Chapter 2: Interacting Rydberg atoms

I. DIPOLE-DIPOLE AND VAN DER WAALS INTERACTIONS

In the previous chapter, we have seen that Rydberg atoms are very sensitive to external electric fields, with their polarizability scaling with the principal quantum number like n^{*7} . This is also the case when the electric field is generated by the charge distribution of the Rydberg electron of another atom, therefore we can expect Rydberg atoms to exhibit very strong interactions. To understand the interactions between Rydberg atoms, we will assume in the following that they are separated far enough so that their electron wave functions do not overlap. As the Rydberg wave functions decay exponentially at large distances, it is possible to express this in terms of a single quantity, the Le Roy radius R_{LR} [1], which is given by

$$R_{LR} = 2\left(\sqrt{\langle n_1, l_1 | r^2 | n_1, l_1 \rangle} + \sqrt{\langle n_2, l_2 | r^2 | n_2, l_2 \rangle}\right),\tag{1}$$

where $|n_i, l_i\rangle$ refers to the electron eigenstate of the *i*th atom. Treating the Rydberg electrons as hydrogenic, we obtain for the expectation value (see [2])

$$\langle r^2 \rangle = \frac{n^2}{2} [5n^2 + 1 - 3l(l+1)].$$
 (2)

For example, the Le Roy radius of two rubidium atoms in the 43S state is $R_{LR} = 532$ nm. In this regime, the interaction potential between two atoms separated by a distance R can be expressed as an Laurent series in R,

$$V(\mathbf{r}_1, \mathbf{r}_2) = \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|} = -\sum_{n=1}^{\infty} \frac{C_n}{R^n}.$$
(3)

The first two terms of the series correspond to the Coulomb and charge-dipole interaction, respectively, and therefore vanish for neutral atoms. The first contribution therefore comes from the dipole-dipole interaction, which is given by

$$V(\mathbf{r}_1, \mathbf{r}_2) = (1 - 3\cos^2 \vartheta_{ij}) \frac{d_i d_j}{R^3},\tag{4}$$

where d_i and d_j are the electric dipole operators and ϑ_{ij} is the angle between the interatomic axis and the quantization axis of the atoms. The higher order terms can be expressed in terms of a series expansion involving spherical harmonics [3],

$$V(\mathbf{r}_1, \mathbf{r}_2) = \sum_{l,L=1} \frac{V_{lL}(\mathbf{r}_1, \mathbf{r}_2)}{R^{l+L+1}}$$
(5)

$$V_{lL}(\mathbf{r}_1, \mathbf{r}_2) = \frac{(-1)^L 4\pi}{\sqrt{(2l+1)(2L+1)}} \sum_m \sqrt{\binom{l+L}{l+m}} \binom{l+L}{L+m} r_1^l r_2^l Y_{lm}(\mathbf{r}_1) Y_{L-m}(\mathbf{r}_2).$$
(6)

In the following we will concentrate on two atoms in the same s state $|r\rangle$, i.e., $n_1 = n_2 \equiv n$. In this case, the main contribution comes from a single combination of p states $|r'r''\rangle = |n'_1, p_1, n'_2, p_2\rangle$. In the case of rubidium, we have $n'_1 = n$ and $n'_2 = n - 1$, as the difference in the quantum defect for s and p states is close to 0.5. The energy difference between the states $|rr\rangle$ and $|r'r''\rangle$ is the Frster defect δ_F . Then, we can write the interaction Hamiltonian in the basis consisting of $|rr\rangle$ and $|r'r''\rangle$ as

$$H = \begin{pmatrix} \delta_F & \frac{d_{r'r'}d_{rr''}}{R^3}\sqrt{D_{\varphi}} \\ \frac{d_{r'r'}d_{rr''}}{R^3}\sqrt{D_{\varphi}} & 0 \end{pmatrix},$$
(7)

where the coefficient $D_{\varphi} = 3$ follows from the angular part of the dipole operators [4]. The eigenvalues of the interaction Hamiltonian are

$$V_{int}(R) = \frac{\delta_F}{2} \pm \frac{1}{2} \sqrt{\delta_F^2 + 4 \frac{(d_1 d_2)^2 D\varphi}{R^6}}.$$
(8)

