Investigation of Retroreflection Scheme for Optical Phase Conjugation by Degenerate Four-Wave Mixing

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***Abstract*—We present theoretical and experimental data which show that the value of the phase conjugate reflectivity by degenerate four-wave mixing depends on whether the retroreflection or counterpropagating schemes are used. It is also shown that the retroreflection method may result in decay constants which are not related to the decay of the gratings formed in the nonlinear medium.**

I. Introduction

FOR our studies in optical phase conjugation by degenerate four-wave mixing, we have utilized two different experimental geometries [1]–[3]. The first makes use of two separate counterpropagating (CP) pump beams, intersecting with a probe beam in a nonlinear optical medium to produce four-wave mixing. In the second scheme, often used because of its simplicity [4]–[7], the second pump beam is the back reflection of the forward pump beam by a mirror located close to the nonlinear medium. We designate this the retroreflection (RR) scheme. When the retroreflecting mirror is located close to the medium, the probe beam will be reflected back into the region irradiated by the two pump beams, causing additional interaction as compared to the CP scheme. This second interaction region has a considerable influence on the magnitude as well as the decay of the phase conjugate reflectivity.

It has been shown [1] that phase conjugation reflectivity obtained by the RR scheme is increased by a factor 4β (where β is the single-pass transmission of the nonlinear medium), compared to the CP scheme assuming that the laser power used in both schemes is the same, and the two pump beams in the CP scheme have equal intensities. This is true only when the reflected probe beam does not intersect the pump beams again.

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Here we show that under the same assumption, but when the reflected probe beam interacts again with the pump beams, this factor can be increased up to $4\beta(1 + \beta)^2$. It is important to note that if one does not account for this fact, it may lead to wrong values for $\chi^{(3)}$.

In addition to this, we show that the "time constants" of the decay of the phase conjugate reflectivity, measured as a function of the delay of the second pump beam, are smaller in the RR scheme than the equivalent decay times obtained by the CP method. In the RR scheme, this decay curve is measured by varying the distance between the nonlinear medium and the retroreflection mirror [6], [7]. We show that the decays obtained by this method may not be the true decay of the induced gratings or width of the pump beam autocorrelation function, but may be inherent to the experimental system and bear no relation to the phase conjugate reflectivity decay. Finally, experimental data are shown for the dependence of the width of the phase conjugated pulse on the distance between the nonlinear medium and retroreflector in the RR scheme. These considerations are relevant to picosecond phase conjugation because the necessity for temporal overlap of the pump pulses implies a short distance between the mixing medium and the retroreflecting mirror.

The basic pulse interaction for the RR system is shown in Fig. 1(a). The equivalent scheme, presented in Fig. 1(b), provides a useful picture of what happens when the probe beam is reflected into the interaction region. When the probe beam is reflected by the mirror, there will be two regions, *ABCD* and *AMNQ*, where gratings will be formed and a phase conjugated signal created. The origin of the different diffraction gratings and the direction of the resulting phase conjugated beams are summarized in Table I. After reflection from the mirror, the conjugate signals originating in the *AMNQ* region propagate in the same direction as signals from the *ABCD* region. The constructive interference of these fields leads to an increase in phase conjugation efficiency.

There are two factors which influence this process. The first factor is due to the decrease in overlapping volume while increasing the distance between the retroreflecting mirror and the nonlinear medium. Examination of Fig. 1 makes it obvious that the size of the *AMNQ* region de-

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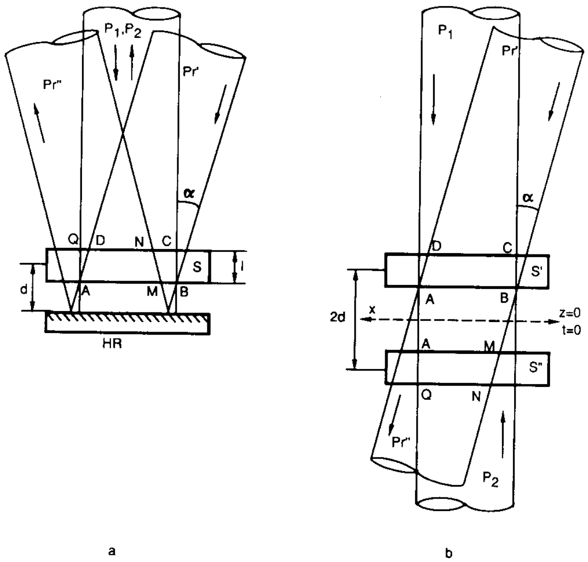

