Twyman–Green Interferometry for Semi-Quantitative Analysis of Homogeneity and Nonlinearity of a Mixing Crystal

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Abstract
Application of Twyman–Green interferometry is extended to achieve semi-quantitative assessment of optical qualities of a nonlinear crystal. Interferograms of a proustite crystal are analyzed and effects of crystal imperfections on the conversion efficiency of a parametric upconversion system are evaluated.

I. Introduction
Optical parametric upconversion systems may be designed to change infrared images into visible pictures. We have performed a detailed study (Schow, 1977; Riazi, 1978) of parametric upconverters and have analyzed the factors which determine their range of applications. The quantum efficiency is generally very low for sum frequency upconversion processes, and certain applications require complex schemes, such as resonance of pump beam or the infrared image (Schow, Riazi, Gandhi & Grow, 1981a, b; Riazi, Christensen & Gandhi, 1979, 1981), to increase the efficiency. The conversion efficiency is also a direct function of both linear and nonlinear characteristics of the mixing crystal. Optical parameters of an actual crystal can deviate significantly from values stated in the literature (in regard to ideal crystals of the same type). Therefore, in the study of upconversion systems, the need arises to evaluate the optical qualities of the mixing crystal and to quantify various effects due to crystal imperfections. Use of a Twyman–Green interferometer for qualitative appraisal of composition variations along the length of a crystal has been described elsewhere (Zernike & Midwinter, 1967). In this paper we extend the application of Twyman–Green interferometry to achieve semi-quantitative information regarding both linear and nonlinear characteristics of a crystal. First, we describe a parametric upconverter and present both the actual and the expected theoretical efficiencies of the system. Then, by analyzing the interferograms of the crystal, we explain the discrepancy between experimental and theoretical values of efficiency.

II. A parametric upconversion system
Fig. 1 shows the block diagram of a parametric upconverter. Gold reflectors $M_1$ and $M_2$ are used to direct the beam of a CO$_2$ laser at $\lambda_p = 10.6 \mu$m into the mixing crystal (proustite). The pump laser is a 250 ns pulsed dye laser (Phase-R model DL-2100C) operating at $\lambda_p = 0.697 \mu$m and repetition rate 0.5 Hz. A narrow-band interference filter is used to ensure a high rejection of laser fluorescence. A quarter-wave plate and a Nicol prism are used to achieve proper polarization of the laser beam. A set of lenses is employed to expand the pump beam in order to fill the aperture of the crystal. Inside the crystal the infrared and pump beams mix, producing a sum frequency beam at $\lambda_s = 0.654 \mu$m. The CO$_2$ polarization is ordinary, the pump is also ordinary, and the upconverted signal is extraordinary. A set of filters and polarizers are placed...
between the output of the crystal and input of the image intensifier to allow passage only of the upconverted beam. The conversion efficiency of the system can be determined experimentally from the relationship (Schow, Riazi, Gandhi & Grow, 1981a)

\[ \eta_T = \frac{P_s \lambda_s}{P_{ir} \lambda_{ir}}, \]  

(1)

where \( P_s \) is the average upconverted power at threshold (i.e. the minimum power which can be detected by the image intensifier) and \( P_{ir} \) is the corresponding threshold value of infrared power. Here \( P_{ir} = 0.707 \) W, and \( P_s = 0.762 \) pW, consequently \( \eta_T = 6.64 \times 10^{-14} \). The conversion efficiency (when phase mismatch is zero) can be also determined theoretically from the relationship (Zernicke & Midwinter, 1967)

\[ \eta_T = \frac{512 \pi^2 d^2 L^2 P_p T}{C n_r n_p \rho \lambda_{ir} \lambda_{ir}}, \]  

(2)

[in c.g.s. units; to convert to SI units divide by \( 64\pi e_0 \)]

where \( d \) is the effective nonlinear coefficient of the crystal, \( L \) is the crystal length, \( P_p / A \) is the average pump power density, \( C \) is the speed of light, \( T \) is the net transmissivity of all optical components (i.e. lenses, filters, polarizers) at sum frequency, and \( n_r, n_p, \) and \( n_s \) are the indices of refraction of the crystal at infrared, pump, and sum frequencies, respectively. For this system we have: \( d = 7.54 \times 10^{-8} \) e.s.u. \( (31.6 \text{ pm } \text{V}^{-1}) \) [for type I phase matching with two ordinary rays and one extraordinary (Zernike & Midwinter, 1967), \( d = d_{31} \sin \theta - d_{22} \cos \theta \sin(3\varphi) \) for class 3m crystals such as proustite. Here azimuthal angle \( \varphi = 90^\circ \), phasematching angle \( \theta = 31.5^\circ \), nonlinear coefficients (Chemla, Kupecek & Schwartz, 1973) \( d_{31} = 40 \times 10^{-9} \) e.s.u. \( (16.7 \text{ pm } \text{V}^{-1}), \) and \( d_{22} = 64 \times 10^{-9} \) e.s.u. \( (26.8 \text{ pm } \text{V}^{-1}), \) so that \( d = 7.54 \times 10^{-8} \) e.s.u. \( (31.6 \text{ pm } \text{V}^{-1}) \)], \( L = 1.85 \text{ cm}, \ P_p / A = 175 \text{ W } \text{m}^{-2}, \ n_r = 2.716, \ n_p = 2.969, \ n_s = 2.952, \lambda_{ir} = 10.6 \mu \text{m}, \lambda_s = 0.654 \mu \text{m}, \ C = 3 \text{ Tm s}^{-1}, \ T = 0.258; \) consequently, \( \eta_T = 2.78 \times 10^{-9} \). The ratio between the theoretical and experimental results is

