# Observation of Ramsey's Interference Fringes in the Profile of Doppler-Free Two-Photon Resonances*

M. M. Salour and C. Cohen-Tannoudji†
*Department of Physics and Gordon McKay Laboratory, Harvard Universtiy, Cambridge, Massachusetts 02138*
(Received 13 January 1977)

Using two coherent time-delayed short pulses, we have demonstrated that one can obtain, in the profile of Doppler-free two-photon resonances, interference fringes with a splitting $1/2T$ ($T$, delay time between the two pulses) much smaller than the spectral width $1/\tau$ of each pulse ($\tau$, duration of each pulse). This method should lead to a number of important improvements in the presently available techniques of ultrahigh-resolution laser spectroscopy of atoms and molecules.

In this Letter we report an experiment which demonstrates that, in the same way that the diffraction pattern through two spatially separated slits exhibits interference fringes within the diffraction profile corresponding to a single slit, by exciting atoms with two time-delayed coherent laser pulses one can obtain, in the profile of the Doppler-free two-photon resonances, interference fringes with a splitting $1/2T$ ($T$, delay between the two pulses) much smaller than the spectral width $1/\tau$ ($\tau$, duration of each pulse).

For two very sharp levels such as ground states or metastable states (an important example being the $1S_{1/2}$-$2S_{1/2}$ transition of H), the use of a single laser beam leads to linewidths which are generally limited by the inverse of the transit time of atoms through the laser beam. Rather than increasing this time by expanding the laser beam diameter (with a subsequent loss of intensity), Baklanov, Dubetskii, and Chebotayev[1] have proposed the use of two spatially separated beams leading to structures in the profile of the resonance having a width determined by the time of flight between the two beams. The experiment described below, however, deals with short-lived atomic states (lifetime $\sim 5\times10^{-8}$ sec), and very short pulses (duration $\sim 5\times10^{-9}$ sec), so that the transit time through the laser beam ($\sim 10^{-7}$ sec) plays no role in the problem. This explains why, instead of two spatially separated beams, we use two time-delayed short pulses. These experiments can be considered as optical analogs of the Ramsey method of using two separated rf or microwave fields in atomic-beam experiments.[2]

In order to obtain interference fringes in the profile of a two-photon resonance, two important requirements must be fulfilled. First, each pulse must be reflected against a mirror placed near the atomic cell in order to submit the atoms to a pulsed standing wave and to suppress in this way any dephasing factor due to the motion of the atoms: The probability amplitude for absorbing two counterpropagating photons is proportional to $e^{i(\omega t-kz)}e^{i(\omega t+kz)}=e^{2i\omega t}$ and does not depend on the spatial position $z$ of the atoms. Second, the phase difference between the two pulses must remain constant during the whole experiment: Important phase fluctuations between the two pulses will wash out the interference fringes, since any phase variation produces a shift of the whole interference structure within the diffraction background. This means that the experiment must be done not with two independent pulses, but with two time-delayed pulses having a constant phase difference during the entire experiment. Furthermore, these two pulses have to be Fourier-limited; that is, their coherence time should be longer than their duration.

A first possible method for obtaining such a sequence of two phase-locked and Fourier-limited pulses is to start with a cw-dye-laser wave (with a very long coherence time) and to amplify it with two time-delayed pulsed amplifiers. Even if the cw laser delivers a very weak intensity in the spectral range under study, one can, with a sufficient number of synchronized pulsed amplifiers, get enough power eventually to observe the two-photon resonance. The time dependence of the amplified wave has the shape represented by the solid lines of Fig. 1(a). The two pulses are two different portions of the same sinusoid (represented in dotted lines), and they obviously have the same phase, which is that of the cw carrier wave. In such a case, a very simple calculation[3] shows that the Doppler-free probability $|b_2|^2$ for having the atom excited in the upper state after the second pulse is related to the probability $|b_1|^2$ of excitation after the first pulse by

$$|b_2|^2=4|b_1|^2[\cos(\omega_0-2\omega)T/2]^2,$$

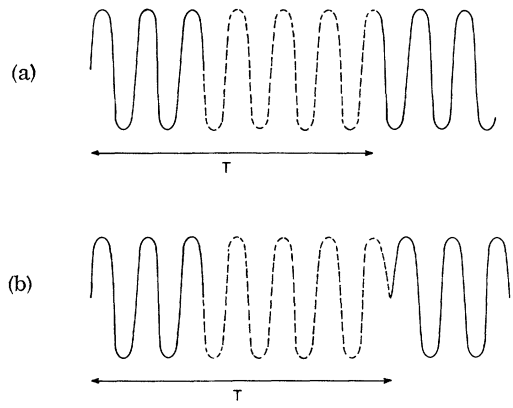


FIG. 1. (a) Two Fourier-limited time-delayed pulses generated by amplifying the same cw wave by two time-delayed independent pulsed amplifiers. They clearly have the same phase which is that of the cw carrier. (b) Two pulses originated from the same Fourier-limited pulse by using a delay line. The second one is just the time translation of the first one by an amount $T$. The phase difference between the two pulses clearly depends on $\omega T$.

