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Spectral linewidth broadening from pair fluctuations in a frozen Rydberg gas

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Spectral linewidth broadening in Rydberg gases, a phenomenon previously attributed to the many-body effect, was observed experimentally almost a decade ago. The observed linewidth was typically 80–100 times larger than the average interaction strength predicted from a binary interaction. The interpretation of such a phenomenon is usually based on the so-called diffusion model, where the linewidth broadening mostly originates from the diffusion of excitations. We present a model calculation to show that diffusion is not the main mechanism of the linewidth broadening. We find that the rare pair fluctuation at small separation is the dominant factor contributing to this broadening. Our results give a width of about 20–30 times larger than the average interaction strength. More importantly, by turning off the diffusion process, we do not observe an order-of-magnitude change in the spectral linewidth.

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Rydberg gases have attracted renewed interest in recent years due to the unprecedented advancement in laser cooling and trapping [1–9]. Rydberg atoms, possessing a large dipole moment and long lifetime, can interact with each other coherently for relatively long times, which make them a potential candidate for quantum information processing [10,11]. There have been many experiments exploring the quantum many-body effects in Rydberg gases—e.g., spectral linewidth broadening [1–3], number correlation [6,12,13], and collective excitation [8]. Many experimental results can be understood from the well-known dipole blockade effect: when two Rydberg atoms are close enough, the dipolar interaction will shift them out of resonance with the external driving laser; thus, double excitation is greatly suppressed.

In this paper, we are interested in the unusual linewidth broadening which was observed in experiments [1,2]. To be specific, we will consider the following two cases. (I) The main process is $|np\rangle+|np\rangle\leftrightarrow|ns\rangle+|(n+1)s\rangle$ for experiment [1], where the principal quantum number n is 23 and the maximal gas density is around 10^{10} cm^{-3}. np, ns, and $(n+1)s$ are abbreviated as p, s, and s', respectively. (II) The main process is $|(n+1)s\rangle+|n's\rangle\leftrightarrow|np\rangle+|(n'+1)p\rangle$ for experiment [2], where the principal quantum numbers n and n' are 24 and 33, respectively, and the maximal gas density is around 10^{9} cm^{-3} for each of the s states. $(n+1)s$, $n's$, np, and $(n'+1)p$ are abbreviated as s, s', p, and p', respectively. For both cases, they express the creation process; e.g., in case (I), one atom makes a downward transition from the Rydberg state $|p\rangle$ to $|s\rangle$, and the other atom makes an upward transition from $|s'\rangle$ to $|p\rangle$—that is to say, creating ss' from a pp pair. The detuning between $|pp\rangle$ and $|ss'\rangle$ is controlled by a static electric field, and the transition is allowed with dominant dipole moments μ_{ps} and $\mu_{ps'}$. Here $\mu_{\alpha\beta}$ denotes the transition dipole moment between states $|\alpha\rangle$ and $|\beta\rangle$. Similar notations will apply to case (II). In addition to the above creation processes, there also exist the exchange process—e.g., $|p\rangle+|s\rangle\leftrightarrow|s\rangle+|p\rangle$. Different from the creation process, the exchange process is always resonant and it describes the hopping of excitation in the whole gas. For this reason, we will also call it a diffusion process.

A rough estimate from the binary interaction will give a linewidth of the order of the average interaction strength, $\bar{V}_1=\mu_{ps}\mu_{ps'}n$ or $\bar{V}_2=\mu_{ps}\mu_{p's'}n$ for the two cases, respectively, with n the average density of the gas. However, the experimentally observed linewidths are typically $\sim 100\bar{V}_1$ for the first case and are $\sim 80\bar{V}_2$ for the second case. Previous explanations are based on a diffusion model, where resonant processes like $|p\rangle+|s\rangle\leftrightarrow|s\rangle+|p\rangle$ form a diffusion band and play a dominant role in the broadening. In this model, even at large detuning, there are still some pairs of atoms close enough to perform the creation process. Because the hopping of excitation can happen in the whole gas, the diffusion evacuates the excitations so that each pair of close atoms can react several times, analogous to autocatalytic processes in chemistry. As pointed out in Ref. [1], the band formed by the diffusion of $|ss'\rangle$ is coupled to the state $|ss'\rangle$ and the existence of this band broadens this population transfer [1,14], which shows that the broadening should be a result of the many-body effect. However, we believe that it is mostly a two-body effect arising from the density fluctuation, as we will show below by simple theoretical reasoning and numerical simulations.

