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Controlling the interactions between cold Rydberg atoms by a time-varying electric field

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Abstract. Long-range interactions between cold Rydberg atoms are being investigated for neutral-atom quantum computing, quantum simulations, phase transitions in cold Rydberg gases and other applications. Fine tuning of the interaction strength can be implemented using Förster resonances between Rydberg atoms controlled by an electric field. The observation of the Stark-tuned Förster resonances between Rydberg atoms excited by narrowband cw laser radiation requires usage of a Stark-switching technique in order to excite the atoms first in a fixed electric field and then to induce the interactions in a varied electric field, which is scanned across the Förster resonance. The application of the radio-frequency field causes additional Förster resonances between collective states, whose line shape depends on the interaction strengths and time. Spatial averaging over the atom positions in a single interaction volume yields a cusped line shape of the Förster resonance. We present a detailed experimental and theoretical analysis of the line shape and time dynamics of the Stark-tuned Förster resonances $Rb(nP_{3/2}) + Rb(nP_{3/2}) \rightarrow Rb(nS_{1/2}) + Rb([n+1]S_{1/2})$ for two Rb Rydberg atoms interacting in a time-varying electric field.

1. Introduction

Long-range interactions between highly excited Rydberg atoms are of interest for quantum information processing with single neutral atoms [1, 2]. Resonant dipole-dipole interaction between atoms in an identical $nL$ Rydberg state can be implemented via Stark-tuned Förster resonances [3-7]. A Rydberg state should be exactly midway between two other Rydberg states of the opposite parity to induce a Förster resonance (figure 1a).

In our recent experiments [2, 7-9] we used three-photon excitation of the $Rb(nP)$ Rydberg atoms by narrowband cw lasers. Scanning the electric field by just 50 mV/cm shifts the Rydberg level out of resonance with the laser radiation, as the cw lasers have line widths below 1 MHz [7]. In order to avoid this problem, we applied a Stark-switching technique [10, 11] to excite the atoms first in a fixed electric field and then to induce the interactions in a lower electric field, which is scanned across the Förster resonance, as shown in figure 1b.

In this report, we present a detailed experimental and theoretical analysis of the Förster resonance line shapes and time dynamics in a time-varying electric field used for the Stark switching. The resonance under study is the Förster resonant energy transfer $Rb(nP_{3/2}) + Rb(nP_{3/2}) \rightarrow Rb(nS_{1/2}) + Rb([n+1]S_{1/2})$ due to dipole-dipole interaction of two Rb Rydberg...
Figure 1. (a) Förster resonance Rb\((nP)\)\(\rightarrow\)Rb\((nP)\)+Rb\((nS)\)+Rb\([n+1]S\) for two Rb Rydberg atoms. (b) Timing diagram of the laser and electric-field pulses. (c) Collective states of the Förster resonance Rb\((37P)\)+Rb\((37P)\)\(\rightarrow\)Rb\((37S)\)+Rb\((38S)\) in the dc electric field. Intersection at 1.79 V/cm corresponds to a Stark-tuned Förster resonance. (d) Collective states of the Förster resonance Rb\((39P)\)+Rb\((39P)\)\(\rightarrow\)Rb\((39S)\)+Rb\((40S)\) in the dc electric field. A dc Stark-tuned Förster resonance is impossible, but it can be induced by adding a \(\sim 100\)-MHz radio-frequency field (green arrow) that binds the collective states [8, 9].

atoms (figure 1a) in a small single laser excitation volume of a frozen Rydberg gas. The energy detuning of this resonance, \(\hbar \Delta = E(nS_{1/2}) + E([n+1]S_{1/2}) - 2E(nP_{3/2})\), is controlled by a weak dc electric field \(F\). The energy shift of a Rydberg level \(nL\) with nonzero quantum defect is quadratic and is defined by its polarizability, \(\delta E_{nL}/\hbar = -\alpha_{nL}F^2/2\). The detuning is then given by

\[
\Delta = \Delta_0 + (\alpha_{nP} - \frac{1}{2}\alpha_{nS} - \frac{1}{2}\alpha_{[n+1]S}) F^2.
\]  

