Resonant Dipole-Dipole Energy Transfer in a Nearly Frozen Rydberg Gas

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In a room temperature vapor of Rb Rydberg atoms, resonant dipole-dipole energy transfer occurs via binary collisions. In contrast, in the 300 μK vapor produced by a magneto-optical trap, the atoms are nearly frozen in place and many atoms interact simultaneously, as in an amorphous solid. As a result of the simultaneous multiple atom interactions, the energy transfer resonances are broadened substantially in frequency. [S0031-9007(97)04909-0]

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In practical terms, a gas is frozen if over the time scale of interest the constituent atoms or molecules move only small distances relative to their separation. In such a case, they can be expected to behave as an amorphous solid. Elegant examples of a frozen gas are the well defined spatial structures of cold ions observed in ion traps [1,2]. With neutral atoms, it is more difficult to realize a frozen gas due to the fact that the interactions between neutral atoms are of shorter range than the Coulomb interaction. Cold Rydberg atoms, with their large sizes and dipole moments, appear to be natural candidates for making a frozen neutral gas. Using them, one can reasonably expect to reach a regime where collective, many-atom, effects lead to qualitatively different behavior. Interesting examples are superradiance and the observation of a Mott transition [3].

Here, we report the results of experiments on resonant dipole-dipole energy transfer among 300 μK Rb Rydberg atoms in a magneto-optical trap (MOT). In these experiments, the dipole-dipole interaction of the atoms approaches their thermal energy. At room temperature 300 K, resonant energy transfer occurs by a well understood binary collision process, but it is qualitatively different at 300 μK. In the latter case, it is clearly not a binary process but a many-atom process which has some similarities to the formation of energy bands in a solid.

The specific process we have examined is, for two atoms, the process

$$\text{Rb } 25s_{1/2} + \text{Rb } 33s_{1/2} \longrightarrow \text{Rb } 24p_{1/2} + \text{Rb } 34p_{3/2}\,. \tag{1}$$

As shown in Fig. 1, it is resonant at the electric fields $E = 3.0$ and 3.4 V/cm, where the $25s_{1/2}$-$24p_{1/2}$ and $33s_{1/2}$-$34p_{3/2}$ intervals are equal. Because of the splitting of the $|m_j| = \frac{1}{2}$ and $\frac{3}{2}$ levels of the $34p_{3/2}$ state, there are two resonances. Binary resonant energy transfer collisions such as the one of Eq. (1) have been studied extensively, and the cross section σ is given by [4]

$$\sigma = \mu_A \mu_B / v\,, \tag{2}$$

where μ_A and μ_B are the $25s$-$24p$ and $33s$-$34p$ electric dipole matrix elements and v is the collision velocity. The duration of the collision, τ, is the inverse of the linewidth, $\Delta\nu$, of the collisional resonance and is given by

$$\tau = 1/\Delta\nu = \sqrt{\mu_A \mu_B}/v^{3/2} = \sqrt{\sigma}/v\,. \tag{3}$$

The linewidth depends on the collision velocity of the atoms but not on their density. At room temperature 300 K, using the values $\mu_A = 492ea_0$ and $\mu_B = 126ea_0$, we calculate $\sigma = 10^{-8}$ cm^2 and $\Delta\nu = 380$ MHz.

Previous experiments have shown that, when the temperature is reduced from 300 K to 1 K, the linewidth of the resonance drops sharply, as shown in Eq. (3) [5]. If we apply Eqs. (2) and (3) to the energy transfer of Eq. (1) at 300 μK, we find $\sigma = 10^{-5}$ cm^2 and $\Delta\nu = 12$ kHz. Typical Rydberg atom densities in the MOT are 10^9 cm^{-3}, implying a mean interatomic spacing of 10^{-3} cm, which is larger than the impact parameter $\sqrt{\sigma}$. Under such conditions, an energy transfer can no longer occur by the same binary collision process, but how it should change is not *a priori* obvious. It is likely

