Suppression of interactions in multimode random lasers in the Anderson localized regime

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Understanding random lasing is a formidable theoretical challenge. Unlike conventional lasers, random lasers have no resonator to trap light, they are highly multimode with potentially strong modal interactions, and they are based on disordered gain media, where photons undergo random multiple scattering. Interference effects notoriously modify the propagation of waves in such random media, but their fate in the presence of nonlinearity and interactions is poorly understood. Here, we present a semiclassical theory for multimode random lasing in the strongly scattering regime. We show that Anderson localization, a wave interference effect, is not affected by the presence of nonlinearities. To the contrary, its presence suppresses interactions between simultaneously lasing modes. Consequently, each lasing mode in a strongly scattering random laser is given by a single long-lived, Anderson localized mode of the passive cavity, the frequency and wave profile of which do not vary with pumping, even in the multimode regime when modes spatially overlap.
intensities via their self- and cross-saturation through the spatial hole burning effect. Our analysis of the lasing theory of refs 24 and 25, augmented by wavefunction correlations in the Anderson localized regime26, provides a straightforward analytical understanding of the suppression of modal interactions. A single-pole approximation is rigorously justified, even in the multimode regime, because of the exponentially small broadening of localized modes and their exponentially small coupling to only an algebraic number of CF modes. The absence of modal interaction we predict is thus totally unexpected. A similar system, with the same gain medium and cavity shape and size, but without or with little scattering, exhibits strong mode competition as standard, in particular with shifts in mode frequencies and the disappearance (and possible reappearance) of certain modes as the pump strength increases17. 

Modal interactions are turned on once the CF modes are broadened, which can occur either at weaker disorder when their localization length increases, or when their lifetime is limited by photon absorption. The linear non-interacting behaviour of lasing modes we predict is the trademark of Anderson localization in random lasers, and we propose to monitor lasing frequencies as a function of pump strength to detect Anderson localization in experiments on random lasers.

As a starting point we take the Maxwell–Bloch equations (MBE), a set of three coupled nonlinear differential equations for the electric field, the polarization and the population inversion in the gain medium27 (Supplementary Section SI). For simplicity we consider either a one-dimensional system or a two-dimensional system with electromagnetic field transverse electric with polarization perpendicular to the gain medium, or transverse magnetic. In those instances, it is sufficient to consider scalar fields. To find steady-state solutions, refs 24 and 25 first write both the polarization and either a one-dimensional system or a two-dimensional system of the vector medium27 (Supplementary Section SI). For simplicity we consider a set of three coupled nonlinear differential equations for the electric field. The linear non-interacting behaviour of lasing modes we define as the population inversion density of the active medium. Our analysis of the lasing theory of refs 24

\[
\frac{d}{dt}\psi_\mu(x,k_\mu) = D_0\sum_{n=1}^{M_\mu} \overline{\mathbf{a}_n} \psi_n(x,k_\mu)
\]

(2)

of the lasing modes \(\Psi_\mu(x,k_\mu)\) over \(M_\mu\) elements of a truncated basis of CF modes \(\{|\psi_\nu(x,k_\mu)\}_{\nu=1}^{M_\mu}\). When \(N\) modes are lasing, \(M \geq N\), and in the Anderson localized regime, we will see that the structure of the threshold matrix \(T\) is such that \(M_\mu = 1\) for each lasing state. In equation (1), \(D_0\) gives the overall pump strength, which we define as the population inversion density of the active medium. The threshold matrix \(T = \Lambda T\) is the product of a diagonal and a non-diagonal matrix,

\[
\Lambda_{nm} = \delta_{nm} \frac{1}{k_{\mu} k_{\mu}} \frac{\gamma_{\perp}}{ck_{\mu} - ck_{\mu} \alpha_{0} + i\gamma_{\perp}}
\]

(3a)

\[
\mathbf{T}_{mn} = \int_{\mathbb{R}} dx \frac{F(x)}{1 + h(x)} \psi_\mu(x,k_\mu) \psi_\mu(x,k_\mu)
\]

