Dark States. From Quantum Optics to Ultracold Atoms and Molecules

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Purpose of this lecture

Describe an experiment, first performed in 1976 in Pisa, and where absorption of light by certain states, called “dark states”, is blocked by a quantum destructive interference effect.

Give a physical interpretation of this effect

Show that dark states have played an essential role in the discovery of several new physical effects:

- Electromagnetically Induced Transparency
- Slow Light
- Subrecoil Laser Cooling and Connection with Anomalous Random Walks (“Lévy Flights”)
- Stimulated Raman Adiabatic Passage (STIRAP)
- Ultracold Molecules
The experiment of Adriano Gozzini in Pisa

Optically pumped sodium vapor put in a gradient of magnetic field parallel to the z-axis. The splitting between 2 Zeeman sublevels $g_1$ et $g_2$ depend on $z$

An RF field with frequency $\nu$ induces resonant transitions between $g_1$ and $g_2$ only at the point $z_0$ where their splitting between is equal to $h\nu$

Modification of the populations of $g_1$ and $g_2$ in $z_0$ resulting in a change of the light intensity emitted by the atoms in $z_0$

Analogy with Magnetic Resonance Imaging (MRI). Spatially resolved magnetic resonance
Bright resonances and dark resonances

Bright resonance: appears at the point $z_0$ where the splitting between $g_1$ and $g_2$ is equal to $\hbar \nu$.

Dark resonance: appears also in the absence of RF, but only if the laser contains at least 2 modes with frequencies $\omega_1$ and $\omega_2$. The dark resonance appears at the point $z'_0$ where the splitting between $g_1$ and $g_2$ is equal to $\hbar (\omega_1 - \omega_2)$: Raman resonance condition

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

Theoretical approaches

Optical Bloch Equations

Equations of evolution of the density matrix of a 3-level atom $g_1$, $g_2$, $e$ excited by 2 laser beams

$$E_1 \exp[i(k_1 \cdot r - \omega_1 t)] + \text{c.c.} \quad \text{and} \quad E_2 \exp[i(k_2 \cdot r - \omega_2 t)] + \text{c.c.}$$

exciting respectively the 2 transitions $g_1 \rightarrow e$ and $g_1 \rightarrow e$

Atom dressed by 2 types of photons $\omega_1$ and $\omega_2$

J. Dalibard, S. Reynaud, C. Cohen-Tannoudji
Basic physical effect

1- We first consider the situation at time \( t=0 \).

The atom is put in a linear superposition of \( g_1 \) and \( g_2 \)

\[
|\psi\rangle = c_1 |g_1\rangle + c_2 |g_2\rangle
\]

such that the 2 absorption amplitudes \( g_1 \rightarrow e \) and \( g_2 \rightarrow e \) interfere destructively

\[
c_1 \vec{D}_{eg_1} \cdot \vec{E}_1 + c_2 \vec{D}_{eg_2} \cdot \vec{E}_2 = c_1 \Omega_1 + c_2 \Omega_2 = 0
\]

\( c_i \) : Amplitude for the atom to be in state \( g_i \) (\( i=1,2 \))

\( \Omega_i \) (Rabi frequencies): Amplitudes for the atom to absorb a photon \( \omega_i \) from the state \( g_i \) (\( i=1,2 \)). \( D \) : atomic dipole moment

The state \( c_1 |g_1\rangle + c_2 |g_2\rangle \) is called "dark state"

The atom is put in a linear superposition of states which cannot absorb light because of destructive interference

Phenomenon called “Coherent Population Trapping” (CPT)
2- If a state is dark at \( t=0 \), can it remain dark at a later time \( t \)?

The coefficients \( c_i \) acquire phase factors \( \exp(-iE_i t/\hbar) \) due to the energies \( E_i \) of the states \( g_i \). The laser fields \( E_i \) acquire phase factors \( \exp(-i\omega_i t) \) due to their frequencies \( \omega_i \).

The total absorption amplitude thus becomes:

\[
\begin{align*}
    c_1 \exp(-iE_1 t/\hbar) \Omega_1 \exp(-i\omega_1 t) + c_2 \exp(-iE_2 t/\hbar) \Omega_2 \exp(-i\omega_2 t)
\end{align*}
\]

It still vanishes, as in \( t=0 \), if and only if the 2 phase factors multiplying \( c_1 \Omega_1 \) and \( c_2 \Omega_2 \) are the same, i.e. if:

\[
    \frac{E_1}{\hbar} + \omega_1 = \frac{E_2}{\hbar} + \omega_2 \iff \hbar(\omega_1 - \omega_2) = E_2 - E_1
\]

One recovers the Raman resonance condition and one understands why the dark resonance only appears when this condition is fulfilled.

