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Radiation Matter Interaction
Exam

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I - Bloch vector representation of a two-level system

1. The most general form of a two-level system eigenstate is:

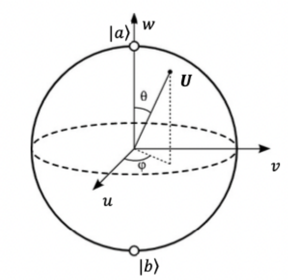
$$|\psi\rangle = \cos\left(\frac{\theta}{2}\right)e^{+i\frac{\phi}{2}}|a\rangle + \sin\left(\frac{\theta}{2}\right)e^{-i\frac{\phi}{2}}|b\rangle$$

We note $\hat{\sigma}$ the corresponding density matrix operator.

Let us consider the so-called Bloch vector $\boldsymbol{U}\begin{pmatrix} u \\ v \\ w \end{pmatrix}$, the coordinates of which are defined as

$$\begin{cases} u = \dfrac{1}{2}(\sigma_{ab} + \sigma_{ba}) \\ v = \dfrac{1}{2i}(\sigma_{ab} - \sigma_{ba}) \\ w = \dfrac{1}{2}(\sigma_{aa} - \sigma_{bb}) \end{cases}$$

By using the general expression of $|\psi\rangle$, compute the expression of u, v and w as a function of θ and ϕ. Show that the extremity of $\boldsymbol{U}$ spans the surface of a sphere with radius $1/2$. Draw a sketch displaying the angles θ and ϕ.



$$\hat{\sigma} = \begin{pmatrix} |c_a|^2 & c_a c_b^* \\ c_b c_a^* & |c_b|^2 \end{pmatrix} = \begin{pmatrix} \cos^2\left(\frac{\theta}{2}\right) & \sin\left(\frac{\theta}{2}\right)\cos\left(\frac{\theta}{2}\right)e^{-i\phi} \\ \sin\left(\frac{\theta}{2}\right)\cos\left(\frac{\theta}{2}\right)e^{+i\phi} & \sin^2\left(\frac{\theta}{2}\right) \end{pmatrix} = \begin{pmatrix} \cos^2\left(\frac{\theta}{2}\right) & \frac{1}{2}\sin\theta e^{-i\phi} \\ \frac{1}{2}\sin\theta e^{+i\phi} & \sin^2\left(\frac{\theta}{2}\right) \end{pmatrix}$$

$$\begin{cases} u = \dfrac{1}{2}(\sigma_{ab} + \sigma_{ba}) = \sin\left(\dfrac{\theta}{2}\right)\cos\left(\dfrac{\theta}{2}\right)\left(\dfrac{e^{+i\phi} + e^{-i\phi}}{2}\right) = \dfrac{1}{2}\sin\theta\cos\phi \\ v = \dfrac{1}{2i}(\sigma_{ab} - \sigma_{ba}) = \sin\left(\dfrac{\theta}{2}\right)\cos\left(\dfrac{\theta}{2}\right)\left(\dfrac{e^{+i\phi} - e^{-i\phi}}{2i}\right) = \dfrac{1}{2}\sin\theta\sin\phi \\ w = \dfrac{1}{2}(\sigma_{aa} - \sigma_{bb}) = \dfrac{1}{2}\left(\cos^2\dfrac{\theta}{2} - \sin^2\dfrac{\theta}{2}\right) = \dfrac{1}{2}\cos\theta \end{cases}$$

$$|\boldsymbol{U}| = \sqrt{u^2 + v^2 + w^2} = \sqrt{\frac{1}{4}\sin^2\theta(\cos^2\phi + \sin^2\phi) + \frac{1}{4}\cos^2\theta} = \frac{1}{2}$$

