



HAL
open science

Threshold of a Photonic Random Laser with Cold Atoms

Luis S. Froufe-Pérez, William Guerin, Rémi Carminati, Robin Kaiser

► **To cite this version:**

Luis S. Froufe-Pérez, William Guerin, Rémi Carminati, Robin Kaiser. Threshold of a Photonic Random Laser with Cold Atoms. 2008. hal-00343356v1

HAL Id: hal-00343356

<https://hal.science/hal-00343356v1>

Preprint submitted on 1 Dec 2008 (v1), last revised 3 Jun 2009 (v4)

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Threshold of a Photonic Random Laser with Cold Atoms

Luis S. Froufe-Pérez¹, William Guerin², Rémi Carminati³ and Robin Kaiser^{2*}

¹*Instituto de Ciencia de Materiales de Madrid, CSIC, Sor Juana Inés de la Cruz 3, Cantoblanco, Madrid 28049, Spain,*

²*Institut Non Linéaire de Nice, CNRS and Université de Nice Sophia-Antipolis, 1361 route des Lucioles, 06560 Valbonne, France,*

³*Laboratoire d'Optique Physique, ESPCI and CNRS, 10 rue Vauquelin, 75231 Paris Cedex 05, France*

(Dated: December 1, 2008)

We address the problem of achieving a photonic 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.

PACS numbers: 42.25.Dd, 42.55.Zz

Random lasing occurs when the optical feedback due to multiple scattering (or "radiation trapping") 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], as well as to understand the fundamentals of random lasing [6, 7]. State-of-the-art random lasers [8] are usually based on condensed matter systems and they share a number of common features. 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. Radiation trapping [9] as well as lasing [10, 11] have been demonstrated in such a medium. One advantage is the ability to easily characterize and model the microscopic properties of the medium, 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 [12], we will consider only incoherent (intensity) feedback.

Let us first remind the Letokhov's threshold in the general case, before applying it to an atomic system. We consider a homogenous, disordered and active medium of size L . The random lasing threshold is governed by two characteristic lengths: the scattering mean free path ℓ_{sc} [13, 14] and the linear gain length ℓ_g ($\ell_g < 0$ in the case of absorption). 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 critical length is given by

[1, 15]

$$L_{\text{eff}} > \beta\pi\sqrt{\ell_{sc}\ell_g/3}, \quad (1)$$

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 [13]. 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 related to the other lengths by $\ell_{\text{ex}}^{-1} = \ell_{sc}^{-1} - \ell_g^{-1}$.

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 at frequency ω is then given by $\ell_{\text{ex}}^{-1} = \rho k \times \text{Im}[\alpha(\omega)]$ and $\ell_{sc}^{-1} = \rho k \times k^3/6\pi \times |\alpha(\omega)|^2$ [16] ($k = \omega/c$ is the wave number of the associated optical transition). We define a dimensionless atomic polarizability $\tilde{\alpha}$ such that $\alpha = \tilde{\alpha} \times 6\pi/k^3$, and omit the dependence on ω in the following. We shall use the on-resonance optical thickness, which is an intrinsic parameter of the cloud and reads $b_0 = \rho\sigma_0 L$, where $\sigma_0 = 6\pi/k^2$ is the resonant scattering cross-section (without pump laser). Using these quantities, Eq. (1) reduces to

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

Moreover, we have $\eta = 1 + 2\xi/[b_0|\tilde{\alpha}|^2 + 2(\beta - 1)\xi]$ with $\xi \simeq 0.71$ for $L > \ell_{sc}$ [17, 18]. Note that deeply in the diffusive regime ($L \gg \ell_{sc}$), $\eta \sim 1$.

Eq. (2) 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 a transmission larger than one, *i.e.* single-pass amplification, 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 (that exhibits "Mollow gain"), for which the normalized atomic polarizability at frequency ω can be written analytically (assuming a weak "probe" intensity) [19],

$$\tilde{\alpha}(\delta, \Delta, \Omega) = -\frac{1}{2} \frac{1 + 4\Delta^2}{1 + 4\Delta^2 + 2\Omega^2} \times \frac{(\delta + i)(\delta - \Delta + i/2) - \Omega^2\delta/(2\Delta - i)}{(\delta + i)(\delta - \Delta + i/2)(\delta + \Delta + i/2) - \Omega^2(\delta + i/2)}. \quad (3)$$

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 $\text{Im}(\tilde{\alpha}) > 0$ [11, 19].

