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Effects of electric fields on ultracold Rydberg atom interactions

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Abstract

The behaviour of interacting ultracold Rydberg atoms in both constant electric fields and laser fields is important for designing experiments and constructing realistic models of them. In this paper, we briefly review our prior work and present new results on how electric fields affect interacting ultracold Rydberg atoms. Specifically, we address the topics of constant background electric fields on Rydberg atom pair excitation and laser-induced Stark shifts on pair excitation.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

There has been increasing interest in ultracold Rydberg atoms over the last several years. The interest is largely the result of the unique properties of the long-range interactions between Rydberg atoms. At ranges exceeding $10\ \mu\text{m}$, the potential energies associated with Rydberg atom interactions can be greater than the kinetic energy of the atoms in an ultracold gas ($T \leq 1\ \text{mK}$). Rydberg atom interactions can also be anisotropic and can be tuned with electric fields and state selection of the atom's principal quantum number, n [1]. All of these properties have interesting consequences. One important consequence, central to this field, is that Rydberg atom interactions can be used to create a single collective atomic excitation in a macroscopic volume on the order of microns in a gas [2]. This phenomenon is referred to as the 'Rydberg atom dipole blockade' and is important because it provides a pathway to quantum mechanically entangle clusters of atoms, a task that has proven extremely difficult to accomplish by any means [3]. Because of this collection of extraordinary features, ultracold Rydberg gases are a valuable system for investigating a host of interesting topics such as quantum dipolar gases [4], fundamental experiments on quantum mechanics [5], the

development of quantum devices like quantum gates [6] and single photon sources [7], many-body phenomena [8], and exotic forms of matter like long-range Rydberg molecules [9]. All of these research directions have been vigorously pursued in recent years. However, the study of ultracold Rydberg gases is still quite new, the first experiments having taken place a little over 10 years ago [10], and despite the intense recent activity many of the promising directions have yet to be fully explored. As a result, one can anticipate not only continuing successes in current research areas but also the emergence of new research directions that use ultracold Rydberg atoms.

A starting point for understanding many of the research topics that use ultracold Rydberg atoms is the interaction between Rydberg atom pairs. A key element for understanding Rydberg atom pair interactions, which we address here, is how they are affected by electric fields. In general, although not always, electric fields are more important than magnetic fields for Rydberg atom experiments [1]. Rydberg atoms are particularly sensitive to electric fields. Since a Rydberg electron ranges far from its nucleus, the atom is highly polarizable and relatively small electric fields, on the order of $10\text{--}100\ \text{mV cm}^{-1}$, can lead to significant perturbations. In

fact, the Rydberg atom polarizability scales with the principal quantum number as n^7 , so the effect of an electric field can vary widely depending on the Rydberg state. Because stray electric fields are present in all experiments, laser fields are used to excite and probe Rydberg atoms, electric fields are used to detect Rydberg atoms via pulsed field ionization (PFI) and laser fields are used to trap atoms which are subsequently excited to Rydberg states, perturbation due to electric fields is often significant in experiments and cannot be easily eliminated. Electric fields, though, are not always a burden for cold Rydberg atom experiments. Electric fields can also be useful, since, for example, they can be used to manipulate the interactions between Rydberg atoms. It is therefore important to understand the effects of electric fields so that at minimum, the experimental complications can be managed and at best experiments can be optimized.

In this paper, we describe some of the most important effects electric fields have on ultracold Rydberg atoms and how ultracold Rydberg atoms interact. In the next section, we explain some of the ways that constant background electric fields can affect interactions between pairs of Rydberg atoms [11–14]. In the following section, we address how laser fields, through the ac-Stark effect, can influence the excitation of pairs of Rydberg atoms [15]. Finally, we summarize and discuss the relationships between these effects. We will confine our discussion to alkali atoms.

2. The effect of constant electric fields on Rydberg atom pair interactions

Rydberg atom pair interactions can be measured so that experiments can be directly compared to calculations. Figure 1 shows two spectra taken by scanning a narrow bandwidth, ~ 1 MHz, laser to excite ultracold Cs Rydberg atoms to measure the number of ions created after PFI. Figure 1 is an example of experimentally observed spectra of an ultracold Rydberg gas. The large peaks in the spectra are due to the creation of cold Cs 66D Rydberg atoms. The more interesting features, from the perspective of Rydberg atom interactions, are the small ones that are labelled with the asymptotic Rydberg atom pair states, 65D + 67D for example, with which they are associated [14, 16]. The spectral features labelled 65D + 67D are the result of non-resonant two-photon absorption of the laser light by Cs $6P_{3/2}$ atom pairs. The label indicates that one atom is in the 65D Rydberg state asymptotically and the other atom is in the 67D state asymptotically. The absorption of two photons by two $6P_{3/2}$ atoms in a Cs magneto-optic trap (MOT) produces a pair molecular state consisting of two excited interacting Cs Rydberg atoms. The same types of features have been observed in Rb ultracold Rydberg gases [14, 17]. The second qualitative observation about the spectral features associated with the 65D + 67D pair excitation is to note that the energy shifts with the background electric field. The energy shifts are large on the scale of the kinetic energy and spectral bandwidth of the laser. These observations are direct experimental evidence of interacting Rydberg atom pairs and can be used as benchmark data to test calculations of Rydberg atom pair

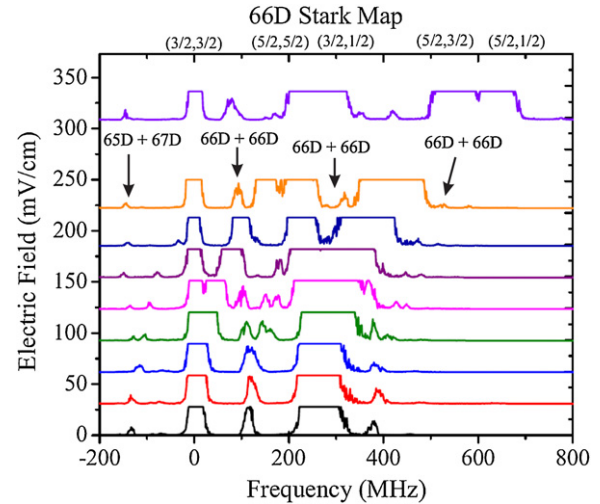


Figure 1. The spectra of a Cs ultracold Rydberg gas as a function of an applied constant electric field around the 66D Rydberg state. A laser operating at ~ 508 nm is scanned and Rydberg atoms are detected via pulsed field ionization. The signal is proportional to the number of Rydberg atoms or Rydberg atom pairs excited.

interactions. In this section, we will address this topic and explain how to calculate the Rydberg atom pair interactions and the energy shifts of those interactions in the presence of a constant or slowly varying background electric field. Our goal is to give new researchers in this field a picture of Rydberg atom pair interactions so that they can understand them both quantitatively and qualitatively.

Cold Rydberg atom pair interactions are typically dominated by near zone, non-radiative, Coulombic interactions. Coulombic interactions are due to the quantum mechanical fluctuations of the charge distributions of the two interacting atoms which cause separation dependent, R , energy shifts of the atomic energy levels. In an overly simplified but useful picture of the interactions, the charge fluctuations of one atom produce an electric field at the location of the second atom which causes an energy shift of the second atom's quantum state. Simultaneously, the charge fluctuations of the second atom produce an electric field at the position of the first atom that leads to energy shifts of atom 1. Since atomic Rydberg states have large polarizabilities, they are very sensitive to these electric fields and we expect and observe strong interactions at large internuclear separations, typically larger than the atomic kinetic energy at $R \sim 10 \mu\text{m}$. The Rydberg atom interactions lead to forces between two Rydberg atoms that can be described by a potential that depends on R and the orientation of two atoms relative to some fixed axis. The atoms can be attracted or repelled from each other depending on the relative phases of the charge fluctuations and strength of their couplings to different states. The charge fluctuations can lead to resonant exchange of energy if they can resonantly excite the other atom. Resonant energy exchange typically leads to strong interactions at large R . Resonant energy exchange, however, is not necessary for Rydberg atom interactions to be strong because of the large density of Rydberg states which leads to many atomic states being relatively near to resonance. In addition to being strong at

large separations, the Rydberg atom interactions can also be different depending on the orientation of the two atoms because the charge fluctuations produced are anisotropic, depending on the atomic states involved. The important fluctuations of the charge distributions almost always occur at frequencies such that the atoms are close enough to each other so the electric field does not have to propagate from one atom to the other as electromagnetic radiation. This latter point is what is meant by the words ‘near zone.’

