

Dark Resonances

From Optical Pumping to Cold Atoms and Molecules

Claude Cohen-Tannoudji
Collège de France and Laboratoire Kastler Brossel*
Département de Physique de l'Ecole Normale Supérieure
24 rue Lhomond F - 75231 Paris Cedex 05

November 11, 2009

1 Introduction

Coherent Population Trapping (CPT) is a very beautiful example of quantum interference effect. It was discovered in 1976 in the group of Adriano Gozzini in Pisa [1]. “Dark resonances” were observed in optical pumping experiments and subsequently interpreted as a quenching of fluorescence due to a destructive interference between two absorption amplitudes connecting two ground state sublevels g_1 and g_2 to an excited sublevel e . The atom can absorb light from g_1 and jump to e . Similarly, it can absorb light from g_2 and jump to e . But, if it is in a certain linear superposition of g_1 and g_2 , $c_1g_1 + c_2g_2$, the two absorption amplitudes $g_1 \rightarrow e$ and $g_2 \rightarrow e$ interfere and cancel out. The fluorescence stops. The state $c_1g_1 + c_2g_2$ from which light cannot be absorbed is called a “dark state”.

Since that time, dark resonances and dark states appeared to play an essential role in several new physical effects, like Electromagnetically Induced Transparency (EIT), Stimulated Raman Adiabatic Passage (STIRAP), Velocity Selective Coherent Population Trapping (VSCPT) and subrecoil laser cooling, dark resonances in photo-association spectra. We try in this paper to give a review of the various developments having occurred in this research field, putting the emphasis on the interpretation of the physical phenomena. We will not enter into technical details and our bibliography cannot be considered as exhaustive.

*Unité de Recherche de l'Ecole normale supérieure et de l'Université Pierre et Marie Curie, associée au CNRS. Institut Francilien de Recherches sur les Atomes Froids (IFRAF).

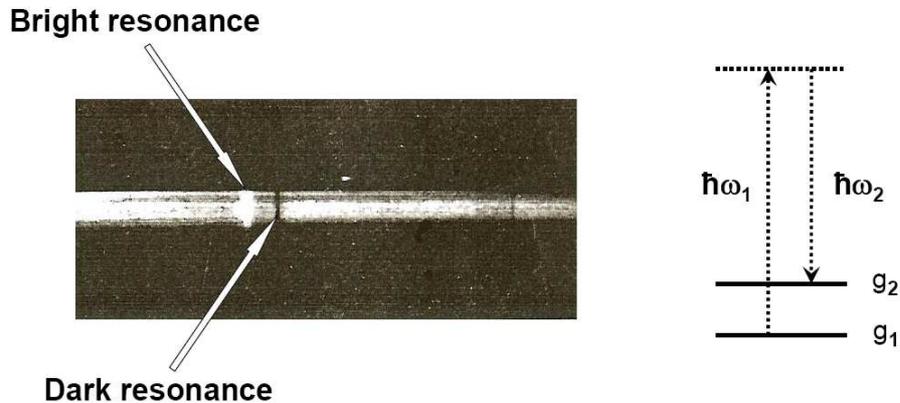


Figure 1: Bright and dark resonances observed in the fluorescence light emitted by a sodium vapor put in a spatially inhomogeneous magnetic field. The dark resonance appears at a point where the frequency difference $\omega_1 - \omega_2$ between two laser modes matches the frequency splitting between two Zeeman sublevels g_1 and g_2 .

2 Early works on dark resonances

The Pisa experiment [1] was using an optically pumped sodium vapor put in a spatially inhomogeneous magnetic field along the z -axis. The splitting between Zeeman sublevels is thus z -dependent. If one applies a radiofrequency field with frequency ω_{RF} , it induces resonant transitions between two Zeeman sublevels g_1 and g_2 only at the point z where $E_{g_2} - E_{g_1} = \hbar\omega_{RF}$. The population difference between g_1 and g_2 is modified by the RF resonant transitions and this results in a modification of the fluorescence light (generally an increase) at the points where the resonance condition is fulfilled. One observes a series of bright lines in the fluorescence emitted along the path of the laser beam in the cell (see Fig.1), forming a spatially resolved RF spectrum.

The important result of this experiment was the appearance of dark resonances (no fluorescence) at certain points along the path of the laser beam (see Fig.1), which remain present if the RF field is switched off, provided however that the laser beam is multimode. It was readily found that dark resonances appear for values of z such that the frequency difference between two laser modes is equal to the frequency splitting between two Zeeman sublevels g_1 and g_2 belonging to the hyperfine states of the sodium atom:

$$E_{g_2} - E_{g_1} = \hbar\omega_1 - \hbar\omega_2 \quad (1)$$

This equation is a resonance condition for the stimulated Raman transitions between g_1 and g_2 . One photon is absorbed in one laser mode, another one emitted in a stimulated way in another mode, the atom going from one Zeeman

sublevel to another one with conservation of the total energy.