To better understand the diffusion process, we consider a toy model for the process $|s\rangle+|p\rangle\leftrightarrow|p\rangle+|s\rangle$. The Hamiltonian is

$$H=\sum_{j,k} V_{\mathrm{dip}}(\vec{r}_j-\vec{r}_k), \tag{1}$$

which is purely a diffusion process under the dipolar interaction

$$V_{\mathrm{dip}}(\vec{r})=c_d\frac{1-3\cos^2\theta}{r^3}. \tag{2}$$

$\vec{r}=\vec{r}_1-\vec{r}_2$, $\cos\theta=\hat{z}\cdot\hat{r}$, and $c_d=\mu_{sp}^2$. The energy scale is chosen to be $\bar{V}=\mu_{sp}^2 n$, the inverse of which sets up the time scale. To ease our discussion, we consider only one s atom and a bunch of p atoms with zero magnetic moment—i.e., magnetic quantum number $m=0$. In Fig. 1, we show the probability of finding an s state on the initial s atom as a function

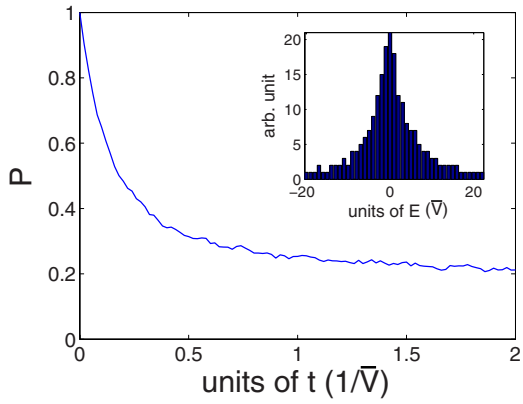

FIG. 1. (Color online) The probability of finding an s state on the initial s atom as a function of time. The inset shows the histogram of the eigenenergy. The simulation is carried out with 256 atoms and over 1000 spatial configurations. Energy is in units of $\bar{V}$ and time is in units of $1/\bar{V}$ (see text).

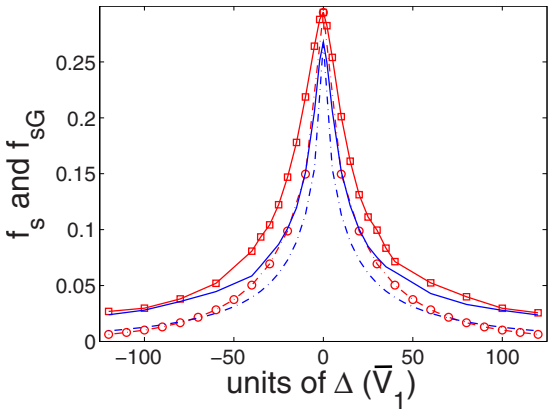

FIG. 2. (Color online) f_s and $f_{s,G}$ as a function of Δ for $N=10$ atoms averaged over 1000 spatial configurations. Results with marked (unmarked) lines include (exclude) diffusion. Solid and dash dotted lines are for the case of homogeneous and Gaussian convolution, respectively. Parameters used: $T=3.4$, $\sigma=500$, $\mu_{sp}=1.02$, and $\mu_{sp'}=0.98$. The results are insensitive to σ. Δ is in units of $\bar{V}_1$ (see text).

of time—i.e., $P=|\langle\psi(t)|\psi(t=0)\rangle|^2$, where $|\psi(t)\rangle$ is the many-body wave function at time t. We can see that the probability decays smoothly and saturates to a finite value. At $t\sim 0.2$, there is about a 50% chance that the s state has drifted away. So the characteristic time of diffusion is on the order of $1/10$. In addition, we show the histogram of the eigenenergy in the inset of Fig. 1 by directly diagonalizing the Hamiltonian (1). We find that the width of this diffusion band is roughly 5, corresponding to $1/t$. This poses questions on the original explanation of the band diffusion model. Since for large detuning—e.g., $\Delta=40\bar{V}_1$—in order to make a non-negligible transfer from pp to ss', the interaction strength between them should be of the same order as Δ. In this case, pp and ss' should be split by an amount of $\sim\Delta$. That is to say, the manifold of pp and ss' will be in a large detuning to the band. Thus, the state ss' with a large detuning from the band will not decay into it. Therefore, the explanation of broadening from this diffusion band model is questionable.

