

Many-Body Effects in a Frozen Rydberg Gas

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We studied the properties of a cold ($\sim 100 \mu\text{K}$) and dense ($\sim 10^{8-10} \text{ cm}^{-3}$) atomic Rydberg Cs gas, and found that the observed widths and shapes of resonances in population transfers cannot be explained in the framework of a usual gas model. We propose a “frozen Rydberg gas” model, where the interplay between two-body and many-body phenomena affects in an unexpected way the width and the shape of spectral lines. [S0031-9007(97)04903-X]

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Recent demonstrations of Bose-Einstein condensation have pointed out the role of collisional processes in the evaporative cooling method [1–3]. Relatively dense cold atomic samples have revealed, in turn, a rich variety of new phenomena in atomic collision [4], and in particular a strong excitation exchange effect. Usually one expands the physical characteristics of a gas in the power series in density that corresponds to one-, two-, three-, etc., body phenomena. The excitation exchange is governed by the two-body interactions, whereas the many-body effects in sparse systems are usually considered as small. But does such a gas at low temperature and density remain a sparse system? In this Letter we show that the situation may change completely for an ensemble of ultracold atoms when the typical collision time considerably exceeds the time corresponding to the inverse of typical interaction energy between two neighboring atoms. In this regime the many-body phenomena play an equally important role. Observation of such phenomena becomes possible due to the recent development of a laser cooling technique, which brings new tools for atomic and molecular physics.

We report the first results of experiments performed with cold cesium atoms in Rydberg states along with a theoretical model and show how the interplay between two-body and many-body processes affects, in an unexpected way, the width and the shape of the resonances in the population transfer induced by the energy exchange. The fact that the excited Cs atoms obtained by pulsed laser irradiation are slow and are excited to high Rydberg states plays the crucial role: the large size, the large dipole moment, and the long lifetime of the Rydberg atoms allow one to reach experimental conditions where many-body phenomena become important.

The atomic sample contains $N_d \sim 10^8 - 10^{10}$ highly excited Cs atoms per cm^3 at a temperature $T \sim 100 \mu\text{K}$ in a chosen Rydberg p -state with the principal quantum number $n = 20 - 30$. The typical displacement $\Delta R = v\tau \sim 10 - 300 \text{ nm}$ of atoms moving at an average velocity $v \sim 10 \text{ cm/s}$ during the experiment time of $\tau \sim 0.1 - 3 \mu\text{s}$ is much less than the average distance $\bar{R} \sim (3/4\pi N_d)^{1/3} = 5 - 20 \mu\text{m}$ between neighboring atoms. It is of the same order of magnitude as the size of the Rydberg orbits $R_0 \approx 4n^2 \text{ a.u.} \sim 80 \text{ nm}$ (for $n \sim 20$), whereas the nuclear

de Broglie wavelength is about $\lambda_{dB} \sim 30 \text{ nm}$. It means that one can ignore completely the motion of atoms and consider the atomic ensemble as a “frozen Rydberg gas” [5].

We trace the number of atoms in s states created in the energy exchange process [7]

$$\text{Cs}_A[np_{3/2}] + \text{Cs}_B[np_{3/2}] \rightarrow \text{Cs}_A[ns] + \text{Cs}_B[(n+1)s], \quad (1)$$

when one of the atoms (A) makes a downward transition from the Rydberg state $|np_{3/2}\rangle$ to a lower Rydberg state $|ns_{1/2}\rangle$, whereas the other atom (B) makes an upward transition $|np_{3/2}\rangle \rightarrow |(n+1)s_{1/2}\rangle$. For each of the atoms it corresponds to an allowed dipole transition, and hence the typical interaction $V_{AB} = \mu_A \mu_B / R_{AB}^3$ depends on the matrix elements μ_A and μ_B of the dipole moment and the distance R_{AB} between the atoms. The reaction of Eq. (1) is tuned into resonance by a Stark shift of the $p_{3/2}$ state in the static electric field \mathcal{E} , as is shown in the level scheme in Fig. 1(a). Aside from the creation of ss' couple, that is, two s states in Eq. (1), the processes of excitation exchange with other atoms (C, D , etc.)

