotic value \(\lambda_p^\text{eq} (A_{sp})^{-1}\) in a far-field region, with \(\lambda_p^\text{eq}\) and \(A_{sp}\) being denoted as the far-field limits of the pumping rate and the spontaneous emission rate, respectively. It is worth noting that when the dye molecules are close to the metallic surface, the enhancement of the pumping rate is limited but the energy absorption rate becomes divergent, and thus expression (7) presents a minus population inversion, resulting in laser quenching. Because the net emission power of laser photons per unit volume is characterized by:

\[
\Delta N B_0 I(\omega) \propto \Delta N |E_p P_{\text{em}^{\text{eq}}}|^2 \tag{4}
\]

with \(P_{\text{em}^{\text{eq}}}\) representing the stimulated transition dipole moment at laser frequencies, by substituting expressions (4) and (7) into this expression, we assert that in the vicinity of Ag nanoparticles, the enlarged population inversion of the laser dye, multiplied by the enhanced near-fieldtriggering stimulated transitions, would cause substantial lasing. This mechanism is the key to understanding why coherent lasing resonances can be so easily achieved in Ag-nanoparticle-based amplifying media.

When the pump energy was moderate and the intensity of the applied electric field surpassed 0.25 \(\text{V} \mu\text{m}^{-1}\), the lasing spectra exhibited an intriguing frequency-splitting, i.e. two remarkable spike-groups with a wavelength-separation...
of several nanometers appearing at the top of each emission spectrum. As shown in figure 5(a), a pair of maximum emission spikes belonging to the two separated spike-groups, respectively, are denoted by $P_1$ and $P_2$, in which the former almost always localized at 610 nm, and the latter tended to settle in the vicinity of 607 nm, and experienced a slight red shift with an increase in external voltage. The insert in the main panel shows the wavelength separation as a function of the applied transverse voltage. (b) The evolution of the emission intensity against the voltage. The wavelength separation as a function of the applied transverse voltage is shown in figure 5(b). Using a weak electric field of below 0.25 V $\mu$m$^{-1}$, we only obtained flat spontaneous emission spectra, no matter how strong the pumping was. When the applied electric field was increased beyond 0.5 V $\mu$m$^{-1}$, the emission spectra exhibited a pronounced random lasing property, and the peak of the emission intensity experienced a rapid climb. By continuously modulating the voltage upwards, we found that any further increases in emission intensity became less significant and eventually negligible. This indicates that introducing dielectric perturbation into the host medium could effectively control the lasing resonances of the NLC-Ag-DCM sample.

Figure 5. (a) The main panel shows the electric field dependence of the emission spectra, featured by striking frequency splitting under stronger applied fields, and the insert gives the corresponding wavelength separation as a function of the applied transverse voltage. (b) The evolution of the emission intensity against the transverse voltage exhibits switching behavior at around 15 V. All the experimental data were collected from a pumped strip ~1.8 mm in length and ~0.20 mm in width, with the error bars corresponding to the standard deviations.

To investigate the influence of the direct fluctuation of the NLC host on the emission properties, we let the transverse electric field increase from 0 volts to 60 volts at a fixed pumping strength. The evolution of the emission intensity against the transverse voltage is shown in figure 5(b). Using a weak electric field of below 0.25 V $\mu$m$^{-1}$, we only obtained flat spontaneous emission spectra, no matter how strong the pumping was. When the applied electric field was increased beyond 0.5 V $\mu$m$^{-1}$, the emission spectra exhibited a pronounced random lasing property, and the peak of the emission intensity experienced a rapid climb. By continuously modulating the voltage upwards, we found that any further increases in emission intensity became less significant and eventually negligible. This indicates that introducing dielectric perturbation into the host medium could effectively control the lasing resonances of the NLC-Ag-DCM sample.

In contrast to the concept that a robust and steady scattering mechanism is favorable in supporting coherent feedback for lasing resonances [27–29], the transient and dynamic one in the NLC medium actually disturbed any coherent feedback and quenched the lasing resonances. There is a simple understanding that the dependence of lasing intensity on the external electric field can be interpreted by looking into the light scattering details of the NLC. A differential cross section of light scattering per unit volume of the NLC is given by [30]:

$$
\frac{d\sigma}{d\Omega} = \frac{k^4k_i^2}{16\pi^2} \sum_{\alpha=x,y} \left[ (\mathbf{p}_i \cdot \mathbf{e}_a)(\mathbf{p}_i \cdot \mathbf{e}_s) + (\mathbf{p}_i \cdot \mathbf{e}_s)(\mathbf{p}_i \cdot \mathbf{e}_a) \right]^2 I_s(q),
$$

