

Atomic State Transfer via Adiabatic Passage

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Abstract

In this paper I seek to explain how the valence electron of an atom may be transferred from one energy level to another reliably via adiabatic passage. First I state and derive the adiabatic theorem. Next I derive Rabi Flops- the oscillation of an electron between two energy levels as a result of bombardment by a laser. The state of a two-level system, as the atom will be described by, may be represented on a Bloch Sphere, so I discuss the transformation from the math behind Rabi Flops to the Bloch Sphere visual representation.

When a laser impinges upon an atom, the eigenstates become superpositions of the ground and excited energy states that oscillate in time. I show that if the laser begins highly detuned and sweeps slowly through resonance that the electron state is carried from the ground state into the excited state with a very high probability.

Adiabatic Theorem

The Adiabatic Theorem states that if a Hamiltonian is varied slowly from one form to another, that the n^{th} eigenstate of the original Hamiltonian will be shifted to the n^{th} eigenstate of the resulting Hamiltonian. This theorem requires that the eigenstates be non-degenerate.

This theorem may be derived by looking for a separable solution to a Hamiltonian that varies slowly in time, with a time-dependant energy eigenvalue. Typically when this approach is taken, it is done for a time-independent Hamiltonian. For a time-varying Hamiltonian, it is necessary to allow the spatial component of the wavefunction to vary in time. This approach only makes sense if the states themselves vary slowly in time since:

$$\Psi_n(\vec{r}, t) = \phi_n(\vec{r}, t)T_n(t) \longrightarrow i\hbar(\dot{\phi}_n T_n + \phi_n \dot{T}_n) = \hat{H}\phi_n T_n$$

Here, we have assumed that the Hamiltonian has Eigenvalues with corresponding eigenfunctions Ψ_n . By requiring that $\dot{\phi}T \ll \phi\dot{T}$, we may reproduce the typical separable equations. This condition is met if the Hamiltonian varies slowly in time. The equations produced are:

$$\hat{H}(\vec{r}, t)\phi = E_n(t)\phi_n$$

$$i\hbar\dot{T}_n = E_n(t)T_n$$

Thus it was reasonable to assume that the Hamiltonian had time-varying eigenvalues earlier. This gives the time-dependant phase of the wavefunction, thus the general wavefunction may be written as

$$\Psi(\vec{r}, t) = \sum_n c_n(t) \phi_n(\vec{r}, t) e^{-i\Theta_n(t)} \quad \text{where} \quad \Theta_n(t) = \frac{-1}{\hbar} \int_0^t E_n(t') dt'$$

Plugging this form of $\Psi(\vec{r}, t)$ into the Schrodinger equation gives:

$$i\hbar \sum_n [\dot{c}_n \phi_n + c_n \dot{\phi}_n + i c_n \phi_n \dot{\Theta}_n] e^{i\Theta_n} = \sum_n c_n \hat{H}(t) \phi_n e^{i\Theta_n}$$

The last term on the left side of this equation cancels the right side because $\hat{H}(t) \phi_n = E_n \phi_n$ and $-\hbar \dot{\Theta}_n = E_n$, leaving:

$$\sum_n \dot{c}_n \psi_n e^{i\Theta_n} = - \sum_n c_n \dot{\psi}_n e^{i\Theta_n}$$

Multiplying both sides by ψ_m and integrating over the spatial coordinate gives:

$$\dot{c}_m = - \sum_n c_n \langle m | \dot{\psi}_n \rangle e^{i(\Theta_n - \Theta_m)}$$

Now we take a detour to derive an expression for the right side of this equation that we can show is equal to zero, so that the coefficients, c_m , are constant in time, proving that the n^{th} state of the initial Hamiltonian becomes the n^{th} state of the final Hamiltonian.

$$\frac{d}{dt} \langle H | n \rangle = \frac{d}{dt} \langle E_n | n \rangle$$

$$\dot{H} | n \rangle + H | \dot{n} \rangle = \dot{E}_n | n \rangle + E_n | \dot{n} \rangle$$

Multiply on the left side by $\langle m |$:

$$\langle m | \dot{H} | n \rangle + \langle m | H | \dot{n} \rangle = \dot{E}_n \delta_{mn} + \langle m | E_n | \dot{n} \rangle$$