If it is not fulfilled, a state which is dark at a certain time \( t=0 \) will no longer be dark at a later time, and it will absorb light.
Variations of the fluorescence rate $R_F$ with the detuning $\Delta$ from the unperturbed Raman resonance

One varies the splitting between $g_1$ and $g_2$, $\omega_1$ and $\omega_2$ being fixed. Or one sweeps $\omega_2$, $\omega_1$ being fixed.

The dark resonance has a width $\Gamma'$ determined by the relaxation time in the ground state, much smaller than the width $\Gamma$ determined by the lifetime of the excited state. It is their very small width which explains the interest of dark resonances for applications.

Example of application:

« Electromagnetically Induced Transparency (EIT) »

An atomic vapor with a great optical depth for a laser $\omega_1$ exciting the transition $g_1 \rightarrow e$ becomes transparent for this laser if one adds a second laser $\omega_2$ exciting the transition $g_2 \rightarrow e$ with a frequency fulfilling the Raman resonance condition. The index of refraction for the laser $\omega_1$ varies very rapidly in the neighborhood of the dark resonance $\Rightarrow$ slow light.
SUBRECOIL LASER COOLING
3- What happens if the atom is moving with a velocity \( v \)?

\[ r = v \, t \]

We first use a classical treatment of atomic motion. Replacing \( r \) by \( vt \) in \( \exp[i(k_i \cdot r - \omega_i t)] \), one gets additional phase factors \( \exp(ik_i \cdot vt) \) multiplying \( c_i \Omega_i \).

If the state was dark for all \( t \) when \( v=0 \) and if \( k_1 \cdot v \neq k_2 \cdot v \), the phase shifts of \( c_1 \Omega_1 \) and \( c_2 \Omega_2 \) due to the motion will not be the same and the state will no longer be dark when \( v \neq 0 \).

Coherent population trapping is velocity selective

“Velocity Selective Coherent Population Trapping (VSCPT)”

Interpretation in terms of Doppler effect: \( k_1 \cdot v \) et \( k_2 \cdot v \) are the Doppler shifts of \( \omega_1 \) and \( \omega_2 \) due to the motion. They subtract in the Raman process and change the detuning \( \Delta \) which becomes \( \Delta+(k_1 \cdot v - k_2 \cdot v) \) different from \( \Delta \) if \( k_1 \cdot v \neq k_2 \cdot v \).

Case where \( |k_1| = |k_2| \)

Coherent population trapping is velocity selective only if \( k_1 \) and \( k_2 \) are not parallel, for example opposite.
- One realizes a situation where the fluorescence rate $R$ depends on $v$ and vanishes for $v=0$. Atoms with zero velocity (or with $v$ close to 0) no longer absorb photons and are protected from the « bad effects » of light (random recoils due to the 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 processes can make them falling in the region close to $v=0$ where they remain trapped and accumulate.

- The longer the interaction time $\theta$, the narrower the interval $\delta v$ in which the atoms can remain trapped during this time.

One can have $\delta p = m \delta v < \hbar k$ (subrecoil cooling)
Coherent population trapping (continued)

4- Is it correct to describe atomic motion classically?

If \( \delta p < \hbar k \), the coherence length \( \xi \) of the atomic wave packets becomes greater than the wavelength \( \lambda \) of the lasers and the atom can no longer be considered as localized at point \( r \) in these waves

\[
\delta p < \hbar k \quad \rightarrow \quad \xi = \frac{\hbar}{\delta p} > \frac{2\pi}{k} = \lambda
\]

On the other hand, the 2 absorption amplitudes from \( g_1 \) and \( g_2 \) can interfere only if they reach the same final state \( e, p \) (atom in the state \( e \) with momentum \( p \)). Because of momentum conservation these 2 amplitudes must start from \( g_1, p - \hbar k_1 \) and \( g_2, p - \hbar k_2 \) and the energies of must include not only the internal energy \( E_{gi} \) but also the external kinetic energy \( (p-\hbar k_i)^2/2m \).

The dark state is thus a superposition of 2 states differing not only by the internal state \( g_i \), but also by the external state \( p - \hbar k_i \):

\[
|\psi\rangle = c_1 |g_1, \vec{p} - \hbar \vec{k}_1\rangle + c_2 |g_2, \vec{p} - \hbar \vec{k}_2\rangle, \text{ avec (résonance Raman)}
\]

\[
E_{g_1} + \left(\frac{\vec{p} - \hbar \vec{k}_1}{2m} + \hbar \omega_1 = E_{g_2} + \left(\frac{\vec{p} - \hbar \vec{k}_2}{2m} + \hbar \omega_2
\right)
\]
First demonstration of VSCPT-1D

The 2 lasers have the same frequency ($\omega_1=\omega_2$), opposite directions ($k_1=-k_2$) and polarizations $\sigma_+$ and $\sigma_-$.

