Motion in an ultralong-range potential in cold-Rydberg-atom collisions

V. A. Nascimento, M. Reetz-Lamour,* L. L. Caliri, A. L. de Oliveira,† and L. G. Marcassa
Instituto de Física de São Carlos, Universidade de São Paulo, Caixa Postal 369, 13560-970, São Carlos-SP, Brazil
(Received 25 October 2005; published 14 March 2006)

In a recent letter Li *et al.* [Phys. Rev. Lett. **94**, 173001 (2005)] have studied experimentally the role of dipole-dipole interactions in the ionization of Rydberg atoms and the formation of a cold plasma. The authors associated their observations with atomic motion due to such interaction. In this work, we adapt the dynamical model proposed by De Oliveira *et al.* [Phys. Rev. Lett. **90**, 143002 (2003)] based on a two atom interaction, which reproduce qualitatively well some experimental results observed by Li and co-workers. This comparison presents clear evidences of motion in such ultralong range potential between cold-Rydberg atoms, which may be important for some applications, as quantum computation.

DOI: 10.1103/PhysRevA.73.034703 PACS number(s): 34.60.+z, 32.80.Pj

In the field of cold-Rydberg atoms several research topics have been investigated and discovered. Ultracold plasmas [1], many body diffusion of excitation [2], Rydberg state lifetime measurements [3], population of high-angular-momentum states through free charge [4], molecular cross-over resonances [5], and local dipole blockade [6] are among them. Besides, there are also several interesting theoretical proposals, which have not been observed so far. As an example, transport properties of ultracold gases doped with ions [7], quantum computing [8], and the creation of "trilobite" Rydberg molecules [9]. All these experiments and proposals rely on the unique Rydberg atom properties at low temperature, which have opened up new research fields.

Recently, Li *et al.* [10] have studied experimentally the role of dipole-dipole interactions in the ionization of Rydberg atoms and the formation of a cold plasma. In a particular experiment, they prepared the Rydberg atoms in the $39S_{1/2}$ state, and using a microwave frequency, they drove the transition from a $39S_{1/2}$ to a $39P_{3/2}$ state. They observed an asymmetric line shape in the ion formation spectrum; which was explained using a two-atom picture. In their picture, the atomic pair starts the collision in the $39S_{1/2}$-$39S_{1/2}$ potential curve. At a given internuclear separation, the atomic pair absorbs a microwave photon and is excited to the $39S_{1/2}$-$39P_{3/2}$ potential curve. If the photon is set to the blue of the atomic transition, the pair will be excited to a repulsive potential and the atoms will not collide. If the microwave photon is set to the red of the atomic transition, the pair will be excited to an attractive potential and the atoms will accelerate towards each other, and at short range they will ionize, presenting an asymmetrical line shape. From such a picture, the authors have concluded that ultralong-range potentials cause cold-Rydberg atoms to move; and this effect cannot be neglected for future applications [8].

The existence of such ultralong-range potentials between Rydberg atoms was proposed by Boisseau and co-workers [11]. In that paper, the authors calculated the dispersion coefficients C_5, C_6, and C_8 for two rubidium atoms considered in the same nP state. In a more recent paper, this calculation was extended to other states as well as to other atomic species [12]. Our group has also used such a calculation to interpret the results of a time resolved experiment of energy-transfer collisions using cold-rubidium-Rydberg atoms in a magneto-optical trap (MOT) [13]. In such experiment, we have monitored the exit collisional channel population as a function of time due to energy transfer collision between two Rydberg atoms in the same nP state. The semiclassical model proposed takes into account the motion of the atomic collisional pairs under the influence of an ultralong-range potential and radiative decay. The comparison between the experimental results and the model shows evidences of atomic motion in such potentials.

In this paper, we adapt the model from Ref. [13] for the experiment carried out by Li *et al.* [10]. The model allows us to predict the frequency dependence of the ion signal, and the comparison with the experimental result shows a good agreement. This comparison suggests also that the Rydberg atoms move due to the dipole interaction and that several orbits may take place before the collision ends. We start by presenting our model [13,14], followed by the discussions, and the final conclusion remarks.

