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Coherent interactions in pump–probe absorption measurements: the effect of phase gratings

S. L. Palfrey and T. F. Heinz

IBM Thomas J. Watson Research Center, P.O. Box 218, Yorktown Heights, New York 10598

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We show that in order to describe completely the coherent coupling between the pump and probe pulses in a pump–probe measurement of transient absorption, the influence of induced phase gratings must be included. The importance of phase gratings is demonstrated experimentally for the case of a bleachable dye and analyzed in terms of transient four-wave mixing. These results are relevant to the interpretation of pump–probe measurements on all time scales performed with pulses from a single laser, particularly when the pulse duration is comparable with the material response time.

1. INTRODUCTION

In transient absorption measurements performed with pulses from a single laser, it has been recognized for some time that a coherent interaction between the pump and probe pulses can alter the observed probe transmission when the two pulses overlap temporally in the sample.[1–4] A complete understanding of this interaction is crucial in the measurement of ultrafast relaxation times comparable with the laser pulse duration. For the usual geometry in which the pump and probe cross at an angle, this interaction can be described in the following way. When the pulses are mutually coherent in the sample, they set up a spatial modulation in the medium's optical properties. This induced grating can then scatter pump radiation into the probe beam and thereby affect the probe intensity. For transform-limited pulses, the scattered pump radiation will be in phase with the probe if the grating is formed by a modulation in the absorption coefficient (an amplitude grating). If, however, the grating results from a modulation in the refractive index (a phase grating), the scattered radiation will be in quadrature with the probe.[5] These considerations imply that only amplitude gratings and not phase gratings will have an appreciable effect on the probe signal.

We demonstrate here that induced phase gratings can also lead to a strong coherent coupling of the pump and probe pulses, provided that the pulses exhibit some degree of phase modulation.[6] In a system with a finite relaxation time, the phase grating will build up gradually and reflect the average phase difference between the electric fields of the pump and probe pulses. If this phase difference remains constant, the pump radiation scattered from the induced phase grating will be precisely in quadrature with the probe beam. If, on the other hand, the phase difference between the pulses changes in time, pump light scattered from this grating will reflect this phase shift and will no longer remain in quadrature with the field of the probe beam. In this manner, the pump radiation scattered from the induced phase grating can interfere with the probe beam and strongly influence its intensity.

In this paper we present an analysis of this effect in terms of four-wave mixing and illustrate its significance with measurements of transient bleaching in dyes. We find that this previously unnoticed interaction can have a strength comparable with that associated with the amplitude grating. The influence of the phase grating is, of course, particularly pronounced in the wing of an absorption where changes induced in the refractive index exceed those in the absorption coefficient. Our measurements indicate that, even for nearly transform-limited pulses from a well-mode-locked laser, the residual phase modulation can be sufficient to make the effect of the phase grating significant. The distinctive feature of the phase grating is that it manifests itself as a component of the probe signal that is antisymmetric with respect to the delay time of the probe pulse. This result is important for the correct interpretation of pump–probe measurements in the region of short delay times.

2. THEORY

The coherent and incoherent signals obtained in pump–probe experiments can be understood by considering the nonlinear response of the medium to the two pulses. The contribution of the phase grating can be described within the framework previously developed for treating the amplitude grating.[2–4] We restrict ourselves to the regime of a weak nonlinear response in the sample, in which case the coherent coupling is a form of transient four-wave mixing. The phase grating is associated with the real part of the third-order nonlinear susceptibility, while the amplitude grating arises from its imaginary part. To analyze this transient four-wave mixing process we first derive the time dependence of the nonlinear polarization and then calculate the influence of this polarization on the transmitted probe energy. For the case of a system with rapid dephasing, the third-order polarization is given by the product of the electric field and the change in the linear polarizability arising from population changes induced by the field at earlier times.[7] If $\mathbf{E}(t)$ is the complex field envelope for an implicit $e^{-i\omega t}$ time dependence, then the nonlinear polarization at frequency ω is given by

$$P_i^{(3)}(t) = E_j(t) \int_{-\infty}^{\infty} dt' E_k^*(t') E_l(t') [A'_{ijkl}(t - t') + iA''_{ijkl}(t - t')]. \quad (1)$$

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Here $A'(t-t')$ and $A''(t-t')$ are proportional to the real and imaginary parts of the third-order susceptibility, respectively, and describe the response of the real and imaginary parts of the linear susceptibility at time t to the field at some earlier time t'.