Here, \(\Delta_0\) is the detuning in a zero electric field (for example, \(-103\) MHz for \(n=37\) and \(+74\) MHz for \(n=39\)). The detuning can be tuned to zero for Rydberg states with \(n \leq 38\), as shown in figure 1c for the \(37P_{3/2}(|M_J| = 1/2)\) state, while for states with \(n \geq 39\), the dc electric field only increases \(\Delta\), as shown in figure 1d for the \(39P_{3/2}(|M_J| = 1/2)\) state. For the latter, a Förster
resonance can be obtained by adding a 90–100 MHz rf field that induces rf transitions between collective states and compensates for the Förster energy defect, as demonstrated in our recent papers [8, 9].

Our previous numerical Monte Carlo simulations using the Schrödinger’s equation [5, 12] and our experimental data [5, 8, 9] have shown that spatial averaging over the random positions of \( N = 2 - 5 \) Rydberg atoms interacting in a single laser excitation volume results in cusp-shaped Förster resonances as soon as the resonances saturate and the interaction time is long enough. Cusp-shaped Förster resonances were also observed recently at high Rb Rydberg atom density in Ref. [13] and an analytical model has been proposed. Three-body Förster resonances in Cs Rydberg states with \( \Delta M_J \) randomly placed in a single excitation volume. Our Förster resonance induces transitions between Rydberg states with \( \Delta M_J = 2 \) interacting in a single laser excitation volume results in cusp-shaped Förster resonances.

2. Time dynamics and line shapes of the Förster resonances

2.1. Theory with density-matrix equations

We consider an example of the Stark-tuned Förster resonance for two Rb Rydberg atoms randomly placed in a single excitation volume. Our Förster resonance induces transitions between Rydberg states with \( \Delta M_J = 0 \). This corresponds to the \( z \)-oriented dipoles, and the operator of the dipole-dipole interaction is

\[
\hat{V} = \frac{\hat{d}_a \hat{d}_b}{4\pi\varepsilon_0} \left[ \frac{1}{R_{ab}^3} - \frac{3}{2} \left( \frac{Z_{ab}^z}{R_{ab}^3} \right)^2 \right],
\]

where \( \hat{d}_a,b \) are the \( z \) components of the dipole-moment operators of the two interacting atoms \( a \) and \( b \), \( Z_{ab}^z \) is the \( z \) component of the vector connecting the two atoms \( \mathbf{R}_{ab} \) (\( z \) axis is chosen along the dc electric field), and \( \varepsilon_0 \) is the dielectric constant.

In order to calculate the time evolution of the populations in the two Rydberg atoms interacting via a Förster resonance, we elaborate a density-matrix model, which takes into account the additional broadening due to unresolved hyperfine structure and fluctuations of the controlling dc electric field. Our considerations are limited to the case of \( N=2 \) interacting Rydberg atoms, when approximate analytical solutions can be found and compared with our experimental data.

Let us denote the lower energy state 37S as state 1, the middle state 37P_{3/2} as state 2, and the upper state 38S as state 3, as shown in figure 2a. Then, for two interacting Rydberg atoms with dipole-dipole matrix element \( \hat{V} \) given by equation (2), [22] is the initial collective state populated by a short laser pulse at \( t = 0 \), and [13] and [31] are the two equally populated final states having a small energy detuning \( \Delta \) from state [22] (figure 2b). A simpler effective two-level system, shown in figure 2c, with a reduced dipole-dipole matrix element \( \sqrt{2} \hat{V} \) can replace the three-level system of figure 2b in the theoretical calculations, because collective states [13] and [31] are equivalent and behave identically. We now denote state [22] as state \( a \) in the effective two-level system and the symmetric composite state \( (|13\rangle + |31\rangle)/\sqrt{2} \) as state \( b \) in figure 2c. The other, antisymmetric composite state \( (|13\rangle - |31\rangle)/\sqrt{2} \) is not considered, as it is unaffected by the operator of the dipole-dipole interaction \( \hat{V} \) and can be excluded from the analysis.

