3(b), 3(c), and 3(d), one could induce a 90° (i.e., 180° for two-photon) phase shift between every other pair of coherent pulses into the sample cell. By subtracting the resulting fluorescence from pairs of coherent pulses with and without the phase shift, one could eliminate the diffraction background and thus automatically double the interference signal. A detailed study of this scheme will appear elsewhere.[3]

To conclude, we mention a possible variant of the previous method. Instead of two pulses, one could use a sequence of N equally spaced pulses obtained, for example, by sending the initial pulse into a confocal resonator.[6] The optical analog of such a system would be a grating with N lines. One should expect in that case that the frequency spectrum experienced by the atom would be a series of peaks (N times narrower in width), separated by the frequency interval $1/T_{\text{eff}}$.

We are grateful to our colleagues at Ecole Normale Supérieure (Paris), Joint Institute for Laboratory Astrophysics, and to Professor N. Bloembergen, Professor V. P. Chebotayev, Professor T. W. Hänsch, Professor F. M. Pipkin, Professor N. F. Ramsey, Mr. J. Eckstein, and Mr. R. Teets for many stimulating discussions. One of us (C.C.-T.) thanks Harvard University and Massachusetts Institute of Technology for their warm hospitality during his stay in Cambridge.

*Supported in part by the Joint Services Electronics Program.

†Permanent address: Laboratoire de Spectroscopie Hertzienne de l'Ecole Normale Supérieure, associé au Centre National de la Recherche Scientifique et Collège de France, 75231 Paris Cedex 05, France.

[1]Ye. V. Baklanov, B. Ya. Dubetskii and V. P. Chebotayev, Appl. Phys. 9, 171 (1976); Ye. V. Baklanov, B. Ya. Dubetskii, and V. P. Chebotayev, Appl. Phys. 11, 201 (1976); B. Ya. Dubetskii, Kvantovaya Elektron. (Moscow) 3, 1258 (1976) [Sov. J. Quantum Electron. 6, 682 (1976)]. Note that the method of using spatially separated laser fields has very recently been applied to saturated absorption spectroscopy by J. C. Bergquist, S. A. Lee, and J. L. Hall, Phys. Rev. Lett. 38, 159 (1977).

[2]N. F. Ramsey, *Molecular Beams* (Oxford Univ. Press, New York, 1956); also, C. O. Alley, in *Quantum Electronics, A Symposium* (Columbia Univ. Press, New York, 1960), p. 146; M. Arditi and T. R. Carver, IEEE Trans. Instrum. IM-13, 146 (1964).

[3]M. M. Salour, to be published.

[4]M. M. Salour, Bull. Am. Phys. Soc. 21, 1245 (1976).

[5]For the two pulses of Fig. 1(b), the result of the calculation is $|b_2|^2 = 4|b_2|^2|\cos\omega_0 T/2|^2$, so that $|b_2|^2$ does not depend on ω, with ω_0 and T fixed. Note, however, that the interference fringes reappear when T alone (or ω_0 alone) is varied.

[6]R. Teets, J. Eckstein, and T. W. Hänsch, this issue [Phys. Rev. Lett. 38, 760 (1977)].

Coherent Two-Photon Excitation by Multiple Light Pulses*

R. Teets,† J. Eckstein, and T. W. Hänsch
Department of Physics, Stanford University, Stanford, California 94305

(Received 20 December 1976)

We have studied Doppler-free two-photon excitation of atoms with a train of phase-coherent standing-wave light pulses, originating from the same laser pulse. Quantum interference effects produce narrow spectral fringes which have a physical origin similar to that of Ramsey fringes. Linewidths much less than the Fourier-transform limit of an individual light pulse have been observed for the sodium 3S-5S transition; and a dramatic enhancement of the resonant signal is possible.

