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Optical pumping in dense atomic media: Limitations due to reabsorption of spontaneously emitted photons

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Abstract. – Resonant optical pumping in dense atomic media is discussed, where the absorption length is less than the smallest characteristic dimension of the sample. It is shown that reabsorption and multiple scattering of spontaneous photons (radiation trapping) can substantially slow down the rate of optical pumping. A very slow relaxation out of the target state of the pump process is then sufficient to make optical pumping impossible. As model systems an inhomogeneously and a radiatively broadened 3-level system resonantly driven with a strong broad-band pump field are considered.

Introduction. – Optical pumping is an established technique in atomic and molecular physics to selectively populate or depopulate specific states or superpositions [1,2]. It is based on the absorption of photons of a specific mode and subsequent spontaneous emission into many modes. The dissipative nature of the latter part makes it possible to transform mixed into pure atomic states. From this there results the importance of optical pumping for state preparation in systems with a thermal distribution of population and for laser cooling [3].

The maximum achievable rate of pumping is determined by the escape time of the emitted photons, which in optically thin media is given by the free-space radiative lifetime. When the medium becomes optically thick, however, *i.e.* when the absorption length becomes smaller than the smallest sample dimension, the escape time of photons can be substantially reduced. This phenomenon, known as radiation trapping [4], is due to reabsorption and multiple scattering of spontaneously emitted photons and can drastically reduce the rate of optical pumping in dense media. These limitations could be of major importance in many different fields as, for instance, near-resonance linear and nonlinear optics in dense media [5,6] or the realisation of Bose condensation by velocity selective coherent population trapping (VSCPT) [7].

To describe the reabsorption and multiple scattering of photons we here utilize a recently developed approach to radiative interactions in dense atomic media [8]. In this approach a nonlinear and nonlocal single-atom density matrix equation is derived which generalizes the linear theory of radiation trapping [4] to the nonperturbative regime. As a model system a 3-level Λ configuration driven by a strong broad-band field is considered and the limits of i) large inhomogeneous and ii) purely radiative broadening are studied.

Let us consider the Λ-type system shown in fig. 1. A strong driving field with (complex) Rabi frequency $\Omega(t)$ couples the lower state $|c\rangle$ to the excited state $|a\rangle$, which spontaneously decays into $|c\rangle$ and $|b\rangle$. Since $|b\rangle$ is not coupled by the driving field, this results in optical

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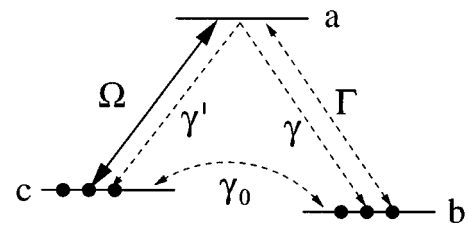

Fig. 1. – Optical pumping in a Λ system.

pumping from $|c\rangle$ to $|b\rangle$. We also take into account a possible finite lifetime of the target state described by a population exchange between the lower states at rate γ_0.

It was shown in [8] that the effect of the incoherent background radiation can be described by additional (nonlinear and nonlocal) pump and relaxation rates and level shifts in the single-atom density matrix equation. If we assume orthogonal dipole moments or sufficiently different frequencies of the two optical transitions, the level shifts are negligible. Also if the driving field is strong, the incoherent photons do not affect the pump transition $a \leftrightarrow c$. Thus we are left with a pump and decay rate $\Gamma(t)$ on the $a \leftrightarrow b$ transition and the effective single-atom equations of motion read in a rotating frame:

$$\dot{\rho}_{aa} = -(\gamma + \gamma' + \Gamma)\rho_{aa} + \Gamma\rho_{bb} + i(\Omega^*\rho_{ac} - \text{c.c.}), \tag{1}$$

$$\dot{\rho}_{cc} = \gamma'\rho_{aa} + \gamma_0\rho_{bb} - \gamma_0\rho_{cc} - i(\Omega^*\rho_{ac} - \text{c.c.}), \tag{2}$$

$$\dot{\rho}_{ac} = -(i\Delta_{ac} + \Gamma_{ac})\rho_{ac} + i\Omega(\rho_{aa} - \rho_{cc}). \tag{3}$$

Δ_{ac} is the detuning of the drive field from resonance and Γ_{ac} is the respective coherence decay rate. It should be noted that Γ is a function of the density matrix elements of all other atoms, and hence eqs. (1)-(3) are nonlinear and nonlocal.