In a pump–probe measurement, the total electric field $\mathbf{E}$ consists of the sum of the pump field $\mathbf{E}_1$ and the probe field $\mathbf{E}_2$. The rate of energy loss in the probe beam resulting from this polarization is proportional to $\mathrm{Im}[\mathbf{E}_2^* \cdot \mathbf{P}^{(3)}]$ at each point in the sample. The total change in the probe energy then follows from integrating this quantity over time and over the sample volume. For pump and probe beams that can be described with planar wave fronts, the result of the integration over a plane transverse to the beams is that only the terms of $\mathbf{P}^{(3)}$ with the same wave vector as the probe affect the probe energy. Furthermore, if the angle between the two beams is sufficiently small and the sample is optically thin, the contribution to the signal integrated over time will be the same for each transverse plane in the sample.[8] If we take the pump and probe fields to be polarized in the x direction, we find that the change in the probe energy is proportional to

$$S = \mathrm{Im}\Big\{\int_{-\infty}^{\infty} E_2^*(t)E_1(t)\int_{-\infty}^{\infty} E_1^*(t')E_2(t') \times [A'_{xxxx}(t-t') + iA''_{xxxx}(t-t')]\mathrm{d}t'\mathrm{d}t + \int_{-\infty}^{\infty} E_2^*(t)E_2(t)\int_{-\infty}^{\infty} E_1^*(t')E_1(t') \times [A'_{xxxx}(t-t') + iA''_{xxxx}(t-t')\mathrm{d}t'\mathrm{d}t\Big\} \tag{2}$$

We now consider the usual case in pump–probe measurements where the probe is a copy of the pump delayed by time τ, i.e.,

$$E_1(t) = E(t) \tag{3a}$$

and

$$E_2(t) = E(t-\tau)e^{i\omega\tau}. \tag{3b}$$

The observed change in probe energy is then given by

$$S(\tau) = \gamma(\tau) + \beta_1(\tau) + \beta_2(\tau), \tag{4}$$

where

$$\gamma(\tau) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} |E(t-\tau)|^2|E(t')|^2 A''_{xxxx}(t-t')\mathrm{d}t'\mathrm{d}t, \tag{4a}$$

$$\beta_1(\tau) = \mathrm{Re}\Big\{\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} E^*(t-\tau)E(t)E^*(t') \times E(t'-\tau)A''_{xxxx}(t-t')\mathrm{d}t'\mathrm{d}t\Big\}, \tag{4b}$$

and

$$\beta_2(\tau) = \mathrm{Im}\Big\{\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} E^*(t-\tau)E(t)E^*(t') \times E(t'-\tau)A'_{xxxx}(t-t')\mathrm{d}t'\mathrm{d}t\Big\}. \tag{4c}$$

This result contains both the incoherent and coherent contributions. The incoherent term $\gamma(\tau)$ is given by the convolution of the pulse-intensity autocorrelation with the response function for the induced bleaching (or absorption). Note that effects arising from changes in the real part of the susceptibility do not enter here, nor does $\gamma(\tau)$ depend in any way on the phase of the electric-field envelope. While the contribution of $\gamma(\tau)$ will persist as long as the material response remains, the coherent terms $\beta_1(\tau)$ and $\beta_2(\tau)$ are present only when the pump and probe pulses overlap temporally and are mutually coherent in the sample. Without this coherence, the fields of the pulses cannot build up a stable diffraction grating, and these terms will be washed out. The contribution of $\beta_1(\tau)$ results from the amplitude grating in the sample and has been discussed thoroughly elsewhere.[1–4] The new feature of this calculation, as represented by the term $\beta_2(\tau)$, is the explicit inclusion of the effects of the phase grating.