$$\begin{aligned} \text{Cs}_C[np_{3/2}] + \text{Cs}_A[ns] &\rightarrow \text{Cs}_C[ns] + \text{Cs}_A[np_{3/2}] \\ \text{Cs}_D[np_{3/2}] + \text{Cs}_B[(n+1)s] &\rightarrow \text{Cs}_D[(n+1)s] \\ &+ \text{Cs}_B[np_{3/2}] \end{aligned} \quad (2)$$

are also possible. They are always resonant and have the same order of magnitude as the process in Eq. (1) but do not create new ss' couples. They allow for the migration of s states over the “frozen” atoms. We therefore have to consider a novel quantum system of Rydberg atoms at rest, which evolves in the course of the energy exchange processes Eqs. (1) and (2). One can say in the spirit of the concept of quasiparticles that the ss' couples are created and that each of the s states independently moves over a media of a frozen Rydberg gas. This system resembles an amorphous glass, where the centers are randomly distributed over the sample.

The basic scheme of the experiment is as follows. The cold Cs atoms are produced in a vapor-loaded MOT cell [8] created at the intersection of three pairs of mutually orthogonal, counterpropagating $\sigma^+ - \sigma^-$ -laser

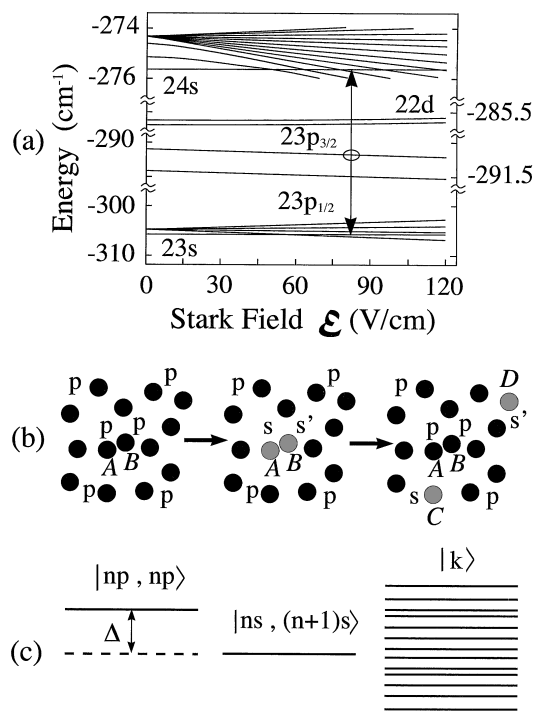


FIG. 1. Level diagram (a) for an isolated Cs atom in an electric field \mathcal{E} for the principal quantum number $n = 23$. Rydberg states s , p , and d are shown along with the adjacent Stark manifold. The avoided crossings of the levels and manifolds are located far enough from the resonances to exclude any influence (for details see Ref. [6]). The splitting of the $|m| = 1/2$ and $|m| = 3/2$ states of the $np_{3/2}$ level, which is of the order of 1.2 GHz, is not shown. Illustration (b) of the elementary act of excitation exchange process which includes creation of an ss' couple at a pair of closely spaced Rydberg atoms ($A + B$) and subsequent evacuation of these states to atoms C , D , etc. at longer distances, and the corresponding level scheme (c) for the pair ($A + B$) embedded in the system of other atoms (OA). We denote the eigenstate of OA projected to the Hilbert subspace of a single ss' couple by the symbol $|k\rangle$. Quantum transitions to this band models the diffusion of s states (compare to Ref. [11]).

beams (4 mW/cm^2), at the zero magnetic field point of anti-Helmholtz coils device. The residual pressure is 2×10^{-9} Torr. The cooling laser beams are split from a single mode laser beam (SDL 5412-H1, 100 mW, $\lambda \sim 852 \text{ nm}$) injection locked to a master diode laser. The master laser (SDL 5412-H1, 100 mW) is stabilized by optical feedback from an extended grating-ended cavity. Long-term stabilization is ensured by locking the laser frequency to a saturated absorption line in a cesium vapor cell. The laser frequency is tuned about 1.5 natural linewidths below the $6^2S_{1/2}(F = 4) \rightarrow 6^2P_{3/2}(F' = 5)$ transition. A repumping laser beam (SDL 5712-H1, 100 mW) tuned to the $6^2S_{1/2}(F = 3) \rightarrow 6^2P_{3/2}(F' = 4)$ transition is superimposed on two of the cooling laser beams. Under these conditions, the FWHM dimension of the cold sample is in the range of $600\text{--}1000 \mu\text{m}$, and the number of atoms in the trap is between 1×10^7 and 5×10^7 , leading thus to a density of up to 10^{11} cm^{-3} .