Let us now consider two regimes of this interaction Hamiltonian, depending on the strength of the interaction $V_{dd} = d_1 d_2 / R^3$ compared to the Frster defect δ_F . At short distances, we have $V_{dd} \gg \delta_F$, therefore the eigenvalues of H are given by

$$V_{int}(R) = \pm \frac{d_{r'r'}d_{rr''}}{R^3} \sqrt{D_{\varphi}}.$$
(9)

As the dipole-dipole interaction is so strong that it mixes the electronic eigenstates, the interaction decays like $1/R^3$ even though the unperturbed eigenstates do not have a finite electric dipole moment. The other regime is where the atoms are so far apart that $V_{dd} \ll \delta_F$. Then we can perform a Taylor expansion of $V_{int}(R)$, obtaining

$$V_{int}(R) = \pm \frac{(d_1 d_2)^2 D_{\varphi}}{\delta_F R^6}.$$
 (10)

This interaction is a van der Waals interaction decaying like $1/R^6$. In the limit $R \to \infty$, the negative eigenvalue connects to the unperturbed state $|rr\rangle$. From this, we can read off the van der Waals coefficient C_6 to be

$$C_{6} = \frac{(d_{1}d_{2})^{2}D_{\varphi}}{\delta_{F}}.$$
(11)

Note that δ_F may be negative for certain combination of states, in this case the van der Waals interaction is repulsive. Consequently there is a crossover from a resonant dipole interaction at short distances to a van der Waals interaction at large distances taking place at a critical radius r_c , which is given by

$$r_c = \sqrt[6]{\frac{4(d_1d_2)^2}{\delta_F^2}}.$$
(12)

In the case of two Rb atoms in the 43s state, the van der Waals coefficient is given by $C_6 = 1.7 \times 10^{19}$. The behavior of the interaction energy depending on the interatomic separation is shown in Fig. 1. For comparison, the van der Waals coefficient for the ground state of Rb is $C_6 = 4707$ [5].

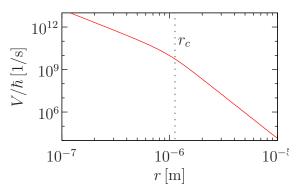


FIG. 1: Dependence of the interaction potential V on the interatomic separation r for the 43s state. The crossover from a dipolar to a van der Waals interaction takes place at r_c .

This huge difference suggests a dramatic scaling of the van der Waals coefficient with the principal quantum number n^* . Each transition dipole moment scales as $d_i \sim n^{*2}$, resulting in a n^{*8} dependence from the dipole matrix elements. However, the Frster defect δ_F has the same scaling as the energy splitting between neighboring Rydberg states, $\delta \sim n^{*-3}$. Overall, this results in a scaling of the van der Waals coefficient like $C_6 \sim n^{*11}$.

Finally, Rydberg atoms in states with nonzero angular momentum quantum number l have a finite quadrupole moment. The quadrupole-quadrupole interaction decays like $1/R^5$, therefore it will become larger than the van der Waals interaction at very large distances. The distance where both interactions are equal can essentially be calculated as [4]

$$\frac{\langle r^2 \rangle^2}{R^5} = \frac{\langle r \rangle^4}{\delta_F R^6}.$$
(13)

The expectation values are comparable, resulting in $R = 1/\delta_F$. For typical Frster defects on the order of a few GHz, this translates to a distance of $R \approx 100 \,\mu\text{m}$, where both interaction are already vanishingly small. Therefore, one can simply neglect the quadrupole-quadrupole interaction in most cases.