The first theoretical treatment of dark resonances [2] was using optical Bloch equations (OBE). A three-level atom $\{g_1, g_2, e\}$ is interacting with two laser beams: $\vec{E}_1 \exp(i \vec{k}_1 \cdot \vec{r} - \omega_1 t)$ exciting only the transition $g_1 \leftrightarrow e$ and $\vec{E}_2 \exp(i \vec{k}_2 \cdot \vec{r} - \omega_2 t)$ exciting only the transition $g_2 \leftrightarrow e$. Within the rotating wave approximation, OBE become a set of first order coupled differential equations with time-independent coefficients which can be solved exactly. One finds that, when the detuning Δ from the resonance Raman condition is equal to 0, the population σ_{ee} of the excited state e vanishes: there is no fluorescence. Simultaneously, the off diagonal element $\sigma_{g_1 g_2}$ of the atomic density matrix σ between g_1 and g_2 takes a large value. This means that atoms are put in a linear combination of the two lower states g_1 and g_2 . The OBE approach gives a quantitative description of the dark resonances but the physical reason why condition (1) is essential for the quenching of fluorescence does not appear clearly. We present in the following section a simple physical interpretation of this Raman resonance condition. Let us also mention that dark resonances can be simply related to the radiative cascade of the atom "dressed" by the two types of photons ω_1 and ω_2 (see for example [3], chapter VI and [4])

3 Interpretation of the Raman resonance condition

We consider in this section an atom at rest in $\vec{r} = \vec{0}$. We will discuss later on the case of a moving atom.

(i) *Expression of the dark state at time $t = 0$.*

Consider an atom in the state:

$$|\psi(t=0)\rangle = c_1 |g_1\rangle + c_2 |g_2\rangle \quad (2)$$

$c_1(c_2)$ is the amplitude for the atom to be in $g_1(g_2)$. Let us introduce the Rabi frequencies characterizing the interaction with the two laser fields:

$$\Omega_i(t=0) = -\vec{D}_{eg_i} \cdot \vec{E}_i / \hbar \quad i = 1, 2 \quad (3)$$

where \vec{D}_{eg_i} is the matrix element of the dipole moment operator between e and g_i . The amplitude to have an absorption process from g_i to e is equal to the amplitude c_i for the atom to be in g_i times the amplitude to absorb a photon from g_i which is proportional to Ω_i . If the amplitude c_1 and c_2 appearing in Eq.(2) are such that:

$$c_1 \Omega_1 + c_2 \Omega_2 = 0 \quad (4)$$

the two absorption amplitudes interfere destructively and the atom cannot be excited. A state (2) obeying condition (4) is called a dark state.

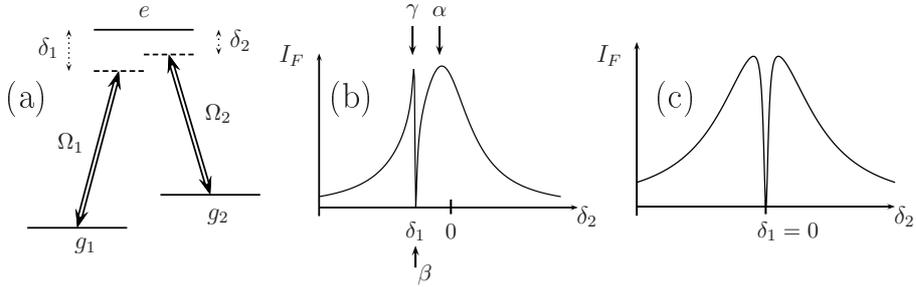


Figure 2: (a): detuning δ_1 and δ_2 of the 2 lasers from the frequencies of the 2 atomic transitions $e \leftrightarrow g_1$ and $e \leftrightarrow g_2$. δ_1 is fixed and δ_2 is varied. (b): Variations of the fluorescence intensity R_F with δ_2 for a non zero value of δ_1 . The vertical arrows indicate the resonant excitation of the transition $e \leftrightarrow g_2$ (α) which appears when $\delta_2 \simeq 0$, the dark resonance corresponding to the Raman resonance between unperturbed states (β) which appears when $\delta_2 = \delta_1$, the resonant Raman transition between perturbed states (γ) (c): Variations of R_F with δ_2 when $\delta_1 = 0$.

(ii) *If a state is dark at time $t = 0$, does it remain dark at a later time?*

We suppose that the state remains dark as time goes on and we try to find the condition for this to happen. If the state remains dark, this means that there is no interaction between the atom and the laser fields and the time evolution of the atom is a free evolution as if the laser fields were switched off. The coefficients c_i of the state (2) acquire a phase factor $\exp(-i E_i t / \hbar)$ where E_i is the unperturbed energy of the state g_i , without any light shift. The laser fields E_i acquire a phase factor $\exp(-i \omega_i t)$ due to their frequencies ω_i . The total absorption amplitude thus becomes:

$$c_1 \exp(-i E_1 t / \hbar) \Omega_1 \exp(-i \omega_1 t) + c_2 \exp(-i E_2 t / \hbar) \Omega_2 \exp(-i \omega_2 t). \quad (5)$$

It remains equal to 0, like in $t = 0$, only if the two phase factors multiplying $c_1 \Omega_1$ and $c_2 \Omega_2$ are the same, i.e. if:

$$(E_1 / \hbar) + \omega_1 = (E_2 / \hbar) + \omega_2 \Leftrightarrow E_2 - E_1 = \hbar (\omega_1 - \omega_2). \quad (6)$$

One recovers the Raman resonance condition (1) and one understands now why the dark resonance appears only when this condition is fulfilled. If it is not fulfilled, a state which is dark at a given time does not remain dark at a later time and can then absorb light. This approach shows also clearly that the resonance Raman condition involves the unperturbed atomic states without any light shifts. In equation (1), the atomic energies E_1 and E_2 are unperturbed energies.