2. Now, we assume that $|\psi\rangle$ describes the eigenstate of a two-level system interacting with a sinusoidal electromagnetic field with angular frequency ω. The optical Bloch equations, in the absence of relaxations, are:

$$\begin{aligned}
\dot{\sigma}_{aa} &= -i\frac{\Omega_1}{2}(\sigma_{ba}e^{+i\omega t} - \sigma_{ab}e^{-i\omega t}) \\
\dot{\sigma}_{bb} &= +i\frac{\Omega_1}{2}(\sigma_{ba}e^{+i\omega t} - \sigma_{ab}e^{-i\omega t}) \\
\dot{\sigma}_{ab} &= +i\omega_0\sigma_{ab} - i\frac{\Omega_1}{2}e^{+i\omega t}(\sigma_{bb} - \sigma_{aa}) \\
\dot{\sigma}_{ba} &= -i\omega_0\sigma_{ba} + i\frac{\Omega_1}{2}e^{-i\omega t}(\sigma_{bb} - \sigma_{aa})
\end{aligned}$$

where Ω_1 is the Rabi frequency and ω_0 is the Bohr frequency of the transition. Use the rotating frame transformation to eliminate the rapid time-dependence in the previous equation set. What is the physical meaning of this transformation ?

The rotating-frame transformation consist in removing the rapid time dependence of the σ_{ii},

$$\begin{aligned}
\tilde{\sigma}_{ab} &= \sigma_{ab}e^{-i\omega t} \\
\tilde{\sigma}_{ba} &= \sigma_{ba}e^{+i\omega t} \\
\tilde{\sigma}_{aa} &= \sigma_{aa} \\
\tilde{\sigma}_{bb} &= \sigma_{bb}
\end{aligned}$$

It consists in writing the coherence terms in the frame that is rotating at the field frequency ω. The system becomes a coupled set of differential equations with constant coefficients:

$$\begin{cases}
\dot{\tilde{\sigma}}_{aa} = -i\frac{\Omega_1}{2}\left(\tilde{\sigma}_{ba} - \tilde{\sigma}_{ab}\right) \\
\dot{\tilde{\sigma}}_{bb} = +i\frac{\Omega_1}{2}\left(\tilde{\sigma}_{ba} - \tilde{\sigma}_{ab}\right) \\
\dot{\tilde{\sigma}}_{ab} = +i\omega_0\tilde{\sigma}_{ab} - i\frac{\Omega_1}{2}(\tilde{\sigma}_{bb} - \tilde{\sigma}_{aa}) \\
\dot{\tilde{\sigma}}_{ba} = -i\omega_0\tilde{\sigma}_{ba} + i\frac{\Omega_1}{2}(\tilde{\sigma}_{bb} - \tilde{\sigma}_{aa})
\end{cases}$$

3. By use of the results obtained in question 2, write the differential equations that are obeyed by $\tilde{u}$, $\tilde{v}$ and $\tilde{w}$, the three components of $\tilde{\boldsymbol{U}}$ in the rotating frame. Show that $\tilde{\boldsymbol{U}}$ undergoes a precession around a vector $\boldsymbol{\Omega}$ whose coordinates will be given. What is the frequency of this precession ?

$$\tilde{u} = \frac{1}{2}(\tilde{\sigma}_{ab} + \tilde{\sigma}_{ba})$$
$$\tilde{v} = \frac{1}{2i}(\tilde{\sigma}_{ba} - \tilde{\sigma}_{ab})$$
$$\tilde{\omega} = \frac{1}{2}(\tilde{\sigma}_{aa} - \tilde{\sigma}_{bb})$$

By analogy with a classical field, we can write:

$$\frac{\mathrm{d}\tilde{\boldsymbol{U}}}{\mathrm{d}t} = \boldsymbol{\Omega} \times \tilde{\boldsymbol{U}} = \begin{pmatrix} \Omega_x \\ \Omega_y \\ \Omega_z \end{pmatrix} \times \begin{pmatrix} \tilde{u} \\ \tilde{v} \\ \tilde{w} \end{pmatrix} = \begin{pmatrix} \Omega_y \tilde{w} - \Omega_z \tilde{v} \\ \Omega_z \tilde{u} - \Omega_x \tilde{w} \\ \Omega_x \tilde{v} - \Omega_y \tilde{u} \end{pmatrix}$$

we deduce that

$$\boldsymbol{\Omega} = \begin{pmatrix} \Omega_1 \\ 0 \\ \omega_0 - \omega \end{pmatrix}$$

The Bloch vector $\boldsymbol{U}$ is precessing around the effective magnetic field $\boldsymbol{\Omega}$ with the frequency $\Omega = \sqrt{\Omega_1^2 + (\omega_0 - \omega)^2}$.

