Vacuum Radiative Level Shift and Spontaneous-Emission Linewidth of an Ato.n in an Optical Resonator

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(Received 24 August 1987)

The center frequency and linewidth of the $^1S_0$-$^1P_1$ resonance line of barium atoms placed near the center of a concentric optical resonator are studied as functions of cavity tuning. Shifts in the transition center frequency, due to radiative level shifts, and changes in linewidth, due to enhanced and suppressed spontaneous emission, are observed. A QED calculation which explicitly includes the resonator mode density gives good agreement with the data.

PACS numbers: 31.30.Jv, 32.70.Jz, 32.80.−t, 42.50.−p

The radiative decay and level shifts of atomic states are well understood when the atom is in free space, beginning with the work of Weisskopf and Wigner[1] for the description of spontaneous emission and with Bethe's calculation[2] of the Lamb shift for radiative level shifts. Recently, much attention has focused on the changes in spontaneous-emission rates which occur for an atom in a cavity, and this has now been demonstrated experimentally.[3-7] In addition to changes in spontaneous-emission rates, it may be expected that radiative level shifts will also be modified when an atom is placed in a cavity.[8] This paper presents the first observation of changes in the radiative level shift of an atom in a resonator. We also directly observe, for the first time, changes in the natural linewidth of a transition. A QED calculation, which explicitly includes the resonator density of modes, is presented and gives good agreement with the results.

The changes in radiative processes which occur in a resonator may be attributed to a change in mode density. In particular, the spontaneous-emission rate Γ and level shift $\delta\omega$ of an atom in the excited state $|E\rangle$ are given by[9]

$$\Gamma = 2\pi \int\int \frac{|\boldsymbol{\mu}_{EG}\cdot\boldsymbol{\epsilon}_k|^2}{\hbar^2}\frac{2\pi\hbar\omega_k}{V}\delta(\omega_{EG}-\omega_k)\rho(\omega_k,\mathbf{k})d\Omega_k d\omega_k, \tag{1}$$

$$\delta\omega = \sum_I \int\int \frac{|\boldsymbol{\mu}_{EI}\cdot\boldsymbol{\epsilon}_k|^2}{\hbar^2}\frac{2\pi\hbar\omega_k}{V}\frac{1}{\omega_{EI}-\omega_k}\rho(\omega_k,\mathbf{k})d\Omega_k d\omega_k, \tag{2}$$

where it is assumed that $|E\rangle$ only decays to a single lower state $|G\rangle$, and the sum is over all states of the atom $|I\rangle$; $\hbar\omega_{EI} = E_E + \hbar\delta\omega - E_I$ is the perturbed energy difference between $|E\rangle$ and $|I\rangle$, $\boldsymbol{\mu}_{EI}$ the dipole matrix element between those states, V the quantization volume, and $\rho(\omega_k,\mathbf{k})$ the number of modes per unit frequency interval per unit solid angle, and the integral includes a sum over the two possible polarizations $\boldsymbol{\epsilon}_k$ for each $\mathbf{k}$. Ordinarily free space is considered, for which $\rho_{\text{free}}(\omega) = V\omega^2/(2\pi)^3c^3$; the insertion of this into Eq. (1) gives the familiar result $\Gamma_{\text{free}} = 4\mu_{EG}^2\omega_{EG}^3/3\hbar c^3$. The corresponding result for $\delta\omega_{\text{free}}$ diverges because the mass renormalization term has been neglected.[9] However, in our experiment we will only be concerned with the difference $\Delta\omega_{\text{cav}} = \delta\omega_{\text{cav}} - \delta\omega_{\text{free}}$ between the shift with the atom in the cavity and that with the atom in free space. For this difference it may easily be seen that mass renormalization is unimportant.