where $\omega_0$ is the Bohr frequency of the atomic transition, $\omega$ the laser frequency, and $T$ the time delay between the two pulses. If one varies $\omega$, with $\omega_0$ and $T$ fixed, one gets within the diffraction profile (associated with $|b_1|^2$) interference fringes described by the last oscillatory term, with a splitting in $\omega$ determined by $1/T_{\text{eff}}$, where $T_{\text{eff}}=2T$. The important point is that the central fringe is exactly centered at half the Bohr frequency ($\omega=\frac{1}{2}\omega_0$). As long as the two pulses are phase-locked, any small variation of $T$ is not important since it does not change the position of the central fringe.

A second method, which is experimentally simpler and which we have used,[4] is to start with a Fourier-limited pulse (obtained by amplifying a cw wave) and then to generate from it two pulses in an optical delay line. The time dependence of the resulting wave has the shape represented by the solid lines of Fig. 1(b). The second pulse is just the time translation of the first one by an amount $T$ ($T>\tau$). However, the sinusoid extrapolated from the first pulse (dotted lines) does not generally overlap the second pulse, which means that the two pulses do not generally have the same phase, unless $T$ is an integral number of optical periods $2\pi/\omega$ (i.e., $e^{i\omega T}=1$). The important point is that the phase difference between the two pulses is $\omega$-dependent in such a way that the interference fringes would disappear if $\omega$ is varied, with $T$ being fixed.[5] One possible scheme (which will be described later) for locking the phases of the two pulses, and consequently for having the interference fringes reappear (centered exactly at $\omega=\frac{1}{2}\omega_0$), is to vary $T$ simultaneously (as $\omega$ is varied) in such a way that $e^{i\omega T}$ remains equal to 1.

Figure 2 shows the experimental setup. We utilize a single-mode cw dye laser (Spectra Physics 580A, frequency-stabilized to a pressure-tuned reference etalon, and pumped by a Spectra Physics 164 $Ar^+$ laser), which is tuned to the $3^2S$-$4^2D$ two-photon transition in sodium (5787.3 Å). (Lifetime of $4^2D$ is $\sim 5\times10^{-8}$ sec.) The output of this cw dye laser (40 mW, 2 MHz) is then amplified

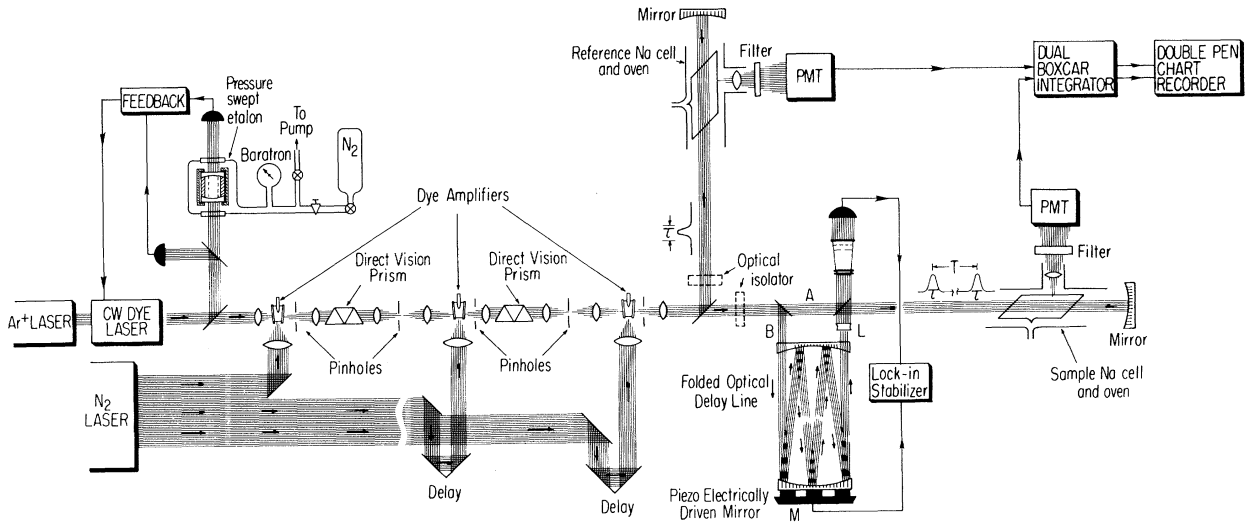


FIG. 2. The experimental setup.