One reason that the contribution of the phase grating to the coherent coupling may not have been considered previously is that in the frequently discussed case of a real pulse envelope (transform-limited pulses), $\beta_2(\tau)$ is identically zero. Intuitively, this result is expected, since for a real field envelope the polarization associated with the real part of the nonlinear susceptibility will always be in phase with the probe field and will therefore not affect the probe energy to first order. However, for non-transform-limited pulses, the relative phases of the fields vary as the polarization builds up with time. The real part of the nonlinear susceptibility can then lead to a component of the polarization in quadrature with the probe field and thereby induce a change in the probe energy.

An important observation about the contribution of $\beta_2(\tau)$ to the coherent signal is that it will have a very different form from that of $\beta_1(\tau)$. In particular, if we replace τ by $-\tau$ in Eqs. (4b) and (4c) we find that, while $\beta_1(\tau)$ is symmetric about zero delay, $\beta_2(\tau)$ is antisymmetric. This is, to our knowledge, the first time it has been explicitly noted that the coherent signal may be asymmetric. The antisymmetry of the β_2 term can be understood in the following way. In the case of rapid dephasing, the real part of the nonlinear susceptibility cannot affect the energy stored in the medium. Within our model, therefore, any effects arising from the phase grating will conserve the energy in the two pulses and can only give rise to an energy transfer between the two beams. Since exchanging $-\tau$ for τ in Eq. (4) is equivalent to exchanging the pump and the probe, it must therefore also change the sign of β_2. As a consequence of the form of $\beta_2(\tau)$, we note that the peak of the coherent signal will not generally occur at zero delay time as it would in the absence of the phase grating. Depending on the form of the electric-field envelope and the relative sign of the real and imaginary parts of the nonlinear susceptibility, the peak of the coherent contribution may be either at negative or positive delay times. However, since $\beta_2(\tau)$ vanishes for $\tau = 0$, the previously noted relation that the incoherent and coherent signals are equal at zero delay time still applies.[1,2]

3. EXPERIMENT

In order to examine the influence of phase gratings on the coherent interaction of pump and probe pulses experimentally, we have measured the probe transmission in a well-characterized system both near resonance and in the wing of the resonance. By tuning the frequency of the laser, we could observe the response when the phase grating had a lesser or

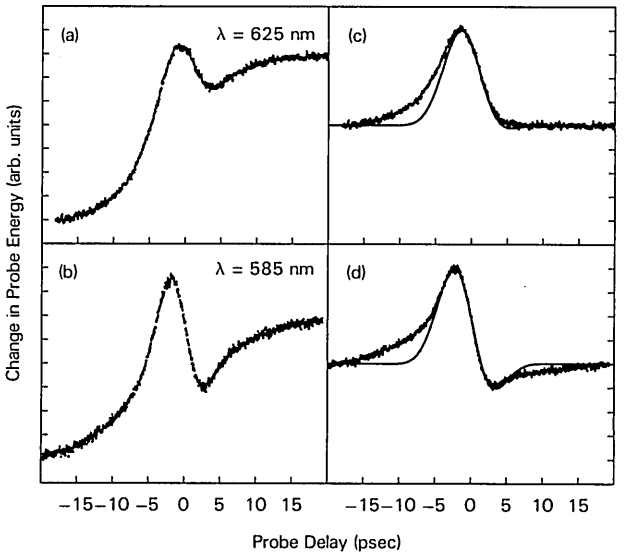

Fig. 1. Probe transmission in Nile blue for nearly transform-limited pulses taken at (a) 625 nm and (b) 585 nm; (c) and (d) show the data corresponding to (a) and (b) after subtracting the incoherent signal as well as the results of the theoretical fits (solid lines).

