Suppression of Spontaneous Decay at Optical Frequencies: Test of Vacuum-Field Anisotropy in Confined Space

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The radiative decay of Cs atoms excited into the $5D_{5/2}$ level and passing between two metallic mirrors spaced by a 1.1-μm gap is observed to depend upon their angular momentum. Spontaneous emission at a wavelength of 3.49 μm is suppressed for the substates with maximum angular momentum normal to the mirrors, which survive without substantial decay during $\sim$13 natural lifetimes. The radiation rate is modified by application of a magnetic field which mixes sublevels having different lifetimes in the gap. This experiment illustrates the anisotropy of the vacuum field in confined space.

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Spontaneous emission from an excited electronic state reflects the properties of the surrounding vacuum-field fluctuations. By placing the radiator near a metallic surface or in a cavity, one can modify the spectral density of these fluctuations and either enhance[1] or inhibit[2] spontaneous emission. Evidence for such effects came first from fluorescence-rate measurements performed on complex molecules radiating near a surface.[3] Interest in these phenomena was recently renewed when it became possible to study them with higher precision on simpler, more fundamental, and better controlled systems. Cavity-induced enhancement[4] and inhibition[5] of spontaneous emission have been observed in microwave transitions between Rydberg levels of alkali-metal atoms and between cyclotron levels of a free electron in a Penning trap.[6] In those low-frequency experiments the cavity sizes were of order one-tenth of a millimeter[5] or larger[6] and the spontaneous-emission rates to be modified by the cavity were quite small (10- to 10^3-s^{-1} range).

In this Letter, we report the suppression of spontaneous emission from an excited atom at a much higher frequency (near infrared, 3.49-μm wavelength radiation) and in a cavity a hundred to a thousand times smaller. The excited level ($5D_{5/2}$ in Cs) is a tightly bound electronic state having a natural lifetime in the microsecond range ($\tau_0=\Gamma_0^{-1}=1.6\ \mu$s).[7] The atoms are passed through an 8-mm-long metallic tunnel made of two parallel mirrors, separated by a 1.1-μm gap. Spontaneous emission is turned off for some substates of the $5D_{5/2}$ level which become stable during the time they are in the tunnel. Other sublevels have their lifetimes modified to a lesser extent and decay before emerging from the cavity. We have used a magnetic field, making an angle with the direction normal to the mirrors, to study the dependence of the spontaneous-emission rate upon the polarization of the excited atoms in the gap. This has allowed us to demonstrate the anisotropy of the vacuum-field fluctuations between the two closely spaced mirrors.

The Cs energy levels relevant for our experiment are shown in Fig. 1(a). The $5D_{5/2}$ level decays with a branching ratio of 1 to the $6P_{3/2}$ level, emitting 3.49-μm

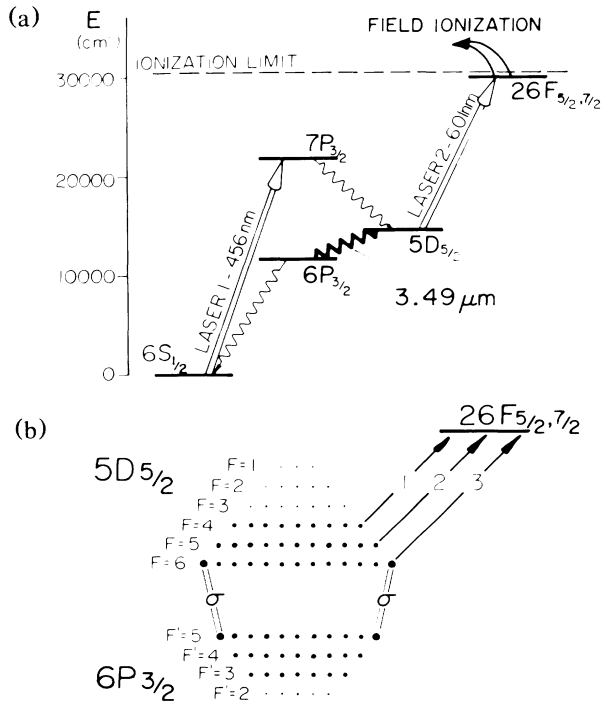

FIG. 1. (a) Cesium energy diagram showing the levels and optical transitions relevant to this experiment. (b) Closeup showing the hyperfine structure of the $5D_{5/2}$ and $6P_{3/2}$ states.

