A cold-atom random laser

Q. Baudouin, N. Mercadier, V. Guarrera, W. Guerin and R. Kaiser
Institut Non Linéaire de Nice, CNRS and Université Nice Sophia-Antipolis, 1361 route des Lucioles, 06560 Valbonne, France

In this supplementary online material, we describe the models used in this study.

I. ATOMIC MODEL

We describe the atom-light interaction by a semi-classical model based on the optical Bloch equations (OBEs). We have to consider five atomic levels: the two ground states $|F = 2, 3\rangle$ (denoted hereafter $|2\rangle$ and $|3\rangle$) and the three excited states $|F' = 1, 2, 3\rangle$ (denoted $|1'\rangle$, $|2'\rangle$ and $|3'\rangle$). They are coupled to each other by three optical fields: the external Raman laser (subscript Ra in the following), the external optical pumping (OP) and the self-generated "random-laser" (RL) around the Raman gain frequency, see Fig. S1. The Raman laser is detuned by Δ_{Ra} from the $|3\rangle \to |2'\rangle$ transition, the random laser has a detuning Δ_{RL} from the $|2\rangle \to |2'\rangle$ transition, and the optical pumping has a detuning Δ_{OP} from the $|2\rangle \to |3'\rangle$ transition. The linewidth Γ of all optical transitions is $\Gamma/2\pi = 6.07$ MHz.

In first approximation, the detunings Δ_{Ra} and Δ_{RL} are equals (and noted Δ and δ for simplicity in Fig. S1 and the main paper). However, the frequency for which the random laser starts is not exactly given by the bare-atom two-photon resonance condition. One reason for this are the various light-shifts of the atomic levels coupled to different lasers. As a consequence, it is necessary to let Δ_{Ra} and Δ_{RL} be independent parameters in the evaluation of the

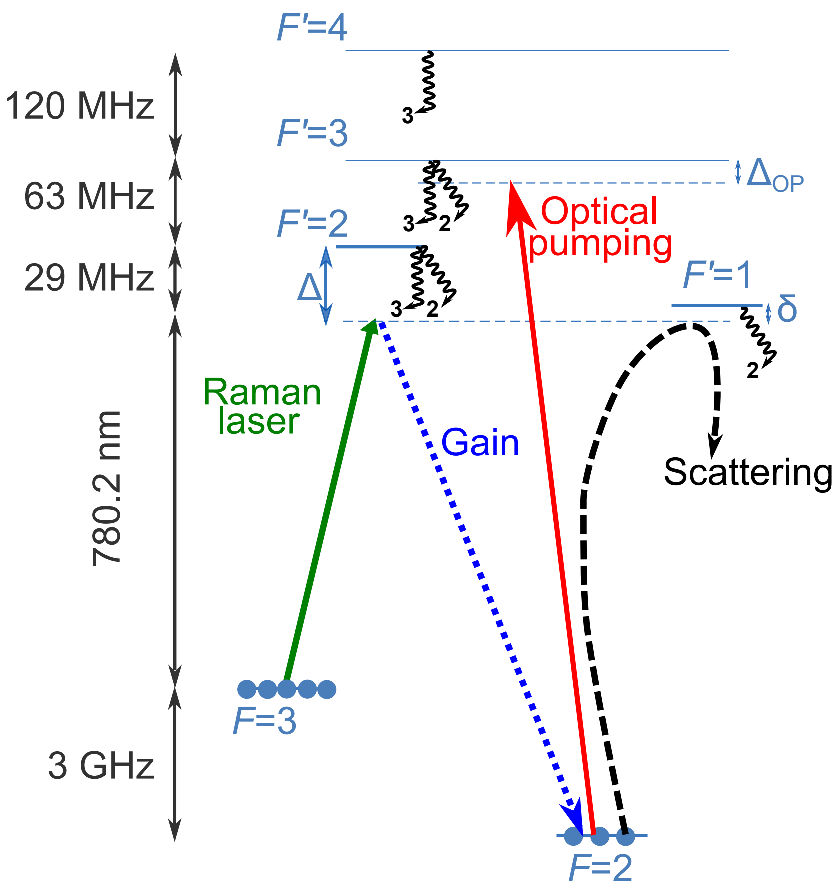

Fig. S 1: **Considered atomic levels and optical fields.** The level $|4'\rangle$ is only used in the laser-cooling process and is thus not included in our model. The five other levels are taken into account as well as the three optical fields: the Raman laser, the optical pumping and the "random-laser" line, where Raman gain and scattering are combined.

atomic response. Finally, we define also these detunings relatively to the $|1'\rangle$ state and note $\delta_{\mathrm{Ra,RL}} = \Delta_{\mathrm{Ra,RL}} + 4.8\Gamma$, where 4.8Γ is the hyperfine splitting between the $|1'\rangle$ and $|2'\rangle$ states.