The most convenient form of equations for the density-matrix elements \( \rho_{ij} \) is the optical Bloch equations [15]:

\[
\frac{d}{dt} \rho_{ij} = \sum_{kl} (\Gamma_{kl}^{ij} - \Gamma_{ij}^{kl}) \rho_{kl} + \sum_{kl} W_{ij,kl} \rho_{kl} - \sum_{kl} W_{kl,ij} \rho_{kl} + \sum_{kl} F_{ij,kl} \rho_{kl},
\]

where \( \Gamma_{kl}^{ij} \) is the rate of population transfer from state \( k \) to state \( l \) to state \( j \), \( W_{ij,kl} \) is the rate of excitation from state \( k \) to state \( l \) to state \( j \), and \( F_{ij,kl} \) is the rate of excitation from state \( k \) to state \( l \) to state \( j \).
Figure 2. (a) Rydberg states related to the Förster resonance Rb\((nP) + Rb(nP) \rightarrow Rb(nS) + Rb\((n+1)S\)\)). (b) Collective states of two Rydberg atoms interacting via Förster resonance with dipole-dipole matrix element \(V\). The energy defect of the Förster resonance \(\Delta\) is controlled by the electric field. (c) Effective two-level system can replace the three-level system in the theoretical calculations, as collective states \(|13\rangle\) and \(|31\rangle\) are equivalent and behave identically. The reduced dipole-dipole matrix element is now \(\sqrt{2}V\).

\[
\begin{align*}
\dot{\rho}_{aa} &= i\sqrt{2} \Omega (\rho_{ab} - \rho_{ba}), \\
\dot{\rho}_{bb} &= i\sqrt{2} \Omega (\rho_{ba} - \rho_{ab}), \\
\dot{\rho}_{ab} &= -i(\Delta + \Gamma/2) \rho_{ab} + i\sqrt{2} \Omega (\rho_{aa} - \rho_{bb}), \\
\dot{\rho}_{ba} &= i(\Delta - \Gamma/2) \rho_{ba} + i\sqrt{2} \Omega (\rho_{bb} - \rho_{aa}).
\end{align*}
\]

Here, \(\Omega = V/\hbar\) is the matrix element of the dipole-dipole interaction in circular frequency units. In order to take into account the additional broadening \(\Gamma\) phenomenologically, equations (3) are modified using the method we have applied previously in Ref.[7] to account for the finite laser linewidths in a four-level theoretical model of the three-photon laser excitation of Rydberg states. In the equations for the coherences, we add the terms with \(\Gamma/2\) in order to introduce additional coherence decay.

The two-atom signal \(S_2\) measured in our experiments is a fraction of Rydberg atoms in the final state \(37S\) or a population of the final state \(37S\) per atom, which is calculated for the interaction time \(t\) as

\[
S_2 = \frac{1}{2} \rho_{bb}(t).
\]

In order to find \(\rho_{bb}(t)\), equations (3) can be solved analytically with the initial conditions \(\rho_{aa}(0) = 1; \rho_{bb}(0) = \rho_{ab}(0) = \rho_{ba}(0) = 0\). Their solution reduces to finding the roots of a cubic equation, if \(\Omega, \Delta, \) and \(\Gamma\) are independent of time [9]. The resulting general analytical expressions are rather complicated, however, and in what follows we consider the analytical solutions only for some particular cases.

2.2. Theory for the Förster resonance amplitude

The exact analytical solution of equations (3) for the time evolution of the Förster resonance amplitude \(S_2(\Delta = 0)\) is given by

\[
S_2(\Delta = 0) = \frac{1}{4} - \frac{1}{4} e^{-\Gamma t/4} \left[ \text{ch} \left( \sqrt{\Gamma^2/16 - 8\Omega^2 t} \right) + \frac{\Gamma/4}{\sqrt{\Gamma^2/16 - 8\Omega^2 t}} \text{sh} \left( \sqrt{\Gamma^2/16 - 8\Omega^2 t} \right) \right].
\]

At the weak dipole-dipole interaction \(8\Omega^2 \ll \Gamma^2/16\), equation (5) reduces to
Figure 3. (a) Comparison between approximate (equation (6), blue line) and exact (equation (5), green line) analytical calculations of the time dependence for Förster resonance amplitude at the weak dipole-dipole interaction of two frozen Rydberg atoms. (b) The same for the strong interaction and equations (5) and (7).

