Transient absorption by a Rydberg atom in a resonant cavity

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A theoretical analysis is presented describing the interaction of the field in a high-Q microwave cavity with an atom that is suddenly excited to the lower of two Rydberg levels that are resonantly coupled by the field. It is found that the field initially in the cavity is canceled by interference with the source field emitted by the atom. The absorption may be characterized as a "darkness wave packet" emitted by the atom and reflected by the cavity walls.

The development of short-pulse laser excitation has led to the production of quantum states that are not well described by the familiar stationary energy eigenstates. It has been shown that in many cases the states produced are localized wave packets propagating in classical orbits.[1] These waves packets may describe Rydberg electrons localized in the radial coordinate[1-3] or the angular coordinates,[4] an electron undergoing photoionization,[5] an internuclear coordinate in a molecule,[6] or a single photon.[7] The concept of a localized wave packet state is not a new one; Schrödinger and Lorentz introduced it in the very earliest days of quantum theory.[8] But, the production of a well-localized packet requires the simultaneous and coherent excitation of many different eigenstates. The development of picosecond and femtosecond laser technology makes it practical to excite such linear combinations.

In this paper we will study another problem in which a localized wave packet state is produced. In our model a high-Q microwave cavity is prepared initially in a state with one photon in one cavity mode, and all of the other modes unexcited. A single atom is introduced into the cavity. The atom is initially in its ground state. A short laser pulse is then used to excite the atom into a Rydberg state. The state is the lower of two Rydberg states that are resonantly coupled by the microwave mode that is initially excited. If the laser pulse is short in duration compared with the round-trip time for a photon in the cavity, transient absorption occurs as the atom absorbs a microwave photon.

In order to treat transient effects in the cavity we must extend the usual single-mode Jaynes-Cummings model to include many cavity modes. Recently, we have considered the problem of transient spontaneous emission by a single atom in a cavity. In that paper we developed the appropriate multimode equations.[7] Since that analysis applies directly to the present problem we will not repeat the entire calculation, but will instead simply write down the relevant equations from that paper, and show the solutions we obtained by numerical integration. We will use the Schrödinger representation and write the state vector in the form

$$|\psi(t)\rangle = b_e(t)\,|e,\{0\}\rangle + \sum_n b_n(t)\,|g,1_n\rangle \;, \qquad (1)$$

where the state $|e,\{0\}\rangle$ has the atom excited and no photons in any mode, while the state $|g,1_n\rangle$ has the atom in its lower resonant level and one photon in the nth cavity mode. We substitute this state vector into Schrödinger's equation to generate the equations

$$\dot{b}_e + i\omega_a b_e = (i/2)\sum_n \Omega_n b_n \qquad (2a)$$

and

$$\dot{b}_n + i\omega_n b_n = (i/2)\Omega_n b_e \;, \qquad (2b)$$

where ω_a is the atomic resonance frequency, ω_n is the frequency of the nth cavity mode, and Ω_n is the Rabi frequency associated with one photon in the nth cavity mode. We have applied the boundary conditions

$$b_e(0)=0, \quad b_n(0)=\delta_{m,n} \;, \qquad (2c)$$

where the m on the δ function denotes the one particular mode that is initially excited.

To simplify the calculation we have taken the atom to be in the center of a spherical cavity. The transition is in resonance with the twentieth mode of the cavity, and there is initially one photon in this mode. We simply numerically integrate Schrödinger's equation, (2a) and (2b), for the two atomic levels coupled to the first 100 cavity modes. The amplitudes b_e, b_n are then used directly to calculate the expectation value of the field intensity in the cavity using the simple relation

$$\langle \psi(0)\,|\,\hat{\mathbf{E}}^{(-)}(r,t)\cdot\hat{\mathbf{E}}^{(+)}(r,t)\,|\,\psi(0)\rangle = \left| \sum_n \frac{\omega_n}{c}\,\mathbf{A}_n(r)\,b_n(t) \right|^2 , \qquad (3)$$

where $\hat{\mathbf{E}}^{(\pm)}(r,t)$ are the positive and negative parts of the Heisenberg picture electric field operators, and $\mathbf{A}_n(r)$ is the classical mode function for the nth mode. This expression is derived explicitly in Appendix B of Ref. 7. It should be noted that this simple relation does not hold generally, but only in the case when there is a single photon in the cavity.