(8)

where $q = k_i - k_s$, with $k_i$ and $k_s$ represent the propagation vectors for any incident and scattered light, respectively; the unit vectors $\mathbf{p}_i$ and $\mathbf{p}_s$ characterize the polarization directions of the incident and scattered light, respectively; the longitudinal unit vector $\mathbf{e}_a$, as matching the oriented electric field, dictates the average orientation of the NLC director field $\mathbf{n} = \mathbf{n}(x, y, z, t)$ in thermal equilibrium, and $I_s(q)$ denotes the time average values for the Fourier components of the spatial correlations of two transverse director fluctuations, which can be calculated from the following variation of the total Frank free energy [9, 31, 32]:

$$
\delta G = \int_V \delta g_{\text{bulk}} \, d\tau
$$

(9)

over the scattering region. The Frank free energy density involved in such a variation is given by:

$$
\delta g_{\text{bulk}} = \frac{1}{2} \left[ k_{11} (\nabla \cdot \mathbf{n})^2 + k_{13} (\nabla \times \mathbf{n})^2 \right] + \text{Const},
$$

(10)

in which we add the last director-dependent term coming from the dielectric polarization caused by the applied electrical field $\mathbf{E}_{ap}$ along the $z$-axis; $k_{11}, k_{12}$ and $k_{13}$ are the standard Frank elastic constants corresponding to splay, twist, and bend deformations, respectively. The director field is assumed to deviate from the average direction by small fluctuations, but to conserve the length, i.e. $\mathbf{n} = \mathbf{e}_z + \delta \mathbf{n} = (\delta h_x, \delta h_y, 1 + \delta h_z)$ and $\mathbf{n} \cdot \mathbf{n} = 1$. 

\[\text{Laser Phys. Lett. 11 (2014) 115814}\]
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Starting from the equation (9), following a similar procedure introduced in reference [30], and then utilizing the energy equipartition theorem, we obtain:

\[ I(q) = k_B T \left( k_1 g_1^2 q^2 + k_2 g_2^2 q^2 + k_3 g_3^2 q^2 + g_4 E_{q}^2 \right)^{-1}, \]

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(11)
in which \( k_B \) is the Boltzmann constant and \( T \) is the absolute temperature. Combining expressions (8) and (11), we see that the director fluctuations and, consequently, the scattering cross section \( d\sigma/d\Omega \) monotonically decline with an increase in the applied electrical field strength. This means that the dynamic dielectric perturbation in an NLC host can be controlled by modulating the external fields.

The fluctuated dielectric perturbations impact the optical properties of the NLC-Ag-DCM sample in at least two aspects: One is damaging the steady interferences between the different particle plasmon–polariton fields, which washes away the coupled plasmonic resonances; the other is diminishing the total emission intensity of both coherent and incoherent light by scattering events. Thus, we relate the observed laser quenching to the damage of the coupled plasmonic resonances, and the dimming of total emission by dynamic light scattering, which takes some of the photons out of the emission beam. Furthermore, the diminishing of the lasing spikes at low strengths of the applied field suggests that the multiple scattering mechanism, originating from the simple thermal fluctuation of the NLC director orientation, fails to create sufficient coherent feedback to develop lasing resonances under the present pumping condition of nanosecond pulses, if not in more general circumstances.

### 4. Conclusions

To sum up, we demonstrated an electrical control over plasmonic random lasing resonances which had never been performed previously. The localized plasmon–polariton field of the silver nanoparticles distributed in the gain medium of the dye-doped NLC contributed to a strong enhancement of the stimulated emission, and thus facilitated the lasing process. In contrast, the dynamic light scattering in the NLC host, which was essentially a result of the thermal fluctuation of the director orientation and the dynamic dissipation due to the NLC’s viscosity, tended to broadening and quenching the lasing resonances. The switching behavior on the random lasing roots in the competition between these two opposite mechanisms, which could be ruled by modulating the director fluctuation strength with the applied electric fields. The disappearance of the lasing resonances in the absence of any applied fields also meant that the intrinsic multiple dynamic light scattering of the NLC materials barely provided coherent optical feedback for random lasing. With the ultimate goal of accurately predicting and designing lasing resonances for general amplifying media modified by metal nanoparticles, further investigations are on the horizon.

### Acknowledgments

The authors thank Professor H Zhong and Dr G Yang for their helpful assistance in the synthesizing and characterizing of the silver nanoparticles. This work was supported by the NSFC Grant (Nos. 10874016 and 11474021) and the special research fund of the Hunan Key Laboratory of Optoelectronic Integration and Optical Manufacture.

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