(3b)

where \(F(x)\) is the pump profile, \(c\) is the speed of light, \(\alpha_0\) is the atomic frequency of the two-level active medium, and \(\gamma_{\perp}\) is the polarization relaxation rate. The integral runs over volume \(C\) (length in dimension \(d = 1\), area in \(d = 2\) of the gain medium). Nonlinearities arise from the hole-burning denominator

\[
h(x) = \sum_{\nu} \Gamma(k_\nu) |\Psi_\nu(x)|^2
\]

(4)

where the sum runs over all lasing modes and \(\Gamma(k) = \gamma_{\perp}/[(ck - \omega_0)^2 + \gamma_{\perp}^2]\). The lasing modes are self-consistently determined by the nonlinear eigenvalue problem of equation (1). Solutions exist only for discrete real values of \(k_\mu\), giving the lasing frequencies \(\Omega_\mu = ck_\mu\). The wavenumber of the CF modes \(k_{\mu} \approx k_{\mu} + i\kappa_{\mu}\), where \(k_{\mu}, \kappa_{\mu} \in \mathbb{R}\), are complex because of the openness of the cavity, \(\kappa_{\mu} = c\tau_{\mu}\) with the lifetime \(\tau_{\mu}\).

The presence of modal interactions is reflected in the structure of the lasing modes, for example, the number \(M_\mu\) of CF states significantly contributing to a single lasing mode in the expansion of equation (2). If the expansion is dominated by a single component, the lasing mode resembles a non-interacting CF mode; however, if the expansion runs over many non-vanishing components, the lasing mode is in general very different from non-interacting CF states, signalling the onset of modal interactions. We next proceed to show that \(M_\mu \approx 1\) in the strongly localized regime, by analysing the structure of the threshold matrix \(T = \Lambda T\). In Supplementary Section III, we argue that matrix elements \(T_{mn}\) are non-zero only when CF states \(\psi_\mu\) and \(\psi_\nu\) are centred within one localization length of one another. Furthermore, the non-vanishing matrix elements fluctuate pseudo-randomly as a function of the indices \((m,n)\). In the Anderson localized regime, most CF modes are long-lived, \(\kappa_{\mu} \approx \frac{1}{2} \exp[-L/\xi]\) and consequently \(|\Lambda_{nm}| \approx (k_{\mu} \gamma_{\perp}/2\exp[L/\xi]/(ck_{\mu} - \alpha_0 + i\gamma_{\perp})|\) is exponentially large at \(k_{\mu} = k_{\mu} \approx k_{\mu}\). One then expects that lasing frequencies are very close to those of certain CF modes, \(k_{\mu} \approx k_{\mu}\), and that the expansion (2) for the corresponding lasing mode is dominated by a single component \(a_{\mu}^{(m)}\). We show perturbatively that this is indeed the case as long as \(\exp[-L/\xi] \ll 1\). Keeping only a single component in equation (1) gives the single-pole approximation,

\[
(a_{\mu}^{(m)}) = 0 \text{ for } n \neq m_0\]

(5)

At this level, one has

\[
D_0 = (T_{m_0,m_0} - (a_{\mu}^{(m)})^2) \ll 1
\]

1. Leading-order corrections are obtained with the approximation

\[
(a_{\mu}^{(m)}) = D_0 T_{m_0,m_0} (a_{\mu}^{(m)}) + D_0 \sum_{n \neq m_0} T_{m_0,n} (a_{\mu}^{(m)})
\]

(6)

From equations (5) and (6), we conclude that corrections to the single-pole approximation are negligible when

\[
\Xi = \sum_{n \neq m_0} \Lambda_{nm} T_{m_0,n} T_{nm_0} \ll 1
\]

(7)

with \(\Lambda_{mm}\) and \(T_{mn}\) defined in equations (3a) and (3b). We next analyse the conditions under which equation (7) is satisfied.