Thus, for $m \neq n$:

$$\langle m | \dot{H} | n \rangle = (E_n - E_m) \langle m | \dot{n} \rangle$$

Plugging this into the expression for \dot{c}_m from before gives:

$$\dot{c}_m = - \sum_{n \neq m} c_n \frac{\langle m | \dot{H} | n \rangle}{E_n - E_m} e^{i(\Theta_n - \Theta_m)} - c_m \langle m | \dot{m} \rangle$$

If \dot{H} is very small, then the sum vanishes, leaving

$$\dot{c}_m = c_m \langle m | \dot{m} \rangle \implies c_m(t) = c_m(0) e^{i\gamma_m(t)}$$

$$\gamma_m(t) = i \int_0^t \langle m(t') | \dot{m}(t') \rangle dt'$$

This integral must be completely imaginary because the states are orthogonal to each other and $\langle m | \dot{m} \rangle = \langle \dot{m} | m \rangle^*$:

$$\frac{d}{dt} \langle m | m \rangle = 0 = \langle m | \dot{m} \rangle + \langle \dot{m} | m \rangle = 2\text{Re}[\langle m | \dot{m} \rangle]$$

thus c_m merely picks up a phase as the Hamiltonian is changed. This phase is referred to as "Berry's Phase" and is measurable. The important feature of this derivation, however, is that the magnitude of each coefficient is left unchanged, so the n^{th} eigenstate of the original Hamiltonian becomes the n^{th} eigenstate of the new Hamiltonian, so long as the change happens slowly.

Rabi Flops

If an atom is subjected to a laser that is resonant with one of its electron transitions, then the electron will oscillate between the ground state and excited state of that transition. If the laser is detuned from that resonance frequency, the electron will no longer be able to fully populate the excited state, but will still oscillate between the ground state and a superposition of the two states. This discovery comes from solving a Hamiltonian that is comprised of the energies associated with each atomic energy level added to the interaction energy of the atom with an external oscillating electric field.

To derive this, we begin with a general statement: If you have a Hamiltonian whose energies and eigenstates are known ($\hat{H}_o |n\rangle = E_n |n\rangle$), then these can be used to describe the state of a new Hamiltonian, $H = H_o + H'$. Since the original state kets are not eigenstates of H , their coefficients will vary in time.

$$i\hbar \frac{\partial}{\partial t} |\Psi\rangle = H |\Psi\rangle$$

$$|\Psi\rangle = \sum_k c_k(t) |k\rangle e^{-i\omega_k t}$$

Multiplying on both sides of the Schrodinger equation by $\langle j |$ gives:

$$i\hbar \langle j | \frac{\partial}{\partial t} |\Psi\rangle = \langle j | (H_o + H') \sum_k c_k(t) |k\rangle e^{-i\omega_k t}$$

$$i\hbar[\dot{c}_j - i\omega_j c_j]e^{-i\omega_j t} = c_j E_j e^{-i\omega_j t} + \sum_k c_k \langle j|H'|k\rangle e^{-i\omega_k t}$$

Since $E_j = \hbar\omega_j$, the second term on the left cancels the first term on the right:

$$i\hbar\dot{c}_j = \sum_k c_k \langle j|H'|k\rangle e^{i(\omega_j - \omega_k)t}$$

In the interest of the Rabi problem, we apply the above equation to a system with only two states, the ground energy state ($|g\rangle$) and the excited energy state ($|e\rangle$). The additional Hamiltonian is the interaction energy of the atom with n external oscillating electric field, $H' = -eE_o \cos(kz - \omega_l t)\hat{\epsilon}\cdot\vec{r}$, with polarization $\hat{\epsilon}$:

$$i\hbar\dot{c}_g(t) = c_e(t)\langle g|H'|e\rangle e^{-i\omega_a t}$$

$$i\hbar\dot{c}_e(t) = c_g(t)\langle e|H'|g\rangle e^{i\omega_a t}$$

ω_a is the resonant frequency of the transition that we are describing with this Hamiltonian. In order to uncouple these first order differential equations, we make the rotating wave approximation to simplify the $\cos(kz - \omega_l t)e^{i\omega_a t}$ term:

$$\cos(kz - \omega_l t)e^{i\omega_a t} = \frac{e^{i(kz - (\omega_l - \omega_a)t)} + e^{-i(kz - (\omega_l + \omega_a)t)}}{2} \approx \frac{e^{i(kz - (\omega_l - \omega_a)t)}}{2}$$

This approximation is valid when $\omega_l - \omega_a \ll \omega_l + \omega_a$, which conveniently is the situation that we are interested in when describing a stimulated electron transition. When this condition is met, the exponential containing the $\omega_l + \omega_a$ term oscillates much faster than the other term, and thus averages out over a short period of time.