It has recently been demonstrated that changes in spontaneous emission can occur when an atom is placed in an open optical resonator with degenerate modes.[7] In the present experiment, atoms are placed near the center of a concentric resonator of mirror separation L and reflectivity R. Provided the atoms are displaced by a distance from the center less than $r_0 = [\lambda L(1-R)]^{1/2}$, where λ is the emission wavelength, the eigenfrequencies of the resonator modes which interact with the atom are completely degenerate. This means that the line-shape function of the cavity as seen by the atoms is given by the Airy function,

$$\mathcal{L}(\omega) = \frac{(1+F)^{1/2}}{1+F\sin^2(\omega L/c)}, \tag{3}$$

irrespective of the propagation direction of the mode $\mathbf{k}$ in the resonator, where the parameter $F = 4R/(1-R)^2$ is related to the finesse $\mathcal{F}$ by $F = (2\mathcal{F}/\pi)^2$. From a ray-optics point of view, this may be seen by our noting that every ray emitted by the atom will return to it after one round trip, and that the round trip phase of all such rays is identical.

The effect of the resonator is to modify the mode density over that part of the solid angle $\Delta\Omega_{\text{cav}}$ controlled by the resonator, with the mode density over the remaining solid angle $\Delta\Omega_{side}$ unchanged. This may be accounted for in Eqs. (1) and (2) by our making the replacement

$$\rho_{cav}(\omega,\mathbf{k}) = \begin{cases} \rho_{free}(\omega)\mathcal{L}(\omega), & \mathbf{k} \text{ in } \Delta\Omega_{cav}, \\ \rho_{free}(\omega), & \mathbf{k} \text{ in } \Delta\Omega_{side}, \end{cases} \quad (4)$$

where the normalization of $\mathcal{L}(\omega)$ is chosen such that the average mode density over one free spectral range is the same as for free space. Substituting $\rho_{cav}(\omega,\mathbf{k})$ in Eq. (1) gives

$$\Gamma_{cav} = \Gamma_{free}\{1 + [\mathcal{L}(\omega_{EG}) - 1] f(\Delta\Omega_{cav})\}, \quad (5)$$

where $f(\Delta\Omega_{cav})$ is the fraction of the total free-space spontaneous emission ordinarily emitted into the solid angle $\Delta\Omega_{cav}$. For the case of circular mirrors of half-angle θ and a $\Delta m = 0$ transition with polarization perpendicular to the cavity axis,

$$f(\Delta\Omega_{cav}) = 1 - \tfrac{3}{4}\cos\theta - \tfrac{1}{4}\cos^3\theta.$$

If also $\theta \ll 1$, $f(\Delta\Omega_{cav}) = (3/8\pi)\Delta\Omega_{cav}$. This result [Eq. (5)] was derived from a different point of view in Ref. 7.

Referring now to Eq. (2), it is easy to see that the only contribution to the difference in frequency shifts $\Delta\omega_{cav}$ occurs near a resonance, $\omega_k \simeq \omega_{EG}$. Therefore, substituting in Eq. (2) for $\rho_{cav}(\omega,\mathbf{k})$ and $\rho_{free}(\omega,\mathbf{k})$, we find

$$\Delta\omega_{cav} = \Gamma_{free} \frac{f(\sigma\Omega_{cav})}{4} \frac{F\sin(2\omega_{EG}L/c)}{1 + F\sin^2(\omega_{EG}L/c)}. \quad (6)$$

Also, if $|G\rangle$ is the ground state, the shift in transition frequency is entirely due to the level shift $\Delta\omega_{cav}$.

Physically, the level shifts are due to contributions from the emission and reabsorption of virtual photons at all possible frequencies. In free space, virtual photons are emitted with essentially equal probability at frequencies slightly above and below resonance, resulting in no net contribution to the frequency shift from a small region of frequency near the atomic resonance. However, in a cavity, the virtual photon emission may be enhanced by a cavity resonance on one side of the atomic resonance relative to the other, resulting in a net contribution to the shift.

In the above it was assumed that the mirrors have perfectly shaped surfaces, that the atoms are located at distances $r \ll r_0$ from the center of the cavity, and that Doppler shifts are negligible. If these conditions are not satisfied, they may be accounted for by modification of the line-shape function $\mathcal{L}(\omega)$. In general, these effects will broaden the peaks in $\mathcal{L}(\omega)$ and reduce the amplitude of its modulation.