Fig. 1. Basic four-wave mixing geometry using retroreflection of the forward pump (a) and its equivalent scheme used in the theoretical calculations (b). *HR*: high reflector; *S*: nonlinear mixing medium; P_1, P_2: forward and backward pump beams; P_r', P_r'' probe beam before and after reflection on HR.

TABLE I
ORIGIN AND DIRECTION OF PHASE CONJUGATED SIGNALS IN RR SCHEME FOR THE CASE THAT THE PROBE BEAM IS REFLECTED BACK INTO THE PUMP REGION OF THE NONLINEAR MEDIUM

Region	Type of Grating	Written by	Readout by	Direction
ABCD	Coarse	P1, Ppr'	P2	Away from
	Fine	P2,Ppr'	P1	the mirror
AMNQ	Coarse	P2, Ppr''	P1	Towards the
	Fine	P1, Ppr''	P2	mirror

pends on the angle between the pump and the probe beam and on the distance between the sample and the mirror. When the mirror is at the nearest position, the output reflected signal will be $(1 + \beta)^2$ times more intense than the reflectivity from the *ABCD* region alone. As the distance increases, the phase conjugated signal decreases, even in cases where the grating in the medium is long lived. The distance and the respective time delay which is necessary for the interference to occur is defined as the overlapping time delay τ_{0v} which is equal to $w_0/c \tan \alpha$ where w_0 is the radius of the beam and α is the angle between forward pump and probe beam. It can be seen that when the pulse length or decay time is much longer than τ_{0v}, some type of decay curve will be observed which may not be due to the grating lifetime or correlated with optical phase conjugation mechanisms.

The second factor depends upon the temporal overlap of the phase conjugated pulses generated in both regions. The characteristic delay at which the two pulses will not interfere is approximately equal to the geometrical length of the laser pulses $c\tau_p$ (τ_p being the FWHM of the pulse). The first factor is influential only when the diameter of the beams is small, namely, $w_0 < \tau_p c \tan \alpha$, while the second factor depends only on the duration of the pulses.

II. THEORETICAL MODEL

We developed a theoretical model based on the geometrical scheme shown in Fig. 1(b) which is equivalent to the actual experimental scheme we used [Fig. 1(a)]. This scheme consists of two samples situated at distance $2d$. We assume that the angle α is small enough ($\alpha < l/w_0$) in order to consider overlapped areas which have apparent rectangular shape. We also assume the small-signal approximation so we can neglect nonlinear effects and pump depletion. In addition to this, we only consider the case where the width of the laser pulses is much larger than the sample thickness ($l < \tau_p c$).

Consider the next form of the electric field of the interacting beams:

$$E_1 = A_1 \Phi_1(t + z/c)\, F_1(r) \cos(\omega t - k_1 z) \tag{1a}$$

$$E_2 = A_2 \Phi_2(t - z/c)\, F_2(r) \cos(\omega t - k_2 z) \tag{1b}$$

$$E_{pr} = A_{pr} \Phi_{pr}(t + z/c)\, F_{pr}(r) \cos(\omega t - k_{pr} z) \tag{1c}$$

where A_i, Φ_i, and F_i are, respectively, the electric field amplitude, the temporal profile, and the spatial transverse profile of the beam i, the index i being 1 and 2 for the forward and backward pump beams and pr for the probe beam. If the pump and probe pulses all originate from the same source, all factors Φ_i and F_i are equal to each other: $\Phi_1 = \Phi_2 = \Phi_{pr} = \Phi$ and $F_1 = F_2 = F_{pr} = F$. As usual in degenerate four-wave mixing, $k_1 = -k_2$ and $k_{pr} = k_1 \cos \alpha$.