We are here interested in genuine optical pumping and therefore consider a broad-band pump [9], *i.e.* $\Omega(t)$ is assumed to have a vanishing mean value and Gaussian δ-like correlations $\left\langle \Omega^*(t)\Omega(t')\right\rangle = R\,\delta(t-t')$. Formally intergating eq. (3), substituting the result back into eqs. (1) and (2), and averaging over the Gaussian distribution of the pump field leads to the rate equations

$$\dot{\rho}_{aa} = -(\gamma + \gamma' + \Gamma)\rho_{aa} + \Gamma\rho_{bb} - R(\rho_{aa} - \rho_{cc}), \tag{4}$$

$$\dot{\rho}_{cc} = \gamma'\rho_{aa} + \gamma_0\rho_{bb} - \gamma_0\rho_{cc} + R(\rho_{aa} - \rho_{cc}). \tag{5}$$

Collective decay rate. – We now have to determine the collective rate Γ. Γ is proportional to the spectrum of the incoherent field at the position $\vec{r}_0$ and the resonance frequency ω of the atom under consideration [8]:

$$\Gamma(\omega, t) = \frac{\wp^2}{\hbar^2}\widetilde{D}(\vec{r}_0, \omega; t) = \frac{\wp^2}{\hbar^2}\int_{-\infty}^{\infty} \mathrm{d}\tau\, \langle\langle \hat{E}^-(\vec{r}_0, t)\hat{E}^+(\vec{r}_0, t+\tau)\rangle\rangle\, e^{i\omega\tau}. \tag{6}$$

Here $\hat{E}^\pm$ are the positive and negative frequency parts of the field operators, $\wp$ is the dipole matrix element of the atomic transition, and $\langle\langle AB\rangle\rangle \equiv \langle AB\rangle - \langle A\rangle\langle B\rangle$. $\widetilde{D}(\omega)$ can be obtained by summing the spontaneous emission contributions of all atoms propagated through the medium [8]:

$$D(1,1) = \iint \mathrm{d}3\,\mathrm{d}4\, D^{\mathrm{ret}}(1,3)\left(D^{\mathrm{ret}}(1,4)\right)^* \Pi^{\mathrm{s}}(3,4). \tag{7}$$

Here $D^{\mathrm{ret}}(1,2)$ is the retarded propagator of the electric field inside the medium, which obeys

a Dyson-equation in self-consistent Hartree approximation:

$$D^{\mathrm{ret}}(1,2) = D_0^{\mathrm{ret}}(1,2) - \iint \mathrm{d}3\,\mathrm{d}4\, D_0^{\mathrm{ret}}(1,3)\,\Pi^{\mathrm{ret}}(3,4)\,D^{\mathrm{ret}}(4,2). \tag{8}$$

In eqs. (7) and (8) the numbers $1, 2\ldots$ stand for $\{\vec{r}_1, t_1\}, \{\vec{r}_2, t_2\}\ldots$, and the intergrations extend over time from $-\infty$ to $+\infty$ and over the whole sample volume. D_0^{ret} is the free-space retarded propagator of the electric field. For simplicity we here have disregarded polarisation. We have also introduced the atomic source correlation

$$\Pi^{\mathrm{s}}(\vec{r}_1, t_1; \vec{r}_2, t_2) = \frac{\wp^2}{\hbar^2} \sum_j \big\langle\big\langle \sigma_j^\dagger(t_1)\sigma_j(t_2)\big\rangle\big\rangle\, \delta(\vec{r}_1 - \vec{r}_j)\,\delta(\vec{r}_2 - \vec{r}_j) \tag{9}$$

and the atomic response function

$$\Pi^{\mathrm{ret}}(\vec{r}_1, t_1; \vec{r}_2, t_2) = \frac{\wp^2}{\hbar^2}\,\Theta(t_1 - t_2) \sum_j \big\langle\big[\sigma_j^\dagger(t_1), \sigma_j(t_2)\big]\big\rangle\, \delta(\vec{r}_1 - \vec{r}_j)\,\delta(\vec{r}_2 - \vec{r}_j)\,, \tag{10}$$