(iii) *Various resonances appearing in the fluorescence signal.*

The previous discussion allows one to understand the variations of the fluorescence light R_F emitted by the atom when the detuning $\delta_1 = \omega_1 - \omega_{eg_1}$ of the first laser from the frequency ω_{eg_1} of the transition $e \leftrightarrow g_1$ is kept fixed while the other detuning $\delta_2 = \omega_2 - \omega_{eg_2}$ is scanned (see Fig. 2a). Consider first the case where the detuning δ_1 is different from zero (Fig. 2b). A broad resonance (vertical arrow α) appears in the variations of R_F with δ_2 when $\delta_2 \simeq 0$ and corresponds to the resonant excitation of the transition $e \leftrightarrow g_2$ by the laser 2. The width of this resonance is equal to the natural width Γ of the excited state e . A narrow dip (vertical arrow β) with a vanishing of R_F appears when $\delta_2 = \delta_1$, i.e. when the Raman resonance condition between unperturbed states is fulfilled. This is the dark resonance. Finally, we have a narrow peak (vertical arrow γ) appearing when the Raman resonance condition between perturbed states is fulfilled. Resonant stimulated transitions between the light shifted states g_1 and g_2 are induced by the 2 lasers and modify R_F . The dip β and the peak γ of Fig. 2b are narrow because their width is determined by the coherence relaxation time in the ground state which is (for low enough laser intensities) much longer than the radiative lifetime $1/\Gamma$ of the excited state e . Note finally the asymmetry of the variations of R_F with δ_2 in the neighborhood of the dark resonance. They can be interpreted in terms of Fano profiles [5]. When $\delta_1 = 0$ (see Fig. 2c), the curve giving R_F versus δ_2 becomes symmetric with still a narrow dip going to zero, corresponding to the dark resonance appearing inside a broad structure corresponding to the resonant excitation of the transition $e \leftrightarrow g_2$ by the laser 2¹.

Note finally that the dark resonance exists only if the two laser fields are coherent. If their phases changes in a random way, the phase difference between the two absorption amplitudes appearing in Eq. (5) changes also in a random way so that the destructive interference cannot hold for all times. If the frequency splitting between g_1 and g_2 falls in the radiofrequency or in the microwave domains, a convenient way for generating the two frequencies ω_1 and ω_2 is to start with a single laser beam and to modulate its amplitude or its frequency at a frequency near $\omega_1 - \omega_2$, in order to generate sidebands. An important advantage of this method is that, even if the laser has a frequency jitter, this jitter is the same for the central frequency and the sidebands so that it cancels out in the difference $\omega_1 - \omega_2$. The position of the dark resonance is thus insensitive to the frequency jitter, which explains the great interest of dark resonances for high resolution laser spectroscopy (for a review see [?]).

¹Note that we suppose here that the two lasers have not an intensity high enough to saturate the atomic transitions. The Rabi frequencies are small compared to Γ . If the laser 1 saturates the atomic transition $e \leftrightarrow g_1$, one observes an Autler-Townes doublet in the variations of R_F with δ_2 , consisting in two peaks separated by an interval equal to the Rabi frequency Ω_{eg_1} (see for example [3], Chapter VI). But we have always $R_F = 0$ when the resonance Raman condition between unperturbed states is fulfilled, i.e. when $\delta_2 = \delta_1 = 0$. The destructive interference between the two absorption amplitudes $g_1 \rightarrow e$ and $g_2 \rightarrow e$ still exists but we no longer have a narrow dip around $\delta_2 = 0$.

4 A few applications of dark resonances.

(i) *Electromagnetically induced transparency (EIT).*

Consider an atomic vapor of three-level atoms $\{e, g_1, g_2\}$, having a great optical depth for a laser ω_1 exciting the transition $e \leftrightarrow g_1$, so that this laser beam is fully absorbed by the vapor. If one adds a second laser beam coherent with the first one and exciting the transition $e \leftrightarrow g_2$, with a frequency ω_2 such that the resonance Raman condition between unperturbed states is fulfilled, the absorption coefficient vanishes because of the dark resonance and the vapor becomes transparent. For a review on this effect see Ref. [6].

(ii) *Slow light.*

With the narrow dip in the absorption coefficient due to the dark resonance is associated a rapid variation of the refractive index of the vapor in the vicinity of the dip. This can change dramatically the group velocity v_g of light in the vapor. Using dark resonances for reducing v_g is one of the methods which have been used to produce slow light [7].

(iii) *Stimulated Raman adiabatic passage (STIRAP).*

In a dressed atom description of coherent population trapping (see for example [3], chapter VI and [4]), the dark state is a linear combination of states labelled by an atomic quantum number, g_1, g_2 or e , and by the numbers N_1 and N_2 of photons in the two laser modes:

$$|\psi_D\rangle = c_1 \Omega_1 |g_1, N_1 + 1, N_2\rangle + c_2 \Omega_2 |g_2, N_1, N_2 + 1\rangle \quad (7)$$

This state is not coupled by the interaction Hamiltonian V to the state $|e, N_1, N_2\rangle$ and is an eigenstate of the unperturbed Hamiltonian H_0 (the two states appearing in (7) are two degenerate states of H_0 , which corresponds to the resonance Raman condition between unperturbed states). The energy separation between the dark state and the other dressed states (eigenstates of the total Hamiltonian $H_0 + V$) is on the order of the Rabi frequencies Ω_1 and Ω_2 and detunings δ_1 and δ_2 . If one sweeps slowly enough Ω_1 and Ω_2 , i.e. the intensities of the lasers, the system remains in the dark state corresponding to the instantaneous values of Ω_1 and Ω_2 .

