Enhancement of Rydberg Atom Interactions Using ac Stark Shifts

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(Received 23 December 2006; published 17 May 2007)

The ac Stark effect was used to induce resonant energy transfer between translationally cold ^{85}Rb Rydberg atoms. When a 28.5 GHz 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. 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.

DOI: 10.1103/PhysRevLett.98.203005 PACS numbers: 32.80.Rm, 32.30.−r, 34.20.Cf, 39.30.+w

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 ω, with amplitude ε_z in the z direction, is given by (see, for example, Ref. [4]):

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

where E_m refers to the energy of state $|m\rangle$ and μ_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 [5,6].

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]. 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 μs pulse of laser light. Before excitation, the magnetic and electric fields are reduced to less than 0.02 G and 0.05 V/cm, respectively. Approximately 3 μs 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 μs. 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(b).

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, suggesting that it is due to spatial inhomogeneity of the dressing fields over the cold atom sample. 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.* [8], 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 $49s_{1/2} - 50s_{1/2}$ 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

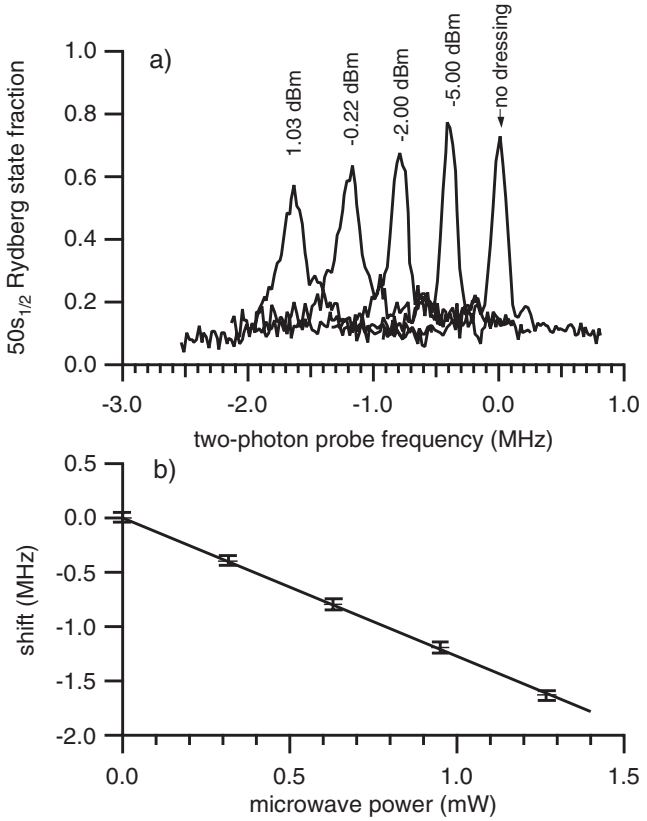

FIG. 1. (a) Observation of the two-photon $49s_{1/2} - 50s_{1/2}$ microwave transition for different powers of a 28.5 GHz dressing field. The probe frequency shown on the horizontal axis is twice the applied frequency, offset by 66.012 926 GHz. The synthesizer power settings are indicated. (b) Shift in the line centers $49s_{1/2} - 50s_{1/2}$ as a function of synthesizer power. Based on a calculated differential shift of $-0.195\ \mathrm{GHz/(V/cm)^2}$ from Eq. (1), we can find the calibration factor relating the synthesizer power to ε_z^2.