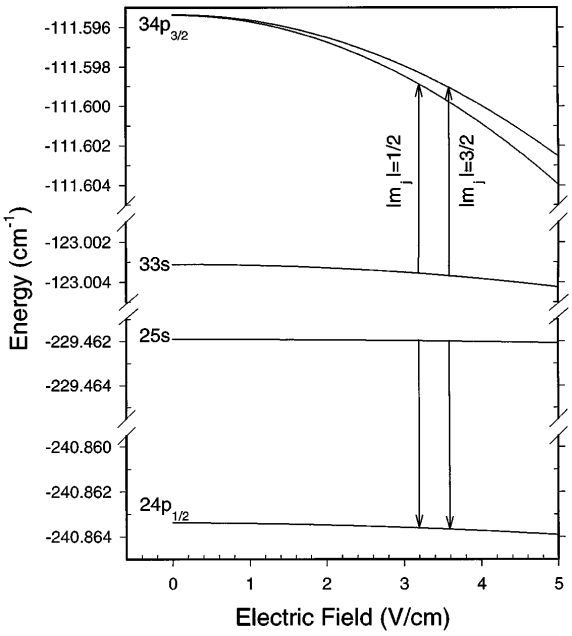

FIG. 1. Energy levels of Rb in an electric field showing the two energy transfer resonances of Eq. (1).

to become more quasistatic. For example, line broadening in a dense, 10^{12} cm^{-3}, thermal 300 K Rydberg atom sample can be described with a quasistatic picture [6].

In the 300 μK experiment, the ^{85}Rb atoms are held in a MOT, which is housed in a stainless steel vacuum chamber at a background pressure of 10^{-8} torr [7]. Other measurements suggest that the temperature in the MOT is not higher than 300 μK [8,9]. The chamber is at room temperature, and Rb is kept in a sidearm reservoir which is held at 40 °C. We use diode lasers for the trapping and repumping beams, all of which have typical linewidths, powers, and diameters of 3 MHz, 10 mW, and 5 mm. The magnetic field gradients in the horizontal and vertical directions are 5 and 10 G/cm, respectively. By imaging the radiation from trapped atoms onto a detector array, we have determined the full width at half maximum of the cloud of atoms in the trap to be 0.7 mm, and the number of atoms in the trap to be as high as 10^7, leading to a density of 10^{10} cm^{-3}. The trap volume is midway between a pair of vertical copper plates 1.909 cm apart. In the center of the left-hand plate, there is a $\frac{1}{4}$ in. diameter array of twenty 0.020 in. diameter holes through which ions produced by field ionization are extracted. A dual microchannel plate detector is placed just outside the left-hand plate to detect the ions.

We excite the Rb atoms from the $5p_{3/2}$ state to the $25s$ and $33s$ states with two pulsed dye lasers pumped by a Nd:YAG (yttrium aluminum garnet) laser running at a 20 Hz repetition rate. The dye lasers have 0.1 cm^{-1} linewidths and transfer up to 10% of the trap population to each of the two Rydberg states, leading to maximum Rydberg state densities of $N_0 = 10^9$ cm^{-3}. We allow the atoms to interact for up to 3 μs in a static field and then apply a 1 μs rise time high voltage pulse to the right-hand plate to field ionize the excited atoms. The amplitude of the field pulse is 550 V/cm, which allows easy separation of the ionization signals from the $34p$ and $33s$ states. We detect the signal from the $34p$ state atoms, and we scan the value of the static field through the resonances of Fig. 1 over many shots of the laser to observe the energy transfer resonances.

To do the experiment at 300 K, we simply turned off the trapping lasers and magnetic fields and excited the background Rb atoms to the $5p_{3/2}$ state with a pulsed dye laser and then to the $25s$ and $33s$ states as described earlier. In this case, the density of Rb Rydberg atoms was 2×10^9 cm^{-3}. At 300 K, the resonance is 250 MHz wide (FWHM), in reasonable agreement with the calculated value. Based on the observed collision rate and Rb number density, we estimate $\sigma = 10^{-8}$ cm^2, also in agreement with expectations.

