Measurement of $\chi^{(3)}$ and phase shift of nonlinear media by means of a phase-conjugate interferometer

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We have constructed a new interferometer that uses two phase-conjugate mirrors. This device allows for the measurement of the relative phase shift and the ratio of the nonlinear susceptibilities of materials. We have been able to measure weak reflectivity signals not detectable by normal degenerate four-wave mixing methods. It has been determined that the $\chi^{(3)}$ value of colloid semiconductor glass OG530 at 532 nm is 7×10^{-12} esu.

Recent advances in nonlinear-optical devices have made it possible to measure nonlinear properties of materials with high accuracy.[1] The study of nonlinear susceptibilities and the measurement of $\chi^{(3)}$ coefficients have become easier and more accurate by the use of interferometric methods.[2–4] These methods additionally make possible the measurement of the sign of $\chi^{(3)}$ components.[4–7]

In this Letter we describe the use of a modified Twyman–Green (MTG) interferometer, which includes two phase-conjugate mirrors. As will be seen, this interferometer makes possible the accurate measurement of the relative phase shift between the two phase conjugators and the ratio of their susceptibilities.

A schematic representation of the MTG interferometer is shown in Fig. 1. The phase-conjugate mirrors are based on degenerate four-wave mixing in a retroreflection scheme.[8,9] The pump and probe beams each are split into two beams by a splitter, BS, and follow the two paths shown in Fig. 1. After interacting at the phase-conjugate mirrors, PC1 and PC2, the retroreflected beams recombine at BS. The signal generated by the recombination of the pump beams constitutes the conventional Twyman–Green interferometer, which we designate pump interferometer PI. The phase-conjugate interferometer, CI, consists of the two phase-conjugate signals generated at the conjugate mirrors PC1 and PC2 that interfere with each other at BS. The interdependency of the two interferometers as seen in Fig. 1 is such that alignment of one interferometer, PI or CI, results in the automatic alignment of the other.

The best way to understand the data from the MTG interferometer is to introduce a phase shift α into one of the arms of the interferometer and analyze the interference pattern of PI and CI. The phase shift can be induced by moving one of the mirrors. The interference signal of the pump interferometer, I_{PI}, is

$$I_{\mathrm{PI}} = I_1 + I_2 - 2\sqrt{I_1 I_2}\cos\left[\frac{4\pi}{\lambda}(l_2 - l_1) + \alpha\right], \qquad (1)$$

where I_1 and I_2 are the intensities of the returning pump beams that follow the optical path lengths l_1 and l_2 of arms 1 and 2, respectively. The phases of the two conjugate waves, $\varphi_{c,1}$ and $\varphi_{c,2}$, are

$$\varphi_{c,1} = \varphi_{0,1} + \varphi_{f,1} + \varphi_{b,1} - \varphi_{\mathrm{pr}} - \frac{2\pi}{\lambda} d_1$$

and

$$\varphi_{c,2} = \varphi_{0,2} + \varphi_{f,2} + \varphi_{b,2} - \varphi_{\mathrm{pr}} - \frac{2\pi}{\lambda} d_2, \qquad (2)$$

where $\varphi_{0,1}$ and $\varphi_{0,2}$ are the absolute phase shifts of the phase conjugators, which depend on the type of diffraction grating created by pump and probe beams,[6] φ_f and φ_b are phases of the forward and backward pump beams, respectively, φ_{pr} is the input phase of the probe beam relative to the pump beam at the plane of the beam splitter, and d_1 and d_2 are the beam-splitter-to-conjugator optical distances. Subscripts 1 and 2 refer to the optical paths of arms 1 and 2, respectively. For each arm the following expressions hold:

$$\varphi_{b,1} + \varphi_{f,1} = \frac{4\pi}{\lambda} l_1, \qquad \varphi_{b,2} + \varphi_{f,2} = \frac{4\pi}{\lambda} l_2 + \alpha. \qquad (3)$$