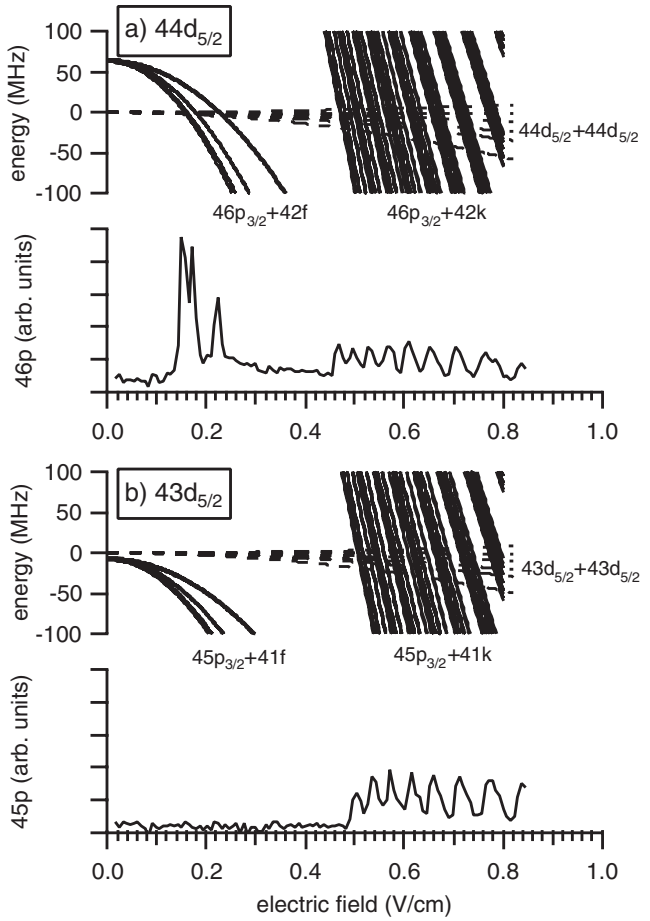

FIG. 2. dc electric-field-induced resonant energy transfer spectra, with atoms initially in the (a) $44d_{5/2}$ and (b) $43d_{5/2}$ states. Also shown are the calculated total energy of several two atoms states, relative to the zero field energy of $nd_{5/2} + nd_{5/2}$, where $n = 44$ and 43. For the initial states $nd_{5/2} + nd_{5/2}$ (dashed lines), we plot all magnetic sublevel possibilities for each of the two atoms involved ($m_j = 1/2, 3/2, 5/2$). In the case of the final states, we consider the different possibilities for the two-atom energies depending on the magnetic sublevel of the $(n + 2)p_{3/2}$ states ($m_j = 1/2,\ 3/2$) and the $(n - 2)f$ and $(n - 2)k$ states ($m_j = 1/2, \ldots, 7/2$). The calculations follow the procedures of Ref. [8] and use the spectroscopic data of Refs. [9,10].

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:

$$nd_{5/2} + nd_{5/2} \rightarrow (n + 2)p_{3/2} + (n - 2)f_{5/2,7/2}. \qquad (2)$$

For $n = 44$ the energy of the final state is approximately 60 MHz higher than that of the initial state (calculated using the spectrosopic data of Refs. [9,10]). 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(a). In particular, atoms are excited to $nd_{5/2}$ Rydberg states and are 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. [11] 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 ω, 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 has been computed as a function of frequency using Eq. (1). Figure 3

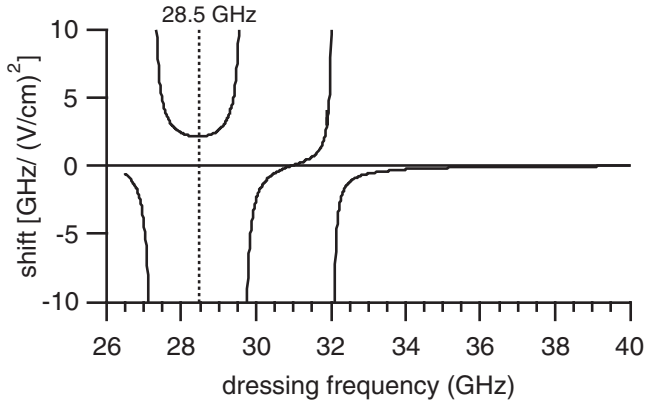

FIG. 3. Calculated difference in the ac Stark shifts of the final and initial states of the process $43d_{5/2} + 43d_{5/2} \rightarrow 45p_{3/2} + 41f_{5/2}$ (all in $m_j = 1/2$) as a function of microwave frequency. With no microwave field, the final state is lower in energy than the initial state by 6.0 MHz and 8.3 MHz (for $41f_{5/2}$ and $41f_{7/2}$, respectively) [9,10]. Therefore, frequencies where the shift is positive in this figure (i.e., 28.5 GHz) will push this process closer to resonance.