Many nonlinear media (e.g., semiconductor glasses, polydiacetylenes) indicate the simultaneous presence of two components in the time decay of $\chi^{(3)}$, a fast component in the picosecond time domain, and a much slower one in the nanosecond region [4], [7], [10]–[12].

For semiconductor glasses (OG 530 type was used in our experiment), the relative contribution of the fast and slow components of $\chi^{(3)}$ depends on the energy density of the pump pulses [11]. At high density, the fast component is predominant; at low density, the slow component is. We will consider two opposite cases: 1) a very fast response of the nonlinear medium, much faster than the pulse length, and 2) the case when the medium response is slower than the time between laser pulses.

In the fast response case, the amplitude shape of the conjugated pulse $\mathcal{E}'$ in the *ABCD* region can be found by following the standard procedures [1]:

$$\mathcal{E}'(t, r) = Q_{RR} \gamma \Phi^2(t + d/c)\, \Phi(t - d/c)\, F^3(r) \tag{2}$$

where $Q_{RR} = A_1^2 A_{pr} \beta(1 - \beta)/\ln \beta$ [14] and γ is the nonlinear coefficient $\gamma = (\pi\omega/cn)\chi^{(3)}$. In fact, $\gamma = \gamma_c + \gamma_f$ where γ_c and γ_f are the nonlinear contributions due to coarse and fine gratings, respectively. The signal from the second interaction volume $AMNQ$ has a different form:

$$\mathcal{E}''(t, r) = \beta Q_{\mathrm{RR}} \gamma \Phi^2(t - d/c)\, \Phi(t + d/c) \cdot F^2(r)\, F(r + s) \tag{3}$$

where s is the displacement of the reflected probe beam center from the center of the pump beams in the plane of the sample, $s = 2d \tan \alpha$.

The intensity of the conjugated signal will depend upon the constructive interference of both conjugated waves:

$$\mathcal{I} \propto \left[\mathcal{E}'(t + d/c, r) + \mathcal{E}''(t - d/c, r)\right]^2. \tag{4}$$

This expression allows us to calculate the change of conjugated pulse shape and maximum intensity versus distance between the sample and the RR mirror. Integrating over $t, x, y,$ $(r = \sqrt{x^2 + y^2})$, we can get the energy of the conjugated signal. Let us assume a Gaussian shape of both the temporal and spatial profile of the laser pulses:

$$\Phi(t) = \exp(-t^2/T^2)$$

where T is the half width at $1/e$ amplitude ($T = \tau_p/\sqrt{2 \ln 2}$) and

$$F(r) = \exp(-r^2/w_0^2)$$

where w_0 is the beam radius at $1/e$ amplitude. Integration of (4) leads to

$$W_{\mathrm{RR}}(\tau) \propto (Q_{\mathrm{RR}}\gamma)^2 \exp\left(-\frac{4\tau^2}{3T^2}\right)\left[1 + \beta^2 \exp\left(-\frac{4\tau^2}{3\tau_{\mathrm{ov}}^2}\right) + 2\beta \exp\left(-\frac{8\tau^2}{3T^2}\right) \exp\left(-\frac{5\tau^2}{6\tau_{\mathrm{ov}}^2}\right)\right] \tag{5}$$

where τ is the time delay of the second pump beam, $\tau = 2d/c$, and $\tau_{\mathrm{ov}} = w_0/c \tan \alpha$. It is interesting to compare (5) to the corresponding expression for the counterpropagating scheme:

$$W_{\mathrm{CP}}(\tau) \propto (Q_{\mathrm{CP}}\gamma)^2 \exp\left(-\frac{4\tau^2}{3T^2}\right) \tag{6}$$

where $Q_{\mathrm{CP}} = A_1 A_2 A_{pr} \sqrt{\beta}(1 - \beta)/\ln \beta$. We see that decay curve for RR scheme is the sum of three exponential terms. Fig. 2 shows some of the decay curves obtained for different ratios of τ_{ov} and T. For comparison, the decay curve obtained by the use of the CP scheme is shown too (lower curve). One sees that due to the interference terms, the reflectivity using the RR scheme apparently decays faster than using the CP scheme. The ratio of maximum reflectivities for both schemes at zero delay ($\tau = 0$) will be

$$W_{\mathrm{RR}}(0)/W_{\mathrm{CP}}(0) = (Q_{\mathrm{RR}}/Q_{\mathrm{CP}})^2 (1 + \beta)^2. \tag{7}$$