At the plane of the beam splitter, the phases of the conjugate signals will be $\varphi_{c,1} + (2\pi/\lambda)d_1$ and $\varphi_{c,2} + (2\pi/\lambda)d_2$. The CI output will then be

$$I_{\mathrm{CI}} = I_{C,1} + I_{C,2} - 2\sqrt{I_{C,1} I_{C,2}} \cos\left[\frac{4\pi}{\lambda}(l_2 - l_1) + \alpha + \varphi_{0,2} - \varphi_{0,1}\right]. \qquad (4)$$

Comparison of Eqs. (1) and (4) shows that the interference signals at the two outputs are described by the same function of the induced phase shift α. The only difference is a phase shift $\varphi_{0,2} - \varphi_{0,1}$. This phase shift is equal to the difference of the absolute phase of the conjugate signals in the two nonlinear media. This means that, if the absolute phase shift in one of the two conjugate materials is known, then the absolute phase shift of the second medium can be determined easily.

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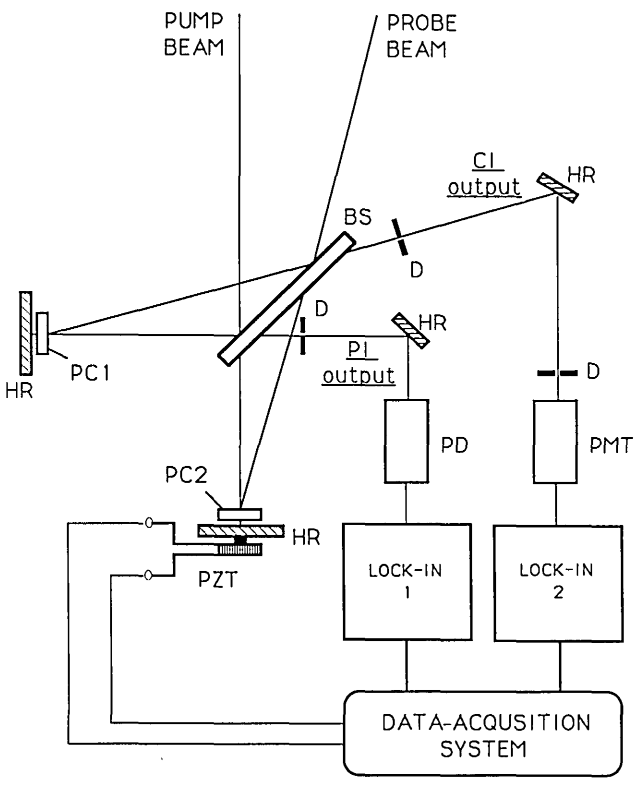

Fig. 1. Experimental system used for measurements of phase shift and $\chi^{(3)}$ ratio. HR's, highly reflecting mirrors; D's, diaphragms.

In the case when the same medium is used in both arms, $\varphi_{0,2} = \varphi_{0,1}$, the interference signals at both outputs should be exactly in phase.

The ratio between the maximum and the minimum of the interference pattern can be used to find the ratio of $\chi^{(3)}$ in both conjugate media. Let $A_{c,1}$ and $A_{c,2}$ be the conjugate amplitudes from the two conjugators. In the weak-reflectivity case, neglecting pump depletion for the quasi-collinear retroreflection geometry,[9] the conjugate amplitude $A_{c,i}$ is

$$A_{c,i} = -\gamma_i S_i G_i A_{p,i}{}^2 A_{\mathrm{pr},i}, \qquad i = 1, 2, \tag{5}$$

where G_i is the absorption factor, $G_i = \beta_i(1 - \beta_i{}^2)/|\ln \beta_i|$ (β_i is the single-pass transmission of the conjugate medium), γ_i is the nonlinear coupling coefficient, $\gamma_i = (\pi\omega/cn)\chi_i^{(3)}$, S_i is the thickness of the conjugate medium, and $A_{p,i}$ and $A_{\mathrm{pr},i}$ are the amplitudes of the pump and probe beams, respectively. The ratio M of maximum to minimum signals from the conjugate interferometer is

$$M = \frac{I_{\max}}{I_{\min}} = \left(\frac{1 + A_{c,2}/A_{c,1}}{1 - A_{c,2}/A_{c,1}}\right)^2. \tag{6}$$

