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Quantization of Atomic Motion in Optical Molasses.

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Abstract. – We present a quantum treatment of laser cooling of neutral atoms. The cooling mechanism studied is the «Sisyphus» process for 1-dimensional optical molasses. We first derive the energy eigenstates for the atom moving in the potential associated with the light-shifts due to the laser. Then, taking into account optical pumping, we calculate the steady-state populations of these quantum levels in the secular approximation. This approach allows us to determine the atomic eigenfrequencies in the optical wells, as well as momentum and position distributions. In particular, the minimum r.m.s. atomic momentum is $\simeq 6$ single-photon momenta; this result was not accessible to previous semi-classical treatments.

Laser cooling of free atoms in «optical molasses» has led in the recent years to extremely low atomic kinetic temperatures, in the micro-Kelvin range [1-3]. We present here a theoretical approach to this cooling; it is based on a quantum treatment of the external atomic motion for the case of 1-dimensional optical molasses, formed by the superposition of two opposing travelling waves. Our approach is in some ways similar to the one developed for trapped ions [4], although in our case the trapping potential is created by the laser light itself. Therefore this potential depends on the atomic internal state and is periodic, so that the energy spectrum consists of bands. From the steady-state populations of these energy levels, we then obtain the position and momentum distributions of the atoms inside the molasses. We also discuss various ways for measuring this energy spectrum.

We study here the simple case of «Sisyphus» cooling for an atom with a transition between a $J_g = 1/2$ ground state and a $J_e = 3/2$ excited state (fig. 1*a*)) [5]. The laser field is formed by the superposition of two plane waves, with a wavelength $\lambda = 2\pi/k$, travelling along the $+z$ and $-z$ directions, and respectively polarized along Ox and Oy. The resulting polarization of the light presents a spatially periodic gradient of ellipticity: for a convenient choice of phases, it is σ_- at $z = 0$, linear at $z = \lambda/8$, σ_+ at $z = \lambda/4$, ….

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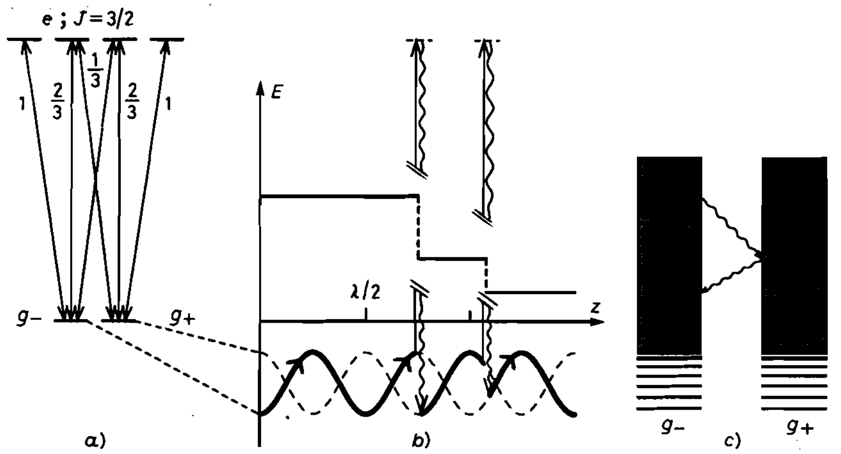

Fig. 1. – *a*) Atomic-level scheme and intensity factors (square of the Clebsch-Gordan coefficients) for a $J_g = 1/2 \leftrightarrow J_e = 3/2$ transition. *b*) Semi-classical description of Sisyphus cooling, with a «typical» random path showing a decrease of the total atomic energy (kinetic + potential) as the atom moves in the optical bipotential $U_\pm(z)$. *c*) Quantum description of Sisyphus cooling, showing the energy jumps sketched in *b*) as transition between eigenstates of the total atom-laser Hamiltonian. The energy spectrum corresponds to $U_0 = 100E_R$. In the steady state, atoms are mostly localized in the wells, *i.e.* more than half of the population is on the two lowest bands.

We restrict ourselves to the low saturation domain:

$$s_0 = \frac{\Omega^2/2}{\delta^2 + \Gamma^2/4} \gg 1\,, \tag{1}$$

which is known experimentally to lead to the lowest temperatures. When (1) is fulfilled, the atoms remain mainly in their internal ground-state sublevels. In (1), $\Omega = -2dE_0/\hbar$ is the Rabi frequency characterizing the coupling between the atomic dipole d and the field amplitude E_0 in each travelling wave, Γ is the natural width of the atomic excited state, and $\delta = \omega_L - \omega_A$ is the detuning between the laser (ω_L) and the atomic (ω_A) frequencies.

