Enhancement of Rydberg atom interactions using ac Stark shifts

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The ac Stark effect was used to induce resonant energy transfer between translationally cold Rydberg atoms. The $^{85}$Rb Rydberg atoms were obtained by laser excitation of cold atoms from a magneto-optical trap. Using two-photon microwave spectroscopy of the $49s_{1/2} - 50s_{1/2}$ transition, the field strength of a 28.5 GHz dressing field was calibrated. When this dressing field was set at specific field strengths, the two-atom dipole-dipole process $43d_{5/2} + 43d_{5/2} \rightarrow 45p_{3/2} + 41f$ was dramatically enhanced, due to induced degeneracy of the initial and final states. A series of observed resonant field strengths correspond to different magnetic sublevel possibilities for the initial and final states. These agree well with calculated resonance fields based on a perturbative ac Stark shift formula. This method for enhancing interactions is complementary to dc electric-field-induced resonant energy transfer, but has more flexibility due to the possibility of varying the applied frequency.

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The large transition dipole moments of Rydberg atoms make them much more sensitive to electric fields than less excited atoms. For example, the dc polarizabilities of low-angular momentum Rydberg states scale like $n^7$, where $n$ is the principal quantum number [1]. This high sensitivity can be exploited for various means. For instance, energy transfer between Rydberg atoms may often be tuned into resonance using dc electric fields [2].

Rydberg atoms are also sensitive to small oscillating electric fields. For example, microwave dressing fields may be used to modify the dc polarizabilities of Rydberg states [3]. This could be used to reduce the influence of electric field inhomogeneities on the dephasing of a Rydberg atom qubit.

The present work demonstrates the use of weak microwave dressing fields to tune electric dipole-dipole interactions between Rydberg atoms into resonance, in a case where this cannot be accomplished by dc fields. We exploit the latitude to change both the amplitude and frequency of the dressing fields to create interatomic interactions which are much stronger than could otherwise be achieved.

Our results may be understood in terms of the ac Stark shifts of the relevant states. The ac Stark shift of a state $|\phi\rangle$, in an electric field oscillating at angular frequency $\omega$, with amplitude $\varepsilon_z$ in the $z$ direction, is given by (see, for example, Ref. [4]):

$$\Delta E_\phi = \frac{1}{2} \varepsilon_z^2 \sum_{m \neq n, n = 0} \frac{(E_m - E_n)|\langle \phi | \mu_z | m \rangle|^2}{(E_\phi - E_m)^2 - (\hbar \omega)^2}$$

(1)

where $E_m$ refers to the energy of state $|m\rangle$ and $\mu_z$ is the electric dipole moment in the $z$ direction. The modification of resonant energy transfer between Rydberg atoms by microwave fields has been previously studied in a strong field regime, where the Floquet description was more suitable [2, 3].

The ac Stark shifts of Rydberg states can be probed using microwave spectroscopy. For our purposes, this provides a useful check on the validity of Eq. 1 and allows us to calibrate the applied field strengths when studying interatomic interactions. Our apparatus has been described previously [7, 8]. A standard vapor cell magneto-optical trap (MOT) acts as a source of cold $^{85}$Rb atoms. These are excited to $49s_{1/2}$ Rydberg states using a 1 $\mu$s pulse of laser light. Before excitation, the magnetic and electric fields are reduced to less than 0.02G and 0.05 V/cm respectively. Approximately 3 $\mu$s after excitation, a 28.5 GHz ac dressing field is turned on. While the dressing field is on, a $\approx 33$ GHz probe drives the $49s_{1/2} - 50s_{1/2}$ two-photon transition. The probe pulse lasts 6 $\mu$s. The dressing field is then switched off and a selective field ionization pulse [1] is applied to measure the $49s_{1/2}$ and $50s_{1/2}$ populations. By scanning the probe frequency between laser shots and collecting the resulting spectra, we can measure the difference in the ac Stark shifts of the two states involved in the transition (see Fig. 1). As Eq. 1 indicates, the shifts should scale linearly with applied dressing field power. This is verified in Fig. 1.