The experimental apparatus is illustrated in Fig. 1. A beam of barium atoms is collimated by aperture A1 (1 mm diam) and intercepted by a beam from a cw dye laser, and then recollimated by a second aperture A2 (25 μm diam) and intercepted by a second beam from the same laser. The mean thermal speed of the atoms is 2.4×10^4 cm/s. The laser is tuned near the $^1S_0$-$^1P_1$ transition of ^{138}Ba at $\lambda = 553$ nm. The $^1P_1$ state has a free-space radiative linewidth of 19 MHz.[10] Two regions of excited atoms are thus created, one ("region 1") outside the cavity, and a second ("region 2") inside the cavity. The excited atoms inside the cavity are confined to a region extending $\simeq 30$ μm in each dimension, and are carefully positioned at the center of the cavity. The laser is linearly polarized perpendicular to the resonator axis, and since the ^{138}Ba isotope has zero nuclear spin and is reasonably well resolved from other isotopic components, only a single $\Delta m = 0$ transition is excited. The laser power in both beams is kept well below saturation; the beam in region 2 is focused to a diameter of 30 μm and has a power of 0.02 μW. The sideways fluorescence is collected by optical fiber bundles OFB1 and OFB2 and detected by photomultiplier tubes PMT1 and PMT2.

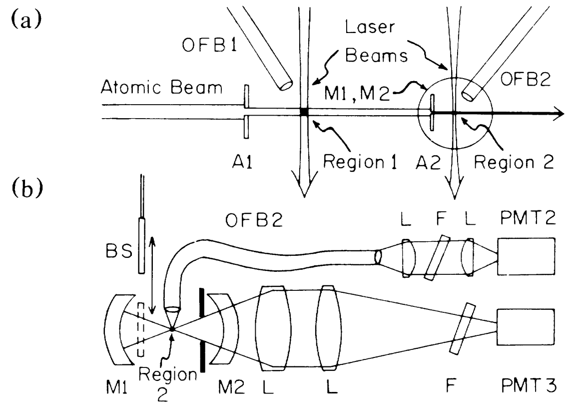

FIG. 1. Experimental apparatus. (a) Atomic-beam excitation geometry. (b) View from side of cavity. PMT1, not shown, detects light from optical fiber bundle OFB1. L, lens; F, interference filter.

The concentric cavity mirrors M1 and M2 have a radius of curvature of 2.50 cm, corresponding to $L = 5.00$ cm and a free spectral range (FSR) $\Delta\nu_{FSR} = c/2L = 3000$ MHz. Their clear diameter is 1.88 cm, so that $\theta = 22°$. The mirrors are coated with a thin bare aluminum film of reflectivity $R = 0.65$, corresponding to $F = 21.2$. The fluorescence emerging through one of the mirrors is collected by an $f/1.2$ camera lens and focused onto a third photomultiplier tube PMT3. The cavity is carefully aligned to concentricity with three independent piezoelectric transducers attached to cavity mirror M1. A small ($\lesssim 1$ free spectral range) linear displacement of M1 from this position tunes the cavity. In order to compare the cavity-modified quantities to those in free space, a movable beam stop BS may be inserted between the atoms and the mirror M1, thus removing the effect of the cavity.

In the experiment, the cavity tuning is held fixed and the fluorescence intensity from all three channels is simultaneously recorded as a function of laser frequency. A typical scan for the fluorescence out the end of the

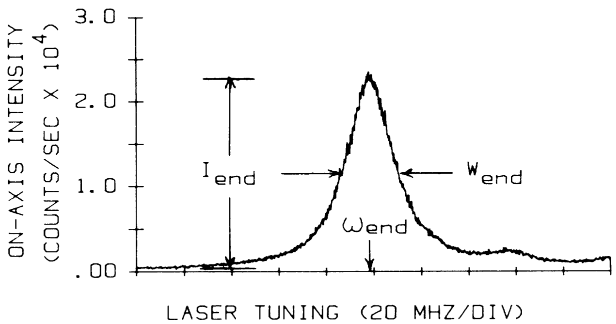

FIG. 2. Spontaneous-emission intensity out the end of the cavity vs laser tuning. The feature on the high-frequency side of the transition is the next isotopic component.

cavity versus laser tuning is shown in Fig. 2. For each such scan, the peak intensity I, width W (full width at half maximum), and frequency shift $\Delta\omega'$ are measured for the fluorescence out of both the ends of the cavity ($I_{\rm end}$, $W_{\rm end}$, and $\Delta\omega'_{\rm end}$) and the sides of the cavity ($I_{\rm side}$, $W_{\rm side}$, and $\Delta\omega'_{\rm side}$). Here $\Delta\omega'=\Delta\omega-\Delta\omega_0$, where $\Delta\omega=\omega-\omega_{\rm free}$ is the difference between the center frequency ω for the atoms inside the cavity (region 2) and the center frequency $\omega_{\rm free}$ for the atoms outside the cavity (region 1); $\Delta\omega_0$ is the same quantity with the cavity blocked.[11] The main purpose of region 1 is to provide an accurate frequency reference for the measurement of $\Delta\omega'$. The experiment is then repeated at a succession of different cavity tunings.

