Collective Absorption of Blackbody Radiation by Rydberg Atoms in a Cavity: An Experiment on Bose Statistics and Brownian Motion

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Absorption of blackbody radiation by Rydberg atoms in a resonant cavity is shown to be a collective process in which the atoms behave as a Bose gas. The equilibrium energy of this gas was measured and found equal to twice the photon energy (factor of 2 accounting for atomic transition degeneracy). It is shown that this experiment exhibits the Brownian motion of the atomic-system Bloch vector.

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We have measured the absorption of blackbody radiation by Rydberg atoms in a resonant cavity. We have observed that the number ΔN of absorbing atoms is limited to twice the number $\bar{n} = [\exp(h\nu/k_B T) - 1]^{-1}$ of blackbody photons in the cavity (k_B and h, Boltzman and Planck constants; T, temperature of radiation; ν, transition frequency). In this process, the atomic sample evolves in a *collective* mode and behaves as a single quantum system exhibiting basic effects of Bose-Einstein statistics and Brownian motion. The atoms in this experiment define a universal radiation thermometer in which the absolute temperature is directly deduced from a millimeter-wave photon counting measurement, the calibration being independent of the probability rate of the Rydberg transition. These features make this study quite different from previous ones which have recently investigated the absorption of blackbody radiation by Rydberg atoms.[1]

Our experimental arrangement is sketched in Fig. 1. A thermal beam of Na atoms is excited by a laser pulse (5 ns duration) to the $|30S_{1/2}\rangle$ level. Excitation occurs in a millimeter-wave cavity which can be tuned to the transition to the higher lying $|30P_{1/2}\rangle$ level ($\nu = 134$ GHz; $\lambda = c/\nu = 2.25$ mm; equivalent temperature $h\nu/k_B = 6.45$ K). The semicofocal Fabry-Perot cavity has an intermirror length $L \sim 1.3$ cm, a Gaussian mode waist $w_0 = 3$ mm, and $Q = 5 \times 10^3$. The effective volume V of the mode is $V = \pi L w_0^2/2 = 0.18$ cm^3. The radiation in the cavity is initially in equilibrium at room temperature ($T_0 = 300$ K). The atoms interact with the thermal field in the cavity during an average transit time $\Delta t = 2.5$ μs, then drift out and pass between two condenser plates where they are field ionized. The resulting electrons are detected by an electron multiplier (EM). The time-varying ionization field,[2] starting a time $t_0 = 20$ μs after the laser pulse, reaches the threshold for ionization of the $30S_{1/2}$ and $30P_{1/2}$ levels at different times, producing two electron peaks, each being proportional to the population of the corresponding level at time t_0. It is essential in this work to measure these populations precisely. We reduce the signal to very low atom counts by attenuating the laser beam, and measure the signals corresponding to single-atom events which yield the absolute EM gain at its maximum voltage. We then determine the gain versus voltage curve and from it we deduce the absolute number of electrons received by the EM. The number of atoms leaving the cavity is obtained by correcting for losses due to spontaneous emission during the time $t_0 - \Delta t$. The accuracy of the atom number measure is estimated as $\pm 25\%$.

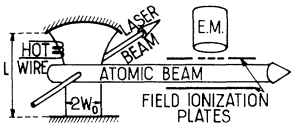

FIG. 1. Experimental arrangement for Rydberg-atom blackbody radiation absorption in a resonant cavity.

To tune the cavity on and off resonance, we either sweep its frequency by varying L, or Stark shift the atomic frequency by perturbing the atoms with a small dc electric field (we use an electrode not shown in Fig. 1). Both procedures have the same effect. When the cavity is off resonance, we observe, after averaging over 100 laser pulses, the typical EM output versus time signal of Fig. 2(a) (solid line): The large peak corresponds to the $30S_{1/2}$ level and the small one to the $30P$ level populated from the $30S_{1/2}$ level by the absorption of the blackbody radiation in the modes transverse to the cavity axis (during time Δt) and outside the cavity (time $t_0 - \Delta t$). About 10% of the atoms are transferred by these nonresonant processes at 300 K. For a resonant cavity [Fig. 2(a), dashed line], an increase ΔN of the number of

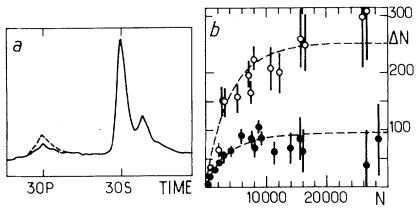

FIG. 2. (a) Time-resolved field-ionization signals corresponding to resonant (dashed line) and off-resonant (solid line) cavity ($N \sim 400$ atoms). (b) Plots of ΔN vs N for two radiation temperatures. Solid circles: $T = 300$ K; open circles: $T_{rad} \sim 900$ K, corresponding to a hot wire at 1600 K.

