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Threshold of a Random Laser with Cold Atoms

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We address the problem of achieving an optical random laser with a cloud of cold atoms, in which gain and scattering are provided by the same atoms. The lasing threshold can be defined using the on-resonance optical thickness b_0 as a single critical parameter. We predict the threshold quantitatively, as well as power and frequency of the emitted light, using two different light transport models and the atomic polarizability of a strongly-pumped two-level atom. We find a critical b_0 on the order of 300, which is within reach of state-of-the-art cold-atom experiments. Interestingly, we find that random lasing can already occur in a regime of relatively low scattering.

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Random lasing occurs when the optical feedback due to multiple scattering in a gain medium is strong enough so that gain in the sample volume overcomes losses through the surface. Since its theoretical prediction by Letokhov [1], great efforts have been made to experimentally demonstrate this effect in different kinds of systems [2, 3, 4, 5, 6], as well as to understand the fundamentals of random lasing [7, 8]. The broad interest of this topic is driven by potential applications (see [9] and references therein) and by its connections to the subject of Anderson localization [10]. State-of-the-art random lasers [9] are usually based on condensed matter systems, and feedback is provided by a disordered scattering medium, while gain is provided by an active material lying in the host medium or inside the scatterers. In general, scattering and gain are related to different physical entities.

Another system that can be considered for achieving random lasing is a cold atomic vapor, using magneto-optical traps, where radiation trapping [11] as well as lasing [12, 13] have been demonstrated. One advantage is the ability to easily model the microscopic response of the system components, which can be extremely valuable to fully understand the physics of random lasers. However, in such system, the ability to combine gain and multiple scattering at the same time is not obvious, as both should be provided by the same atoms. The purpose of this Letter is to address this issue quantitatively. Note that even though new interesting features appear when coherent feedback is involved [14], we will consider only incoherent (intensity) feedback.

Following Letokhov's theory, we consider a homogeneous, disordered and active medium of size L . The random lasing threshold is governed by two characteristic lengths: the elastic scattering mean free path ℓ_{sc} [15, 16] and the linear gain length ℓ_g ($\ell_g < 0$ in the cases of absorption or inelastic scattering). In the diffusive regime, defined as $L \gg \ell_{sc}$, the lasing threshold is reached when the unfolded path length, L^2/ℓ_{sc} , becomes larger than the gain length. More precisely, the threshold is given by

[1, 17] $L_{\text{eff}} > \beta\pi\sqrt{\ell_{sc}\ell_g/3}$, where β is a numerical factor that depends on the geometry of the sample ($\beta = 1$ for a slab, $\beta = 2$ for a sphere), and $L_{\text{eff}} = \eta L$ is the effective length of the sample, taking into account the extrapolation length [15]. Another important length scale is the extinction length, as measured by the forward transmission of a beam through the sample, $T = e^{-L/\ell_{\text{ex}}}$. The extinction length is given by $\ell_{\text{ex}}^{-1} = \ell_{\text{sc}}^{-1} - \ell_g^{-1}$.

Let us consider now a homogeneous atomic vapor, constituted by atoms of polarizability $\alpha(\omega)$ at density ρ , submitted to a homogeneous pump field. The extinction length and the scattering mean free path are related to their corresponding cross-section σ by $\ell_{\text{ex,sc}}^{-1} = \rho\sigma_{\text{ex,sc}}$, with $\sigma_{\text{ex}}(\omega) = k_0 \times \text{Im}[\alpha(\omega)]$ and $\sigma_{\text{sc}}(\omega) = k_0^4/6\pi \times |\alpha(\omega)|^2$ [18]. As we consider only quasi-resonant light, we use only the wave vector $k_0 = \omega_0/c$ with ω_0 the atomic frequency. We also define a dimensionless atomic polarizability $\tilde{\alpha}$ such that $\alpha = \tilde{\alpha} \times 6\pi/k_0^3$ (we omit the dependence on ω in the following). As it is an intrinsic parameter of the cloud, it is convenient to use the on-resonance optical thickness $b_0 = \rho\sigma_0 L$, where $\sigma_0 = 6\pi/k_0^2$ is the resonant scattering cross-section (without pump laser). Using these quantities, the threshold condition writes

$$\eta b_0 > \frac{\beta\pi}{\sqrt{3|\tilde{\alpha}|^2 (|\tilde{\alpha}|^2 - \text{Im}(\tilde{\alpha}))}}. \quad (1)$$