We also limit our treatment to situations where the average Doppler shifts can be neglected compared to Γ. This means that we do not take into account here «Doppler cooling» [4], and that we are left only with «polarization gradient cooling» [5, 6]. The effect of the atom-field coupling on the atomic dynamics can then be split into two parts [7].

First, the reactive part of this coupling (light-shift) consists in a periodic potential $U_\pm(z)$, depending on the atomic ground-state sublevel $g_\pm$:

$$U_\pm(z) = \frac{U_0}{2}(-2 \pm \cos(2kz)) \qquad \text{with } U_0 = -\frac{2}{3}\hbar\delta s_0\,. \tag{2}$$

For instance, at $z = 0$ where the light is σ_-, the sublevel g_- is three times more light-shifted ($-3U_0/2$) than the level g_+ ($-U_0/2$) because of the intensity factors sketched in fig. 1*a*); this is reversed at $z = \lambda/4$ where the light is σ_+.

Second, the atom-field coupling also has a dissipative part because of processes involving the absorption of a laser photon and the emission of a spontaneous photon. This dissipative coupling causes in particular real transitions to occur, via optical pumping, between g_+ and g_- and thus gives the states finite lifetimes.

If one treats classically the motion of the atom in the bi-potential $U_\pm(z)$, the analysis of Sisyphus cooling is then straightforward [5]. Take a negative detuning δ so that $U_0 > 0$, and suppose that the atom starts on level g_- (see fig. 1*b*)). As the atom moves in $U_-(z)$, it may undergo a transition to g_+. This transition occurs preferentially where the laser light is mostly σ_+ polarized, which corresponds to the tops of $U_-(z)$. The atom is then put in a valley for $U_+(z)$. This transition decreases the potential energy of the atom, while leaving its kinetic energy unchanged, if one neglects the momentum of the fluorescence photon involved in the process. From g_+, the same sequence can be repeated so that, on the average, the atom ascends more hills than it descends, which damps its velocity.

We now turn to the quantum treatment of Sisyphus cooling. We start with the master equation describing the time evolution of the atomic density matrix, including both internal and external degrees of freedom [8]. If (1) is fulfilled, we can adiabatically eliminate optical coherences as well as the excited-state part of the density matrix. We then get an equation of motion involving only the ground-state part σ of the density matrix and acting in the space generated by the vectors $|g_\pm, p\rangle$ (atom in internal state $g_\pm$ and with momentum p) [9]:

$$\dot{\sigma}(t) = \frac{1}{i\hbar}[H_0, \sigma(t)] + (\dot{\sigma}(t))_{\text{relax}} \tag{3}$$

with

$$H_0 = \frac{P^2}{2M} + U_+(Z)|g_+\rangle\langle g_+| + U_-(Z)|g_-\rangle\langle g_-| \tag{4}$$

and

$$(\dot{\sigma}(t))_{\text{relax}} = -\frac{\gamma_0}{2}(A\sigma(t) + \sigma(t)A) + \gamma_0 \int_{-\hbar k}^{\hbar k} \mathrm{d}p' \sum_m N_m(p') B_m^\dagger \exp[-ip'Z/\hbar]\,\sigma(t) \exp[ip'Z/\hbar] B_m. \tag{5}$$

M is the atomic mass, P and Z are the momentum and position operators of the atomic centre of mass and $\gamma_0 = 2\Gamma s_0/9$. The Hamiltonian H_0 contains the atomic kinetic energy and the potential energy corresponding to the reactive part of the atom-field coupling. The relaxation term (5), corresponding to the dissipative part of this coupling, has two contributions: the first one describes the departure from a given level, for instance from $|g_+, p_1\rangle$ to other levels, either $|g_+, p_2\rangle$ or $|g_-, p_2\rangle$, *via* an absorption-spontaneous emission cycle. The second term in (5) describes the reverse process, *e.g.*, the feeding of $|g_+, p_1\rangle$ from either $|g_+, p_2\rangle$ or $|g_-, p_2\rangle$. The integral is taken over the momentum p' of the spontaneous photon along Oz; the index m represents the projection of the photon angular momentum also along Oz and the functions $N_m(p')$ stand for the normalized distribution pattern for the spontaneous photon [8]. The operators A and B_m can be written as

$$A = \frac{3}{2}((2 - \cos 2kZ)|g_+\rangle\langle g_+| + (2 + \cos 2kZ)|g_-\rangle\langle g_-|), \tag{6a}$$

$$B_0 = \cos(kZ)|g_+\rangle\langle g_-| + \sin(kZ)|g_-\rangle\langle g_+|, \tag{6b}$$

$$B_1 = \frac{\sin(kZ)}{\sqrt{2}}(3|g_+\rangle\langle g_+| + |g_-\rangle\langle g_-|); \quad B_{-1} = \frac{\cos(kZ)}{\sqrt{2}}(|g_+\rangle\langle g_+| + 3|g_-\rangle\langle g_-|). \tag{6c}$$