As shown in Fig. 1 the linewidth of the transitions increase as they shift. By varying the Rydberg density and probe power, we have determined that this is not due to interatomic interactions or power broadening. The observed broadening is very sensitive to the alignment of the horn that launches the dressing microwaves towards the atoms. By varying the position of this horn, the broadening can be made significantly worse than shown in Fig. 1. Thus, we believe that the broadening with increasing power is due to spatial inhomogeneity of the dressing fields over the cold atom sample (this is complicated, due to the presence of reflections and low-Q resonances within the vacuum chamber). Minimizing this inhomogeneity is a major technical challenge in using dressing fields.

By evaluating the matrix elements in Eq. 1 using the techniques of Zimmermann et al. [4], we can compute the
frequency dependence of the ac Stark shift. In particular, as the denominator of Eq. 1 suggests, the shift direction may be reversed by changing ω. Spectroscopy probes the differential shift between the two levels involved, and this may also be reversed. For example, we have observed that the 49s1/2 − 50s1/2 transition is shifted to higher frequency with a dressing field of 37.45 GHz, and that this shift is also proportional to dressing power. This ability to change the direction of differential energy shifts with the applied frequency is essential to what follows.

It is well-known that dc Stark shifts may be used to enhance the interactions between Rydberg atoms [1, 2]. For example, consider the following resonant energy transfer process in Rb, which may be driven by the dipole-dipole interaction:

\[ nd_{5/2} + nd_{5/2} \rightarrow (n + 2)p_{3/2} + (n - 2)f_{5/2, 7/2} \]  

(2)

For \( n = 44 \) the energy of the final state is approximately 60 MHz higher than that of the initial state (calculated using the spectroscopic data of Ref.'s [10, 11]). However, as an electric field is applied, the 42f states exhibit strong, quadratic Stark shifts to lower energies. This tunes the process into resonance, as shown in Fig. 2. In particular, atoms are excited to \( nd_{5/2} \) Rydberg states and allowed to interact in the presence of a weak dc field. By varying the dc electric field between laser shots, and detecting the \((n+2)p\) population by selective field ionization, resonant energy transfer spectra may be obtained (see Ref. [12] for more details). As Fig. 2 illustrates, at \( n = 44 \) the resonance condition is turned on by a weak electric field, but for \( n = 43 \) it is tuned further out of resonance. In this case the initial state is higher in energy than the final state by \( \approx 10 \) MHz.

Since dc fields cannot tune \( 43d_{5/2} + 43d_{5/2} \rightarrow 45p_{3/2} + 41f_{5/2, 7/2} \) into resonance, we consider using ac fields. In particular, we expect that the flexibility in the choice of \( \omega \), which allows shift directions to be reversed, could be...
beneficial. To illustrate this, the difference in ac Stark shifts between the final and initial states have been computed as a function of frequency using Eq 1. Figure 3 illustrates that over certain frequency ranges the ac field would shift the initial and final states closer into resonance – which could not be achieved with a dc electric field (see Fig. 2).

To experimentally test this idea, we have looked for the resonant energy transfer process: 43d_{5/2} + 43d_{5/2} \rightarrow 45p_{3/2} + 41f_{5/2} (all in m_f = 1/2) as a function of microwave frequency. With no microwave field, the final state is lower in energy than the initial state by 6.6 MHz and 8.3 MHz (for 41f_{5/2} and 41f_{7/2} respectively) [14, 11]. Therefore, frequencies where the shift is positive in this figure (i.e. 28.5 GHz) will push this process closer to resonance.

By equating the experimentally observed shift of these transitions with those based on the prediction of Eq 4, we can determine the factor relating \( \varepsilon_z^2 \) to the applied microwave power. This calibration can be compared with the field calibration obtained from the 49s_{1/2} – 50s_{1/2} line discussed at the beginning of this paper. It is essential that these calibrations are done at the same dressing field frequency, due to frequency dependent losses, reflections and low-Q resonances within the vacuum chamber. These calibrations agree to within 20%. Discrepancies may be due to the distribution of the probe field, which will have a slightly different spatial distribution over the trapped atom cloud at these two different frequencies.