\[ R = \frac{\eta_T^{\text{experimental}}}{\eta_T^{\text{theoretical}}} = \frac{6.64 \times 10^{-14}}{2.78 \times 10^{-9}} = 2.39 \times 10^{-5}. \]

III. Analysis of crystal nonlinear properties

We have employed a Twyman–Green interferometer to evaluate the nonlinear characteristics of the crystal. Our experimental setup is illustrated in Fig. 2. The beam from a He–Ne laser is expanded, polarized, and split into two beams. One beam passes through the crystal, which is placed inside a capacitor, and then it is reflected by mirror \( M_1 \) toward the observation screen. The second beam crosses the beam splitter, is reflected by \( M_2 \), returns, is reflected on the semireflecting face of the beam splitter, and it is superposed on the first beam, thereby resulting in fringes on the observation screen. The resultant fringe patterns, when there is no voltage applied across the crystal and when there is an applied voltage of \( 7.5 \text{ kV} \), are illustrated in Figs. 3(a) and (b), respectively. Analysis of Fig. 3 indicates that a voltage of \( 7.5 \text{ kV} \) results in a phase shift of \( \pi/2 \) rad. Fig. 4 shows that the crystal is
oriented such that the applied voltage has Y and Z components, and the laser beam is polarized in the YZ plane. For this configuration, we have (Yariv, 1967)

\[ A(1/n^2)_1 = r_{13}E_3 - r_{12}E_2, \]

where \( n \) is the index of refraction, \( r_{ij} \) is the electro-optic tensor, and subscripts 1, 2, and 3 correspond to \( x, y, \) and \( z \), respectively. Also,

\[ \Delta n_1 = -\frac{n^3}{2}A\left(\frac{1}{n^2}\right)_1 = -\frac{n^3}{2}(r_{13}\sin \theta - r_{12}\cos \theta)E_0. \]

For our case, \( n_1 = 2.97, \theta = 31.5^\circ, r_{13} = 2.78 \text{ pm V}^{-1}, \) and \( r_{12} = 3.21 \text{ pm V}^{-1} \) (Trevelyan, 1969), then

\[ \Delta n_1 = 1.68 \times 10^{-11}E_0, \]

where \( E_0 \) is the free-space electric field between the electrodes. The two-pass phase shift is related to the change in the index of refraction by

\[ \Delta \phi = 4\pi H\Delta n_1/\lambda, \]

where \( H \) is the spacing between the electrodes. Here \( \lambda = 0.6328 \mu \text{m}, \Delta n_1 = 1.68 \times 10^{-11}E_0, \) and \( E_0H = V_0 = 7.5 \text{ kV}, \) therefore the theoretical phase shift is

\[ \Delta \phi = 0.8 \pi. \]

The electro-optic coefficient of our crystal is smaller than values given in the literature (for optimum proustite) by the ratio

\[ \frac{r_{13}\text{[experimental]}}{r_{13}\text{[theoretical]}} = \frac{r_{12}\text{[experimental]}}{r_{12}\text{[theoretical]}} = \frac{\Delta \phi\text{[experimental]}}{\Delta \phi\text{[theoretical]}} = \frac{0.5 \pi}{0.8 \pi}. \]

Consequently (see Appendix A), the efficiency is reduced by a factor of

\[ \left[ \frac{d\text{[experimental]}}{d\text{[theoretical]}} \right]^2 = \left( \frac{0.5 \pi}{0.8 \pi} \right)^2 = 0.391 \]

due to nonlinear coefficients which do not meet the values stated in the literature.

### IV. Test of homogeneity of the crystal

The experimental procedure for analyzing the linear properties of the crystal is similar to that of Fig. 2, except it is no longer necessary to apply a voltage across the crystal. The desired interferograms are presented in Fig. 5. Distortion in the fringes of Figs. 5(b) and (c), and also dissimilarity of fringe patterns with polarization, indicate that the crystal is inhomogeneous. It is not possible to quantify directly effects due to linear deficiencies; however, to give useful insight, we present an analysis of local phase mismatch in terms of local index variation. Consider

![Fig. 4. Schematic diagram of crystal orientation employed for evaluation of nonlinear coefficients.](image)

![Fig. 5. Twyman–Green interferograms (a) when crystal is absent, (b) for ordinary ray, and (c) for extraordinary ray.](image)
Fig. 6 which is a superposition of nulls of Figs. 5(b) and (c). A and B are two points with a phase difference of π rad. For optimum phase matching at A:

\[(\Delta k)_A = 2\pi \left( \frac{n_s - n_{ir}}{\lambda_s} - \frac{n_{ir} - n_p}{\lambda_{ir}} \right) = 0.\]