where $\sigma_j = |b\rangle_{jj}\langle a|$ is the spin-flip operator of the j-th atom and Θ is the Heaviside step function. In terms of the σ's the dipole operator of the j-th atom reads $d_j = \wp(\sigma_j + \sigma_j^\dagger)$. The names reflect the physical meaning of the quantities (9), (10). The Fourier transform of Π^{s} is proportional to the spontaneous emission spectrum of the atoms and that of Π^{ret} gives the susceptibility of the medium. Equations (7) and (8) represent a nonperturbative summation of the spontaneous radiation contributions of all atoms propagated through the medium. It assumes a Gaussian statistics, which is however a good approximation for the background radiation.

The Dyson equation (8) was solved in [8] with some approximations in a macroscopic (continuum) limit where $\Pi(\vec{r}_1, t_1; \vec{r}_2, t_2) = \int \mathrm{d}^3\vec{r}\, P(\vec{r}, t_1, t_2)\,\delta(\vec{r}_1 - \vec{r})\,\delta(\vec{r}_2 - \vec{r})$. This yielded for the collective decay rate

$$\Gamma(\omega; t) = \frac{\wp^2\omega^4}{(6\pi)^2\epsilon_0^2 c^4} \int_V \mathrm{d}^3\vec{r}\, \frac{e^{2q_0''(\vec{r},\omega;t)r}}{r^2}\, \widetilde{P}^{\,\mathrm{s}}(\vec{r}, \omega; t), \tag{11}$$

where $r = |\vec{r}_0 - \vec{r}|$ is the distance bewteeen the source and the probe atom. The probability that a photon reaches the probe atom is determined by the absorption coefficient

$$q_0''(\vec{r}, \omega, t) = \frac{\hbar\omega}{3\epsilon_0 c}\,\mathrm{Re}\,\left[\widetilde{P}^{\mathrm{ret}}(\vec{r}, \omega; t)\right]. \tag{12}$$

One can easily calculate the atomic source and response functions for the Λ system of fig. 1,

$$\widetilde{P}^{\mathrm{ret}}(\vec{r}_j, \omega, t) = \frac{\wp^2}{\hbar^2} N\, \overline{\frac{\rho_{aa}^j(t) - \rho_{bb}^j(t)}{\Gamma_{ab} + i(\omega - \omega_{ab}^j)}}\,, \tag{13}$$

$$\widetilde{P}^{\,\mathrm{s}}(\vec{r}_j, \omega, t) = \frac{2\wp^2}{\hbar^2} N\, \overline{\frac{\rho_{aa}^j(t)\Gamma_{ab}}{(\Gamma_{ab})^2 + (\omega - \omega_{ab}^j)^2}}\,, \tag{14}$$

where N is the density of atoms, ω_{ab}^j is the resonance frequency of the j-th atom, Γ_{ab} the coherence decay rate of the corresponding transition and the overbar denotes averaging over a possible inhomogeneous distribution of frequencies.

At this point we shall distinguish two limiting cases. We first consider the limit of large Doppler broadening and secondly the case of purely radiative broadening.

