Coherent Transients by Optical Phase Switching: Dephasing in $LaCl_3:Pr^{3+}$

A. Z. Genack and D. A. Weitz
Exxon Research and Engineering Company, Linden, New Jersey 07036

and

R. M. Macfarlane and R. M. Shelby
IBM Research Laboratory, San Jose, California 95193

and

A. Schenzle
Fachbereich Physik, Universität Essen, D-4300 Essen, West Germany
(Received 24 March 1980)

A novel coherent transient effect is observed following a rapid shift in the relative phase of an exciting laser and sample polarization. An analytical expression is derived for the signal which is a sum of exponentially decaying terms with characteristic times involving both T_1 and T_2. Phase switching is a powerful approach to the study of optical relaxation and is applied here to measure dephasing in $LaCl_3:Pr^{3+}$.

PACS numbers: 42.65.Gv, 42.10.Mg, 42.60.Fc, 76.30.Kg

Studies of optical coherent transients provide detailed dynamical information about selectively excited states in atoms, molecules, and solids. In the past, a variety of transient effects has been generated by bringing the sample in or out of resonance with the laser by switching either the laser amplitude,[1,2] the frequency of the sample polarization,[3] or the laser frequency.[4-7] Such transients are induced by changing the light-matter interaction in a time short compared to the homogeneous dephasing time of the excited transition. In this Letter, we present a new and powerful approach to transient spectroscopy in which the relative phase of the laser and sample polarization is switched.[8] Once the phase shift is accomplished, the laser and sample are again at the original frequency and a transient signal is detected as the sample polarization relaxes towards equilibrium. Unlike other coherent transient effects which can be used to measure relaxation rates, the laser interacts resonantly with the sample while the signal is detected. We present here an analytical solution for the phase-switched transient and show that this method can give the relaxation times of both the diagonal and off-diagonal elements of the density matrix, T_1 and T_2. With this method, coherent transient spectroscopy can be performed in the infrared (IR), visible, and ultraviolet on a time scale of ~50 psec to microseconds. The method should also make possible the observation of ultrafast transients with rf and microwave radiation. In this Letter, we confirm the theory by results in I_2 vapor and by measurements of dephasing in the $^3H_4$-$^1D_2$ transition of $LaCl_3:Pr^{3+}$ at low temperatures.

The power of the phase-switching method lies in its simplicity. The phase may be switched by applying a voltage step to a modulator placed in the laser beam. For the most rapid electronic switching a velocity-matched, extracavity modulator can be used to switch the phase in a time equal to the step rise time which can be $\lesssim 50$ psec. Since the required phase change is no more than π rad, a lower modulation voltage is required for observing transient signals with phase switching as compared to extracavity frequency switching.[6,7] The reduced voltage requirement is particularly attractive in the infrared where large voltages are necessary to shift the optical phase of long-wavelength radiation and should make possible the observation of direct absorption transients three orders of magnitude faster than has been demonstrated to date.[3,4] Even faster transients may be observed utilizing the ac Stark effect induced by a picosecond laser to switch the phase of the sample relative to an exciting cw laser.

The small field emitted by the sample propagates collinearly with the laser beam and at the same frequency; thus, the laser serves as the local oscillator for homodyne mixing to greatly enhance the detection sensitivity. We note that since the signal is unmodulated a reduced detection bandwidth is required for observing phase switching as compared to frequency switching. Thus the method of optical phase switching is ideally suited for rapid transients and yet is equally applicable to studying dephasing on all

longer time scales.

As an example of the transients observed in phase switching, the signal obtained for the $^3H_4$-$^1D_2$ transition at 6011 Å in $LaCl_3$:0.1% Pr^{3+} at 1.6 K is shown in Fig. 1(a). A 295-V step was applied to a 1039 D Lasermetrics deuterated ammonia dehydrogen phosphate (AD*P) modulator to change the phase by π rad and the signal shown was obtained from an average of 20 shots. We have observed similar signals in LaF_3:Pr^{3+} when the sample phase was switched by a pulsed Stark field. Using a CdTe extracavity modulator, we have also observed IR phase-switched transients in the 0-1 ν_3 transition of SF_6 with a CO_2 laser operating on the $P(18)$ transition at 10.6 μm.