Suppose that one starts in a state for which $\Omega_1 = 0$ and $\Omega_2 \neq 0$. The state g_1 is then a dark state because an atom in g_1 cannot absorb a photon ω_2 . If one slowly decreases Ω_2 and increase Ω_1 until Ω_2 vanishes, the dark state will evolve adiabatically to a dark state for which $\Omega_1 \neq 0$ and $\Omega_2 = 0$. The system will have been transferred adiabatically from g_1 to g_2 . Note that this way of changing Ω_1 and Ω_2 is counterintuitive for transferring the atom from g_1 to g_2 . One would rather think that laser ω_1 should be first used to transfer the atom from g_1 to e , the laser ω_2 being then used to transfer by stimulated emission the atom from e to g_2 . In fact, we don't use in STIRAP any real absorption or stimulated emission process. One follows adiabatically a dark state where the

excited state e is never populated. The advantage of STIRAP, first introduced in molecular physics [8] (for a review on STIRAP, see also [9]), is to avoid any spontaneous emission process from e , which would populate rovibrational states other than g_2 where one wants to transfer molecules selectively.

5 Velocity selective coherent population trapping (VSCPT).

In section 3, we have supposed that the atom is at rest in $\vec{r} = \vec{0}$ and we have shown that, if the state is dark at $t = 0$, it remains dark at a later time only if the resonance Raman condition between unperturbed states is fulfilled. We consider now the case where the atom is moving with a velocity \vec{v} and we first use here a classical treatment of atomic motion consisting of replacing \vec{r} by $\vec{v}t$. We will discuss later on situations where a quantum treatment of atomic motion becomes necessary.

Replacing \vec{r} by $\vec{v}t$ in the exponentials $\exp(i\vec{k}_i \cdot \vec{r} - \omega_i t)$ with $i = 1, 2$ appearing in the expression of the two laser electric fields gives rise to additional phase factors $\exp(i\vec{k}_i \cdot \vec{v}t)$ multiplying $c_1\Omega_1$ and $c_2\Omega_2$ in (5). If the state is dark at $t = 0$, it remains dark at a later time for a moving atom only if

$$\vec{k}_1 \cdot \vec{v} = \vec{k}_2 \cdot \vec{v} \quad (8)$$

The physical interpretation of this condition is clear. For a moving atom, the apparent laser frequencies are Doppler shifted by amounts equal to $\vec{k}_1 \cdot \vec{v}$ and $\vec{k}_2 \cdot \vec{v}$, respectively. In a Raman stimulated process these two Doppler shifts add with a negative sign. The resonance Raman condition remains fulfilled only if the two Doppler shifts are equal.

An interesting situation occurs when

$$\vec{k}_1 \cdot \vec{v} \neq \vec{k}_2 \cdot \vec{v} \quad (9)$$

The state which is dark when $\vec{v} = \vec{0}$ is no longer dark when the atom is moving. An atom at rest cannot absorb light. But, as soon as it is moving, photon absorption processes can take place. Coherent population trapping becomes velocity selective. The absorption rate $R(v)$ is then velocity dependent. It vanishes for $v = 0$ and its variations with v have the shape represented in Fig. 3a.

In a one-dimensional situation, a convenient way for having condition (9) fulfilled is to take two counterpropagating laser beams. The difference between the two Doppler shifts takes then the highest possible value leading to the highest velocity dependence for the dark states. If the two laser beams are copropagating, the difference between the two Doppler shifts is on the order of the Doppler width of the $g_1 \leftrightarrow g_2$ transition. If this transition falls in the microwave domain, this Doppler width can be smaller than the width of the dark resonance (inverse of the coherence time in the ground state). One can therefore ignore

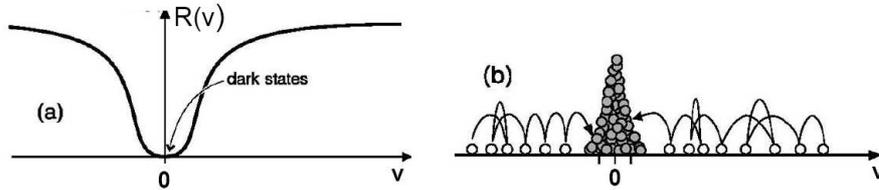


Figure 3: Principle of laser cooling by velocity selective coherent population trapping (VSCPT). (a): the fluorescence rate $R_F(v)$ vanishes for $v = 0$. (b): The atoms perform a random walk in velocity space and accumulate in the vicinity of $v = 0$.

the effect of atomic motion. Most experimental implementations of the applications discussed in the previous section use this configuration of copropagating beams. Using laser cooled atoms still improves the situation by reducing the contribution of the Doppler width to the width of the dark resonance.

6 Laser cooling based on VSCPT.

(i) Principle of the method.

An atom interacting with laser beams undergoes fluorescence cycles consisting of absorption processes followed by spontaneous or stimulated emission processes. Since spontaneous photons can be emitted in random directions, the atomic momentum changes in a random way after each spontaneous emission process by an amount on the order of the photon momentum $\hbar k$. The motion of the atom in momentum space is thus a random walk with steps on the order of $\hbar k^2$. Since this random change of momentum cannot be controlled, it seems at first sight impossible to reduce the momentum spread of an ensemble of atoms below the recoil limit $\hbar k$.

Suppose now that the atom interacts with a VSCPT laser configuration leading to velocity dependent absorption rate $R(v)$ represented in Fig. 3a. Atoms with zero velocity (or with v very close to $v = 0$) are protected from the “bad effects” of light, associated with the random recoils due to spontaneous emission processes following the absorption of photons. Atoms with non zero velocity undergo fluorescence cycles. The velocity changes associated with the random recoils due to spontaneous emission can make them falling in the region close to $v = 0$ where they remain trapped and accumulate. This cooling scheme is thus based, not on a friction mechanism, but on a velocity dependent random

²In most usual laser cooling schemes, there is in addition a friction force which damps the mean value of the atomic momentum.

walk in velocity space with a trap near $v = 0$ in this space where atoms can accumulate.