To briefly describe the model, we start by supposing a pair of atoms at an internuclear separation R_0 in the presence of a microwave field of frequency ω_m and intensity I. The number of colliding atomic pairs (N_{at}) between R_0 and R_0+dR_0 excited from the $39S_{1/2}$-$39S_{1/2}$ potential curve to the $39S_{1/2}$-$39P_{3/2}$ attractive potential curve potential is proportional to

$$N_{at} \propto 4\pi R_0^2 dR_0 \varepsilon(R_0,\omega_m,I). \quad (1)$$

The first term ($4\pi R_0^2 dR_0$) accounts for the number of colliding atomic pairs at internuclear separation R_0. The second term is the excitation rate, which is given by

$$\varepsilon(R_0,\omega_m,I) = \frac{I/I_s}{1 + \frac{4}{\Gamma^2}\left(\Delta + \frac{C_3}{\hbar R_0^3}\right)^2 + I/I_s}, \quad (2)$$

where the constant C_3 characterizes the long-range part of the excited molecular attractive potential, ω_0 is the atomic

---

*Permanent address: Physikalisches Institut der Albert-Ludwigs Universität, Freiburg, Germany.

†Permanent address: Universidade do Estado de São Catarina, Joinville, SC, Brazil.

resonant frequency, Γ is the atomic spontaneous decay rate, $\Delta = \omega_m - \omega_0$ is the microwave detuning, and I_s is the saturation intensity of the transition. If we consider that the saturation broadening is small compared with the detuning, the microwave frequency will be able to excite only the atomic pairs that obey the resonance condition $\Delta = -C_3/\hbar R_0^3$. For this condition, the number of colliding atomic pairs in the excited state will be given by

$$N_{at} \propto R_0^2 \left| \frac{d\Delta}{dR_0} \right|^{-1} \propto \Delta^{-2}. \qquad (3)$$

After the excitation, the atoms will accelerate towards each other. During this motion, it is necessary to take into account the probability of the colliding pairs to survive spontaneous decay and to a small internuclear separation. At this point, it is important to define a maximum distance for the collision to produce an ion (R_{ion}). If the colliding atomic pair decays at an internuclear distance larger than R_{ion}, no ions will be produced. If the colliding atomic pair decays at an internuclear distance smaller than R_{ion}, one of the atoms will be ionized. Therefore we can define two time intervals: (i) t_0 is the classical time to go from $R=R_0$ to $R=R_{ion}$; and (ii) t_1 is the time to go from $R=R_{ion}$ to $R=0$. Therefore the probability (P_r) that a pair will decay spontaneously at an internuclear separation smaller than R_{ion} is given by

$$P_r = e^{-2\Gamma t_0}(1 - e^{-4\Gamma t_1}), \qquad (4)$$

where the factor of 2 accounts for the fact that either of the two atoms may decay. This probability (P_r) accounts only for half a vibrational cycle in this potential. To account for several vibrations of the pair in the potential, one has to rewrite Eq. (4) as [14]

$$P_r = \frac{\sinh(2\Gamma t_1)}{\sinh[2\Gamma(t_0 + t_1)]}, \qquad (5)$$

where $t_0 + t_1 \cong 0.746\sqrt{\mu R_0^5/2C_3} = (-\Delta_\tau/\Delta)^{5/6}/\Gamma$ and μ is the reduced mass. We are interested in the situation that $R_0 \gg R_{ion}$, which implies that $t_0 \gg t_1$ and that t_1 is independent of detuning. In this situation, the number of ions produced by collisions (N_{ions}) will be proportional to the product of the number of colliding atomic pairs in the excited state [Eq. (3)] and the probability of reaching a small internuclear separation [Eq. (5)], which is given by

$$N_{ions} \propto \frac{1}{\Delta^2 \sinh[2(-\Delta_\tau/\Delta)^{5/6}]}. \qquad (6)$$

If we consider only a single orbit in the excited state potential, then $N_{ions} \propto \exp[2(-\Delta_\tau/\Delta)^{5/6}]/\Delta^2$. We should point out that for large detunings, the multiple and single orbits models predict a $1/\Delta^{7/6}$ and $1/\Delta^2$ detuning dependence, respectively. In Fig. 1, we show that the experimental result from Ref. [10] presents a $1/\Delta^{1.05\pm0.05}$ dependence for large detunings, which is very close to the value predicted by the multiple orbits model. In the inset, we show also the fitting for both models, where the multiple orbits model shows better agreement with the experimental results than the single orbit model. Unfortunately, we cannot compare the parameter Δ_τ obtained from the fitting with the theoretical because the broadening of the atomic transition. Nevertheless, we believe that the multiple orbits model has the main physical insights.