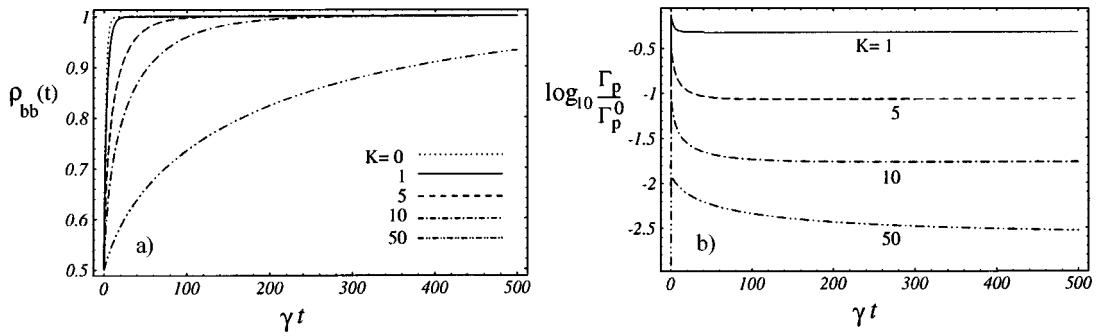

Fig. 2. – (a) Time evolution of population of level $|b\rangle$ for $\gamma_0 = 0$, $R/\gamma = 10$ and $\gamma'/\gamma = 1$ for different density parameters $K = g\,N\lambda^2 d_{\rm eff}$, $g = \gamma/\sqrt{2\pi}\Delta_{\rm D}$. (b) Corresponding effective rate of optical pumping.

Inhomogeneously broadened system. – The approach of [8] is based on the Markov approximation of a spectrally broad incoherent radiation. This approximation is justified for example in an inhomogeneously broadened system. We therefore discuss first the case of large Doppler broadening. If we are interested in the population dynamics on a time scale slow compared to velocity changing collissions, we may set $\rho^j_{\mu\mu}(t) = \overline{\rho^j_{\mu\mu}(t)} \equiv \rho_{\mu\mu}(\vec{r}_j, t)$ and thus have the same population dynamics in all velocity classes. Since Γ depends on the populations of all atoms, eqs. (4) and (5) are nonlocal. In the case of a constant density of atoms and a homogeneous pump field, Γ and hence all density matrix elements will be approximately homogeneous. We therefore make a simplifying approximation and disregard the space dependence. The volume integral is then carried out by placing the probe atom in the center of the sample. This yields for a Gaussian Doppler distribution of width $\Delta_{\rm D} \gg \gamma$

$$\frac{\Gamma(\omega,t)}{\gamma} = \frac{\rho_{aa}(t)}{\rho_{bb}(t) - \rho_{aa}(t)} \left[1 - \exp\left[-H(t)e^{-\Delta^2/2\Delta_{\rm D}^2}\right]\right], \tag{15}$$

where $\Delta = \omega - \omega^0_{ab}$ is the detuning from the atomic resonance at rest, and $H(t) = K\,[\rho_{bb}(t) - \rho_{aa}(t)]$. $K = g\,N\lambda^2 d_{\rm eff}$ with $g = \gamma/\sqrt{2\pi}\,\Delta_{\rm D}$ characterizes the number of atoms within one relevant velocity class in a volume given by the wavelength squared and the effective escape distance $d_{\rm eff}$. In deriving (15) we have used the relation between the free-space radiative decay rate γ and the dipole moment $\wp$: $\wp^2 = 3\pi\hbar\epsilon_0 c^3\gamma/\omega^3$ [10]. $d_{\rm eff}$ corresponds for a long cylindrical slab to the cylinder radius; for a thin disk to its thickness and for a sphere to its radius.

Averaging over the inhomogeneous velocity distribution of the atoms eventually yields

$$\begin{aligned}\Gamma(t) = \overline{\Gamma(\omega,t)} &= \int_{-\infty}^{\infty} {\rm d}\omega\, \frac{1}{\sqrt{2\pi}\Delta_{\rm D}} e^{-\Delta^2/2\Delta_{\rm D}^2}\, \Gamma(\omega,t) \\ &= \gamma\, \frac{\rho_{aa}(t)}{\rho_{bb}(t) - \rho_{aa}(t)} \frac{1}{\sqrt{\pi}} \int_{-\infty}^{\infty} {\rm d}y\, e^{-y^2} \left[1 - \exp\left(-H(t)e^{-y^2}\right)\right].\end{aligned} \tag{16}$$

In fig. 2a we have shown the population in the target state $|b\rangle$ as function of time starting from equal populations of levels $|c\rangle$ and $|b\rangle$ at $t = 0$. Here we have assumed that the target state is stable, *i.e.* $\gamma_0 = 0$. One recognizes that optical pumping is considerably slowed down already for values of K on the order of 10, which usually corresponds to much less than one atom per λ^3. The slow-down of pumping is further illustrated in fig. 2b, where the effective

