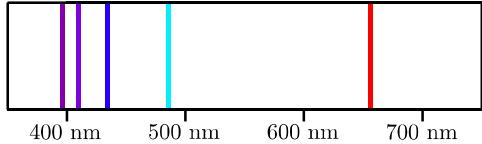


3 Rydberg Atoms

In a nutshell, a Rydberg atom is an atom whose valence electron is in a state with very high principal quantum number $n \gg 1$. As a consequence, these atoms possess very exaggerated properties, such as very long lifetimes and, most importantly, extremely large dipole moments which lead these atoms to display interactions among them which are very strong over very long distances.



Historically, the first time this concept appeared was in 1885, with Balmer's formula for the wavelengths of the visible absorption lines of hydrogen, given by

$$\lambda = \frac{bn^2}{n^2 - 4}$$

with $b = 3645.6 \text{ \AA}$. Actually, this formula gives the wavelengths corresponding to electronic transitions from a state of hydrogen with $n = 2$ to higher lying levels with principal quantum number n , as it can be seen writing it as

$$\frac{1}{\lambda} = \frac{4}{b} \left(\frac{1}{4} - \frac{1}{n^2} \right) .$$

Similar formulas were found for other series by Lyman, Brackett, Paschen ... and then summarized by Johannes Rydberg in 1890 as

$$\frac{1}{\lambda} = \frac{R_y}{hc} \left(\frac{1}{n_2^2} - \frac{1}{n_1^2} \right)$$

where n_1 and n_2 represent the initial and final state of the electron, respectively. Here, $R_y = 13.6 \text{ eV}$ is the Rydberg unit of energy, which also determines the binding energy of the electron in the hydrogen atom

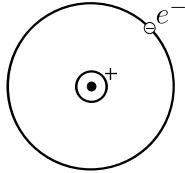
$$W = -\frac{R_y}{n^2},$$

which is the minimum energy necessary for an electron in a state with principal quantum number n to leave the atom (obtained by putting n_2 to infinity).

3.1 Properties of Rydberg atoms

In this subsection we will get to know a few of the very exaggerated properties that make Rydberg atoms unique.

3.1.1 Binding energy



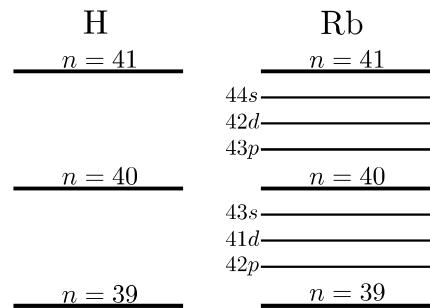
The description of alkali atoms such as Rb, Na and K with their valence electron in a Rydberg state is similar to the hydrogen, as they are both to first order formed by an electron orbiting a core of net positive $+e$ charge. The difference between the two is that in the alkali atom the positive core is formed by a nucleus with charge $+Z$ and $Z - 1$ electrons.

Hence, it has a structure and it can only be considered point-like, as a single proton, when the electron is very far away from the nucleus. This is actually the case when the electron is in a high angular momentum state (i.e., with $l \geq 4$). Here, the basic properties of the two systems are the same, since in these states the orbits of the valance electron are such that they do not penetrate the ionic core, and hence they experience a potential very similar to the usual Coulomb one ($\propto -1/r$). On the other hand, when l takes low values (e.g. $l = 0, 1, 2 \dots$, corresponding to s, p or d states), the valence electron penetrates the inner core, probing its structure, and, consequently, the wave functions and energy levels of an alkali Rydberg atom differ from their hydrogen counterparts.

This effect can be accounted for in the binding energy of an alkali Rydberg atom, which is accurately described by

$$W = -\frac{R_y}{n^{*2}} = -\frac{R_y}{(n - \delta_l)^2}.$$

Here, we have introduced δ_l , which stands for the so-called *quantum defect*, and $n^* = n - \delta_l$, called the *effective principal quantum number*. The quantum defect δ_l is particularly high for low l quantum states. The consequence of this quantum defect in the spectrum of an alkali atom is that, while in the hydrogen all states with the same principal quantum number n form degenerate manifolds, In the alkali atoms this degeneracy is broken, and the low l states are notably shifted in energy, as shown schematically in the figure.



3.1.2 Dipole matrix elements

Transitions between atomic states primarily occur due to coupling with the electric dipole moment $\vec{d} = -e\vec{r}$ (remember section 1.3). In order to find the scaling of this dipole moment with the principal quantum number in the case of a Rydberg state, let us consider the situation where the electron performs a transition between two states with the same n and l differing by one, such that the transition is dipole permitted, i.e.

$$|n, l, m_l\rangle \longrightarrow |n, l \pm 1, m_l\rangle .$$

Note that the energy difference between these two levels is not very large. The transition dipole moment here is calculated as

$$\begin{aligned}\langle \vec{d} \rangle &= \langle n, l, m_l | \vec{d} | n, l \pm 1, m_l \rangle = -e \langle n, l, m_l | \vec{r} | n, l \pm 1, m_l \rangle \\ &= -e \left[\langle n, l, m_l | r \sin(\theta) \cos(\varphi) \hat{x} + r \sin(\theta) \sin(\varphi) \hat{y} + r \cos(\theta) \hat{z} | n, l \pm 1, m_l \rangle \right],\end{aligned}$$

where we have separated the components of the dipole into each individual direction, i.e. $\langle \vec{d} \rangle = \langle d_x \rangle \hat{x} + \langle d_y \rangle \hat{y} + \langle d_z \rangle \hat{z}$.