A. Reduction from five to four levels

Including the $|1'\rangle$ in a fully coherent model leads to the appearance of coherence effects when we compute the scattering cross-section around the random-laser frequency: the contribution of the $|2\rangle \to |2'\rangle$ and $|2\rangle \to |1'\rangle$ transitions interfere, yielding sharp variations with the frequency [1]. We neglect these effects as we assume them to be irrelevant in the experiment because the coherence between the $|1'\rangle$ and $|2'\rangle$ states is expected to be destroyed by the multiple scattering undergone by the random-laser light.

We thus choose to describe by OBEs the coupling between the 4 levels $|F=2\rangle$, $|F=3\rangle$, $|F=2'\rangle$, $|F=3'\rangle$ and include the contribution of the $|1'\rangle$ state only as a supplementary scattering term, which adds incoherently to the off-resonant scattering provided by the $|2'\rangle$ state. We thus write for the total scattering cross-section,

$$\sigma_{\mathrm{sc,tot}}(\delta_{\mathrm{RL}}) = \sigma_{\mathrm{sc}}(\delta_{\mathrm{RL}}) + \mathcal{C}\langle\tilde{\rho}_{22}\rangle \frac{\sigma_0}{1+4\delta_{\mathrm{RL}}^2/\Gamma^2} , \tag{1}$$

where σ_{sc} and $\tilde{\rho}_{22}$ are, respectively, the scattering cross-section and the population of the $|2\rangle$ state, derived from a four-level model (without the $|1'\rangle$ level) as detailed below. The resonant scattering cross-section is $\sigma_0 = 3\lambda^2/2\pi$ with $\lambda = 780.2$ nm the wavelength of the D_2 line and $\mathcal{C} = \frac{1}{3}\frac{2F'+1}{2F+1} = \frac{2}{15}$ corresponds to the average coupling of the $|2\rangle \to |1'\rangle$ transition assuming a statistical mixture of the Zeeman sublevels [2]. For $\delta_{\mathrm{RL}} \sim 0$ and the parameters used in the experiment, the second term of Eq. (1) is the dominant contribution.

B. Optical Bloch equations for the four-level system

We derive the OBEs from the time dependent Schrödinger equation $i\hbar\frac{d\rho}{dt} = [H,\rho]$, where ρ is the density matrix and $H = H_{\mathrm{at}} + H_{\mathrm{int}}$ the total Hamiltonian, sum of an atomic part and of an interaction part, which reads $H_{\mathrm{int}} = \hbar\sum_{i,j}\Omega_{ij}(|i\rangle\langle j| + |j\rangle\langle i|)$, where Ω_{ij} is the Rabi frequency of the optical field that couples the levels i and j. We choose all Ω_{ij} as real numbers and note $\Omega_{\mathrm{Ra}} = \Omega_{32'}$, $\Omega_{\mathrm{RL}} = \Omega_{22'}$, and $\Omega_{\mathrm{OP}} = \Omega_{23'}$.

We add optical relaxations, use the rotating wave approximation and after making the following substitutions,

$$\begin{aligned}
\tilde{\rho}_{ii} &= \rho_{ii} \quad \text{for } i = \{2,3,2',3'\} , \\
\tilde{\rho}_{23'} &= \rho_{23'}e^{-i\Delta_{\mathrm{OP}}t} , \\
\tilde{\rho}_{22'} &= \rho_{22'}e^{-i\Delta_{\mathrm{RL}}t} , \\
\tilde{\rho}_{23} &= \rho_{23}e^{-i(\Delta_{\mathrm{RL}}-\Delta_{\mathrm{Ra}})t} , \\
\tilde{\rho}_{3'2'} &= \rho_{3'2'}e^{-i(\Delta_{\mathrm{RL}}-\Delta_{\mathrm{OP}})t} , \\
\tilde{\rho}_{3'3} &= \rho_{3'3}e^{-i(\Delta_{\mathrm{RL}}-\Delta_{\mathrm{Ra}}-\Delta_{\mathrm{OP}})t} , \\
\tilde{\rho}_{2'3} &= \rho_{2'3}e^{i\Delta_{\mathrm{Ra}}t} ,
\end{aligned}$$