4. We focus on the case where the electric field frequency is resonant with the Bohr frequency of the transition, with the system initially in the ground state $|a\rangle$. Give the equation of motion of $\tilde{\boldsymbol{U}}$. How is the precession modified as compared to the case treated in question 3? What is its new frequency? What is the time evolution of the excited state population?

If the electric field is resonant with the Bohr frequency, this means that $\omega_0 - \omega = 0$ and thus,

$$\frac{\mathrm{d}}{\mathrm{d}t}\tilde{\boldsymbol{U}} = \begin{pmatrix} 0 \\ -\Omega_1 \tilde{\omega} \\ \Omega_1 \tilde{v} \end{pmatrix}$$

It is easy to find that

$$\left| \begin{array}{l} \tilde{u} = 0 \\ \tilde{v} = -\frac{1}{2}\sin(\Omega_1)t \\ \tilde{w} = \frac{1}{2}\cos(\Omega_1)t \end{array} \right.$$

$\boldsymbol{\Omega}$ is along the OX axis of the rotating frame and $\boldsymbol{U}$ precesses around $\boldsymbol{\Omega}$, in the (y, z) plane, at the Rabi frequency Ω_1.The population of the excited state is

$$\tilde{\sigma}_{bb} = \sin^2\left(\frac{\Omega_1}{2}t\right)$$

II - Optical pumping

We will study in this exercise the effect of "optical pumping" of an atom. We will show that the selection rules governing the interaction of an atom with circularly polarized light lead, under continuous illumination of the ground-to-excited state transition, to a non-equilibrium distribution of population among the sub-levels of the ground state.

We consider an atomic transition between a ground state of total angular momentum $J_a = 1/2$ and an excited state with total angular momentum $J_b = 1/2$. In the absence of incident light, the two ground-state sub-levels, which have the same energies, are equally populated: half of the atoms are initially in each of the states $m_a = -1/2, +1/2$. The quantization axis is chosen to be along the z direction.

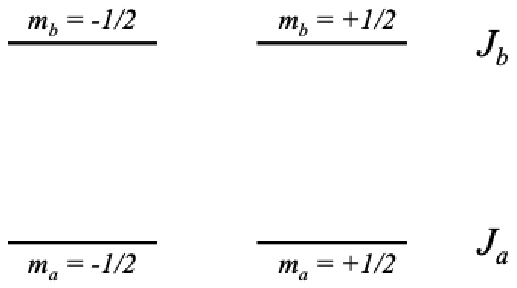


Figure 1 : diagram of ground and excited states of the studied atom/

1. The population of atoms interacts with a σ^+ circularly polarized light, the frequency of which is resonant with the $J_a = 1/2 \to J_b = 1/2$ transition. Which one of the two possible excited states is populated through light absorption? Justify your answer by discussing the optical selection rules.

   The transition induced by σ^+ is $|m_a = -1/2\rangle \to |m_b = +1/2\rangle$, we have the electric dipole operator of a σ^+ polarized light that is,

   $$\langle f|\, W + |i\rangle \propto \langle n_f, l_f, m_f|\, x + iy\, |n_i, l_i, m_i\rangle \propto \int \sin\theta \mathrm{d}\theta \mathrm{d}\varphi Y_{l_f}^{m_f *}(\theta,\varphi) Y_1^1(r) Y_{l_i}^{m_i}(\theta,\varphi)$$

   which by using the property of the spherical harmonics shows that $m_f = m_i + 1$.