radiation which can be cut off in a micron-wide cavity. We define the normal to the mirrors as the "quantization axis" Oz. The radiation rate Γ_{F,M_F} of each $|F,M_F\rangle$ hyperfine sublevel of the $5D_{5/2}$ state [$F=1$ to 6, $-F \leq M_F \leq F$; see Fig. 1(b)] can be divided into a σ and a π contribution, corresponding respectively to the emission of photons polarized parallel and perpendicular to the mirror surface ($\Gamma_{FM_F}=\Gamma^{(\sigma)}_{FM_F}+\Gamma^{(\pi)}_{FM_F}$). σ and π contributions are due respectively to $\Delta M_F=\pm 1$ and $\Delta M_F=0$ transitions to the final $6P_{3/2}$, $F'M'_F$ state ($F'=2$ to 5). In free space the total emission rate is the same for all substates, because the vacuum fluctuations are isotropic ($\Gamma_{FM_F}=\Gamma_0$). Between two ideal mirrors with a spacing $d<\lambda/2$, the vacuum-field mode density drops to zero for σ polarization, whereas it is enhanced by the factor $3\lambda/4d$ (2.38 in our experiment) for π polarization. Each $|F,M_F\rangle$ substrate thus acquires a modified emission rate $\Gamma^{(\mathrm{cav})}_{FM_F}=(3\lambda/4d)\Gamma^{(\pi)}_{FM_F}$ which, depending upon the ratio of the π to σ emission channels for this particular state, can be either smaller or larger than Γ_0. Of special interest are the $F=6$, $M_F=\pm 6$ states which decay only via σ transitions to the $|6P_{3/2}, F'=5, M_{F'}=\pm 5\rangle$ sublevels. The radiation rate for these states is $\Gamma^{(\mathrm{cav})}_{6,\pm 6}=0$. For some other substates the rates are $\Gamma^{(\mathrm{cav})}_{6,\pm 5}=0.40\Gamma_0$; $\Gamma^{(\mathrm{cav})}_{6,\pm 4}=0.72\Gamma_0$; ...; $\Gamma^{(\mathrm{cav})}_{6,0}=1.3\Gamma_0$; $\Gamma^{(\mathrm{cav})}_{5,\pm 5}=0.56\Gamma_0$; $\Gamma^{(\mathrm{cav})}_{4,\pm 4}=0.96\Gamma_0$....

An equivalent way of looking at these effects is to consider the interaction of the atomic dipole with its images in the mirrors.[8] The interaction with the walls not only modifies the emission process (dissipative part of the coupling) but also induces energy shifts of the atomic states (dispersive part). The derivative of these shifts with respect to the atomic position corresponds to the van der Waals force pulling the atom toward the mirrors. In our tunnel, this force is estimated to deflect about 80% of the thermal Cs beam onto the surfaces. Spontaneous-emission suppression can be observed only on the 20% of the atoms which escape collision with the walls. The length l of our tunnel is a compromise: It is much longer than the 0.6-mm distance traveled by the atoms during the $5D_{5/2}$ natural lifetime, yet short enough that we do not lose all the atoms to the walls through the van der Waals attraction.

Our setup is sketched in Fig. 2. The experiment is performed on a Cs atomic beam. We employ two single-mode cw lasers, one for preparation and one for detection of the atoms in the $5D_{5/2}$ level. Laser 1 (Coherent Radiation Innova 20/CR 699-21 dye laser system) is tuned to 456 nm and excites Cs from the ground state, $6S_{1/2},F=4$, to the $7P_{3/2},F=5$ hyperfine level, from which about 13% of the atoms are transferred by spontaneous cascade into the $5D_{5/2}$ level. Laser 1 can be frequency-locked to the Cs transition with the help of an auxiliary Cs beam. The population of the $5C_{5/2}$ level is monitored with laser 2 (Spectra Physics 171/380D dye laser system) tuned around 601 nm, which excites the atoms from the $5D_{5/2}$ level into the $26F$ Rydberg level.