transferred atoms is observed. The variation of ΔN as a function of the total number N of atoms is plotted in Fig. 2(b) (solid circles). The data are processed by a LSI 11 computer. Between laser pulses, the computer drives the Stark-shifting field on and off. In each case, it determines the $30S_{1/2}$ and $30P$ populations at time Δt and measures ΔN by the difference between $30P_{1/2}$ populations corresponding to on- and off-resonant signals. Statistics of 300 events for each N yields an average $\langle \Delta N \rangle$ corresponding to the solid circles in Fig. 2(b) (uncertainties on $\langle \Delta N \rangle$ are shown by error bars). We see that $\langle \Delta N \rangle$ increases with N for small systems but that for $N \gtrsim 5000$ it reaches a limit $\langle \Delta N \rangle_{lim} = 80 \pm 20$. This saturation, typical of the experiment, corresponds to a strong decrease of the relative transfer $\langle \Delta N \rangle / N$ for large N, making the resonant enhancement increasingly difficult to observe. $\langle \Delta N \rangle_{lim}$ is independent of cavity characteristics (it is the same for another cavity configuration with $Q = 10^4$). On the other hand, it clearly varies with radiation temperature. To show this effect, we couple the cavity to a small electrically heated tungsten wire whose temperature T_w, controlled by its resistance, is varied from $T_0 = 300$ K to $T_1 = 1600$ K. The open circles in Fig. 2(b) show the results for $T_w = T_1$. The new limit is $\langle \Delta N \rangle_{lim} = 270 \pm 60$.

Cavity enhancement of blackbody absorption is not unexpected. In this respect, this experiment is the counterpart of the "absorption inhibition" effect[3] observed for Rydberg atoms between metallic plates which act as a waveguide beyond cutoff for the absorption frequency. In that case, the plates decreased the density of available radiation modes and the thermal-field absorption was accordingly diminished. In our case, the cavity increases the spectral density of radiation resonant with the atoms and amplifies the absorption. The surprising point is that this enhancement is, even for relatively large atomic systems, limited to a very small absolute number of absorbed photons. Let us make a very simple estimate of the thermal-field absorption rate $W_B^{(cav)}$ by an atom in the cavity. It is the product of a coefficient $C = \pi d^2/\epsilon_0 \hbar^2$ (d is the electric dipole matrix element between the levels, equal to 420 a.u. for this transition) and the blackbody-radiation spectral density in the cavity, $I_B^{(cav)} = (\bar{n} Q/\pi V)[\exp(h\nu/k_B T) - 1]^{-1}$. Introducing the average number $\bar{n}$ of photons in the cavity, we can write $W_B^{(cav)} = C I_B^{(cav)} = d^2 Q \bar{n}/\epsilon_0 \hbar V$. At room temperature, $\bar{n} = 47$ and $W_B^{(cav)} \sim 2 \times 10^4$ sec^{-1}, corresponding to a total absorption probability $W_B^{(cav)} \Delta t \sim 5\%$ during the atom-cavity interaction. If the N atoms were coupled *independently from each other to* the cavity mode at 300 K, a number of them $\Delta N \sim 0.05N$ would be excited. ΔN would increase linearly with N, not saturate as observed for high N. Moreover, $\Delta N/N$ should increase with T and tend towards $\frac{1}{2}$ at high temperature ($T \sim 3000$ K corresponds to $\bar{n} \sim 500$ and $W_B^{(cav)} \Delta t \sim 50\%$). A tentative explanation for the limiting behavior of ΔN is that the atoms "cool" the radiation field by their own absorption.[4] This hypothesis does not hold however, since the cavity filling time $T_{cav} = Q/2\pi\nu = 6$ ns is 400 times shorter than Δt. The cavity walls can produce $\bar{n} \Delta t / T_{cav}$ photons during Δt, i.e., a number large enough to excite several thousand atoms.