In order to calculate each of these components (here, for example, the z -component), we need to consider that the wave functions, just like in the hydrogen case, can be separated into radial and angular coordinates, i.e.

$$|n, l, m_l\rangle \propto R_{nl}(r) Y_{lm}(θ, φ),$$

such that

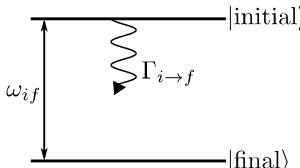
$$\langle d_z \rangle \propto \int_{\Omega} d\Omega Y_{lm_l}(\theta, \varphi) \cos(\theta) Y_{l \pm 1 m_l}^*(\theta, \varphi) \int_0^{\infty} dr r^3 R_{nl}(r) R_{nl \pm 1}^*(r) .$$

Here, one can see that the first term (angular component) is independent of the principal quantum number n . On the other hand, for large enough orbital quantum numbers l , the two radial components are extremely similar, such that the second component is very close to the average distance of the electron to the nucleus, $\langle r \rangle$, which in turn is proportional to n^2 . Since here $n \gg 1$, this gives rise to *very strong dipole moments*. Note, however, that these are not permanent dipoles, but induced dipoles due to the electron being most of the time very far ($\langle r \rangle$ can be up to a micrometer) from the nucleus.

3.1.3 Lifetime

Another attractive feature of Rydberg atoms (i.e. atoms with the valence electron in a Rydberg state) is that these states have very long radiative lifetimes. This seems perhaps counter-intuitive at first, because the electron is so highly excited and consequently close to the ionization threshold. We can estimate the scaling of the lifetime with n by considering what we know from section 1.3.

Let us consider a two-level system as the one in the figure. We calculated that the rate of decay from the initial to final state is given by



$$\Gamma_{i\rightarrow f} \propto \omega_{if}^3 \left| \langle \vec{d} \rangle_{if} \right|^2 ,$$

where $\hbar\omega_{if}$ is the energy difference between the two states and $\langle \vec{d} \rangle_{if}$ is the corresponding transition dipole moment. Let us now consider the scaling of these observables to obtain the one of the lifetime of the initial state, τ , which is inversely proportional to the decay rate.

First, for a state with large orbital quantum number (l close to n), we have calculated that $\left| \langle \vec{d} \rangle_{if} \right| \propto n^2$. Moreover, here the quantum defect is very small and hence the energy difference between states with the same n scales as $\omega_{if} \propto 1/n^3$. This overall gives that

$$\Gamma_{i\rightarrow f}^{(l\approx n)} \propto \frac{1}{n^9} n^4 = \frac{1}{n^5} .$$

On the other hand, when $l = 0, 1, \dots$ the calculation needs to be done considering the specific form of wave functions of the alkali atom, slightly different from the hydrogen ones. This calculation results in

$$\Gamma_{i\rightarrow f}^{(l\approx 0)} \propto \frac{1}{n^3} .$$

Hence, the lifetimes of the Rydberg states $\tau \propto 1/\Gamma_{i\rightarrow f}$ grow rapidly with the principal quantum number, between the two limiting scalings n^5 and n^3 . As an example, for a rubidium atom:

$$n = 60 \quad l = 1 \longrightarrow \tau \approx 7.2 \text{ }\mu\text{s}$$

$$n = 60 \quad l = 54 \longrightarrow \tau \approx 70 \text{ ms}$$

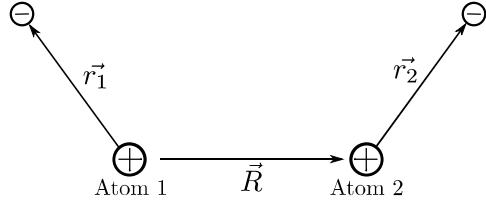
in both cases having values orders of magnitude larger than the lifetimes of low-lying

excited states (for example 11 ns for the first excited state of rubidium), and in some cases larger than the timescale in which experiments with cold atoms are realized, making them effectively stable over the experimental time.

3.2 Interactions between Rydberg atoms

In this section we will study how two or more Rydberg atoms interact with each other. We will then study the consequences of these interactions in the dynamics of a Rydberg gas driven by a laser field, in particular focussing on the phenomenon so-called Rydberg blockade.