We investigate the linewidth problem of cases (I) and (II) by direct numerical simulations. To do this, we randomly put several atoms in a cubic box and assume each atomic state $|p\rangle$ or $|p'\rangle$ has no magnetic moment. The numerical schemes are as follows. We first calculate the full dynamics with a given detuning for each spatial configuration of atoms with a fixed evolution time. We then average over spatial configurations to obtain the excitation probability as a function of detuning from which we can extract the linewidth. In order to minimize the finite-size effect, we adopt wrap boundary conditions.

We first focus on case (I) and discuss case (II) subsequently. The energy and time scale are the same as those in the toy model. In case (I), each atom can be in the state p, s, or s'. The Hamiltonian is found to be

$$H=\sum_{jk}[V_{jk}e^{-i\Delta t}|p_jp_k\rangle\langle s_js'_k|+V'_{jk}|p_js_k\rangle\langle s_jp_k|+V''_{jk}|p_js'_k\rangle\langle s'_jp_k|]+\text{H.c.},$$

describing the processes

$$p+p\rightarrow s+s', \tag{3}$$

$$p+s\rightarrow s+p, \tag{4}$$

$$p+s'\rightarrow s'+p, \tag{5}$$

where process (3) is not always resonant and its detuning Δ is controlled by an electric field, while processes (4) and (5) are always resonant. V_{jk}, V'_{jk}, and V''_{jk} all take the form of $V_{\text{dip}}(\vec{r}_j-\vec{r}_k)$ with corresponding $c_d=\mu_{sp}\mu_{sp'}$, μ_{sp}^2, and $\mu_{sp'}^2$ for processes (3)–(5), respectively. Initially all of the atoms are in the state p, and they evolve under the dipolar interaction for a fixed time T. We are interested in the yield of the s atom (f_s) as a function of detuning, from which we can extract the linewidth.

We perform calculations with up to $N=10$ atoms and average over the initially random atom positions many times. Under wrap boundary conditions, our results already show convergent behavior for $N=8$ atoms. Extrapolation to $N=\infty$ will give about a 15% difference from that of $N=10$. As we will see later, this difference is not crucial to our conclusion as we are interested in the order-of-magnitude difference. Our results are shown as solid lines in Fig. 2. For the marked solid line, we include all three processes, while for the unmarked solid line, we only consider process (3); i.e., we effectively turn off the diffusion process. The extracted linewidths are about 35 and 25 for the two cases, respectively. This shows that process (3) already gives a width an order of magnitude larger than the average interaction strength and the diffusion process further broadens the linewidth by roughly 50%. So the unusual linewidth broadening mostly comes from process (3).

In real experiments, the density is not uniform. Therefore, we need to take into account the density profile of the atomic cloud. We assume it as a Gaussian form with a width σ, so

the density is written as $n(r)=2\sqrt{2}\bar{n}e^{-r^2/\sigma^2}$, where $\bar{n}$ is the average density. Accordingly, the excitation fraction with a Gaussian convolution ($f_{s,G}$) is given by

$$f_{s,G}=\frac{\int f_s[\Delta/\bar{V}\times\bar{V}/V(r)]n(r)r^2dr}{\int n(r)r^2dr}$$
$$=\frac{\int f_s[\Delta/\bar{V}\times e^{r^2/\sigma^2}/2\sqrt{2}]e^{-r^2/\sigma^2}r^2dr}{\int e^{-r^2/\sigma^2}r^2dr}. \quad (6)$$

Here we assume that the density varies slowly on the length scale we considered. The results for $N=10$ atoms are shown as the dash-dotted lines in Fig. 2. The marked dash-dotted line is the result including all three processes, while the unmarked dash-dotted line is the result only including process (3). The extracted linewidth with diffusion is about 30, which is a few times smaller than the results in Ref. [1]. The one extracted from the calculation without diffusion is about 20, again demonstrating that diffusion is not the main mechanism in the broadening. We also note that the curves are not perfectly symmetric around $\Delta=0$ due to the anisotropy of the dipolar interaction. However, this difference is too small to be detected under current experimental conditions.

