Rydberg Cold Collisions Dominated by Ultralong Range Potential

A. L. de Oliveira,* M. W. Mancini, V. S. Bagnato, and L. G. Marcassa

Instituto de Física de São Carlos, Universidade de São Paulo, Caixa Postal 369, 13560-970, São Carlos, São Paulo, Brazil

(Received 13 September 2002; published 10 April 2003)

In this work we measure the time evolution of the population resulting from energy-transfer collisions as a function of the energy difference between the entrance and exit collisional channels using a sample of cold Rydberg atoms produced in a rubidium magneto-optical trap. The $34S_{1/2}$ population, produced by collisions between atoms in the $33P_{3/2}$ state, is monitored as a function of time through the pulsed-field ionization technique. The experimental results are compared with a recent published model based on a two-body interaction considering an attractive potential [Phys. Rev. A **65**, 023405 (2002)]; which is calculated according to a recent Letter by Boisseau *et al.* [Phys. Rev. Lett. **88**, 133004 (2002)]. The agreement is remarkable, which suggests the existence of such ultralong range potential proposed by Boisseau *et al.*

DOI: 10.1103/PhysRevLett.90.143002 PACS numbers: 32.80.Pj, 32.80.Rm, 34.20.Cf

In the last decade, developments for cooling and trapping neutral atoms [1] have led to several important achievements like Bose-Einstein condensation in cold trapped alkalis [2] and the high resolution spectroscopy of bound states in cold molecules [3]. More recently, such techniques were also applied to experiments involving ultracold plasmas [4], Rydberg cold collisions [5], and Rydberg lifetime measurements [6]. Thermal Rydberg atoms have been studied in depth both theoretically and experimentally [7], but at low temperature the magnitude of their properties make them excellent candidates for several experiments. In the literature there are several theoretical proposals which rely on such properties. For example, transport properties of ultracold gases doped with ions [8], quantum computing [9], and the creation of "trilobite" Rydberg molecules [10].

In a recent Letter, Boisseau and co-workers [11] propose the existence of ultralong range potential between two Rydberg atoms, as well as long range and shallow wells supporting bound states. In that Letter, the authors calculated the dispersion coefficients C_5, C_6, and C_8 for two rubidium atoms considered in the same nP state. In another recent paper, our group [12] carried out a time resolved experiment of energy-transfer collisions using cold rubidium Rydberg atoms in a magneto-optical trap (MOT). We have monitored the $31S_{1/2}$ population as a function of time originated from collisions between the $29P_{3/2}$ and $29P_{1/2,3/2}$ states. The experimental results were qualitatively reproduced by a semiclassical model [13] which took into account the dynamics of the collisional process under the influence of a $1/R^5$ potential and radiative decay. The agreement was not better due to the lack of knowledge about potential between Rydberg atoms.

In this Letter, we studied the time evolution of an energy-transfer collision using Rydberg atoms produced in a cold rubidium sample held in a MOT. This collision process is studied as a function of the energy difference between the entrance and exit collisional channels; the energy difference is tuned by applying a static electric field using the Stark effect. We compare the experimental results with our previous model [12] considering the ultralong range potential calculated according Boisseau and co-workers [11], and a very good agreement is observed. The obtained results can be considered as experimental evidence of the existence of such ultralong range potential. More specifically, we have studied the following collisional process in rubidium (Rb):

$$33P_{3/2} + 33P_{3/2} \rightarrow 33S_{1/2} + 34S_{1/2}. \quad (1)$$

The electric field lifts the degeneracy of the azimuthal orbital angular momentum quantum number (m). There are three possible collisional channels involving such states: (i) $|m_j| = 1/2 + 1/2$ (around 8 V/cm), (ii) $|m_j| = 1/2 + 3/2$ (around 8.5 V/cm), and (iii) $|m_j| = 3/2 + 3/2$ (around 9 V/cm) where the voltages in parentheses represent the necessary Stark shift to tune each channel. For simplicity we have restricted ourselves to $|m_j| = 1/2 + 3/2$. In Fig. 1, we show the energy difference between the entrance channel ($33P_{3/2} + 33P_{3/2}$) and the exit channel ($34S_{1/2} + 33S_{1/2}$) for $|m_j| = 1/2 + 3/2$ [14]. From Fig. 1 and energy consideration, it is clear that the collisional channel for $|m_j| = 1/2 + 3/2$ is open for an electric field below 8.5 V/cm and closed for values above it.