Although the experiment is done with no deliberately applied dc electric fields, they are difficult to avoid, since they vary from day to day. Therefore, the frequencies of each of the 43d_{5/2} – 45p_{3/2} and 43d_{5/2} – 41f_{5/2,7/2} transitions are measured, with no dressing field present. These give the experimental difference in energies between the final and initial states of Eq. 4 for the field conditions of the experiment. We find that this “energy defect”, \( \delta E \approx -7.4 \pm 0.1 \) MHz for the 41f_{5/2} case and \( \approx -9.6 \pm 0.1 \) MHz for the 41f_{7/2} case. These differ from the values of \(-6.0(5)\) MHz and \(-8.3(4)\) MHz obtained from the constants of Ref’s [10] and [11]. Although a small electric field explains some of this discrepancy, the shifts of the 43d_{5/2} – 45p_{3/2} and 43d_{5/2} – 41f_{5/2,7/2} lines are not consistent with the same field. In addition, the field required to explain the shift of the 43d_{5/2} – 45p_{3/2} line exceeds the uncertainty in the electric field zero (\( \pm 0.05 \) V/cm), suggesting that a slight adjustment of the spectroscopic constants may be necessary.

From the experimentally observed energy defects we can calculate the resonance field strengths for the different magnetic sublevel possibilities. The ac Stark shifts
for a state $|\phi\rangle$ may be written as $\Delta E_\phi = k_\phi \varepsilon_z^2$, where $k_\phi$ is computed using Eq. [1] For a process like Eq. [2] ($\langle \phi | + | b \rangle \rightarrow | c \rangle + | d \rangle$) the resonance fields may be computed by rearrangement of: $\delta E + [k_c + k_d - k_a - k_b] \varepsilon_z^2 = 0$. Due to the selection rules for the dipole-dipole interaction, not all final and initial state magnetic sublevel combinations are coupled. In Fig. 4 the vertical lines indicate all calculated resonance fields consistent with $\Delta m_j = 0, \pm 1$ for each atom. Although many of the resonances are unresolved, the general agreement is good, and the highest field resonances are in clear agreement with the calculation.

Some caution is required in applying Eq. [1] to the $| f_{3/2, 2, 1/2} \rangle$ states, due to the small energy separation of the two fine structure components (2.3 MHz). The fine structure splittings of the other relevant states, $| f_{5/2, 1/2} \rangle$ and $| g_{3/2, 5/2} \rangle$, are significantly larger. When the Stark shifts become comparable to the splitting of the two fine structure components, Eq. [1] is not valid. This is entirely analogous with the perturbative calculation of polarizabilities in the dc case. To examine this issue, a Floquet calculation has been implemented (see, for example, Ref. [13]). This calculation shows that for the field strengths at the resonance locations, the perturbative calculation is accurate on the scale of Fig. 4.

The modification of resonant energy transfer between Rydberg atoms due to strong microwave fields has been reported [3, 4]. In this case, the primary observation was that an integer number of microwave photons can either be given up or gained in resonant energy transfer to account for the energy defect between the initial and final states. Thus, the exact frequency of the ac field plays an important role. The microwave power determines the number of “sidebands” present (the number of photons lost or gained in the collision). This can be accurately described using Floquet theory [4] (although the ac Stark effect does play a minor, observable role). In the present work – where the energy level shifts are perturbative – the important tuning parameter is the microwave power. The frequency is not as important – it should be set within a range to give the desired shift the correct sign. However, it should not be too close to any resonance, as this will prevent the microwave fields from being turned on and off adiabatically.

Rydberg atom interactions have recently received considerable attention in the context of quantum information processing with neutral atoms. For example, the dipole-dipole interaction between Rydberg atoms has been proposed as means of allowing clouds of cold atoms to store qubits, using a process known as dipole blockade [14]. Lukin et al. [14] considered using long-range resonant electric dipole interactions. However, initial experiments in Rb have focused on non-resonant van der Waals interactions [15, 16, 17]. Recently, local blockade has been observed in Cs using resonant dipole-dipole interactions between Rydberg atoms [18]. In Rb, several groups have identified Eq. [2] as a strong resonant process [19, 20, 21]. As the experimental results of the present work indicate, this process may be shifted into resonance by either dc or ac electric fields (Fig.’s 2 and 4). This would enhance the blockade effect. Possible advantages of ac fields over dc fields include the capacity to turn interactions on and off very quickly (due to the modulation capabilities of the source), and the ability to induce interactions at arbitrary dc fields.

In summary, perturbative ac fields have been demonstrated to enhance the interactions between Rydberg atoms by making them resonant. The frequency dependence of the ac Stark shift allows this to be accomplished with more versatility than the dc Stark shift.

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