The magnetic field is equal to 0 so that:

$$E_{g_+} = E_{g_-}$$

The Raman resonance condition writes $p=0$ and the dark state is:

$$|\psi\rangle = (1/\sqrt{2}) \left[ |-1\rangle \otimes |-\hbar k\rangle + |+1\rangle \otimes |+\hbar k\rangle \right]$$

Entangled state with a double peak momentum distribution.

Improvement of the experiment

One no longer uses a transversally cooled atomic beam.

The atoms are stopped, trapped in a MOT, cooled with 2 laser beams with opposite directions, with polarizations $\sigma_+$ and $\sigma_-$, and then dropped on a multi channel detector.

The interaction time $\theta$ can be much longer.

A single drop

80 drops

Adiabatic transfer

By adiabatically changing the relative intensity of the 2 lasers, one changes the coefficients $c_1$ and $c_2$ and one transfers the total population of the 2 peaks into a single one.

Same idea as STIRAP "STImulated Raman Adiabatic Passage") in molecular physics, allowing one to transfer all molecules from a level to another one, while remaining always in a dark state.

Generalizations of VSCPT to 2D and 3D

Difficulty of the traditional methods of quantum optics and of numerical calculations for a quantitative study of these effects.
Quantum Monte-Carlo simulation of 1D-VSCPT

Calculation by the dressed atom approach of the distribution of the time intervals between 2 successive spontaneous emissions of photons

Between 2 emissions, the momentum quantum number remains constant and changes in a random way after each emission


Anomalous random walk along the time axis dominated by a few rare events where \( p \) remains close to zero.

Self-similarity at all scales

Analogy with other situations involving ‘Lévy flights’
François BARDOU
Random sequence of events \( \{M_i\} \)

Occurring at random times \( T_0 = 0, T_1, T_2, ... T_{i-1}, T_i, ... \)

\[
\begin{array}{cccccc}
M_0 & M_1 & M_2 & M_3 & M_{i-1} & M_i \\
\hline
T_0 & T_1 & T_2 & T_3 & T_{i-1} & T_i \\
\end{array}
\]

\( \tau_i \): Time interval between two successive events \( M_{i-1}, M_i \)

The \( \tau_i \)'s are independent random variables, all distributed according to the same law \( P(\tau) \)

Random walk along the \( t \)-axis with steps \( \tau_i \):

\[
T_1 = \tau_1 \quad T_2 = \tau_1 + \tau_2 \quad T_3 = \tau_1 + \tau_2 + \tau_3 \quad T_N = \sum_{i=1}^{N} \tau_i
\]

\( T_N \) is a sum of \( N \) independent random variables

Distribution of the sums \( T_N \)

If \( \langle \tau \rangle \) et \( \langle \tau^2 \rangle \) are finite, this distribution is a gaussian

Central limit theorem
What happens for distributions laws when \( P(\tau) \) has power law tails?

While being normalizable, \( P(\tau) \) can have infinite mean value or variance. This is for example the case when:

\[
P(\tau) \sim \frac{C}{\tau^{1+\mu}} = \frac{\mu \tau_b^\mu}{\tau^{1+\mu}}
\]

If \( 0 < \mu \leq 1 \) \( \langle \tau \rangle = \infty \) and \( \langle \tau^2 \rangle = \infty \)

If \( 1 < \mu \leq 2 \) \( \langle \tau^2 \rangle = \infty \)

The central limit theorem no longer applies

**Lévy laws**

For such laws, Paul Lévy has shown that \( T_N \) (after a certain scaling) is distributed, in the limit \( N \to \infty \), according to a universal law depending only on \( \mu \)

One can show also that \( T_N \) is dominated by a small number of terms. Importance of rare events.
Example of numerical simulation

Variations of $T_N/\tau_b$ with $N$ when $\mu = 1/2$

- A single event contributes to a large part of $T_N$
- Self-similar structure at all scales
Sprinkling distribution $S(t)$ (also called « renewal density »)

$S(t)$ is given by a sum of convolution products of $P(t)$, which can be easily calculated by a Laplace transform

$$S(t) = P(\tau) + P(\tau) \otimes P(\tau) + P(\tau) \otimes P(\tau) \otimes P(\tau) + \ldots$$

If $\langle \tau \rangle$ is finite

$$S(t) = 1/\langle \tau \rangle$$

If $\langle \tau \rangle$ is infinite, with power laws of $P(\tau)$ decreasing as $1/\tau^{\mu+1}$

$$S(t) \propto \frac{1}{t^{1-\mu}} \quad (\mu < 1)$$

When $t$ increase, the probability to draw a large value of $t$ increases and the sprinkling decreases.