For each couple of pumping parameters $\{\Delta, \Omega\}$, the use of the polarizability (3) into the threshold condition (2) allows to calculate 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 [20], 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 to fulfill the diffusive condition. The minimum optical

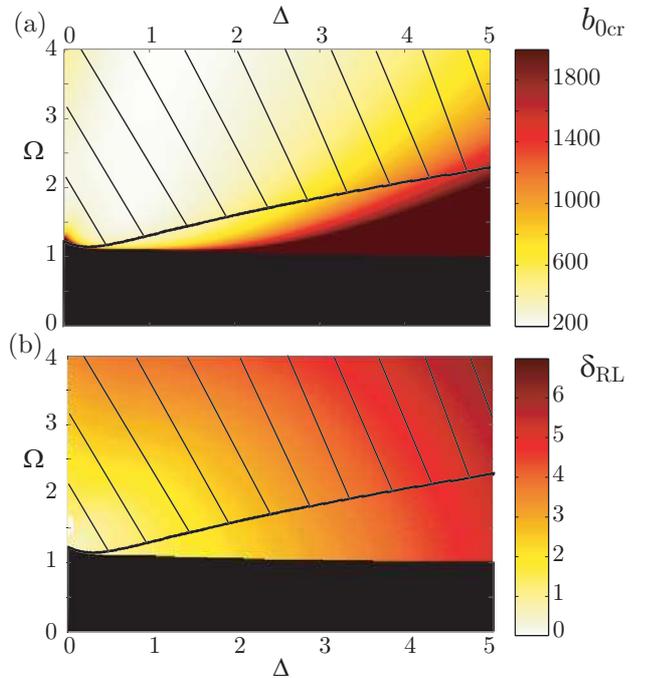


FIG. 1: Threshold of random lasing based on Mollow gain [19], calculated for each pumping parameters Δ (detuning) and Ω (Rabi frequency) with Eqs. (2,3). Only the $\Delta > 0$ part is represented. (a) Critical optical thickness b_{0cr} to allow lasing. (b) Detuning δ_{RL} of the random laser from the pump frequency. The black area corresponds to a forbidden region (no gain). The hatched part corresponds to parameters for which the diffusion approximation is not reliable.

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. [21], that is based on the radiative transfer equation (RTE). The RTE is a Boltzmann-type transport equation [22], that has a larger range of validity with respect to the ratio L/ℓ_{sc} than the diffusion equation [23].

Letokhov's diffusive theory [1, 15] and the RTE-based theory [21] 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 [17] (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

$$\Psi_s(z, \mathbf{u}, t) = \phi_{\kappa, s}(\mathbf{u}) \exp(i\kappa z) \exp(st), \quad (4)$$

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 $\kappa \ell_{sc} < \pi/2$ [17, 21]:

$$\frac{s_0(\kappa)}{\kappa c} = \frac{1}{\kappa \ell_g} - \left(\frac{1}{\kappa \ell_{sc}} - \frac{1}{\tan(\kappa \ell_{sc})} \right) \quad (5)$$

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 $\kappa \ell_{sc} \ll 1$ [23]. A first order expansion of Eq. (5) yields:

$$\frac{s_0^{(\text{DA})}(\kappa)}{\kappa c} = \frac{1}{\kappa \ell_g} - \frac{\kappa \ell_{sc}}{3}, \quad (6)$$

where the superscript (DA) stands for diffusion approximation. The threshold condition $s_0^{(\text{DA})}(\kappa = \pi/L_{\text{eff}}) = 0$ leads to Eq. (1), 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 Eqs. (5-6)) is the same in both models. Secondly, the scattering contribution (second term in Eqs. (5-6)) 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 atoms 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.

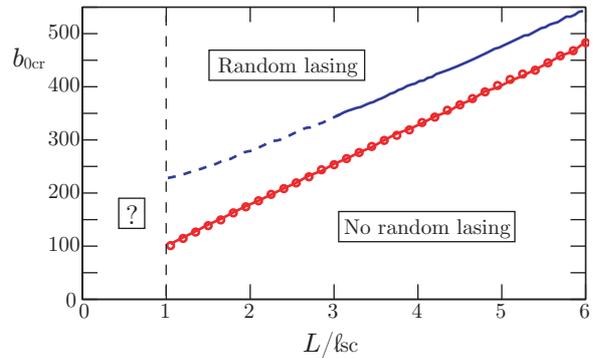


FIG. 2: Critical optical thickness for different geometries and transport model. The lower, red curve corresponds to the slab geometry (width L), with RTE model (continuous line) and diffusive model (open circle). The upper, blue curve corresponds to the sphere geometry (diameter L), with the diffusive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

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. (3)) to obtain the polarizability at the lasing frequency, including the gain saturation induced by the random laser intensity. Above threshold, the laser intensity, described by its normalized Rabi frequency Ω_{RL} , is determined by cancelling s_0 (that 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 intensity. As gain should compensate losses, the emitted power P_{RL} is proportional to the gain cross-section, *i.e.* $P_{\text{RL}} \propto (|\tilde{\alpha}|^2 - \text{Im}(\tilde{\alpha})) |\Omega_{\text{RL}}|^2$.