Note that the longer the interaction time θ , the narrower the interval δv around $v = 0$ in which the atoms can remain trapped during θ . One can thus have, after a long enough interaction time, $\delta p = m\delta v < \hbar k$. Contrary to other cooling schemes where fluorescence cycles never stop, VSCPT cooling can be subrecoil.

(ii) *Necessity of a quantum treatment of atomic motion.*

If $\delta p < \hbar k$ (subrecoil cooling), the coherence length $\xi = \hbar/\delta p$ of the atomic wave packets becomes greater than the wavelength $\lambda = 2\pi/k$ of the lasers and the atom can no longer be considered as localized at point \vec{r} in these waves. A quantum description of the translational degrees of freedom of the atom becomes then necessary.

On the other hand, the two absorption amplitudes from g_1 and g_2 can interfere only if they reach the same final state e, \vec{p} (atom in the internal state with a momentum \vec{p}). Because of momentum conservation, these two amplitudes must start from $g_1, \vec{p} - \hbar\vec{k}_1$ and $g_2, \vec{p} - \hbar\vec{k}_2$. The energies of these two states must also include, not only the internal energy E_{g_i} , with $i = 1, 2$, but also the translational kinetic energy $(\vec{p} - \hbar\vec{k}_i)^2/2m$. The dark state is thus a superposition of two states differing, not only by the internal state g_i , but also by the translational state $\vec{p} - \hbar\vec{k}_i$:

$$|\psi_D\rangle = c_1 |g_1, \vec{p} - \hbar\vec{k}_1\rangle + c_2 |g_2, \vec{p} - \hbar\vec{k}_2\rangle, \quad (10)$$

the Raman resonance condition between unperturbed states becoming now:

$$E_{g_1} + (\vec{p} - \hbar\vec{k}_1)^2/2m + \hbar\omega_1 = E_{g_2} + (\vec{p} - \hbar\vec{k}_2)^2/2m + \hbar\omega_2 \quad (11)$$

(iii) *Experimental observation.*

Subrecoil cooling by VSCPT was first proposed and demonstrated in [10]. For detailed calculations, see [11] and [12]. Another method for producing a velocity dependent absorption rate $R(v)$ having the shape represented in Fig. 3a has been proposed and demonstrated in [13]. It is not based on dark resonances, but uses sequences of stimulated Raman and optical pumping pulses.

The experiments using VSCPT have been performed on helium atoms in the 2^3S_1 metastable state excited by 2 coherent counterpropagating laser beams with the same frequency, close to the frequency of the $2^3S_1 \leftrightarrow 2^3P_1$ transition at 1083 nm, and with σ_+ and σ_- polarizations, respectively. One can easily show that only 3 atomic states are involved in the cooling process: $|g_1\rangle = |2^3S_1, M = -1\rangle$, $|g_2\rangle = |2^3S_1, M = +1\rangle$, $|e\rangle = |2^3P_1, M = 0\rangle$ (see Fig. 4a). So, we have exactly the three-level situation considered above. If the static magnetic field is equal to 0, the 2 lower states $M = \pm 1$ have the same energy. Since the 2 laser frequencies are the same, the Raman resonance condition (11)

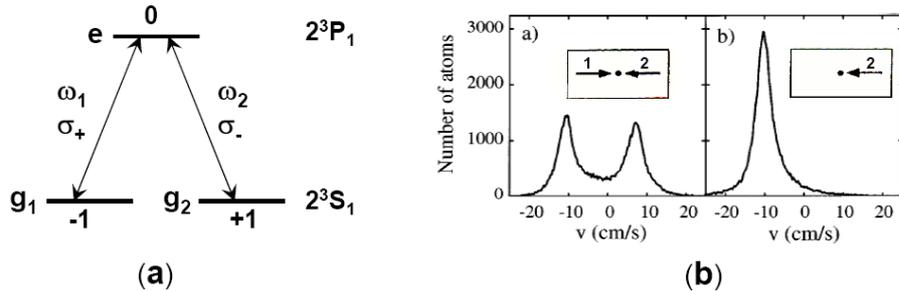


Figure 4: VSCPT experiment. (a): three Zeeman sublevels involved in the VSCPT cooling experiment described in this section. (b): the left part shows the double peak velocity distribution obtained in this experiment. The fact that the two peaks are resolved proves the subrecoil character of the cooling. The right part of the figure shows that a single peak without any loss of atoms is obtained when one of the 2 lasers is switched off slowly.

then gives $p = 0$ and the dark state is:

$$|\psi_D\rangle = \frac{1}{\sqrt{2}} [|M = -1, -\hbar k\rangle + |M = +1, +\hbar k\rangle] \quad (12)$$

This is an internal-external entangled state with a double peak momentum distribution. After a series of experimental improvements[14], momentum distributions with a clear double peak structure have been obtained (see left part of Fig. 4b). The fact that two peaks are clearly resolved means that their width δp is smaller than their splitting $2\hbar k$. In fact, the width of the peaks of Fig. 4b is essentially determined by the experimental resolution. A more precise experiment based on a direct measurement of the spatial correlation function of the atoms[15] showed that the momentum width δp was about 30 times smaller than $\hbar k$ (temperatures in the nanokelvin range). By switching off slowly one of the two laser beams, one can transform adiabatically the dark state from a double component state, like (12), into a single component one (see right part of Fig. 4b) without any loss of atoms. The physical interpretation of this transformation is analogous to the one given for STIRAP.