In Fig. 2, we show the 300 μK resonances using several densities of trapped atoms. The highest density is $N_0 = 10^9$ cm^{-3} in each of the $33s$ and $25s$ Rydberg states, and the densities shown in Fig. 2 are N_0, $0.77N_0$, $0.46N_0$, and $0.19N_0$. The number density was varied

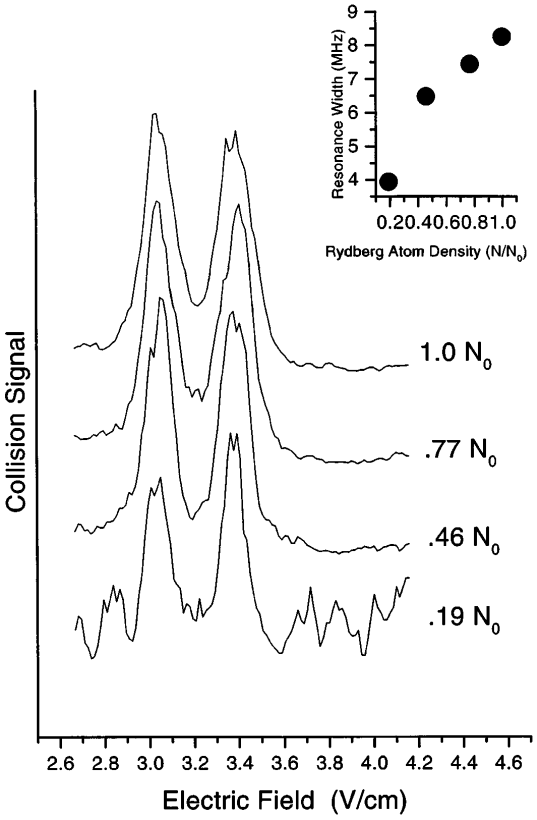

FIG. 2. The $25s_{1/2} + 33s_{1/2} \rightarrow 24p_{1/2} + 34p_{3/2}$ resonances at 300 μK observed in the MOT at four densities, $0.19N_0$, $0.46N_0$, $0.77N_0$, and N_0. The inset shows the width of the observed resonances vs relative density.

by changing the intensity of the repumping laser. Other trap parameters, and the temperature, were unchanged. The observed signals increased monotonically with rates which decreased with time constants of order 1 μs. For each of the traces of Fig. 2, energy transfer was allowed to occur for 3 μs after laser excitation before the field ionization pulse was applied. With the number density N_0, approximately 20% of the $33s$ atoms had undergone transitions to the $34p$ state in 3 μs.

The conversion from field to frequency at $E = 3.4$ V/cm in Fig. 2 is 1 V/cm = 53 MHz, and the resonance widths increase from 3.9 to 8.4 MHz as the number density is increased from $0.19N_0$ to N_0, as shown explicitly by the inset. The typical observed linewidth, 5 MHz, is almost 3 orders of magnitude larger than the expected 12 kHz linewidth for a binary collision. Furthermore, the linewidths of the energy transfer resonances increase with number density, unlike those of binary collisional resonances. Similar broadening of Cs energy transfer resonances has been observed by Mourachko *et al.* [10]. By examining other energy transfer resonances occurring at higher static fields, we have determined the electric field inhomogeneity to be less than 1 part in 10^3, which corresponds to a 0.15 MHz contribution to the observed

linewidths. To determine the effect of the magnetic field gradient, we added magnetic field pulses of up to 3 G, and, by observing shifts in the resonances, set an upper limit of 0.5 MHz on the magnetic field's contribution to the linewidth of the observed resonances. We believe the field inhomogeneities to contribute approximately 1 MHz to the observed widths.