« Aging » of the system
Modelization of subrecoil cooling

- Trapping zone of dimension $p_{\text{trap}} \ll \hbar k$ around $p=0$
  At the next spontaneous emission, the atom leaves the trap
- The distribution of the trapping times $t$ has decreases with power-law tails in $\tau^{-\mu}$ where $\mu = D/\alpha$ ($D$ is the dimensionality and $\alpha$ the exponent of the absorption probability of a photon $R(p) = C p^\alpha$. For exemple, if $D=1$, $\alpha=2$, $\mu=1/2$
- Calculation of the sprinkling distribution $S(t)$ in the trap
- Calculation of the momentum distribution $P(p,\theta)$ of the atoms after an interaction time $\theta$
  $$P(p,\theta) \propto \int_0^\theta dt S(t) \exp\left[-R(p)(\theta - t)\right]$$
  $\exp\left[-R(p)(\theta - t)\right]$: Probability to remain in the trap during $(\theta - t)$
- Obtention of analytical expressions which be difficult to derive with traditional methods
Example: Measurement of the Fourier transform of $P(p, \theta)$ and comparison with theoretical predictions

B. Saubamea
M. Leduc
C. Cohen-Tannoudji

$T = 4nK$

Full lines: Predictions of Lévy statistics
Dotted lines: Best fit with the T.F. of a Lorentzian
Experimental points with error bars
ULTRACOLD MOLECULES
How to cool molecules

The absence of « closed transitions » in molecules makes it difficult to apply laser cooling. Other more efficient methods have been developed.

1- Paramagnetic molecules trapped and cooled by contact with a cold helium gas which is then pumped (J. Doyle)

2- Stark or Zeeman deceleration of molecules by pulsed inhomogeneous electric or magnetic fields (G. Meijer M. Raizen)

3- Sweeping a Feshbach resonance to obtain a Feshbach molecules which is then transferred to the ground state by the STIRAP method

4- One or Two photon photoassociation
Feshbach Resonances
Feshbach Resonances

The 2 atoms collide with a very small positive energy $E$ in a channel which is called “open”

The energy of the dissociation threshold of the open channel is taken as the zero of energy

There is another channel above the open channel where scattering states with energy $E$ cannot exist because $E$ is below the dissociation threshold of this channel which is called “closed”

There is a bound state in the closed channel whose energy $E_{bound}$ is close to the collision energy $E$ in the open channel
Physical mechanism of the Feshbach resonance

The incoming state with energy $E$ of the 2 colliding atoms in the open channel is coupled by the interaction to the bound state $\varphi_{\text{bound}}$ in the closed channel.

The pair of colliding atoms can make a virtual transition to the bound state and come back to the colliding state. The duration of this virtual transition scales as $\hbar / |E_{\text{bound}} - E|$, i.e., as the inverse of the detuning between the collision energy $E$ and the energy $E_{\text{bound}}$ of the bound state.

When $E$ is close to $E_{\text{bound}}$, the virtual transition can last a very long time and this enhances the scattering amplitude.

Analogy with resonant light scattering when an impinging photon of energy $\hbar \nu$ can be absorbed by an atom which is brought to an excited discrete state with an energy $\hbar \nu_0$ above the initial atomic state and then reemitted. There is a resonance in the scattering amplitude when $\nu$ is close to $\nu_0$. 
Sweeping the Feshbach resonance

The total magnetic moment of the atoms are not the same in the 2 channels (different spin configurations). The energy difference between the them can be varied by sweeping a magnetic field.
Scattering length versus magnetic field

\[ a \]

- \( a > 0 \) Repulsive effective long range interactions
- \( a = 0 \) No interactions Perfect gas
- \( a < 0 \) Attractive effective long range interactions

\( B_0 \) : value of B for which the energy of the bound state, in the closed channel (shifted by its interaction with the continuum of collision states in the open channel) coincides with the energy \( E \sim 0 \) of the colliding pair of atoms

Near \( B = B_0 \), \(|a|\) is very large
Strong interactions
Strong correlations

The bound state exists only in the region $a > 0$. It has a spatial extension $a$ and an energy $E_b = -\frac{\hbar^2}{ma^2} \propto -(B - B_0)^2$

Weakly bound dimmer with universal properties
Quantum "halo" state or "Feshbach molecule"
If $B_0$ is swept through the Feshbach resonance from the region $a < 0$ to the region $a > 0$, a pair of colliding ultracold atoms can be transformed into a Feshbach molecule.

