Inhibited Spontaneous Emission

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The radiative properties of an atom in a cavity differ fundamentally from the atom's radiative properties in free space. Spontaneous emission is inhibited if the cavity has characteristic dimensions which are small compared to the radiation wavelength, and enhanced if the cavity is resonant. The cavity causes slight shifts in the energies of the atom, analogous to radiative shifts. Experiments are proposed for observing these effects.

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Spontaneous emission is the most visible manifestation of the dynamical interaction between matter and the vacuum; the primal role of vacuum fields in driving every excited atom to its ground state is often regarded as a basic fact of nature. The purpose of this Letter is to point out that vacuum states can be radically altered by a cavity and that it should be possible to realize experimental conditions in which an atom is effectively decoupled from the vacuum and cannot radiate. In such a situation the atom's excited state is, to first approximation, a real eigenstate which is devoid of the usual radiative energy width. The energy levels are slightly altered by the cavity, and the Lamb shift is modified. By changing the experimental conditions, the rate of spontaneous emission can be vastly increased compared to the free-space value.

Our starting point is the observation made many years ago by Purcell[1] that the spontaneous emission rate A for a two-state system is increased if the atom is surrounded by a cavity tuned to the transition frequency, ν. If the quality factor of the cavity is Q, then the spontaneous rate in the cavity is $A_c \simeq QA$. Physically, the cavity enhances the strength of the vacuum fluctuations at ν, increasing the transition rate. Conversely, the decay rate decreases when the cavity is mistuned.[2] If ν lies below the fundamental frequency of the cavity, spontaneous emission is significantly inhibited. In the case of an ideal cavity, no mode is available for the photon and spontaneous emission cannot occur.

The rate A for spontaneous transitions from initial state $|i\rangle$ to final state $|f\rangle$ can be written by the "golden rule"[3]

$$A = [\,|\langle f|H|i\rangle|^2/\hbar^2]\rho(\nu)\,.$$

Here $|i\rangle$ is the excited state of the atom in the absence of any photons, $|f\rangle$ is the final state of the atom with a single photon, H is the Hamiltonian for the atom-field interaction, $\rho(\nu)$ is the density of photon states, and the matrix element is volume normalized. In free space $\rho_s = 2(4\pi\nu^2/c^3)$; the factor of 2 is due to the two allowed polarization states. In a cavity the density of modes is essentially (1 mode)$\Delta\nu^{-1}V^{-1}$, where $\Delta\nu = \nu/Q$ is the cavity's resonance width and V is the cavity's volume. Taking $V \simeq (\lambda/2)^3 = (c/2\nu)^3$, we have $\rho_c \simeq 8\nu^2 Q/c^3$. Under the assumption that the cavity mode is doubly degenerate, then $\rho_c/\rho_s \simeq (2/\pi)Q$ and $A_c \simeq QA$ as previously described.

The presence of any conducting surfaces near the radiator affects the mode density and the radiation rate. Parallel conducting planes can somewhat alter the emission rate but can only reduce the rate by a factor of 2 because of the existence of TEM modes[4] which are independent of the separation.[2] In order to eliminate spontaneous emission every propagating mode must be suppressed. A completely enclosed perfectly conducting cavity would accomplish this, but a waveguide below cutoff could also serve the purpose. The waveguide, which can be viewed as a cavity with ends removed to infinity, is experimentally attractive because atoms can pass through it freely.

The wave vector for the fundamental mode of a waveguide is $k = (2\pi/c)(\nu^2 - \nu_0^2)^{1/2}$, where ν_0 is the fundamental guide cutoff frequency. Imposing periodic boundary conditions, $kL = 2\pi n$, where L is a length long compared to the diameter of the waveguide, yields for the fundamental mode

$$\rho_0(\nu) = \frac{2}{V}\frac{dn}{d\nu} = \frac{4}{cA_g}\frac{\nu}{(\nu^2 - \nu_0^2)^{1/2}}\,.$$

A_g is the area of the guide; $A_g \simeq \zeta(c/2\nu_0)^2$, where ζ is a numerical constant of order unity. The factor of 4 results from the twofold degeneracy

between positive and negative values of n, and the twofold degeneracy in polarization which is appropriate for a circular waveguide. Near cutoff the waveguide causes a resonancelike enhancement of the spontaneous emission rate. If $\nu < \nu_0$, $\rho_0(\nu) = 0$ and spontaneous emission cannot occur.

