creased breaking occurs sooner.

Via computations, we have demonstrated the existence of V states—a new class of stationary nonlinear dispersive wave solutions of the Euler equations in two dimensions. The analytic solution to this bifurcation problem may be obtained from the nonlinear integro-differential equation (10) or by the methods of complex analysis. The stability of V states to small perturbations of arbitrary symmetry is an open question.

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[3]N. J. Zabusky, M. H. Hughes, and K. V. Roberts, "Contour Dynamics for the Euler Equations in Two Dimensions" (to be published). The present improved version of the code uses the following N-node discretized version of Eq. (6):

$$\vec{u}(x_m, y_m) = \sum_1^N \Delta u_n (\vec{e}_x \cos\theta_n + \vec{e}_y \sin\theta_n),$$

where Δu_n is obtained by integrating Eq. (6) over a straight line segment of slope $\tan\theta_n$ which connects nodes n and $n+1$. The $2N$ differential equations

$$(\dot{x}_m \vec{e}_x + \dot{y}_m \vec{e}_y) = \vec{u}(x_m, y_m) \quad (m = 1, 2, \ldots, N)$$

are advanced in time using a second-order predictor-corrector algorithm.

Optical Coherent Transients by Laser Frequency Switching: Subnanosecond Studies

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By extending the laser-frequency-switching technique to a 100-psec time scale, we have observed for the Na D_1 line the first-order free-induction decay, its inhomogeneous dephasing time T_2^*, its interference with the nonlinear free-induction decay, and a 1.8-GHz interference beat of the ground-state hfs. Detailed theoretical predictions of these new coherence effects are faithfully observed.

The method of *laser frequency switching*,[1] utilized recently in generating coherent optical transients, has provided new ways for examining the dynamic interactions occurring in molecules,[1] solids,[2] and atoms.[3] In this work, the frequency of a cw dye laser is abruptly switched by means of voltage pulses applied to an intracavity electro-optic phase modulator. A resonant sample in the path of this light exhibits coherent transients which are detected in the forward beam, allowing dephasing and population decay times to be measured thus far in the range 1 μsec to 5 nsec.

In the present study, laser frequency switching is extended to a time scale of 100 psec. This fiftyfold increase in time resolution is achieved without sacrificing the previous advantages of heterodyne detection, high sensitivity, and the ability to monitor the entire class of coherent optical transients by preselecting the voltage pulse sequence. Hence, quantitative studies of coherent optical transients in this time domain are now feasible.

New coherence effects may also arise at these short times as illustrated here for the optical free-induction decay[4] of an inhomogeneously broadened transition. This transient displays a polarization containing both a first-order and a nonlinear laser field dependence having different decay times, heterodyne beat frequencies, and laser tuning characteristics. The first-order free-induction decay (FID), which was predicted,[5] decays rapidly in the time of an inverse inhomogeneous linewidth T_2^* and is observed in the time domain for the first time by laser frequency switching. The well-known nonlinear FID[4] may be long-lived with a decay time determined by the power-broadened homogeneous linewidth. We view these two forms of FID as the transient analogs of steady-state linear and nonlinear (hole-burning) laser spectroscopy of an inhomogeneously broadened transition. Furthermore, the increased time resolution permits the first observations of very-high-frequency interference beats, for example, due to the 1.8-GHz hfs splitting of

the sodium ground state.

The above properties of FID follow from a density-matrix solution[5] of the Schrödinger wave equation where we assume that a Doppler-broadened atomic transition $1\rightarrow 2$ with center frequency ω_{21} is coherently prepared in steady state by a cw-laser beam. The system then radiates the FID transient when the laser frequency is suddenly switched from $\Omega\rightarrow\Omega'$. We make use of an earlier derivation[5] of the Doppler-averaged polarization and express the FID heterodyne beat signal E_b^2 in terms of an error function of complex argument $w(x+iy)$[6,7]:

$$E_b^2(L,t)=KE_0^2\exp\left(\frac{-t}{T_2}\right)\exp\left[-\left(\frac{\sigma t}{2}\right)^2\right]\exp[-i(\omega_{21}-\Omega')t]$$
$$\times\left\{(1+\eta)w\left(\frac{i\sigma t}{2}+\frac{i\Gamma-\delta}{\sigma}\right)+(\eta-1)w\left(-\frac{i\sigma t}{2}+\frac{i\Gamma+\delta}{\sigma}\right)\right\}+\text{c.c.} \quad (1)$$

Here, all frequencies are in angular units; the dipole dephasing time is T_2; the population decay time T_1; the Doppler linewidth is σ; we define the inhomogeneous dephasing time $T_2^*\equiv 2/\sigma$; the quantity $\delta\equiv\omega_{21}-\Omega$; the power-broadened linewidth is $\Gamma\equiv(1/T_2^2+\chi^2T_1/T_2)^{-1/2}$; the Rabi frequency is $\chi\equiv\mu_{12}E_0/\hbar$, where E_0 is the laser field amplitude and μ_{12} the transition matrix element; and the saturation parameter $\eta\equiv 1/(T_2\Gamma)$. This expression is nearly exact within the rotating-wave approximation and the slowly varying envelope approximation for an optically thin sample of length L.