The details about our experimental setup and detection technique are described elsewhere [12]. In Fig. 2 we show the time evolution of the $34S_{1/2}$ state population for three values of static electric field, 8.19, 8.36, and 8.48 V/cm. Clearly, the time evolution depends on the static electric field or, in other words, on the energy difference between the entrance and exit collisional channels. This fact was not observed in our previous experiment [12]. It is also clear that as we approach the resonance (the energy difference equals zero) the peak in the time evolution

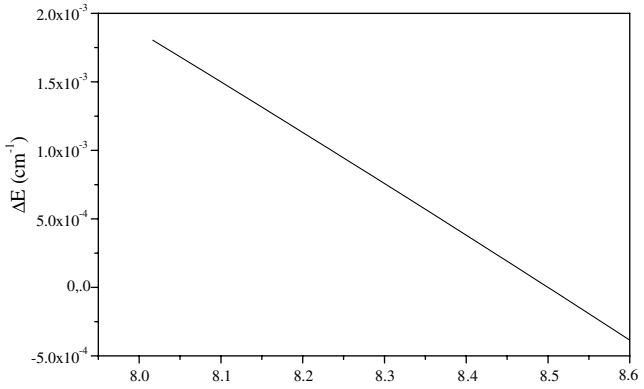

FIG. 1. Energy difference between the entrance $33P_{3/2} + 33P_{3/2}$ and exit $34S_{1/2} + 33S_{1/2}$ channels, for $|m_j| = 1/2 + 3/2$.

shifts towards longer times. In the inset of Fig. 2 we show the $34S_{1/2}$ state population as a function of the static electric field, for a delay time of 2 μs. According to Fig. 1, the collisional channel should not take place for an electric field larger than 8.5 V/cm, because above this value the energy difference would be negative. However, we observe that there is ion signal above 8.5 V/cm. The process still may be possible due to the fact that the atoms have an initial velocity. The energy difference from the electric field of 8.5 to 8.6 V/cm is on the order of 4×10^{-4} cm^{-1}, which is consistent with atoms at 400 μK.

To explain such behavior we have used the model presented in our previous work [12], which is based on a binary collision and is adapted from the Gallagher-Pritchard model for cold collisions [13]. However, we have considered the potential for the $33P_{3/2} + 33P_{3/2}$ channel calculated by Boisseau and co-workers [11]. Because of the relative importance of the terms in the

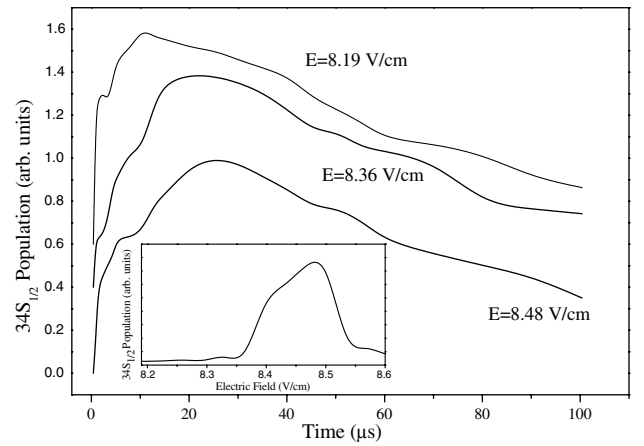

FIG. 2. Time evolution of the population in the $34S_{1/2}$ state, for three fixed values of electric field: 8.19, 8.36, and 8.48 V/cm. In the inset, the $34S_{1/2}$ state population is shown as a function of the static electric field, for a delay time of 2 μs.

potential, we will consider here only the C_5 and C_6 coefficients and neglect the C_8 coefficient. Under this potential the atoms are classically accelerated towards each other until they reach an internuclear separation where the attractive potential curve, which asymptotically connected to $33P_{3/2} + 33P_{3/2}$, crosses the one connected to $33S_{1/2} + 34S_{1/2}$. At this internuclear separation, the atomic pair may change potential, ending the collision process in the $33S_{1/2} + 34S_{1/2}$ state, and the $34S_{1/2}$ population is detected. During this collision, spontaneous decay may happen and shall be considered here. To calculate the time evolution of the $34S_{1/2}$ state population, we also need to know the potential for the $34S_{1/2} + 33S_{1/2}$ channel. This potential will be calculated in the near future [15], and here we will consider it flat.