At the trap position, a static electric field $\mathcal{E} = 30\text{--}100 \text{ V/cm}$ and a pulsed high voltage field can be applied by means of a pair of electric field grids spaced by 15 mm. The $6P_{3/2}$ atoms are excited in the presence of static field by a pulsed (7 ns, $<1 \text{ mJ}$) dye laser running at 10 Hz repetition rate, which is pumped by the third harmonic of a Nd-YAG laser and tuned to the transition $6P_{3/2} \rightarrow np_{3/2}$ ($n = 22\text{--}26$, $\lambda \sim 518 \text{ nm}$). This excitation does not significantly change the atomic velocity. After the excitation the atoms evolve during about $2 \mu\text{s}$, and when a quasi-steady-state regime attains, we apply a high voltage pulse of a 300 ns risetime and selectively ionize the upper s state. The ions are expelled out of the interaction region and detected by a pair of microchannel plates. The signal from the ion detector is recorded with a gated integrator. By slowly sweeping the field \mathcal{E} we chirp the detuning of the reaction in Eq. (1) and observe ion peaks corresponding to resonances of different magnetic components m of the initial $np_{3/2}$ state—either $|m| = 1/2$ or $|m| = 3/2$. Control of the Rydberg atom density is performed by attenuating the dye laser.

In Fig. 2 we show typical resonance lines for the transition of Eq. (1). One sees that the linewidth and the line shape changes with the density of Rydberg atoms. Indeed, in Fig. 2(a) one sees a narrow peak with a linewidth of 35 MHz FWHM, which is close to the resolution limit of $\sim 20 \text{ MHz}$. At higher densities Figs. 2(b)–2(d), a progressive broadening of the lines up to 200 MHz is evident. One clearly sees the transformation of the line shape with

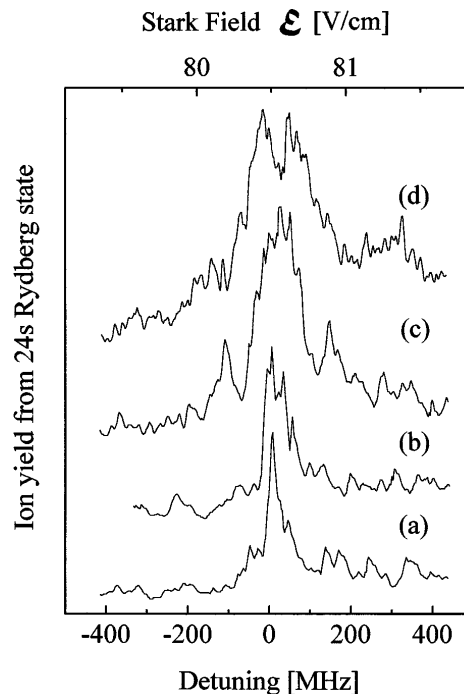


FIG. 2. Energy transfer resonance Eq. (1) for $23p_{3/2}$ state with $|m| = 1/2$. The ionic yield for the $24s_{1/2}$ state is shown as a function of the detuning controlled by \mathcal{E} field. Estimated densities of Cs[$23p_{3/2}, |m| = 1/2$] atoms: (a) $4 \times 10^8 \text{ cm}^{-3}$, (b) $7 \times 10^8 \text{ cm}^{-3}$, (c) $4 \times 10^9 \text{ cm}^{-3}$, and (d) 10^{10} cm^{-3} .

the density: At the highest densities the resonance is split in two peaks and at intermediate densities small side bands around the resonance peak can be seen. This result is totally different from the case of two-body resonant collisions in a dilute gas for the regime with mean free time exceeding the time of observation, where the linewidth and the line shape do not depend on the atomic density N_d and the peak maximum grows as N_d^2 . Hence, the observed density dependence of the lines is a signature of many-body effects.