2. We assume that atoms can relax from the excited state determined in question 2 to their ground state by spontaneous emission. What are the possible final ground-states of the relaxation? What is the polarization of the emitted light?

   The possible final ground-state of this process is $|m_a = \pm 1/2\rangle$ and thus it's two possible polarizations : σ^- or π.

3. Under continuous illumination with σ^+ circularly polarized light a steady state is reached. Experimentally it is found that the incoming light is no longer absorbed. The progress

towards the steady state can be monitored by measuring the transmission of the pumping light (see figure below). As a consequence, the σ^+ circularly polarized emission goes to zero intensity. What does this suggest for the population of the ground state levels? Explain the absorption-emission process that is responsible for the evolution of the system towards this non-equilibrium optically pumped distribution in the ground state?

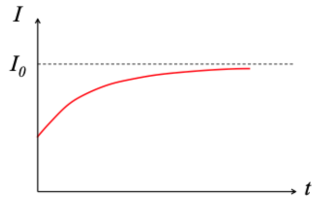


Figure 2 : Temporal evolution of the transmitted light intensity. In the steady-state the absorption goes to zero, i.e. $I=I_0$.

III - Rotation-vibration spectrum Diatomic molecules

The nuclear Hamiltonian of a diatomic molecule is written:

$$-\frac{\hbar^2}{2\mu}\frac{1}{r}\frac{\partial^2}{\partial r^2}r + \frac{\hbar^2}{2\mu r^2}L^2 + V(r)$$

where r is the inter-nuclear distance, and μ is the reduced mass of the two-body system. L is the angular momentum operator and $V(r)$ is the potential energy of interaction that is assumed to be harmonic.

1. By looking for the solution of the corresponding Schrödinger equation under the form:

$$\chi_{n,l,m}(r,\theta,\phi) = \frac{1}{r}u_n(r)Y_l^m(\theta,\phi)$$

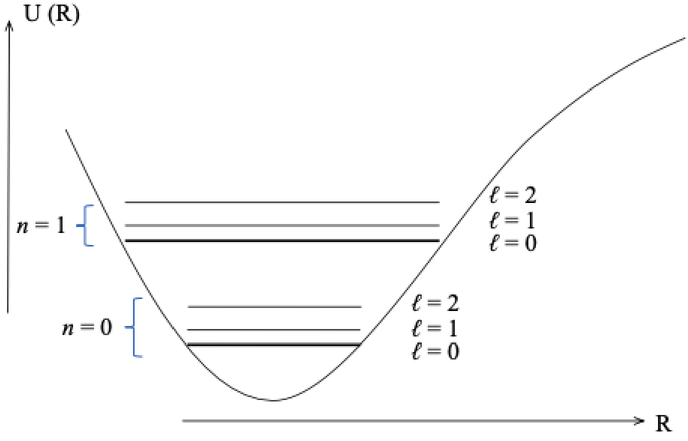
where $u_n(r)$ is the eigenfunction of a one-dimensional harmonic oscillator and $Y_l^m(\theta,\phi)$ is a spherical harmonic, compute the expression of the energy eigenvalues $E_{n,l}$ (assume that the centrifugal energy does not depend on r and is equal to its value evaluated at the average inter-nuclear distance r_0). Identify the vibrational and rotational terms. Make a schematic diagram of the energy spectrum.