II. THE RYDBERG BLOCKADE MECHANISM

Let us now have a look how the presence of interactions modifies the excitation dynamics. As before, we will assume resonant laser excitation between the ground state $|g\rangle$ and a single Rydberg state $|r\rangle$. Furthermore, we will assume that the atoms are separated by more than r_c so that the treatment in terms of a van der Waals interaction is appropriate. Therefore, the system is fully described by the states $|gg\rangle$, $|gr\rangle$, $|rg\rangle$, and $|rr\rangle$. The Hamiltonian in this basis is of the form

$$H = \frac{\Omega}{2} \left(|g\rangle \langle r| \otimes 1 + 1 \otimes |g\rangle \langle r| + \text{H.c.} \right) - \frac{C_6}{R^6} |rr\rangle \langle rr|$$

= $\frac{\Omega}{2} \left(|gg\rangle \langle gr| + |gg\rangle \langle rg| + |gr\rangle \langle rr| + |rg\rangle \langle rr| + \text{H.c.} \right) - \frac{C_6}{R^6} |rr\rangle \langle rr|.$ (14)

We can simplify the problem by noting that the state $|-\rangle = (|gr\rangle - |rg\rangle)/\sqrt{2}$ does not take part in the dynamics, i.e., it is an eigenstate of the Hamiltonian with an eigenvalue of zero. We can therefore introduce an effective three-level system consisting of the states $|gg\rangle$, $|+\rangle = (|gr\rangle + |rg\rangle)/\sqrt{2}$, and $|rr\rangle$. In this new basis, the Hamiltonian becomes

$$H = \frac{\sqrt{2}\Omega}{2} \left(|gg\rangle\langle +| + |+\rangle\langle rr| + \text{H.c.} \right) - \frac{C_6}{R^6} |rr\rangle\langle rr|.$$
(15)

Note the enhancement of the Rabi frequency by a factor of $\sqrt{2}$ in this basis. The resulting dynamics depends on the strength of the interaction compared to the Rabi frequency. In the weakly interacting regime given by $|C_6|/R^6 \ll \Omega$, the system will undergo slightly perturbed Rabi oscillations with Rabi frequency Ω , but the qualitative picture is hardly modified compared to the single atom case. However, in the regime of strong interactions denoted by $|C_6|/R^6 \gg \Omega$, the system behaves differently. The first excitation from $|gg\rangle$ to $|+\rangle$ is unaffected by the interaction. The second excitation from $|+\rangle$ to $|rr\rangle$ is off-resonant because of the strong interaction, see Fig. 2. Effectively, the $|rr\rangle$ state is decoupled from the dynamics, as it can never be reached. This decoupling of the doubly excited state is called the "Rydberg blockade".

We can reduce the description to a two level system consisting only of $|gg\rangle$ and $|+\rangle$, governed by the Hamiltonian

$$H = \frac{\sqrt{2\Omega}}{2} \left(|gg\rangle \langle +| + \text{H.c.} \right).$$
(16)

The dynamics of this Hamiltonian again produces Rabi oscillations, however with two important differences to the non-interacting case. First, the maximum probability to find an

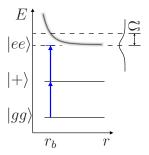


FIG. 2: For two spatially close atoms only one can be excited as the strong interaction shifts the doubly excited state out of resonance.

atom in the Rydberg state, p_r is 1/2, as the $|+\rangle$ state has only one of the two atoms in the Rydberg state. Second, the Rabi frequency is enhanced by a factor of $\sqrt{2}$, resulting in

$$p_r(t) = \frac{1}{2}\sin^2\left(\sqrt{2}\Omega t\right). \tag{17}$$

The distance at which the blockade sets in can be determined by setting the interaction strength equal to the Rabi frequency. This results in a blockade radius r_b given by

$$r_b = \sqrt[6]{\frac{|C_6|}{\Omega}}.$$
(18)

In typical experiments, r_b is on the order of 5–10 μ m.

If we stop the laser excitation process at the time $t = \pi/\sqrt{8\Omega}$, the system will be in the state $|+\rangle = (|gr\rangle + |rg\rangle)/\sqrt{2}$. This state has some very interesting properties. Remarkably, this state cannot be written as a product state of the form

$$|\psi\rangle = |\phi\rangle \otimes |\chi\rangle. \tag{19}$$

This can be proven by noting that for each product state $|\psi\rangle$, there exists a unitary transformation $U = U^{(1)} \otimes U^{(2)}$ that transforms the product state $|gg\rangle$, i.e.,

$$|\psi\rangle = U^{(1)} \otimes U^{(2)} |gg\rangle.$$
⁽²⁰⁾

 $U^{(1)}$ and $U^{(2)}$ are unitary 2 × 2 matrices, and we can carry out the matrix multiplication explicitly using their matrix elements $U^{(i)}_{\alpha\beta}$,

$$U^{(1)} \otimes U^{(2)} |gg\rangle = U^{(1)} \left[U^{(2)}_{gg} |gg\rangle + U_{rg} |gr\rangle \right]$$