The results are summarized in Fig. 3. The top set of data (a) shows the peak height $I_{\rm end}$, the second set (b) shows the peak height $I_{\rm side}$, the third set (c) shows the widths $W_{\rm end}$ and $W_{\rm side}$, and the last set (d) shows the observed frequency shifts $\Delta\omega'_{\rm end}$ and $\Delta\omega'_{\rm side}$, all as functions of cavity tuning. In each case the straight line shows the same quantities $I^0_{\rm side}$, $I^0_{\rm end}$, and W_0 observed with the cavity blocked, thus giving the free-space values of these quantities. (Here $I^0_{\rm end}$ was corrected for the attenuation of the remaining mirror.) The width $W_0=\Gamma_{\rm free}+\Gamma_0$ contains a contribution Γ_0 from a combination of transit-time broadening, laser frequency jitter, and Doppler broadening.

The curves for each set of data show a theoretical fit by the functions,

$$I_{\rm end}=I^0_{\rm end}\frac{\Gamma_{\rm free}(\Gamma_{\rm free}+\Gamma_0)}{\Gamma_{\rm cav}(\Gamma_{\rm cav}+\Gamma_0)}\mathcal{L}(\omega_{EG}), \tag{7}$$

$$I_{\rm side}=I^0_{\rm side}\frac{\Gamma_{\rm free}(\Gamma_{\rm free}+\Gamma_0)}{\Gamma_{\rm cav}(\Gamma_{\rm cav}+\Gamma_0)}, \tag{8}$$

$$W=\Gamma_{\rm cav}+\Gamma_0, \tag{9}$$

$$\Delta\omega'=\Delta\omega_{\rm cav}, \tag{10}$$

where $\Gamma_{\rm cav}$ and $\Delta\omega_{\rm cav}$ are given by Eqs. (5) and (6), $\Gamma_{\rm free}=19$ MHz, and the measured values of $f(\Delta\Omega_{\rm cav})=0.106$, $\Gamma_0=5.0$ MHz, and $I^0_{\rm end}$ and $I^0_{\rm side}$ are used.[12] The parameter F was adjusted, to produce the best fit to the data, at $F=8.0$. This is somewhat less than the ideal value; the reduced value takes into account the various broadening mechanisms, as discussed earlier. Good agreement with the theory is obtained.

