Measurement of optical dephasing by double phase switching

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We show that the optical dephasing time T_2 can be obtained from a measurement of the time delay T_0 between two shifts of $\pm\pi/2$ in the phase of the exciting laser for which a null occurs in the jump in the nonlinear transmission through a sample. The phase shift is imposed upon a single-mode cw laser by applying a rectangular voltage pulse to an electro-optic phase modulator placed in the laser beam. The time delay at which the null occurs is proportional to T_2, which is given by the expression $T_2=(2/\ln 2)T_0$. The method is demonstrated in atomic sodium and iodine vapor and the separation of third-order optical transients from linear transients induced by the phase shift is discussed. An analytic expression for the jump in the nonlinear transmission, induced by the second phase shift, as well as for the transient in transmission following a single phase shift of a laser which weakly saturates an optically thin sample is derived. It is also shown that optical dephasing rates may be inferred from the variation of the integral of the transmitted light in a double-phase-switching experiment.

INTRODUCTION

In a recent paper we explored the nature of the nonlinear transient induced in light transmitted through an absorbing sample by a sudden shift in phase of the exciting cw laser.[1] We found that, because the laser continues to drive the sample resonantly after the phase shift, the subsequent reestablishment of steady state is influenced by the relaxation time for level population, T_1 in addition to the phase coherence time T_2. For the case of an inhomogeneously broadened two-level system, the transient consists of a discontinuous rise followed by a sum of exponentially decaying terms involving T_1 and T_2 in separate terms. Except for the case $T_2 \ll T_1$, for which the transient signal reduces to a single exponential decay, a complicated transient results. In this paper we show that the optical dephasing rate can be determined by introducing a second phase shift at time $t=T$ which is equal and opposite to the initial shift at $t=0$. The dephasing time T_2 is measured independently of T_1 by employing phase shifts of $\pm\phi=\pm\pi/2$ rad and finding the value of T for which the jump in the nonlinear signal at T is zero.[2] The time at which this null occurs $t=T_0$ is shown to be directly proportional to T_2 and the method is demonstrated in atomic sodium and iodine vapor.

The use of a phase-switched sequence of coherent excitation pulses was originally developed in nuclear magnetic resonance (NMR) in connection with the techniques of spin-locking,[3] rotary-echo,[4] and multiple-pulse NMR.[5] Recently the optical analog of the rotary echo has been demonstrated.[6,7] In that experiment a modulated echo appears at a time 2τ following a phase switch applied at a time τ after the laser frequency is switched into resonance with a group of molecules in an inhomogeneously broadened transition. Our experiments, utilizing either a single or double phase switch differ from the NMR experiments and their optical analogs in two fundamental respects. First, only phase switching is employed—the sample is not switched into or out of resonance by pulsing the amplitude or frequency of the laser in the course of the experiment. Rather, cw excitation is used, and the sample is in steady state before the first phase switch is applied. Second, the intensity of the exciting radiation is sufficiently low that the transition is not power broadened. The effect of a shift in the laser phase on optical propagation in optically dense media has been studied previously.[2,8–10] Here we will restrict ourselves to the study of optically thin samples in which the laser field is not appreciably attenuated.

CONCEPT

A schematic diagram of the experimental arrangement is given in Fig. 1. The laser phase is shifted oppositely during the rising and falling edges of a rectangular voltage pulse of variable width T applied to an electro-optical phase modulator placed in the laser beam before the sample. A low power, single-mode cw laser beam is used so that power broadening is minimized and only the lowest-order nonlinear term in the polarization need be considered. In addition, we will only consider samples for which the homogeneous linewidth is a small fraction of the total inhomogeneous linewidth Δ_0, so that $T_2^* \sim 1/\Delta_0 \ll T_2$, where T_2^* is the time scale of the decay of the linear transient. This condition ensures that we can separately examine the nonlinear transient once the linear transient has decayed.