These experiments are one-dimensional cooling experiments. It has been shown[16] that, for a $J_g = 1 \leftrightarrow J_e = 1$ transition, like the one studied here, VSCPT cooling can be extended to two and three dimensions. The dark state is then isomorphic to the laser configuration, which means that, if we have six laser beams propagating along the positive and negative directions of three perpendicular axis, the dark state is a linear superposition of six atomic wave packets propagating along the same directions with different internal states. Three-dimensional VSCPT cooling experiments have been done[17]. When they are combined with STIRAP type adiabatic transfers, they provide the possibility to manipulate three-dimensional wave packets in the nanokelvin range.

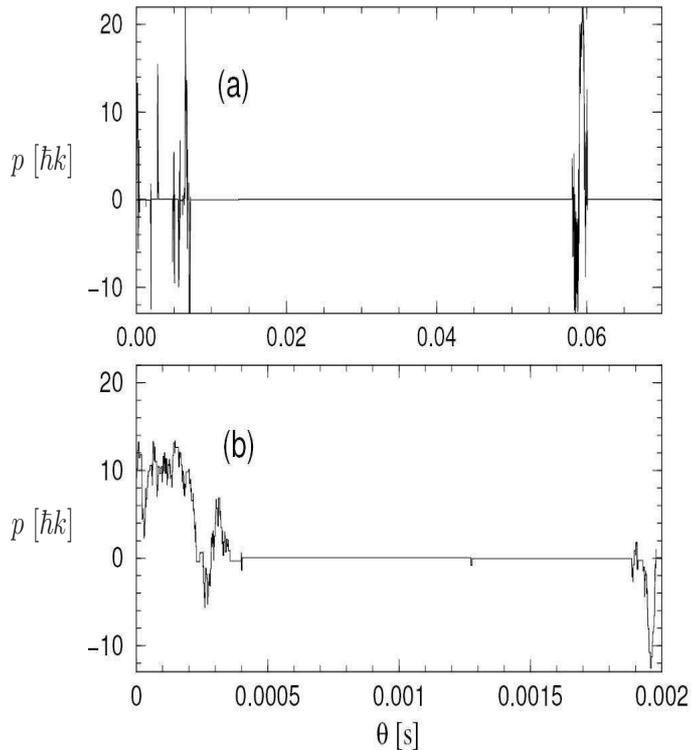


Figure 5: (a): Monte-Carlo simulation of 1D-VSCPT of metastable helium atoms. Each vertical discontinuity corresponds to a spontaneous emission jump during which the atomic momentum changes abruptly. The random walk is clearly dominated by a few rare events. (b): zoomed part of the sequence.

(iv) Monte-Carlo simulations and connection with Lévy flights.

In a full quantum treatment of atomic motion, the external atomic degrees of freedom must be treated quantum-mechanically and the atomic density matrix σ_A must have two types of quantum numbers, internal (i, j) and external (p, p') :

$$\langle i, p | \sigma_A | j, p' \rangle$$

The equations of motion of these matrix elements are called “generalized optical Bloch equations (GOBE)”. They are more complex than usual optical Bloch equations where atomic motion is treated semi-classically and which involve only internal quantum numbers.

Because of their complexity, it is in general impossible to get analytical solutions of the GOBE. Furthermore, subrecoil cooling has no steady-state solution, and it is thus very difficult to make simple predictions concerning the long time

limit where these mechanisms are most interesting. A numerical integration of the GOBE also raises serious practical problems in this long time. The momentum distribution then contains a series of very narrow peaks, analogous to delta functions, and requires a very large number of discrete values for its description in a numerical calculation. It is very difficult to predict if the weight of the area below these narrow peaks, which is related to the proportion of cooled atoms, tends to a finite value or to zero.

Other approaches have been tried for investigating these problems, in particular quantum Monte Carlo simulations using the waiting time distribution giving the distribution of the time intervals between two successive spontaneous emission processes. For example, for 1D-VSCPT of metastable helium, the dressed atom approach has been used for calculating this distribution [18, 19]. Between 2 spontaneous emissions, the atom evolves in the family of three states $\{|e, p\rangle; |g_1, p - \hbar k\rangle; |g_2, p + \hbar k\rangle\}$ coupled by absorption and stimulated emission processes. After each spontaneous emission process, whose probability of occurrence can be calculated by diagonalizing a 3×3 matrix, p changes in a random way and the system enters a new family of three states labelled by the new value of p , and so on. Figure 5 shows the random time evolution of p obtained from such a simulation. It clearly suggests an anomalous random walk along the time axis dominated by a few rare events occurring when p becomes close to zero. In fact, there are about 4000 time intervals in the simulation of Fig. 5.a, and a single of them lasts a time which is on the order of three quarters of the total interaction time. The zoom shown in Fig. 5.b exhibits a similar structure and demonstrates a self-similarity of the random walk at all scales.

All these results have suggested the existence of a close connection between subrecoil cooling and random processes dominated by a few rare events for which Gaussian statistics cannot be applied. In fact, one can show that the distribution of the time intervals spent by the atom in a small interval around $v = 0$ is a broad distribution with power-law tails. The distribution is so broad that it can have no first and second moments. A completely new approach using Lévy statistics which is well adapted for analyzing these broad distributions has been introduced for analyzing the long time limit of subrecoil cooling [19]. It is described in detail in [20], which contains also a tutorial chapter on Lévy statistics. Let us just mention here that it has provided quantitative analytical predictions for the main features of the cooled atoms, for example their momentum distribution, and that these predictions have been checked experimentally[21].