We further consider the motional effect on linewidth broadening. To do this, we give a constant speed v_s for each atom, but with random direction. For $v_s=0.05$, the atom moves 0.1 (average distance) at the end of the simulation, which is still in the so-called "frozen" gas regime. Our results give an additional broadening of about 20%, so the motional effect is not important as expected.

Following similar procedures, we investigate case (II). Now each atom can be in the state s, s', p, or p'. The Hamiltonian is found to be

$$H=\sum_{jk}[V_{jk}e^{-i\Delta t}|p_jp'_k\rangle\langle s_js'_k|+V'_{jk}|p_js_k\rangle\langle s_jp_k|+V''_{jk}|p'_js'_k\rangle\langle s'_jp'_k|]$$
$$+\text{H.c.},$$

describing the processes

$$s+s'\rightarrow p+p', \quad (7)$$

$$p+s\rightarrow s+p, \quad (8)$$

$$p'+s'\rightarrow s'+p', \quad (9)$$

with a dipolar interaction. Our numerical results are shown in Fig. 3. In this case, the width with (without) diffusion is found to be about 20 (10). Therefore, the diffusion does not cause an order-of-magnitude change in the linewidth broadening.

So what causes the broadening? It is nothing unusual, but the rare pair fluctuation at small distances. To see this point, we calculate $P(|\Delta|)$, which is the probability distribution of nearest-neighboring atoms with absolute interaction strength not larger than the absolute detuning (see Fig. 4). $P(|\Delta|)$ can be found with the help of

$$P(|\Delta|)=\int_0^{|\Delta|}\frac{dP(|V|\leq|\Delta'|)}{d|\Delta'|}d|\Delta'|. \quad (10)$$

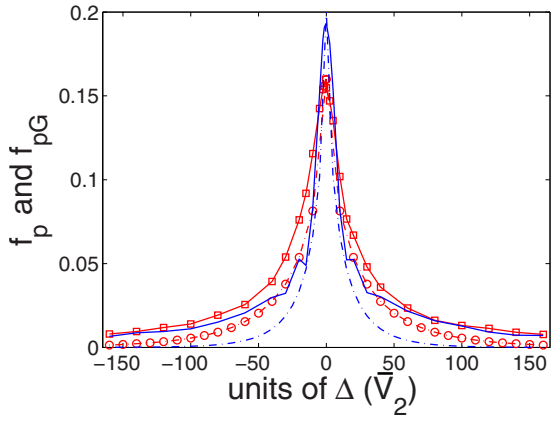

FIG. 3. (Color online) f_p and $f_{p,G}$ as a function of Δ for $N=20$ atoms averaged over 1000 spatial configurations. Results with marked (unmarked) lines include (exclude) diffusion. Solid and dash-dotted lines are for the case of homogeneous and Gaussian convolution, respectively. Parameters used: $T=0.36$, $\sigma=500$, $\mu_{sp}=2$, and $\mu_{s'p'}=0.5$. The results are insensitive to σ. The initial populations of s and s' are the same. Δ is in units of $\bar{V}_2$ (see text).

In a homogeneous system, according to the Erlang distribution [15], the nearest pair distribution is $\propto e^{-4\pi r^3/3}$ with unit density. Therefore, for an isotropic interaction $V_{\text{iso}}=1/r^3$ with $c_d=1$ (e.g., $\bar{V}=1$), we have

$$\frac{dP(|V_{\text{iso}}|\leq|\Delta|)}{d|\Delta|}=\frac{4\pi}{3|\Delta|^2}e^{-4\pi/3|\Delta|}, \quad (11)$$

while for the while dipolar interaction V_{dip} with $c_d=1$ (e.g., $\bar{V}=1$), we have

$$\frac{dP(|V_{\text{dip}}|\leq|\Delta|)}{d|\Delta|}=\frac{4\pi}{3|\Delta|^2}\int_0^1 dx|1-3x^2|e^{-(4\pi/3|\Delta|)|1-3x^2|}. \quad (12)$$