In the asymptotic limit[7] as $t\rightarrow\infty$, Eq. (1) yields

$$E_b^2(L,t)=4(\eta-1)E_0^2K\exp(-t/T_2)\exp(-\Gamma t)\exp(-\delta^2/\sigma^2)\cos(\Omega-\Omega')t, \quad (2)$$

the anticipated nonlinear FID transient[4,5] where the emission occurs at the initial laser frequency Ω and produces with the laser beam a heterodyne beat of frequency $\Omega-\Omega'$. Note that the signal is nonlinear in the laser intensity due to the factor $E_0^2(\eta-1)$, the signal is absorptive, and the decay is an exponential with decay rate $1/T_2+\Gamma$.

In the short-time limit $t<2\Gamma/\sigma^2$, we expand the error function[7]

$$w(z)=1+(2iz/\sqrt{\pi})+\ldots$$

for $z<1$, retaining only the first two terms to obtain the initial behavior of the first-order FID:

$$E_b^2(L,t)\cong 4E_0^2K\exp\left(\frac{-t}{T_2}\right)\exp\left[-\left(\frac{\sigma t}{2}\right)^2\right]$$
$$\times\left\{\frac{1}{T_2}\left(\frac{1}{\Gamma}-\frac{2}{\sqrt{\pi}\sigma}\right)\cos(\omega_{21}-\Omega')t-\frac{2}{\sqrt{\pi}}\frac{\omega_{21}-\Omega}{\sigma}\sin(\omega_{21}-\Omega')t\right\}. \quad (3)$$

This radiation peaks at the Doppler line center at frequency ω_{21} and produces with the laser beam a beat of frequency $\omega_{21}-\Omega'$. The signal amplitude is linear in the laser intensity and contains both an absorptive and a dispersive part, which depends on the initial laser frequency through the factor $\omega_{21}-\Omega$. Here, the decay is a Gaussian, $\exp[-(\sigma t/2)^2]$. Consequently, at short times the first- and higher-order FID terms interfere in a variety of interesting ways, dependent on the particular conditions of light intensity and laser tuning. Numerical solutions of the general expression (1) are obtained using an error function subroutine, and the results of computer plots are given in Fig. 1 for the Na D_1 line where $(\Omega-\Omega')/2\pi=5$ GHz, $\sigma=5.8$ GHz, $\Gamma=1.6$ GHz, $\chi=2.3$ GHz, $T_2=32$ nsec, and $\eta=0.019$. At lower light intensity when $\chi=0.8$ GHz, the interferences are less striking and more closely resemble the observations of Fig. 2, which we now consider.

With the present optics, light from a Coherent 599-cw dye laser is frequency switched by a traveling wave electro-optic phase modulator[8] that is external to the laser cavity. This beam excites a sample resonantly and with the forward FID emission strikes a fast GaAs photodiode. The photodetector of 0.002-in. diam has a 30-psec response time or less and is mounted directly on an S-4 sampling head (25 psec response) of a Tektronix 7904 oscilloscope with a 7S11 sampling unit. The time-averaged transient signals are then sampled and stored digitally using a local computer.

The optical phase modulator is a lithium tantalate crystal ($0.5\times 0.65\times 50$ mm^3) mounted in a

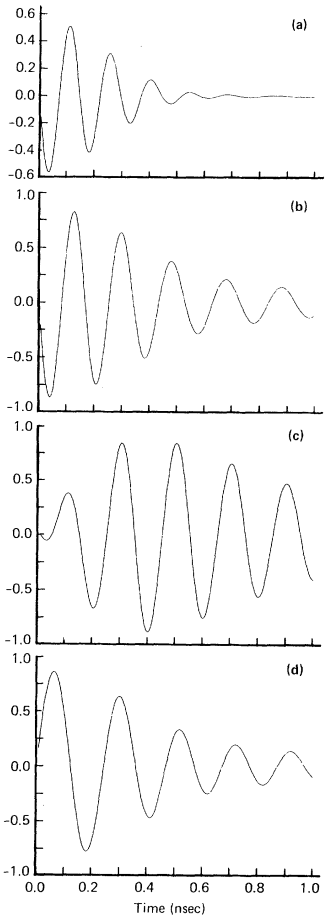

FIG. 1. Numerical solutions of the FID heterodyne beat signal $E_b^2(L,t)$ of Eq. (1) where $\chi = 2.3$ GHz, $(\Omega-\Omega')/2\pi = 5$ GHz, and $(\omega_{21}-\Omega)/2\pi$ in GHz equals (a) 1.5, (b) 1.0, (c) 0.0, and (d) -1.0. See text for other parameters.

stripline microwave transmission line. A square wave dc voltage pulse (Tektronix 109 pulser with a 700-Hz repetition rate) applied to this configuration propagates with the light wave down the length of the crystal in a time of 1.5 nsec, changing the optical refractive index from n_0 to n_1. Since the propagation velocities of the optical (c) and dc (v) waves differ, the light will experience a time-varying refractive index and hence a frequency shift

$$\Omega - \Omega' = \Omega \frac{n_1 - n_0}{n_0} \frac{v}{c \pm v}, \tag{4}$$

where the waves may travel either in the same direction (minus sign) or opposite direction (plus sign). Frequency shifts in the range 0 to 10 GHz have been achieved with a duration of 1.5 nsec, a rise time of ~ 100 psec, and at a rate of 17 MHz/V.