It is clear that the energy transfer does not occur by simple binary collisions, but how does it occur? We begin by noting the relevant magnitudes of the problem. For an average interatomic spacing $r_0 = (\frac{3}{4}\pi N_0)^{1/3}$, the typical dipole-dipole interaction $\mu_A\mu_B r_0^3 = 0.24$ MHz. At $T = 300$ μK, $kT = 6$ MHz, and the collision velocity $v = \sqrt{8kT/\pi M} = 38$ cm/s, where k is Boltzmann's constant and M is the reduced mass of the two colliding atoms, both ^{85}Rb in this case. Over 3 μs, the typical time scale for the experiment, the atoms move 1×10^{-4} cm or $0.1r_0$.

There are three pairwise dipole-dipole couplings which exist in the system;

$$\text{Rb } 25s_{1/2} + \text{Rb } 33s_{1/2} \longrightarrow \text{Rb } 24p_{1/2} + \text{Rb } 34p_{3/2}\,, \tag{4a}$$

$$\text{Rb } 25s_{1/2} + \text{Rb } 24p_{1/2} \longrightarrow \text{Rb } 24p_{1/2} + \text{Rb } 25s_{1/2}\,, \tag{4b}$$

and

$$\text{Rb } 33s_{1/2} + \text{Rb } 34p_{3/2} \longrightarrow \text{Rb } 34p_{3/2} + \text{Rb } 33s_{1/2}\,. \tag{4c}$$

Equation (4a), which is identical to Eq. (1), is the dipole-dipole coupling responsible for the observed resonant energy transfer from the two s states to the two p states. It is only resonant at the fields $E = 3.0$ and $E = 3.4$ V/cm and has the strength $\mu_A\mu_B/r^3$, where r is the spacing between the two atoms. The dipole-dipole couplings of Eqs. (4b) and (4c) are resonant at all electric fields, have magnitudes μ_A^2/r^3 and μ_B^2/r^3, and are the same as those responsible for self-broadening of resonance line transitions [11] and the suppression of superradiance [12].

A qualitative description of the process which reproduces the major features of our observations starts with the assumption that the atoms are frozen in place. Some pairs of atoms are closely spaced, by r_c, where $r_c < r_0$, have strong couplings, and are able to undergo the transition of Eq. (4a) far off resonance, by up to $\mu_A\mu_B/r_c^3$, leading to the large observed linewidths. There are too few of these atoms, though, to give the magnitude of the signal observed. However, the pairs of close atoms oscillate at frequency $\mu_A\mu_B/r_c^3$ between the $25s$ and $33s$ states and the $24p$ and $34p$ states, spending about half of their time in the p states. While in the p states, resonant energy transfer can occur to other $33s$ and $25s$ atoms, which are the mean distance r_0 away, by the processes of Eqs. (4c) and (4b), respectively. This transfer occurs at rates given approximately by μ_A^2/r_0^3 and μ_B^2/r_0^3. For a given pair of close atoms, this entire process is repeated many times over the 3 μs course of the experiment, and p state population slowly expands outward from the close atoms. The linewidth is determined by the coupling of the close atoms, and the time scale for the development of the signal and its magnitude are determined by the couplings due to the average spacing.

If these two processes were independent, the line shape would be determined entirely by the probability of finding pairs spaced by r_c, and a line shape with wings falling off as $1/\Delta$, where Δ is the detuning from resonance, would result. However, a four atom model shows that these two processes are not independent. For simplicity, we label the atomic $25s$, $24p$, $33s$, and $34p$ states s, p, s', and p', respectively. Consider initially exciting two atoms to each of the s and s' states, to produce the state $sss's'$ of the system. The wave function of the system is given by the ordered product of the atomic wave functions at the four spatial sites. The second and third atoms are close together, being spaced by r_c, while the other spacings are the average spacing r_0. We assume that the tuning electric field is slightly below that required for the resonant energy transfer of Eq. (4a). If we ignore the dipole-dipole couplings of Eq. (4), the energies would be as shown in Fig. 3(a), where the energy level spacing Δ is the detuning from the resonance of Eq. (4a). The six states shown in Fig. 3(b) are the eigenstates, and no evolution from the initial state $sss's'$ occurs.