Higher modes in the waveguide contribute to the spontaneous rate for frequencies above their cutoff values. If ν_j is the cutoff frequency for the jth mode, then the total mode density $\rho_g(\nu)$ is

$$\rho_g(\nu) = \frac{16\nu_0^2}{\zeta c^3} \sum_j \frac{\nu}{(\nu^2 - \nu_j^2)^{1/2}} .$$

The sum is over all modes for which $\nu_j < \nu$. As the frequency increases and more and more modes are included, $\rho_g(\nu)$ approaches the free-space mode density, as Fig. 1(a) shows. The cavity changes the spontaneous emission rate by the ratio $R(\nu) = \rho_g(\nu)/\rho_s(\nu)$, which is plotted in Fig. 1(b).

A realistic treatment of spontaneous emission in a waveguide must take into account resistive losses. (Radiative losses are expected to be small because of the guide's large length-to-diameter ratio.) The quantum mechanical theory of radiation in a damped system is beyond the scope of this Letter, but the important features can be understood from simple qualitative arguments. In the vicinity of cutoff a waveguide can behave like a cavity with $Q \simeq a/\delta$, where a is the characteristic dimension of the waveguide and δ is the skin depth. As a result the singularities in $R(\nu)$ become finite with a height of approximately Q. There is a nonzero response in the cutoff region, $\nu < \nu_0$, where $R(\nu) \sim 1/Q$. Thus the spontaneous decay rate is decreased compared to free space by a factor of order Q in the region $\nu < \nu_0$, and increased by a factor of Q in the regions $\nu \simeq \nu_0$.

The experimental challenge in demonstrating inhibited spontaneous emission is to find a system which has a spontaneous transition with a wavelength long enough to allow construction of a practical waveguide, but a lifetime short enough for the atom to radiate before escaping from the apparatus. Electric dipole transitions between Rydberg states can satisfy these requirements. In order to avoid radiative decay by optical or other short-wavelength transitions, it is essential to employ states which have but one allowed radiative decay channel. This suggests employing states of the maximum angular momentum, $l = n - 1$, where n is the principal quantum number. To make the discussion concrete, we shall consider the transition $|n, n-1, n-1\rangle \rightarrow |n-1, n-2, n-2\rangle$, where the quantum numbers are $|n, l, m\rangle$. This transition corresponds classically to dipole radiation by a charge in a circular orbit. The wavelength and spontaneous lifetimes are approximately $\lambda = 4.6 \times 10^{-6} n^3$ cm and $\tau = 9.3 \times 10^{-11} n^5$ s. For example, for the transition $n = 25 \rightarrow 24$, $\lambda = 0.07$ cm and $\tau = 900$ μs. A typical thermal atom travels 40 cm in this lifetime. The cutoff diameter of the waveguide is 0.04 cm, and a Q in excess of 10^3 should be attainable.

Spontaneous emission for $\Delta m = -1$ transitions by atoms near the axis of a circular waveguide occurs via coupling to the $TE_{1,j}$ and $TM_{1,j}$ modes.