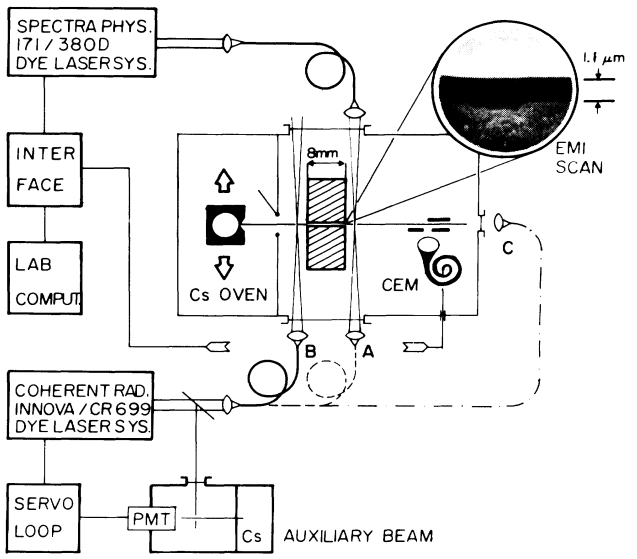

FIG. 2. Experimental setup. Inset: A scanning electron microscope picture of the 1.1-μm tunnel exit.

This state is ionized by a 1-kV/cm electric field applied in front of a Channeltron electron multiplier. By scanning ν_2 across the $5D_{5/2}\rightarrow 26F$ line and recording the electron count rate, we measure the relative populations of the various hyperfine sublevels F of the $5D_{5/2}$ state. Both laser beams are guided to the apparatus by optical fibers. The beam waists of the cylindrically focused fiber outputs intersect the atomic beam along 5-mm-high, 30-μm-wide lines parallel to the tunnel entrance and exit slits.

The mirror tunnel[9] is made from two fused-silica blocks stacked against each other but separated by thin Ni foil spacers. The 30×8-mm^2 mirror surfaces are plane to better than a few hundred angstroms over their whole area and are coated with a 300-Å-thick gold coating, providing a 96% reflection coefficient for 3.49-μm radiation. The blocks have very sharp edges as shown in the insert of Fig. 2, which presents an electron microscope image of a portion of the tunnel exit. The blocks are pressed against each other by springs whose tension is adjusted until the desired 1.1-μm spacing is achieved. The mirrors are cooled to about 5 °C so that most of the Cs atoms colliding with the tunnel walls stick to them and do not scatter randomly inside the gap.[9]

The tunnel subtends an angle 1.4×10^{-4} radian so that our 25-μm-wide Cs oven slit (15 cm away from the mirrors) must be centered with a precision of ± 15 μm on the tunnel axis. Final alignment is performed by our optimizing the Cs atom transmission. For this purpose the optical fiber for laser 1 is placed in the downstream position A (see Fig. 2) after the tunnel exit and lasers 1 and 2 are superimposed so that the 456- and 601-nm transitions are simultaneously driven in atoms emerging from the gap. Now the lasers and the Channeltron system

merely serve to detect ground-state Cs atoms. Translating the oven in the direction perpendicular to the tunnel axis gives a triangular atomic transmission profile and we set the oven position for maximum signal.[9]

The rms velocity $\bar{v}$ of the atoms transmitted by the tunnel is an important parameter for our data analysis. In order to measure it, the beam from laser 1 is made collinear with the atomic beam by our moving the fiber into position C (see Fig. 2) and its frequency ν_1 is tuned across the $(6S_{1/2},F=4)\rightarrow(7P_{3/2},F=5)$ line. This provides a Doppler-shifted spectrum from which we deduce the velocity distribution of the atoms emerging from the gap. For an oven temperature $T=350$ K we find $\bar{v}=390$ m/s, much larger than the 300 m/s measured when a wide slit is used to collimate the beam. We attribute the increase in $\bar{v}$ to the fact that slow atoms in the thermal distribution are more likely to collide with the mirrors as a result of the van der Waals attraction. The average tunnel crossing time for the detected atoms is $\tau=l/\bar{v}=20.5\ \mu s=12.8$ natural lifetimes of the $5D_{5/2}$ state.

Figure 3(a) displays a recording of the spectrum obtained with laser 2 back in position A, by our tuning ν_2 across the $5D_{5/2}\rightarrow 26F$ line while keeping ν_1 fixed on the $(6S_{1/2},F=4)\rightarrow(7P_{3/2},F=5)$ transition. A 2.4-G magnetic field is applied perpendicular to the mirrors (see below). The $7P_{3/2},F=5$ level can only decay to the $F=6$, 5, and 4 hyperfine levels of the $5D_{5/2}$ state and thus only the three corresponding components appear in the absorption spectrum. The Zeeman components of the hyperfine lines are not resolved in the 2.4-G field. The largest peak (absorption from $F=6$) corresponds to an electron counting rate of about 2000 cps, for an oven temperature of 350 K. We estimate that we detect one atom out of 500, so that about 10^6 atoms per second actually emerge from the gap.