3.2.1 Dipole-dipole Interactions

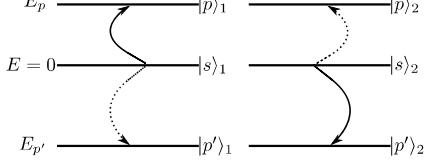


Above we have a schematic of two Rydberg atoms separated by a distance \vec{R} . Each atom as a nucleus and valence electron separated by the relative distances \vec{r}_1 and \vec{r}_2 (for atom 1 and 2, respectively). One can obtain the interaction between the two atoms (here represented for simplicity as dipoles) simply by considering the Coulomb interactions between the charges and using that $|\vec{R}| \gg |\vec{r}_1|, |\vec{r}_2|$:

$$\begin{aligned}
 V &= e^2 \left[\frac{1}{|\vec{R}|} + \frac{1}{|\vec{R} + \vec{r}_2 - \vec{r}_1|} - \frac{1}{|\vec{R} + \vec{r}_2|} - \frac{1}{|\vec{R} - \vec{r}_1|} \right] \\
 &\stackrel{(*)}{=} e^2 \left[\frac{1}{R} + \frac{1}{R\sqrt{1 + \frac{r_1^2 + r_2^2 - 2\vec{r}_1 \cdot \vec{r}_2}{R^2} + 2\frac{\vec{r}_2 \cdot \hat{R} - \vec{r}_1 \cdot \hat{R}}{R}}} - \frac{1}{R\sqrt{1 + \frac{r_2^2}{R^2} + 2\frac{\vec{r}_2 \cdot \hat{R}}{R}}} - \frac{1}{R\sqrt{1 + \frac{r_1^2}{R^2} - 2\frac{\vec{r}_1 \cdot \hat{R}}{R}}} \right] \\
 &\underset{\text{up to } 1/R^3}{\approx} (R \gg r_1, r_2) \approx e^2 \frac{[\vec{r}_1 \cdot \vec{r}_2 - 3(\vec{r}_1 \cdot \hat{R})(\vec{r}_2 \cdot \hat{R})]}{R^3}.
 \end{aligned}$$

Note that this is simply a multipole expansion. Since the atoms are neutral, the 1st and 2nd order terms are zero and the $1/R^3$ term is the dominant one.

3.2.2 Simple model for interactions



In a real atom, the spectrum can be very complicated and the amount of levels one may need to consider to obtain a detailed and realistic model for the interactions, large. However, let us consider a simplified model that gives a quite accurate depiction of the interactions.

To do so, we consider two atoms with three energy levels, $|s\rangle \equiv |ns\rangle$, $|p\rangle \equiv |np\rangle$ and $|p'\rangle \equiv |n'p\rangle$. Both p -states can be dipole-coupled to the s -state. For simplicity, let us put the energy of the s -state to zero, and $E_p > 0$ and $E_{p'} < 0$.

Now we will consider the possible states of the two atoms, and to do so we consider that the atoms are both initially in the $|s\rangle$ state. The only states directly coupled to this state are states where both atoms are in a p -state. Hence, we can consider as our basis the states

$$\begin{aligned} |S\rangle &\equiv |s\rangle_1 \otimes |s\rangle_2 \\ |P\rangle &\equiv |p\rangle_1 \otimes |p\rangle_2 \\ |P'\rangle &\equiv |p'\rangle_1 \otimes |p'\rangle_2 \\ |PP'\rangle &\equiv \frac{1}{\sqrt{2}} [|p\rangle_1 \otimes |p'\rangle_2 + |p'\rangle_1 \otimes |p\rangle_2] . \end{aligned}$$

In order to further simplify our model, we will consider a regime in which

$$\delta = E_p + E_{p'} \ll 2E_p, 2E_{p'} .$$

As a consequence, $|P\rangle$ and $|P'\rangle$ are well out of resonance and the transition probabilities to these states is negligible. Within these approximations, the Hamiltonian describing the interaction between the states $|S\rangle$ and $|PP'\rangle$ is:

$$H_{\text{int}} = \hbar \begin{pmatrix} 0 & V \\ V & \delta \end{pmatrix}$$

where we have used as basis states $|S\rangle \equiv \begin{pmatrix} 0 \\ 1 \end{pmatrix}$, $|PP'\rangle \equiv \begin{pmatrix} 1 \\ 0 \end{pmatrix}$.

In order to obtain the actual interactions between two Rydberg atoms, we need to

diagonalize this Hamiltonian, which gives the eigenenergies

$$E_{\pm} = \frac{\delta}{2} \pm \frac{1}{2} \sqrt{\delta^2 + 4V^2} ,$$

where one can see that the energy of the pair of states depends upon the separation between the atoms via V . The specific form of the spatial dependence can be derived in two distinct regimes:

i) *Long range* ($V(R) \ll \delta$)

Here the energies $E_{\pm} = \frac{\delta}{2} \pm \frac{\delta}{2} \sqrt{1 + \frac{4V^2}{\delta^2}}$ can be expanded for large δ as

$$\begin{aligned} E_+ &\approx \delta + \frac{V^2}{\delta} \\ E_- &\approx -\frac{V^2}{\delta} \end{aligned}$$