The results of these calculations are shown in Figs. 1(a)–1(e). The quantity plotted is the expectation value of the normally ordered intensity of the field in the cavity. We have multiplied this intensity by the square of the

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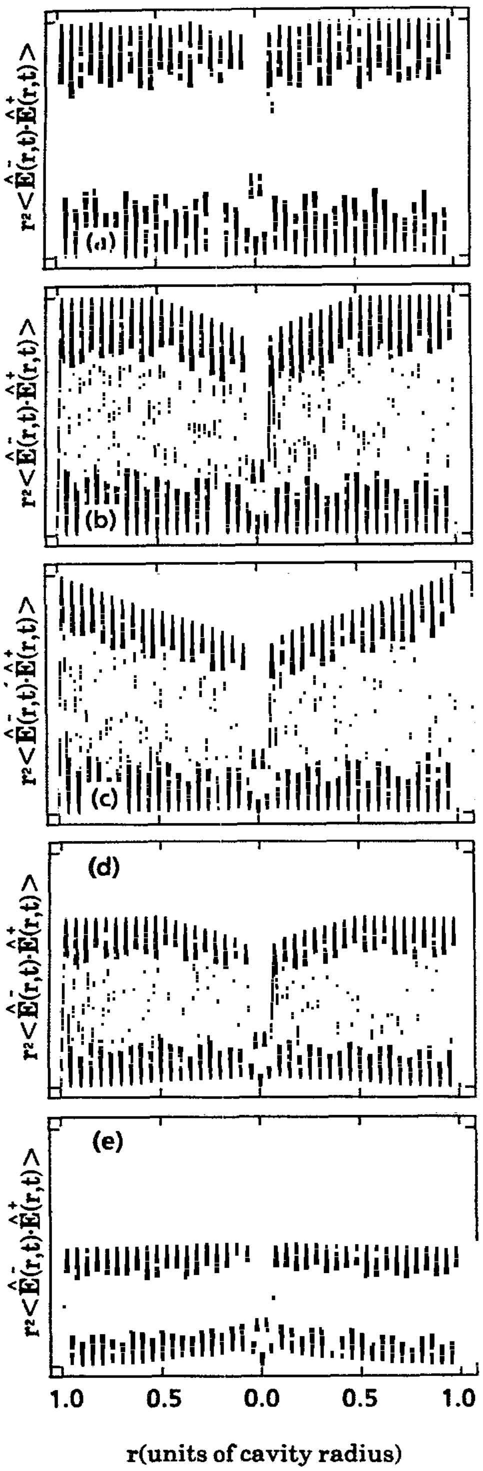

FIG. 1. Expectation value of field intensity vs distance from center of cavity for various times after the absorption begins. (a) The intensity distribution is plotted for $t=0$. (b) The intensity distribution is plotted for a time one-fourth of a cavity-round-trip time. The darkness wave has reached halfway to the cavity wall. (c) The intensity distribution as the darkness wave reaches the wall. (d) The intensity distribution for a time three-fourths of a round-trip time. The outgoing and reflected darkness waves overlap. (e) The intensity distribution after one round-trip time.

distance from the center of the cavity in order to take out the geometrical effects of the spreading wave front. This intensity is plotted as a function of the distance from the center of the cavity where the atom is located. Initially, the field is in a Fock state with one photon in the twentieth mode. The intensity distribution for this case is shown in Fig. 1(a).