Constant background electric fields can affect the charge fluctuations by shifting the atomic energy levels and mixing together different electric field free atomic states. The energy shifts and atomic state mixing change the spectrum of the charge fluctuations and how they couple different states. An electric field can also polarize a sample of Rydberg atoms inducing large dipole moments in the atoms. This causes the atoms to behave as permanent dipoles. Accordingly, there is an additional anisotropic dipole–dipole interaction between Rydberg atom pairs that is induced by an electric field of sufficient strength to polarize the Rydberg atoms.

The qualitative discussion presented in the last several paragraphs can be quantified for the calculation of Rydberg atom interaction potentials. To describe our calculations of Rydberg atom pair interactions, we start by taking the perspective that the interactions perturb the two Rydberg atoms. The reason we take this approach is that many of the applications of Rydberg atoms focus on the atoms themselves and not the molecular pair states. The interactions that lead to dipole blockade, for example, have largely been viewed as an energy shifting of two, or more, excitation states out of resonance with a laser, in a volume determined by the dipole blockade radius which is determined by the laser bandwidth, atomic linewidth and interaction strength [2]. An equally valid alternative approach to using atomic basis states is to use a molecular basis. Since the choice of basis is one of convenience, we will not discuss using a molecular basis here. As a consequence of our choice of basis, the asymptotic energies of the atomic Rydberg states are used to label each adiabatic pair state. If an electric field is present, the pair state is labelled by the state with the dominant electronic character at infinite separation. One advantage of this scheme is that we avoid correlating molecular state labels with asymptotic atomic pairs. The pair energies at infinite separation are just the sums of each of the atomic energies, which are well known from quantum defect measurements and include atomic fine structure. Using the energies determined by experimental measurements of the quantum defects for the separated atoms classifies the calculation as semi-empirical. The basis we have introduced here is a j_1 – j_2 coupling scheme, as described by Herzberg [18], where j_i is the sum of orbital and spin angular momentum of the electron for atom i . Each individual atom maintains strong LS coupling and the molecular states conserve the projection quantum number along the internuclear axis, $m_{j1} + m_{j2} = M$. The M quantum number is usually labelled Ω in molecular physics.

Because the Rydberg atom pair interactions are governed by charge fluctuations, it is natural to use a multipole expansion in R^{-1} to calculate the moments of the charge fluctuations

to obtain the interaction potentials [19]. This is a standard approach to the problem of interacting atoms at internuclear separations where exchange interactions are negligible. When two Rydberg atoms are separated by distances of several μm or greater, as in typical cold trapped Rydberg gases, dipole and quadrupole interactions dominate. Neglect of higher multipoles than the quadrupoles can be justified using the scaled van der Waals coefficients [20] or by, for example, calculating the octopole interactions and verifying that they can be neglected [12]. Calculating the octopole interactions and verifying that they are insignificant allows one to safely neglect higher order multipole interactions. As an example, even at the lowest R considered in the calculation of pair states around $89\text{D} + 89\text{D}$ for Cs, the energy shifts due to octopole interactions were on the order of 10^{-4} cm^{-1} , which is 1% of the combined dipole and quadrupole contributions at $R = 3 \mu\text{m}$ [12]. The octopolar interactions quickly dropped to below 10^{-6} cm^{-1} at $R = 6 \mu\text{m}$ for this particular case. Similar results, with distances scaled to the changing principal quantum number, were obtained for Rb Rydberg atom pair calculations for $n \sim 37$ [11]. There has not yet appeared any experimental evidence that octopolar interactions are important. In contrast, some experimental evidence exists that quadrupolar interactions are important for collisions that occur between Rydberg atoms [11]. Of course, using a multipolar expansion also helps us to decrease the number of states involved in a calculation due to the multipolar transition selection rules. However, the multipolar selection rules for field-free Rydberg states can be badly broken in a background electric field.

The interactions between the Rydberg atoms can span the entire range of resonant to non-resonant cases depending on the atomic energy levels involved [12]. Purely non-degenerate interactions yield a potential with an R^{-6} dependence while purely degenerate interactions yield an R^{-3} dependence. For most Rydberg states, because there is a high density of states, both degenerate and non-degenerate interactions are important. This is a complication for Rydberg atom interactions since generally there are many near-degenerate atomic energy levels that can exchange energy. Sums of many near-degenerate interactions can actually dominate or significantly change a true resonant interaction. Additionally, states which are degenerate at large R may not be degenerate at shorter ranges where several multi-polar interactions may play a role and the atomic states have been mixed by the interactions.

One important advantage to recognize when approaching the problem of calculating Rydberg atom pair interactions is that the transition strength between atomic Rydberg states due to multipolar interaction decreases as the difference between principal quantum numbers, Δn , of the two states increases [1, 21]. The decrease in transition strength with Δn can be understood qualitatively by recalling that for large n the oscillator strength of a transition due to dipolar interaction from state $|nlm\rangle$ to $|n'l'm'\rangle$ averaged over all l, l', m , and m' for large n decreases as the energy separation of the two states to the third power:

$$F_{n'n} \propto \left(\frac{Ry}{E'_n - E_n} \right)^3 \frac{1}{n^6} = \frac{n^3}{8(n' - n)^3}. \quad (1)$$

Coupled with the fact that the Rydberg atom energies can be calculated very accurately so that asymptotic near resonances for pair states can be identified before a calculation is carried out and the difficulty of treating degeneracies perturbatively, the decrease of transition strength with Δn points towards using matrix diagonalization to calculate Rydberg atom interaction potentials. This approach suggests expanding the basis states around some energetic region and truncating it when a desired level of convergence is reached.

A perturbative approach to the calculation of the Rydberg atom interaction potentials for a pair of atoms in R^{-1} will give van der Waals coefficients [20]. However, because of the large density of Rydberg atom energy states, this approach is not accurate for most cases of interacting Rydberg atoms, particularly in the presence of electric fields. Near-resonant interactions become non-negligible and lead to avoided crossings at large R which are important to calculate accurately to make quantitative comparisons to experiment. We have calculated Rydberg atom pair interactions by diagonalization of the Hamiltonian in a truncated basis. The size of the basis is determined by the convergence of the calculated pair interaction potentials [12]. Matrix diagonalization can accurately account for the off-resonant, near-resonant and resonant interactions at the same time. For cases where an electric field is present and assumed to lie along the internuclear axis, the calculation is simplified because M is a good quantum number [12]. Convergence on the level of 10^{-4} cm^{-1} is still only acquired by large pair basis sizes of ~ 6000 close-lying states for $M = m_{j1} + m_{j2} = 0$. For higher M , slightly smaller basis sizes can be used, for example ~ 4000 basis states at $M = 5$ for Cs pairs around $89D + 89D$ [12]. The calculations are computationally intensive but can be easily parallelized in a straightforward way for a cluster architecture. In the case of a background electric field strong enough to polarize the atoms or if the interactions are significantly anisotropic, larger basis set sizes are required and the calculations become more technically challenging. Angular dependence for these calculations is addressed in more detail at the end of this section. The range of R over which this overall method is valid is determined by the near zone, or long wavelength, approximation and the R where exchange and Coulomb repulsion of the two alkali Rydberg electrons become significant. When the large matrix diagonalization method is required, as opposed to perturbation theory, is a more complicated question. The answer depends on how accurately the potentials must be calculated to answer a specific experimental question. For collisions and the description of exotic bound states, a perturbative approach is unlikely to yield sufficient accuracy as these phenomena involve dynamics at short range where many curves interact with each other. On the other hand, a careful application of perturbation theory may work at long range if accuracy at the several MHz level can be tolerated. If perturbation theory is applied, it is important to sum over the interactions with all of the most important states. This procedure can be as difficult as simply doing the matrix diagonalization. Particularly problematic are quadrupolar interactions between near degenerate states at long range and large background

electric fields which mix and shift the atomic states. However, in some simple cases such as S states used in many dipole blockade experiments, where the actual extent of the blockade volume is not important, perturbation theory can likely be applied in a straightforward manner.