Moreover, here the eigenstate $|-\rangle \approx |S\rangle$, such that here two atoms in the ns -state experience an energy shift level determined by the *van-der-Waals potential*:

$$V_{vdW} = -\frac{C_6}{R^6} ,$$

with $C_6 = \alpha^2/\delta$ being the van-der-Waals coefficient and

$$\alpha \equiv \langle s | e^2 \left(\vec{r}_1 \cdot \vec{r}_2 - 3 (\vec{r}_1 \cdot \hat{R}) (\vec{r}_2 \cdot \hat{R}) \right) | PP' \rangle$$

the *polarizability*. Note now, that since we know that the transition dipole moment of a Rydberg atom is proportional to n^2 , the polarizability α is proportional to n^4 . Moreover, we also know that the energy difference between nearby levels goes as $\delta \propto 1/n^3$. Hence,

$$C_6 \propto n^{11}$$

which in turn gives rise to very strong interactions, even at large distances. As an example, consider that for Rb, two atoms in the $43s$ state give rise to

$$C_6 = -4.45 \cdot 10^{-27} \text{ MHz} \cdot \text{m}^6$$

which, for two atoms at $R = 3 \mu\text{m}$ gives an interaction of

$$V_{vdW} = 3.35 \text{ MHz} ,$$

orders of magnitude larger than between ground state atoms.

ii) *Short range* ($V(R) \gg \delta$)

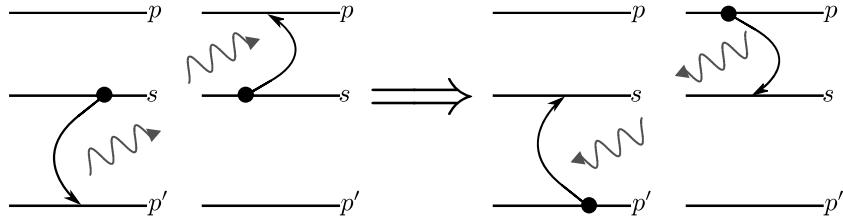
In this limit, the expansion of the energies for small δ gives:

$$E_{\pm} \approx \pm V = \pm \frac{C_3}{R^3} .$$

This is the resonant *dipole-dipole interaction* regime, where $C_3 = \alpha$, such that

$$C_3 \propto n^4 .$$

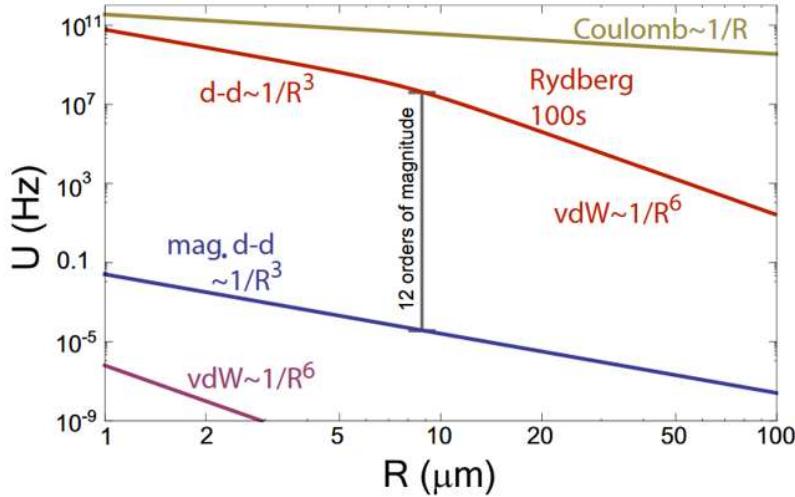
Here, the eigenstates of the interactions are superpositions of $|S\rangle$ and $|PP'\rangle$. In this limit, the interactions can be better understood as exchange interactions between these two states:



The transition between the two regimes $1/R^3$, $1/R^6$ occurs at the van der Waals radius

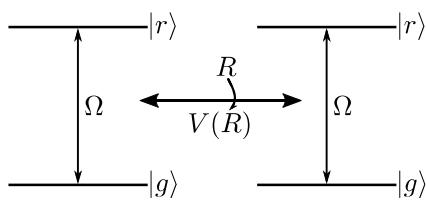
$$V(R_{vdW}) = \delta \Rightarrow R_{vdW} = \sqrt[6]{\left| \frac{C_6}{\delta} \right|} \propto n^{7/3} \quad (*)$$

An estimate of the interactions strengths can be found below, where a comparison with the interactions between the Rydberg and ground state interactions is shown. Moreover, here one can see clearly the onset of the van der Waals radius (where the gradient of the interactions change). Finally, note that there is an angular dependence of the interaction that we have not discussed in depth. This can become important, particularly for high l states, where the interactions can become highly anisotropic, or in the presence of external fields that fix the dipole direction.