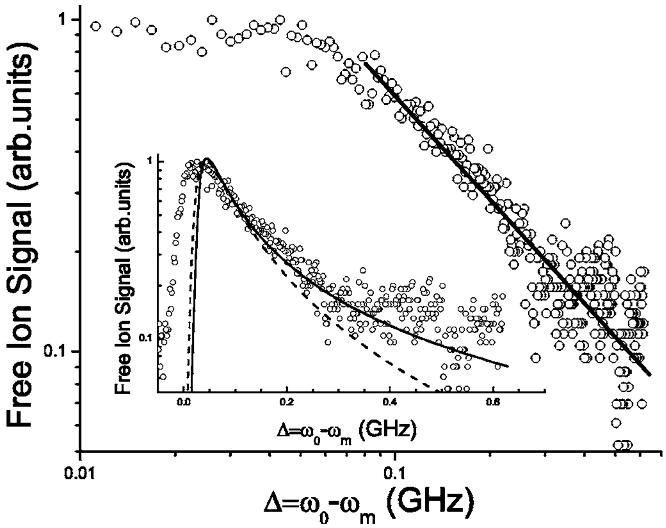

FIG. 1. Free ion spectrum as a function of the microwave frequency. For large detunings, the signal presents a $1/\Delta^{1.05\pm0.05}$ dependence. In the inset, the dashed and full lines are the model considering single and multiple orbits, respectively.

In conclusion, the results observed in Ref. [10] show clearly that the Rydberg atoms move due to the dipole interaction and several orbits may take place before the collision ends. Therefore we believe the semiclassical picture is appropriate to describe such an effect. We believe that such motion due to large internuclear forces cannot be neglected for future applications of cold-Rydberg atoms, especially involving quantum computation [8].

This work received financial support from FAPESP (Fundação de Amparo à Pesquisa do Estado de São Paulo) and Capes/DAAD.

[1] T. C. Killian, M. J. Lim, S. Kulin, R. Dumke, S. D. Bergeson, and S. L. Rolston, Phys. Rev. Lett. **86**, 3759 (2001).

[2] I. Mourachko, D. Comparat, F. de Tomasi, A. Fioretti, P. Nosbaum, V. M. Akulin, and P. Pillet, Phys. Rev. Lett. **80**, 253 (1998); W. R. Anderson, J. R. Veale, and T. F. Gallagher, *ibid.* **80**, 249 (1998).

[3] K. M. F. Magalhães, A. L. de Oliveira, R. A. D. S. Zanon, V. S. Bagnato, and L. G. Marcassa, Opt. Commun. **184**, 385 (2000); A. L. de Oliveira, M. W. Mancini, V. S. Bagnato, and L. G. Marcassa, Phys. Rev. A **65**, 031401 (2002).

[4] S. K. Dutta, D. Feldbaum, A. Walz-Flannigan, J. R. Guest, and G. Raithel, Phys. Rev. Lett. **86**, 3993 (2001).

[5] S. M. Farooqi, D. Tong, S. Krishnan, J. Stanojevic, Y. P. Zhang, J. R. Ensher, A. S. Estrin, C. Boisseau, R. Côté, E. E. Eyler, and P. L. Gould, Phys. Rev. Lett. **91**, 183002 (2003).

[6] D. Tong, S. M. Farooqi, J. Stanojevic, S. Krishnan, Y. P. Zhang, R. Côté, E. E. Eyler, and P. L. Gould, Phys. Rev. Lett. **93**, 063001 (2004); K. Singer, M. Reetz-Lamour, T. Amthor, L. G. Marcassa, and M. Weidemüller, *ibid.* **93**, 163001 (2004).

[7] R. Côté and A. Dalgarno, Phys. Rev. A **62**, 012709 (2000).

[8] D. Jaksch, J. I. Cirac, P. Zoller, S. L. Rolston, R. Côté, and M. D. Lukin, Phys. Rev. Lett. **85**, 2208 (2000).

[9] C. H. Greene, A. S. Dickinson, and H. R. Sadeghpour, Phys. Rev. Lett. **85**, 2458 (2000).

[10] W. Li, P. J. Tanner, and T. F. Gallagher, Phys. Rev. Lett. **94**, 173001 (2005).

[11] C. Boisseau, Ionel Simbotin, and Robin Côté, Phys. Rev. Lett. **88**, 133004 (2002).

[12] K. Singer, J. Stanojevic, M. Weidemüller, and R. Côté, J. Phys. B **38**, 295 (2004).

[13] R. A. D. S. Zanon, K. M. F. Magalhães, A. L. de Oliveira, and L. G. Marcassa, Phys. Rev. A **65**, 023405 (2002); A. L. de Oliveira, M. W. Mancini, V. S. Bagnato, and L. G. Marcassa, Phys. Rev. Lett. **90**, 143002 (2003).

[14] M. G. Peters, D. Hoffmann, J. D. Tobiason, and T. Walker, Phys. Rev. A **50**, R906 (1994).