If, as stated previously, the laser power is the same in both schemes and the two pump beam intensities are equal when using the CP method, then $(Q_{\mathrm{RR}}/Q_{\mathrm{CP}})^2 = 4\beta$ and the ratio of reflectivity of the RR and CP scheme is $4\beta(1 + \beta)^2$.

In the second case where we consider only the slow component of $\chi^{(3)}$, the intensity of phase conjugate re-

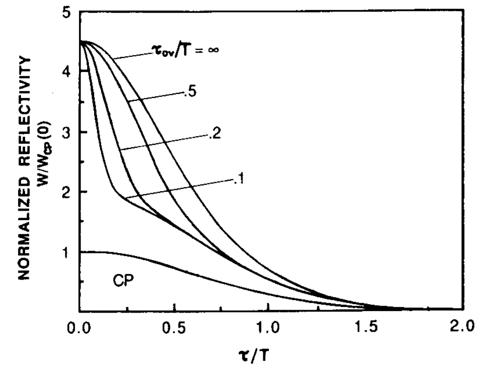

Fig. 2. Calculated phase conjugated reflectivity as a function of the delay of the backward pump pulse. All curves are normalized to the reflectivity in the CP scheme at zero time delay, i.e., $W_{\mathrm{CP}}(0)$. Curves in the RR scheme are shown for different values of τ_{ov}/T where T is the bandwidth at $1/e$ amplitude, according to (5). The nonlinear susceptibility $\chi^{(3)}$ is assumed to be instantaneous and the sample transmission was taken as 50 percent. The curve for the CP scheme is shown for comparison.

flectivity will be a function of four terms connected with the gratings listed in Table I:

$$\mathcal{I} \propto \left[\mathcal{E}_c'(t, r) + \mathcal{E}_f'(t + d/c, r) + \mathcal{E}_c''(t - d/c, r) + \mathcal{E}_f''(t, r)\right]^2 \tag{4a}$$

where f and c correspond to fine and coarse grating. Prime (′) and double prime (″) correspond to the $ABCD$ and $AMNQ$ volume.

$$\mathcal{E}_c'(t, r) = Q_{\mathrm{RR}} \gamma_c \exp\left(-\frac{t^2}{T^2} - 3\frac{r^2}{w_0^2}\right),$$

$$\mathcal{E}_f'(t + d/c, r) = Q_{\mathrm{RR}} \gamma_f \exp\left[-\frac{\tau^2}{T^2} - \frac{(t + d/c)^2}{T^2} - 3\frac{r^2}{w_0^2}\right],$$

$$\mathcal{E}_c''(t - d/c, r) = Q_{\mathrm{RR}} \gamma_c \exp\left[-\frac{(t - d/c)^2}{T^2} - 2\frac{r^2}{w_0^2} - \frac{(r - s)^2}{w_0^2}\right],$$

$$\mathcal{E}_f''(t, r) = Q_{\mathrm{RR}} \gamma_f \exp\left[-\frac{t^2 + \tau^2}{T^2} - 2\frac{r^2}{w_0^2} - \frac{(r - s)^2}{w_0^2}\right].$$

The change of the phase-conjugated pulse energy W_{RR} as a function of second pulse delay is obtained by integration of (4a):

$$W_{\mathrm{RR}} \propto Q_{\mathrm{RR}}^2 \left\{\gamma_c^2 + \gamma_f^2 \exp\left(-2\frac{\tau^2}{T^2}\right)\left[1 + \beta^2 \cdot \exp\left(-\frac{4\tau^2}{3\tau_{\mathrm{ov}}^2}\right) + 2\beta \exp\left(-\frac{\tau^2}{8T^2} - \frac{5\tau^2}{6\tau_{\mathrm{ov}}^2}\right)\right]\right.$$