we obtain the following equations for the populations,

$$\frac{d\tilde{\rho}_{22}}{dt} = \Gamma t_{2'2}\tilde{\rho}_{2'2'} + \Gamma t_{3'2}\tilde{\rho}_{3'3'} + i\frac{\Omega_{\mathrm{OP}}}{2}(\tilde{\rho}_{23'} - \tilde{\rho}_{3'2}) + i\frac{\Omega_{\mathrm{RL}}}{2}(\tilde{\rho}_{22'} - \tilde{\rho}_{2'2}) , \tag{2}$$

$$\frac{d\tilde{\rho}_{2'2'}}{dt} = -\Gamma\tilde{\rho}_{2'2'} - i\frac{\Omega_{\mathrm{Ra}}}{2}(\tilde{\rho}_{32'} - \tilde{\rho}_{2'3}) - i\frac{\Omega_{\mathrm{RL}}}{2}(\tilde{\rho}_{22'} - \tilde{\rho}_{2'2}) , \tag{3}$$

$$\frac{d\tilde{\rho}_{3'3'}}{dt} = -\Gamma\tilde{\rho}_{3'3'} - i\frac{\Omega_{\mathrm{OP}}}{2}(\tilde{\rho}_{23'} - \tilde{\rho}_{3'2}) , \tag{4}$$

$$1 = \tilde{\rho}_{22} + \tilde{\rho}_{2'2'} + \tilde{\rho}_{3'3'} + \tilde{\rho}_{33} , \tag{5}$$

where t_{ij} is the probability that an atom in state i decays into state j [2, 4]. The coherence terms are given by

$$\frac{d\tilde{\rho}_{23'}}{dt} = i\frac{\Omega_{\rm OP}}{2}(\tilde{\rho}_{22} - \tilde{\rho}_{3'3'}) - i\frac{\Omega_{\rm RL}}{2}\tilde{\rho}_{2'3'} - \tilde{\rho}_{23'}(\Gamma/2 + i\delta_{\rm OP}) \ , \tag{6}$$

$$\frac{d\tilde{\rho}_{22'}}{dt} = i\frac{\Omega_{\rm RL}}{2}(\tilde{\rho}_{22} - \tilde{\rho}_{2'2'}) + i\frac{\Omega_{\rm Ra}}{2}\tilde{\rho}_{23} - i\frac{\Omega_{\rm OP}}{2}\tilde{\rho}_{3'2'} - \tilde{\rho}_{22'}(\Gamma/2 + i\Delta_{\rm RL}) \ , \tag{7}$$

$$\frac{d\tilde{\rho}_{23}}{dt} = -i\frac{\Omega_{\rm OP}}{2}\tilde{\rho}_{3'3} + i\frac{\Omega_{\rm Ra}}{2}\tilde{\rho}_{22'} - i\frac{\Omega_{\rm RL}}{2}\tilde{\rho}_{2'3} - i\tilde{\rho}_{23}(\Delta_{\rm RL} - \Delta_{\rm Ra}) \ , \tag{8}$$

$$\frac{d\tilde{\rho}_{3'2'}}{dt} = i\frac{\Omega_{\rm Ra}}{2}\tilde{\rho}_{3'3} - i\frac{\Omega_{\rm OP}}{2}\tilde{\rho}_{22'} + i\frac{\Omega_{\rm RL}}{2}\tilde{\rho}_{3'2} - \tilde{\rho}_{3'2'}[\Gamma + i(\Delta_{\rm RL} - \Delta_{\rm OP})] \ , \tag{9}$$

$$\frac{d\tilde{\rho}_{3'3}}{dt} = -i\frac{\Omega_{\rm OP}}{2}\tilde{\rho}_{23} + i\frac{\Omega_{\rm Ra}}{2}\tilde{\rho}_{3'2'} - \tilde{\rho}_{3'3}[\Gamma/2 + i(\Delta_{\rm RL} - \Delta_{\rm Ra} - \Delta_{\rm OP})] \ , \tag{10}$$

$$\frac{d\tilde{\rho}_{2'3}}{dt} = i\frac{\Omega_{\rm Ra}}{2}(\tilde{\rho}_{2'2'} - \tilde{\rho}_{33}) - i\frac{\Omega_{\rm RL}}{2}\tilde{\rho}_{23} - \tilde{\rho}_{2'3}(\Gamma/2 - i\Delta_{\rm Ra}) \ , \tag{11}$$

with $\tilde{\rho}_{ji} = \tilde{\rho}^*_{ij}$.