= $U^{(1)}_{gg} U^{(2)}_{gg} |gg\rangle + U^{(1)}_{gg} U^{(2)}_{rg} |gr\rangle + U^{(1)}_{rg} U^{(2)}_{gg} |rg\rangle + U^{(1)}_{rg} U^{(2)}_{rg} |rr\rangle.$ (21)

The $|+\rangle$ state does not have any $|gg\rangle$ component, so either $U_{gg}^{(1)}$ or $U_{gg}^{(2)}$ would have to be zero. But any of these choices will also cause the $|gr\rangle$ or $|rg\rangle$ component to vanish, making it impossible to write $|+\rangle$ as a product state. Such quantum states that cannot be written as product states are *entangled* [6]. Entanglement is a type of quantum correlation that is not found in classical system and is a key ingredient for a quantum computer. The state $|+\rangle$ is actually a maximally entangled state because the two atoms are perfectly anticorrelated. This possiblity to create entangled quantum states using the Rydberg blockade has resulted in proposals to use Rydberg atoms as the building blocks of a quantum computer [7], and the first proof-of-principle experiments have already been carried out [8].

As the blockade radius r_b can be much larger than the typical interparticle distance that can be achieved with laser-cooled atoms, it is natural to ask what happens when there are more than two atoms located inside the blockade radius. For N atoms that can be either in the $|g\rangle$ or in the $|r\rangle$ state, the tensor product Hilbert space contains 2^N basis states. This exponential scaling of the complexity of the problem cannot be underestimated: For N = 300, the number of basis states already exceeds the number of atoms in the universe! Nevertheless, we can expand each state according to

$$|\psi\rangle = \sum_{k=1}^{2^{N}} c_{k} |k\rangle \tag{22}$$

$$|k\rangle = \prod_{i=1}^{N} |\alpha_i\rangle_i,\tag{23}$$

where α_i is either g or r. The Hamiltonian for this quantum many-body system can be written as

$$H = \frac{\Omega}{2} \sum_{i} \left(|g\rangle_i \langle r|_i + \text{H.c.} \right) + \sum_{i < j} V_{ij} |r\rangle_i |r\rangle_j \langle r|_i \langle r|_j, \qquad (24)$$

where V_{ij} is the strength of the van der Waals interaction between atoms *i* and *j*. Now, we are interested in the blockaded regime, where all the interaction strengths are much larger than the Rabi frequency, i.e., $V_{ij} \gg \Omega$. Then, the dynamics of the system is restricted two the manifold of zero or one Rydberg excitation. We can simplify the problem further by going to a basis of collective states given by:

$$|G\rangle = \prod_{i=1}^{N} |g\rangle_i = |g_1 g_2 g_3 \dots g_N\rangle$$
(25)

$$|R\rangle = \frac{1}{\sqrt{N}} \sum_{i} |g_1 \dots r_i \dots g_N\rangle.$$
(26)

The $|R\rangle$ state is a symmetric superposition of a single Rydberg excitation. In order to express the Hamiltonian in the new basis, we need to calculate the transition matrix element $\langle G|H|R\rangle$, which is given by

$$\langle G|H|R\rangle = \frac{\Omega}{2} \langle G|\sum_{i=1}^{N} |g_i\rangle \langle r_i|R\rangle = \frac{\Omega}{2} \sum_{i=1}^{N} \frac{1}{\sqrt{N}} = \frac{\sqrt{N}\Omega}{2}.$$
 (27)

In the collective basis, the Hamiltonian is given by

$$H = \frac{\sqrt{N\Omega}}{2} \left(|G\rangle \langle R| + \text{H.c.} \right), \qquad (28)$$

as the matrix elements to all other states in the single excitation manifold are zero. Consequently, we obtain collectively enhanced Rabi oscillation by a factor of \sqrt{N} , leading to a probability to find an atom in the Rydberg state that is given by

$$p_r(t) = \frac{1}{N} \sin^2 \left(\sqrt{N\Omega t}\right).$$
(29)

It is also possible to use these collective quantum states as the computational basis in a quantum computer [9].