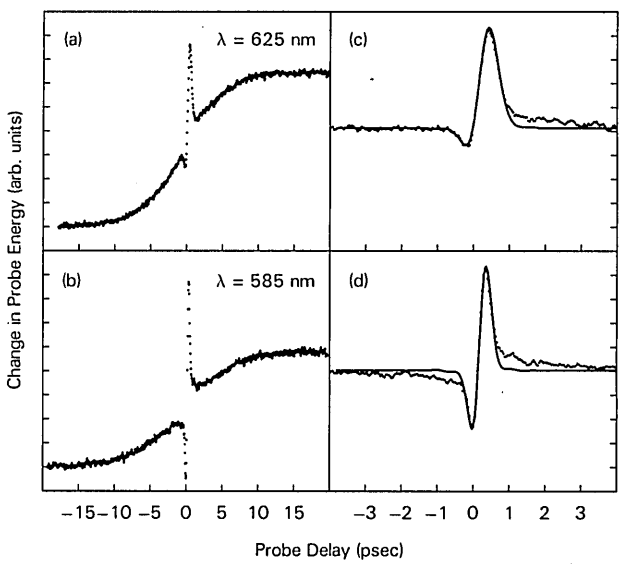

Fig. 2. Same as Fig. 1, but with pulses spectrally broadened in an optical fiber.

greater strength compared with that of the amplitude grating. The sample used in our experiments was a thin (200-μm) jet of Nile blue dye dissolved in ethylene glycol. The nonlinear interaction arose from an induced bleaching in the $S_0 \rightarrow S_1$ transition. For simplicity of interpretation, the dye concentration was held sufficiently low so that the jet was optically thin. The laser excitation was produced by a cavity-dumped, synchronously pumped mode-locked dye laser that yielded 6-psec pulses at a 4-MHz repetition rate. The pump and probe were obtained by splitting the output in two and passing one of the beams through a variable delay line. For the measurements presented here, the pump and probe were polarized parallel to each other and the two beams were made to cross at an angle of about 10°. The spot size of the beams on the sample was approximately 25 μm, which for our excitation (~10 nJ/pulse) only gave rise to a weak bleaching. To ensure further that these data were obtained in a regime where an analysis in terms of four-wave mixing is valid, we checked that the probe signal was proportional to the product of the pump and probe intensities, as predicted by Eq. (4).

We have performed these measurements both with nearly transform-limited pulses and with highly non-transform-limited pulses so as to illustrate the behavior of the coherent interaction. In Figs. 1(a) and 1(b) we display data for the probe signal as a function of delay time τ of the probe that were obtained with the pulses coming directly from the laser. Measurement of the spectrum of these pulses indicated that their time–bandwidth product was about a factor of 2 larger than the transform-limited value for a sech^2 pulse. Figure 1(a), taken with the laser tuned to 625 nm, shows the response near the peak of the dye absorption at 645 nm; Fig. 1(b) corresponds to excitation at 585 nm, on the high-frequency wing of the absorption. The analogous measurements obtained with pulses having a reduced coherence length are given in Figs. 2(a) and 2(b). These strongly chirped pulses were produced by passing the output of the mode-locked dye laser through a single-mode optical fiber. The resulting self-phase modulation led to roughly a factor of 10 increase in the pulse bandwidth. The group-velocity dispersion in the fiber gave rise to a mild concomitant broadening of the pulse envelope to about 10 psec FWHM.