We can susbtitute in Schrödinger's equation,

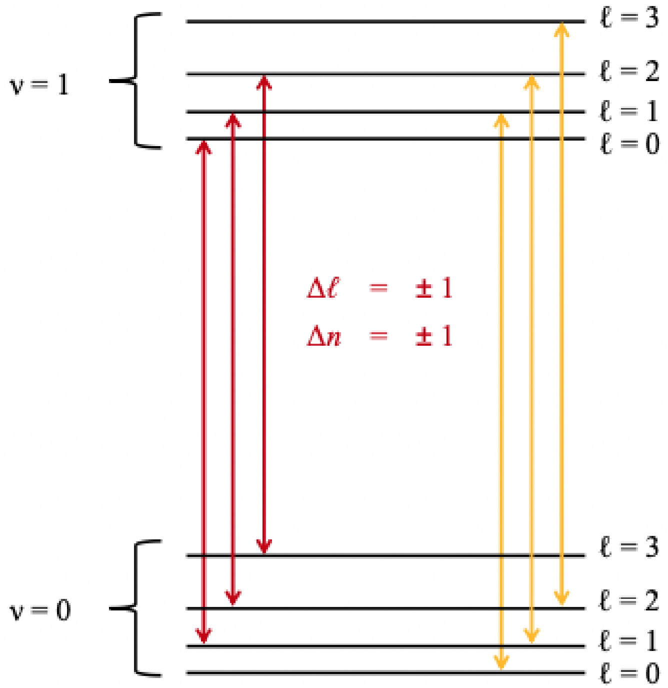
$$\left[-\frac{\hbar^2}{2\mu}\frac{1}{r}\frac{\partial^2}{\partial r^2}r + \frac{\hbar^2}{2\mu r^2}L^2 + V(r)\right]\left(\frac{1}{r}u_n(r)Y_l^m(\theta,\phi)\right)$$

and we remember that $L^2 Y_l^m = l(l+1)\hbar^2 Y_l^m$ and assuming that $U(r_0) = 0$ alongside that $r \approx r_0$ we get,

$$E_{\nu,l} = \hbar\omega_0(\nu + 1/2) + \frac{\hbar^2 l(l+1)}{2\mu r_0^2}$$



2. Within the dipolar approximation, recall the selection rules for vibrational and rotational transitions, between the eigenstates labelled by n (harmonic oscillator) and l (rotation of a molecule with moment of inertia I).



3. The figure below displays the infrared absorption spectrum of HBr. Label the absorption lines with vibrational and rotational quantum numbers.

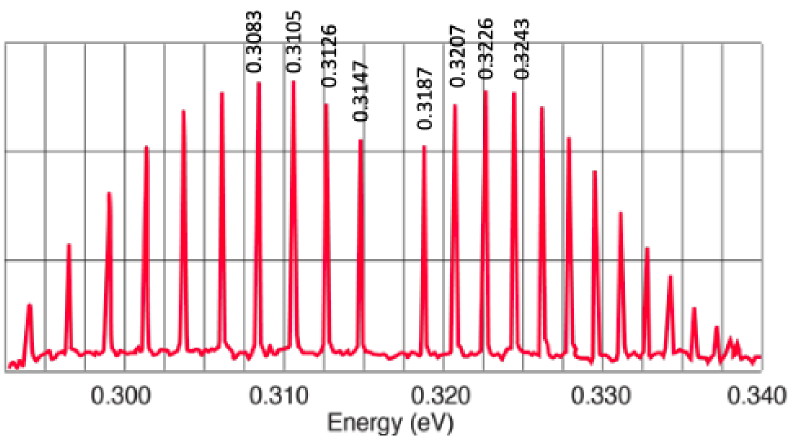


Figure 3 : Sketch of the vibration-rotation spectrum of HBr

left is P-branch, meaning $l_f = l_i - 1$. Right is R-branch, meaning $l_f = l_i + 1$.

4. What is the origin of the decreasing peak intensity on both sides of the spectrum?

5. Show that, for the intense peaks in the doublets, the rotational constant $B = \frac{\hbar^2}{2I}$ is equal to ≈ 1 meV. What is the value of the moment of inertia I ?

6. Calculate the reduced mass and calculate the interatomic radius R_0 from the value of I.

7. From the vibrational frequency ω, show that the force constant k is equal to 379 Nm^{-1} (use the first peak of the R band to determine the vibrational transition frequency $\hbar\omega$). $M_{\mathrm{Br}} = 80$ u, $M_{\mathrm{H}} = 1$ u with 1 u$= 1.66 \times 10^{-27}$ kg.