Moreover, we have $L/\ell_{sc} = b_0|\tilde{\alpha}|^2$ and $\eta = 1 + 2\xi/[L/\ell_{sc} + 2(\beta - 1)\xi]$ with $\xi \simeq 0.71$ for $L > \ell_{sc}$ [19, 20]. Note that deeply in the diffusive regime ($L \gg \ell_{sc}$), $\eta \sim 1$.

Eq. (1) is the first result of this Letter. It shows, in the diffusive regime, the existence of a threshold of random lasing as soon as the medium exhibits gain, i.e., $|\tilde{\alpha}|^2 - \text{Im}(\tilde{\alpha}) > 0$. This threshold is given by a critical on-resonance optical thickness, expressed as a function of the atomic polarizability only. Interestingly, the condition $\text{Im}(\tilde{\alpha}) < 0$, corresponding to single-pass amplification ($T > 1$), is not a necessary condition.

The previous result is general and does not depend on a particular pumping mechanism or atomic model. Let

us now specify a gain model that will allow numerical evaluations of the lasing threshold and of the features of the emitted light. We shall use the simplest case of strongly-pumped two-level atoms, for which the normalized atomic polarizability at frequency ω can be written analytically (assuming a weak “probe” intensity) [21],

$$\tilde{\alpha}(\delta, \Delta, \Omega) = -\frac{1}{2} \frac{1 + 4\Delta^2}{1 + 4\Delta^2 + 2\Omega^2} \quad (2)$$

$$\times \frac{(\delta + i)(\delta - \Delta + i/2) - \Omega^2\delta/(2\Delta - i)}{(\delta + i)(\delta - \Delta + i/2)(\delta + \Delta + i/2) - \Omega^2(\delta + i/2)}.$$

In this expression, $\Delta = (\omega_p - \omega_0)/\Gamma$ is the normalized detuning between the pump frequency ω_p and the atomic transition ω_0 of linewidth Γ , $\delta = (\omega - \omega_p)/\Gamma$ is the normalized detuning between the considered “probe” frequency and the pump, and Ω is the Rabi frequency, normalized by Γ , associated with the pump-atom interaction. For a strong enough pumping power, this atomic polarizability allows for single-pass gain, when $\text{Im}(\tilde{\alpha}) > 0$. This gain mechanism is referred as “Mollow gain” [13, 21] and corresponds to a three-photon transition (population inversion in the dressed-state basis).

For each couple of pumping parameters $\{\Delta, \Omega\}$, the use of the polarizability (2) into the threshold condition (1) allows the calculation of the critical on-resonance optical thickness b_0 as a function of δ . Then, the minimum of b_0 and the corresponding δ determine the optical thickness b_{0cr} that the cloud must overcome to allow lasing, and the frequency δ_{RL} of the random laser at threshold. The result is presented in Fig. 1 for a spherical geometry ($\beta = 2$). The result for b_{0cr} is independent of the sign of Δ and we only show the region $\Delta > 0$. The minimum optical thickness that allows lasing is found to be $b_{0cr} \approx 200$ and is obtained for a large range of parameters, approximately along the line $\Omega \approx 3\Delta$. The optimum laser-pump detuning is near the gain line of the transmission spectrum, i.e., $\delta_{RL} \sim \text{sign}(\Delta)\sqrt{\Delta^2 + \Omega^2}$ (a small shift compared to the maximum gain condition is due to the additional constraint of combined gain and scattering).