4. DISCUSSION

The importance of the pulse coherence properties on the probe signal at short delay times is seen clearly by comparing Figs. 1 and 2. Since the data in Fig. 2 were taken with pulses having a greatly increased bandwidth but a similar intensity envelope, the dissimilarity of the two figures must be attributed almost entirely to the coherent coupling terms. The qualitative features of these data can be explained by reference to Eq. (4). For the Nile blue solution under study, all the relaxation processes are either extremely fast (vibrational) or slow (electronic, rotational) so that $A'_{xxxx}(t)$ and $A''_{xxxx}(t)$ can be approximated by step functions. In this limit, the incoherent contribution $\gamma(\tau)$ is given by the integral of the pulse intensity autocorrelation function. The symmetric part of the coherent signal, $\beta_1(\tau)$, varies in this case like the square of the electric-field autocorrelation.

We have isolated the coherent contribution to the data by subtracting the incoherent signal calculated from the experimentally determined intensity autocorrelation. The results are indicated in Figs. 1(c), 1(d), 2(c), and 2(d). In agreement with the theory, we observe that with the nearly transform-limited pulses the width of the coherent signal is approximately the same as the pulse duration, and for the spectrally broadened pulses of Fig. 2 its reduced width is comparable with the inverse spectral width of 0.8 psec. The antisymmetric contribution to the coherent signal in these data is apparent from the figure. We see not only that the relative size of the asymmetry depends on the wavelength of the pulse but additionally that its sign is different for the two types of pulses.

The effect of the wavelength on the relative contributions of the phase and amplitude gratings can be understood by considering how the absorption and emission spectra of the dye determine the real and imaginary parts of the nonlinear susceptibility, $\chi^{(3)}$. As an approximation to the Nile blue

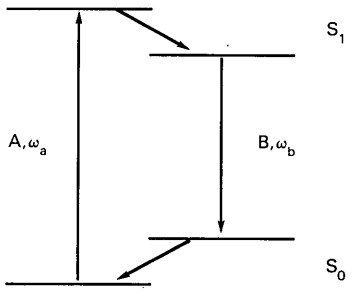

Fig. 3. Four-level model of the Nile blue energy-level structure.

energy-level structure, we introduce the four-level system shown in Fig. 3. In this model there is a transition at frequency ω_a (determined from the absorption spectrum) from the bottom of S_0 to the middle of S_1 and a second transition at ω_b (determined by the emission spectrum) from the bottom of S_1 to the middle of S_0. We assume that the nonradiative intraband relaxations are instantaneous on the time scale of the pulse, so that only these transitions contribute to the resonant part of the nonlinearity. The nonlinear susceptibility is then determined by the influence of the population loss from the bottom of S_0 on the linear susceptibility associated with the transition at ω_a and the influence of the population at the bottom of S_1 on the linear susceptibility associated with the transition at ω_b. We find that the resonant behavior is described by

$$\chi^{(3)} \sim -\left(\frac{A}{\omega-\omega_a+i\Gamma_a}+\frac{B}{\omega-\omega_b+i\Gamma_b}\right)\Delta N, \qquad (5)$$

where A and B are the oscillator strengths for the two transitions, Γ_a and Γ_b are the transition half-widths, and ΔN is the decrease in population of the ground state. If we assume that the strengths and widths of the two transitions are equal, we infer from Eq. (5) that the ratio of the real and imaginary parts of $\chi^{(3)}$ is equal to the ratio of the detuning from the average of the two resonance frequencies to the half-width of the lines. For the case of Nile blue, the absorption maximum lies at ~645 nm and the emission line is Stokes shifted by about 45 nm. Taking the linewidths to be about 70 nm, we then predict that at 625 and 585 nm this ratio will be −1.2 and −3.4, respectively. According to this model, we expect that the coherent term associated with the phase grating becomes arbitrarily large with respect to both the amplitude grating and incoherent terms as we travel further and further off resonance. It should be noted, however, that the coupling mechanism discussed here requires a change in population and, hence, will become weak off resonance. At some point, the contribution from the instantaneous response of the nonlinear susceptibility would also have to be considered. We also find that the point at which the phase grating disappears should be shifted significantly to the red of the center of the absorption line.