The atom then begins absorbing, and in Fig. 1(b) we see the intensity distribution after one-fourth of a cavity round-trip time. The field intensity is reduced out to a distance half way to the cavity wall. A very convenient way of understanding this reduction is to realize that the atom responds to the pre-existing field in the cavity by generating a field which is 180° out of phase with the pre-existing field. This radiated field travels out in the cavity just cancelling the first field. It appears as though a "darkness wave" were propagating out from the atom.[9] The envelope of the darkness wave is approximately linearly decreasing with distance. The reason for this peculiar shape is that the amplitude of the radiated field at the atom is growing linearly with time as would be expected from perturbation theory. As this field propagates out from the atom it produces a field distribution whose *amplitude* decreases linearly with distance. The radiated field is initially much weaker than the pre-existing field. The total intensity of the field in the cavity is the square of the sum of the amplitudes of the two fields, so the largest contribution by the radiated field to the intensity is the cross term containing the product of the amplitude of the pre-existing field and the radiated field. This term is proportional to the amplitude of the radiated field, so it depends linearly on distance from the atom.

Figure 1(c) shows the intensity distribution at one-half a round-trip time in the cavity. The darkness wave has made it to the cavity wall. Of course, the radiated field reflects off of the cavity wall just as any other field does, so that it comes back toward the atom. In Fig. 1(d) we see the field after three-fourths of a round-trip time. The linearly increasing outward-going field combines with the reflected field, which has just the opposite slope, producing a constant field intensity in the overlap region. Figure 1(a) shows the intensity distribution at a time exactly one cavity round trip after the atom began absorbing. The field is approximately uniform throughout the cavity, but with an amplitude reduced from its initial value.

One can continue this analysis for times of the order of a Rabi period or longer, but the results will not in general continue in the simple intuitive fashion they do in the first round trip. As the radiated field constitutes a larger and larger portion of the total field in the cavity, the simple linear envelope shape is lost, and the obvious causality of the darkness wave packet is obscured. How soon this complexity enters the picture depends on the size of the dipole moment for the resonant transition. Over the range of atomic Rydberg states ($10<n<\infty$) the dipole moment varies enough so that the ratio of the Rabi period to the cavity round-trip time ranges from 10^{-8} to 10^{-4}. The example in Fig. 1 is for a ratio of Rabi period to cavity-round-trip time equal to 0.1. We have chosen this unphysically large dipole moment to increase the amplitude of the darkness wave so that it is easier to see in the graphs. In the real cases the linear shape of the darkness wave packet persists over many round trips before it becomes such a large perturbation that quadratic terms become important. There are closely related problems in which the Rabi frequency is large and new types of phenomena occur. These are problems in which the single atom is replaced by a collection of superradiant atoms, or in which the single-photon field is replaced by a highly populated single-mode field. We will not discuss these problems here but will leave them for a later publication.

Our treatment of the transients in the cavity field includes many modes. Ones immediate intuition would suggest that the process of absorption of a single photon from a particular cavity mode does not involve any of the other, initially unpopulated, modes in the cavity. But of course, that is not true. The only way to produce a sharp wave front in the cavity is to excite many modes. As soon as the atom is placed in the resonant state it has a nonzero probability of scattering the single photon into any of the other cavity modes. It is these small but nonzero probability amplitudes that produce the transient darkness wave packet. To illustrate this process we have plotted the logarithm of these probabilities in Figs. 2 and 3.

In Fig. 2(a) we show the probabilities at a time one-twentieth of a cavity round trip time after the process begins. The central spike, which is truncated in the figure, is the probability for a photon in the initially excited twentieth mode. This probability is still almost exactly one. But, there is probability of order 10^{-7} of excitation of all of the other modes up to $n=100$. The structure in the spectrum is determined by two things: the natural line shape of the atomic transition, and the shape of the laser pulse that placed the atom in the resonant lower state. Here we have assumed an instantaneous excitation leading to the sinc-function-like oscillations in the spectrum. In Fig. 2(b) is shown the same energy spectrum at a time one-tenth of a round trip time into the absorption.