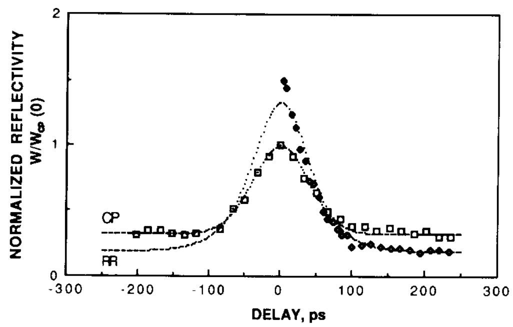

Fig. 3. Theoretical and experimental dependance of the decay of the phase conjugate reflection as a function of the delay of the backward pump pulse in the CP scheme and the RR scheme. Dotted lines represent the calculated curves, normalized to the $W_{CP}(0)$ point; (□) experimental results for CP scheme and (◇) experimental results for RR scheme.

$$+ 2\gamma_c\gamma_f \exp\left(-\frac{9\tau^2}{8T^2}\right)\left\{1 + \beta^2 \exp\left(-\frac{4\tau^2}{3\tau_{ov}^2}\right) + \beta \exp\left(-\frac{5\tau^2}{6\tau_{ov}^2}\right)\left[\exp\left(\frac{\tau^2}{8T^2}\right) + \exp\left(-\frac{3\tau^2}{8T^2}\right)\right]\right\}\Bigg\}. \quad (8)$$

$W_{RR}(\tau)$ and $W_{CP}(\tau)$ normalized to $W_{CP}(0)$ are shown in Fig. 3 when the ratio γ_c/γ_f equals 1.3. This figure shows that a fictitious fast component is present even though we assumed in our calculations to have only a slow component. The contrast ratio between the signal at zero delay and that at delays much longer than τ_{ov} and τ_p depends on the ratio γ_c/γ_f:

$$W_{CP}(0)/W_{CP}(\infty) = (\gamma_f + \gamma_c)^2/\gamma_c^2 \quad (9)$$

$$W_{RR}(0)/W_{RR}(\infty) = (1 + \beta)^2(\gamma_f + \gamma_c)^2/\gamma_c^2. \quad (10)$$

The ratio between reflectivities for both schemes at zero delay is again defined by (7).

III. Experimental

The experimental system used to obtain the data presented in this paper is shown in Fig. 4. A CW mode-locked Nd/YAG laser (Spectra-Physics, model 3000) emits 1.06 μm, 100 ps laser pulses at a repetition rate of 82 MHz. These pulses were frequency doubled by a KTP crystal and separated from the fundamental beam by a harmonic separator (HS). An average green power of 600 mW was obtained, equivalent to an energy of 7 nJ per pulse. The duration of the pulses was determined by a Hamamatsu synchroscan streak camera (M1995, connected to an EG&G OMA system) and found to be 75 ± 5 ps. In addition to this, the coherence length of the pulses was determined by Michelson interferometry [13]. The coherence function of the SH pulses is shown in Fig. 5. Assuming a Gaussian temporal pulse profile, this coherence time corresponds to a transform-limited pulse duration of 64.5 ps [8]. We conclude from these data that our pulses are nearly transform limited and have a Gaussian