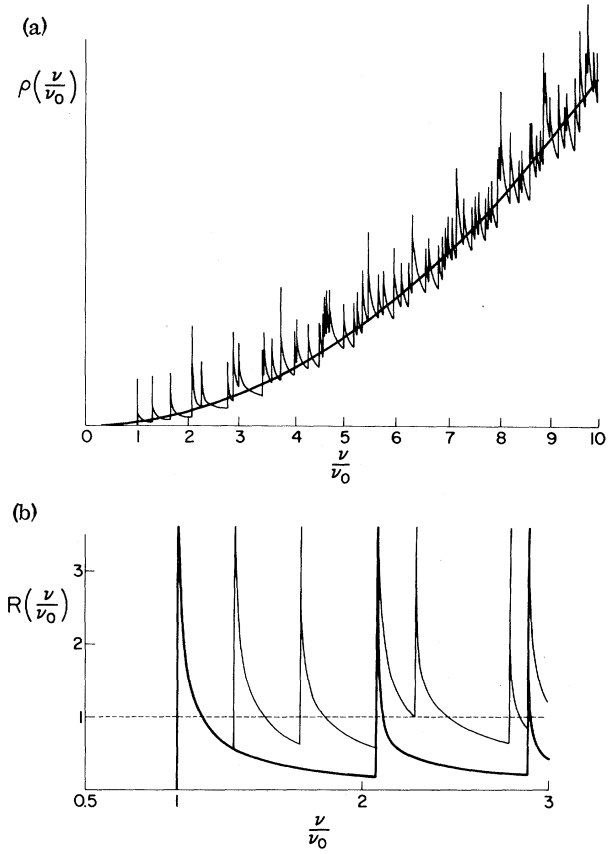

FIG. 1. (a) Mode density in a perfectly conducting cylindrical waveguide. Frequency is in units of the lowest cutoff frequency of the waveguide, $\nu_0 = 0.29c/a$, where a is the radius. For clarity, the singularities have been truncated. The smooth curve represents the mode density in free space. (b) Ratio of waveguide mode density to free-space mode density. The heavy line is for the modes which couple to a $|\Delta m| = 1$ transition for an atom on axis: $TE_{1,j}$ and $TM_{1,j}$. The change in the spontaneous emission rate from the free-space value is proportional to the ratio $R(\nu)$.

The density ratio $R(\nu)$ for these modes is shown in Fig. 1(b). (It is interesting to note that the spontaneous emission rate into these modes can also be inhibited by a significant factor, greater than 4, in several frequency ranges above ν_0.) In principle, spontaneous emission due to quadrupole and higher-order processes can occur even when the dipole radiation is inhibited by the cavity, but their rates are so low that they should not cause difficulty. Another possible source of confusion, stimulated emission by blackbody radiation, can be avoided by operating the cavity at cryogenic temperatures.

Observing enhanced spontaneous emission is a much simpler experimental task than observing inhibited emission; one can emply high-n transitions whose natural lifetimes are so long that spontaneous emission would normally be unobservable. This allows a general scaling up of the waveguide diameter and relieves the need to employ the highest angular momentum states. Such an experiment is similar in concept to the "one-atom maser" proposed by Haroche.[5] The one-atom maser consists of a Rydberg atom in a very-high-Q etalonlike resonator. The distinction between the two proposals lies in the choice of cavity mode. In a fundamental mode cavity the spontaneous rate is completely determined by the cavity; there is no threshold for enhancement. For a high-mode optical resonator, enhanced emission into a single mode must compete with spontaneous emission in all the other modes of space, and it can only be observed if the cavity Q is sufficiently large.

Numerous experimental schemes are possible for studying the effect of a waveguide on the spontaneous emission rate. The initial- and final-state distribution can be determined by selective field ionization,[6] a sensitive technique which works well at the single-atom level. The primary problem in observing inhibited spontaneous emission is to produce Rydberg atoms in the "circular" state. A number of schemes are possible and experimental work is in progress.

An atom surrounded by a cavity forms a coupled quantum system, and the atom's energy levels are inevitably shifted by the cavity. The leading interaction is essentially a dipolar Van der Waals attraction between the atom and the conducting walls. As Casimir and Poulder pointed out in a well-known paper,[7] Van der Waals interactions arise from alterations in the zero-point energy of a system. An atom in a cavity has many features in common with the hypothetical system Casimir and Poulder employed to motivate their discussion: an atom interacting with an infinite perfectly conducting plane. The behavior of an atom in a cylindrical waveguide can be understood qualitatively by considering an atom at the center of a spherical cavity. Such a model gives the correct order of magnitude of the level shifts, though the perturbation does not have the proper angular dependence.