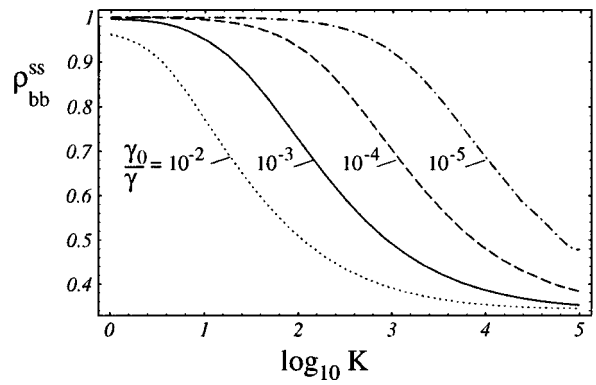

Fig. 3. – Stationary population in level $|b\rangle$ for $R/\gamma = 10$, $\gamma'/\gamma = 1$ and different values of γ_0 as a function of density parameter K.

pump rate defined as

$$\Gamma_{\mathrm{p}} \equiv -\frac{\mathrm{d}}{\mathrm{d}t}\ln[\rho_{aa} + \rho_{cc}] \tag{17}$$

is plotted normalized to the value in an optically thin medium ($\Gamma_{\mathrm{p}}^0 = \gamma/2$). One can see that the optical pump rate approaches a constant asymptotic value, which for $K \gg 1$ and large pump rates R is given by

$$\Gamma_{\mathrm{p}}^{\mathrm{as}} = \frac{\gamma}{2K\big(\pi \ln K\big)^{1/2}} \ll \frac{\gamma}{2}\,. \tag{18}$$

Since we have assumed in the plots of fig. 2 an infinitely long-lived target state ($\gamma_0 = 0$), all populations eventually end up in $|b\rangle$. However, if γ_0 is nonzero and in particular if it becomes comparable to the asymptotic rate Γ_p^{as}, the steady-state populations of all states equalize. In this case optical pumping is less and less efficient and becomes eventually impossible. This is illustrated in fig. 3, where the stationary population in state $|b\rangle$ is shown as a function of the density parameter K for different values of γ_0.

Radiatively broadened system. – We now discuss the case of a radiatively broadened system. In analogy to the case of inhomogeneous broadening, we find for the spectral distribution

$$\frac{\Gamma(\omega,t)}{\gamma} = \frac{\rho_{aa}(t)}{\rho_{bb}(t) - \rho_{aa}(t)}\left[1 - \exp\left[-H(t)\frac{\gamma_{ab}\Gamma_{ab}}{\Gamma_{ab}^2 + \Delta^2}\right]\right], \tag{19}$$

where $\Delta = \omega - \omega_{ab}$, $\Gamma_{ab} = \gamma_{ab} + \Gamma$, and $\gamma_{ab} = (\gamma + \gamma' + R + \gamma_0)/2$, and $H(t) = \widetilde{K}\,[\rho_{bb}(t) - \rho_{aa}(t)]$. Here $\widetilde{K} = \widetilde{g}\,N\lambda^2 d_{\mathrm{eff}}$ with $\widetilde{g} = \gamma/2\pi\gamma_{ab}$. As opposed to the corresponding relation in the inhomogeneous case, eq. (19) determines the collective decay rate only implicitly, and Γ needs to be calculated self-consistently. For small atomic densities or $\rho_{aa} \approx \rho_{bb}$ the exponential function in eq. (19) can be expanded into a power series. The first nonvanishing term found from this has the same spectral shape than the single-atom response function. In such a case the Markov approximation used in [8] is no longer valid and the approach is quantitatively incorrect. We shall nevertheless use it and discuss the range of validity afterwards.

We find that in the case of radiative broadening the rate of optical pumping decreases exponentially with the density parameter as opposed to $[N\lambda^2 d_{\mathrm{eff}}]^{-1}$ in the inhomogeneous case. For sufficiently large pump rates R and stable target state ($\gamma_0 = 0$) the asymptotic rate of optical pumping is here