We focus our attention on the discontinuity in the nonlinear transient signal ΔS_2 at $t=T$, when the second phase switch is applied, because a null must exist in ΔS_2 at some time $t=T_0$. The time T_0 depends only upon T_1, T_2, and ϕ, and this should give an independent determination of relaxation in the sample. The existence of a null is evident from considering two limiting cases illustrated in Fig. 2. For $0 \le t \le T \ll T_1, T_2$ the nonlinear polarization has not yet responded much to the phase change of the driving field. Therefore, when the field returns to its original phase at $t=T$, the system is returned nearly to its initial state and only a small change in the signal level occurs. This requires a second discontinuity at $t=T$ equal but opposite in sign to the initial jump at $t=0$

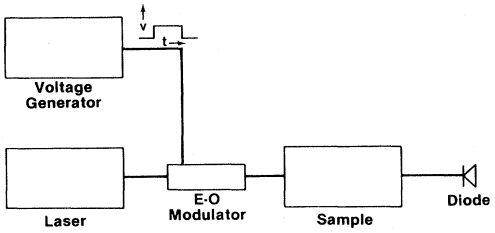

FIG. 1. Schematic diagram of apparatus used in double phase switching. Equal and opposite phase shifts of the laser light are produced by applying a rectangular voltage pulse to an electro-optic modulator placed in the laser beam. Transmission through the sample is detected in a photodiode.

$$\Delta S_2 = -\Delta S_1 \ .$$

On the other hand, for $t \gg T_1, T_2$ the system reaches steady state with respect to the phase-switched field. The state prior to the second phase switch, therefore, is identical to the state before the first one, aside from an unimportant overall phase factor. When the phase is switched again at $t=T$ the signal will have to be identical to the one at $t=0$ and

$$\Delta S_2 = \Delta S_1 \ .$$

Since $\Delta S_2(T)$ is a continuous function of the delay time which changes sign

$$\Delta S_2(T) = \Delta S_1(0) \times \begin{cases} -1, & t \to 0 \\ +1, & T \to \infty \end{cases}$$

there must be a finite time $T=T_0$ where the discontinuity $\Delta S_2(T_0)$ vanishes.

The functional dependence of ΔS_2 on T, ϕ, T_1, and T_2 is obtained in the Appendix. There, the response of a two-level optically thin sample to the time-dependent field

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

with

$$E^{\pm} = E \times \begin{cases} 1, & t < 0 \\ e^{\pm i\phi}, & 0 < t < T \\ 1, & t > T \ . \end{cases} \tag{1}$$

is given.

In order to interpret the present experimental results, it will suffice to have a qualitative understanding of the various components of the transient signal. The linear response of the sample polarization follows the transient field in Eq. (1) in a time $\sim T_2^*$ and so, in the limit $\Delta_0 \to \infty$, the response is instantaneous and no transient is observed since the laser intensity is constant. In this limit, the first-order response contains no information about the dynamics of the sample, but merely reflects the modulation of the exciting laser field. The third-order polarization, which is the lowest-order nonlinear term of the sample response, is continuous and relaxes towards steady state in a time $\sim T_2$. The transmitted light due to this term is a homodyne beat of the continuous emission due to the sample and the piecewise continuous field in Eq. (1) and exhibits a discontinuity at $t=0$ and T. In interpreting our experiments, we will ignore the rapidly decaying contribution of the linear polarization and look for the contribution of the third-order polarization to the light transmitted through the sample. This is characterized by a step in transmission at $t=0$ and T that decays in a time $\sim T_2 > T_2^*$. It is this nonlinear response which is illustrated in Fig. 2. For the special choice of $\phi=\pi/2$, the time T_0 ($\phi=\pi/2$), at which a null occurs in ΔS_2, becomes independent of T_1 as shown in the Appendix. The dephasing time is obtained in Eq. (A10) and is

$$T_2 = (2/\ln 2) T_0(\pi/2) \ . \tag{2}$$

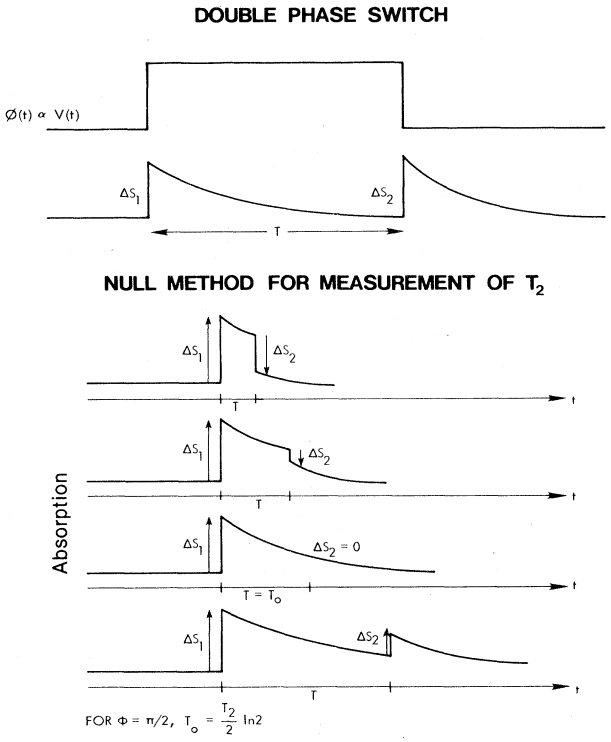

FIG. 2. A schematic representation of the nonlinear transient absorption for various delay times T between the two phase shifts. Rapidly decaying linear transient following each phase shift is not shown. Transient for T less than, equal, and greater than the time T_0 at which a null occurs in ΔS_2 is illustrated.