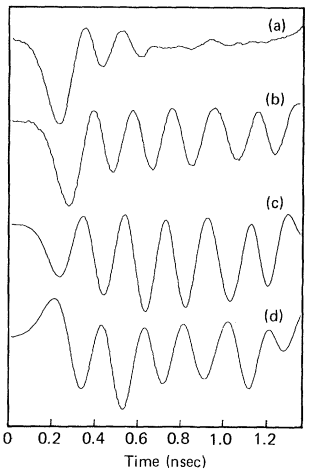

FIG. 2. Experimental FID heterodyne beat signals where $\chi = 0.8$ GHz and $(\omega_{21}-\Omega)/2\pi$ in GHz equals (a) 1.5, (b) 0.7, (c) 0.0, and (d) -1.0. The laser frequency shift is $(\Omega-\Omega')/2\pi = 5$ GHz, corresponding to a 335-V dc square-wave pulse. The gain in (a) is 2×.

In testing the above FID theory, we examined the behavior of the sodium D_1 line at 16 956.16 cm^{-1} using a 10-cm path length of Na vapor at $\sim 2\times10^{-6}$ Torr with 5% of the laser beam being absorbed. Figure 2 shows the observed FID heterodyne beat signals for different values of $\omega_{21} - \Omega$ where the frequency jump $(\Omega - \Omega')/2\pi = 5$ GHz and the laser power density is 3 W/cm² ($\eta = 0.055$). Note that the large frequency switch makes it possible for the first time to switch completely outside the Doppler linewidth, which for Na is 0.77 GHz. We see that as the initial laser frequency Ω is tuned through the Doppler line shape, the FID changes remarkably, displaying a rich assortment of new effects, in agreement with the results of Eqs. (1)–(3) and Fig. 1. Thus, in Fig. 2(a) where $(\omega_{21} - \Omega)/2\pi = 1.5$ GHz the detuning is large enough so that only the first-order FID is evident and only the *off-resonant velocity groups* contribute. The decay, which obviously is nonexponential, is complete in 500 psec, as expected since $T_2^* = 340$ psec. Because of the high laser intensity, the signal is dispersive and thus reverses sign when the laser is tuned to the other side of the Doppler peak, as in Fig. 2(d), where $(\omega_{21} - \Omega)/2\pi = -1$ GHz, in agreement with (3) and Fig. 1(d). Furthermore, the first-order FID beat frequency of 6.5 GHz in Fig. 2(a) is given by $\omega_{21} - \Omega'$, and is confirmed in other experiments by varying Ω'.

On the other hand, the nonlinear FID becomes more prominent when $\omega_{21} - \Omega$ is small, as it is in

Figs. 2(b)–2(d), because of the large-amplitude preparation of the *resonant velocity group*. Compare with Figs. 1(b)–1(d). The decay, which persists beyond the first-order FID, is exponential in agreement with Eq. (2), being given by $\sim e^{-\chi t}$ in this power-broadened regime with $\chi = 0.8$ GHz. Note that the decay is faster in Fig. 1 as $\chi = 2.3$ GHz. The observed beat frequency of 5 GHz is given by the laser frequency jump $(\Omega - \Omega')/2\pi$ and is verified in other experiments where this quantity is varied. These signals are also absorptive since their phase is invariant to laser tuning. In Fig. 2(c) where $\omega_{21} - \Omega = 0$, a dramatic interference between the linear and nonlinear FID occurs near the time origin, as in Fig. 1(c). Thus, all of the predictions of Eq. (1) seem to be obeyed.

In addition, in Fig. 2(d) where $\omega_{21} - \Omega = -1.0$ GHz, the laser is tuned midway between the two hyperfine components producing a modulated pattern, not present in Fig. 1, due to a 1.8-GHz interference beat of the ground-state hyperfine splitting. The origin of this beat is being studied further through its Fourier transform spectrum. Quantum beats have been observed previously in the upper state by spontaneous emission, in the lower or upper state by modulated photon echoes, and in superfluorescence but at frequencies which are at least one order of magnitude smaller.[9]

In the case of extreme homogeneous broadening when $\Gamma/\sigma \gg \sigma t$, Eq. (1) simplifies to give the NMR result

$$E_b^2(L,t) = KE_0^2 \exp\left(-\frac{t}{T_2}\right) \exp[-i(\omega_{21} - \Omega')t] \frac{1/T_2 - i\delta}{\Gamma^2 + \delta^2} + \text{c.c.}, \tag{5}$$

and then T_2 may be obtained in the absence of the above interference phenomena.

In extending this subnanosecond technique, it is evident that other coherent transients involving pulse preparation such as adiabatic rapid passage and photon echoes can be applied in quantitative optical dephasing studies where the time scale may be reduced even further.

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