We now look for the steady-state solution of (3). Here, we will restrict our analysis to the situation where the Hamiltonian part of (3) is predominant over the relaxation part. This requires that a typical Bohr frequency of the Hamiltonian H_0, *i.e.* the oscillation angular frequency ω_{osc} of the atom in the bottom of the wells of $U_\pm(z)$, be much larger than the typical damping rate γ_0:

$$\frac{\omega_{\text{osc}}}{\gamma_0} = \sqrt{\frac{27\hbar k^2|\delta|}{M s_0 \Gamma^2}} = 6\frac{|\delta|}{\Gamma}\sqrt{\frac{E_{\text{R}}}{U_0}} \gg 1, \tag{7}$$

where $E_R = \hbar^2 k^2/2M$ is the recoil energy. We see from (7) that our treatment is valid, for a given potential depth U_0, in the limit of large detunings. This is known experimentally to lead to the lowest temperatures [3]. Classically, this situation corresponds to the case of particles performing a large number of oscillations on a given potential $U_+(z)$ or $U_-(z)$ before jumping to the other one.

When (7) is satisfied, the procedure is straightforward. We first look for the eigenstates and the energy spectrum of H_0. We find, as usual for periodic potentials, alternating bands of allowed and forbidden energies (fig. 1*c*)). The eigenstates can be labelled as $|n, q, \varepsilon\rangle$, where n is an integer $\geqslant 0$ labelling the band, and where $\varepsilon = \pm 1$ stands for the internal state $g_\pm$. q is the Bloch index, chosen in the first Brillouin zone $(-k < q \leqslant k)$, and which takes discrete values since we use here standard periodic boundary conditions in a box with a size large compared to the spatial period $\lambda/2$. We note that the two states $|n, q, \pm\rangle$ have the same energy $E_{n,q}$ due to the symmetry between $U_\pm$. This eigenvalue problem can be cast into a universal one (Mathieu equation) if one expresses both U_0 and $E_{n,q}$ in terms of the recoil energy E_R (fig. 2*a*)) (¹). For a «typical» laser cooling situation, $U_0 = 100E_R$, obtained with cesium atoms for instance with $\delta = -20\Gamma$ and $\Omega = 1.5\Gamma$, one finds $\omega_{osc}/2\pi \simeq 40$ kHz, with 6 bands corresponding to bound states ($E_{n,q} < -U_0/2$); the width of the lowest band, $n = 0$, is extremely small ($< 10^{-6} E_R$). The number of such «bound bands» increases as $\sqrt{U_0/E_R}$, as does the splitting $\hbar\omega_{osc}$ between two adjacent bands.

Now we take into account the relaxation part of (3), which causes transitions between the various $|n, q, \varepsilon\rangle$ (fig. 1*c*)). Since γ_0 is very small compared to ω_{osc}, we can use a secular approximation and assume that σ is diagonal in the basis $|n, q, \varepsilon\rangle$ in steady state (²). Averaging (5) in a given $|n, q, \varepsilon\rangle$, we find the following relation between the steady-state

