

## Modeling many-particle mechanical effects of an interacting Rydberg gas

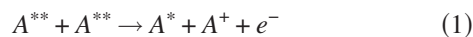
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In a recent work [Phys. Rev. Lett. **98**, 023004 (2007)], we investigated the influence of the attractive van der Waals interaction on the pair distribution and Penning ionization dynamics of ultracold Rydberg gases. Here we extend this description to atoms initially prepared in Rydberg states exhibiting repulsive interaction. We present calculations based on a Monte Carlo algorithm to simulate the dynamics of many atoms under the influence of both repulsive and attractive long-range interatomic forces. Redistribution to nearby states induced by blackbody radiation is taken into account, changing the effective interaction potentials. The model agrees with experimental observations, where the ionization rate is found to increase when the excitation laser is blue detuned from the atomic resonance.

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Ultracold Rydberg gases represent a unique system for studying atomic many-body dynamics on experimentally observable time scales. Among many other applications [1,2], these dilute and yet strongly interacting gases are the starting point for the formation of ultracold plasmas [3–7]. While the cold ground-state atoms can be considered to be fixed in space, highly excited atoms are accelerated due to their long-range interaction potentials. In a previous work we have shown how the attractive long-range van der Waals interaction influences the distribution of pair distances during excitation and is responsible for the initial collisional ionization in an ultracold Rydberg gas [8]. Ions produced by the Penning ionization process



were seen to appear earlier when the excitation laser was red detuned from the atomic resonance in a system with purely attractive interactions. As explained in [8], a red-detuned laser preferentially excites close pairs, which in turn collide on shorter time scales. In that work, the question remained of how collisional ionization can be described in a system with only repulsive van der Waals interaction (as is the case for Rydberg  $S$  states). Experimental observation showed that the gas was ionizing on longer time scales compared to the attractive case, and ions appeared earlier when the excitation was blue detuned [8]. Collisional ionization requires attractive potentials; thus we must assume that some atoms are transferred to other states after some time, thereby experiencing an attractive dipole-dipole interaction with their neighbors. Figure 1(a) illustrates this procedure. Apart from state-changing collisions with electrons, which already require some free charges in the cloud [7], the main source for redistribution of Rydberg states is blackbody radiation [9]. An experiment enforcing redistribution with microwave fields has shown that the resulting attractive forces lead to interaction-induced collisions [10].

However, a simple two-atom picture as shown in Fig. 1(a) with both atoms initially experiencing repulsive forces is not sufficient to explain our observations, since by the time a redistribution to an attractive potential takes place, initially close atoms have already been accelerated too far away from each other to return within observable time scales. In order to describe the system more realistically, one has to take into account the dynamics of many particles under the influence of repulsive interactions.

Here we show how the experimental observations can be

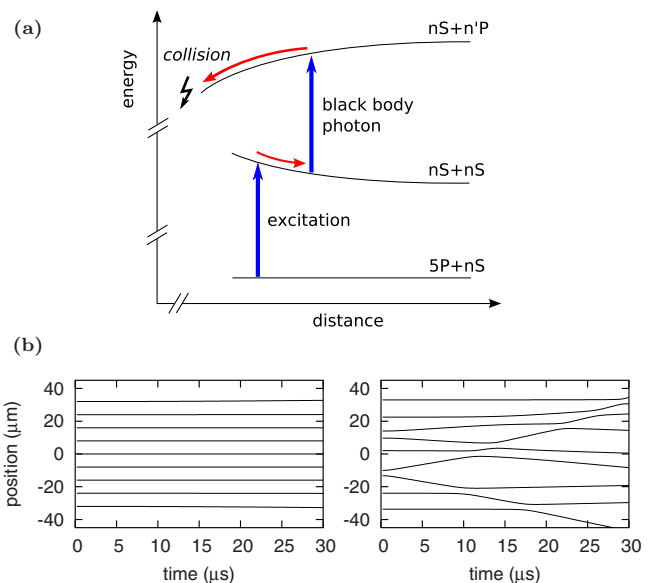


FIG. 1. (Color online) (a) Rydberg atoms excited in a repulsive potential. Blackbody photons can transfer one atom to a dipole-coupled state, allowing for attractive interaction. (b) One-dimensional calculation of particle trajectories to illustrate the idea of many-particle repulsive interaction. When all particles have the same (large) initial distance from their neighbors, they will be only slightly repelled (left graph). When some close pairs exist in the beginning, there will repeatedly be atoms coming close to each other over a long time scale (right graph). Here an average initial distance of  $8 \mu\text{m}$  and a van der Waals potential with  $C_6 = -10^{21}$  a.u. are assumed.