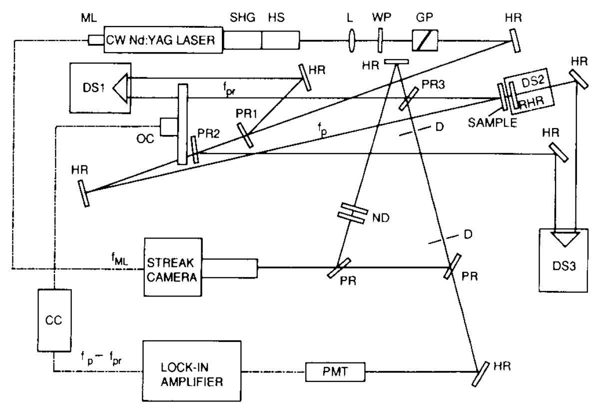

Fig. 4. Experimental setup used for the investigation of phase conjugation by degenerate four-wave mixing using the counterpropagating (CP) and the retroreflection (RR) scheme. *ML*: mode locker (82 MHz); *SHG*: second harmonic; *HS*: harmonic separator; *WP*: half-wave plate; *GP*: glan prism; *HR*: high reflectors (532 nm); *PR*: partially reflecting mirrors; *RHR*: removable highly reflecting mirror; *DS*: delay stages; *OC*: differential optical chopper; *CC*: chopper controller; *PMT*: photomultiplier; *ND*: neutral density filters; *D*: diaphragms.

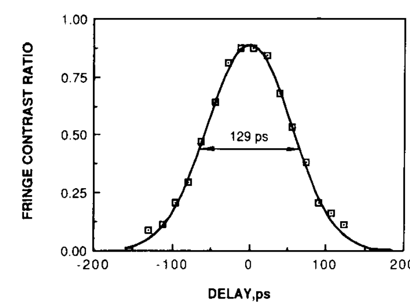

Fig. 5. Coherence function depicted as a plot of the fringe contrast ratio versus delay between picosecond pulses (532 nm) as measured by a Michelson interferometer. The points are the experimental data, and the solid curve represents a Gaussian function having an FWHM of 129 ps.

profile. A Gaussian pulse shape is therefore adopted in constructing our theoretical model and calculations.

The laser beam is collimated by a lens *L* (2 m focal length). The power could be adjusted by a variable attenuator, which consisted of a half-wave plate (WP) and Glan prism polarizer (GP). Our experiment is designed in such a way that we can switch from the RR scheme to the CP scheme simply by removing the retroreflecting mirror RHR behind the nonlinear mixing medium. Therefore, this arrangement allowed us to make a very accurate comparison of the two methods for producing OPC by DFWM. In our experiments, a semiconductor colloid glass filter (Schott 0G 530) was used as phase conjugating medium. The sample thickness was 3 mm; the transmission was 56 percent. The probe beam was created by beamsplitter *PR*1 ($R = 0.16$) and intersects the forward pump beam at an angle of 2.5°. Using this angle and the radius of the beams, we calculated the τ_{ov} for the RR scheme to be 90 ps. *PR*2 ($R = 0.46$) is used to create the second pump beam for the CP scheme. The polarizations of all the beams are parallel. The timing of the forward pump beam and the probe were adjusted by means of delay stage *DS*1. The time dependence of the phase conjugated reflectivity was probed by scanning the delay stage *DS*2 in the RR scheme and delay stage *DS*3 in the CP scheme. The phase conjugate signal was separated from the probe beam by a beamsplitter *PR*3 ($R = 0.3$). The energy of the phase conjugated pulses was detected by a photomultiplier and measured by a lock-in amplifier (EG&G, model 5209), which was referenced to the difference of the modulation frequencies of the pump and the probe beams. A single wheel mechanical chopper (Stanford Research SR540) allows chopping the two beams at two different frequencies. This ensures a constant difference frequency, which in our experiments was 333 Hz. This detection scheme allowed for an almost complete rejection of stray light and a virtually background-free measurement of the phase conjugated signal. Part of the phase conjugated beam was split off by beamsplitter *PR*1 and directed towards the synchroscan streak camera. The signals from the streak camera were digitized by an optical multichannel analyzer and fed to a DEC LAB-23 minicomputer. The resulting pulse shapes were deconvoluted for the response curve of the streak camera.

IV. Results and Discussion

Fig. 3 shows experimental results obtained both with the CP scheme and the RR scheme using a semiconductor colloid glass as a nonlinear medium. It is worthwhile to compare the ratio of experimental reflectivities obtained at zero delay and maximum delay for the two schemes to the theoretical prediction.