There is another interesting feature: one may expect the linewidth to be of the order of typical size of the interaction $V = \mu_A \mu_B \bar{R}^{-3}$, where $\bar{R} = n^{-1/3}$ is the mean distance between neighboring Rydberg atoms. However, the observed lines are much larger. In Fig. 2(d) one sees that at the density $N_d = 10^{10} \text{ cm}^{-3}$ the observed linewidth $\delta \sim 200 \text{ MHz}$ while the mean interaction $V = 1.5 \text{ MHz}$ is 2 orders of magnitude smaller. One could assume that such broad lines result from the rare fluctuations corresponding to pairs of atoms at small separations $R_\delta \sim \bar{R}(V/\delta)^{1/3} \ll \bar{R}$. The estimate $N_d R_\delta^3$ shows that such pairs represent at most 1% of the ensemble. Although this assumption may give the correct linewidth, it will explain neither the observed ionic yield $\sim 10\%$ nor the transformation of the line shape. Analysis of other phenomena, such as the inhomogeneity of the electric or magnetic fields, the presence of photoions [9], and black body radiation, shows that they do not give any line broadening effects larger than 20 MHz, and no significant population transfer due to the Dicke superradiance has been detected [10]. We thus have encountered a novel situation where the line shapes of the frozen Rydberg gas depend on the density, while the linewidth is much larger than that suggested by the mean distance. Note that this situation is not typical of just our particular system, since similar phenomena have been observed for rubidium atoms by Anderson *et al.* [11].

The interpretation of the results relies on the following model. We assume that the observed broad lines indeed appear due to a relatively small number of pairs of close atoms that perform the rapid excitation exchange of Eq. (1). However, each pair is an open system embedded in a larger system of other Rydberg atoms [see Fig. 1(b)], and the slow processes in Eq. (2) allow for the diffusion of ss' couple out of the closely spaced pair of atoms. Each of these pairs may therefore undergo the reaction several times. The process has some analogy to autocatalytic processes in chemistry: the fluctuations are responsible for the ignition of the reaction and the diffusion evacuates the reaction products. Although the diffusion is a slow process in comparison to the interaction between the atoms of the pair, its contribution to the evolution of system is crucial since it acts as a "bottle neck" for the evacuation of s states out of the pairs, thus affecting the width and the line shape of the resonance energy exchange, Eq. (1).

A consistent description of the open system can rely neither on the Schrödinger equation nor on the standard density matrix approach with exponential relaxation, since

the diffusive evacuation is nonexponential: the probability $p(t)$ for the ss' couple to remain at the pair of close atoms after time t in the course of a d -dimensional diffusion has a power-law character $p(t) \sim (Dt)^{-d/2}$, with the diffusion coefficient D dependent on the density of Rydberg atoms. However, one still can assume the Markovian character of the process and separate the problem into two parts, first, by considering the unitary evolution of a single ss' couple and finding the probability of the elementary act of its creation followed by the diffusive evacuation, and, second, by describing the result of multiple repetition of this elementary act which yields many ss' couples.

The level scheme in Fig. 1(c) illustrates the elementary act. The energy exchange in the pair of atoms $A + B$ leads to an interaction of the initial state $|np, np\rangle$ with the state $|ns, (n+1)s\rangle$, whereas the diffusion of ss' couple is described as a transition to a band. As the only essential variable which describes the diffusion, we take the probability amplitude $G(t - t_1) = [p(t - t_1)]^{1/2} = D^2 [D(t - t_1)]^{-d/4}$ for the ss' couple to return at the pair at time t , after it has left this pair at the time t_1 [12]. The corresponding Schrödinger equations reads

$$i\hbar \dot{\psi}_{pp}(t) = \Delta \psi_{pp}(t) + \frac{\mu_A \mu_B}{R_{AB}^3} \psi_{ss'}(t), \quad (3)$$

$$i\hbar \dot{\psi}_{ss'}(t) = \frac{\mu_A \mu_B}{R_{AB}^3} \psi_{pp}(t) + \int_{-\infty}^t G(t - t_1) \psi_{ss'}(t_1) dt_1,$$

where ψ_{pp} and $\psi_{ss'}$ are the amplitudes of states $|np, np\rangle$ and $|ns, (n+1)s\rangle$, respectively, and Δ is \mathcal{E} -field controlled detuning between these states. Equation (3) can be solved by the standard Fourier-transform method, which yields the probability $\rho(t, \Delta) = |\psi_{pp}(t, \Delta)|^2$ for atoms A and B to remain at their initial states after the time t .