To verify that the asymmetric curves of Figs. 1 and 2 can be explained by the effects of the phase grating, we have carried out a numerical calculation of the coherent signal for some model pulse envelopes. Since we were interested only in showing a qualitative agreement between the theory and experiment, we assumed a sech^2 form for the pulse-intensity envelope with a duration determined by the FWHM of the measured intensity autocorrelation. The pulse phase modulation was represented by a linear frequency sweep, which should approximate the chirp of the pulse broadened in the fiber.[9] The complex field envelope of the pulses coming directly from the laser, which had a bandwidth of ~3 cm^{-1}, was not known. Using an input pulse with the appropriate duration and spectrum, and the response functions $A'_{xxxx}(t)$ and $A''_{xxxx}(t)$ equal to the unit step function, we integrated Eqs. (4b) and (4c) numerically. The data were fit by fine tuning the spectral width of the pulse to match the temporal width of the coherent signal and then adjusting normalization factors on A' and A'' to match the asymmetry in the data.

The predictions of Eq. (4), indicated for the spectrally broadened pulses in Figs. 2(c) and 2(d), show good agreement with the experimental data considering the simplified form assumed for the laser pulse. To fit the data at 585 nm we used a pulse having a positive frequency sweep with a bandwidth of 52 cm^{-1} and a ratio of A' to A'' of −4.2. The best agreement for the data at 625 nm was obtained with a 32-cm^{-1} bandwidth and a ratio of A' to A'' of −1.3. These ratios agree remarkably well with those predicted by the four-level model given above. The difference in the spectral widths needed for the two fits can be ascribed to the lower laser power at the longer wavelengths, which resulted in less spectral broadening in the fiber; both of the assumed values agree well with the estimated pulse bandwidths. For the case of the nearly transform-limited pulses coming directly from the laser, we have fitted the data in Figs. 1(c) and 1(d) by taking pulses with a 3.5-cm^{-1} bandwidth (a factor of 2 from the transform limit). The opposite sign of the asymmetry compared to Fig. 2 is accounted for by using a negative frequency sweep. At the 585-nm wavelength the best agreement required a ratio of A' to A'' of −2.6; at 625 nm the ratio was −0.85. Given the crude nature of the model of the pulse envelopes, the values of A' and A'' found here agree quite well with those determined above. We conclude from these simple calculations that the observed asymmetries in the coherent signal are well accounted for by the effects of the phase grating. Furthermore, these calculations demonstrate quite clearly that these effects can be as large as or larger than the amplitude-grating effects, even when the pulses are close to transform limited.

5. CONCLUDING REMARKS

In this paper, we have explicitly demonstrated the influence of phase gratings in transient absorption measurements of dye molecules. Our result should, however, be applicable to pump–probe measurements in other material systems. We expect a coherent contribution from the phase grating to exist whenever a phase grating can be induced in the sample and the laser pulses display some degree of phase modulation. Although the response functions may take somewhat different forms, this conclusion holds as well for different experimental geometries, such as with perpendicularly polarized pump and probe or with collinear, copropagating beams. One case, however, where the phase-grating contribution cannot exist is in the so-called equal-pulse-correlation technique.[10] In this method the energy in both the pump and probe beams is measured, and, consequently, energy transfer between the two is not be detected.

While the measurements that we have presented here were obtained with picosecond pulses, the general analysis applies as well to experiments in the femtosecond regime, provided that the dephasing rates in the sample are rapid compared

with the pulse duration. Since the phase and amplitude gratings arise from the nonlinear response of the system within the pulse envelope, their contributions may depend on the experimental time scale. In the simple four-level model introduced above, it is apparent that for measurements with pulses shorter than the intraband relaxation time, the transition occurring at the lower energy (denoted by B in Fig. 3) does not contribute to the third-order susceptibility. Therefore, assuming that only the two transitions A and B are present, the phase grating should vanish. For the case of dyes, of course, transitions between other levels within the two bands can contribute to the nonlinear susceptibility, so that a phase grating would still be induced even for short laser pulses. For measurements of the relaxation of photoexcited carriers in semiconductors, the same four-level model might still be applied, but since the crystal momentum must be conserved, only the two transitions shown would be allowed. Hence, for time scales faster than any intraband relaxation, only amplitude gratings should be significant. If, on the other hand, the pulses were long compared to the time in which the electrons and holes relax outside the bandwidth of the excitation, the coherent signal would result primarily from phase-grating effects.