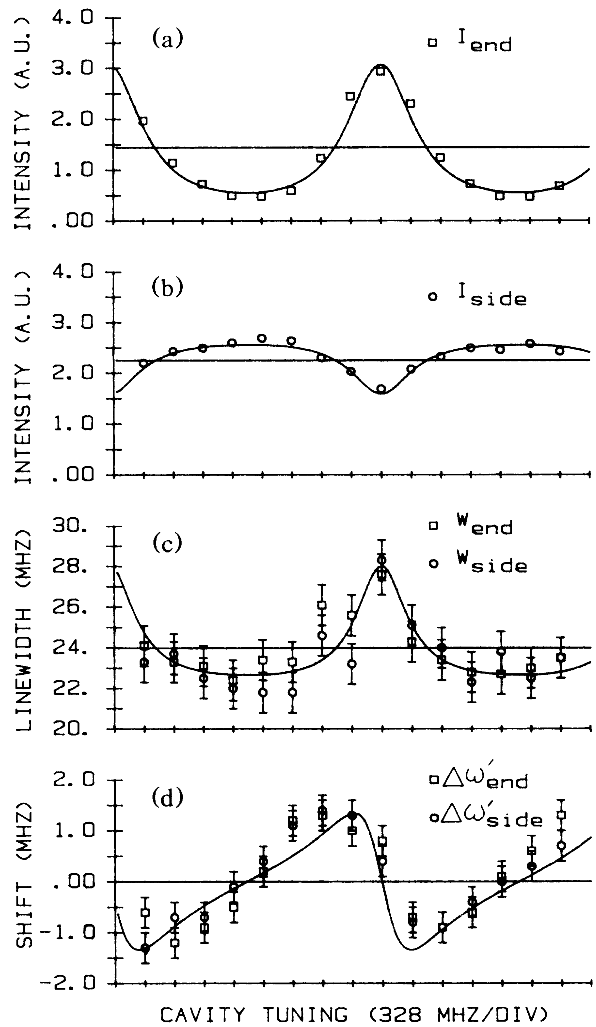

FIG. 3. Observed intensities, linewidths, and frequency shifts as functions of cavity tuning. The cavity length decreases from left to right.

There are a number of important features to notice about the data. First, the intensity out of the sides of the cavity decreases when the intensity out the ends of the cavity increases. This decrease is *not* a "spatial redistribution" of the spontaneous-emission probability. It arises only because of a decrease in the excited-state atomic population, caused by an increase in the total decay rate Γ.[12] Second, the linewidth of the transition increases or decreases in direct proportion to the total spontaneous-emission rate. This confirms that the enhanced and inhibited decay of the atoms is spontaneous rather than stimulated. Finally, the radiative shift vanishes when the atomic resonance either coincides with a cavity resonance, or is exactly between two cavity resonances, and the transition shifts to the blue when the nearest cavity mode is tuned to the red, and vice versa. This may be understood by our viewing the atom as interacting primarily with the nearest cavity mode; the atom-cavity mode coupling pushes their eigenfrequencies apart.

Several points should be emphasized. First, the laser is not directly coupled into the cavity; the cavity perturbs the atom, and the laser is only used to probe the atom. Second, the experiment is carried out under true single-atom conditions: The density of the beam is $\simeq 10^8-10^9$ atoms/cm^3, resulting in $\simeq 1-10$ atoms in the resonator at any given time. Further, even if several atoms are present, they will not interact appreciably since the focused beam waists of the generated fields do not coincide. As emphasized above, the *net* radiative lifetime has changed, as the changes in linewidth confirm. Finally, we note that the radiative level shifts of an atom may be important in precision spectroscopy, if one hopes to use suppressed spontaneous emission to narrow a spectral line. Our experiment shows that, in the case of an open degenerate optical resonator, the radiative level shift vanishes when the greatest suppression occurs: when the atomic frequency is between two cavity modes.

In conclusion, we have demonstrated that the energy levels of an atom may be radiatively shifted by a change in the density of vacuum modes of the field when the atom is placed in a resonator. We have also directly shown that, when the atom's spontaneous-emission rate is enhanced or suppressed, the natural linewidth of the transition increases or decreases in direct proportion. We find that these effects are well described by a QED calculation in which the mode density of free space is replaced by that of the resonator.

We would also like to acknowledge the valuable contributions of J. J. Childs, Eugene Gath, and C. R. Monroe. This work was performed at the Massachusetts Institute of Technology Laser Research Center, a National Science Foundation Regional Instrumentation Facility. This work was supported by the National Science Foundation under Grant No. PHY-8313248.

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[11]The frequency shift $\Delta\omega_0 = +1.8$ MHz is due to a difference in the slight residual first-order Doppler shift between region 1 and region 2. This shift occurs because the laser beams in the two regions are not exactly perpendicular to the atomic beam, so that a small component of the atomic velocity appears along the propagation direction of the laser beam.

[12]The expressions for I_{end} and I_{side} are derived by our noting that $I_{\mathrm{side}} \propto n_e$, whereas $I_{\mathrm{end}} \propto n_e \gamma_{\mathrm{cav}} \propto n_e \mathcal{L}(\omega_{EG})$, where n_e is the excited-state population and γ_{cav} the partial spontaneous-emission rate of the atom into the cavity. The assumption of unsaturated excitation of a homogeneously broadened transition gives $n_e \propto \sigma/\Gamma \propto [(\Gamma+\Gamma_0)\Gamma]^{-1}$, where σ is the absorption cross section. Note that this effect may be viewed as a variation of the transition saturation intensity as a function of cavity tuning.