We have obtained an analytical solution for the signal produced in an optically thin, inhomogeneously broadened two level system when the laser phase is instantaneously shifted. In the calculation, we consider relaxation that occurs in the presence of the driving field and begin by treating the response of a single frequency component of the inhomogeneously broadened line which is off resonance from the driving field by Δ. It is convenient to express the field whose phase is instantaneously shifted by φ at $t=0$ as

$$E(t)=2E\sin[\omega t+\varphi(t)]=-i(E^+e^{i\omega t}-E^-e^{-i\omega t}),$$

with

$$E^\pm=\begin{cases}E, & t<0,\\ Ee^{\pm i\varphi}, & t>0.\end{cases} \tag{1}$$

The components of the oscillating $P^\pm(t)$ corresponding to $E^\pm(t)$ are obtained by solving the Bloch equations, which may be written as[9]

$$\begin{aligned}\dot{P}^\pm&=(\pm i\Delta-1/T_2)P^\pm+2(\mu^2/\hbar)NWE^\pm,\\ N\dot{W}&=-N(W-W_0)/T_1-(E^+P^-+E^-P^+)\hbar,\end{aligned} \tag{2}$$

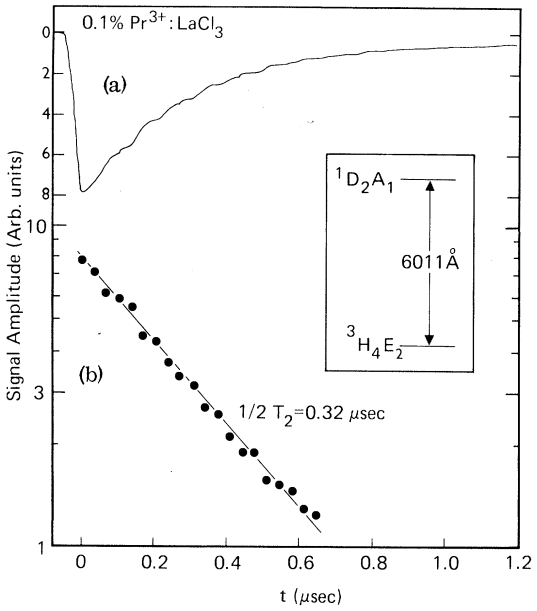

FIG. 1. Phase-switched decay in $LaCl_3$:0.1% Pr^{3+} at 1.6 K; (a) signal, and (b) logarithm of signal, giving $T_2=0.64$ μsec (homogeneous linewidth 500 KHz).

where μ is the dipole matrix element between the ground and excited state which is assumed to be real, N is the density of molecules for which the states involved in the transition are occupied, W describes the population inversion for the isochromat and W_0 is the equilibrium value of the inversion in the absence of the field. We solve these equations for the excitation conditions described in Eq. (1), and assume the system is in steady state at $t=0$. The signal we observe for $t>0$ is the homodyne beat between the laser field and the sample emission which is proportional to the polarization, i.e., $S(\Delta,t)\propto[P^+(\Delta,t)E^-+P^-(\Delta,t)E^+]$.

With use of a Laplace transform to solve Eq. (2), the signal is

$$S(\Delta,t)=4\pi(\omega n/c)W_0NL\chi^2e^{-i\varphi/2}\sin^2\tfrac{1}{2}\varphi$$

$$\times\sum_{j=1}^{3}\{1-\chi^2(T_1/T_2)[\Delta^2+(1/T_2)^2+\chi^2T_1/T_2]^{-1}+1/T_1Z_j\}\frac{(Z_j+i\Delta+1/T_2)e^{Z_jt}}{(Z_j-Z_{j-1})(Z_j-Z_{j+1})}+\text{c.c.}, \tag{3}$$

where $\chi=2E\mu_{12}/\hbar$ is the Rabi flopping frequency, n is the index of refraction of the medium, L is the length of the sample, and Z_j are the roots of the characteristic equation

$$F(Z)=[Z+(1/T_1)][Z+(1/T_2)]^2+\Delta^2[Z+(1/T_1)]+\chi^2[Z+(1/T_2)]. \tag{4}$$

Simple solutions for these roots can only be calculated in the cases (i) $T_1=T_2$, (ii) $\Delta=0$, and (iii) $\chi=0$. The results for the first two cases will be given elsewhere[10] and the third case is considered here as the starting point for a perturbation treatment in the low-power limit $\chi^2T_1T_2\ll1$. In this regime, power broadening is negligible and the intrinsic relaxation times T_1 and T_2 are exhibited in the signal. In

this case the roots of $F(z)$ may be expanded as

$$Z_1 = -1/T_1 - \chi^2\delta/(\delta^2+\Delta^2) + O(\chi^4),$$
$$Z_{2,3} = -1/T_2 \pm i\Delta \pm i\chi^2/2(\Delta \pm i\delta) + O(\chi^4), \tag{5}$$

where $\delta = 1/T_2 - 1/T_1$. To obtain the nonlinear phase-switched transient, one must collect all terms up to second order in χ in Eq. (3) and integrate the result over the inhomogeneous line profile. For excitation at the center of a Gaussian line and in the limit where the inhomogeneous linewidth, Δ_0, is much greater than $1/T_2$, we find