Consider a one-electron atom with an instantaneous dipole moment $-e\vec{r}$ centered in a cavity of radius a. The induced field is $\vec{E} = -e\vec{r}/a^3$, and the atom-field interaction is $V = -e^2r^2/a^3$. For a "circular" state $r^2 \simeq n^4a_0^2$, where a_0 is the Bohr radius, and $V = -(e^2/a_0)(a_0/a)^3n^4$. The energy shift between level n and $n-1$ is $\Delta V \simeq -4(e^2/a_0)(a_0/a)^3n^3$. If the cavity is near cutoff, then $a \simeq \lambda/2\pi \simeq n^3a_0/\alpha$ and $\Delta V \simeq -4(e^2/a_0)\alpha^3n^{-6}$. The fractional change in energy is $4\alpha^3n^{-3}$. For $n = 25$, the fractional change is 10^{-10}. By operating on a high-n transition in a small-diameter cavity, the fractional shift can be enhanced. It is interesting to note that retardation corrections to the Van der Waals interaction are negligible since the radiative modes are suppressed. In principle the cavity also alters the Lamb shift, though the effect is very small.

Experimental studies of inhibited spontaneous emission seem worthwhile in view of the novelty of the situation and the ongoing controversies about the physical nature of the vacuum fields.[8] Any scheme for suppressing spontaneous emission is potentially useful to the art of precision measurements, though perturbations due to the atom-cavity interaction may limit such applications. In any case, observations of inhibited or enhanced spontaneous emission would provide an interesting bridge between atomic physics and macroscopic electrodynamics.

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Polarized Sodium Nuclei Produced by Laser Optical Pumping with Velocity Changing Collisions

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Theoretical and experimental studies of nuclear polarization induced by single-mode–laser optical pumping are reported. The nuclear polarization is shown to depend on the product of laser intensity and buffer-gas pressure. In ^{23}Na it approaches one at modest laser intensities (~ 100 mW/cm^2) for pressures as small as a few hundred millitorr.

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It has recently been emphasized that achieving complete nuclear polarization in dense alkali vapors is important for producing intense polarized light-ion beams by charge exchange[1] and polarized targets for parity-nonconservation studies.[2] The present paper demonstrates that single-mode–laser optical pumping in combination with velocity-changing collisions (vcc) can be used to attain a high degree of nuclear polarization in alkali vapors. General theoretical results are presented for alkalilike systems and confirmed in sodium, where almost complete nuclear polarization is achieved. Several authors have studied vcc processes in laser saturation spectroscopy[3,4] and in laser optical pumping.[5,6] Complete optical pumping has been achieved at high buffer-gas pressures (collision-broadened regime, where vcc processes have no influence),[7,8] but the use of laser optical pumping with vcc, at very low buffer pressures, to produce nuclear polarization has not been studied in detail previously.

The laser optical-pumping process, in which nuclear polarization is achieved through the coupling between electron and nuclear spins, has been referred to as laser-induced nuclear orientation (LINO).[9] Ordinarily, single-mode laser radiation interacts only with a small fraction of the thermal velocity distribution, of order $\gamma/ku \sim 10^{-2}$, with γ and ku the homogeneous and Doppler widths, respectively. This results in poor Doppler coverage, hence incomplete nuclear polarization. Furthermore, in alkali atoms the atomic population is distributed in two ground-state hyperfine (hf) levels, the Doppler-broadened absorption profiles of which overlap in the cases of Li and Na.

The present experiments use vcc induced by argon buffer-gas atoms to attain single-mode laser optical pumping of the entire ground-state population. In the vcc regime velocity changes cause every alkali atom to be at the resonant velocity at some time during its diffusion through the laser beam. Furthermore, this spatial diffusion considerably retards wall relaxation and greatly extends the laser-atom interaction time. Because of large collision cross sections high vcc rates can be achieved at low (< 1 Torr) buffer pressures, where collisional broadening is negligible.

Consider a gas of three-level atoms with closely spaced or degenerate levels 1 and 2 optically con-