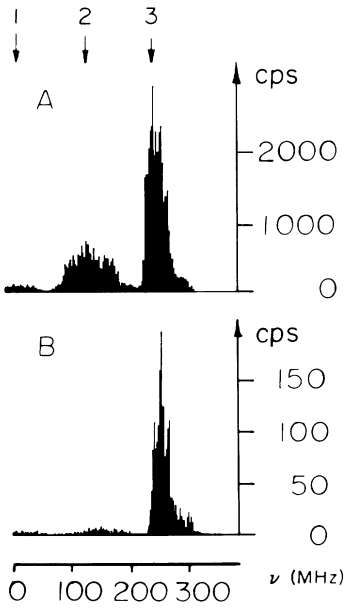

FIG. 3. Spectra of the $5D_{5/2}\rightarrow 26F$ transition recorded with laser 1 respectively in positions A and B (see Fig. 2). Recording B is evidence for suppression of spontaneous decay from the $5D_{5/2}$, $F=6$ level in the gap. (Vertical axis: Channeltron counting rate; hyperfine components 1, 2, and 3 defined in Fig. 1.)

Figure 3(b) shows the spectrum obtained under the same conditions as Fig. 3(a), except the laser 1 is now moved upstream into position B, exciting the atoms into the $5D_{5/2}$ level before they enter the tunnel. This signal is due to atoms that have survived the tunnel crossing —12.8 natural lifetimes—without radiating. The probability for a Cs atom in the $5D_{5/2}$ level to stay excited that long in free space is only 2.8×10^{-6}. Since the signal essentially consists of the single peak No. 3, we observe that spontaneous emission is suppressed only for the highest angular-momentum state, $F=6$. We estimate that the excitation process prepares at most 68% of the F=6 atoms in the long-lived $M_F=\pm6$ states. Of these, no more than 15% survive the propagation in free space before entering the gap and between the gap exit and laser beam 2 (1.2 mm total distance). Combining these two figures with the measured Fig. 3(b)-to-Fig. 3(a) peak ratio of $(7\pm1)\%$, we arrive at an upper limit of $0.4\Gamma_0$ for the $M_F=\pm6$ state radiation rate between the mirrors. The actual decay rate is probably smaller because this analysis neglects other loss factors, some of which are discussed below.

The magnetic field dependence of the $5D_{5/2}$ state transmission through the tunnel provides a dramatic test of the anisotropy of the spontaneous-emission process between the mirrors. The resonance of Fig. 3(b) completely disappears when the directing field perpendicular to the mirrors is not applied, whereas the corresponding spectrum in Fig. 3(a) is not modified. In the absence of the directing field, the atoms are in fact subjected in the gap to the residual laboratory field $B_x=0.36$ G, which is parallel to the mirrors. This field mixes the $M_F=\pm6$ state with the much shorter-lived $|M_F|<5$ states of the $F=6$ hyperfine level. As a result, the effective lifetime of the $M_F=\pm6$ states is reduced to the extent that no excited atoms survive the gap. We have studied this magnetic mixing effect by recording the excited-state transmission signal as a function of the angle θ between the total magnetic field **B** and the direction normal to the mirrors. The results are shown in Fig. 4. When the field component B_z along Oz is much larger than B_x, **B** is essentially normal to the mirrors ($\theta\sim0$ or $\theta\sim180°$) and a large excited-state transmission is observed. When B_z/B_x is reduced, the signal diminishes, practically vanishing when the angle between **B** and Oz is larger than 30°. The solid line in Fig. 4 corresponds to a calculation in which we have used the Lamb-Bethe theory,[10] together with the $\Gamma^{(cav)}_{FM_F}$ rates discussed above, to compute the effective lifetime of the magnetically mixed levels. (We have also taken into account the change in the relative