$$S(t) = \begin{cases} 4\pi^{3/2}(\omega n/c)W_0 NL\chi^2/[\,\Delta_0(1+\chi^2 T_1 T_2)^{1/2}\,] = S(0), & t<0, \\ S(0) + 4\pi^{3/2}(\omega n/c)(W_0/\Delta_0)NL\chi^4 T_1 T_2 \sin^2\tfrac{1}{2}\varphi & \\ \quad \times\left[\dfrac{1}{1-\epsilon/2}\exp\left(\dfrac{-2t}{T_2}\right) - \dfrac{\epsilon}{1-\epsilon}\exp\left(\dfrac{-2t}{T_2}\right) + \dfrac{\epsilon}{(1-\epsilon)(2-\epsilon)}\exp\left(\dfrac{-t}{T_1}\right)\right], & t>0, \end{cases} \tag{6}$$

where $\epsilon = T_2/T_1$.

We note that the normalized signal is characterized by only two parameters, T_1 and T_2, and also that the function is well behaved for $\epsilon = 1$ and $\epsilon = 2$. The transient is seen to be a nonlinear effect and the signal is proportional to the laser intensity squared while the terms linear in the laser intensity vanish in the limit $\Delta_0 \to \infty$. The discontinuity at $t = 0$, $\Delta S = S(0^+) - S(0^-)$, is analogous to the first-order effect in the free induction decay which displays a rapid change on the time scale Δ_0^{-1},[7,10] which was set equal to zero in the present calculation. The $\sin^2\frac{1}{2}\varphi$ dependence of the signal amplitude in Eq. (6), which is characteristic of the phase-switched transient, is confirmed in Fig. 2, which shows the jump in signal in iodine vapor as a function of phase shift imposed by a variable voltage pulsed across an extracavity AD*P phase modulator.

In the limit $T_2 \ll T_1$, the change in signal after the phase switch is

$$\Delta S(t) = 4\pi^{3/2}(\omega n/c)(W_0/\Delta_0)NL\chi^4 T_1 T_2 \sin^2\tfrac{1}{2}\varphi \exp(-2t/T_2), \tag{7}$$

and the dephasing time T_1 is given directly. We note that the signal in Eq. (7) decays at twice the dephasing rate $1/T_2$. This is because during the preparation time ($t<0$) a hole whose width j is the homogeneous linewith is imposed on the inhomogeneous absorption line, and after the phase shift ($t>0$) each isochromat relaxes at the rate $1/T_2$. Thus, the contribution from the preparation and probing stages add together to give the overall decay observed experimentally.[11]

The limit $T_2 \ll T_2$ is found to be appropriate to $LaCl_3$:Pr^{3+} as well as many other solids at low temperatures. In $LaCl_3$:0.1% Pr^{3+} we measure a phase-switched transient decay of 0.32 μsec (see Fig. 1) using preparation times of 2–10 μsec obtained by acousto-optically gating the light intensity at 50 Hz repetition rate. Signals were measured in the regime where their amplitude was proportional to the square of the laser power, i.e., in the absence of power broadening. The absorption by the sample was 8% and ~5 mW focused to ~1 mm diameter gave ~3% change in absolute transmission immediately after the phase switch. The population decay time $T_1 = 195 \pm 2$ μsec was measured by observing the fluorescence decay. Since the transient decay time is much shorter than T_1, we may assume that $T_2 \ll T_1$ and use Eq. (7) to obtain $T_2 = 0.64$ μsec, corresponding to a homogeneous linewidth full width at half maximum (FWHM) of 500 ± 30 kHz. As expected from Eq. (7) we obtained the same dephasing rate within experimental error from a measurement of free induction decay following laser-frequency switching. Previous work[12] with use of saturation spectroscopy measured a width of 7.5 MHz

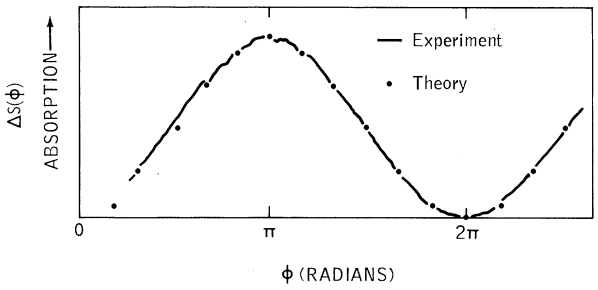

FIG. 2. Change signal $\Delta S(\varphi)$ in I_2 vapor measured as function of phase shift φ induced by voltage step applied to an AD*P modulator (0.011 rad/V at 5900 Å). Results are in good agreement with theoretical prediction, $\Delta S(\varphi) = \sin^2\frac{1}{2}\varphi = \frac{1}{2}(1-\cos\varphi)$.

limited by laser-frequency jitter. In our case laser, jitter contributed no more than ~30 KHz.