As explained in Ref. [12], the first step in the model is to calculate the density of colliding pairs (ΔN) present at an internuclear separation between R_0 and $R_0 + dR_0$. These pairs may reach the crossing point, where the potential change takes place, between t and $t + dt$. The time (t) necessary for a pair to evolve from $R = R_0$ to $R = R_c$ (R_c is the internuclear separation where the potentials cross each other) with an initial velocity equal to zero is given by

$$t = \int_{R_0}^{R_c} \frac{\sqrt{\mu/2}dR}{\sqrt{C_5(\frac{1}{R^5} - \frac{1}{R_0^5}) + C_6(\frac{1}{R^6} - \frac{1}{R_0^6})}}, \qquad (2)$$

where μ is the reduced mass of the atomic pair. We have verified that for $t > 1$ μs Eq. (2) can be approximated by

$$t = \frac{B(1/2, 7/10)}{5}\left(\frac{\mu R_0^7}{2C_5}\right)^{1/2}, \qquad (3)$$

where $B(z, w)$ is the beta function [16]. It is important to point out that our measurements are for $t > 2$ μs. And for the considered potential R_c varies from $9 \times 10^3 a_0$ to $20 \times 10^3 a_0$, and the atoms for which the travel time is larger than 2 μs are separated for more than $30 \times 10^3 a_0$. Next we have to take into account the probability of the colliding pair to survive spontaneous decay and to reach the short range part of the potential. At the crossing point, the pair has a probability q to change the potential curve. To account for several oscillations of the pair in the attractive potential before the pair decays or changes potential, one has the probability (P_r) that a pair will survive spontaneous decay and change its potential as

$$P_r = \frac{e^{-2t/\tau}q}{1 - (1 - q)e^{-4t/\tau}}, \qquad (4)$$

where τ is the lifetime of the initial state, while the factor of 2 accounts for the fact that either of the two atoms may decay.

Finally, to account for the atomic pairs that change potential for $t < \tau_1$, where τ_1 is an arbitrary time, one

must integrate over time from $t = 0$ to $t = \tau_1$. To account for the fraction of the population which changed potential at time t and survived spontaneous decay in the $34S_{1/2}$ state until $t = \tau_1$ we add the term $e^{-(\tau_1 - t)/\tau'}$. Therefore, the total number of pairs in the $33S_{1/2} + 34S_{1/2}$ potential as a function of the delay τ_1 is given by

$$N(\tau_1) = \frac{4\pi}{7} n^2 \left(\frac{2C_5}{\mu}\right)^{3/7} \left(\frac{5}{B(1/2, 7/10)}\right)^{6/7} \times \int_0^{\tau_1} t^{-1/7} \frac{e^{-2t/\tau} q}{1 - (1 - q)e^{-4t/\tau}} e^{-(\tau_1 - t)/\tau'} dt, \quad (5)$$

where τ' is the lifetime of the $34S_{1/2}$ state.

We have fitted our results with Eq. (5), considering the measured values of the lifetime for the $33P$ and $34S$ states [6] and q as a free parameter. Figure 3 shows the theoretical curve (dashed line) predicted by Eq. (5) together with the experimental results (solid line) for an electric field of 8.5 V/cm. In the inset, we show the variation of q as a function of the electric field, which shows that as we approach $\Delta E = 0$, q increases approaching 0.5. This fact is consistent with the fact that in Fig. 2 the maximum in the population signal shifts to larger times as we move towards $\Delta E = 0$. This can be explained as follows: If q is small, it is necessary that the atomic pairs undergo several oscillations before changing potential. Only atoms that are at small internuclear separation can oscillate enough times before spontaneous decay happens. The atomic pair far apart from the crossing region decay before undergoing the necessary oscillations. Therefore, only atomic pairs at small internuclear separation can contribute to the total signal, and the peak in the population is located at small time values. As q increases, the contribution of the atoms that are at long internuclear separation increases also and the peak shifts toward greater delay time values. We should point out that the multiple oscillations are essential to explain the time evolution of the collision as a function of the electric field. The parameter q may be calculated using the Landau-Zener model [17]. According to this model, the probability of changing potential is given by