Neglecting retardation effects gives an upper bound on R set by the reduced wavelength $R_{\text{max}} = \bar{\lambda} = \lambda/2\pi$ [22], where λ is the transition wavelength between two states of the Rydberg atoms. There are many states that contribute to a particular interatomic potential so there are many different transitions, and consequently λ , that play a role in an interaction potential calculation. The validity of the long wavelength approximation needs to be checked for each set of coupled Rydberg states used in a calculation. However, the long wavelength approximation turns out to be an accurate assumption for almost all Rydberg atom states. For Rb, a transition from the continuum to $18S$ is approximately 500 cm^{-1} . This corresponds to a wavelength of $20 \mu\text{m}$, or a reduced wavelength of $3.2 \mu\text{m}$. Such a transition, because of the large implicit change in n , will most likely not make a significant contribution to any Rydberg atom pair potentials. For a transition between $n = 20$ and $n = 18$, the reduced wavelength is $12 \mu\text{m}$. These principal quantum numbers, $n = 18$ and $n = 20$, are rather small for most recent ultracold Rydberg atom experiments. At $n = 50$, more typical of current experiments, transition frequencies are $\sim 5 \text{ cm}^{-1}$ between neighbouring states giving a reduced wavelength of 2 mm ! Transitions between adjacent Rydberg states for $n > 50$ have even smaller energy differences due to the n^{-3} dependence of the energy separations. These extreme examples of transitions give a measure of how accurate the near-zone approximation works. It is useful to keep in mind that this approximation is most likely to break down at small n . For $n > 40$, it is unlikely that this approximation will break down for realistic experimental conditions for states and associated transitions that make significant contributions to the interatomic potentials.

A conservative estimate of the lower bound for R is the LeRoy radius [23], where $R_{\text{LR}} = 2(\sqrt{\langle\alpha|r^2|\alpha\rangle} + \sqrt{\langle\beta|r^2|\beta\rangle})$. In this expression, $|\alpha\rangle$ and $|\beta\rangle$ represent sets of atomic quantum numbers. The LeRoy radius is typically a crude estimate of where exchange interactions become non-negligible. For $89D$ pairs we calculated $R_{\text{LR}} \sim 2.5 \mu\text{m}$ [12]. Since the average distance between atoms in a cold Rydberg gas at typical experimental number densities of $\sim 10^6\text{--}10^{10} \text{ cm}^{-3}$ is $\sim 3\text{--}25 \mu\text{m}$, the relevant range of R for most experiments meets the LeRoy criterion. A better estimate of the inner R where the calculations are valid is obtained by calculating the direct and exchange integrals for the Coulomb interaction between the two Rydberg electrons. Because the electrons are in diffuse orbitals and the multipolar interactions are large, this reduces the internuclear separations where the calculations are useful. We are currently in the process of including the electron repulsion in our calculations, but this work is preliminary.

Figures 2, 3 and 4 show potential energy curves calculated for Cs and Rb at background electric fields of 0, 0.5 and 2 V cm^{-1} , respectively, around the $37D + 37D$ asymptote for

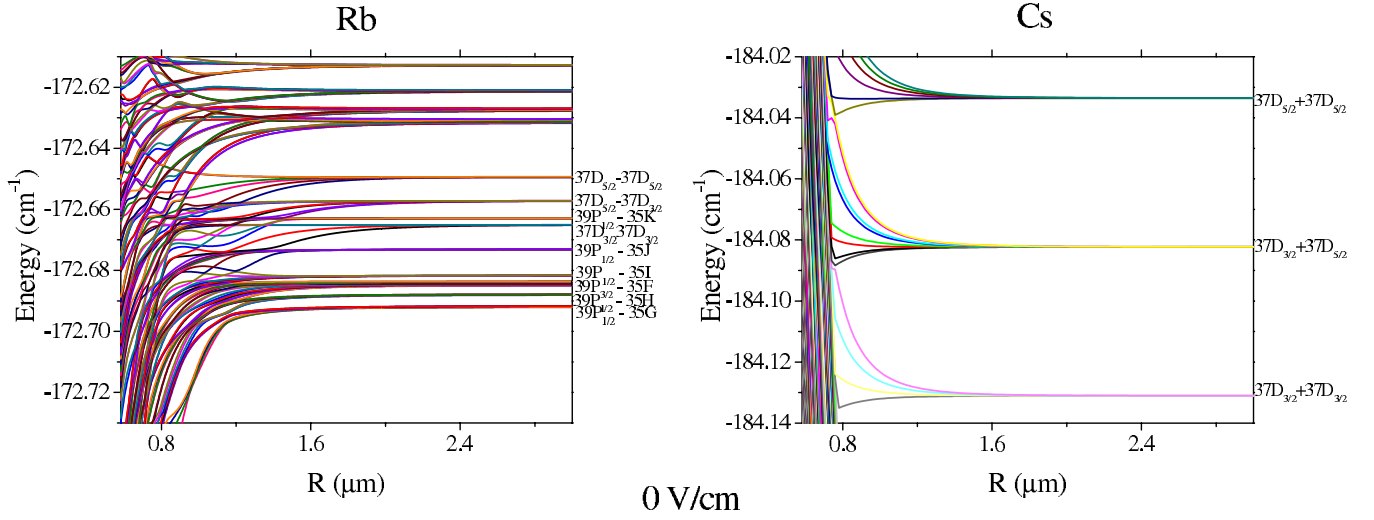


Figure 2. Rydberg atom interaction potentials calculated as described in the text for Cs and Rb Rydberg atom pairs around the $37D + 37D$ asymptote. The calculation is done for the case of no background electric field.

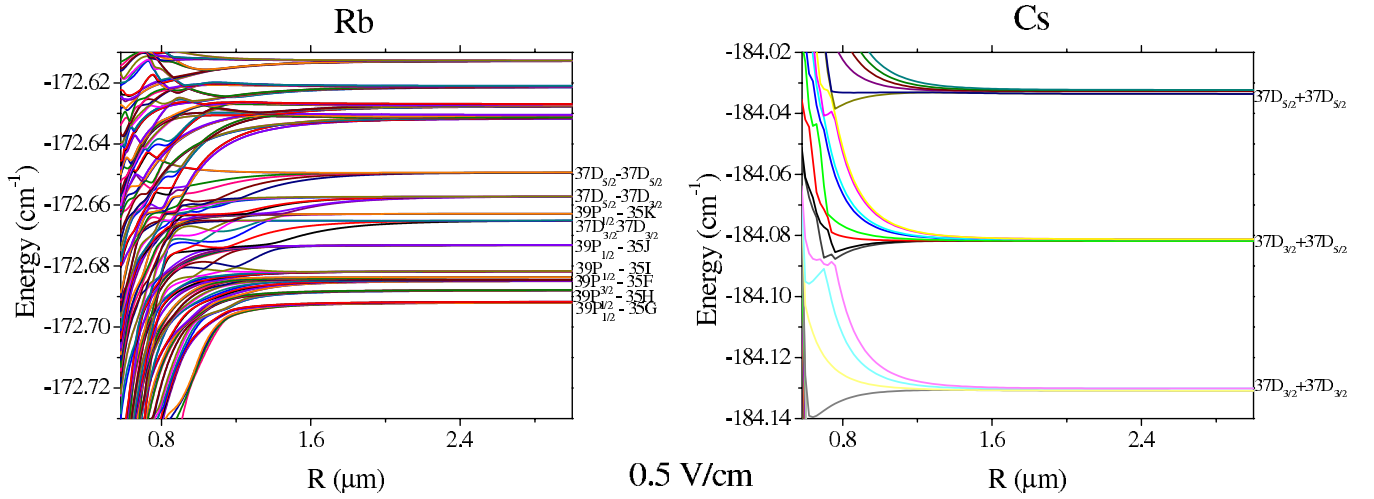


Figure 3. Rydberg atom interaction potentials calculated as described in the text for Cs and Rb Rydberg atom pairs around the $37D + 37D$ asymptote. The calculation is done for the case of an electric field of 500 mV cm⁻¹.