For \(T_{m_0,n} T_{mm_0}\) not to vanish, we need to consider overlapping CF states \(m_0\) and \(n\), which have their support within one localization length. At energy \(h\kappa_\mu\), they must therefore differ in energy by at least the CF mode spacing within one localization volume29, in our case \(\Delta_{\nu} = \hbar/(b_1^2 \Delta_0 + b_2^2)\), \(b_1 = 2\) and \(b_2 = 2\pi\). We have the upper bound \(|\Lambda_{nm}| \approx \hbar (\kappa_{\mu}/(2\hbar h\kappa_\mu + \Delta_{\nu}) \times \gamma_{\perp}/(ck_{\mu} - \alpha_0 + i\gamma_{\perp}))|\) for \(n \neq m_0\) in equation (7), and thus \(|\Lambda_{nm}/\Lambda_{mm}^{1/2} \approx 2 \gamma_{\perp}/(ck_{\mu}^{1/2}) \exp[-L/\xi]/[1 + 2b_{\nu}/(ck_{\mu}^{1/2})]|\). Next, averages \(T_{m_0,n} T_{mm_0}\) of the product of matrix elements of \(T\) can be evaluated, under the assumption that CF states are sufficiently close to localized modes of a disordered closed system. This
systems, whose lifetimes are long enough, generate generic diffusive modes. The lifetime of such modes can be estimated from the single-pole approximation. Corrections from the single-pole approximation are still negligible if the condition expressed in equation (7) is satisfied and that a criterion is the localization length of cavity modes is numerically estimated with the weakly scattering, delocalized regime. Accordingly, we propose that the criteria for the absence of interactions in random lasers are $\xi \ll L$ in $d = 1$ and $\xi \ll L$, $k_f L \ll L/\xi$ in $d = 2$. While we expect that the $d = 3$ criterion is the same as in two dimensions, we recall that an analysis of the MBE for vector fields is possible in this case.

Corrections to the single-pole approximation become non-negligible when the mode lifetime becomes shorter. We briefly discuss two cases when this happens. First, in the Anderson localized regime, CF modes have an exponentially long lifetime, but their broadening can be limited by photon absorption or leakage. Both effects can be included in the above estimate via the substitution $\kappa_{muv} \simeq \xi^{-1} \exp[-L/\xi] \rightarrow \kappa_{muv} \simeq (c \tau_{\text{eff}})^{-1}$. This leads to $|\Lambda_{muv}/\Lambda_{muv}|_{\text{eff}} \lesssim h/\Delta_{\tau_{\text{eff}}}$ and corrections to the single-pole approximations become of the order $E \sim (k_f \xi)/\Delta_{\tau_{\text{eff}}}$. We conclude that photon absorption turns on the interaction in the localized regime, when it reduces the mode lifetime such that $\tau_{\text{eff}} \lesssim (k_f \xi)/k_f c$. Second, mode lifetimes are shorter and wavefunction correlations are different in the $d = 2$ diffuse, delocalized regime, where all CF modes overlap. From ref. 28, we estimate (Supplementary Section SIII) $E \sim h/(k_f L)/\Delta_{\tau_{\text{out}}}$. The level spacing $\Delta_{\tau} = \hbar c/2 \pi k_f L^2$. Prelocalized states exist in such systems, whose lifetimes are long enough, $\tau_{\text{preloc}} \gtrsim h/\Delta$; that deviations from the single-pole approximation are still negligible if $f(k_f \xi, k_f L) \ll \Delta_{\tau}/\tau_{\text{preloc}}/h$. Reference 17 on the other hand suggests that lasing in diffusive random lasers can be supported by standard generic diffuse modes. The lifetime of such modes can be estimated by the classical diffusion time $\tau_{\text{cl}} = L^2/c \ell$ through the system, and the dimensionless measure of corrections to the single-pole approximation becomes $\Xi \sim k_f \xi \ell f(k_f \xi, k_f L) > 1$. This agrees with the observed strong modal interactions reported in ref. 17 for a diffusive laser with $k_f \xi \ell > 1$. We caution however that our estimates all rely on the assumption that CF modes have the same wavefunction correlations as modes of closed, non-absorbing Anderson localized systems. This is no longer true in the weakly scattering, delocalized regime.