We find the steady-state solution of the OBEs by numerically solving the corresponding linear system. It allows us to compute all the relevant atomic quantities, in particular the atomic polarizability at the random-laser frequency,

$$\alpha = \frac{6\pi}{k_0^3}\frac{\rho_{22'}}{\Omega_{\rm RL}/\Gamma} = \frac{6\pi}{k_0^3}\tilde{\alpha}, \tag{12}$$

where $k_0 = 2\pi/\lambda$ and $\tilde{\alpha}$ is dimensionless. The elastic scattering cross-section is then given by

$$\sigma_{\rm sc} = \frac{k_0^4}{6\pi}|\alpha|^2 = \sigma_0|\tilde{\alpha}|^2 \ , \tag{13}$$

the extinction cross-section by $\sigma_{\rm ext} = k_0{\rm Im}(\alpha) = \sigma_0{\rm Im}(\tilde{\alpha})$ and the gain cross-section by $\sigma_{\rm g} = \sigma_{\rm sc} - \sigma_{\rm ext}$ [3]. The cross-sections that are relevant for the other fields are computed in the same way.

Finally, due to energy conservation, the total fluorescence emitted by the cloud is equal to the extinction of the two external beams,

$$P_{\rm F} = \int n(\vec{r})d\vec{r}\,(\sigma_{\rm ext,Ra}I_{\rm Ra} + \sigma_{\rm ext,OP}I_{\rm OP}) \ , \tag{14}$$

where I denotes the respective incoming intensities.

Note that the OBEs allows us to compute the elements of the density matrix when all the detunings and Rabi frequencies, and in particular $\Omega_{\rm RL}$, are specified. To compute $\Omega_{\rm RL}$, we need to introduce self-consistent models that couple the EBOs with the light transport in the sample.

II. SELF-CONSISTENT MODEL WITH AMPLIFIED SPONTANEOUS EMISSION

We first consider a situation where light transport is ballistic, that is, neglecting random walk by scattering. This is relevant for the regions 1 of Fig. 2. For simplicity, we consider a spherical atomic cloud of homogeneous density n (the two external lasers have also homogeneous intensities).

Starting with $\Omega_{\rm RL} = 0$ in the EBOs, we compute a source term corresponding to spontaneous Raman scattering, given by $P_0 = \Gamma t_{2'2}\tilde{\rho}_{3'3'}$ [second term of the right-hand-side of Eq. (2)], taken at the detuning $\Delta_{\rm RL}$ of maximum gain. We then consider an atom at the center of the cloud (radius R) and compute the intensity at the RL frequency emitted by the other atoms. Taking into account the extinction along the ballistic path of the light, we integrate the source term P_0 described above,

$$I_{\rm RL} = \int \frac{nP_0}{4\pi r^2}\exp\left(-n\sigma_{\rm ext}r\right)r^2\sin(\theta)drd\theta d\phi = \frac{P_0}{-\sigma_{\rm ext}}\left[\exp(-n\sigma_{\rm ext}R) - 1\right] \ . \tag{15}$$

Note that $2n\sigma_{\rm ext}R = b_0{\rm Im}(\tilde{\alpha})$ so that the dependence on the optical thickness b_0 is explicit. Note also that, in the absence of scattering, $-\sigma_{\rm ext} = \sigma_{\rm g}$. One can see that $I_{\rm RL}$ increases with b_0 only if $\sigma_{\rm ext} < 0$, i.e., in presence of gain. This increase is smooth with b_0, without any visible threshold [Fig. S2].

From this result we can inject $\Omega_{\rm RL} = \Gamma\sqrt{I_{\rm RL}/(2I_{\rm sat})}$ in the EBOs, with $I_{\rm sat} = 6.4$ mW/cm^2. Since the value of $\Omega_{\rm RL}$ changes the atomic response (due to saturation and optical pumping), the source term and the gain cross-section depend on $\Omega_{\rm RL}$. We thus iterate this procedure until $\Omega_{\rm RL}$ converges to a stable value corresponding to the

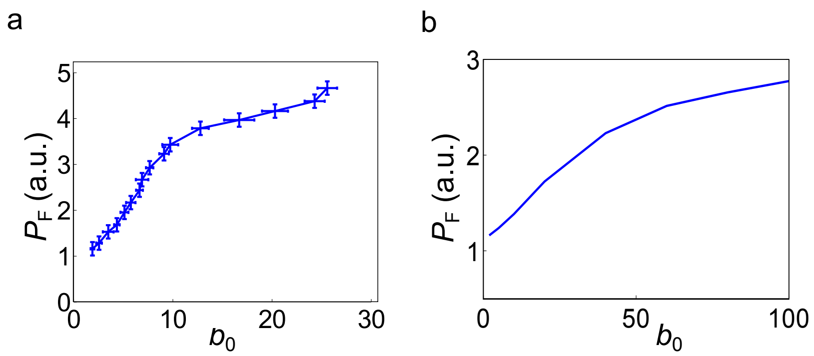

Fig. S 2: **Amplified stimulated emission (ASE) due to Raman gain. a,** Fluorescence power at $\delta = -2\Gamma$ as a function of the optical thickness. The data are those of Fig. 2. At this detuning, scattering due to the $|1'\rangle$ excited state is small and Raman gain explains the increase of the total fluorescence as b_0 increases with a constant number of atoms. Vertical error bars correspond to the noise on the data and horizontal error bars to shot-to-shot fluctuations of b_0. **b,** Solution of the ASE model, without any adjustable parameter.