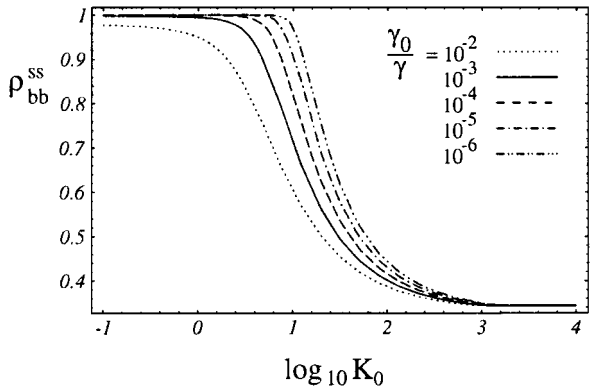

Fig. 4. – Same as fig. 3 for radiatively broadened system; $K_0 = N\lambda^2 d_{\rm eff}/2\pi$, $R/\gamma = 10$, $\gamma'/\gamma = 1$.

$$\Gamma_{\rm p}^{\rm as} = \frac{\gamma}{2}\exp\left[-\widetilde{K}\right]. \tag{20}$$

Physically, this is due to the fact that here the incoherent photons are in resonance with all atoms, which drastically increases the scattering probability. As a consequence, much smaller decay rates γ_0 out of the target state are sufficient to make optical pumping impossible. This is illustrated in fig. 4, where we have plotted the stationary population in state $|b\rangle$ as a function of the density parameter $K_0 = N\lambda^2 d_{\rm eff}/2\pi$ for different values of γ_0.

In order to check the validity of the Markov approximation, we have shown in fig. 5 the stationary normalized spectral distribution $\Gamma(\omega)/\Gamma$ for $K_0 = 1$, 10 and 100 and $\gamma_0/\gamma = 10^{-4}$. Also plotted is the atomic absorption spectrum for $K_0 = 1$ (solid line). One recognizes that the spectrum of the background radiation has only a slightly larger width than the atomic response for $K_0 = 1$. In this case the Markov approximation is not valid. The situation however improves when the density is increased. Thus fig. 4 has only qualitative character for lower densities.

Summary. – We have shown that resonant optical pumping in a dense atomic medium is substantially different from optical puming in dilute systems. When the absorption length of spontaneously emitted photons process becomes less than the minimum escape distance, these

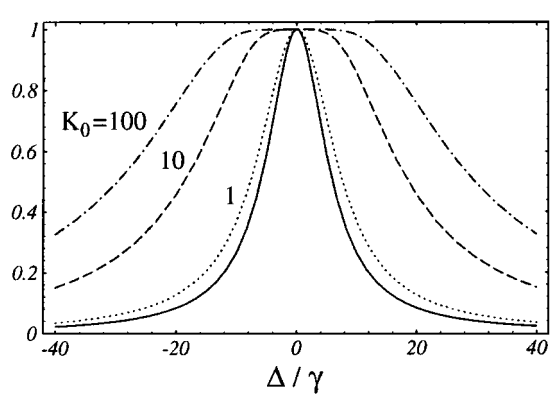

Fig. 5. – Spectral distribution of incoherent background radiation for $R/\gamma = 10$, $\gamma' = \gamma$, $\gamma_0/\gamma = 10^{-4}$, and $K_0 = 1$ (dotted), $K_0 = 10$ (dashed) and $K_0 = 100$ (dash-dotted). Also shown is the normalized absorption spectrum for $K_0 = 1$.

photons are trapped inside the medium and cause repumping of population. This leads to a considerable slow-down of the transfer rate and can make optical pumping impossible if the target state of the pump process has a finite lifetime. The effect is much less pronounced in inhomogeneously broadened systems due to the reduction of the spectral density of background photons.

These results may have some important consequences. It is practically impossile to use resonant optical pumping in media with $N\lambda^3 \sim 1$. This sets strong limits to the possibility to prepare pure states or coherent superpositions in systems with initial thermal occupation of states, such as hyperfine ground levels of alkali at room temperature. Even though the above analysis did not take into account quantum properties of the atoms and considers only resonant pumping, the results indicate that it may be very difficult to achieve Bose condensation via VSCPT in optical lattices [11]. Also the present results show that electromagnetically induced transparency (EIT) [12] in dense media cannot be understood as the result of optical puming into a dark state. Essential for EIT in dense media is an entirely coherent evolution [13] via stimulated adiabatic Raman passage [14]. Some of these aspects will be discussed in more detail elsewhere.

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