\[ S_{\text{weak}}^2(\Delta = 0) \approx \frac{1}{4} \left[ 1 - e^{-16\Omega^2 t/\Gamma} \right]. \]  

(6)

The amplitude slowly goes to its steady-state value 1/4 as \( t \) increases. For the strong dipole-dipole interaction \( 8\Omega^2 \gg \Gamma^2/16 \), the damped Rabi-like oscillations appear, while the steady-state value is also 1/4:

\[ S_{\text{strong}}^2(\Delta = 0) \approx \frac{1}{4} \left[ 1 - e^{-\Gamma t/4} \cos \left( 2\sqrt{2}\Omega t \right) \right]. \]  

(7)

Figure 3a demonstrates the comparison between the approximate (equation (6), blue line) and exact (equation (5), green line) analytical calculations of the time dependence for the Förster resonance amplitude at the weak dipole-dipole interaction of two spatially frozen Rydberg atoms. Although there is some discrepancy at the short interaction time due to unaccounted coherence in equation (6), the overall agreement is rather good. Figure 3b shows the same for the strong interaction and demonstrates almost perfect agreement between equations (5) and (7). The calculated time dependences in figure 3 is what can be observed with two Rydberg atoms, interacting in two spatially separated optical dipole traps, as in the experiment of Ref. [16].

In our experiments, the two atom positions are not fixed. The interaction term \( \Omega \) in equations (6) and (7) has a fluctuating value that results in decoherence and washing out the Rabi-like oscillations in equation (7), even at \( \Gamma = 0 \). To calculate the resonance amplitude measured in our experiments, equations (6) and (7) should be averaged over all possible spatial positions of the two interacting atoms in the excitation volume, which is formed by the two intersecting laser beams. In Ref. [12] we have shown that the averaging can be done using the nearest-neighbor probability distribution [17] with the average distance between nearest-neighbor atoms \( R_0 \approx \left[ 3/(4\pi n_0) \right]^{1/3} \) at volume density \( n_0 \). Using this method, we find the approximate analytical solutions to the averaged amplitudes of equations (6) and (7) [9]:

\[ < S_{\text{weak}}^2(\Delta = 0) > \approx \frac{1}{4} \left[ 1 - e^{-[0.44\Omega_0^2 t/\Gamma]} \right]^{1/3}, \]  

(8)

\[ < S_{\text{strong}}^2(\Delta = 0) > \approx \frac{1}{4} \left[ 1 - e^{-0.55\Omega_0 t - \Gamma t/4} \right], \]  

(9)

where

\[ \Omega_0 = \frac{\sqrt{2}d_1 d_2}{4\pi \varepsilon_0 \hbar R_0^3} \]

is the orientation-averaged reduced interaction matrix element at the average distance \( R_0 \). Here, \( d_1 \) and \( d_2 \) are the dipole moments of transitions \( nP_{3/2} (M_J = 1/2) \rightarrow nS_{1/2} (M_J = 1/2) \) and...
\( nP_{3/2} (M_J = 1/2) \rightarrow (n+1)S_{1/2} (M_J = 1/2) \). The fitting coefficients 0.44 and 0.55 have been found from the numerical simulations.

Figure 4a demonstrates the comparison between the analytical (equation (8), blue line) and numerical (green line) calculations of the time dependence for Förster resonance amplitude at the weak dipole-dipole interaction of two spatially averaged Rydberg atoms. Again, there is some discrepancy at the short interaction time, but at the longer times the agreement is good. Figure 4b shows the same for the strong interaction and demonstrates good agreement between the analytical calculations with equation (9) and numerical results. We note that the time dependences of the averaged amplitudes in equations (8) and (9) significantly differ from those in equations (6) and (7) for two spatially fixed atoms. This should be taken into account when theory is compared with experiment.

Figure 5. (a) Comparison between analytical (equation (10), blue line) and numerical (green line) calculations of the Förster resonance line shape at the weak dipole-dipole interaction of two frozen Rydberg atoms. (b) The same for the strong interaction and equation (13).