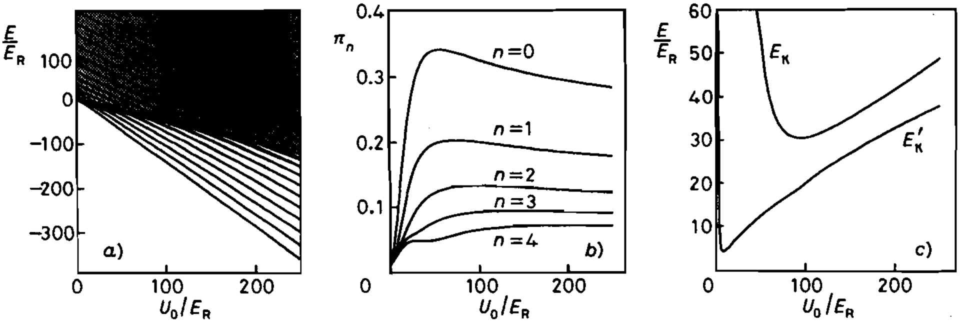

Fig. 2. – *a*) Band structure of the energy spectrum of H_0, plotted as a function of the potential depth U_0. The shaded areas correspond to allowed energies. For a given U_0, the energies above $-U_0/2$ corresponding to an above-barrier motion (see eq. (2)) are mostly allowed (quasi-free motion). On the opposite, energy bands corresponding to a bound classical motion ($-3U_0/2 < E < -U_0/2$) are very narrow except in the immediate vicinity of $-U_0/2$. *b*) Steady-state population of the various energy bands, as a function of U_0. *c*) Steady-state kinetic energies $E_K = p^2_{r.m.s.}/2M$ and $E'_K = \delta p_e^2/2M$ (where δp_e is the halfwidth at $1/\sqrt{e}$ of the momentum distribution) as a function of U_0. These two quantities would be equal for Gaussian momentum distributions.

(¹) A similar treatment has been applied to the case of a 2-level atom moving in a standing wave, in the absence of spontaneous emission [10].

(²) If σ is a solution of (3) in steady state, then $T\sigma T^\dagger$, where T is the $\lambda/2$ spatial translation operator, is also a solution. The uniqueness of steady state therefore implies $\sigma = T\sigma T^\dagger$, so that any nondiagonal matrix element inside a band $\langle n, q_1, \varepsilon|\sigma|n, q_2, \varepsilon\rangle$ is zero, although the secular approximation argument cannot be applied to it. On the other hand, one can show that the spatial coherence of σ in steady state is restricted to a fraction of wavelength because of spontaneous emission processes.

population $\Pi_{n,q,\varepsilon}$:

$$0=\dot{\Pi}_{n,q,\varepsilon}=-\gamma_{n,q,\varepsilon}\Pi_{n,q,\varepsilon}+\sum_{n',q',\varepsilon'}\gamma(n',q',\varepsilon'\rightarrow n,q,\varepsilon)\Pi_{n',q',\varepsilon'}\,, \tag{8a}$$

$$\gamma_{n,q,\varepsilon}=\gamma_0\langle n,q,\varepsilon\,|\,A\,|\,n,q,\varepsilon\rangle\,, \tag{8b}$$

$$\gamma(n',q',\varepsilon'\rightarrow n,q,\varepsilon)=\gamma_0\int_{-\hbar k}^{\hbar k}\mathrm{d}p'\sum_m N_m(p')\,|\,\langle n',q',\varepsilon'\,|\,B_m\exp[ip'Z/\hbar]\,|\,n,q,\varepsilon\rangle\,|^2\,. \tag{8c}$$

This expresses an equilibrium between the population leaving $|n, q, \varepsilon\rangle$ (first term of (8*a*)) and the population arriving in $|n, q, \varepsilon\rangle$ (second term of (8*a*)). By introducing a closure relation into (8*c*), we can show that $\gamma_{n,q,\varepsilon}$ calculated from (8*b*) is equal to the sum over the primed variables of the $\gamma(n, q, \varepsilon\rightarrow n', q', \varepsilon')$. Equation (8*a*) associated with the normalization condition allows one to completely determine the various populations $\Pi_{n,q,\varepsilon}$ in equilibrium, and therefore to characterize the steady state completely. This steady state usually depends on two parameters, Rabi frequency Ω and detuning δ, or equivalently U_0 and γ_0. We have already noticed that the energy spectrum of H_0 depends only on U_0. Now, we see that γ_0 can be factorized in eq. (8*a*), which means that the steady-state populations $\Pi_{n,q,\varepsilon}$ do not depend on γ_0 in the limit (7). In this limit, U_0 is therefore the only important parameter to characterize the steady state.

On the other hand, the rates (8*b*) and (8*c*) are proportional to γ_0, which indicates that the «cooling time» varies as γ_0^{-1}. Two types of transitions are involved in the relaxation process. Consider first a transition where there is a change of internal state: $g_\varepsilon\rightarrow g_{-\varepsilon}$. These are, as we have seen in the semi-classical approach, at the origin of the Sisyphus cooling (fig. 1*b*, *c*)). The corresponding departure rate $\bar{\gamma}_{n,q,\varepsilon}$ from a given level $|n, q, \varepsilon\rangle$ to all $|n', q', -\varepsilon\rangle$ is found to be equal to the average in the state $|n, q, \varepsilon\rangle$ of the semi-classical rates of transfer $\gamma_0\cos^2 kz$ ($\gamma_0\sin^2 kz$) from g_+ to g_- (g_- to g_+). If these transitions $\varepsilon\rightarrow-\varepsilon$ were the only ones, one would find a strong accumulation of the atoms in the lowest band $n=0$, where $\bar{\gamma}_{n,q,\varepsilon}$ is very small. Actually, this strong accumulation in the lowest level is partially counterbalanced by transitions $\varepsilon\rightarrow\varepsilon$ with different values of q and n. These transitions correspond to a heating, due for instance to the randomness of the momenta of the emitted fluorescence photons.