The above paradox is resolved by noticing that the atoms are not absorbing *independently* from each other, but rather *collectively:* The dipoles induced on different atoms by the fluctuating thermal field have an absolute random phase, but their phases *relative to each other* are correlated because of the coherent spatial structure of the cavity mode. As a result, the atomic sample acquires a global dipole proportional to N. The fluctuating dipole couples to the cavity walls and undergoes a collective radiative damping with a characteristic rate[5] $T_R^{-1} \simeq d^2 Q N/\hbar \epsilon_0 V$. Radiative damping competes with the absorption of thermal radiation, and the absorption probability is limited to a value of the order of $W_B^{(cav)} T_R$ (not $W_B^{(cav)} \Delta t$). The atomic (d^2) and cavity (Q) parameters are eliminated; the limit is $W_B^{(cav)} T_R \sim \bar{n}/N$. The absolute number of absorbing atoms $\Delta N = W_B^{(cav)} T_R N$ approximately equals the photon occupation number, a quite unexpected result.

The existence of a limit independent of the system's specific parameters (d^2, Q) suggests that

this result can be derived by purely statistical arguments. Consider first the simple case where the Rydberg atoms are prepared inside the cavity in a volume much smaller than λ. The absorption process in the cavity then leaves the atoms *indistinguishable*. Since the initial state (all atoms in $30S_{1/2}$ level) is invariant by atom exchange, throughout the interaction with the cavity the sample remains in a state fully symmetrical with respect to atom permutations. There are $N+1$ such symmetrical states (called Dicke states[6]), differing from each other only by the occupation numbers in the lower and upper levels. The atomic system, restricted to these states, behaves as a single quantum object with nondegenerate energy levels (not as a "classical" gas of N independent atoms with 2^N states and large degeneracies). The thermal equilibrium of this quantum system is quite easy to determine. The Dicke states make a scale of equidistant levels with an elementary spacing $h\nu$ equal at resonance to the energy scale of the harmonic-oscillator cavity mode. When the two systems are put in contact, they tend to an equilibrium corresponding to the same population distribution in each. Thus, the average excitation of the Rydberg gas tends *exactly* towards $\bar{n}$. This result holds even if the atoms are not in a small volume. The Dicke states are then replaced by $N+1$ collective superposition states with probability amplitude reflecting the atom distribution in the mode, all the above arguments remaining valid. The atomic indiscernability in this absorption process is typical of a Bose gas. The heat capacity of a Bose gas being much smaller than the one of a classical degenerate gas, we understand that the atoms thermalize by absorbing a very small energy and by removing a very small entropy from the field.

We have implicitly assumed so far that the Rydberg levels are nondegenerate. In fact, the $30S_{1/2} \rightarrow 30P_{1/2}$ transition results from the superposition of two independent components, $|30S_{1/2}, M_J = \mp\frac{1}{2}\rangle \rightarrow |30P_{1/2}, M_J = \pm\frac{1}{2}\rangle$, induced by the two σ_+ and σ_- components of the radiation field in the cavity (if we quantize along the cavity axis, the transverse Gaussian field is obviously a σ_+, σ_- superposition). Each of these components corresponds to a collective atomic mode which, at equilibrium, absorbs $\bar{n}$ photons. The limit is thus in fact $2\bar{n} \sim 2k_B T/h\nu$. This value (94 atoms at room temperature) agrees well with the measured one (80 ± 20 atoms). The $\langle \Delta N \rangle_{\lim}$ values obtained for other tungsten-wire temperatures can thus be considered as *absolute* radiation-temperature measurements. We get $T_{rad} = 900$ K for the open circles in Fig. 2(b), which means that the radiation reaches a temperature equal to about half that of the hot wire, a result consistent with estimates based on simple cavity coupling models.

In order to analyze this process more quantitatively, let us describe the $N/2$-atom system associated to each (σ_+ or σ_-) transition as a constant-length angular-momentum-like vector $\vec{J}$ evolving in an abstract space. This so called Bloch vector[7] is constructed by symmetrical superposition of $N/2$ spin-$\frac{1}{2}$-like two-level systems. The initial position of this vector corresponds to all spins aligned in the "down" position (defined as the Oz axis) and the Bloch vector points along this direction at $t = 0$. Because of blackbody radiation perturbation, the transverse components J_x and J_y of $\vec{J}$—which represent two out-of-phase components of the global atomic electric dipole—acquire small values $J_i = N\theta_i/4$ $(i = x, y)$, where the θ_i are independent fluctuating Bloch angles. Since $J^2 = J_x^2 + J_y^2 + J_z^2$ is conserved, J_z is at the same time reduced by a small amount $\Delta J_z = N(\theta_x^2 + \theta_y^2)/8$, related to the number of excited atoms by $\Delta N/2 = 2\Delta J_z$. Each θ_i can be shown to obey a damped pendulum equation (valid only at the classical limit $N, \bar{n} \gg 1$):