We have studied Doppler-free two-photon excitation of atoms with a train of phase-coherent standing-wave light pulses, produced by multiple reflections of a single laser pulse inside an optical resonator. Quantum interference effects result in narrow spectral lines, whose widths depend only on the resonator losses and on the natural transition linewidth, not on the laser linewidth. The resonant excitation probability is proportional to the square of the number of pulses incident during the atomic relaxation time; and a dramatic signal enhancement over single-pulse excitation is possible. We have observed these coherence effects for the sodium 3S-5S transition.

The physical origin of the narrow interference fringes is similar to that of Ramsey fringes.[1] The light field induces coherent atomic oscillations at the two-photon resonance frequency, and the incremental effect of each pulse depends on the phase of its light field relative to these atomic oscillations. It has recently been pointed out[2] that Doppler-free two-photon excitation with standing light waves should permit the observation of optical Ramsey fringes with two spatially

separated fields interacting with an atomic beam, because the phase of the sum frequency is not affected by the first-order Doppler effect. (The atoms in a standing wave see opposite Doppler shifts for the two traveling-wave components.) Optical Ramsey fringes have very recently been observed with two phase-coherent light pulses, separated in time rather than in space.[3]

The presently proposed technique of multiple-pulse interference can provide sharp lines rather than the sinusoidal pattern of two-field Ramsey fringes. This is of particular interest for spectroscopic applications, because it makes it possible to resolve closely spaced line components. Also the large possible signal enhancement is advantageous for the study of weak transitions or for excitation with ultraviolet light where powerful pulsed lasers often with large linewidths are required for the efficient generation of the tunable radiation by nonlinear frequency mixing. The signal enhancement should also permit the use of larger, less intense beams, thus alleviating the problems of ac Stark shifts[4] and transit-time broadening. The large enhancement can even make two-photon excitation attractive for effective pumping of excited states for such applications as selective photochemistry.

To describe the excitation process, we refer to the experimental configuration shown in Fig. 1. A laser pulse, tuned to excite a two-photon transition between atomic levels $|g\rangle$ and $|f\rangle$, enters a resonator of length L, for example, through a partially transmitting mirror, and bounces back and forth, producing a train of phase-coherent pulses, separated in time by $\tau = 2L/c$. The atoms are placed near one mirror, where the incident and reflected fields form a standing wave. The fluorescence from the upper state is monitored as a function of the resonator length.

To show how the multiple pulse excitation leads to narrow linewidths and large signals, we calculate the excitation probability, using perturbation theory. Since we are only interested in the Doppler-free part, we ignore the velocity dependence of the atoms. (The motion of the atoms does not cause dephasing since the atoms see opposite phase shifts for the two counterpropagating waves in the standing-wave field.)

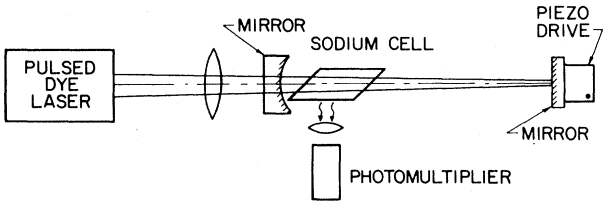

FIG. 1. Experimental arrangement.

The field for the train of pulses in the resonator is

$$\vec{E}(t) = \sum_{n=0}^{\infty} \vec{E}_0(t - n\tau) \exp[-i\omega(t - n\tau)](R_1 R_2)^{n/2} + \text{c.c.}, \quad (1)$$

where τ is the roundtrip time in the resonator, R_1 and R_2 are the mirror reflectivities, ω is the laser frequency, and $\vec{E}_0(t)$ is the (slowly varying) envelope function for a single laser pulse inside the resonator. We note that the pulse does not have to be transform-limited. The detailed spatial field distribution is unimportant in the limit of small excitation probabilities, as long as only the fundamental TEM_{00} cavity modes are excited. The interaction Hamiltonian $-e\vec{r}\cdot\vec{E}$ can be written as $V\exp(-i\omega t) + V^*\exp(i\omega t)$, where V is a slowly varying function of time.