In order to calculate numerically the steady-state populations, we have taken into account the first 80 bands, with 6 values for q in each band corresponding to the discrete values of q for our chosen boundary conditions. We have checked that a larger quantization volume makes no noticeable difference. On the other hand, 80 bands, among which less than 10 are bound, are required to get accurate values for steady-state r.m.s. momenta. The variations of the total populations π_n of the first five bands *vs.* U_0 are shown in fig. 2*b*): for instance, π_0 is maximal around $U_0\simeq 60E_{\mathrm{R}}$, and reaches 0.34; in this case, the total population of unbound bands ($E_{n,q}\geqslant -U_0/2$) is 0.2, which suggests a spatial localization of the atoms. This is confirmed by the calculation of the spatial distribution $\rho(z)$ using the z representation of the $|n, q, \varepsilon\rangle$. The modulation $\eta=\max(\rho)/\min(\rho)$ of these distributions follows the empirical law $\eta\simeq(U_0/10E_{\mathrm{R}})^{1/4}$, from $U_0=20E_{\mathrm{R}}$ up to the largest U_0 that we have considered, *i.e.* $1000E_{\mathrm{R}}$. Such a localization of atoms in 3D Na molasses has recently been observed [11]. The set of $\Pi_{n,q,\varepsilon}$ also gives the momentum distribution in steady state. We have plotted in fig. 2*c*) the average kinetic energy, $E_{\mathrm{K}}=p^2_{\mathrm{r.m.s.}}/2M$, as a function of U_0. It is minimal for $U_0\simeq 95E_{\mathrm{R}}$, $p_{\mathrm{r.m.s.}}$ being then of the order of $5.5\hbar k$. We recall that one finds experimentally for 3D Na or Cs molasses a minimum $p_{\mathrm{r.m.s.}}$ of 3 or $4\hbar k$ [3, 12] (^{3}).

(^{3}) Experimental measurements are usually done by a determination of the width of momentum distributions, which is more in the spirit of the curve E'_{K} of fig. 2*c*).

These results can be compared with the ones given by usual semi-classical approaches [5, 6]. We note that the validity condition (7) is the reverse of the one required for these approaches, where one assumes that the atoms move sufficiently slowly so that their internal state can follow nearly adiabatically their external motion; when (7) holds, it is not possible to describe the atomic motion in terms of a friction force linear in velocity and a constant momentum diffusion coefficient. Actually, in spite of these opposite conditions on $\omega_{\text{osc}}/\gamma_0$, both approaches predict a similar linear dependence of E_K with U_0, for large U_0; the slopes are comparable, 0.14 here (fig. 2*c*)) instead of 0.19 semi-classically [5]. On the other hand, even when (7) holds, a semi-classical treatment is possible. It uses as a slow variable the total atomic energy instead of the atomic velocity and gives results close to the ones obtained here [9].

In addition to momentum or position distributions, this approach gives other observable quantities of laser-cooled atomic samples, more deeply connected to the quantization of atomic motion. The fluorescence spectrum of the atoms inside the molasses [11] can give a way for observing this discrete structure of the energy spectrum. It requires that the induced radiative width $\simeq \gamma_0$ of the ground-state sublevels should be smaller than the splitting between the bands, $\simeq \omega_{\text{osc}}$, which just corresponds to the condition of validity of our treatment (7). One could also measure, inside the molasses, the absorption or the amplification of a weak probe laser with a frequency shift from ω_L of the order of $\pm\,\omega_{\text{osc}}$.

Finally, we note that our treatment can be generalized to more complicated physical situations. We have recently extended it to the case of a $J_g = 1 \leftrightarrow J_e = 2$ transition in the same laser configuration. The generalization of this treatment to 2 and 3 dimensions is also under way. The diagonalization of H_0 is then much more complicated because the eigenvalue problem is not factorizable into 2 or 3 1D problems.

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