$$\dot{\theta}_i + \theta_i/2T_R = (d/\hbar)E_B{}^i(t), \tag{1}$$

with T_R being defined as above and the $E_B{}^i(t)$ being two $\pi/2$-out-of-phase components of the blackbody field, resonant with the atomic transition. Equation (1) is identical to the Langevin equation of a Brownian particle of velocity $v = c\theta_i$ and mass $m = Nh\nu/2c^2$, subjected to a restoring force $(\theta_i/2T_R)$ and to a fluctuating force F such that $F/mc = dE_B{}^i/\hbar$. This force is produced by "collisions" with the photon reservoir at temperature T. Its correlation time is $T_{cav} \ll T_R$. Within a few T_R, each degree of freedom of this particle acquires an average kinetic energy $m\bar{v}^2/2 = h\nu\bar{\theta}_i{}^2/4$ equal to $k_B T/2$. We thus have $2(\Delta J_z)_{av} = N(\theta_x^2 + \theta_y^2)/4 = k_B T/h\nu$, which, taking into account the transition degeneracy, yields again $\langle \Delta N \rangle_{\lim} \sim 2k_B T/h\nu$. This description—equivalent to the previous one for $N, \bar{n} \gg 1$—portrays absorption in the cavity as the Brownian motion of a Bloch vector immersed in a thermal bath of blackbody photons. If Δt is shorter than T_R, the system does not reach its equilibrium and one has $\langle \Delta N \rangle = 2\bar{n}[1 - \exp(-\Delta t/T_R)]$. In this experiment, Δt is fixed and T_R can be changed by varying N. $\langle \Delta N \rangle$ appears thus, below saturation, as an exponential function of N [dashed lines in Fig. 2(b)].

It is also interesting to measure the fluctuations around equilibrium, which are typical of a Brownian motion. A detailed study of the pulse-to-pulse random variations of ΔN around $\langle \Delta N \rangle$ should allow us to probe the large fluctuations of the photon number in the cavity mode and to reconstruct their Bose-Einstein probability distribution. Of course the phenomenon described here is general, not *a priori* restricted to Rydberg atoms. For ordinary atoms with $d \sim 1$ a.u. however, $\Delta t/T_R$ is very small unless N is of the order of $\sim 10^9$. The relative excitation $\langle \Delta N \rangle / N = \bar{n}/N$ is then undetectable ($\sim 10^{-7}$): Only Rydberg atoms can produce Bloch vectors "light" enough to exhibit Brownian motion. There is a link between this work and Rydberg maser studies[7] where the atoms are prepared in the upper level of the transition resonant with the cavity. The Bloch vector at $t = 0$ is then in the up position; its superradiant decay towards the down direction is triggered by the blackbody field and it obeys an equation similar to Eq. (1). It is striking that the atomic indiscernability, responsible for "superradiance" when the system is initially excited, leads to a kind of "subabsorption" when it starts from its lower state.

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Bond-Breakage Mechanism in the Predissociation of the $\tilde{A}$ State of N_2O^+: New Information from High Resolution Spectroscopy

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A recent study of high-resolution laser spectroscopy in a fast N_2O^+ beam indicates that the hyperfine structure of the $\tilde{A}(100)$ state is mainly due to Fermi contact interaction on the outer nitrogen nucleus. This new spectroscopic result is crucial for the interpretation of the laser predissociation of isotopically substituted N_2O^+ with a tandem mass spectrometer. Under such circumstances the fast-ion-beam laser-spectroscopy method yields new information on the bond-breakage mechanism during predissociation.

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In this communication, we report the analysis of a very-high-resolution photodissociation spectrum of the $\tilde{A}(100)$-$\tilde{X}(000)$ system of N_2O^+ in the near ultraviolet. Obtained by fast-ion-beam laser spectroscopy (FIBLAS), with a setup (see Carré *et al.*[1]) which reproduces, in a high energy range, the familiar tandem mass spectrometer geometry, this spectrum represents the first example of a fully resolved hyperfine structure in the electronic excited state of an asymmetrically triatomic molecular ion. Two main conclusions arise from our study of isotopically substituted $^{15}N_2O^+$, $^{14}N^{15}NO^+$, and $^{15}N^{14}NO^+$: First, we demonstrate that the hyperfine structure of the molecule is essentially due to the external nucleus; second, we present evidence that in the predissociation of the $\tilde{A}(100)$ state the molecule dissociates by ejection of this external nucleus, thus without "scrambling." This last mechanism was invoked by Berkowitz and Eland[2] in their