The obtained critical optical thickness is achievable with current technology [22], showing that random lasing is possible in a system of cold atoms with Mollow gain. As this result has been obtained using the diffusion approximation, the condition $L/\ell_{sc} = b_0|\tilde{\alpha}|^2 \gg 1$ must be satisfied. This is not the case in the full range of random lasing parameters that we have found. For example, with $\Delta \approx 1$ and $\Omega \approx 3$, the critical optical thickness is almost minimum, $b_{0cr} = 213$, but $L/\ell_{sc} \approx 0.44$. In this case, the threshold defined by Eq. (1) is at best unjustified, at worst wrong. In order to identify in Fig. 1 the region in which the approach should be valid, we have hatched the area corresponding to $L/\ell_{sc} < 3$. Note that random lasing is still expected in this region, but for a larger on-resonance optical thickness, that would allow

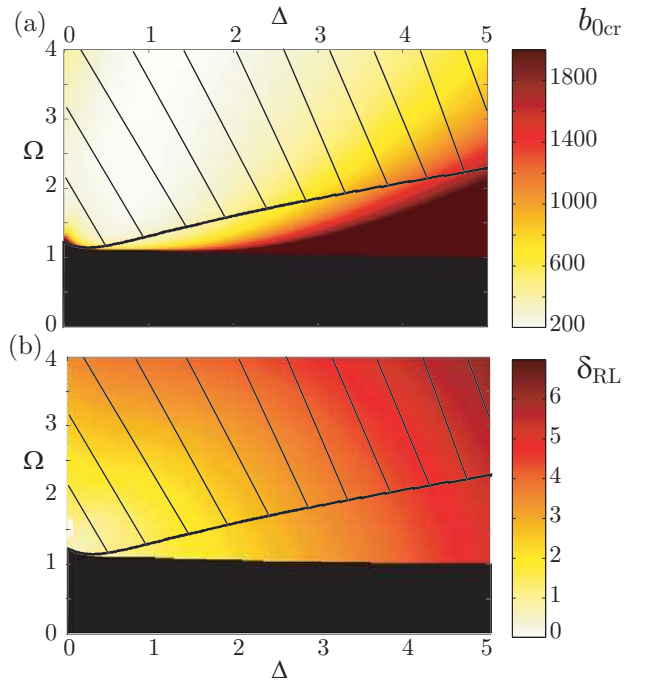


FIG. 1: Threshold of random lasing based on Mollow gain [21], calculated for each pair of pumping parameters Δ (detuning) and Ω (Rabi frequency) with Eqs. (1,2). Only the $\Delta > 0$ part is represented. (a) Critical optical thickness b_{0cr} to allow lasing. (b) Detuning δ_{RL} of the random laser from the pump frequency. The black area corresponds to a forbidden region (no gain). The hatched part corresponds to parameters for which the diffusion approximation is *a priori* not reliable.

the fulfillment of the diffusive condition. The minimum optical thickness in the region of parameters compatible *a priori* with the diffusion approximation is 347, and is located in the vicinity of $\{\Delta = 1, \Omega = 1.2\}$.

This first evaluation demonstrates the need for a more refined transport model. In the following, we use the approach introduced in Ref. [23], that is based on the radiative transfer equation (RTE). The RTE is a Boltzmann-type transport equation [24], that has a larger range of validity with respect to the ratio L/ℓ_{sc} than the diffusion equation [25].

Letokhov’s diffusive theory [1, 17] and the RTE-based theory [23] of random lasing both rely on a modal expansion of the solution of the transport equation. In order to compare the predictions of both models, we focus on the slab geometry ($\beta = 1$) since the modal expansion of the RTE is well known in this case [19] (to our knowledge, no simple expansion is available for a sphere in the RTE approach). The modal approach consists in looking for solutions of the form $\Psi_s(z, \mathbf{u}, t) = \phi_{\kappa, s}(\mathbf{u}) \exp(i\kappa z) \exp(st)$, where $\Psi(z, \mathbf{u}, t)$ is the specific intensity (z is the distance from the slab surface and \mathbf{u} denotes a propagation direction). For a given real κ , $s(\kappa)$ and $\phi_{\kappa, s}$ form a set of eigenvalues and eigenfunctions of the RTE. If one denotes

by $s_0(\kappa)$ the eigenvalue corresponding to the mode with the longest lifetime in the passive system, a laser instability appears when $s_0(\kappa) > 0$ in the presence of gain. The lasing threshold is defined by the condition $s_0(\kappa) = 0$. For isotropic scattering, this eigenvalue has an analytical expression valid for $\kappa \ell_{sc} < \pi/2$ [19, 23]:

$$s_0(\kappa)/c = \ell_g^{-1} - [\ell_{sc}^{-1} - \kappa / \tan(\kappa \ell_{sc})] \quad (3)$$

where c is the energy velocity. For a slab of width L , the dominant mode corresponds to $\kappa = \pi/L_{\text{eff}} = \pi/(L + 2\xi\ell_{sc})$. In practice, this determination of κ is meaningful as long as $\xi = 0.71$ can be taken as a constant (independent on L), which is the case for $L > \ell_{sc}$. This condition sets the limit of accuracy of the modal RTE approach.