EXPERIMENT

We have observed double-phase-switching transients in both sodium and iodine vapor. In these experiments the phase is shifted by applying a rectangular voltage pulse to a Lasermetrics Model 3126B, lithium tantalate, traveling-wave, phase modulator placed in the laser beam. The modulator was composed of two crystals, 25 mm in length with electrodes deposited along the length of the c faces of the crystals and separated by the crystal height of 0.65 mm. The impedence of the modulator is close to 50 Ω.

The rise time for the phase switch is 0.6 nsec which results from the difference in velocity of the light and of the applied voltage across the modulator.

A Tektronix 109 pulse generator with a 150-psec rise time and pulse width determined by the length of charging cable was used for experiments in sodium. A Hewlett Packard 1905 pulse generator with 7-nsec rise and fall times was used for experiments in iodine vapor. The signal was detected with a pin diode coupled to a preamplifier manufactured by B&H which together had a 3-GHz bandwidth. The transients were averaged using a Tektronix R7912 transient digitizer with 500 MHz bandwidth. The rise time observed for the transient is ~ 1 nsec for experiments in sodium and ~ 8 nsec for iodine. A number of experimental artifacts in the transmitted light were observed and eliminated in this experiment. In the absence of a sample a step is observed while the voltage is applied to the modulator and a ringing is seen following the voltage switches. We found that the step can be eliminated by adjusting a linear polarizer placed after the modulator and that the ringing can be eliminated by adjusting the polarization of the light entering the modulator with a polarization rotator and ensuring that the light was not truncated by apertures in the system.

A null in ΔS_1 in iodine is observed as the voltage step height is varied with 53.5 V applied to the modulator and corresponds to a phase shift of 2π as seen in Eq. (A8). The pulse height was set to 13.4 V to achieve a $\pi/2$ phase shift.

The double-phase-shift transient in sodium vapor at a time for which $\Delta S_2 \sim 0$ is shown in Fig. 3. Laser power of 1 mW is expanded to a diameter of 4 cm before entering the sodium cell. The cell is 10 cm long and is heated to 100 °C. With the laser tuned to the center of the sodium-D line, 10% of the light is absorbed. The sharp dip in absorption at T_0 results from reduced absorption during the time the laser phase is shifting. For the duration of the phase shift $\Delta t \sim 0.6$ nsec the laser is frequency shifted by $\Delta\omega \sim \pm\pi/2\Delta t$. Since $\Delta t > T_2^* \approx 100$ psec, the linear component of the sample polarization can approach steady state with the frequency-shifted laser field and absorption of the laser beam is reduced while the laser frequency is shifted off the center of the line. This results in a spike in the signal at $t = T_0$. On the other hand, at $t = 0$ the spike is not evident because the jump in the nonlinear signal ΔS_1 is opposite in sign to the first-order transient. In a subsequent publication we will describe observations of linear-phase-switched transients where the laser phase is shifted in a time much shorter than T_2^*. The slowly decaying nonlinear transient in sodium increased in magnitude by a factor of ~ 4 when the laser power was doubled. This confirmed that only the third-order nonlinear term contributed significantly to the nonlinear response.

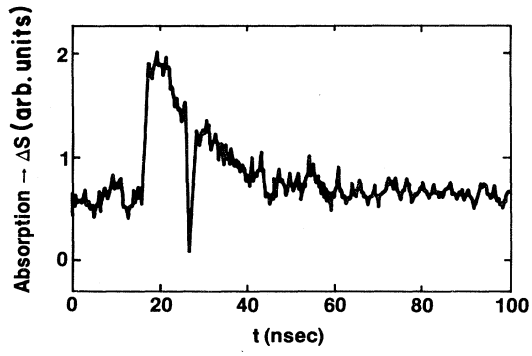

FIG. 3. Double-phase-shift transient in sodium vapor for a delay of 11 nsec between the phase shifts of $\pm\pi/2$. Sharp spike at the time of the second phase shift is due to the linear transient caused by the change in absorption during the time that the laser phase is shifting.