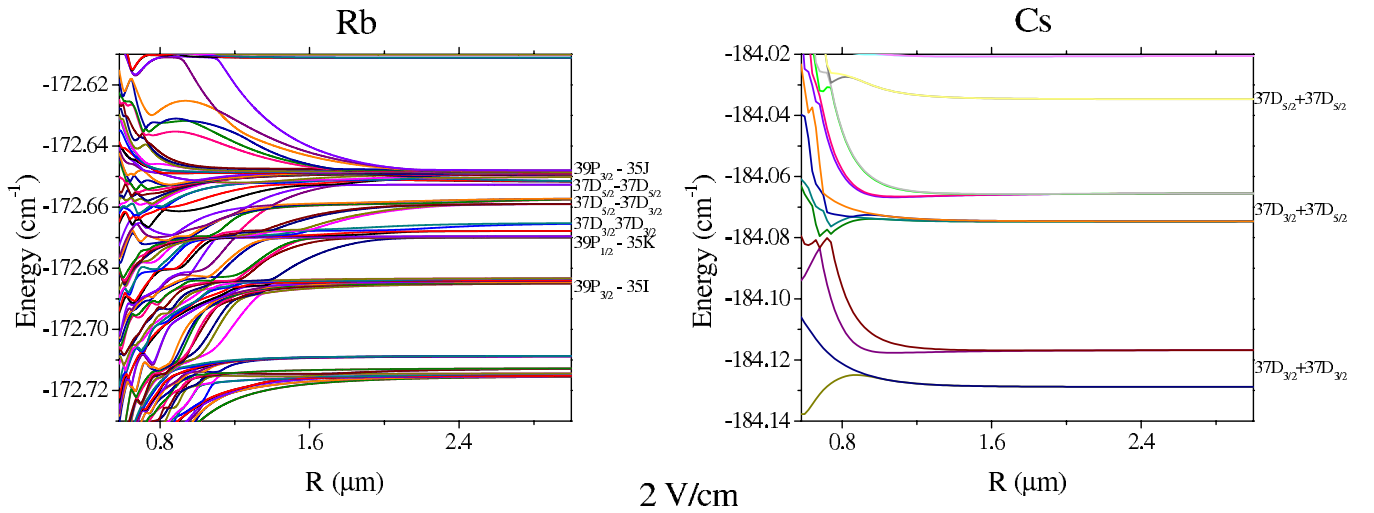


Figure 4. Rydberg atom interaction potentials calculated as described in the text for Cs and Rb Rydberg atom pairs around the $37D + 37D$ asymptote. The calculation is done for the case of an electric field of 2 V cm⁻¹.

$M = 0$ over a range of R where the calculation is valid. We have labelled the states by the asymptotic atomic pair states to which each potential correlates, as described previously. The potential energy curves (eigenvalues) are shown as a function of R . The interaction potentials are calculated in three steps: (1) calculation of Stark-shifted single-atom energies and basis states; (2) selection of an appropriate truncated pair basis; and (3) diagonalization of the Hamiltonian in the Stark-shifted pair basis. The matrix that must be diagonalized can be written in a Stark-shifted pair basis as [12]

$$\langle \tilde{\alpha} | \langle \tilde{\beta} | H(R) | \tilde{\alpha}' \rangle | \tilde{\beta}' \rangle = \delta_{\alpha\alpha'} \delta_{\beta\beta'} E_{\alpha\beta} + \langle \tilde{\alpha} | \langle \tilde{\beta} | H_{DD}(R) + H_{DQ}(R) + H_{QQ}(R) | \tilde{\alpha}' \rangle | \tilde{\beta}' \rangle, \quad (2)$$

where $|\tilde{\alpha}\rangle$ and $|\tilde{\beta}\rangle$ denote sets of atomic quantum numbers and the tilde denotes Stark-shifted states for these calculations. R is taken to be oriented along the quantization axis z , which is also the direction along which the electric field points. The term $E_{\alpha\beta} = E_{\alpha} + E_{\beta}$ denotes the asymptotic $\rightarrow \infty$ Stark-shifted energies including atomic fine structure. $H_{DD}(R)$, $H_{DQ}(R)$ and $H_{QQ}(R)$ correspond to the dipole–dipole, dipole–quadrupole and quadrupole–quadrupole interactions, respectively. It is also possible to calculate the new eigenstates after matrix diagonalization. The eigenstates can be written as

$$|\Psi\rangle(R) = \sum C_{\alpha\beta}(R) |\tilde{\alpha}\rangle |\tilde{\beta}\rangle, \quad (3)$$

where $C_{\alpha\beta}$ is the expansion coefficient in the Stark-shifted pair basis. The eigenstates provide insight into how the interactions mix the asymptotic atomic states. One advantage of calculating the Stark-shifted atomic basis first is that it is easier to separate the effect of the background electric field from that of the interactions. The choice of the electric field lying along the internuclear axis primarily neglects the effect of the static dipolar interaction induced by the electric field. We have estimated this effect to be $\sim 10^{-6}$ of the dipole and quadrupole interactions over the range of internuclear separations shown in the figures, consistent with our prior work [12]. The static dipolar interaction at these electric fields for these states is completely negligible compared to the accuracy of the potential curves and other interactions. However, at larger electric fields, there will be significant angular dependence as we describe later in this section.

Figures 2, 3 and 4 show that the long-range interactions between Rydberg atoms are tunable using electric fields. As the electric field is changed over a relatively small range of values, 0–2 V cm⁻¹, the potential curves change dramatically, particularly for Rb. The potential curves also show that the potentials are atomic species dependent. The differences are mainly due to differences in the quantum defects and fine structure. The fact that high angular momentum states combined with the 39P states of Rb result in near energy degeneracies with the 37D + 37D states of Rb shows how complex Rydberg atom pair interactions can be. The Rb curves, in contrast to Cs, have many avoided crossings at long range and significantly more perturbation. The figures show that a particular species of alkali atom may actually have advantages over another one for an experiment. For example, the lower Cs states at 2 V cm⁻¹ would be better for a dipole blockade experiment than those of Rb at any

electric field and Cs at smaller electric fields. Note that all the potential curves within a narrow bandwidth at the lowest energy asymptote are repulsive for Cs at 2 V cm⁻¹ and these curves are separated by large frequency intervals compared to typical laser bandwidths used for blockade experiments from the other asymptotes. At 2 V cm⁻¹, the blockade distance will have some angular dependence, the blockade volume will not be a sphere, but the angular effect from the polarization of the atoms by the external electric field will be small compared to the ~ 200 MHz height of the lowest barrier. This latter statement can be justified by taking the result found in the section on angular dependence of the potentials and scaling it for $n = 37$. Of course this is only one example, but it illustrates the points that electric fields have a large impact on the shape of Rydberg atom interaction potentials, the interaction potentials at the same quantum numbers for different alkali species are not the same, in fact the interactions may not even be similar, and electric fields can be useful for manipulating the Rydberg atom interactions.

Another interesting and important feature that is shown in figures 2, 3 and 4 is the splitting of asymptotically degenerate Rydberg atom pair states. There are two effects that lead to the splitting and avoided crossings between states that are not dipole coupled at zero electric field. The background electric field causes an R^{-3} resonant coupling between the asymptotically degenerate states and the quadrupole–quadrupole interaction creates an R^{-5} resonant coupling between such states. The first effect is due to the mixing by the Stark effect while the latter effect is a result of the non-vanishing quadrupole or higher moments of the interacting states. Because these interactions tend to mix states with different m_j they tend to suppress the angular dependence of the multipolar interactions when the electric fields are not strong enough to polarize the Rydberg atoms.

To probe these potentials, we have carried out several experiments [11, 14–16]. These experiments have investigated collision dynamics and molecule formation. We have been able to measure the shape of several features of the potentials and test these against our calculations. In [14], Cs Rydberg atom time-of-flight imaging was used to obtain kinetic energy release data on two dissociating atoms after they were excited to a stationary but repulsive region of a potential curve corresponding to Cs 88D + 90D. Remarkably, the data agreed within experimental error. In [16], bound states of Cs Rydberg atom molecules were measured using a form of Coulomb explosion imaging. Again, the data were in excellent agreement with the calculations. Finally, we were also able to quantitatively explain results on Rb Rydberg atom collisions using our calculated Rb potential energy curves [11]. Figure 5 shows measured rate constants for the yield of Rb n P atoms after the excitation of Rb n D + n D states and a calculation based on a Landau–Zener model of those same quantities. Given the complexity of the interatomic potentials and numbers of avoided crossings, explained fully in [11], the agreement is further evidence that our Rydberg potential energy calculations are accurate.

The angular dependence of the Rydberg atom interaction potentials is more difficult to calculate, but is especially

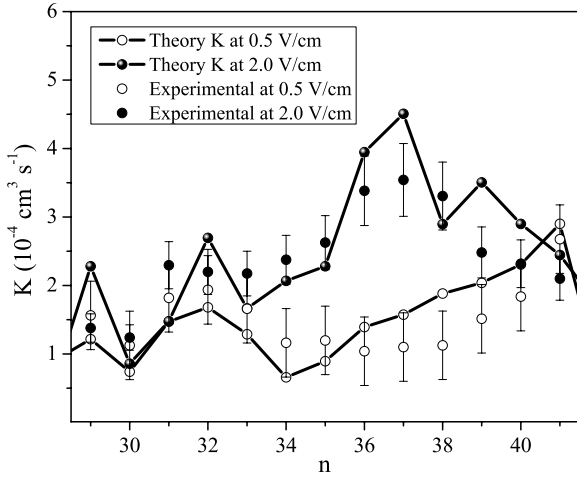


Figure 5. Theoretical and experimental rate constant, K , for the production of Rb $(n+2)P$ Rydberg atoms after the excitation of $nD+nD$ Rydberg atom pairs for 0.5 V cm⁻¹ and 2.0 V cm⁻¹ as a function of n for a PFI delay time of 100 ns.

important in large electric fields when the Rydberg atoms become polarized. The angular dependence of the potentials can also be important in small or zero background electric fields in sensitive experiments that probe interactions on the scale of MHz. Many experiments are not sensitive to angular dependence because they average over all impact parameters of the colliding or interacting Rydberg atoms. Particularly nice results in confined geometries have shown that the angular dependence of Rydberg interactions can be observed [24]. More experiments will no doubt be aimed at measuring the angular dependence of Rydberg atom interactions in the future, so we discuss the calculation of angularly dependent potentials here.