3.2.3 Blockade effect

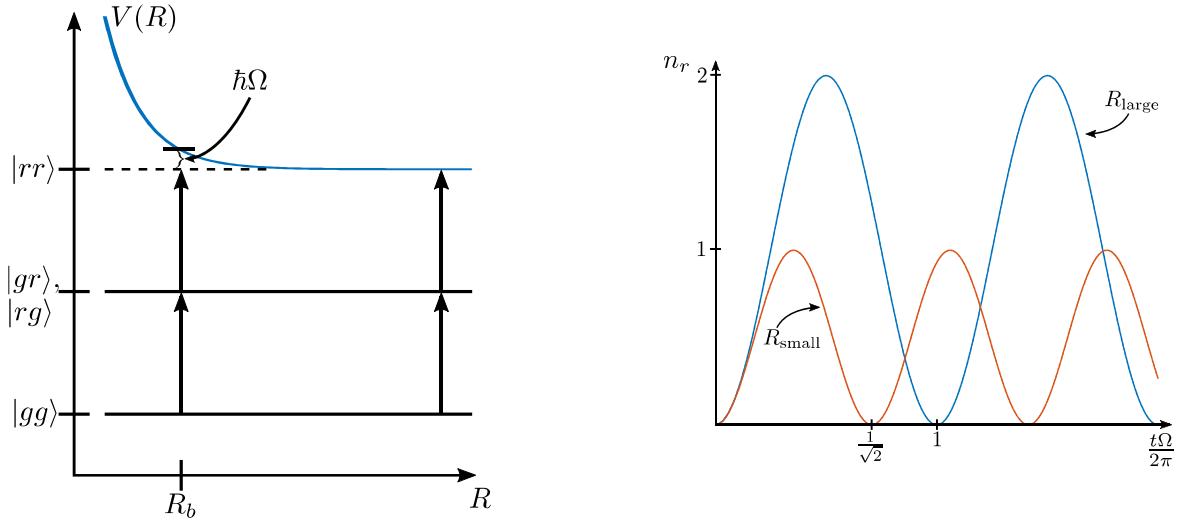
We have now established that for high principal quantum number n the Rydberg interactions lead to a large energy shift of the state where the two atoms are in the Rydberg state. We will now describe the dynamics of a system of two two-level atoms where the lower and upper level are the ground state and a Rydberg one, respectively.



In particular, let us consider a pair of atoms resonantly driven (by a laser), from their ground state $|g\rangle$ to a Rydberg state $|r\rangle$, with a Rabi frequency Ω . If the atoms are very far from each other, both atoms can be excited to the Rydberg state and hence the dynamics will be such that the atoms go from $|gg\rangle$ to $|rr\rangle$ with frequency Ω , as one can see in the figure below. If the atoms are closer together, however, the interaction causes the $|rr\rangle$ state to be detuned out of resonance for the laser, eventually preventing the excitation of the $|rr\rangle$ state. This can be observed by simply solving the Schrödinger equation with Hamiltonian (remember section 1.2):

$$H = \frac{\hbar\Omega}{2} \sum_{i=1,2} (\sigma_i^\dagger + \sigma_i) + V(R)n_1n_2$$

with $\sigma_i = |g_i\rangle \langle e_i|$ and $n_i = \sigma_i^\dagger \sigma_i$ being the operator that counts how many excitations there are in atom i .



This is the so-called *dipole blockade*, or *Rydberg blockade effect*. In the simplest model, the so-called *blockade radius* R_b , i.e., the distance at which we consider the $|rr\rangle$ state out of resonance, can be estimated by equating the van-der-Waals interaction with the power-broadened linewidth of the laser, given by its Rabi frequency, i.e. $V(R_b) = \hbar\Omega$, such that

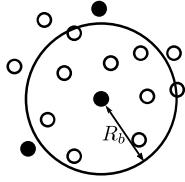
$$R_b = \sqrt[6]{\frac{C_6}{\Omega}}.$$

The consequence of the blockade effect is that within the blockade radius there can only be one Rydberg excitation taking place at a time. Coming back to the dynamics, one can easily obtain from the Hamiltonian (by changing the basis states $|gr\rangle$ or $|rg\rangle$ to their symmetric and antisymmetric superposition states) that in the case of two atoms, the state excited by the laser in the blockaded limit is not $|gr\rangle$ nor $|rg\rangle$, but rather

$$|\Psi_s\rangle = \frac{1}{\sqrt{2}} [|gr\rangle + |rg\rangle] ,$$

i.e. a symmetric superposition between the two singly excited states. Moreover, the frequency at which this state is excited is increased to be $\sqrt{2}\Omega$.

Finally, note that for an ensemble of N_b atoms localized within a radius $R < R_b$ the blockade also applies. Here, as only one atom can be excited within the full blockaded sphere, the system oscillates between the ground state and the symmetric superposition state



$$|g^{N_b-1}r\rangle = \frac{1}{\sqrt{N_b}} \sum_{i=1}^{N_b} |g_1 g_2 \dots r_i \dots g_{N_b}\rangle .$$

The coupling between these two states gives an enhanced collective Rabi frequency

$$\Omega_{N_b} = \sqrt{N_b} \Omega .$$

This effect has been observed in a large amount of experiments, and even exploited for quantum information and computation purposes.

3.3 Rydberg quantum optics

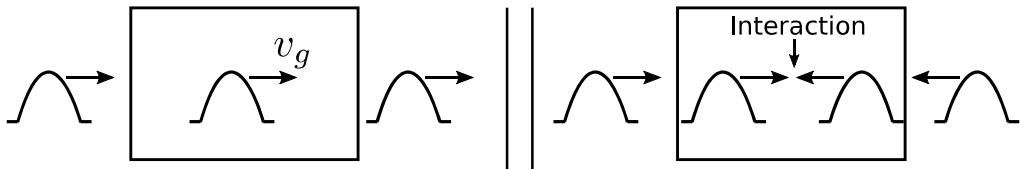
In this section we will focus on one example of application of the interactions between Rydberg atoms (i.e. the Rydberg blockade): the increase of non-linearity of the response of a medium to light.