1) Zero Delay: Taking into account that $\beta = 0.5$ and that for our experiments $(Q_{RR}/Q_{CP})^2 = 0.59$, we calculate [see (7)]

$$W_{RR}(0)/W_{CP}(0)\big|_{th} = 1.33.$$

The experimental ratio is

$$W_{RR}(0)/W_{CP}(0)\big|_{exp} = 1.5 \pm 0.2.$$

This value is very close to the calculated increase in reflectivity. Therefore, in the optimal case where the same laser power is used for both schemes and the two pump beams in the CP scheme have equal intensity, the reflectivity obtained with the RR scheme will be $(Q_{RR}/Q_{CP})^2(1 + \beta)^2 = 4.5$ more then the reflectivity with the CP scheme.

2) Maximum Delay Equals About 260 ps: This delay ensures that the reflected probe beam does not intersect the overlap region of the pump beams anymore. At this delay, the two schemes are equivalent. Since there is no interference in the RR scheme, then the ratio of reflectivities should be

$$W_{RR}/W_{CP} = (Q_{RR}/Q_{CP})^2 = 0.59.$$

This value can be compared to the experimental ratio which was found to be equal to 0.62 ± 0.1.

The reflectivity with the RR scheme is 2.5 more than it should be when the probe beam is not reflected from the mirror. This value is very close to the calculated increase of reflectivity, 2.25. So we can expect that if product $P_1 P_2$ is the same for both schemes, the reflectivity with the RR scheme will be $4\beta(1 + \beta)^2$ more than the reflectivity with CP scheme.

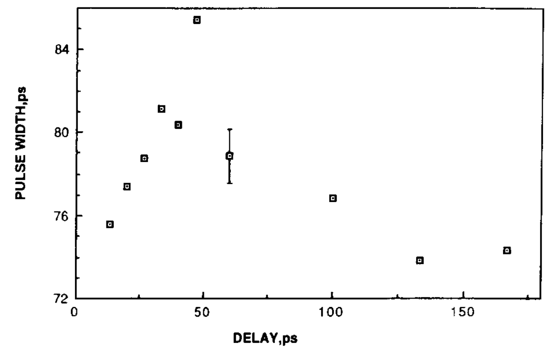

Fig. 6. Change of phase conjugate pulse width as a function of delay for the RR scheme.

From the ratio of the reflectivity at zero and maximum delay, (9), for the CP scheme we calculated the ratio between the contribution of coarse and fine grating in our sample to be $\gamma_c/\gamma_f = 1.3$. Using this value, we were able to calculate the theoretical curves presented by a dotted line in Fig. 3. As it is seen in this figure, there is a good agreement between the predicted value based on our model and the experimental data. The background level at negative delays is due to the fact that the true decay times of the gratings are much longer than the time period between the pulses, and every pulse sees the grating created by previous pulses.

We have also studied the change for the conjugate signals with increasing delay. The experimental results obtained for these changes as a function of delay are shown in Fig. 6. Every experimental point on the plot is an average value of four pulse duration measurements with integration time 0.025 s. At a delay of ~50 ps, the width of the phase conjugated pulse reaches a maximum, and when the conjugated pulse length reaches the laser pulse length (75 ps). The fact that at zero delay the length of the phase conjugated pulse is approximately equal to the pump pulse length indicates that the slow component of $\chi^{(3)}$ decay is predominant in this semiconductor's colloids.

V. Summary and Conclusion

We compared the retroreflection and counterpropagating schemes for phase conjugation by degenerate four-wave mixing. We have shown that 1) the retroreflection scheme is more effective than counterpropagating scheme, and 2) measurement of decay constants using the retroreflection scheme may be difficult to interpret and extreme care should be exercised. In the case where the transverse radius of the pulses is small as compared to the geometrical length of the pulse, the apparent decay curves of the phase conjugate reflection will be shortened. 3) In the retroreflection scheme, the pulse length of the conjugated pulse changes with increasing delay between the retroreflector and sample.

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[14] The factor $\beta(1-\beta)/\ln\beta$ in the expression of Q_{RR} takes into account the influence of the linear absorption β of the nonlinear medium on the phase conjugation efficiency. See [1], pp. 152-153].

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