Substituting $A_{c,i}$ from Eq. (5), we get

$$\chi_2^{(3)} = \chi_1^{(3)} \frac{\sqrt{M} - 1}{\sqrt{M} + 1} \left(\frac{n_1}{n_2}\right) \left(\frac{S_1}{S_2}\right) \left(\frac{G_1}{G_2}\right) F_{1,2}, \tag{7}$$

where

$$F_{1,2} = \frac{A_{p,1}{}^2 A_{\mathrm{pr},1}}{A_{p,2}{}^2 A_{\mathrm{pr},2}}.$$

Equation (7) allows for the measurement of relative values of $\chi^{(3)}$. The factor $F_{1,2}$ can be obtained from power or energy measurements of the pump and probe beams in each arm or from maximum-to-minimum intensity ratio M_{id} of the conjugate interference signal when two identical samples are used in the interferometer. Then

$$F_{1,2} = \frac{\sqrt{M_{id}} + 1}{\sqrt{M_{id}} - 1}. \tag{8}$$

The sensitivity of the MTG interferometer is high because the signal detected is due to the interference of the phase-conjugate waves, while the scattered light does not cause interference and hence is not detected. The weak-signal detection capability becomes apparent if we assume a case when the ratio of conjugate signals is small, $I_{c,2}/I_{c,1} = a \ll 1$. In this case the maximum-to-minimum ratio at the CI output is $1 + 4\sqrt{q}$. For example, if the ratio of conjugate signals, $I_{c,2}/I_{c,1}$, is 2×10^{-4}, the modulation of the CI output signal will be 6% and the period will be the same as the period of the PI output signal. This is equivalent to a signal amplification of 300 times.

We have verified these results by using our interferometer in conjunction with an experimental system similar to the one described earlier.[9] The laser system consisted of a cw mode-locked Nd:YAG laser emitting 1 W of power after frequency doubling to 532 nm. The external angle between the pump (120 mW) and probe (85 mW) beams was 2.5°. PC1 was a 3-mm-thick semiconductor-doped glass filter, OG 530, mounted directly in front of a high reflector with an air gap of 0.1 mm. The position of the high reflector in the second arm was controlled by a piezoelectric transducer, PZT.

To check the interferometer we initially used OG530 in both interferometer arms. The signals from the two outputs of the interferometer were recorded using a photodiode, PD, and a photomultiplier, PMT, integrated with lock-in amplifiers that feed a Macintosh SE computer for data analysis and plotting. The signals recorded simultaneously at both outputs are shown in Fig. 2(a). The output signal as a function of the linearly incremented voltage applied to the PZT was fitted, using a nonlinear least-squares algorithm, to sine functions of the form $a \sin(bV + c) + d$ (where b is the period, V the applied PZT voltage, c the phase shift, and a and d constants). This allowed for an accurate determination of the phase shift and intensity ratio. Following this procedure, we determined a relative phase shift between the conjugate beams from two OG530 filters to be $\varphi_{0,2} - \varphi_{0,1} = 1.6°$. Repeated measurements of the phase shift using identical OG530 samples in both arms showed a reproducibility of ±5°. We believe that this uncertainty originated from photochemical, irreversible changes in the sample owing to the laser exposure. A more detailed description and analysis of this effect will be published shortly.[10] From the data shown in Fig. 2(a) we calculated $F_{1,2}$ to be 1.2 ± 0.1.