3.3.1 Electromagnetically induced transparency

$\Rightarrow \boxed{\chi} \Rightarrow$ As light (an electric field \vec{E}) passes through a medium, it is both attenuated (absorbed) and phase-shifted. This optical response can be characterized in terms of the *susceptibility* χ , which is related to the refractive index $n = \sqrt{1 + \chi}$ of a medium. In particular, the dielectric polarization of a medium can be written as $\vec{P} = \epsilon_0 \chi \vec{E}$. The susceptibility is a complex quantity, with its real part accounting for the phase shift and the imaginary the absorption. The susceptibility can be expressed as a power expansion of the electric field $E = |\vec{E}|$ as

$$\chi = \underbrace{\chi^{(1)}}_{\text{linear optical response}} + \underbrace{\chi^{(2)} E}_{\text{non-linear processes}} + \chi^{(3)} E^2 + \dots .$$

Usually, $\chi^{(1)}$ is the dominant contribution over the higher order effects. However, it is interesting to be able to increase the non-linear response of the medium to be able, for example, to change the velocity of propagation of light in a medium



or to create interactions between two or more photons in a medium, inducing, for example, controlled phase shifts between the light pulses that may find applications in quantum information processes.

When the medium is formed by a gas made out of two-level systems, the susceptibility or response to a probe field close to resonance with the transition is dominated by a large and absorptive $\chi^{(1)}$ linear component. However, non-linear effects can be induced by adding a third level and second (control) field as shown in the figure. In particular, we consider a three-level atom with $|g\rangle$ ground, $|e\rangle$ excited and $|r\rangle$ second excited states separated by $\hbar\omega_{eg} = E_e - E_g$ and $\hbar\omega_{re} = E_r - E_e$. We consider now two lasers which drive the $|g\rangle \rightarrow |e\rangle$ (*probe laser*) and $|e\rangle \rightarrow |r\rangle$ (*control laser*) with detunings $\Delta_p = \omega_{eg} - \omega_p$ and $\Delta_c = \omega_{re} - \omega_c$, respectively (note that we will be interested in the response of the medium to the probe light only). Applying what we know from section 1.2 to the two transitions, we can obtain that the Hamiltonian that describes this physics reads:

$$H = \hbar\Delta_p |e\rangle\langle e| + \hbar(\Delta_p + \Delta_c) |r\rangle\langle r| + \hbar\frac{\Omega_p}{2} \left(\underbrace{|g\rangle\langle e|}_{\sigma_{eg}} + \underbrace{|e\rangle\langle g|}_{\sigma_{eg}^\dagger} \right) + \hbar\frac{\Omega_c}{2} \left(\underbrace{|e\rangle\langle r|}_{\sigma_{er}} + \underbrace{|r\rangle\langle e|}_{\sigma_{er}^\dagger} \right).$$

Now we will also consider that the intermediate $|e\rangle$ state has a decay rate γ . Then, as we know, we need to resort to the *master equation* for the description of the system such that the dynamics are given by

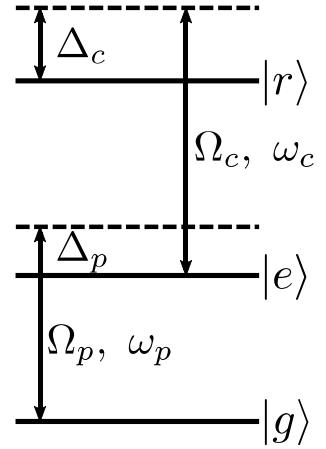
$$\dot{\rho} = -\frac{i}{\hbar} [H, \rho] + \gamma \left(\sigma_{eg}\rho\sigma_{eg}^\dagger - \frac{1}{2} \{ \sigma_{eg}^\dagger\sigma_{eg}, \rho \} \right).$$

Now one can solve this equation and, in particular, find out the *stationary state* of the system (when $\dot{\rho} = 0$). Considering that the polarization of the medium is proportional to the expectation value of the dipole moment of the $|g\rangle \rightarrow |e\rangle$ transition, one can obtain that the χ_{probe} to the probe light is proportional to

$$\Omega_p\chi_{probe} \propto \rho_{eg} = \langle e|\rho|g\rangle .$$

One can solve this easily from the master equation (for example with MATHEMATICA) and find then an expression for χ_{probe} . Since this expression is rather complicated, let us investigate two specific cases:

- i) *Probe-only* ($\Omega_c, \Delta_c = 0$)



Without the control laser, the system reduces to a driven two-level atom. Here, it is simple to calculate that (*)

$$\chi_{\text{probe}}^{2L} \propto \frac{(\Delta_p + i\frac{\gamma}{2})}{\Delta_p^2 + \frac{\gamma^2}{4} + \frac{\Omega_p^2}{2}},$$

a response that, as expected, is largely linear (you can check this by performing a Taylor expansion for small Ω_p and plotting the full result and the first term of the expansion, to see they are extremely similar!) The absorption (imaginary part of the susceptibility), has a Lorentzian shape and has a maximum on resonance, i.e., at $\Delta_p = 0$ (see Figure below, dashed lines).