As long as the laser frequency is tuned away from any intermediate single-photon resonance frequency ω_{mg}, first-order perturbation theory yields only rapidly oscillating state amplitudes. But second-order perturbation theory[5] predicts a slowly varying amplitude C_f of the final state, describing two-photon excitation. Neglecting atomic relaxation, we expect

$$C_f^{(2)}(t) = \int_0^t dt' \sum_m \frac{i}{\hbar^2} \frac{V_{fm} V_{mg} \exp[i(\omega_{fg} - 2\omega)t']}{\omega_{mg} - \omega}. \quad (2)$$

If the successive light pulses do not overlap in time, the integral of Eq. (2) can be replaced by the sum of integrals over individual pulse contributions. Since the successive pulses are replicas of each other, except for a constant amplitude factor, only one such integral needs to be evaluated, corresponding to the state amplitude $C_f(\tau)$ after the first pulse. The total sum is then

$$C_f(\infty) = C_f(\tau) \sum_{n=0}^{\infty} \{e^{i\omega_{fg} n\tau} (R_1 R_2)^n\}. \quad (3)$$

The entire dependence on the laser frequency is contained in $C_f(\tau)$. The sum over the amplitude and phase factors depends only on the atomic resonance frequency ω_{fg} and on the resonator properties, not on the laser frequency ω. Similar summations are encountered in the theory of optical multiple-beam interference,[6] and it is hence not surprising that the excitation $|C_f(\infty)|^2$ versus delay time τ exhibits many of the features of a Fa-

bry-Perot interferogram.

It is not difficult to include in this calculation a (transverse) relaxation rate γ for the oscillating atoms. The total fluorescence from this state $|f\rangle$, excited by one pulse train, is then $P = P_0\eta$, where P_0 is the fluorescence signal produced by a single pulse, and the factor η describes the multipulse interference:

$$\eta = \eta_{\max}[1 + F\sin^2(4\pi L/\lambda_0)]^{-1},$$

$$\eta_{\max} = \frac{(1 + R_1R_2e^{-\gamma\tau})}{(1 - R_1^2R_2^2)(1 - R_1R_2e^{-\lambda\tau})}, \quad (4)$$

$$F = 4R_1R_2e^{-\gamma\tau}/(1 - R_1R_2e^{-\gamma\tau})^2.$$

Here, $\lambda_0 = 2(2\pi c/\omega_{fg})$ is the resonance wavelength of the two-photon transition. The dependence of the signal on the exact resonator length, described by Eq. (4), is similar to the transmission of a Fabry-Perot interferometer, given by the Airy formula of Ref. 6, except that the resonant maxima occur twice as frequently, and the finesse in the limit of zero atomic linewidth drops with R_1R_2, rather than with $(R_1R_2)^{1/2}$. The linewidth (full width at half-maximum) of an individual fringe is approximately

$$(\delta\nu)_{\mathrm{FWHM}} \approx (2/\pi\sqrt{F})(c/4L). \quad (5)$$

The resonant signal enhancement, in the limit of negligible atomic relaxation, is given by $\eta_{\max} \approx 1/(1 - R_1R_2)^2$, which is about equal to the square of the effective number of pulse roundtrips, N^2. In addition, there will be a Doppler-broadened background due to traveling-wave excitation,[7] which reduces the fringe contrast. But this background grows only linearly with N so that the contrast improves with increasing N.

While the coherence aspects of two-photon excitation with multiple pulses can best be understood by working in the time domain, the spectroscopic aspects are clearest in the frequency domain. In some ways the situation resembles two-photon spectroscopy with a multimode laser.[8] Figure 2 shows the intensity spectra of a train of Gaussian light pulses for two different cavity tunings. Resonant two-photon excitation is possible whenever the frequencies of any two peaks add up to the atomic transition frequency ω_{fg}. As illustrated in Fig. 2, this can occur in two ways: Either a peak coincides with $\frac{1}{2}\omega_{fg}$ (solid curve), or the frequency $\frac{1}{2}\omega_{fg}$ falls exactly halfway between two cavity modes (dashed curve). In either case, all the modes under the single-pulse envelope contribute to the excitation. For long pulses with narrow spectral envelope, the excitation in the latter case may be smaller than in the first case.