We numerically checked our theoretical predictions. From the above analysis, dimensionality plays only a limited role in the Anderson localized regime, and for numerical convenience, we take a $1d$ edge-emitting, disordered microcavity laser. The cavity extends from $x = -L/2$, where we impose reflecting boundary conditions to $x = L/2$ where it is open. We fix its length, $L = 2 \mu m$ and the resonant frequency $\omega_0 = c k_0$ with $k L = 50$. The transversal and longitudinal relaxation rates are $\gamma_\perp = 8 \times 10^{-2} \omega_0$ and $\gamma_\parallel = 2 \times 10^{-5} \omega_0$. Disorder is introduced in the cavity by a spatially varying refractive index randomly distributed in the interval $n(x) \in [1.3, 6.7]$ with a spatial correlation $\chi = 8 \text{ nm}$. This rather large variation has been chosen to ensure that with the numerically reachable values of $L$ and $\omega_0$, we are in the Anderson localized regime. The localization length of cavity modes is numerically estimated with the inverse participation ratio

$$I(\Psi) = \left| \int \text{d}x |\Psi(x)|^2 \right|^2 / \int \text{d}x |\Psi(x)|^4$$

which estimates the spatial extent of a wavefunction regardless of its overall normalization. For an exponentially localized mode $\Psi(x) = \exp[i \theta(x)] \exp[-|x|/2\xi]/\sqrt{2\xi}$, one has $I(\Psi) = 4\xi$. In Fig. 1 we show a typical set of data for lasing modes in the strongly disordered, localized regime, indicating the mode's position, spatial extent, lasing frequency and output power. We estimate...
and are localized well inside the cavity, a distance larger than varies from one mode to another. Lasing modes have long lifetimes, are in the Anderson localized regime. As usual, localization lengths with pump strength, with a linear dependence on $D_0$ (modes) or in frequency. Only the modal emission power increases (Fig. 1a). This is even more remarkable, given that several modes Fig. 1b), their frequency (Fig. 1d), nor their spatial extent.

Figure 2 | Overlapping lasing modes in the Anderson localized regime. Spatial mode profile $|\Psi(x)|^2$ of lasing modes for a cavity with the same parameters as in Fig. 1. a, At pump strength $D_0 = 0.04$, there are seven lasing modes. Straight lines are exponential fits giving the indicated localization lengths $\xi$. Fits around $x = -0.2 \mu m$ (not shown) give $\xi \approx 0.06 \mu m$. b, At pump strength $D_0 = 0.4$, three new modes have started to lase (dashed lines); however, lasing modes have not changed their overall profile, but have only been multiplied by a mode-dependent factor.

$\xi \approx 0.03$–0.1 $\mu m$ from Fig. 1a, so that $\xi/L \approx 0.015$–0.05 $\ll 1$ and we are in the Anderson localized regime. As usual, localization lengths vary from one mode to another. Lasing modes have long lifetimes, and are localized well inside the cavity, a distance larger than $\xi$ away from the emitting cavity boundary. We see that as the pump varies by a factor of a thousand and up to twelve modes lase, lasing modes change neither their position (monitored by their centre of mass, Fig. 1b), their frequency (Fig. 1d), nor their spatial extent (Fig. 1a). This is even more remarkable, given that several modes strongly overlap either spatially (for instance the red and brown modes) or in frequency. Only the modal emission power increases with pump strength, with a linear dependence on $D_0$ that is reached quickly after threshold for each lasing mode. This is what is expected for non-interacting modes, given that $D_0$ gives the inversion density of the active medium and is not bound in our approach. Incorporating a finite maximal density in our model would lead to mode saturation, but would also limit the number of lasing modes and thus constrain our investigations of mode interactions.