2.3. Theory for the Förster resonance line shape

The analytical expressions for the line shape of the two-atom Förster resonances at \( \Delta \neq 0 \) are much more complicated than equations (5)–(7). In the case when \( \rho_{bb} \ll \rho_{aa} \) (weak interaction or short interaction time), the line shape for two frozen Rydberg atoms is approximately given by

\[
S_{\text{weak}} \approx \frac{2\Omega^2}{\Delta^2 + \Gamma^2/4} \left( \frac{\Gamma t}{2} + \frac{\Delta^2 - \Gamma^2/4}{\Delta^2 + \Gamma^2/4} \left[ 1 - e^{-\Gamma t/2} \cos (\Delta t) \right] - \frac{\Delta \Gamma}{\Delta^2 + \Gamma^2/4} e^{-\Gamma t/2} \cos (\Delta t) \right)
\]

(10)

Our numerical simulations have shown that this formula is quite precise (error less than 10%) for arbitrary \( \Omega, \Gamma, \Delta \), and \( t \) as far as \( S_2 < 0.1 \). In fact, this is a general formula for the line shape of weak Förster resonances in two frozen Rydberg atoms. Figure 5a presents the comparison between analytical (equation (10), blue line) and numerical (green line) calculations of the Förster resonance line shape at the weak dipole-dipole interaction of two frozen Rydberg atoms and the interaction time \( t \sim 0.5 \mu s \).

In the opposite case of strong interaction \( 8\Omega^2 \gg \Gamma^2/16 \), the line shape is approximately given by
This formula describes the saturation of the Förster resonance accompanied by the damped Rabi-like oscillations. As $t$ or $\Omega$ grow, there appears a flat-top contour with the width of the flat-top part $4\sqrt{2}\Omega$, while the resonance wings are close to a Lorentzian. At $t \to \infty$, equation (11) gives $S^\text{strong}_2 \to 1/4$ independently of $\Delta$. This is because the steady-state solutions $\rho_{aa} = \rho_{bb} = 1/2$; $\rho_{ab} = \rho_{ba} = 0$ of equations (3) are independent of $\Delta$ if $\Gamma \neq 0$. For large $\Delta$, however, the time evolution is very slow, as described by the second term in equation (11). Figure 5b presents the comparison between analytical (equation (11), blue line) and numerical (green line) calculations of the Förster resonance line shape at the strong dipole-dipole interaction of two spatially averaged Rydberg atoms. (b) The same for the strong interaction.

Equation (12) is thus a general formula that describes the cusped line shape of the Förster resonance for two Rydberg atoms randomly positioned in a single interaction volume. It is valid for a broad range of all parameters, and in addition it describes the time dynamics of the Förster resonance for the interaction times $t > 1 \mu s$.

The formation of the cusp-shaped resonance upon spatial averaging can be understood from the fact that at zero detuning, the interaction is long range (resonant dipole-dipole) and it is effective even for the distant atoms, while for nonzero detuning the interaction is short range (vanishingly small) at large distances. In our averaging here, we use not the Lorentz line shape but equation (11) with $\Gamma \to \infty$, as discussed in Ref. [9]. The averaged line shape turns out to be a cusp, which is approximately described by the following formula:
and it is much weaker for the distant atoms [8]. Therefore, upon spatial averaging, the resonance wings are reduced stronger than the resonance center, and this finally results in the cusped line shape.

The only drawback of equation (12) is that it is invalid for the short interaction times ($t < 1 \mu s$), when the Fourier broadening dominates. For the analytical estimates, the averaged Fourier broadening is described with a certain accuracy by equations (10) or (11) if $2\Omega^2$ is replaced by $\Omega_0^2$.

2.4. Experimental Förster resonance line shape

The experiments were performed with cold $^{85}$Rb atoms in a magneto-optical trap (MOT). Our experiments feature atom-number-resolved measurement of the signals obtained from $N = 1 - 5$ detected Rydberg atoms with a detection efficiency of 65% [5, 18]. It is based on a selective field ionization (SFI) detector with channel electron detector (CEM) and post-selection technique. We can record Förster resonances for up to five of the detected Rydberg atoms and compare these with the theoretical simulations.