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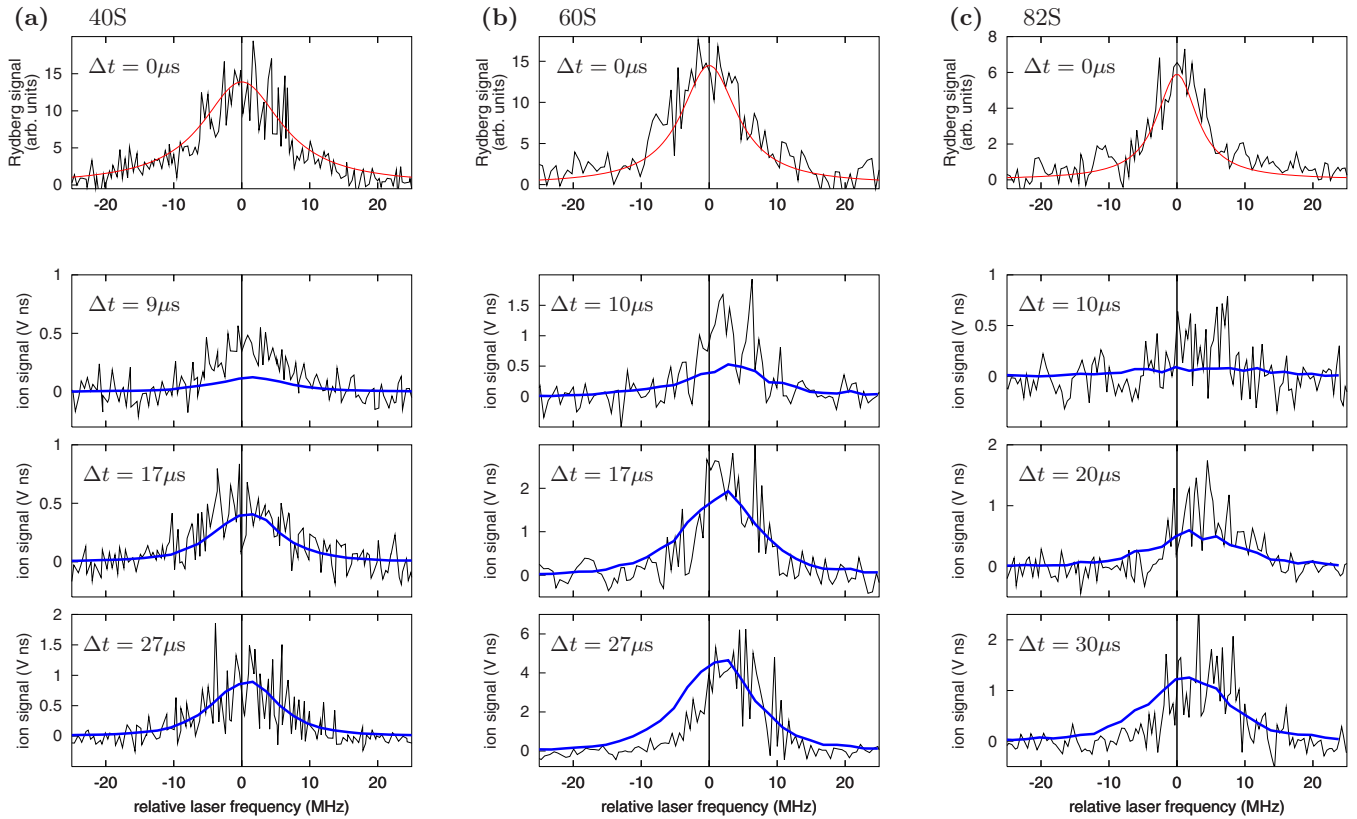


FIG. 2. (Color online) Expected collisional ionization when exciting in a repulsive potential. The upper graphs show the Rydberg signal directly after excitation, including a Lorentz fit to determine the excitation fraction and saturation to be used in the model. The other graphs show the ion signal detected after various delays. The corresponding simulated signals are plotted as bold (blue) lines.  $n =$  (a) 40, (b) 60, and (c) 82.

reproduced by considering blackbody redistribution and motion of many particles. Figure 1(b) illustrates the idea of the many-body aspect in a one-dimensional example: When all particles at a given average density have the largest possible (equal) initial distance from their neighbors, they are only slightly repelled from each other (left graph). Atoms redistributed to other states at any time need a rather long time to be accelerated toward a neighbor, as the separations are always large. In contrast, when some close pairs exist in the beginning, as is the case for blue-detuned excitation, there will constantly be atoms coming close to each other—even on a long time scale [right graph of Fig. 1(b)]. Atoms redistributed to other states at a certain time would then have the chance to collide with their neighbors earlier compared to red-detuned excitation. The same idea holds for the three-dimensional case implemented in our model described below.