We investigate more quantitatively the evolution with pumping of the spatial mode structure. We show in Fig. 2 the spatial profile of lasing modes at two different pump strengths, both in the multimode regime. At moderate pump strength, $D_0 = 0.04$ (Fig. 2a), there are seven lasing modes, which all exhibit exponential Anderson localization. From exponential fittings, we have extracted localization lengths in the range $\xi \in [0.05, 0.3] \mu m$, in good agreement with the values for the inverse participation ratio shown in Fig. 1a. Remarkably enough, we see two strongly overlapping lasing modes (red and brown). Upon increasing the pump strength to $D_0 = 0.4$ (Fig. 2b), three additional modes are lasing (indicated by dashed lines); however, the shape of the lasing modes has not changed at all. Enhanced pumping only increases their overall magnitude by a mode-dependent factor. In particular, neither the brown nor the red lasing mode have been modified by the emergence of the brown violet mode, despite their strong overlap.

Figure 3 next shows in colour scale the normalized spatial profile $|\Psi(x)|^2/\int_\mu dx|\Psi(x)|^2$ of the second lasing mode, corresponding to the red curves in Figs 1 and 2. It is clearly seen that over almost four orders of magnitude for $D_0$, and despite nine mode thresholds being crossed, the mode’s spatial structure remains exactly the same. Particularly interesting is the fact that around $D_0 = 0.3$, a new mode starts to lase (brown curves in Figs 1 and 2), spatially right on top of the mode plotted in Fig. 3a. Despite their strong overlap, these two modes do not interact, which we attribute to strong localization.

Having confirmed our prediction that modal interactions are frozen in the Anderson localized regime, we finally illustrate how they are turned on when the lifetime of the passive cavity modes is reduced, either by absorption or leakage. We achieve this by adding a finite imaginary part to the index of refraction. In Fig. 4a,b, we present data for the same cavity as in Fig. 1, but the index of refraction acquires a finite imaginary part, $\text{Im}[n] = 0.15$. We see that modal interactions are turned on as inverse participation ratios and modal emission power vary significantly with the pump strength. Numerical results presented in Supplementary Section SII further show that lasing frequencies are much more pump-sensitive when modes have a reduced lifetime. We also...
found, but do not show, that the centres of mass of the modes vary significantly with the pump strength when their lifetime is reduced. We finally corroborate our prediction that modal perturbations are turned on in the delocalized regime by simulations of cleaner cavities in Fig. 4c,d. We first observe that fewer modes lase, which we attribute to the presence of stronger mode interactions. The presence of strong interactions is further confirmed by the appearance and disappearance of a lasing mode (blue curves) as the pump increases. Such switching on and off of lasing modes with increasing pump strength has been reported in two-dimensional weakly disordered cavities, where they were due to frequency repulsion. This is not the mechanism at work here, as we found that the blue mode in Fig. 4c,d is separated by about 0.5γ⊥ from the (yellow) lasing mode with closest frequency.

The stationary approach we have followed does not capture effects due to the presence of time-dependent perturbations such as spontaneous emission or noise, which can lead to amplified spontaneous emission (ASE). However, we do not expect ASE to significantly alter our conclusions, first because its effect is weak in strongly scattering systems and becomes even weaker at larger pump strengths away from lasing threshold, and second because additional mode couplings need to be exponential in 〈L/ξ〉 to compensate for the exponentially long lifetime of Anderson localized modes, \(\text{IPR} \propto \exp[〈L/ξ〉]\) in equation (7), and turn on corrections to the single-pole approximation. The presence of noise may broaden lasing modes, and lead to the above discussed reduction in mode lifetime; however, the broadening is usually small in the strongly scattering regime.

The interplay between nonlinear interactions and quantum/wave coherent effects in systems with complex scattering is not well understood. Here, we have shown how strong localization effects freeze mode interactions in multimode lasers, even between modes that have significant spatial or frequency overlap. Our theory shows that Anderson localization justifies a single-pole approximation where modes do not interact, and nonlinearities due to spatial-hole burning only determine which modes lase, and with what threshold and intensity. Our results suggest a novel approach to investigate Anderson localization in strongly scattering random lasers by observing how much lasing frequencies vary with pump strength, which does not require spatially resolving the lasing field.

Note added in proof: We have recently become aware of recent experiments on random lasers in the Anderson localized regime, which may corroborate our predictions.

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Author contributions
P.S. wrote the numerical codes and performed the numerical calculations. Both authors
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Competing financial interests
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