$$q = 2e^{-A}(1 - e^{-A}), \quad (6)$$

where

$$A = \frac{2\pi^2 \Omega^2}{h v |\frac{dV}{dR}|_{R=R_C}}. \quad (7)$$

Here Ω is the coupling between the molecular potentials, v is the atomic velocity at the crossing, and dV/dR is the slope of the difference between the potentials. To calculate q one has to know precisely the potentials at short range in order to calculate Ω, v, and dV/dR. In order to find out if the model used is consistent with the obtained values for q; we consider two approximations: (i) We will consider the potential for the $34S_{1/2} + 33S_{1/2}$ channel as flat, and the potential for $33P_{3/2} + 33P_{3/2}$ as calculated by Côté [15]. Under these considerations we can calculate the velocity and the potential slope at the crossing point. (ii) The coupling between the potentials (Ω) will be considered as $\Omega = \Delta E/D$ (where D is a fitting parameter) [15,18]. In Fig. 4 we show the fitting of q using Eqs. (6) and (7). From the fitting we obtain that $D \simeq 2.9$, producing a coupling which corresponds to a fraction of the energy difference between the asymptotic potentials. This situation is also true in other collisional processes such as fine structure changing, and it is expected to apply to our case as well [15,18]. We believe that if the correct shape for the $34S_{1/2} + 33S_{1/2}$ potential, the actual coupling, and the initial velocity of the atoms were included in the model, a better agreement between theory

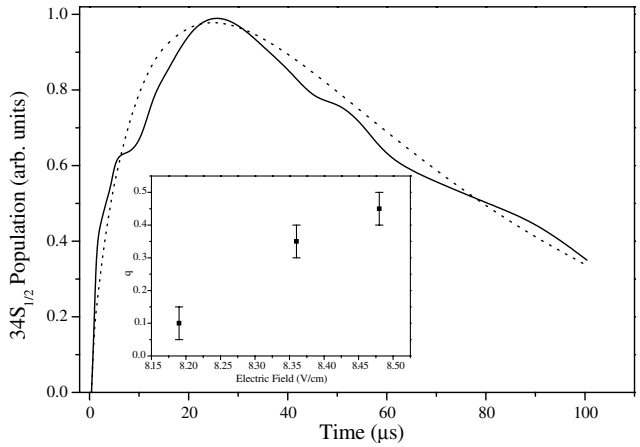

FIG. 3. Time evolution of the population in the $34S_{1/2}$ state for electric field $V = 8.48$ V/cm. The dashed line represents the theoretical prediction, and the solid line corresponds to the experimental results. Inset: Behavior of the probability of changing potential curve q, with the electric field. For $\Delta E = 0$, q approaches to 0.5.

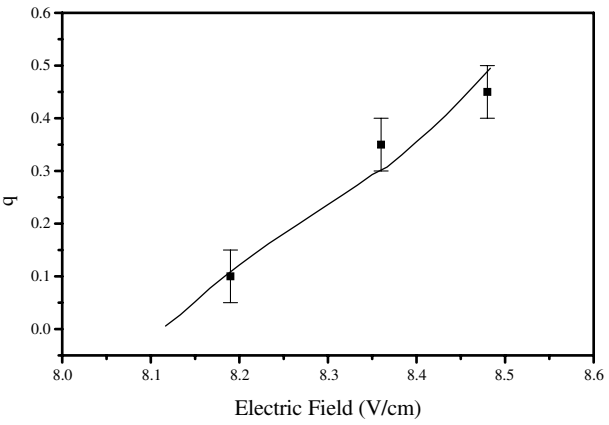

FIG. 4. Probability q, as a function of the electric field near the resonances at electric field values corresponding to channels involving the states $|m_j| = 1/2 + 1/2$ (around 8 V/cm), and $|m_j| = 1/2 + 3/2$ (around 8.5 V/cm). The solid line is the fitting using Eqs. (6) and (7).

and experiment would be observed, and we would be able to reproduce the dependence of the $34S_{1/2}$ population as a function of electric field. In spite of its simplicity, considering only one channel, the model describes all the important physical features of the collisional process. However, we should point out that more channels will contribute to the final products. There will be several crossing points due to the existence of several potential curves and contribution from several partial waves. In principle, we could treat each channel independently, and each channel would contribute to a specific value for q. The time evolution of each channel would be described by Eq. (6) with its particular q. As seen in our results each q will shift the maximum of the time evolution of the products by a bit, but its general behavior will be the same. The final contribution including all channels would be a time evolution with several q's, which can be replaced by an effective value for q.