The excitation of Rb atoms to the $nP_{3/2}(|M_J|=1/2)$ Rydberg states is realized via three-photon transition $5S_{1/2} \rightarrow 5P_{3/2} \rightarrow 6S_{1/2} \rightarrow nP_{3/2}$ (figure 2a) by means of three cw lasers modulated to form $2 \mu s$ exciting pulses at a repetition rate of 5 kHz [7]. Small Rydberg excitation volume of $20 - 40 \mu m$ in size is formed using crossed laser-beam geometry [5]. The laser intensities are adjusted to obtain about one Rydberg atom excited per laser pulse on average.

We use a Stark-switching technique [10, 11] to switch the Rydberg interactions on and off, as depicted in figure 1b. Laser excitation occurs during $2 \mu s$ at a fixed electric field of $5.6 \ V/cm$. Then the field decreases to a lower value near the resonant electric field ($1.79 \ V/cm$ for the $37P_{3/2}$ state), which acts for $3 \mu s$ or less until the field increases back to $5.6 \ V/cm$. Then, $0.5 \mu s$ later, a ramp of the strong field-ionizing electric pulse of $200 \ V/cm$ is applied. The lower electric field is slowly scanned across the Förster resonance and the SFI signals are accumulated for $10^3 - 10^4$ laser pulses. A pulse of the rf field with variable amplitude ($0 - 0.5 \ V/cm$) and frequency ($10 - 100 \ MHz$) can be admixed to the lower dc field.

Figure 7 presents the line shapes of the Förster resonance Rb$(37P)+$Rb$(37P) \rightarrow$ Rb$(37S)+$Rb$(38S)$ recorded in our experiment for $N=2$ detected Rydberg atoms at various $t$. The interaction time $t$ is set by the length of the square-shaped controlling electric-field pulse.

At the long interaction times ($1.975$ and $2.975 \mu s$), the resonance shape and width in figures 7g and 7h almost do not change with $t$. This means that the resonance takes its stationary form, and only its amplitude slowly grows with $t$ according to equation (8). The resonance is cusp shaped, in agreement with equation (12). The narrowest resonance appears at $t=2.975 \mu s$. Its width in the electric-field scale is $16 \ mV/cm$, corresponding to $1.9 \ MHz$ in the detuning scale. The detuning scale is obtained using the calculated polarizabilities of the collective Rydberg states in figure 1c. Our attempts to obtain a narrower resonance by increasing $t$ were not successful. Since the Fourier width of the interaction pulse is $t^{-1} \approx 0.34 \ MHz$ and the estimated average interaction energy is $\Omega_0/(2\pi) \sim 0.25 \ MHz$, they cannot be fully responsible for the $1.9 \ MHz$ resonance width. Therefore, there is an additional broadening $\Gamma/(2\pi) \sim 1 \ MHz$, apparently due to the unresolved hyperfine structure and parasitic ac electric fields. This observation agrees with what we have observed in our previous experiments [5, 8].

The Fourier broadening of the resonances is demonstrated in figures 7a–7e at the shorter interaction times ($< 1 \mu s$). The Fourier width of the interaction pulse is significant at short $t$ and the Förster resonance broadens, while its amplitude decreases.

In figures 7a–7h, we also compare the experimental two-atom spectra at various $t$ with the line shapes numerically calculated with equations (3) and averaged over the random positions of two atoms in a cubic interaction volume. The fitting parameters in the theory were the volume size...
Figure 7. (a)-(h) Comparison between experimental and theoretical line shapes of the Förster resonance $\text{Rb}(37P) + \text{Rb}(37P) \rightarrow \text{Rb}(37S) + \text{Rb}(38S)$ for two Rydberg atoms at various interaction times. (i) Comparison between experiment and theory for the resonance amplitude at various interaction times. (k) Comparison between experiment and theory for the resonance width at various interaction times.