The multipolar interaction Hamiltonian can be written as a series of powers of R^{-1} :

$$H(r_1, \theta_1, \phi_1; r_2, \theta_2, \phi_2; R) \\ \propto \sum_{l_1=1}^{\infty} \sum_{l_2=1}^{\infty} \sum_{m=-l_1}^{+l_1} C_{l_1, l_2, m} \frac{r_1^{l_1} r_2^{l_2}}{R^{l_1+l_2+1}} Y_{l_1}^m(\theta_1, \phi_1) Y_{l_2}^{-m}(\theta_2, \phi_2), \quad (4)$$

where (r, θ, ϕ) are the electronic coordinates for each atom, and R is the internuclear distance. Terms with $l = 1$ are dipolar in nature and terms with $l = 2$ are quadrupolar in nature. For example, the term with $l_1 = 1, l_2 = 2$ would be a part of the dipole–quadrupole interaction. In the case where the electric field is parallel to the internuclear axis, the atomic wavefunctions are written in terms of the same coordinates (r, θ, ϕ) as the Hamiltonian. In this Hamiltonian, θ is measured with respect to the internuclear axis which defines the z -axis. The calculation of the Hamiltonian matrix elements requires calculation of the radial matrix elements and integrals of products of three spherical harmonics. The latter integrals simplify to products of Clebsch–Gordan coefficients:

$$\int_0^\pi \int_0^{2\pi} \sin(\vartheta) Y_{n_1}^{m_1}(\vartheta, \varphi) Y_{n_2}^{m_2}(\vartheta, \varphi) Y_{n_3}^{m_3}(\vartheta, \varphi)^* d\varphi d\vartheta \\ = \sqrt{\frac{(2n_1+1)(2n_2+1)}{4\pi(2n_3+1)}} \langle n_1 n_2 0 0 | n_1 n_2 n_3 0 \rangle \\ \times \langle n_1 n_2 m_1 m_2 | n_1 n_2 n_3 m_3 \rangle. \quad (5)$$

In the case where the electric field is at angle Θ to the internuclear axis, atomic wavefunctions are written in terms of the coordinates (r, θ', ϕ') . These coordinates are measured in a coordinate system with the z -axis along the electric field. The primed coordinate system is thus rotated with respect to the one used in the Hamiltonian, which has its z -axis along the internuclear axis. The rotation that has to be applied to the Hamiltonian to make easy integration possible is one that ensures that the Hamiltonian is also written in terms of the primed coordinates. The unprimed angles appear in the Hamiltonian only as arguments of spherical harmonics. The rotation of spherical harmonics from the unprimed to the primed system is given simply by the identity

$$Y_l^m(\theta, \phi) = \sum_{m'} D_{m'm}^l(\Theta, \Phi, \Xi) Y_l^{m'}(\theta', \phi'), \quad (6)$$

where $D_{m'm}^l(\Theta, \Phi, \Xi)$ are the Wigner rotation matrix elements, expressed in the basis of spherical harmonics. The $D_{m'm}^l(\Theta, \Phi, \Xi)$ depend on the Euler angles; here, Θ should be $\angle(\mathbf{E}, \mathbf{R})$, the angle between the electric field and the internuclear axis of the atomic pair. The expression for the rotation of the spherical harmonic is used to simply replace the spherical harmonics in the Hamiltonian by sums over other spherical harmonics that now depend on the primed coordinates. Note that the rotation conserves l , but mixes m . Thus, dipolar interactions stay dipolar, and quadrupolar interactions stay quadrupolar. However, the resulting rotated Hamiltonian is not block-diagonal with respect to M in the Stark-shifted atomic pair basis as it was if the electric field lies along the internuclear axis. This makes the calculation more difficult because it requires a dramatic increase in the number of basis states. Even on a cluster supercomputer, the Hamiltonian matrix has to be spread out on several nodes. To make matters worse, these calculations have to be carried out at a number of angles sufficient to sample the potential accurately. Figure 6 shows a calculation of the dipolar potential for the 90D + 90D state of Cs at a background electric field of 100 mV cm⁻¹. The upper portion of the figure shows the full potential while the lower part of the figure shows the potential due to the polarization of the atoms for a several R . This latter figure is obtained by subtracting out the multipolar van der Waals potential at the magic angle where the static dipole–dipole interaction is zero. The figure shows that electric fields can polarize a sample of Rydberg atoms so that the static dipole–dipole interaction is as strong or stronger than the van der Waals interactions. This can lead to many interesting phenomena such as the appearance of new phases in a quantum gas or lattice of Rydberg atoms [4].