The null in ΔS_2 for sodium occurs at $T = 11$ nsec. Using Eq. (A10) this gives $T_2 \sim 29$ nsec. This is in reasonable agreement with the measured value of 32 nsec and is consistent with the 16-nsec radiative decay time since $T_2 = 2T_1$ for dephasing limited by the excited-state lifetime.[11]

In Fig. 4 we show results of double phase switching in iodine vapor at a pressure of 30 mTorr for two values of T. The $(v,J) = 2,59 \rightarrow 15,60$ transition of the $X\,^1\Sigma_g^+ \rightarrow B\,^3\Pi_{0^+_u}$ electronic transition, which lies 8 GHz above the lower frequency sodium-D line, was excited. Iodine is not strictly a two-level system since the ground- and excited-state populations decay at different rates due to coupling with a large manifold of levels. Nonetheless, a null in ΔS_2 must exist and is a rough measure of relaxation of the sample. The spike observed immediately following the phase shift arises from an overshoot of the voltage applied to the modulator which is shown in Figs. 4(b) and 4(d). Laser power of 5 mW are used in these experiments. Lenses with focal lengths of 10 cm are used to focus the laser beam before passing through the phase modulator and to recollimate the beam before it enters the 20-cm-long cell. From the results in Fig. 4(a), we obtain a value for $T_0(\pi/2)$ of 75 nsec. Using the right-hand side of Eq. (A10) to define an effective relaxation time, we obtain a time of 215 nsec. The measured dephasing time using photon echoes is 340 nsec.[12] The shorter effective relaxation time measured in the double-phase-switching experiment is expected since the transition was slightly power broadened in this experiment.

Observing ΔS_2 requires a detector time response which is considerably faster than T_2. It is possible, in principle, to observe the intrinsic dephasing even with a detector with a response slower than T_2 by measuring the integral of the double-phase-switched transient as a function of the delay T between the two phase shifts. The contribution of the linear transient to this integral should be independent of T as long as $T > T_2^*$. A comparison of the integrated signal with theory would require the solution for $P^{\pm}(t)$ for $t > T$ which we have not attempted as yet. A simple result, however, is obtained for the case $T_2/T_1 = \epsilon \ll 1$. The integral of the normalized nonlinear transient for a single phase switch is

$$\int_0^\infty g(t)dt = T_2[1+\epsilon/2-\epsilon^2/8+O(\epsilon^3)]\,, \tag{3}$$

thus the integral of the normalized nonlinear transient for $T \gg T_2 T_1$ should be $2T_2$, whereas the integral for $T = T_0$ should be $\simeq T_2$. Thus T_0, and hence T_2, could readily be found in this case.