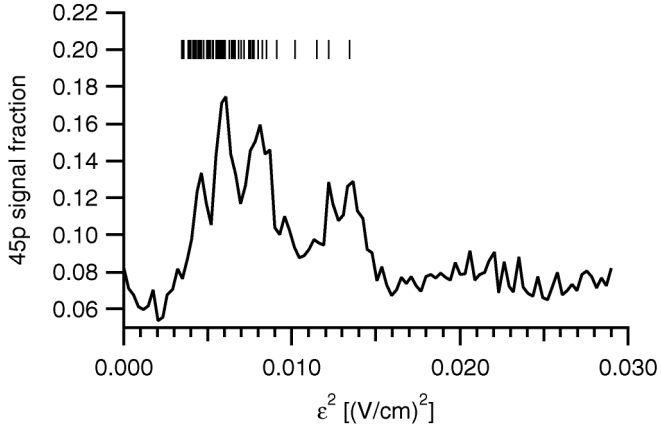

FIG. 4. Observed $45p$ signal as a fraction of total Rydberg signal following a 23 μs waiting period after excitation of $43d_{5/2}$ Rydberg atoms, with a 28.5 GHz field of variable strength present. The field amplitude calibration is discussed in the text. The vertical lines indicate calculated resonance field strengths for the different magnetic sublevel possibilities for the initial and final states of $43d_{5/2} + 43d_{5/2} \rightarrow 45p_{3/2} + 41f_{5/2,7/2}$ (see text for details).

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(b)].

To experimentally test this idea, we have looked for the resonant energy transfer process: $43d_{5/2} + 43d_{5/2} \rightarrow 45p_{3/2} + 41f_{5/2,7/2}$ in the presence of a 28.5 GHz microwave field. The atoms are excited to $43d_{5/2}$ Rydberg states (as discussed above), and the dressing field is turned on 3 μs after excitation. This field is held on for 20 μs until shortly before selective field ionization. This is repeated, scanning the applied microwave power between shots. No deliberate dc electric field is applied. As Fig. 4 indicates, a significant population transfer to the $45p$ state is observed with the microwave field applied. As with the dc case, a series of resonances are observed. This is expected, due to the different magnetic sublevel possibilities for the final and initial states (see below). When the microwave field strength is fixed at a resonance position and the overall Rydberg density of the sample is varied (by changing the excitation laser power), the fraction transferred decreases with reduced density. This confirms that the observed resonances are due to an interatomic process. The transferred fraction grows linearly with density (for small transfer fractions, where depletion of the initial states is not significant). The widths of the resonances also increase with density.

To calibrate the applied fields in Fig. 4, we observe the single-photon transition $43d_{5/2} - 45p_{3/2}$ with a series of dressing field strengths. As with the $49s_{1/2} - 50s_{1/2}$ case, the shifts are observed to be linear in applied microwave power. However, this case is slightly more complicated due to the different magnetic sublevels involved. By equating the experimentally observed shift of these transitions with those based on the prediction of Eq. (1), we can determine the factor relating ε_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 Letter. 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. (2) 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 Refs. [9,10]. 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 (± 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_ϕ is

computed using Eq. (1). For a process like Eq. (2) ($|a\rangle + |b\rangle \rightarrow |c\rangle + |d\rangle$) the resonance fields may be computed by rearrangement of: $\delta E + [k_c + k_d - k_a - k_b]\varepsilon_z^2 = 0$. Because of 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 $41f_{5/2,7/2}$ 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, $45p_{3/2,1/2}$ and $43d_{3/2,5/2}$, are significantly larger. When the Stark shifts become comparable to the splitting of the two fine structure components, Eq. (1) is not valid. To examine this issue, a Floquet calculation has been implemented (see, for example, Ref. [12]). 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 [5,6]. 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 [6] (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 [13]. Lukin *et al.* [13] considered using long-range resonant electric dipole interactions. However, initial experiments in Rb have focused on nonresonant van der Waals interactions [14–16]. Recently, a local blockade has been observed in Cs using resonant dipole-dipole interactions between Rydberg atoms [17]. In Rb, several groups have identified Eq. (2) as a strong resonant process [18]. As the experimental results of the present work indicate, this process may be shifted into resonance by either dc or ac electric fields (Figs. 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. With strong laser fields, this approach could be used to enhance interactions between ground state atoms and/or molecules.

It is a pleasure to acknowledge useful discussions with J. D. Carter. We thank K. Afrousheh for assistance in the early stages of this work. This work was supported by NSERC, CFI, and OIT.

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