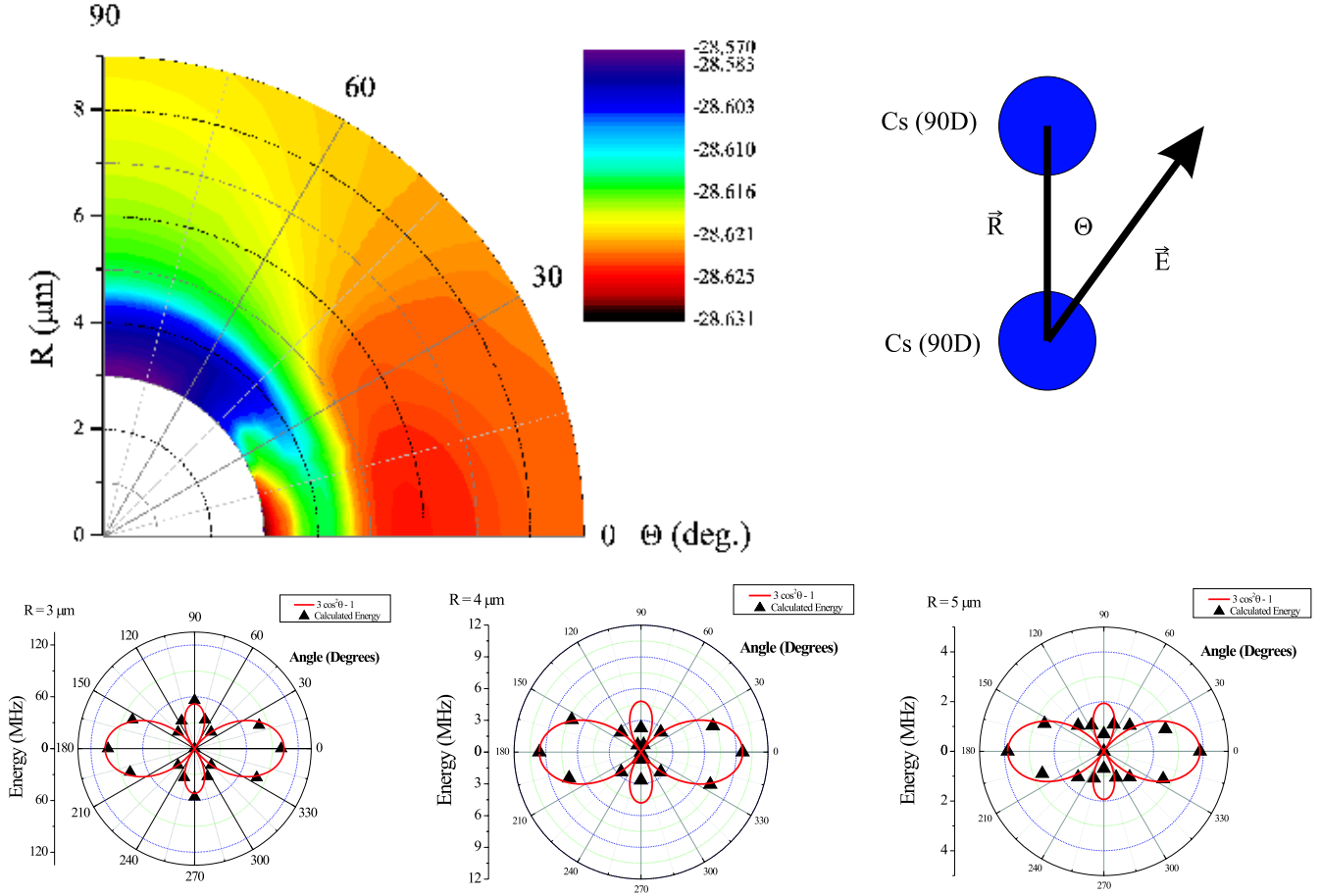


Figure 6. (Top) an angularly dependent Cs Rydberg atom pair potential in an electric field of 100 mV cm^{-1} around the $90\text{D} + 90\text{D}$ state. The potential is shown as a function of R and Θ . The colour map shows the energy in cm^{-1} with the zero of energy placed at two times the ionization limit of the pair state, where both Rydberg electrons are ionized. (Bottom) the static dipole–dipole interaction taken by subtracting out the van der Waals contribution at the angle where the dipole–dipole interaction vanishes for $90\text{D}_{3/2} + 90\text{D}_{3/2}$ for $M = 0$ for three different R . Perturbations of the potentials due to avoided crossings are seen as deviations from the dipolar interaction.

3. The ac Stark effect and excitation of Rydberg atom pairs

Due to the large Rydberg atom polarizability, it was expected that the laser field used to excite Rydberg atoms would play a role in ultracold Rydberg atom experiments. Ates and co-workers [25] were the first to point out theoretically that the Autler–Townes effect, due to the Rydberg excitation laser, can lead to an anti-blockade effect. Using a microwave field after Rb $n\text{D}$ state excitation, Bohlouli-Zanjani and co-workers were able to induce resonant population transfer between Rb Rydberg states in an ultracold Rydberg gas [26]. The first demonstration that the ac Stark effect, due to the Rydberg excitation laser, can affect the preparation of excited Rydberg atom pairs was done by Nascimento and co-workers [15]. In this experiment, population transfer into the $n\text{P}_{3/2}$ state after the excitation of $n\text{S}_{1/2}$ Rb Rydberg atoms was observed. The experimental results showed that the $n\text{P}_{3/2}$ state population was quadratically dependent on the $n\text{S}_{1/2}$ atomic density, indicating a two-body process. The excitation scheme that was used to excite each Rydberg atom of a pair was $5\text{S}_{1/2} \rightarrow 5\text{P}_{3/2} \rightarrow nl$, where nl is the target Rydberg state. Clearly, such a result would naturally be attributed to

binary collisions involving attractive potentials. In this case, however, the interaction between Rb $n\text{S}_{1/2}$ atoms is repulsive according to perturbation theory in the absence of an electric field [20] and the diagonalization method described in the previous section for the background electric fields present in the experiments [12]. The $n\text{P}_{3/2}$ Rydberg atom production is actually due to direct excitation of $n\text{P}_{3/2} + (n - 1)\text{P}_{3/2}$ Rydberg atom pairs. Although this two-photon transition is asymptotically forbidden, it can occur as a result of the ac Stark shifts induced in the intermediate $5\text{P}_{3/2} + 32\text{S}_{1/2}$ state and the dipole–dipole interactions between the pair of Rydberg atoms in the excited state. In this section, we will explain how the ac Stark effect of the excitation laser and the dipole–dipole interactions can affect the preparation of excited Rydberg atoms by further explaining this interesting experimental result.

We start by considering the Rydberg atom pair potentials around the $n\text{S}_{1/2} + n\text{S}_{1/2}$ asymptote for $n \sim 32$ and the R dependent eigenstates of the interacting pairs of atoms. Specifically, we consider the coefficients $C_{\alpha\beta}$ obtained by diagonalization of the Hamiltonian in the Stark-shifted pair basis, as described in the last section of this paper [12]. In figure 7(a), we show the $32\tilde{\text{S}} + 32\tilde{\text{S}}$ pair potentials as well

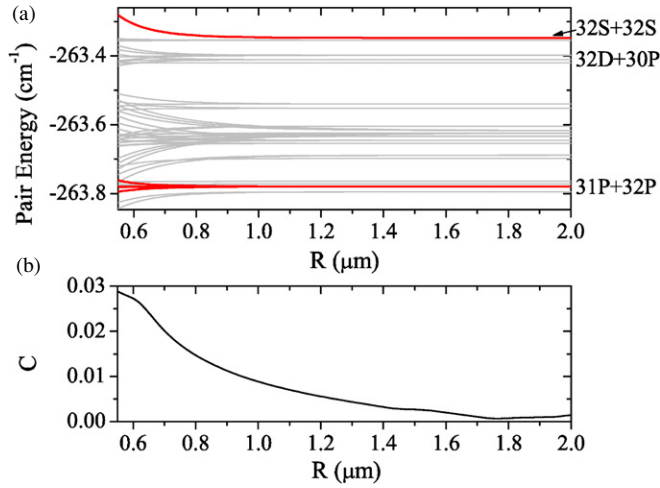


Figure 7. (a) Rubidium $32\tilde{S}_{1/2} + 32\tilde{S}_{1/2}$ and surrounding states for $M = 1$ and a background $\epsilon_b = 500 \text{ mV cm}^{-1}$. The $32\tilde{P} + 31\tilde{P}$ pair state is energetically below $32\tilde{S}_{1/2} + 32\tilde{S}_{1/2}$ for all relevant R . (b) $C(R) = |\langle 32\tilde{S}_{1/2} + 32\tilde{S}_{1/2} | 32\tilde{P}_{3/2} + 31\tilde{P}_{3/2} \rangle|$. The relevant states for the model are highlighted in red (online).

as surrounding states for $M = 1$ in a background electric field of 500 mV cm^{-1} for Rb. The $32\tilde{P} + 31\tilde{P}$ pair state is energetically below the $32\tilde{S} + 32\tilde{S}$ pair state for all relevant R . The projection of the $32\tilde{P} + 31\tilde{P}$ onto the $32\tilde{S} + 32\tilde{S}$ pair is given by $C(R) = |\langle 32\tilde{S}_{1/2} + 32\tilde{S}_{1/2} | 32\tilde{P}_{3/2} + 31\tilde{P}_{3/2} \rangle|$, and is shown in figure 7(b). Since $C(R)$ is nonzero, the dipole transition to the $32\tilde{P} + 31\tilde{P}$ state is not forbidden. The pair potential calculation indicates that $C(R)$ is primarily due to a strong dipole–dipole interaction. $C(R)$ does not depend strongly on the dc electrical field in the range $0\text{--}5 \text{ V cm}^{-1}$. In this case, the oscillator strength for the transition, then, is provided by the small amount of dipole-allowed configuration that is mixed into the state by the Rydberg atom interactions. We have checked the weak dependence on the dc electric field experimentally for Rb by investigating how a range of dc fields affects the observed population transfer. However, the state mixing due to dipole–dipole interaction alone cannot explain the observations as the $32\tilde{P} + 31\tilde{P}$ state is still far detuned from the laser frequencies used in the Rydberg atom pair excitation scheme. The explanation of the observation must also include the excitation dynamics, namely the ac Stark shifts, or Autler–Townes effect, of the laser field.