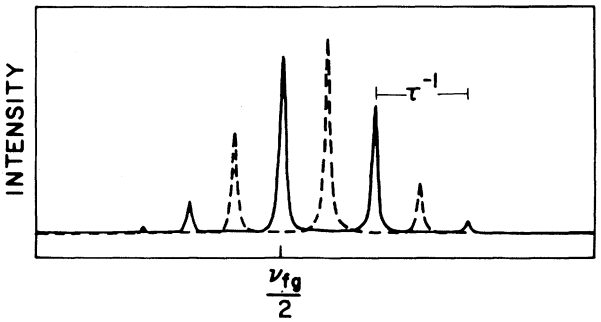

FIG. 2. Intensity spectra of a damped train of Gaussian pulses for a resonator with $R_1R_2 \approx 0.7$. Dashed and solid curves are for two different resonator tunings. The slowly varying envelope corresponds to the spectrum of one individual pulse.

To demonstrate multipulse two-photon excitation in atomic sodium, we used the experimental configuration shown in Fig. 1. Short laser pulses were injected into an optical resonator, containing a sodium cell near one end mirror, to provide a standing-wave field. A nitrogen-laser–pumped dye-laser system[9] was used to produce 10-kW pulses of linearly polarized light at about 6022 Å. The pulses were 6 nsec long and the bandwidth was slightly greater than the Fourier-transform limit of about 170 MHz. The resonator was mounted on a rigid optical bench. Its length was maintained by quartz tubes and Invar rods, which provided mechanical and thermal stability, and an insulating box reduced thermal drifts to a few megacycles per minute. One end mirror was mounted on a piezotranslator, permitting continuous scanning over several wavelengths.

The sodium was contained in an evacuated quartz cell with high-quality Brewster windows in order to minimize losses in the resonator. The cell was heated in an electrical oven to about 120°C, producing a sodium density of 2×10^{10} atoms/cm^3. The laser was tuned to one hyperfine-structure component of the $3S$-$5S$ transition,[7] with an upper-state lifetime of 71 nsec[10]; and the excitation was monitored by observing the ultraviolet cascade fluorescence from the $4P$-$3S$ transition.

Two different resonator configurations were used: a 1-m-long nearly hemispherical cavity and a 2-m-long confocal cavity. The beam-entrance mirror in each case had a transmission of 7%. Light losses at the mirrors and the windows resulted in an effective loss factor R_1R_2 of about

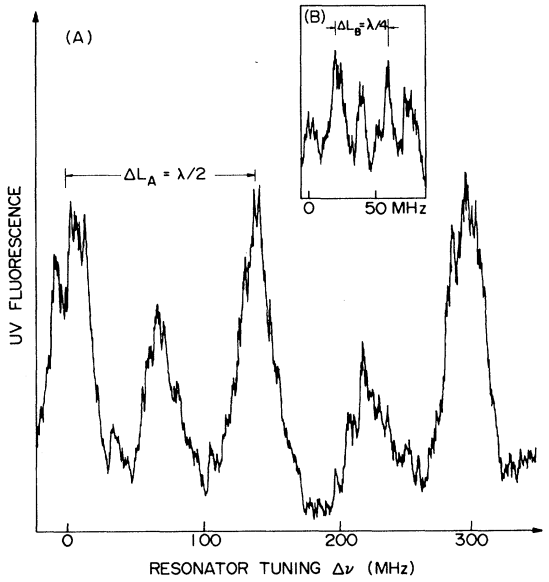

FIG. 3. Multipulse two-photon excitation of the $F = 2$ hyperfine component of sodium $3S$-$5S$. The fluorescence is recorded versus resonator tuning for (a) a 1-m nearly hemispherical resonator and (b) a 2-m confocal resonator.