Another interesting system: Efimov trimmers (R. Grimm)
Observation of universal bound states of 3 atoms in a region \((a < 0)\) where no bound states of 2 atoms can exist

Detection by observation of atom losses near the value of \(a\) where these states are expected to appear (Groups of R. Grimm in Innsbruck and M. Inguscio in Firenze)

Example of a « few body system »
Stimulated Raman Adiabatic Passage (STIRAP)

Klaas Bergmann

Dark state

\[ |\psi_D\rangle = c_1 \, |g_1\rangle + c_2 \, |g_2\rangle \]

\[ c_1 \Omega_1 + c_2 \Omega_2 = 0 \]

\( \Omega_i \) : Rabi frequencies

\[ \frac{c_1}{\Omega_2} = -\frac{c_2}{\Omega_1} \]

\( \Omega_1 \gg \Omega_2 \) \quad The dark state coincides with \( g_2 \)

\( \Omega_2 \gg \Omega_1 \) \quad The dark state coincides with \( g_1 \)

By starting with \( \Omega_1 \gg \Omega_2 \), by reducing adiabatically \( \Omega_1 \) while increasing adiabatically \( \Omega_2 \) until \( \Omega_2 \) becomes much larger than \( \Omega_1 \), atoms are selectively transferred adiabatically from \( g_2 \) to \( g_1 \)

Anti-intuitive order

The system always remains in a dark state, so that \( e \) is never populated (no spontaneous transitions from \( e \)). Selective transfer of molecules from one state to another
Ultracold Molecules in the Ground State

**Method**
- Sweeping a Feshbach resonance and production of $^{40}\text{K}^{87}\text{Rb}$ molecules in a highly excited rovibrational state
- Transfer by STIRAP of this excited state to the rovibrational ground state

The lasers $\omega_1$ and $\omega_2$ are applied in the non intuitive order: the laser $\omega_2$ is first applied and its intensity is decreased while the intensity of the laser $\omega_1$ is increased.

Efficient (56%) and selective transfer (control of the hyperfine and Zeeman levels in which molécules are put)

JILA – Boulder team of D.Jin and J.Ye


Polar $^{40}\text{K}^{87}\text{Rb}$ molecules with a density of $10^{12}$ cm$^{-3}$ at $T = 350$ nK
Why are ultracold molecules interesting

1- High resolution molecular spectroscopy
   More precise fundamental tests (parity, edm,..)

2- Heteropolar ultracold molecules
   Anisotropic and long-range dipole-dipole interactions
   Such molecules put in an optical lattice interact even if they are in different wells

3- Quantum effects in collisions and chemical reactions
   Ultra-cold chemistry
   The de Broglie wavelength of molecules is larger than the distance at which chemical reactions can occur
   ➔ a quantum description of collisions is required
   ➔ new physical effects

4- Quantum gases of ultracold polar molecules
   Domain still unexplored. How does the long range and the anisotropy of interactions modify the superfluidity of such quantum gases
Example of quantum effect in a chemical reaction

\[ {^{40}\text{K}}{^{87}\text{Rb}} + {^{40}\text{K}}{^{87}\text{Rb}} \rightarrow {^{40}\text{K}}{^{40}} + {^{87}\text{Rb}}{^{87}\text{Rb}} \]

The molécules \( {^{40}\text{K}}{^{87}\text{Rb}} \) are fermions

If they are prepared in the same internal state (- 4, 1/2) or (- 4, 3/2), the internal part of the wave function is symetric and the orbital part is thus donc antisymetric. The collision can occur only in the p-wave (l=1)

If the 2 molecules are prepared in different internal states, they can collide in the s-wave

If the collision occurs in the p-wave, a centrifugal barrier appears and prevents the 2 molecules to get close enough to react chemically. The reaction is blocked

There is no barrier in the s-wave and the reaction can occur

A simple flip of the nuclear spin changes the internal state of the molecule and can thus change the reaction speed by a factor 50!

Conclusion

During the last few decades, conservation laws in atom-photon interactions have been used for manipulating atoms, for polarizing them and for cooling them at very low temperatures.

It turns out that quantum interference effects in atom-photon interactions can also be very useful - for overcoming fundamental limits like the single photon recoil limit. - for pointing out interesting connections between laser cooling and the statistics of anomalous random walks - for solving difficulties encountered in the laser cooling of molecules

These new schemes can be considered as a « coherent control », a beautiful illustration of quantum physics
Moshe SHAPIRO
A pioneer in coherent control