The excitation process including the ac Stark effect can be modelled using a density matrix approach. Our theoretical model considers an effective three-level system, which consists of the pair states $5P_{3/2} + 5P_{3/2}$ (state 1), $5P_{3/2} + 32\tilde{S}_{1/2}$ (state 2), and the final $32\tilde{P}_{3/2} + 31\tilde{P}_{3/2}$ pair state (state 3) [27]. In general, the ac Stark effect can lead to much more complicated dynamics involving many more states than 3. However, this system is a highly simplified one that can be investigated to isolate the ac Stark effect. We calculated the excitation probabilities of states around these ones and found them to be two orders of magnitude smaller. These calculations verified that the three-state system described captures the essential physics. The density matrix equations for these three pair states must be solved numerically. The density matrix

equations, after applying the rotating wave approximation, are

$$\begin{aligned}
 \rho'_{33}(t) &= -\gamma_3 \rho_{33}(t) + i\Omega_2(t)(\tilde{\rho}_{32}(t) - \tilde{\rho}_{23}(t)) \\
 \rho'_{22}(t) &= -\gamma_2 \rho_{22}(t) - i\Omega_2(t)(\tilde{\rho}_{32}(t) - \tilde{\rho}_{23}(t)) \\
 &\quad + i\Omega_1(t)(\tilde{\rho}_{21}(t) - \tilde{\rho}_{12}(t)) \\
 \rho'_{11}(t) &= -\gamma_1 \rho_{11}(t) - i\Omega_1(t)(\tilde{\rho}_{21}(t) - \tilde{\rho}_{12}(t)) \\
 \tilde{\rho}'_{32}(t) &= -(\gamma_{31} + i\Delta_2)\tilde{\rho}_{32}(t) + i\Omega_2(t)(\rho_{33}(t) - \rho_{22}(t)) \\
 &\quad + i\Omega_1(t)\tilde{\rho}_{31}(t) \\
 \tilde{\rho}'_{31}(t) &= -(\gamma_{32} + i(\Delta_2 + \Delta_1))\tilde{\rho}_{31}(t) \\
 &\quad + i\Omega_1(t)\tilde{\rho}_{32}(t) - i\Omega_2(t)\tilde{\rho}_{21}(t) \\
 \tilde{\rho}'_{21}(t) &= -(\gamma_{21} + i\Delta_1)\tilde{\rho}_{21}(t) + i\Omega_1(t)(\rho_{22}(t) - \rho_{11}(t)) \\
 &\quad - i\Omega_2(t)\tilde{\rho}_{31}(t)
 \end{aligned} \tag{7}$$

where $\Omega_1(t) = \mu_1 \text{Re} E(t)/2\hbar$, μ_1 is the dipole moment of the $5P \rightarrow 32S$ transition, $\Omega_2(t) = \mu_2 \text{Re} E(t)/2\hbar$, $\mu_2 = C(R)\mu_1$; $\gamma_2 = 2\pi \times 6.06 \text{ MHz}$, $\gamma_{31} = 1/\tau_{32S} + 1/\tau_{31P}$ and τ_{32S} and τ_{31P} are the lifetimes of $32S$ and $31P$ states, respectively. $\gamma_{32} = \gamma_2/2$, $\Delta_1 = 0$ since the laser is on resonance for the $5P \rightarrow 32S$ transition, $\Delta_2 = (E_{32\tilde{P}+31\tilde{P}} - E_{32\tilde{S}+32\tilde{S}})/\hbar$, and $E(t)$ is the electric field due to the pulsed laser. The prime denotes differentiation with respect to time. $E(t)$ is an input parameter in our calculation. To model the non-transform limited pulse, we have used a multi-mode pulse, which is given by

$$\begin{aligned}
 E(t) &= \sqrt{\frac{2I_0}{\epsilon_0 c}} \Delta_t G(t, \Delta_t) \\
 &\quad \times \sum_{n=-(N-1)/2}^{(N-1)/2} \sqrt{\frac{1}{K}} G(n\delta\nu, \Delta_\nu) e^{-i(2\pi n\delta\nu t + \phi_n)},
 \end{aligned} \tag{8}$$

where the mode spacing is $\delta\nu = 240 \text{ MHz}$, $N = 17$ modes, and $\Delta_\nu = 1.5 \text{ GHz}$ is the Gaussian width of the power spectrum. G denotes a normalized Gaussian of specified spectral FWHM, meaning that it integrates over the full frequency range to unity. The resulting intensity profile in time, when averaged over many pulses, is then a Gaussian with $\text{FWHM}_t = \Delta_t = 4 \text{ ns}$. The peak intensity is $I_0 = 780 \text{ MW cm}^{-2}$. K is the normalization factor for the spectral density:

$$K = \sum_{n=-(N-1)/2}^{(N-1)/2} G(n\delta\nu, \Delta_\nu). \tag{9}$$

ϕ_n are random phases for each mode. In figure 8(a), we show a typical pulse used for the calculation. The ac Stark effect as well as other dynamical effects are taken into account automatically by the effective three-level system [27]. The calculation reveals that the ac Stark effect splits the intermediate level $5P_{3/2} + 32\tilde{S}_{1/2}$ as seen by the second photon into multiple resonances or quasi-states [28], as shown in figure 8(b). The maximum frequency shift is $\sim \pm 15 \text{ GHz}$ and corresponds to the maximum Rabi frequency of the laser pulse. The splitting into multiple resonances, instead of just two as expected for continuous wave radiation, is due to the finite duration of the laser pulse. The ac Stark effect shifts the second photon into resonance with the $5P_{3/2} + 32\tilde{S}_{1/2} \rightarrow 32\tilde{P}_{3/2} + 31\tilde{P}_{3/2}$ transition and coupled with the state mixing allows direct excitation of the $32\tilde{P}_{3/2} + 31\tilde{P}_{3/2}$ Rydberg pair state.

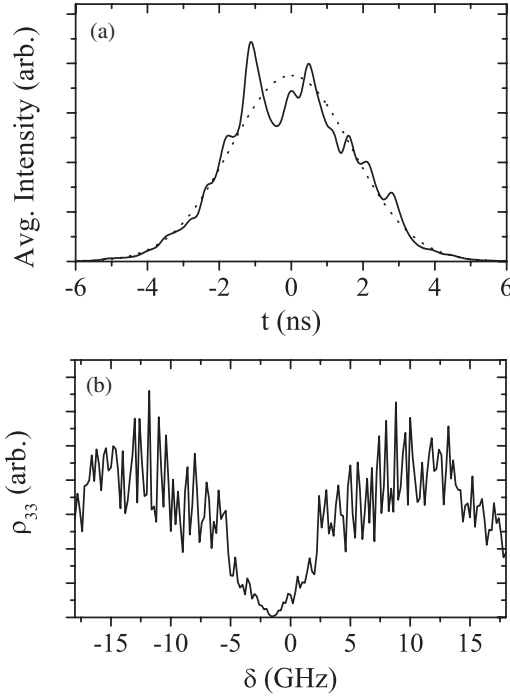


Figure 8. (a) Typical laser pulse used in the calculation. (b) Splitting of the intermediate level $5P_{3/2} + 32S_{1/2}$ due to ac Stark effect.

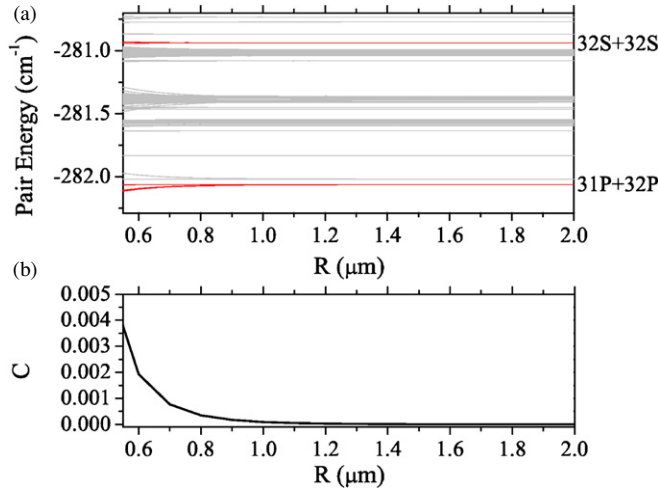


Figure 9. (a) Cesium $32\tilde{S}_{1/2} + 32\tilde{S}_{1/2}$ and surrounding states for $M=1$ and a background $\epsilon_k = 500$ mV cm⁻¹. The $32\tilde{P} + 31\tilde{P}$ pair state is energetically below $32\tilde{S}_{1/2} + 32\tilde{S}_{1/2}$ for all relevant R . (b) $C(R) = |\langle 32\tilde{S}_{1/2} + 32\tilde{S}_{1/2} | 32\tilde{P}_{3/2} + 31\tilde{P}_{3/2} \rangle|$. The relevant states for the model are highlighted in red (online).