steady-state. The total fluorescence, reported in Fig. 3b, can then be computed from Eq. (14). As mentioned above, the frequency of maximum gain is not exactly matching the two-photon resonance condition of the bare atom. We therefore scan the frequency $\Delta_{\rm RL}$ and retain for each value of $\Omega_{\rm RL}$ the frequency providing maximal emission.

Given the simple considered geometry, we expect this model to provide only qualitative results. Nevertheless, it has the advantage of including Raman gain and the saturation effect due to the emitted and amplified light.

Note that another effect can explain an increase of the total fluorescence with the optical thickness b_0, when the Raman-scattered photons from the pump laser are in the regime of multiple scattering on the open $|2\rangle \rightarrow |2'\rangle$ transition. Those photons contribute to optical pumping and can increase the population of the $|F=3\rangle$ state leading to enhanced emission in some cases. This effect has been studied in detail in [4] and is the dominating effect when the Raman laser is tuned very close to resonance ($\Delta_{\rm Ra} \sim 0$, that is, $\delta_{\rm Ra} \sim 4.8\Gamma$). We checked that at the detunings and intensities considered in Figs. 2 and S2, this contribution is negligible.

III. SELF-CONSISTENT MODEL WITH LETOKHOV'S CRITERION FOR THE RANDOM LASER THRESHOLD

When the Raman laser is tuned close to $\delta_{\rm Ra} \sim 0$ (region 2 in Fig. 2), the Raman-scattered light is not in the ballistic regime but undergoes multiple scattering due to the $|2\rangle \rightarrow |1'\rangle$ transition. To describe such multiple scattering, we couple the OBEs to a diffusion equation.

We can then build on Letokhov's results on the diffusion equation with gain: above a critical size for the medium, random lasing starts [5]. With a spherical geometry, the critical radius is given by

$$R_{\rm cr} = \pi \sqrt{\frac{\ell_{\rm sc}\ell_{\rm g}}{3}} \, , \tag{16}$$

where the mean-free-path is given by $\ell_{\rm sc}^{-1} = n\sigma_{\rm sc,tot}$ (Eq. 1) and the gain length by $\ell_{\rm g}^{-1} = n\sigma_{\rm g}$. Here we still suppose a homogeneous density n and we suppose also that the scattering is isotropic so that the transport length equals the scattering mean-free-path. As shown in [6], Letokhov's criterion can be rewritten as a critical optical thickness,

$$b_{0\rm cr} = \frac{2\pi\sigma_0}{\sqrt{3\sigma_{\rm sc,tot}\sigma_{\rm g}}} \, , \tag{17}$$

where the cross-sections are obtained from the EBOs of the 4-level model and additional scattering at the $|2\rangle \rightarrow |1'\rangle$ line as given by Eq. (1).

Based on this threshold criterion, we can compute the emitted light on the random laser line for a given b_0 via the implicit equation $b_{0\rm cr}(\Omega_{\rm RL}) = b_0$, analogous to the condition that gain must exactly compensate losses at the steady state, as in standard lasers. In practice, we start from $\Omega_{\rm RL} = 0$, compute $b_{0\rm cr}$ from the EBOs and Eqs. (1,17), and we increase $\Omega_{\rm RL}$ while $b_{0\rm cr} < b_0$. At each step we choose the random-laser detuning $\Delta_{\rm RL}$ that induces the lowest threshold, since the laser should start with this frequency.

Then, we can use Eq. (14) to compute the total fluorescence, corresponding to the experimental signal, from which we subtract the fluorescence computed with $\Omega_{\mathrm{RL}} = 0$ in order to compare with the measured increase of fluorescence δP_{F} [Fig. 3].

This qualitative model only allows for the computation of the random laser intensity above threshold and does note describe the emitted power below the laser threshold, for which we can use the ASE model described in the previous section. We note that this model also takes into account all saturation effects. In this system, the emitted intensity does not only change the gain, but also the scattering-induced feedback, since it changes the atomic populations.

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