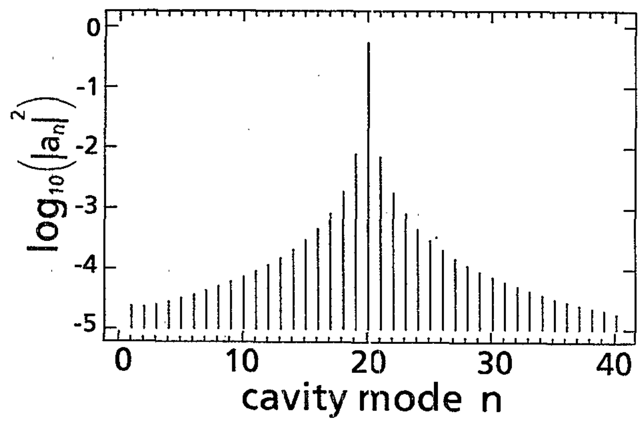

FIG. 3. Energy spectrum of the field after one round trip.

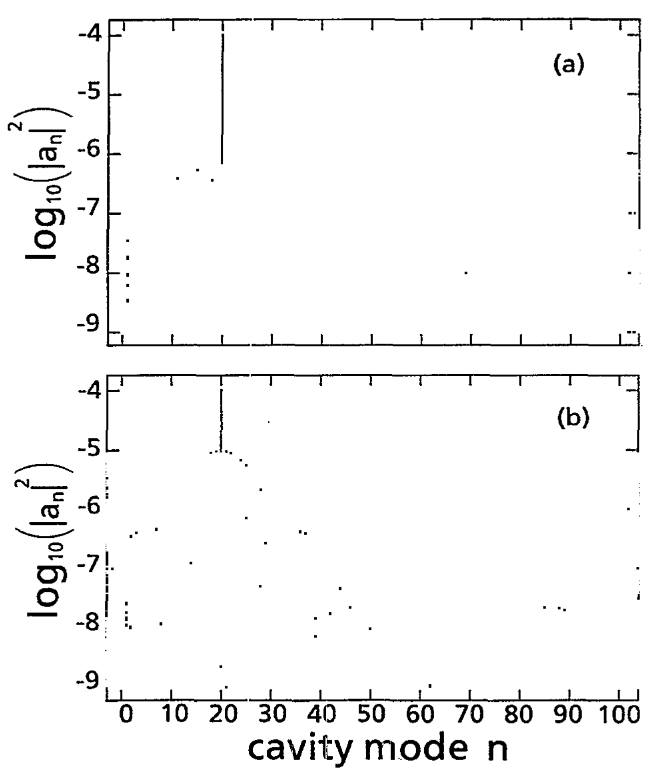

FIG. 2. Energy spectrum of the cavity field. (a) The spectrum after one-twentieth of a round trip. The component at resonance ($n=20$) is truncated. (b) The spectrum after one-tenth of a round trip. The component at resonance is again truncated.

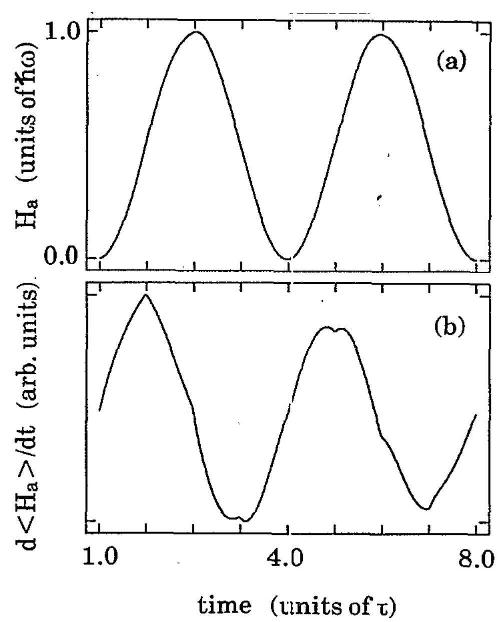

FIG. 4. Response of the atom to the reflected darkness wave packet. Time is measured in units of τ, the cavity-round-trip time. The Rabi period is four round-trip times. (a) The atomic energy is plotted for a period of two Rabi oscillations. The ground-state energy is set to zero. (b) The rate of change of the atomic energy shows kinks at the times when the reflections of the sharp leading edge of the darkness wave packet return from the boundary.