The experimental setup is identical to the one described in [8]. We trap  $^{87}\text{Rb}$  atoms in a standard magneto-optical trap (MOT) at temperatures below  $100\ \mu\text{K}$  and a peak density on the order of  $10^{10}\ \text{cm}^{-3}$ . Excitation to Rydberg states is accomplished by a two-photon excitation scheme, the two transitions  $5S_{1/2} \rightarrow 5P_{3/2}$  and  $5P_{3/2} \rightarrow n\ell$  being realized with two cw laser systems at 780 and 480 nm, respectively. An ultra-stable reference cavity is used to actively stabilize the 480-nm excitation laser. The 780-nm laser illuminates the whole trapped atom cloud homogeneously, while the 480-nm

laser is focused with a waist of  $\sim 37\ \mu\text{m}$  at the center of the MOT. Every 70 ms, the excitation laser is switched on for 200 ns at a given detuning and the gas can then evolve freely for a variable time  $\Delta t$ . After that, an electric field ramp is applied to field-ionize the Rydberg atoms. The ions are detected before the field ramp is switched on (i.e., those produced by collisions) will reach the detector earlier, while Rydberg states ionize at a finite electric field, resulting in a delayed detector signal. The two signals are recorded simultaneously by two boxcar integrators. The Rydberg and ion signals have been measured for different initial Rydberg states (40S, 60S, and 82S), all exhibiting purely repulsive van der Waals interaction [11]. For each of the experiments we have verified that no ions are produced during the excitation. While at  $n=40$  no significant spectral asymmetry of the ion production rate has been observed, at  $n=60$  and 82 we see ions appear earlier on the blue-detuned side of the resonance. The measurements are displayed in Fig. 2. The graphs include Lorentzian fits to the Rydberg excitation line from which we estimate the excitation fraction and saturation used in the corresponding simulation.

Our model considers the interaction-induced forces among a large number of particles and calculates the motion of the atoms by numerically integrating the equations of motion. In the first part of the model, the Rydberg excitation process is simulated. 5000 atoms are randomly placed in a

TABLE I. Model parameters. Rates are given in  $s^{-1}$ , interaction coefficients in atomic units.

| $n$ | $R_{bb}$ | $R_{em}$ | $C_6$                 | $C_3^{eff}$     | $C_3^{max}$       |
|-----|----------|----------|-----------------------|-----------------|-------------------|
| 40  | 12730    | 17325    | $-7.0 \times 10^{18}$ | $2 \times 10^5$ | $8.1 \times 10^5$ |
| 60  | 5680     | 4845     | $-1.0 \times 10^{21}$ | $4 \times 10^6$ | $4.5 \times 10^6$ |
| 82  | 3030     | 1853     | $-3.9 \times 10^{22}$ | $3 \times 10^7$ | $1.7 \times 10^7$ |

cube to represent a density of  $10^{10} \text{ cm}^{-3}$  corresponding to the center of the MOT. The iterative excitation procedure is similar to the one used in Ref. [8]. In each iteration step, each atom  $i$  has a certain probability for excitation or stimulated emission,

$$\mathcal{P}_i = a\mathcal{L}[\delta - V_{vdW}(R_i)]e^{-2(x_i^2+y_i^2)/w^2}. \quad (2)$$

$\mathcal{L}$  represents the laser line shape, determined by the width of the intermediate state  $5P$  [6 MHz full width at half maximum (FWHM)], the widths of the two lasers (1–2 MHz), and the Fourier transform of the excitation pulse ( $\sim 3$  MHz FWHM).  $\delta$  is the detuning from resonance and  $V_{vdW}(R_i)$  is the interaction energy with the nearest Rydberg neighbor. The last factor accounts for the Gaussian intensity distribution of the excitation laser. The factor  $a=0.001$  is a small value to ensure a small number of excitations per iteration step. The procedure is iterated several hundred times to yield the actual percentage of excited atoms observed in the experiment (10–30 % at  $\delta=0$ ). For each atom, the distance  $R_i$  to its nearest Rydberg neighbor is recalculated after each excitation or stimulated emission event. In the second part of the model, the interaction-induced motion of the Rydberg atoms is calculated. During excitation, the interaction between a pair of atoms is assumed to be of van der Waals type,

$$V_{vdW}(R) = -\frac{C_6}{R^6}, \quad (3)$$

as all atoms are in an  $nS$  state. The values for  $C_6$  are calculated using a perturbative approach [11]. Directly after excitation, the atoms are assumed to be at rest, and are then free to move according to the forces caused by the interatomic interaction. These dynamics are simulated by numerical integration of the equations of motion, considering the interactions among all atoms. Whenever the distance between two atoms becomes less than  $4n^2a_0$ , these atoms are assumed to collide and undergo Penning ionization [6,12]. While moving, the atoms can change their internal state in two ways.