The diffusive result is recovered from the RTE approach in the limit $\kappa\ell_{sc} \ll 1$ [25]. A first order expansion of Eq. (3) yields $s_0^{(\text{DA})}(\kappa)/c = \ell_g^{-1} - \kappa^2\ell_{sc}/3$, where the superscript (DA) stands for diffusion approximation. The condition $s_0^{(\text{DA})}(\kappa = \pi/L_{\text{eff}}) = 0$ leads to Letokhov's threshold, with $\beta = 1$.

The comparison between the RTE and diffusive approaches deserves two comments. Firstly, the gain contribution to $s_0(\kappa)$ (first term in Eq. (3)) is the same in both models. Secondly, the scattering contribution (second term in Eq. (3)) is larger in the RTE model by a factor of at most 1.13 (when $L \sim \ell_{sc}$). Thus, the correction introduced by the RTE model, compared to the diffusion approximation, is relatively small, as it corresponds to an increase of $\eta b_{0\text{cr}}$ of at most a few percents. This means that the diffusive model gives accurate results down to $L \sim \ell_{sc}$, and that in cold-atom systems, random lasing can occur even in a regime of low scattering.

In Fig. 2, we compare the minimum optical thickness obtained with both models for the slab geometry and with the diffusive model for the sphere geometry. To put forward the domain of validity in each case, we plot the results as a function of L/ℓ_{sc} . As expected, in the range $L > \ell_{sc}$ the threshold predicted by the RTE for the slab geometry is only slightly larger than the one given by the diffusion approximation, so that the two curves can hardly be distinguished. For the sphere geometry, we dashed the part corresponding to the domain where the diffusive model is *a priori* not reliable, i.e., $L/\ell_{sc} < 3$. Nevertheless, by generalizing the conclusion obtained with the slab geometry, we reasonably expect the threshold to be located between 250 and 300.

Let us now turn to a first characterization of such a random laser. An important quantity to be investigated is the emitted power as a function of the pumping power. In the stationary regime (continuous pumping) we numerically solve the optical Bloch equations for a strongly-pumped two-level atom (without using the weak "probe" approximation that leads to Eq. (2)) to obtain the polarizability at the lasing frequency, including the gain saturation induced by the random laser

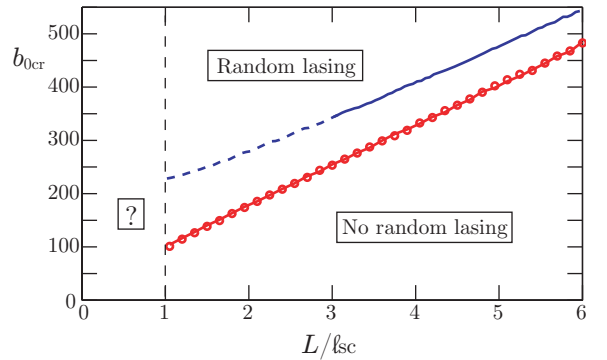


FIG. 2: Critical optical thickness for different geometries and transport models. The lower, red curve corresponds to the slab geometry (width L), with RTE model (continuous line) and diffusive model (open circle). The upper, blue curve corresponds to the sphere geometry (diameter L), with the diffusive model. The part with $L/\ell_{sc} < 3$ is dashed, as the model may not be reliable.

intensity. Above threshold, the laser intensity *in the medium* $I_{\text{RL}}^{(\text{in})} \propto |\Omega_{\text{RL}}|^2$ is determined by the condition $s_0(\kappa, |\Omega_{\text{RL}}|^2) = 0$ (s_0 would be positive without gain saturation). The obtained intensity is analogous to the intra-cavity intensity of a standard laser, and thus does not correspond to the emitted power $P_{\text{RL}}^{(\text{out})}$. At equilibrium, gain compensates losses, and $P_{\text{RL}}^{(\text{out})}$ is equal to the generated power, related to the gain cross-section σ_g , i.e., $P_{\text{RL}}^{(\text{out})} \propto \sigma_g |\Omega_{\text{RL}}|^2$ with $\sigma_g = \sigma_0 (|\tilde{\alpha}|^2 - \text{Im}(\tilde{\alpha}))$.