When we introduce only the strongest coupling, i.e., $\mu_A\mu_B/r_c^3$, between the second and third atoms, the two pairs of states of Fig. 3(b) connected by the long arrows are coupled, leading to two eigenstates which are superpositions of $sss's'$ and $spp's'$ and two which are

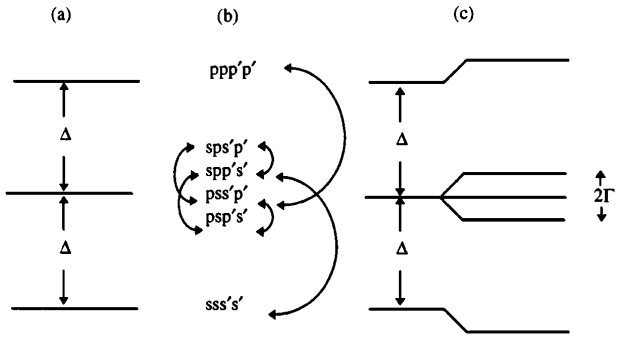

FIG. 3. The energy levels of the system of four atoms when detuned from resonance by Δ. (a) With no dipole-dipole couplings, there are three energies, and the states of (b) are the eigenstates. The four middle states are degenerate. The dipole-dipole interaction of the close second and third atoms, indicated by the long arrows in (b), leads to energy shifts of $\pm\Gamma$ and lifts the degeneracy of the middle four levels, as shown in (c). Note that it does not couple $sss's'$ to $ppp'p'$. The dipole-dipole interactions of Eqs. (4b) and (4c) between atoms of the average spacings, indicated by the short arrows in (b), are required to couple $sss's'$ to $ppp'p'$.

superpositions of $ppp'p'$ and $pss'p'$. These eigenstates are shifted in energy by Γ, as shown in Fig. 3(c), which partially removes the degeneracy of the middle set of levels. Since the initial state is not an eigenstate, a fraction of the state oscillates between $sss's'$ and $spp's'$ in time. This fraction is equivalent to the population transfer and is only significant if $\mu_A\mu_B/r_c^3$ is comparable to Δ. Although it is likely that $\Gamma < \mu_A\mu_B/r_c^3 < \Delta$, their magnitudes cannot be vastly different if there is to be substantial population transfer. Irrespective of the value of $\mu_A\mu_B/r_c^3$, the initial state $sss's'$ is not coupled to $ppp'p'$, so only the close pair of atoms undergoes the transition to the p states. If the average couplings are much smaller, a situation similar to electromagnetically induced transparency results [13].

The average couplings of Eq. (4), of magnitudes $\mu_A\mu_B/r_0^3$, μ_A^2/r_0^3, and μ_B^2/r_0^3, couple all of the states. For example, the couplings of Eqs. (4b) and (4c), μ_A^2/r_0^3 and μ_B^2/r_0^3, couple the middle four states of Fig. 3(b) as shown by the short arrows. The coupling leads to population transfer involving all four atoms only if the eigenstates are significantly mixed, which requires in turn that splittings produced by the weaker couplings be comparable to the intermediate energy splitting Γ. Therefore, $\mu_A\mu_B/r_0^3$, μ_A^2/r_0^3, and μ_B^2/r_0^3 cannot be orders of magnitude smaller than the strong coupling $\mu_A\mu_B/r_c^3$. When more atoms are taken into account, the individual levels of Fig. 3 develop into bands of widths larger than the average coupling [14], which allows the interaction between the close atoms to be stronger relative to the average interaction strength than is allowed in our four atom model.

By lowering the temperature from 300 K to 300 μK, we have observed a qualitative change from binary energy transfer collisions to an energy transfer among many atoms. The dipole-dipole energy transfer in the cold gas can be understood in terms more similar to those used to describe solids. More systematic study of this nearly frozen Rydberg gas are underway, and this frozen Rydberg gas should have a variety of fascinating properties.

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