ii) *Weak probe limit $\Omega_p \ll \Omega_c, \gamma$*

For the full three-level system with a very weak probe laser (for simplicity we put the control laser on resonance, $\Delta_c = 0$), a Taylor expansion for small Ω_p gives a steady-state susceptibility:

$$\chi_{\text{probe}}^{3L} \propto i \overbrace{\frac{1}{\frac{\gamma}{2} + i \left(\Delta_p - \frac{\Omega_c^2}{4(\Delta_p + \Delta_c)} \right)}}^{\chi^{(1)}} + \overbrace{\mathcal{O}(\Omega_p^2)}^{\chi^{(2)\Omega_p}}$$

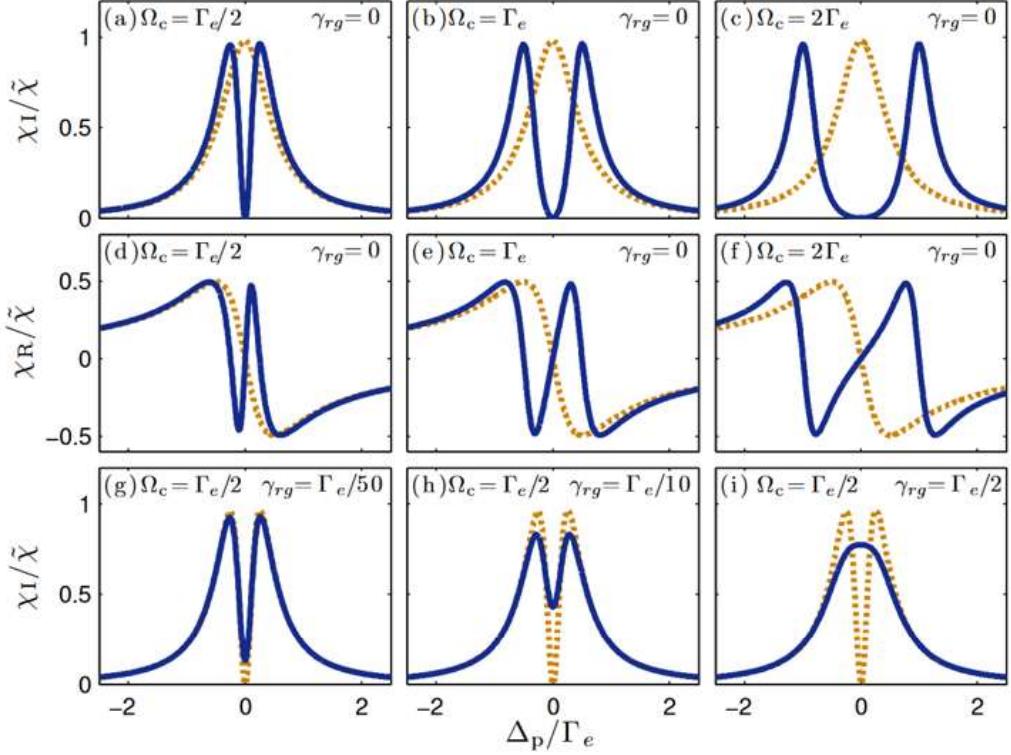
As one can see in the figure (solid lines), this dramatically changes the response of the medium to the light, particularly around resonance, where the absorption (χ_I) of the medium becomes zero, i.e., the medium is completely transparent to the probe light. As Ω_c is further increased the bandwidth of the Electromagnetically Induced Transparency (EIT) also increases.

Moreover, that the group velocity of the light as it passes through the medium is given by

$$v_g = \frac{c}{n(\omega_p) + \omega_p \frac{dn}{d\omega}} .$$

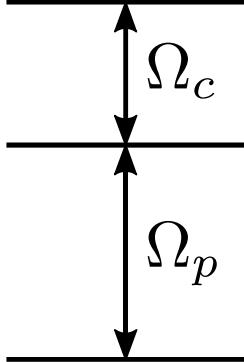
Hence, due to the gradient of the real part of χ changing sign, the group velocity decreases, leading to light being slowed. Finally, the last row of the figure below illustrates how the transparency is destroyed by the addition of decay from the upper level.

Note, however, that even though all of these changes are very interesting, they still only change the linear response of the light, with a very small non-linear contribution.



Alternative view of EIT

To get a better understanding on EIT, it can be useful to think in terms of dressed states. To do so, we diagonalize H when both lasers are on resonance ($\Delta_p = \Delta_c = 0$), where the transparency appears. The eigenstates of the Hamiltonian are given by: (*)



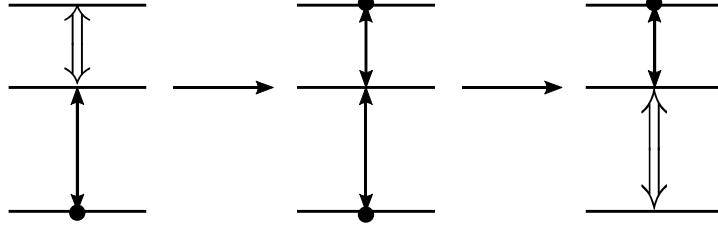
$$|+\rangle = \frac{1}{N_+} \left(\frac{\Omega_c}{\Omega_p} |r\rangle + \frac{\sqrt{\Omega_p^2 + \Omega_c^2}}{\Omega_p} |e\rangle + |g\rangle \right)$$

$$|-\rangle = \frac{1}{N_-} \left(\frac{\Omega_c}{\Omega_p} |r\rangle - \frac{\sqrt{\Omega_p^2 + \Omega_c^2}}{\Omega_p} |e\rangle + |g\rangle \right)$$

$$|D\rangle = \frac{1}{N_0} \left(-\frac{\Omega_p}{\Omega_c} |r\rangle + |g\rangle \right)$$