In another set of experiments we detected interference between the conjugate signals from the OG530

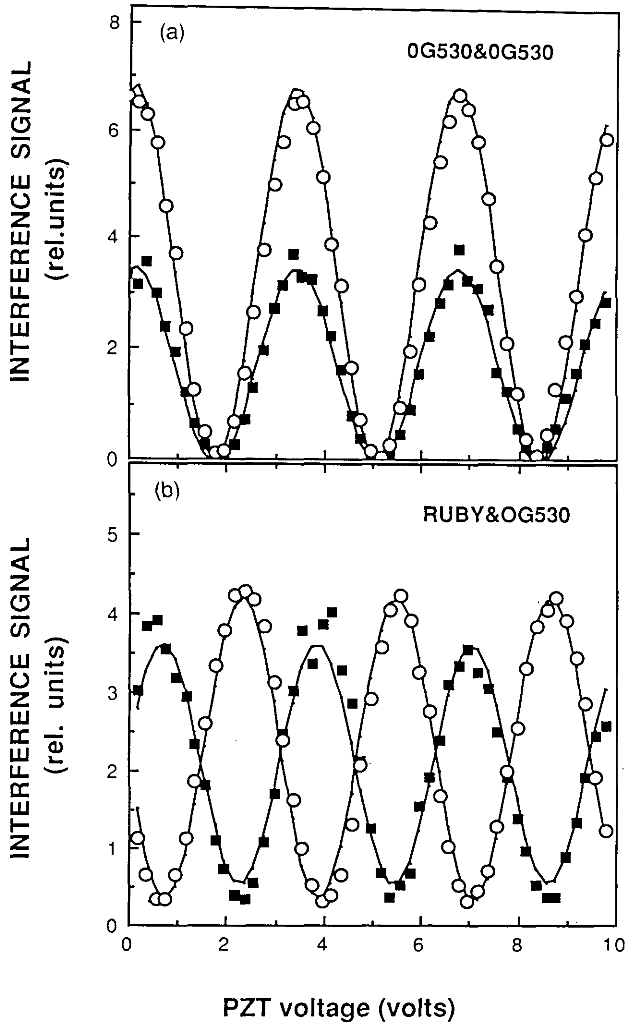

Fig. 2. Interference signal at PI output (O) and CI output (■): (a) when OG530 is used as phase conjugator in both interferometer arms and (b) when OG530 is PC1 and ruby crystal is PC2.

filter and a 5-mm ruby crystal. From the results presented in Fig. 2(b) we see immediately that the interference patterns from the two outputs are shifted by π. This suggests that the signs of $\chi^{(3)}$ in these two conjugators are opposite. Recent data[11,12] show that the real part of $\chi^{(3)}$ in ruby is an order of magnitude larger than the imaginary part. This measured phase shift, π, between conjugate waves in OG530 and ruby indicates that the real part of $\chi^{(3)}$ of OG530 is also much larger than the imaginary part. This result is consistent with the band-filling model.[13,14]

The ability of this interferometer to measure weak signals and $\chi^{(3)}$ ratio was proved in the case when a 2-mm CS_2 cell was used as PC2. The periodic change of CI interferometer output signal was easily detectable. We measured $M = 1.5$. Earlier attempts to record a conjugate signal from CS_2 using conventional schemes for degenerate four-wave mixing phase conjugation with the same laser failed. To our knowledge, no results on CS_2 phase conjugation with a cw or a cw mode-locked laser have been reported. Using the experimental interference data obtained, we calculated from expression (7) that $\chi^{(3)}_{OG530}/\chi^{(3)}_{CS_2} = 19 \pm 3$. The ratio of $\chi^{(3)}$ in OG530 and CS_2 is close to the value previously reported by Gabel *et al.*[15] Using the absolute value for $\chi^{(3)}_{1111}$ in CS_2,[16] we calculated that $\chi^{(3)}$ of OG530 nm is 7×10^{-12} esu.

In conclusion, a new type of phase-conjugation interferometer suitable for measuring $\chi^{(3)}$ from weak conjugate signals has been constructed. This interferometer also makes possible the accurate determination of relative phase shifts and is especially useful when the phase-conjugate reflectivities to be investigated have a low magnitude.

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