In summary, we measured the time evolution of the energy-transfer collision involving cold Rydberg atoms in a sample of trapped ^{85}Rb atoms as a function of the energy difference between the entrance and exit channels of the collisional process. The experimental results were compared to a dynamic model considering the potential calculated in a recent Letter. The results present a behavior which is consistent with the existence of an ultralong range potential. This model is able to reproduce well our experimental data and may provide an alternative explanation for the broad energy width observed in our experiment and in Refs. [5], without invoking many body effects, rather considering only the ultralong potentials and the finite temperature of the atoms. We hope this work will stimulate further theoretical research in this area.

This work received financial support from FAPESP (Fundação de Amparo à Pesquisa do Estado de São Paulo) and Pronex (Programa de Núcleos de Excelência em Óptica Básica e Aplicada). This work was carried out at the Center for Research in Optics and Photonics-CePOF.

*Permanent address: Departamento de Física, Universidade do Estado de Santa Catarina.

[1] J. Weiner, V. S. Bagnato, S. C. Zilio, and P. S. Julienne, Rev. Mod. Phys. **71**, 1 (1999).
[2] M. H. Anderson *et al.*, Science **269**, 198 (1995); K. B. Davis *et al.*, Phys. Rev. Lett. **75**, 3969 (1995); C. C. Bradley, C. A. Sackett, J. J. Tollett, and R. G. Hulet, Phys. Rev. Lett. **75**, 1687 (1995).
[3] F. Fioretti *et al.*, Phys. Rev. Lett. **80**, 4402 (1998); T. Takekoshi, B. M. Patterson, and R. J. Knize, Phys. Rev. Lett. **81**, 5105 (1998); A. N. Nikolov *et al.*, Phys. Rev. Lett. **82**, 703 (1999); C. Gabanini *et al.*, Phys. Rev. Lett. **84**, 2814 (2000).
[4] T. C. Killian *et al.*, Phys. Rev. Lett. **86**, 3759 (2001).
[5] I. Mourachko *et al.*, Phys. Rev. Lett. **80**, 253 (1998); W. R. Anderson, J. R. Veale, and T. F. Gallagher, Phys. Rev. Lett. **80**, 249 (1998).
[6] K. M. F. Magalhães *et al.*, 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).
[7] T. F. Gallagher, *Rydberg Atoms* (Cambridge University Press, Cambridge, 1994).
[8] R. Côté and A. Dalgarno, Phys. Rev. A **62**, 012709 (2000).
[9] D. Jaksch *et al.*, Phys. Rev. Lett. **85**, 2208 (2000).
[10] C. H Greene, A. S. Dickinson, and H. R. Sadeghpour, Phys. Rev. Lett. **85**, 2458 (2000).
[11] C. Boisseau, Ionel Simbotin, and Robin Côté, Phys. Rev. Lett. **88**, 133004 (2002).
[12] R. A. D. S. Zanon, K. M. F. Magalhães, A. L. de Oliveira, and L. G. Marcassa, Phys. Rev. A **65**, 023405 (2002).
[13] A. Gallagher and D. E. Pritchard, Phys. Rev. Lett. **63**, 957 (1989).
[14] M. L. Zimmerman, M. G. Littman, M. M. Kash, and D. Kleppner, Phys. Rev. A **20,** 2251 (1979).
[15] R. Cotê (private communication).
[16] *Handbook of Mathematical Functions With Formulas, Graphs, and Mathematical Tables*, edited by M. Abramowitz and I. Stegun (National Bureau of Standards, Washington, DC, 1964), 3rd ed., p. 258, 6.2.1 subsection.
[17] C. N. Zener, Proc. R. Soc. London A **137**, 696 (1932).
[18] R. Napolitano (private communication).