As $|\Delta|\rightarrow+\infty$, the asymptotic behaviors are $(4\pi/3)|\Delta|^{-2}$ and $(16\pi/9\sqrt{3})|\Delta|^{-2}$ for isotropic and dipolar interactions, respectively. As $|\Delta|\rightarrow 0^+$, they approach 0 and $\sqrt{3}/(4\pi)$ for

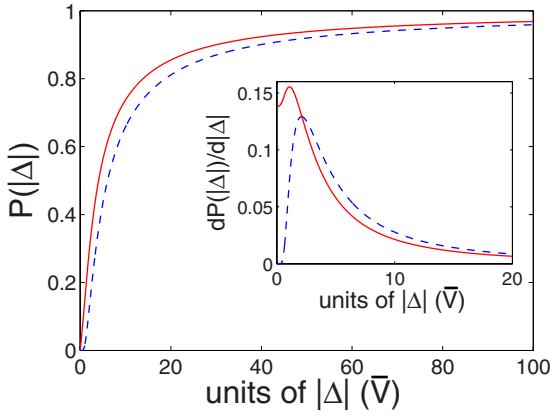

FIG. 4. (Color online) $P(|\Delta|)$ as a function of $|\Delta|$. Blue dashed lines are for the isotropic interaction, and red solid lines are for the anisotropic interaction. Δ is in units of $\bar{V}$ (see text).

isotropic and dipolar interactions, respectively. The remarkable difference at $|\Delta| \to 0^+$ is a signature of the dipolar interaction.

For $|\Delta|=40$, the probability of nearest atom pairs that have an interaction energy larger than $|\Delta|$ is about 10%; i.e., those atom pairs will have a non-negligible contribution to the dynamics. The calculated f_s of case (I) at this detuning is about 8%, close to the estimated value. Therefore, the rare pair fluctuation is the main cause of the linewidth broadening and the diffusion of excitation further increases this broadening by roughly 50%.

For case (II), we can also compute the linewidth (w) for different ratios of s and s'—i.e., w as a function of $\nu \equiv (n_1-n_2)/(n_1+n_2)$, where n_1 (n_2) is the density of s (s') atoms. Our numerical results are shown in Fig. 5. The largest error in our simulation still comes from the finite atom effect, which has been discussed for Figs. 2 and 3. This error does not change much as we vary the ratio of s and s' atoms. Other errors are negligible. We find that w increases as ν increases and saturates at $\nu=\pm 1$. The increasing behavior of w with ν is due to the imbalanced hopping of s and s' atoms ($\mu_{sp}=4\mu_{s'p'}$). The increased ratio in s atoms will thus show a stronger diffusion effect. However, this increase in the linewidth is again not an order-of-magnitude change.

To conclude, we have reexamined the important role that pair fluctuations play in the spectral linewidth broadening of a frozen Rydberg gas. From direct numerical simulations, we find that density fluctuations contribute to a width of roughly 20–30 times the average interaction strength. In addition, by turning off the diffusion process, we did not find an order-of-magnitude change in the linewidth. Therefore, the large linewidth is primarily due to density fluctuations, and the diffusion process is not overwhelmingly dominant as previously suggested. However, our numerical results are only in qualitative agreement with experimental results. The even larger linewidth observed in both cases and the double-peak structure as observed in (I) cannot be explained from current calculations, which encourages further investigations.

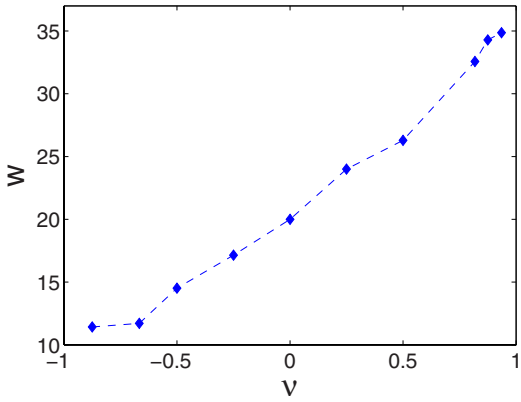

FIG. 5. (Color online) Line width w as function of population ratio ν of s and s' atoms. The same parameters are used as in Fig. 3 except for different ratio of s and s' atoms.

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