7 Dark resonances of ultracold molecules.

One method for preparing ultracold molecules is to start from ultracold atoms and to photo-associate them. Consider for example the case of 2 spin polarized helium atoms in the 2^3S_1 metastable state undergoing a collision. The associated molecular potential is the $^5\Sigma_g^+$ potential. Since the 2 atoms are ultracold, the energy of the collision state, that we will call φ_{coll} , above the dissociation threshold is on the order of $k_B T$ where T is the temperature of the vapor

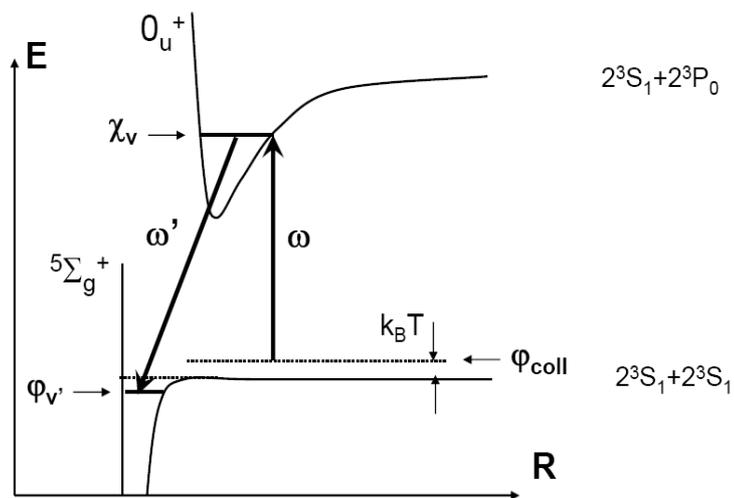


Figure 6: One-photon and two-photon photo-association. In a one-photon photo-association experiment, a pair of colliding atoms in the φ_{coll} state is excited by a laser with frequency ω in a vibrational state χ_v of an excited molecular potential. If a second laser with frequency ω' is added, exciting the transition from χ_v to a vibrational state $\varphi_{v'}$ of the lower molecular potential, the production of molecules in the $\varphi_{v'}$ state is called two-photon photo-association.

(see Fig. 6). During the collision, a laser with frequency ω can excite the pair of colliding atoms from the state φ_{coll} to a bound vibrational state χ_v in an electronically excited molecular potential, for example the 0_u^+ potential, corresponding for large interatomic distances R to two atoms, one in the 2^3S_1 state and one in the 2^3P_0 state (see Fig. 6). This is a one-photon photo-association process. One can also add a second laser, coherent with the first one, with a frequency ω' close to the frequency of the transition connecting the state χ_v to a bound vibrational state $\varphi_{v'}$ in the molecular potential $^5\Sigma_g^+$. This is a two-photon photo-association process.

Two-photon photo-association involves a three-level system $\{\varphi_{\text{coll}}, \varphi_{v'}, \chi_v\}$ analogous to the one considered above and should therefore give rise to dark resonances. There are however an important difference. The state φ_{coll} is not a discrete state but belongs to a continuum, so that the dark resonance should be broad. However, since the atoms are ultracold, their energy spread, on the order of $k_B T$, is very small, and the corresponding broadening of the dark resonance is generally negligible. Second, the three-level system is not closed: the total population can decrease because of Penning ionization processes. One can show[22] that this prevents the fluorescence rate to vanish completely when the Raman resonance condition is fulfilled. The dark resonance still appears as a dip, but this dip does not go to zero.

An experiment of this type has been performed on helium atoms[23]. The state χ_v is the vibrational state $v = 0$ of the 0_u^+ potential. The state $\varphi_{v'}$ is the least vibrational state $v = 14$ of the $^5\Sigma_g^+$ potential. Dark resonances analogous to the ones shown in Fig. 2b and Fig. 2c have been observed³ and their shape found in good agreement with the theoretical predictions of reference [24]. From the position of the dark resonance, one can deduce a very precise value of the binding energy of the least bound state $v = 14$ of the $^5\Sigma_g^+$ potential, and from this binding energy, one can obtain a value of the scattering length a of two helium atoms in the metastable state, more than one hundred times more precise than all previous other measurements. The knowledge of a is important for the interpretation of the experiments having achieved Bose-Einstein condensation of helium atoms in the 2^3S_1 metastable state[25, 26].

In conclusion, one can say that dark resonances, which have been discovered in the early days of optical pumping continue to play an important role in the most recent developments of atomic and molecular physics, like nonlinear optics or ultracold quantum gases. As a recent example of this importance, one can mention a very recent experiment [29] where STIRAP has been used to prepare an ultracold gas (at a temperature of 350 nK) of heteropolar molecules $^{40}\text{K}^{87}\text{Rb}$ in the rovibrational ground state of either the triplet or singlet electronic ground molecular potential. This experiment opens fascinating perspectives due to the fact that heteropolar molecules have a permanent electric dipole moment and have thus anisotropic molecule-molecule interactions with a long range $1/r^3$

³Dark resonances have been also observed by two other groups in two-photon photo-association of alkali atoms[27, 28]

dependence. The density of the molecular gas which has been prepared is quite high (10^{12} cm^{-3}), so that obtaining a molecular condensate with electric dipole-dipole interactions seems within reach.