$26 \times 26 \times 26 \, \mu m^3$ and the additional broadening $\Gamma/(2\pi) = 0.5$ MHz. We also added a background signal $S_2 \approx 0.037$ that appears in the experiments due to parasitic transitions between Rydberg states induced by a 300 K blackbody radiation (BBR). These parameters allowed us to fit the time dependences of the experimental spectra, both for the amplitude (figure 7i) and width (figure 7k) of the two-atom Förster resonance. The amplitude and width are measured with respect to the BBR-induced background signal level.

The time dependence of the amplitude in figure 7i for $t > 1 \, \mu s$ is also well fit by equation (8) at $\Omega_0/(2\pi) = 0.25$ MHz and $\Gamma/(2\pi) = 0.5$ MHz. The time dependence of the width in figure 7k for $t < 1 \, \mu s$ is mainly represented by the Fourier transform width $FWHM/(2\pi) \approx 1/t$. For $t > 1\mu s$, the width becomes nearly constant.
Figure 8. (a)-(d) Comparison between experimental and theoretical line shapes of the inaccessible Förster resonance $\text{Rb}(39P) + \text{Rb}(39P) \rightarrow \text{Rb}(39S) + \text{Rb}(40S)$ induced by the 95 MHz and 100 mV rf field for two Rydberg atoms at various interaction times. (e) Comparison between experiment and theory for the resonance amplitude at various interaction times. (f) Comparison between experiment and theory for the resonance width at various interaction times.

Now we will turn to the "inaccessible" Förster resonance $\text{Rb}(39P_{3/2}) + \text{Rb}(39P_{3/2}) \rightarrow \text{Rb}(39S_{1/2}) + \text{Rb}(40S_{1/2})$ which cannot be tuned by the dc electric field. Its collective energy levels in the dc electric field are shown in figure 1d. The dc field alone increases the energy detuning $\Delta$ and makes the interaction between $\text{Rb}(39P)$ atoms less efficient. However, as we have shown in Refs. [8, 9], the rf field can induce transitions between collective states, so the population transfer at the Förster resonance occurs for the Floquet sidebands irrespective of the possibility to tune it by the dc field. In the present experiment, we applied rf field with 95 MHz frequency and 100 mV amplitude, which created the Floquet sidebands.

In figures 8a–8d, we compare the experimental rf-induced Förster resonances recorded for two atoms at various $t$ to the theoretical line shapes numerically calculated with equations (3) and averaged over random positions of two atoms in a cubic interaction volume. A BBR-induced background signal $S_2 \approx 0.03$ is added to the theoretical spectra. The fitting parameters in the theory were the volume size $16 \times 16 \times 16 \, \mu\text{m}^3$, the additional broadening $\Gamma/(2\pi) = 1 \, \text{MHz}$, and the rf-field strength 150 mV/cm (the latter is 50% larger than the experimentally measured one; this discrepancy remains unclear). These allowed us to fit the time dependences of the experimental spectra both for the amplitude (figure 8e) and width (figure 8f) of the two-atom Förster resonance. The amplitude and width are measured with respect to the BBR-induced background-signal level.

The narrowest resonance appears at $t=2.975 \, \mu\text{s}$. It has a cusped shape, in agreement with equation (12), and its width in the electric-field scale is 18 mV/cm, corresponding to 1.1 MHz in the detuning scale, so this resonance is nearly two times narrower than that for the $37P_{3/2}$ state. The detuning scale is obtained using the calculated polarizabilities of the collective Rydberg states in figure 1d. The main contribution to $\Gamma$ thus comes from the parasitic ac electric fields. That is why in the theory we have to take $\Gamma/(2\pi) = 1 \, \text{MHz}$ (i.e., two times larger than that for the $37P_{3/2}$ state), and a smaller interaction volume to fit the amplitude of the two-atom resonances in figure 8.
3. Conclusions

Our simple theoretical density-matrix model works well and provides correct analytical and numerical results for the time dynamics and cusped line shapes of the Förster resonances in two randomly positioned Rydberg atoms interacting in a single laser-excitation volume.

Good agreement between experiment and theory confirms that any unresolved (hyperfine or Zeeman) structure or parasitic ac Stark broadening of the Förster resonances can be accounted for theoretically by a single parameter $\Gamma$. This significantly reduces the number of equations and simplifies the calculations of the line shapes and time dynamics of the Förster resonances.

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