In order to know if the laser signal can be extracted from the background fluorescence, it is particularly relevant to compare the emitted laser power with the pump-induced fluorescence $P_{\text{Fluo}} \propto \sigma_0 |\Omega|^2 / (1 + 4\Delta^2 + 2|\Omega|^2)$. From this, we compute the ratio

$$\frac{P_{\text{RL}}^{(\text{out})}}{P_{\text{Fluo}}} = \frac{|\Omega_{\text{RL}}|^2}{|\Omega|^2} (|\tilde{\alpha}|^2 - \text{Im}(\tilde{\alpha})) (1 + 4\Delta^2 + 2|\Omega|^2) . \quad (4)$$

We plot the result in Fig. 3 as a function of $|\Omega|^2$, for a pump detuning $\Delta = 1$. To obtain Eq. (4), we assume that both pump and laser intensities are homogeneously distributed across the whole system. We also consider only the optimum random laser frequency, thus neglecting the spectral width of the random laser or any interaction between different random laser frequencies. Hence we neglect several effects as mode competition [26] and inelastic scattering of the laser light. Nevertheless we think that the order or magnitude of the ratio laser-to-fluorescence powers can be realistic for actual experiments, at least as long as only one mode of the laser is active [26]. For the chosen set of parameters, this ratio is more than 5% and hence laser emission should be measurable. Its distinction from the pump-induced fluorescence can be made by looking at the spectrum of the emitted light. Another interesting prediction of this model is that

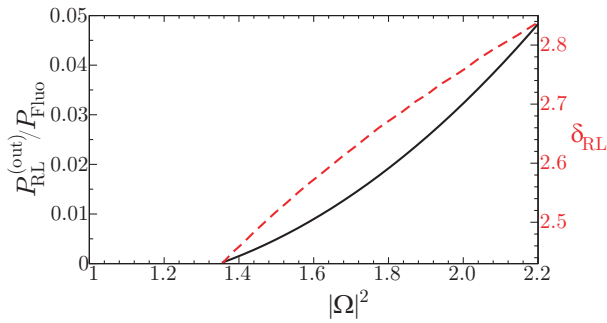


FIG. 3: Continuous line: Emitted random laser power normalized to the pump fluorescence power, as a function of the pump intensity. Dashed line: Normalized laser detuning δ_{RL} . The random medium is a spherical cloud of two level atoms with an on-resonance optical-thickness $b_0 = 650$.

the laser emission frequency shifts as the pump intensity is increased [Fig. 3]. This corresponds to the shift of the maximum gain of the Mollow polarizability.

In summary, we have established the possibility of achieving random lasing with cold atoms. The random laser threshold is described by a single critical parameter, the on-resonance optical thickness b_0 . In the particular case of a gain mechanism based on a strongly-pumped two-level atom (Mollow gain), our model predicts a critical $b_0 \sim 300$. Such an optical thickness is achievable in current cold-atoms experiments, *e.g.* by using crossed dipole traps [22]. We have also determined the basic features of the emitted light above threshold, showing that the random laser emission should be measurable.

Another interesting result is that, due to the large gain, lasing can be obtained with a low feedback (low amount of scattering, *i.e.*, $L \sim \ell_{\text{sc}}$). This regime is similar to that encountered in certain semiconductor lasers with a very poor cavity, and is different from the working regime of random lasers realized to date. This new regime could be numerically investigated by RTE-based simulations [25].