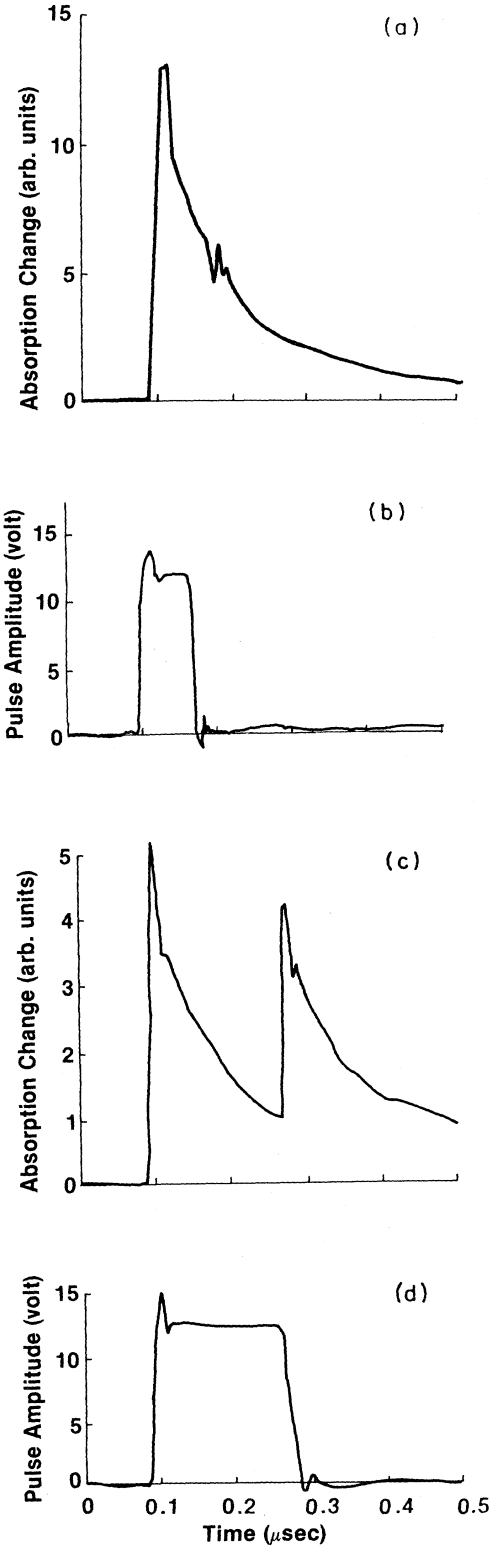

FIG. 4. Double-phase-switching transients in iodine vapor for $T=T_0$ and $T>T_0$ are shown, respectively, in (a) and (c). Respective voltage pulses are shown in (b) and (d).

CONCLUSION

In summary, we have shown that the optical dephasing time in a two-level system may be obtained by measuring the time at which a null occurs in the nonlinear transient following a second phase shift. When $T_2 \ll T_1$ an integral method may be employed and the dephasing time may be inferred from the variation of the integral of the transmitted light in a double-phase-switching experiment with delay time T.

APPENDIX: EXPRESSION FOR NONLINEAR TRANSIENT

In this appendix we obtain expressions for the third-order signal following a single phase shift and for the jump in transmission following the second phase shift. For a two-level system and optically thin sample, the sample polarization may be written as

$$p(z,t)=-i[p^+(t)e^{+i(\omega t-kz)}-p^-(t)e^{-i(\omega t-kz)}] \quad \text{(A1)}$$

with $p^+=N\mu_{21}P^+$ and $p^-=N\mu_{12}P^-$, where N is the density of molecules involved in the transition between the ground and excited states labeled 1 and 2, respectively, k is the wave vector of light in the medium, μ_{21} and μ_{12} are the matrix elements of the molecular dipole moment, and P^+ and P^- are the off-diagonal elements of the density matrix ρ in a frame rotating at the laser frequency

$$\left.\begin{matrix}\rho_{12}\\ \rho_{21}\end{matrix}\right\}=P^{\pm}e^{\pm i(\omega t-kz)} .$$

Maxwell's wave equation, $\partial E_s^{\pm}/\partial z=-2\pi ikp^{\pm}$, then yields $E_S^{\pm}=-2\pi ikLp^{\pm}$ for the rotating components of the field emitted by a sample of length L. Neglecting terms of order E_S^2, the change in transmission due to the presence of the sample is the homodyne signal

$$S=(E^+E_S^-)=-2\pi iNk\mu L(E^+P^-+E^-P^+) . \quad \text{(A2)}$$

In Eq. (A2) we have assumed without loss of generality that $\mu_{12}=\mu_{21}$ is real.

In order to unambiguously separate the time scale of the transient effects related to terms in the polarization which are linear from those that are nonlinear in the field E, we will assume that the inhomogeneous linewidth is infinitely broad, $\Delta_0\to\infty$. The linear polarization then adiabatically follows the field $P^{\pm}\propto E^{\pm}$. It is then convenient to express the off-diagonal elements of the density matrix as the sum of linear and nonlinear components $P^{\pm}=P_L^{\pm}+P_{NL}^{\pm}$. The response of the molecular polarization to the discontinuous phase jumps of the field in Eq. (1) is discontinuous through $P_L^{\pm}$. However, the homodyne signal due to $P_L^{\pm}$ is

$$S_L=(P_L^+E^-+P_L^-E^+)\sim|E^2| , \quad \text{(A3)}$$

which is constant in a phase-switching experiment. Therefore, the transient signal is due to the contributions of $P_{NL}^{\pm}$, which responds continuously to the phase-shifted field. The transient signal is thus

$$S=S_{NL}=P_{NL}^+(t)E^-(t)+P_{NL}^-(t)E^+(t) , \quad \text{(A4)}$$

which is discontinuous whenever $E^{\pm}(t)$ is discontinuous. The jump in signal at $t=T$ is

$$\Delta S_2(T)=S_{\rm NL}(T^+)-S_{\rm NL}(T^-)$$
$$=P^+_{\rm NL}(T)[E^-(T^+)-E^+(T^-)]+\text{c.c.}$$
$$=2iP^+_{\rm NL}(T)e^{i\phi/2}+\text{c.c.} \quad \text{(A5)}$$

Thus, the amplitude of the discontinuity at the second phase switch is obtained once the time dependence of the nonlinear polarization following the first phase switch is calculated. This is done by first solving the Bloch equations for a group of molecules within the inhomogeneous line which are off resonance by the angular frequency Δ from the exciting frequency and then integrating the result over the inhomogeneous line.