0.8, reducing the effective number of roundtrips to about 6. The beam waist at the sodium cell was about 1 mm. For the intensities inside the resonator, the resulting ac Stark shifts are only a few megacycles.[4]

The fluorescence versus resonator tuning for the 1-m hemispherical cavity is shown in Fig. 3(a). The spectrum was recorded at a 15-pps laser repetition rate with a 3-sec time constant during about 150 sec. Since the transverse modes of this resonator are not degenerate, it was essential to mode-match in order to excite predominantly the TEM_{00} mode. The observed linewidth is about 30 MHz, significantly less than the laser bandwidth, but larger than predicted by Eq. (5), largely because of residual excitation of higher-order transverse modes. As predicted, the peaks are separated by $\Delta L = \frac{1}{4}\lambda_0$. Since the laser bandwidth is comparable to the resonator free spectral range, the two different resonance conditions illustrated in Fig. 2, produce differently sized peaks, and result in the observed alternation of intensities. Figure 3(b) shows a corresponding spectrum recorded with the 2-m confocal resonator which does not require mode matching. Here the linewidths are only about 12 MHz, presumably limited by cavity vibrations and ac Stark shifts, but the peaks are separated by only $\Delta L = \frac{1}{8}\lambda_0$, since both even and odd resonator modes were excited.

The interpretation of such a spectrum with its multiple orders is similar to that of a conventional Fabry-Perot spectrum. If two closely spaced lines are present, two combs of resonances appear, separated by the atomic line splitting modulo $c/4L$. In our preliminary experiments, the frequency of the pulsed dye laser was left fixed, and only the external cavity was scanned. For a number of experiments envisioned, it would seem desirable to tune the laser as well. If its frequency were locked to the external resonator, the central fringe of the multipulse interference pattern would always coincide with the atomic transition frequency, as in the double-pulse Ramsey-fringe experiment.[3] One can derive this simply using Eq. (4) by adding the constraint that $L = \frac{1}{2}n\lambda_{\text{laser}}$, where n is integral.

In the present experiment, much laser light is lost when the pulse enters the resonator through one end mirror. In the absence of absorption losses, the input transmittance is $1 - R_1$; and since the excitation is proportional to the intensity squared, the excitation by a single pulse would be decreased by a factor of $(1 - R_1)^2$. However, the observed resonant fluorescence in our experiment was actually comparable to the signal produced by a single unattenuated pulse outside the resonator, giving evidence for the predicted enhancement. Current electro-optic technology makes it entirely feasible to inject a laser pulse into a resonator without attenuation. A low-loss cavity, which permits about 100 pulse roundtrips, would then allow an actual enhancement of the excitation probability by a factor of 10 000.

We are grateful to Dr. Geoffrey H. C. New, Mr. Michael Salour, Professor Claude Cohen-Tannoudji, and Professor Arthur L. Schawlow for numerous stimulating discussions.

*Work supported by the National Science Foundation under Grant No. 14789 and by the U. S. Office of Naval Research under Contract No. N00014-75-C-0841.

†National Science Foundation Predoctoral Fellow.

[1]N. F. Ramsey, *Molecular Beams* (Oxford Univ. Press, London, England, 1956), p. 124.

[2]Ye. V. Baklanov, V. P. Chebotayev, and B. Ta. Dubetsky, Appl. Phys. 11, 201 (1976).

[3]M. Salour, Bull. Am. Phys. Soc. 21, 1245 (1976); M. M. Salour and C. Cohen-Tannoudji, this issue [Phys. Rev. Lett. 38, 757 (1977)]. [*Note added.*—Optical Ramsey fringes have also been frequently observed in saturated absorption with spatially separated fields; J. C. Bergquist, S. A. Lee, and J. L. Hall, Phys. Rev. Lett. 38, 159 (1977)].