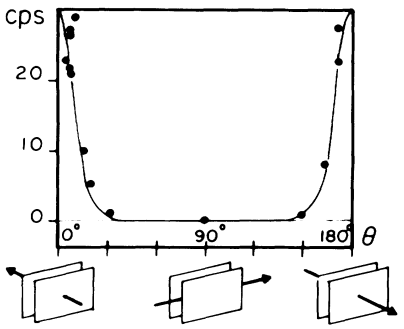

FIG. 4. Excited-state transmission through the tunnel vs the angle θ between the magnetic field and the normal to the mirrors. Solid line indicates theory normalized to maximum counting rate.

preparation efficiencies of the F, M_F states when the angle between **B** and the direction of laser 1 is varied.) The good agreement between theory and experiment indicates that we are in effect measuring here the emission rates $\Gamma_{FM_F}^{(cav)}$ (for $F=6$, $|M_F| \leq 5$).

Finally, we discuss the effect of atomic collisions on our results. The residual pressure of our vacuum chamber is 5×10^{-8} Torr, but we suspect that the background pressure in the tunnel is higher. We attribute the small residual signal from the $F=4$ and $F=5$ hyperfine levels in Fig. 3(b) to collisional transfer of atoms from the long-lived $M_F=\pm6$ states in the last few millimeters before detection. After running the experiment for several hours, we observe a drop of the excited-state transmission signal, probably related to buildup of cesium in the gap. The signal is restored by temperature recycling of the mirrors.

This experiment shows that by the collimation of atomic beams through very narrow slits, it is possible to study how fundamental atomic processes are modified by the boundary conditions of a confined space surrounding the atom.

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[1]E. M. Purcell, Phys. Rev. **69**, 681 (1946).

[2]D. Kleppner, Phys. Rev. Lett. **47**, 233 (1981).

[3]K. H. Drexhage, in *Progress in Optics XII,* edited by E. Wolf (North-Holland, New York, 1974); see also F. DeMartini and G. Innocenti, in *Quantum Optics IV,* edited by J. D. Harvey and D. F. Walls (Springer-Verlag, New York, 1986), who have recently performed experiments on liquid dyes excited between mirrors.

[4]P. Goy, J. M. Raimond, M. Gross, and S. Haroche, Phys. Rev. Lett. **50**, 1903 (1983).

[5]R. G. Hulet, E. S. Hilfer, and D. Kleppner, Phys. Rev. Lett. **55**, 2137 (1985).

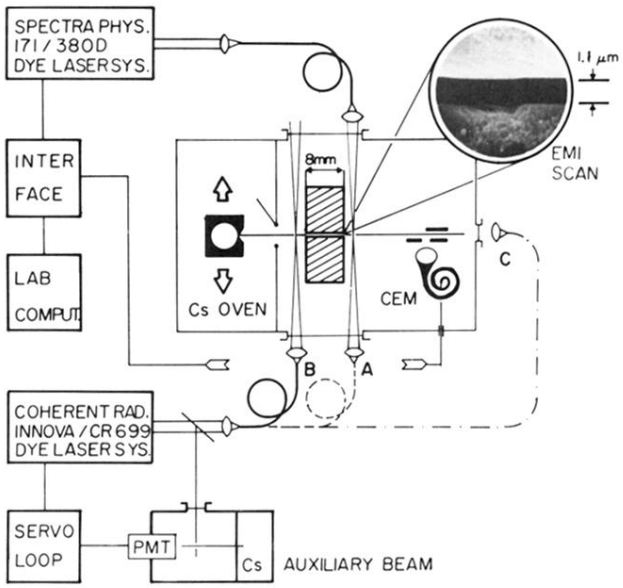
[6]G. Gabrielse and H. Dehmelt, Phys. Rev. Lett. **55**, 67 (1985).

[7]Previous to this work, τ_0 was determined only by theory [$\tau_0=1.43$ μs, with the use of the oscillator strength of M. Fabry, J. Quant. Spectrosc. Radiat. Transfer **16**, 127 (1976)]. In a time-of-flight measurement we have found $\tau_0=1.6(1)$ μs, in reasonable agreement with theory.

[8]P. W. Milonni and P. L. Knight, Opt. Commun. **9**, 119 (1973).

[9]A. Anderson, S. Haroche, E. A. Hinds, W. Jhe, D. Meschede, and L. Moi, Phys. Rev. A **34**, 3513 (1986).

[10]W. E. Lamb, Jr., and R. C. Retherford, Phys. Rev. **79**, 41 (1950).



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