(1) Blackbody-induced redistribution to other Rydberg states. This process will mainly lead to nearby states, because of the higher transition matrix elements. The corresponding rates  $R_{bb}$  are calculated following [9] and listed in Table I.

(2) Spontaneous emission. These atoms are transferred to low-lying states (due to the  $\omega^3$  dependence of the Einstein A coefficient) and do not show any significant long-range interactions with other atoms anymore. They are thus removed from the simulation. The spontaneous emission rates  $R_{em}$ ,

calculated according to [9], are comparable to the blackbody redistribution and must be considered to reproduce the time development of the experimental ion signal (see Table I).

Direct photoionization by blackbody radiation is also included in the model, but the rates for this process are small ( $< 200$  Hz) [13] and are exceeded by the collision rates after the first few microseconds. Two different types of interaction potential are considered in the equations of motion: The (repulsive) interaction between atoms in the initial state is described by the potential in Eq. (3). Attractive potentials are introduced via the dipole-dipole interaction of an atom in the initial state ( $nS$ ) with an atom in a redistributed state ( $n'P$ ), given by

$$V_{dd}(\vec{R}) = \frac{\vec{\mu}_1 \cdot \vec{\mu}_2}{R^3} - 3 \frac{(\vec{\mu}_1 \cdot \vec{R})(\vec{\mu}_2 \cdot \vec{R})}{R^5}, \quad (4)$$

with  $|\vec{\mu}_1|=|\vec{\mu}_2|=\mu_{eff}$  and an effective coefficient  $C_3^{eff}=\mu_{eff}^2$ , representing the average over different electronic states after redistribution, and random angular dipole alignment. The largest possible transition dipole moment  $\mu_{max}$  is found between  $nS$  and  $n'P$  ( $n'=n$ ), which leads to a maximum effective interaction strength  $C_3^{max}=\mu_{max}^2$ . We have simulated the ionization dynamics for the three different principal quantum numbers investigated experimentally. The parameters used in the model are listed in Table I. The effective attractive interaction strengths  $C_3^{eff}$  are chosen to reproduce the time development of the data best. The bold lines in Fig. 2 show the results of the simulation, averaged over 100 runs. In order to compare the line shapes, the simulated traces have been scaled to the experimental data. The scaling factor is the same for all traces belonging to the same principal quantum number  $n$ . To achieve satisfactory overlap of the curves, we used different scaling factors for the three values of  $n$ . We attribute this to a number of effects which are not included in the model: First, the simulation is performed for a constant density, while in the experiment the excitation volume also extends over regions with lower density. The excitation fraction in the center of the cloud can only be roughly estimated, and the excitation is saturated to different amounts for the different measurements. Even more importantly, any secondary effects of the collision products, such as local electric fields and state-changing collisions [7], as well as avalanche ionization effects, are neglected in the model. For higher principal quantum numbers, a considerable amount of atoms may therefore experience a mixing of states in addition to the blackbody redistribution. For this reason, we expect the ion production to be underestimated for higher  $n$ , and accordingly the  $C_3^{eff}$  to be overestimated. In fact, for the  $40S$  state the simulated ionization fraction at  $30 \mu s$  (3.0%) corresponds to the experimental estimate of 4–8 %. At higher  $n$  the ionization fraction is systematically underestimated; for  $82S$  we obtain 1.5% ions from the model, compared to around 15% in the experiment. (The experimental fractions are subject to an uncertainty of about a factor of 2.) At the same time the attractive interaction is systematically overestimated as  $n$  increases (see Table I). If only blackbody redistribution rates were involved in the dynamics,  $C_3^{eff}$  should always be less than  $C_3^{max}$ , the maximum interaction strength

with an energetically close  $P$  state. This suggests that other redistribution processes and the influence of collision products become increasingly important.

Despite these limitations, the qualitative behavior of the dynamics of the system is very well reproduced: The maximum ionization rate appears on the blue-detuned side of the atomic resonance, and this spectral shift increases toward higher  $n$ , as the interaction-induced variation in the pair distance distribution becomes more distinct. The spectral shift of the maximum ion signal is mainly determined by the excitation process, and is insensitive to the choice of attractive potential. It is best reproduced for delay times below  $25 \mu\text{s}$ .

We have presented a Monte Carlo excitation model in combination with a numerical calculation of particle trajectories to explain the ionization behavior of a Rydberg gas initially prepared in a state with repulsive van der Waals interaction. In order to reproduce measurements showing an

increased ionization rate on the blue-detuned wing of the excitation line, it is necessary to consider both blackbody-induced redistribution of states and the motion of many atoms, while two-atom models fail to reproduce the experimental data. For increasing principal quantum number  $n$ , other redistribution processes and the influence of free charges become apparent. These results contribute to a more complete picture of the processes triggering ionization and plasma formation in ultracold Rydberg gases.

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