Here, the only state that emits photons is $|e\rangle$. Hence, $|D\rangle$, which has no overlap with $|e\rangle$ is called a *dark* state. The states $|+\rangle$ and $|-\rangle$ do emit photons, and eventually populate the state $|D\rangle$ more and more, such that in the stationary state only $|D\rangle$ is populated. Since $|D\rangle$ is not coupled to the probe beam, this means that the medium becomes transparent. Note that here,

changing adiabatically from a regime where $\Omega_p \ll \Omega_c$ to one where $\Omega_p \gg \Omega_c$ allows to stay always in the $|D\rangle$ state, which goes from being the $|g\rangle$ state to the $|r\rangle$ state. This method of bringing the population from the ground to an excited state is called STIRAP.



3.3.2 Rydberg EIT

Even though with the EIT one can get a non-negligible amount of non-linearity in the system, it is still very limited. In the following, we will see how to use the Rydberg interactions and, in particular, the blockade effect, to induce even higher non-linearities in the susceptibility and, in turn, use the response of the medium to inform about the interactions between Rydberg states.

To do so, let us consider first two atoms only. Here, the Hamiltonian is very similar to the one we had for one atom, with the difference that the interactions between the two atoms when they are both in the Rydberg state need to be included, and thus

$$H = \frac{\hbar}{2} \sum_{i=1}^2 \left[\Omega_p \left(\sigma_{eg}^{(i)} + \sigma_{eg}^{\dagger(i)} \right) + \Omega_c \left(\sigma_{er}^{(i)} + \sigma_{er}^{\dagger(i)} \right) \right] + V(R) \underbrace{n_r^{(1)}}_{|r_1\rangle\langle r_1|} \cdot \underbrace{n_r^{(2)}}_{|r_2\rangle\langle r_2|},$$

where we have considered $\Delta_p = \Delta_c = 0$ in order to study the transparency on resonance. We may understand the effect of interactions by using again the dressed states. Remember that for N non-interacting atoms the system goes into a product state of all atoms being in the dark state, i.e.,

$$|D\rangle^N = \bigotimes_{i=1}^N |D_i\rangle = \bigotimes_{i=1}^N \frac{1}{N_D} \left(-\frac{\Omega_p}{\Omega_c} |r_i\rangle + |g_i\rangle \right),$$

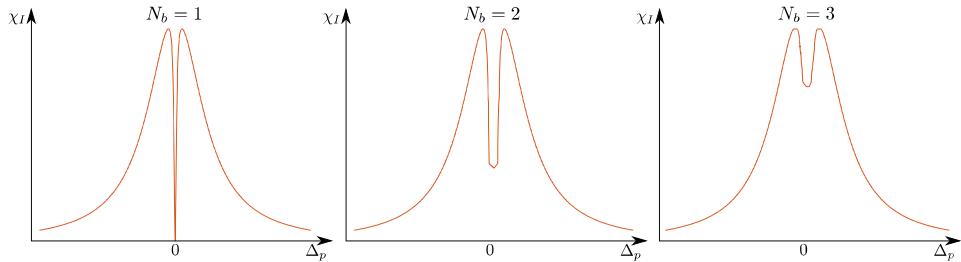
which leads to transparency seen before. As Ω_p increases, more population is transferred to the Rydberg state and the Rydberg atoms start to interact, such that the blockade effect takes place, i.e. there can only be one Rydberg excitation in total in the full gas. This can be understood by considering that the dark state for a blockaded pair of atoms

is

$$|D\rangle^{N=2} = \frac{1}{N_D} \left(\frac{\Omega_p^2}{\Omega_c^2 - \Omega_p^2} |ee\rangle + \frac{\Omega_p \Omega_c}{\Omega_p^2 - \Omega_c^2} (|gr\rangle + |rg\rangle) + |gg\rangle \right).$$

this state scatters

I.e., the state $|ee\rangle$, which scatters on resonance forms now part of the dark state and hence the medium is not transparent anymore. This effect becomes stronger as we increase the number of “blockaded” atoms per blockade sphere N_b , as one can observe solving the master equation and calculating again the susceptibility in the stationary state.



Since this loss of transparency only happens due to the interactions, it is a clear indicator of cooperativity in this system.

Finally, note that the blockade also increases the non-linearity of the medium’s response: As N_b atoms are blockaded, the maximum non-linearity on resonance is enhanced by a factor of N_b , i.e.

$$\chi^{(3)} \propto N_b.$$

For a typical Rydberg experiment, with $n = 60$, a blockade radius of $R_b = 5 \mu\text{m}$ and density $\rho_0 = 10^{12} \text{ cm}^{-3}$, N_b is 1500, which indeed leads to huge non-linearity.

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