At the next step one finds from the probability $\rho(t, \Delta)$ the number of elementary acts $N(t, \Delta)$ that took place during the time t . Indeed, the probability to have no s states up to the time t , given by $\rho(t)$, does not contribute to the total yield $N(t, \Delta)$. The probability $-\dot{\rho}(t_1) dt_1$ to create one ss' couple at time t_1 during the time interval dt_1 multiplied by the probability to have no ss' couples generated later on gives after the integration over dt_1 the probability $P_1(t) = -\int dt_1 \dot{\rho}(t_1) \rho(t - t_1)$ to have at time t only one ss' couple. Analogous expressions, $P_k(t) = (-1)^k \int dt_1 \dots dt_k \dot{\rho}(t_1) \dots \dot{\rho}(t_k) \rho(t - \sum_{m=1}^k t_m)$, can be written for the probabilities of creation of k ss' couples. One finds the total number of ss' couples $N(t, \Delta) = \sum_k k P_k(t)$ by Fourier transform of $P_k(t)$ and summation of the algebraic series. For asymptotically long times it yields

$$N(t, \Delta) \approx \frac{1}{T(\Delta)} t, \quad (4)$$

where $T(\Delta) = \int_{-\infty}^{\infty} \rho(t, \Delta) dt$ represents the lifetime of the initial state.

For further calculations one needs to choose the spatial dimension d of the diffusion process Eq. (2). It is a

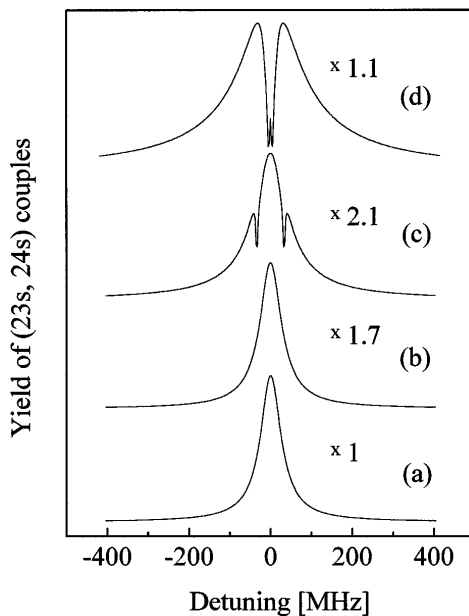


FIG. 3. Theoretical ionic yield rate given by $1/T(\Delta)$ in Eq. (4) at densities (a) $3.8 \times 10^8 \text{ cm}^{-3}$, (b) $7 \times 10^8 \text{ cm}^{-3}$, (c) $3.6 \times 10^9 \text{ cm}^{-3}$, and (d) 10^{10} cm^{-3} .

delicate question which seems to be an interesting physical problem by itself. On the one hand, it is not evident *a priori* that this diffusion is three-dimensional since for the long-range interaction $\sim 1/R^3$ the close and the far neighbors are equally important [12]. On the other hand, for a random atomic distribution the sets of closest atoms may be arranged along random complex-shaped curves or surfaces in $3 - D$ space, given the diffusion occurs among the closest atoms. In the more general situation the dimensionality of these manifolds may even be fractal. To be specific, in this Letter we have chosen $d = 4$. Thereby the $2 \times 2 - D$ diffusion is assumed, which implies that each of the two s states created in the elementary act of Eq. (1) moves independently over a $2 - D$ manifold. There is an excuse for such a random choice: the results are qualitatively similar for other d . In Fig. 3, we show the profiles calculated for different densities of Rydberg atoms for $R_{AB} = 0.9 \mu\text{m}$. One sees the variation of the linewidth with the density, which agrees with the experiment. Moreover, at high densities the calculated spectra show features similar to the experimental ones.

We conclude by summarizing the main results. Coherent evolution of a mesoscopic system of $10^3 - 10^6$ cold Rydberg atoms does not conform to the usual gas model. Peculiarities of the energy exchange in such a system find their explanation in the framework of a frozen gas model, where rare fluctuations of the interatomic distances are responsible for the linewidth, whereas a weak many-body effect of the diffusion of quantum states governs the line shape. Future experiments should check directly the role of the diffusion process and trace the crossover from the collisional regime to the frozen gas regime. Demonstration of such phenomena would be of great importance

for the spectroscopy of ultracold plasmas, metrology, and trapping experiments [13]. One can also note that similar regimes could be of importance in other Rydberg systems, as ZEKE spectroscopy [14], for instance.

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