Finally, let us stress that the model developed here has several limitations, so that the numbers should be considered as first-order estimates. Firstly, we have considered monochromatic pumping, thus neglecting inelastic scattering from the pump. The inelastically-scattered photons may have a non-negligible influence on the atomic response, as shown in [27]. Secondly, the RTE model needs to be extended to a sphere geometry, and to a medium with inhomogeneous density and/or pumping. This would require a full numerical solution of coupled RTEs for the pump and probe beams [23]. We also outline that the case of the Mollow gain was chosen for the sake of simplicity, whereas other gain mechanisms might be more adapted for the search of random lasing, such as Raman gain or parametric gain [13]. Each gain mechanism has its advantages and drawbacks, but the degrees

of freedom they offer, together with the first estimates presented here, make us confident that the experimental realization of a cold-atom random laser is possible with current technology.

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- [1] V. S. Letokhov, *Sov. Phys. JETP* **26**, 835 (1968).
- [2] V. M. Markushev, V. F. Zolin, and Ch. M. Briskina, *Sov. J. Quantum Electron.* **16**, 281 (1986); C. Gouedard *et al.*, *J. Opt. Soc. Am. B* **10**, 2358 (1993).
- [3] N. M. Lawandy *et al.*, *Nature* **368**, 436 (1994); D. S. Wiersma *et al.*, *Nature* **373**, 203 (1995).
- [4] H. Cao *et al.*, *Phys. Rev. Lett.* **82**, 2278 (1999).
- [5] D. S. Wiersma and S. Cavalieri, *Nature* **414**, 708 (2001).
- [6] S. Gottardo *et al.*, *Nature Photon.* **2**, 429 (2008).
- [7] D. S. Wiersma and A. Lagendijk, *Phys. Rev. E* **54**, 4256 (1996).
- [8] A. L. Burin *et al.*, *Phys. Rev. Lett.* **87**, 215503 (2001).
- [9] D. S. Wiersma, *Nature Phys.* **4**, 359 (2008).
- [10] C. Conti and A. Fratalocchi, *Nature Phys.* **4**, 794 (2008).
- [11] A. Fioretti *et al.*, *Opt. Commun.* **149**, 415 (1998); G. Labeyrie *et al.*, *Phys. Rev. Lett.* **91**, 223904 (2003).
- [12] L. Hilico, C. Fabre, and E. Giacobino, *Europhys. Lett.* **18**, 685 (1992).
- [13] W. Guerin, F. Michaud, and R. Kaiser, *Phys. Rev. Lett.* **101**, 093002 (2008).
- [14] H. Cao, *J. Phys. A* **38**, 10497 (2005).
- [15] M. C. W. van Rossum and Th. M. Nieuwenhuizen, *Rev. Mod. Phys.* **71**, 313 (1999).
- [16] We consider only isotropic scattering so that the transport length equals the scattering mean free path [15].
- [17] H. Cao, *Waves Random Media* **13**, R1 (2003).
- [18] A. Lagendijk and B. A. van Tiggelen, *Phys. Rep.* **270**, 143 (1996), section 3.2.2 “Points scatterers” (pp. 164-166).
- [19] K.M. Case and P.F. Zweifel, *Linear transport theory* (Addison-Wesley, 1967).
- [20] K. Drozdowicz, E. Krynicka, and J. Dąbrowska, *App. Rad. Isot.* **58**, 727 (2003).
- [21] B. R. Mollow, *Phys. Rev. A* **5**, 2217 (1972); F. Y. Wu *et al.*, *Phys. Rev. Lett.* **38**, 1077 (1977).
- [22] M. D. Barrett, J. A. Sauer, and M. S. Chapman, *Phys. Rev. Lett.* **87**, 010404 (2001).
- [23] R. Pierrat and R. Carminati, *Phys. Rev. A* **76**, 023821 (2007).
- [24] S. Chandrasekhar, *Radiative Transfer* (Dover, New York, 1960).
- [25] R. Elaloufi, R. Carminati, and J.-J. Greffet, *J. Opt. Soc. Am. A* **21**, 1430 (2004).
- [26] H. E. Türeci *et al.*, *Science* **320**, 643 (2008).
- [27] L. Khaykovich, N. Friedman, and N. Davidson, *Eur. Phys. J. D* **7**, 467 (1999).