The probabilities of the various modes being excited is growing and narrowing up about the resonant mode. In Fig. 3 the energy spectrum is shown for exactly one cavity-round-trip time into the absorption. The probability of the photon being in the resonant mode is now reduced to about 0.51, while the sum of the probabilities that the photon is in one of the other modes is 0.024, and the probability that the atom is excited and there are no photons is 0.46. The structure due to the sudden turn on is now finer than the mode spacing. The fall off of the energy spectrum is now simply the natural line shape.

In a previous paper (Ref. 7) we described the decay of an excited two-level atom in a high-Q cavity, and the process by which the atom becomes aware of the boundaries of the cavity. The picture that emerged from the discussion showed that the atom learns of the boundary conditions by, in effect, interrogating the boundary with its radiated photon wave packet. In the present discussion the initial conditions are different. As we have seen, the atom nevertheless radiates a wave packet. Moreover, this darkness wave packet propagates to the cavity wall, is reflected, and returns to the atom carrying information about the cavity. To demonstrate this we have plotted in Fig. 4 the expectation value of the atomic Hamiltonian $\langle H_a \rangle$ and its time derivative. In the two-level atom approximation the atomic Hamiltonian is proportional to the excited-state atomic population. The atom is in resonance with the excited mode ($n=20$), and the dipole moment was set such that there are exactly four cavity-round-trip times τ per Rabi oscillation. Two Rabi oscillations are plotted in Fig. 4(a). There is no obvious discontinuity in the energy when the wave packet returns. This is because the total field is not discontinuous at this point; only the slope of the field changes discontinuously. This discontinuity is obvious in the rate of change of the atomic energy however. Plotted in Fig. 4(b) is the rate of energy absorption by the atom $d\langle H_a \rangle/dt$. At integer multiples of the cavity round trip sharp kinks are seen. The kinks are due to the sudden return of the radiated wave packet from the boundary.

By extending the Jaynes-Cummings model to include many modes, we have shown that the decrease in cavity field intensity due to transient absorption propagates as a causal wave packet. The wave packet is created by scattering of the initial photon into other cavity modes. The population in these nonresonant modes never becomes large, but their collective effect is essential for maintaining causality in the field propagation. It should be kept in mind that we are describing a state with a single photon in the cavity. The darkness wave carries the information that the probability of detecting a photon in a measurement of the field at point r is decreased from its original value. Since the wave packet is actually the absence of a photon it may properly be described as a "darkness wave packet."

Note added in proof. See also Krökel *et al.*[10]

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[1]J. Parker and C. R. Stroud, Jr., Phys. Rev. Lett. **56**, 716 (1986).

[2]G. Alber, H. Ritsch, and P. Zoller, Phys. Rev. A **34**, 1058 (1986).

[3]J. Parker and C. R. Stroud, Jr., Phys. Scr. **T12**, 70 (1986).

[4]J. A. Yeazell and C. R. Stroud, Jr., Phys. Rev. A **35**, 2806 (1987).

[5]J. Parker and C. R. Stroud, Jr. (unpublished).

[6]M. J. Davis and E. J. Heller, J. Chem. Phys. **71**, 3383 (1979).

[7]J. Parker and C. R. Stroud, Jr., Phys. Rev. A **35**, 4226 (1987).

[8]H. A. Lorentz and E. Schrödinger, in *Letters on Wave Mechanics,* edited by K. Przibram (Philosphical Library, New York, 1967), pp. 55–75.

[9]We have been unable to locate any direct references to "darkness waves" in the literature, but the basic ideas are contained in A. V. Durant, Am. J. Phys. **44**, 630 (1976) [reprinted in *Concepts of Quantum Optics,* edited by P. L. Knight and L. Allen (Pergamon, New York, 1983), p. 155].

[10]D. Krökel, N. J. Hallas, G. Giuliani, and D. Grischkowsky, Phys. Rev. Lett. **60**, 29 (1988).