Another point of interest is how our results might be applied to optical Kerr measurements. Since time-resolved studies of the Kerr effect involve the real part of the nonlinear susceptibility, the analysis of the coherent interaction in these experiments has treated only phase gratings.[11] The generalization of this work to Kerr measurements would therefore be the inclusion of an amplitude grating. In particular, for heterodyning experiments in which the nonlinear polarization is mixed with a phase-shifted probe, any contribution from an amplitude grating would be antisymmetric. However, since Kerr media are frequently transparent, such amplitude effects might be expected to be small.

In conclusion, we have demonstrated that in pump–probe measurements of transient absorption, both amplitude gratings and phase gratings can contribute to the coherent interaction between the pump and probe pulses. The results discussed here, which bear on a wide variety of pump–probe measurements, further complicate the problem of extracting material relaxation times comparable with the pulse duration.

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5. Rigorously speaking, as is clear below, phase and amplitude gratings are associated with variations in the real and imaginary parts of the susceptibility.
6. It should be mentioned that contributions to the coherent signal from a phase grating have been noted previously [C. V. Shank and D. H. Auston, "Parametric coupling in an optically excited plasma in Ge," Phys. Rev. Lett. **34,** 479 (1975)]. The effect that they discuss, however, depends quadratically on the induced nonlinear polarization. This is a higher-order contribution than the interference between the nonlinear polarization and the probe considered here.
7. We neglect in this discussion the off-resonant contribution to the nonlinear susceptibility. Since this term is real and corresponds to an instantaneous response, it will not affect the signal considered here.
8. This analysis can be extended in a straightforward fashion to the case of optically thick samples by integrating the wave equation. See, for example, the treatment in Ref. 3.
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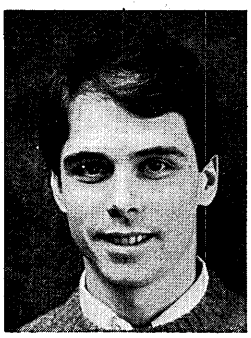
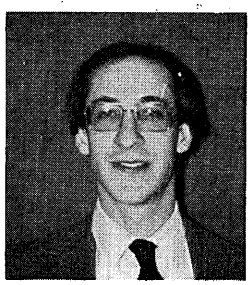
S. L. Palfrey



S. L. Palfrey was born in Lafayette, Indiana, on May 10, 1956. He received the B.A. degree in physics from Oberlin College, Oberlin, Ohio, in 1979 and the A.M. and Ph.D. degrees in physics from Harvard University, Cambridge, Massachusetts, in 1980 and 1984, respectively. Since 1983 he has been a visiting scientist at the IBM Thomas J. Watson Research Center, Yorktown Heights, New York. He is currently working on the development of ultrashort laser sources and the application of these to study atomic, molecular, and condensed-matter systems. Dr. Palfrey is a member of the Optical Society of America, the American Physical Society, and Sigma Xi.

T. F. Heinz



T. F. Heinz was born in Palo Alto, California, on April 30, 1956. He received the B.S. degree in physics from Stanford University in 1978 and was a National Science Foundation and IBM graduate fellow at the University of California, Berkeley, where he received the Ph.D. degree in physics in 1982. In 1983, he joined the staff of the IBM T. J. Watson Research Center, Yorktown Heights, New York, and has been involved in the application of laser techniques for the study of surfaces and dynamical processes.