in three synchronized stages of dye amplifiers pumped by a 1-MW nitrogen laser. Three stages of amplification are necessary to boost the peak output power to an acceptable level for this experiment. The pump light for the amplifiers is geometrically divided between the amplifiers and optically delayed in order to arrive slightly later than the input from the evolving pulse building up from the cw dye laser, maximizing the length of the output pulse. To avoid the undesirable amplified spontaneous emission from the amplifiers, suitable spectral and spatial filters are inserted between stages. (A more detailed description of this oscillator-amplifier dye-laser system will appear elsewhere.[3]) The 75-kW output of the third-stage amplifier is split into two parts. The first part goes into a reference sodium cell; and the second part is further split into another two parts—part $A$ goes directly into a sample sodium cell, and part $B$ is optically delayed in a delay line by a time $T$ ($2T = 17$, 25, and 33 nsec) and is subsequently recombined with part $A$ and focused into the sample sodium cell. We have stabilized the delay line by locking the center fringe of the two cw carrier beams (which is at maximum when the direct and the delayed cw carrier beams are in phase, i.e., when $e^{i\omega T} = 1$) to the motion of the mirror $M$ which is mounted on a piezoelectric ceramic. In this way, the two time-delayed and Fourier-limited pulses are phase-locked during the entire experiment. Furthermore, the delayed beam, with its longer optical path, has wave fronts with a larger radius of curvature than those of the direct beam, so a high-quality lens ($L$) is used to match the curvature of the two wave fronts. Both sodium cells were kept at 130°C ($2\times10^{-6}$ Torr of vapor pressure); they were made of Corning 1720 calcium alumino-silicate glass with optically flat windows and were baked at high temperature and later filled with sodium under very high vacuum to avoid any foreign gas contamination. Note also that to generate the laser standing waves with short pulses, one needs to adjust the back-reflecting mirror so that the time delay between the original pulse and the back-reflected pulse is significantly shorter than the pulse duration $\tau$. The outputs of the two photomultipliers (EMI 9635QB) monitoring the fluorescence from $4P$ to $3S$ at 3303 Å are simultaneously processed by a dual-channel boxcar integrator, and the result is recorded simultaneously by a dual-pen chart recorder.

Figure 3(a) shows the four well-known two-photon resonances of the $3^2S$-$4^2D$ transition of Na,

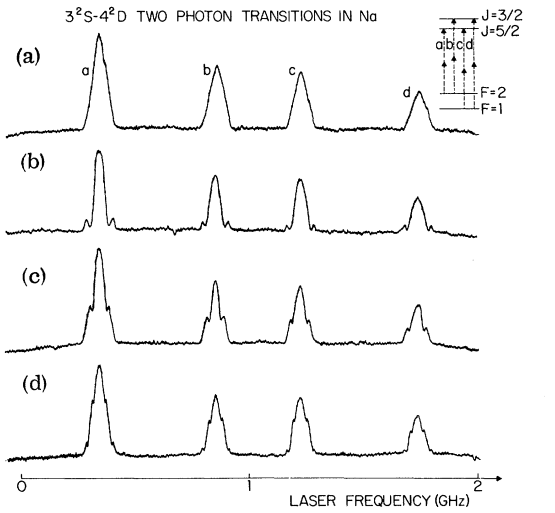


FIG. 3. (a) Recording of the four Doppler-free $3^2S$-$4^2D$ two-photon resonances of Na observed in the reference cell excited with a single pulse. (b)–(d) Same resonances observed in the sample cell excited with two time-delayed coherent pulses. Width of each pulse, $\tau = 8$ nsec; effective delay between two pulses, $\tau_{\text{eff}} = 2T = 17$, 25, and 33 nsec, respectively. The spacing between the fringes, $\Delta\nu \approx 60$, 40, and 30 MHz, respectively, is in good agreement with the theoretical prediction $1/2T$.

observed in the reference cell with a single pulse. Figure 3(b) shows the same four resonances observed in the sample cell which is excited by the two time-delayed coherent pulses with $2T \simeq 17$ nsec. An interference structure clearly appears on each resonance. We have verified that the splitting between the fringes is inversely proportional to the effective delay time $T_{\text{eff}}$ ($T_{\text{eff}} = 2T$) between the two coherent pulses. Clearly it is possible to choose $T$ longer than the radiative lifetime of the excited state, in which case the resolution would be better than the natural linewidth. One must not forget, however, that in such a case only atoms living a time at least equal to $T$ would contribute to the interference effect, which means that the fringes would have a very poor contrast. This is verified in Figs. 3(c) and 3(d), which show the same experimental traces as those of Fig. 3(b). Note, however, that in these cases [Figs. 3(c) and 3(d)] the contrast of the fringes is progressively less than that of Fig. 3(b), where a shorter delay is used. We have also verified that, with the delay line unlocked, the interference fringes disappear (as $\omega$ is varied).

In order to eliminate the diffraction background appearing in each of the four resonances of Figs.

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 $\omega$, with $\omega_0$ and $T$ fixed. Note, however, that the interference fringes reappear when $T$ alone (or $\omega_0$ 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