References

- [1] G. Alzetta, A. Gozzini, L. Moi, G. Orriols, *Il Nuovo Cimento* **36B**, 5 (1976).
- [2] E. Arimondo, G. Orriols, *Lett. Nuovo Cimento* **17**, 333 (1976).
- [3] C. Cohen-Tannoudji, J. Dupont-Roc, et G. Grynberg, “Atoms-Photon Interactions : Basic Processes and Applications” (Wiley, New-York, 1992).
- [4] J. Dalibard, S. Reynaud, C. Cohen-Tannoudji, in “Interaction of Radiation with Matter”, Volume in honour of A. Gozzini, p. 29, ed. by L. Radicati, (Pise, 1987).
- [5] B. Lounis, C. Cohen-Tannoudji, *J. Physique II France*, **2**, 579 (1992).
- [6] E. Arimondo, in *Progress in Optics*, ed. E. Wolf, Vol. 35, p.257 (Elsevier, Amsterdam, 1996).
- [7] S.E. Harris, *Physics Today*, July 1997, p.36.
- [8] L.V. Hau, S.E. Harris, Z. Dutton, C.H. Behroozi, *Nature (London)* **397**, 594 (1999).
- [9] J.R. Kulinski, U. Gaubatz, F.T. Hioe and K. Bergmann *Phys.Rev.* **A40**, 6741 (1989).
- [10] N.V. Vitanov, M. Fleischauer, B.W. Shore and K. Bergmann *Adv.At.Mol.Opt.Phys.* **46**, 55 (2001).
- [11] A. Aspect, E. Arimondo, R. Kaiser, N. Vansteenkiste, and C. Cohen-Tannoudji, *Phys. Rev. Lett.* **61**, 826 (1988).
- [12] A. Aspect, E. Arimondo, R. Kaiser, N. Vansteenkiste and C. Cohen-Tannoudji, *J. Opt. Soc. Am. B* **6**, 2112 (1989).
- [13] C. Cohen-Tannoudji, *Atomic motion in laser light*, in *Fundamental systems in quantum optics*, J. Dalibard, J.-M. Raimond, and J. Zinn-Justin eds, Les Houches session LIII (1990), p.1 (North-Holland, 1992).
- [14] M. Kasevich and S. Chu, *Phys. Rev. Lett.* **69**, p. 1741-1744 (1992).
- [15] F. Bardou, Ph. D. Thesis, University of Paris XI Orsay (1995).
- [16] B. Saubaméa, T.W. Hijmans, S. Kulin, E. Rasel, E. Peik, M. Leduc, and C. Cohen-Tannoudji, *Phys. Rev. Lett.* **79**, 3146 (1997).
- [17] M.A. Ol’shanii and V.G. Minogin, *Optics. Comm.* **89**, 393 (1992).

- [18] S. Kulin, B. Saubaméa, E. Peik, J. Lawall, T. W. Hijmans, M. Leduc, and C. Cohen-Tannoudji, *Phys. Rev. Lett.* **78**, 4185 (1997).
- [19] C. Cohen-Tannoudji, F. Bardou, and A. Aspect, *Review of fundamental processes in laser cooling*, Proceedings of Laser Spectroscopy X (Font-Romeu, 1991), edited by M. Ducloy, E. Giacobino, and G. Camy, p.3 (World Scientific, Singapore, 1992).
- [20] F. Bardou, J.-P Bouchaud, O. Emile, A. Aspect and C. Cohen-Tannoudji, *Phys. Rev. Lett.* **72**, p. 203-206 (1994).
- [21] F. Bardou, J.-P Bouchaud, A. Aspect and C. Cohen-Tannoudji, *Lévy Statistics and Laser Cooling. How Rare Events Bring Atoms to Rest*, Cambridge University Press (Cambridge, 2002).
- [22] B. Saubaméa, M. Leduc, and C. Cohen-Tannoudji, *Phys. Rev. Lett.* **83**, 3796 (1999).
- [23] M. Portier, Ph. D. Thesis, University of Paris VI (2007).
- [24] S. Moal, M. Portier, J. Kim, J. Dugué, U.D. Rapol, M. Leduc, and C. Cohen-Tannoudji, *Phys. Rev. Lett.* **96**, 023203 (2006).
- [25] J.L. Bohn and P.S. Julienne, *Phys. Rev.* **A60**, 414 (1999).
- [26] A. Robert, O. Sirjean, A. Browaeys, J. Poupard, S. Nowak, D. Boiron, C.I. Westbrook and A. Aspect, *Science* **292**, 461 (2001).
- [27] F. Pereira Dos Santos, J. Leonard, J. Wang, C.J. Barrelet, F. Perales, E. Rasel, C.S. Unnikrishnan, M. Leduc, and C. Cohen-Tannoudji, *Phys. Rev. Lett.* **86**, 3459 (2001).
- [28] P. Dumke, J.D. Weinstein, M. Johanning, K.M. Jones and P. D. Lett, *Phys. Rev.* **A72**, 041801(R) (2005).
- [29] K. Winkler, G. Thalhammer, M. Theis, H. Ritsch, R. Grimm and J.H. Denschlag, *Phys. Rev. Lett.* **95**, 063202 (2005).
- [30] K.-K. Ni, S. Ospelkaus, M. H. G. de Miranda, A. Pe'er, B. Neyenhuis, J. J. Zirbel, S. Kotochigova, P. S. Julienne, D. S. Jin, and J. Ye, *Science*, published online Sept. 18, 2008. See also arXiv:0808.2963v2 [*quant - ph*].