The Bloch equations, which describe the time dependence of elements of the density matrix in the presence of the electrometric field $E(t)$, may be written[13]

$$\dot{P}^{\pm}(t)=\left[\pm i\Delta-\frac{1}{T_2}\right]P^{\pm}(t)+2gE^{\pm}W\ , \quad \text{(A6a)}$$

$$\dot{W}(t)=\frac{1}{T_1}(W-W_0)-g[P^+(t)E(t)-P^-(t)E^+(t)]\ , \quad \text{(A6b)}$$

where $W=\rho_{22}-\rho_{11}$ is the inversion, W_0 is the equilibrium value of the inversion in the absence of the laser field, and $g=\mu/\chi$. We solve these equations in the limit $\chi^2T_1T_2\ll 1$, where $\chi=\mu E/\hbar$ is the Rabi flopping frequency. In this limit we may drop terms in $P^{\pm}$ which are higher than third order in E. Using the Laplace transform we solve the Bloch equations [Eq. (A6)] for the exciting field given in Eq. (1) for $t<T$. Integrating over Δ we obtain the result

$$P^{\pm}(t)=\begin{cases}P^{\pm}=\pi W_0\chi^2(1+T_1T_2\chi^2)^{-1/2}, & t<0 \quad \text{(A7a)}\\ \pi W_0\chi(1+T_1T_2\chi^2)^{-1/2}e^{i\phi}-i\pi W_0\sin\phi e^{i\phi}T_1T_2\chi^3(g(t)-2e^{-2t/T_2}) & \\ +i\pi W_0\sin(\phi/2)e^{i\phi/2}T_1T_2\chi^3g(t), & 0<t<T \quad \text{(A7b)}\end{cases}$$

with

$$g(t)=\frac{\epsilon}{(1-\epsilon)(2-\epsilon)}e^{-t/T_1}+\frac{2}{2-\epsilon}e^{-2t/T_2}$$
$$-\frac{\epsilon}{1-\epsilon}e^{-t/T_2}\ ,$$

and $\epsilon=T_2/T_1$.

The change signal following a single phase switch can be shown from Eqs. (A2) and (A7) to be

$$S=\Delta S_1 g(t)$$

with

$$\Delta S_1=4\pi^{3/2}k(W_0/\Delta_0)\text{NL}\chi^4T_1T_2\sin^2\tfrac{1}{2}\phi\ . \quad \text{(A8)}$$

We note that $g(t)$ has the limiting values $g(0)=1$ and $g(\infty)=0$. Using Eq. (A5) the second jump is

$$\Delta S_2(T,\phi)=\Delta S_1[(1-4e^{-2T/T_2}\cos^2\tfrac{1}{2}\phi)$$
$$-2g(T)(1-2\cos^2\tfrac{1}{2}\phi)]\ . \quad \text{(A9)}$$

The second term in the square brackets in Eq. (A9) can be set to zero by choosing $\phi=\pi/2$. ΔS_2 then becomes independent of T_1 with a null at

$$T_2=T_2(2/\ln 2)T_0(\pi/2)\ . \quad \text{(A10)}$$

In general, both T_0 and ΔS_2 $[t=T_0(\pi/2)]$ vary with ϕ. However, for the special case $g(t)=e^{-2t/T_2}$, which is obtained when $T_2\ll T_1$, $\Delta S_2(T_0(\pi/2),\phi)$ is independent of ϕ since

$$\Delta S_2(T_0(\pi/2),\phi)$$
$$=\Delta S_1(\phi)[1-2g(T)](1-2\cos^2\tfrac{1}{2}\phi)=0\ , \quad \text{(A11)}$$

when $g(T_0)=\frac{1}{2}$. Equivalently, we may say that T_0 is independent of ϕ in this case. A variation of $\Delta S_2(T_0(\phi/2))$ with ϕ, therefore, indicates that T_1 is not much larger than T_2.

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