Making this approximation and decoupling the equations for c_g and c_e gives:

$$\ddot{c}_g - i\delta\dot{c}_g + \frac{\Omega^2}{4}c_g = 0$$

$$\ddot{c}_e + i\delta\dot{c}_e + \frac{\Omega^2}{4}c_e = 0$$

where the detuning is defined as $\delta \equiv \omega_l - \omega_a$, and the Rabi Frequency is $\Omega \equiv \frac{-eE_o}{\hbar}\langle e|\hat{\epsilon}\cdot\vec{r}|g\rangle$. The matrix element, $\langle e|\hat{\epsilon}\cdot\vec{r}|g\rangle$, can be found by advanced methods that are not discussed here.

For a state that is initially entirely in the ground state, the solution of these equations is:

$$c_g(t) = \left[\cos\left(\frac{\Omega t}{2}\right) - i\frac{\delta}{\Omega'} \sin\left(\frac{\Omega t}{2}\right) \right] e^{i\frac{\delta}{2}t}$$

$$c_e(t) = -i \frac{\Omega}{\Omega'} \sin\left(\frac{\Omega' t}{2}\right) e^{-i \frac{\delta}{2} t}$$

The oscillation frequency is $\frac{\Omega'}{2} \equiv \frac{1}{2} \sqrt{\Omega^2 + \delta^2}$. It increases as the detuning is increased. There are two prominent features of these equations. One is that the maximum amplitude of the population of the excited state decreases as the detuning is increased. The other is that the oscillation frequency increases as the detuning increases. These oscillating energy states are the eigenstates of the laser-atom Hamiltonian.

Bloch Sphere Representation

By a clever substitution of variables, the Rabi flop equations may be represented as vectors that behave identically to a classical precession problem. To do so, we define the state vector as $\vec{R} \equiv u\hat{u} + v\hat{v} + w\hat{w}$. Where

$$\begin{aligned} u &= c_g c_e^* + c_g^* c_e \\ v &= i(c_g c_e^* - c_g^* c_e) \\ w &= |c_e|^2 - |c_g|^2 \end{aligned}$$

In so doing, the state of the system is represented by a vector of unit length that touches the surface of a sphere (dubbed the Bloch Sphere) centered on the origin. In the ground state, the vector points along the negative w axis. In the excited state, the vector points along the positive w axis. In an even superposition of states the vector lies along the equator of this imaginary sphere, and the phase difference between the ground and excited state is depicted by the polar angle that the vector points at. Of course, any other mixture of states may be represented with this vector.

Rabi Flops may also be represented in the Bloch Sphere picture by defining the Rabi vector, $\vec{\Omega}' \equiv \Omega\hat{x} + \delta\hat{z}$. With this done, the Rabi flop equations reduce simply to $\dot{\vec{R}} = \vec{R} \times \vec{\Omega}'$, the equation for classical precession. The rotating state vector is, as before, an eigenstate of the laser-atom Hamiltonian.

Adiabatic Passage

When an atom is subjected to a laser, the eigenstates of the Hamiltonian cease to be energy eigenstates, rather they become oscillating superpositions of the two (represented on the Bloch Sphere by a state vector that rotates around the Rabi vector). By shining a greatly detuned ($\delta \gg \Omega$) laser on the atom, the eigenstates become tiny rotations of the state vector around the nearly vertical Rabi vector. If the laser is slowly swept through resonance to highly detuned in the opposite direction, then the Hamiltonian shifts adiabatically, and the rotating state vector follows the Rabi vector. At the end

of this transfer, the state vector is making tiny oscillations near the top of the Bloch Sphere, thus it is composed almost entirely of the excited energy state and has a very large probability of being measured in the excited state. This is adiabatic passage.

One caveat that must be addressed in order for this procedure to work is that the duration of adiabatic passage must be small compared to the natural lifetime of the atomic transition to avoid the risk of spontaneous emission during the process. This requires the incoming laser to have a sufficient amount of power, since the Rabi Frequency is dependant upon the power of the laser, and also determines the rate at which the detuning may be shifted.

References

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