As in the much studied $LaF_3:Pr^{3+}$, (where $T_2 = 5.6$ μsec),[13] the homogeneous optical linewidth will be determined at low temperature by magnetic interactions between the Pr^{3+} ion and fluctuating fields due to surrounding nuclear spins. However, the ground state of Pr^{3+} in $LaCl_3$ is a non-Kramers doublet with a much larger (100×) magnetic moment of 1.0 Bohr magneton.[14] The faster optical dephasing observed in this material is consistent with stronger magnetic interactions arising from this large moment. A detailed treatment of this dephasing will require inclusion of Pr^{3+}-Pr^{3+} spin interactions, the effect of random strain fields[12] on the anisotropic magnetic moment, and the effect of the large Pr^{3+} moment on the nuclear spin dynamics.

In conclusion, we have described a new coherent transient effect in which the phase of the coherent exciting radiation is switched relative to that of the sample polarization. An analytical expression for the resulting transient signal has been derived and shown to directly yield the dephasing time T_2 in the limit of low power and $T_2 \ll T_1$. We have observed a phase-switched transient in the $^3H_4$-$^1D_2$ transition of $LaCl_3$:0.1% Pr^{3+} and found $T_2 = 0.64$ μsec. Because of its inherent simplicity and reduced modulation requirements compared to frequency switching, phase switching is a powerful approach to coherent transient spectroscopy in frequency regimes from the rf to the ultraviolet. It is particularly well suited to the measurement of fast dephasing and should considerably decrease the time scale on which direct absorption transients can be measured at infrared and microwave frequencies.

This work was supported in part by the U. S. Air Force Office of Scientific Research under Contract No. F49620-79-C-0108.

[1]N. A. Kurnit, I. D. Abella, and S. R. Hartmann, Phys. Rev. Lett. 13, 567 (1964), and Phys. Rev. 141, 391 (1966).

[2]W. H. Hesselink and D. W. Wiersma, Chem. Phys. Lett. 56, 227 (1978); D. E. Cooper, R. W. Olson, R. D. Wetting, and M. D. Fayer, Chem. Phys. Lett. 67, 41 (1979).

[3]R. G. Brewer and R. L. Shoemaker, Phys. Rev. Lett. 27, 631 (1971).

[4]J. L. Hall, in *Atomic Physics 3*, edited by S. J. Smith, G. K. Walters, and L. H. Volsky (Plenum, New York, 1973), p. 615.

[5]R. G. Brewer and A. Z. Genack, Phys. Rev. Lett. 36, 959 (1976).

[6]A. Z. Genack and R. G. Brewer, Phys. Rev. A 17, 1463 (1978).

[7]R. G. DeVoe and R. G. Brewer, Phys. Rev. Lett. 40, 862 (1978).

[8]A. Z. Genack, D. A. Weitz, and A. Schenzle, Bull. Am. Phys. Soc. 24, 896 (1979).

[9]A. Schenzle and R. G. Brewer, Phys. Rev. A 14, 1756 (1976).

[10]A. Schenzle and A. Z. Geneck, to be published.

[11]F. A. Hopf and R. F. Shea, Phys. Rev. A 7, 2105 (1973).

[12]C. Delsart, N. Pelletier-Allard, and R. Pelletier, Appl. Phys. Lett. 31, 443 (1977).

[13]R. M. Macfarlane, C. S. Yannoni, and R. M. Shelby, Opt. Commun. 32, 101 (1980), and references therein.

[14]C. A. Hutchison and E. Wong, J. Chem. Phys. 29, 754 (1958).

Light-Induced Critical Behavior in the Four-Wave Interaction in Nonlinear Systems

Chr. Flytzanis and C. L. Tang[(a)]

Laboratoire d'Optique Quantique, Ecole Polytechnique, 91128 Palaiseau cedex, France

(Received 5 March 1980)

It is shown that the four-wave interaction of a powerful stationary pump field and a weak signal field in a dielectric modeled with an assembly of anharmonically interacting particles exhibits a behavior radically different from that expected from the usual perturbation theory assuming small oscillation amplitudes. A light-induced critical behavior in the intensity of the reflected phase-conjugated field is obtained as a consequence of the interaction of the pump with the matter.

PACS numbers: 42.65.-k

It is well known that optical nonlinear processes result from the anharmonicity in the equation of motion of bound charges. These processes have almost exclusively been treated within the perturbation scheme.[1,2] We recall that in this scheme the induced polarization, which acts as a nonline-