From an experimental point of view, it is particularly relevant to compare the emitted laser power with the pump-induced fluorescence, in order to know if the laser signal can be extracted from the background fluorescence. In figure 3 we plot the corresponding ratio as a function of the squared normalized pump Rabi frequency, for a pump detuning $\Delta = 1$. This ratio is estimated as

$$\frac{P_{\text{RL}}}{P_{\text{Fluo}}} = \frac{(|\tilde{\alpha}|^2 - \text{Im}(\tilde{\alpha})) |\Omega_{\text{RL}}|^2}{s_p / (1 + s_p)}, \quad (7)$$

where $s_p = 2|\Omega|^2/(1 + 4\Delta^2)$ is the pump saturation parameter. To obtain Eq. (7), 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 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. We note that, for

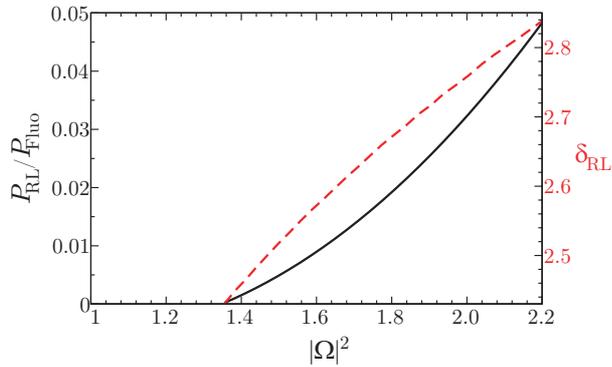


FIG. 3: Continuous line: Emitted random laser power normalized to the pump fluorescence power, as a function of the pump intensity. Dashed line: Normalized laser detuning δ_{RL} . The random medium is a spherical cloud of two level atoms with an on-resonance optical-thickness $b_0 = 650$.

the chosen set of parameters, this ratio is more than 5% and hence laser emission should be measurable. Another interesting prediction of this model is that 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 in a cloud of 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 [20]. We have also determined the basic features of the emitted light above threshold, showing that the random laser behavior 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. Such a cold-atom random laser may have peculiar properties, that makes it a very interesting tool in the field of light propagation in disordered amplifying media. This new regime could be numerically investigated by RTE-based simulations [23].

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 [24]. Secondly, the RTE model needs to be extended to a sphere geometry, and to a medium with inhomogeneous density in order to calculate a more accurate threshold. 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 [11]. 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 state-of-the-art technology.

The authors thank S. Skipetrov, J. J. Sáenz, R. Pierrat and F. Michaud for useful discussions. L.S.F. acknowledges the financial support of Spanish ministry of science and innovation through its Juan de la Cierva program. This work is supported by ANR-06-BLAN-0096.

* Electronic address: Robin.Kaiser@inln.cnrs.fr

- [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. Cavaleri, Nature **414**, 708 (2001).
- [6] D. S. Wiersma and A. Lagendijk, Phys. Rev. E **54**, 4256 (1996).
- [7] A. L. Burin *et al.*, Phys Rev. Lett. **87**, 215503 (2001).
- [8] D. S. Wiersma, Nature Phys. **4**, 359 (2008).
- [9] A. Fioretti *et al.*, Opt. Commun. **149**, 415 (1998); G. Labeyrie *et al.*, Phys. Rev. Lett. **91**, 223904 (2003).
- [10] L. Hilico, C. Fabre and E. Giacobino, Europhys. Lett. **18**, 685 (1992).
- [11] W. Guerin, F. Michaud and R. Kaiser, Phys. Rev. Lett. **101**, 093002 (2008).
- [12] H. Cao, J. Phys. A **38**, 10497 (2005).
- [13] M. C. W. van Rossum and Th. M. Nieuwenhuizen, Rev. Mod. Phys. **71**, 313 (1999).
- [14] We consider only isotropic scattering so that the transport length equals the scattering mean free path [13].
- [15] H. Cao, Waves Random Media **13**, R1 (2003).
- [16] A. Lagendijk and B. A. van Tiggelen, Phys. Rep. **270**, 143 (1996).
- [17] K.M. Case and P.F. Zweifel, *Linear transport theory* (Addison-Wesley, 1967).
- [18] K. Drozdowicz, E. Krynicka and J. Dąbrowska, App. Rad. Isot. **58**, 727 (2003).
- [19] B. R. Mollow, Phys. Rev. A **5**, 2217 (1972); F. Y. Wu *et al.*, Phys. Rev. Lett. **38**, 1077 (1977).
- [20] M. D. Barrett, J. A. Sauer and M. S. Chapman, Phys. Rev. Lett. **87**, 010404 (2001).
- [21] R. Pierrat and R. Carminati, Phys. Rev. A **76**, 023821 (2007).
- [22] S. Chandrasekhar, *Radiative Transfer* (Dover, New York, 1960).
- [23] R. Elaloufi, R. Carminati and J.-J. Greffet, J. Opt. Soc. Am. A **21**, 1430 (2004).
- [24] L. Khaykovich, N. Friedman and N. Davidson, Eur. Phys. J. D **7**, 467 (1999).