III. LIGHT-MATTER INTERACTIONS

So far, we have only looked into the dynamics of the atomic states, ignoring the effects on the light field. However, there are situations where the interplay between Rydberg interactions and coupling between the atoms and the radiation field results in some very interesting consequences. This is particularly true when the light field is so weak that its quantization in terms of single photons becomes relevant.

A. Open quantum systems

To understand the dynamics of an atom coupled to the radiation field, one needs to adopt a description for an open quantum system as atoms in excited states can spontaneously emit photons via an interaction with the vacuum of the radiation field. The most straight-forward way to describe the state of an open quantum system is to use a statistical sum (mixture) of Hilbert space vectors, giving rise to a density matrix of the form

$$\rho = \sum_{i} p_{i} |\psi_{i}\rangle \langle\psi_{i}|, \qquad (30)$$

where $0 \le p_i \le 1$ denotes the probability to find the system in the (pure) quantum state described by the Hilbert space vector $|\psi_i\rangle$. Being probabilities, they are subject to the constraint $\sum_i p_i = 1$, resulting in $\text{Tr}\{\rho\} = 1$ while the non-existence of negative probabilities requires the density matrix to be positive-semidefinite.

As the state of the quantum system is no longer described by a Hilbert space vector, the dynamics is no longer described by the Schrdinger equation. Instead, we have

$$\frac{d}{dt}\rho = \frac{d}{dt}\sum_{i} p_{i}|\psi_{i}\rangle\langle\psi_{i}| = \sum_{i} p_{i}\left[\left(\frac{d}{dt}|\psi_{i}\rangle\right)\langle\psi_{i}| + |\psi_{i}\rangle\left(\frac{d}{dt}\langle\psi_{i}|\right)\right]$$
(31)

$$= -\sum_{i} p_{i} \frac{i}{\hbar} \left(H |\psi_{i}\rangle \langle \psi_{i}| - |\psi_{i}\rangle \langle \psi_{i}| H \right) = -\frac{i}{\hbar} \left[H, \rho \right],$$
(32)

which is called the Liouville-von Neumann equation. Note that this equation is analogous to the Liouville for the classical phase space density, where the commutator is replaced by Poisson brackets.

If we also want to introduce irreversible processes such as spontaneous emission, we have to go beyond the Liouville-von Neumann equation and describe the dynamics in terms of a quantum master equation. In the simplest case, the environment (i.e., the radiation field) is static and does not have a memory of previous irreversible events, such an environment is called to be "Markovian". Then, one can show that the dynamics of the atom coupled to the radiation field is given by a Lindblad master equation

$$\frac{d}{dt}\rho = -i[H,\rho] + \sum_{i=1}^{d^2-1} \gamma_i \left(c_i \rho c_i^{\dagger} - \frac{1}{2} \left\{ c_i^{\dagger} c_i, \rho \right\} \right), \tag{33}$$

where d is the dimension of the Hilbert space of the atom, γ_i are decay rates that describe the frequency of the irreversible events, and the quantum jump operators c_i describe the action of the irreversible event [10]. Note that the jump operators can be non-Hermitian. For a better understanding of the meaning of the Lindblad master equation, it is instructive to look into an alternative formulation in terms of a stochastic process [11]. In this approach, each state $|\psi_i\rangle$ of the statistical ensemble is propagated under the non-Hermitian Hamiltonian

$$H' = H - \frac{i}{2} \sum_{i} \gamma_i c_i^{\dagger} c_i.$$
(34)