[4]P. F. Liao and J. E. Bjorkholm, Phys. Rev. Lett. 33,

128 (1974).
[5]A. Gold, in *Quantum Optics, Proceedings of the International School of Physics, "Enrico Fermi," Course XLII,* edited by R. J. Glauber (Academic Press, New York, 1969), p. 397.
[6]M. Born and E. Wolf, *Principles of Optics* (Pergamon, Oxford, England, 1970), p. 323.
[7]M. D. Levenson and N. Bloembergen, Phys. Rev. Lett. 32, 645 (1974).
[8]D. E. Roberts and E. N. Fortson, Opt. Commun. 14, 332 (1975).
[9]R. Wallenstein and T. W. Hänsch, Opt. Commun. 14, 353 (1975).
[10]D. Kaiser, Phys. Lett. 51A, 375 (1975).

Quantum Beats in Superfluorescence in Atomic Cesium

Q. H. F. Vrehen and H. M. J. Hikspoors
Philips Research Laboratories, Eindhoven, The Netherlands

and

H. M. Gibbs*
Bell Laboratories, Murray Hill, New Jersey 07974
(Received 20 December 1976)

Superfluorescence has been observed at 3 μm on the $7P$-to-$7S$ transition in atomic cesium after 2-ns excitation of a $6S$-to-$7P$ transition. Both in a magnetic field and in zero field, quantum beats have been observed. The beat frequencies correspond not only to initial-level splittings but also to combinations of initial- and final-level splittings. Superfluorescence beats are therefore basically different from single-atom quantum beats.

We report the first observation of quantum beats[1] in superfluorescence (SF),[2-5] i.e., in the cooperative emission of an initially inverted system.[6] SF beats and photon-echo beats[1b] can arise either from a superposition of simultaneously excited upper levels (as for quantum beats in single-atom fluorescence[1a]) or from the beating in the detector of the coherent emissions of two independent transitions (just as the outputs of two lasers can be mixed to produce beats).[1b] The coherence required for beating of independent transitions is introduced by the excitation in the photon-echo case, but in the SF case it arises from the emission process itself.

SF quantum beats may have spectroscopic applications because, unlike single-atom quantum beats, they can occur at lower-state splittings as well as upper-state splittings. Also, almost all of the energy in the inverted transition is emitted along the excited cylinder and in a time much shorter than the natural radiative lifetime, yielding signals much larger than those for ordinary fluorescence. These advantages of coherent emission were also emphasized for photon-echo quantum beats.[1b] The reduced emission time limits the accuracy of the frequency determination, but a signal increase of many orders of magnitude can be achieved on weak transitions[4] before this limitation becomes important. The strong emission intensity in SF can lead to frequency chirps, which must be understood before SF quantum beats can become a technique for high-resolution spectroscopy.

Quantum beats in spontaneous emission are well known and may be observed when two conditions are met.[1] First, the initial state must be prepared as a coherent superposition of substates at a well-defined point in space or time. Second, the various substates must radiate to a common final state (or states). Under such conditions the intensity of the spontaneous emission can reveal beats that correspond to level splittings of the initial state. Since the atoms decay in the absence of any driving field, accurate values for the splittings can be obtained.[1]

In SF, many atoms decay coherently in their common reaction field. A coherent optical field is emitted at the frequencies of the transitions that take part in the SF process.[1b] So the beat frequencies may reflect initial-level splittings, final-level splittings, or combinations of both. As a result, beats in SF can be quite different from those in spontaneous emission—the two conditions above need not be met.

In this Letter, the results of two different experiments are presented. In one experiment, a coherent mixture of initial substates is excited; the observed beats are well understood on the basis of calculations for the beats in spontaneous emission. In the other experiment, two initial