In figure 9, we show, for the case of Cs, the $32\tilde{S} + 32\tilde{S}$ and surrounding states, figure 9(a), as well as the $C(R) = |\langle 32\tilde{S}_{1/2} + 32\tilde{S}_{1/2} | 32\tilde{P}_{3/2} + 31\tilde{P}_{3/2} \rangle|$, figure 9(b). For Cs, the energy splitting, ΔE , between the $32\tilde{P} + 31\tilde{P}$ state and the $32\tilde{S} + 32\tilde{S}$ state is about three times larger than for the same Rb state, resulting in a $C(R)$ about ten times smaller. This difference between Rb and Cs is consistent with a scaling law used in our previous work [15], where $C(R) \propto n^7 \propto 1/\Delta E^{7/3}$. Therefore, we expect the $32P_{3/2}$ signal to be ten times smaller

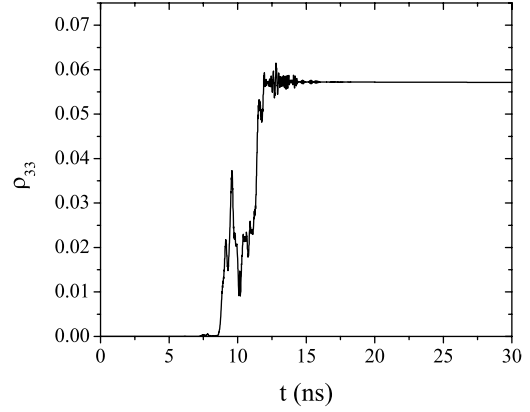


Figure 10. ρ_{33} , the excited state population, as a function of time obtained from the calculation described in the text.

in a Cs ultracold Rydberg gas than in a Rb ultracold Rydberg gas. An experiment of this nature using Cs instead of Rb would have difficulty observing a signal given its presumably low level.

In our calculations, we have observed that the population in the $32\tilde{P}_{3/2} + 31\tilde{P}_{3/2}$ state, observed by measuring the $32P_{3/2}$ population, reaches equilibrium after 15 ns, figure 10. In order to get the total excited $32\tilde{P}_{3/2} + 31\tilde{P}_{3/2}$ state population, it is necessary to integrate from $R = 0.55$ μ m to $R = 1.8$ μ m. The inner cutoff was chosen to be $R = 0.55$ μ m because the potentials are so attractive or so repulsive at smaller R that the pair state is out of resonance with the laser pulse. The outer cutoff corresponds to the distance at which the multipolar interaction becomes smaller than the kinetic energy of the atoms. We obtain an average of $N_{\text{exc}} \sim 5100$ excited pairs per laser pulse with these assumptions. If the maximum density of excited Rydberg atoms is 5×10^9 cm⁻³ as measured in the experiment and considering that the resonant excitation of $32S$ atom pairs is saturated at the intensities used in the experiment, we obtain $N_{32S} = 1.3 \times 10^5$ excited $32S$ atoms per pulse. This leads to an estimated $32P_{3/2}$ to $32S_{1/2}$ signal ratio of 3.9%, which is in excellent agreement with the experimental value of $\sim 2.8\%$ at a PFI detection time of 100 ns [15]. Differences in the measured and theoretical values are thought to be due to errors in finding the number of Rydberg atoms detected and excited in the experiment since the experimentally determined numbers were used for the calculations.

The model that we have described predicts that the population transfer occurs on a short time scale, < 15 ns. After this short period, the nP state population can only decay spontaneously or via blackbody processes. To confirm this, we measured the time evolution of the nP electron signal up to 10 μ s after the laser pulse was applied. It is well known that blackbody radiation can transfer population to the nP state, especially at low Δn [1]. To eliminate the blackbody contribution from the detected nP signal, we used the same technique applied in our work on Rydberg atom $nD+nD$ collisions [11]. We used a third boxcar gate to detect the time-dependent population of the states lying energetically above the nP state N_{up} . Since these upper states in this case can only be excited by blackbody radiation,

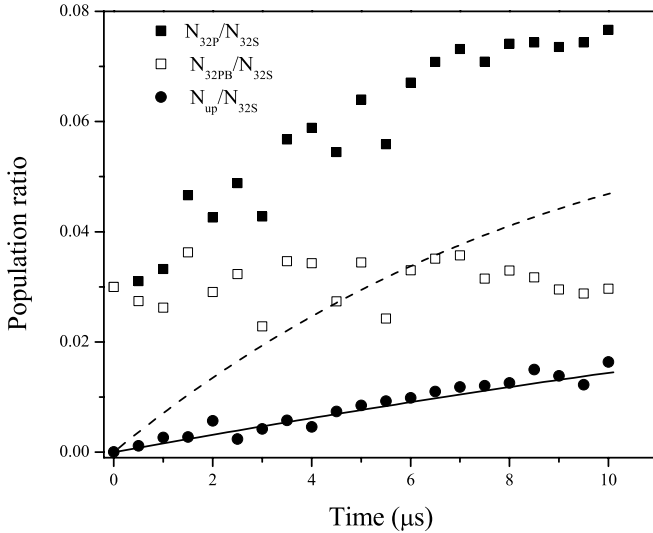


Figure 11. N_{32P} , N_{32PB} and N_{up} populations as a function of time. The full line is the N_{up} population exclusively due to blackbody radiation transfer. The dashed line is the blackbody contribution to the N_{32P} population obtained with the rate equation model.

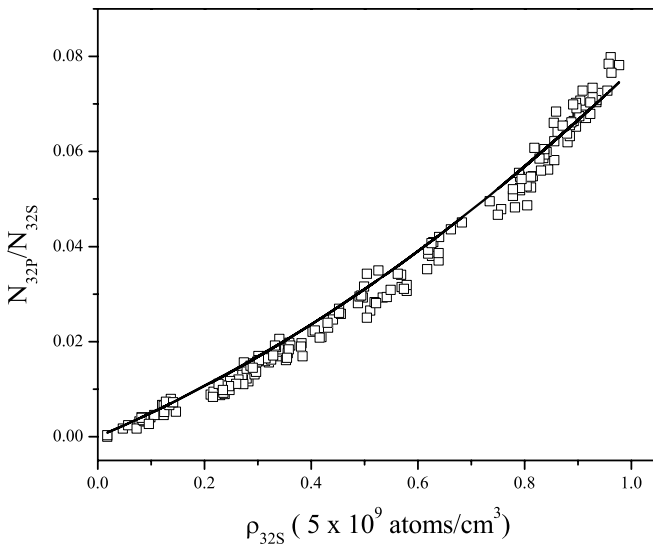


Figure 12. N_{32P} to N_{32S} ratio as a function of 32S atomic density. The full line is a no free parameter model which considers the two-body contribution and the blackbody radiation contribution.

their populations are a nonadiabatic process and spontaneous emission-free measurement of the blackbody transfer. We compared these measurements with a rate equation model for blackbody radiation transfer, which we have also used in [11]. In figure 11, we show the experimental populations for N_{32P} (full square) and N_{up} (full circle) as a function of time delay. The figure shows the theoretical model for blackbody radiation transfer for N_{32P} (dashed line) and N_{up} (full line). The $32P_{3/2}$ state population without blackbody contributions, N_{32PB} (open square), is obtained by subtracting the theoretical $32P_{3/2}$ population due to blackbody transfer from the experimental measurement. The resulting population exhibits a slight decay, which is consistent with the $32P_{3/2}$ state lifetime. After direct excitation and the removal of

the blackbody contribution to the $32P_{3/2}$ population, the remaining fraction of atoms can only spontaneously decay. The behaviour shown in figure 11 is in agreement with our theoretical model, which predicts that the population transfer due to the ac Stark effect of the laser pulse occurs in less than 15 ns. We also emphasise that this behaviour is completely different from the population transfer that occurs after the excitation of Rb $nD+nD$ Rydberg atoms [11].

To further verify our blackbody transfer model, we measured the $32P$ state population as a function of $32S$ atomic density for a time delay of $10\mu s$. In figure 12, we show the $32P_{3/2}/32S$ ratio as a function of ρ_{32S} . The full line is a theoretical prediction with no free parameters, which takes into account the two-body contribution from the Rydberg atom pair excitation at short time delay, < 15 ns, and the blackbody contribution for $10\mu s$. Clearly, these results corroborate our description of the dynamics and the blackbody radiation.

4. Conclusion

To summarize, we have studied the role of the Stark effect on interactions between ultracold Rydberg atoms. We have shown that a small constant electric field can completely change the interactions between pairs of Rydberg atoms, revealing the multilevel character of the potentials and the importance of interactions that occur at short range. The agreement between theory and experiment that we have shown in this work and others gives us confidence that our description of the main physical features is accurate. We expect that the angular dependence of Rydberg atom interactions will be the next important test for our theoretical model, as this is an increasingly interesting avenue to explore. The new time evolution data presented here on the $nP_{3/2}$ atom production after excitation of cold Rb $nS_{1/2}$ Rydberg atoms provide further proof that the model presented in our previous work is accurate [15]. An important conclusion of this paper is that the effects observed for one specific atomic species cannot always be extrapolated to another one in a direct way. For each atomic species and principal quantum number, it is necessary to calculate the Rydberg atom pair interaction potentials before any prediction can be made. This may be a crucial point when comparing experiments performed with different atoms and different Rydberg states.

Acknowledgments

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