For each infinitesimally small timestep τ , the probability of a quantum jump in the *i*th channel is calculated as

$$p_i = \tau \langle \psi_j | c_i^{\dagger} c_i | \psi_j \rangle. \tag{35}$$

If such a quantum jump occurs, the system is brought into the state $|\psi'_j\rangle = c_i |\psi_j\rangle$ and the final result is normalized. Repeating this process over all channels and states in the ensemble then reproduces the Lindblad form of the quantum master equation. Remarkably, this method converges rapidly with the number of states in the ensemble, n. Essentially, the stochastic process performs a Monte-Carlo sampling of the quantum master equation, yielding an error for any observable $\langle O \rangle = \text{Tr}\{O\rho\}$ proportional to $1/\sqrt{n}$. In many cases, this allows the number of samples to be chosen much smaller than the Hilbert space dimension of the problem, d, reducing the computational complexity of simulating the quantum master equation.

IV. RESONANCE FLUORESCENCE

As a first example for an open quantum system, let us consider a single two-level system interacting with the vacuum of the radiation field. Additionally, we will assume that the two-level system consisting of the states $|g\rangle$ and $|e\rangle$ is resonantly driven by an external laser. The Hamiltonian is then given by mation we neglect the fast oscillating term. Then, we can write the laser Hamiltonian as

$$H = \frac{\Omega}{2}(\sigma_+ + \sigma_-), \tag{36}$$

where we have introduced the spin-flip operators $\sigma_+ = |e\rangle\langle g|$ and $\sigma_- = \sigma_+^{\dagger} = |g\rangle\langle e|$. The interaction with the radiation field will lead to spontaneous emission with a rate

$$\gamma = \frac{4}{3}\omega^3 d^2,\tag{37}$$

where ω is the frequency of the transition and d is the dipole matrix element [10]. Spontaneous emission results in the atom changing the state from $|e\rangle$ to $|g\rangle$, so the corresponding quantum jump operator is simply given by σ_{-} . The total quantum master equation then reads

$$\frac{d}{dt}\rho = -i\left[H,\rho\right] + \gamma \left(\sigma_{-}\rho\sigma^{+} - \frac{1}{2}\left\{\sigma_{+}\sigma_{-},\rho\right\}\right).$$
(38)

The interesting aspect about this master equation is that it exhibits a competition between the coherent dynamics generated by the laser and the dissipative dynamics arising from the decay into the vacuum of the radiation field. The solution to this master equation can be found by introducing the Bloch vector $\langle \vec{\sigma} \rangle$ given in terms of the expectation values of the Pauli matrices as [10]

$$\langle \vec{\sigma} \rangle = \begin{pmatrix} \langle \sigma_x \rangle \\ \langle \sigma_y \rangle \\ \langle \sigma_z \rangle \end{pmatrix}.$$
(39)

The equations of motion for the Bloch vector are given by

$$\frac{d}{dt}\langle \vec{\sigma} \rangle = G \langle \vec{\sigma} \rangle + \vec{b},\tag{40}$$

using the matrix G given by

$$G = \begin{pmatrix} -\gamma/2 & 0 & 0\\ 0 & -\gamma/2 & -\Omega\\ 0 & \Omega & -\gamma \end{pmatrix}$$
(41)

and the vector \vec{b} ,

$$\vec{b} = \begin{pmatrix} 0\\0\\-\gamma \end{pmatrix}.$$
(42)

This equation of motion is called the optical Bloch equation. Its stationary state $\langle \vec{\sigma} \rangle_s$ can be found from the condition $d/dt \langle \vec{\sigma} \rangle = 0$ and is given by

$$\langle \sigma_z \rangle_s = -\frac{\gamma^2}{\gamma^2 + 2\Omega^2} \tag{43}$$

$$\langle \sigma_+ \rangle_s = \langle \sigma_- \rangle_s^* = -i \frac{\Omega \gamma}{\gamma^2 + 2\Omega^2}.$$
 (44)

Note that the population of the excited state,

$$p_e = \frac{1}{2} (1 + \langle \sigma_z \rangle_s) = \frac{\Omega^2}{\gamma^2 + 2\Omega^2}$$
(45)

is always less than 1/2 even in the limit of strong driving, i.e., $\Omega \gg \gamma$. Thus, it is not possible to create population inversion in a two level system in the stationary state by coherent driving. The population will merely saturate at $p_e = 1/2$. The off-diagonal elements are related to the linear susceptibility χ , which is defined in terms of the polarization P, according to

$$\langle P \rangle = \frac{\chi E}{4\pi} = \frac{N}{V} d\langle \sigma_+ \rangle, \tag{46}$$

where N/V is the atomic density. Using the relation $\Omega = dE$ and solving for χ , we obtain

$$\chi = 8\pi i \frac{N}{V} d^2 \frac{\gamma}{\gamma^2 + 2\Omega^2}.$$
(47)

As the susceptibility is purely imaginary, the medium formed by the atoms is purely absorptive, with an absorption coefficient that is proportional to the decay rate γ .

V. ELECTROMAGNETICALLY INDUCED TRANSPARENCY

The steady state of the driven two-level atom is not particularly exciting, but this changes drastically when a third level (e.g., a Rydberg state) is introduced. The presence of the third state enables interference effects between different excitation and deexcitation paths and leads to a much richer structure in the susceptibility. In the following, we consider a ladder-type three-level system, consisting of a ground state $|g\rangle$, an intermediate excited state $|e\rangle$, and a Rydberg state $|r\rangle$. Furthermore, we assume laser driving of the transitions between the ground state and the intermediate state and the intermediate state and the Rydberg state. Then, the Hamiltonian is given by

$$H = \begin{pmatrix} 0 & 0 & \frac{\Omega_p}{2} \\ 0 & \delta & \frac{\Omega_c}{2} \\ \frac{\Omega_p}{2} & \frac{\Omega_c}{2} & \Delta \end{pmatrix},$$
(48)

where Δ is the detuning between $|g\rangle$ and $|e\rangle$, and δ is the two-photon detuning between $|g\rangle$ and $|r\rangle$.

On two-photon-resonance, the eigenstates of this Hamiltonian can be expressed by introducing two angles θ and ϕ , which are defined as

$$\tan \theta = \frac{\Omega_p}{\Omega_c} \tag{49}$$

$$\tan 2\phi = \frac{\sqrt{\Omega_p^2 + \Omega_c^2}}{\Delta}.$$
(50)

Then, the eigenstates of the Hamiltonian are given by [12]

$$|a^{+}\rangle = \sin\theta\sin\phi|g\rangle + \cos\phi|e\rangle + \cos\theta\sin\phi|r\rangle$$
(51)

$$|a^{0}\rangle = \cos\theta|g\rangle - \sin\theta|r\rangle \tag{52}$$

$$|a^{-}\rangle = \sin\theta\cos\phi|g\rangle - \sin\phi|e\rangle + \cos\theta\cos\phi|2\rangle.$$
(53)

Remarkably, the state $|a^0\rangle$ does not contain the intermediate state $|e\rangle$. This means that once the system is in the state $|a_0\rangle$, it remains there as spontaneous emission is no longer effective. On the other hand, if we wait long enough, the system will eventually end up in $|a_0\rangle$ as the other two eigenstates of the Hamiltonian can decay via spontaneous emission events. Such a behavior is called "coherent population trapping". Moreover, the once the system is in $|a_0\rangle$, its susceptibility vanishes since we have $\rho_{ge} = \langle a_0 | g \rangle \langle e | a_0 \rangle = 0$. Consequently, $|a_0\rangle$ is called a "dark state" of the dynamics, as the system does not absorb any photons any more. As all incoming photons simply pass through such a medium, this effect is known as "electromagnetically induced transparancy".

Away from the two-photon resonance $(\delta \neq 0)$, we can calculate the susceptibility in the limit of a weak probe laser, i.e., $\Omega_p \ll \Omega_c, \gamma$ [12], obtaining

$$\chi = 4\pi \frac{N}{V} d^2 \left[\frac{4\delta(\Omega_c^2 - 4\delta\Delta)}{|\Omega_c^2 + i2\delta(\gamma + i2\Delta)|^2} + i \frac{8\delta^2\gamma}{|\Omega_c^2 + i2\delta(\gamma + i2\Delta)|^2} \right].$$
(54)

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