The phase mismatch factor at B is given by:

\[(\Delta k)_B = 2\pi \left( \frac{n_s + \Delta n_s - n_{ir} + \Delta n_{ir} - n_p + \Delta n_p}{\lambda_s} - \frac{n_{ir}}{\lambda_{ir}} - \frac{n_p}{\lambda_p} \right) \equiv 2\pi \left( \frac{\Delta n_s}{\lambda_s} - \frac{\Delta n_{ir}}{\lambda_{ir}} - \frac{\Delta n_p}{\lambda_p} \right).\]

Here \(\Delta n\) represents the difference in indices of refraction of crystal at A and B, \(\Delta n_{ir} = \Delta \varphi_{ir} \lambda_{ir} / 2\pi L\), \(\Delta n_p = \Delta \varphi_p \lambda_p / 2\pi L\) (\(\Delta \varphi\) is the phase difference between points A and B) and \(\Delta \varphi_{ir} = \Delta \varphi_p = \pi\). To gain an idea of the value of \(\Delta k\), let \(\Delta n = 0\); then

\[(\Delta k)_B = -2\pi/L.\]

Upconversion efficiency is proportional to \(\text{sinc}^2(L\Delta k/2)\), so that when efficiency is maximum at point A, it is zero at B. The distance between points A and B is 1 mm and the diameter of homogeneity (based on the criterion that the crystal is homogeneous over an area in which the upconverted signal is at least 0.81 of maximum) for the crystal is only 0.5 mm. The crystal is inhomogeneous to the laser beam in cross section, and also along its length, adversely affecting phase matching and efficiency. In the previous section it was shown that a reduction factor of 0.391 in efficiency is attributed to deficient nonlinear properties of the crystal. We have also determined (see Appendix B) that efficiency is reduced by a factor of 0.328 due to high absorption, a factor of 0.391 is accounted for by a deficient nonlinear coefficient, and a factor of \(1.86 \times 10^{-4}\) is due to inhomogeneities of the crystal. The contribution of crystal inhomogeneities to reduction in efficiency was determined by first examining other factors which result in a difference between the theoretical and experimental values of efficiency (as noted earlier, it is not possible to make a direct quantitative assessment of effects due to inhomogeneities). In evaluating the crystal, we have assumed that values given by Chemla, Kupecek & Schwartz (1973) and Trevelyan (1969) for electro-optic and nonlinear coefficients of proustite refer to a crystal of (near) optimum qualities.

V. Discussion and conclusions

Requirements for high crystal quality have been discussed by Zernike & Midwinter (1967). A method which employs powders for evaluation of nonlinearity and phase-matching capabilities of a crystal has also been described previously (Kurtz & Perry, 1968). We have presented for the first time an approach, using Twyman–Green interferometry, to obtain semi-quantitative evaluation of both linear and nonlinear qualities of a crystal. Our method has proven valuable for testing the optical qualities of a crystal employed in an image upconversion system. For a parametric upconverter of actual efficiency lower than the expected value by 2\(39 \times 10^{-3}\), we determined that a reduction of 0.328 is due to high absorption, a factor of 0.391 is accounted for by a deficient nonlinear coefficient, and a factor of \(1.86 \times 10^{-4}\) is due to inhomogeneities of the crystal. The contribution of crystal inhomogeneities to reduction in efficiency was determined by first examining other factors which result in a difference between the theoretical and experimental values of efficiency (as noted earlier, it is not possible to make a direct quantitative assessment of effects due to inhomogeneities). In evaluating the crystal, we have assumed that values given by Chemla, Kupecek & Schwartz (1973) and Trevelyan (1969) for electro-optic and nonlinear coefficients of proustite refer to a crystal of (near) optimum qualities.

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APPENDIX A

Relationship between electro-optic and nonlinear coefficients

The matrix of electro-optic coefficients for a trigonal 3m crystal (such as proustite) is (Chemla, Kupecek & Schwartz, 1973)

\[
\begin{vmatrix}
0 & -r_{22} & r_{13} \\
0 & r_{22} & r_{13} \\
0 & 0 & r_{33} \\
0 & r_{51} & 0 \\
r_{51} & 0 & 0 \\
-r_{22} & 0 & 0
\end{vmatrix}
\]

where

\[
r_{22} = -r_{12}
\]

(also \(r_{12} = r_{61}, r_{51} = r_{42}\) and \(r_{13} = r_{23}\)). The nonlinear coefficients \(d_{ij}\) are related to electro-optic coefficients \(r_{ij}\) by (Zernike & Midwinter, 1967).
From (3), (A1), and (A2), it follows that

\[
\frac{d_{31\text{ experimental}}}{d_{31\text{ theoretical}}} = \frac{d_{22\text{ experimental}}}{d_{22\text{ theoretical}}} = \frac{5}{8}.
\]

The effective nonlinear coefficient is given by

\[
d = d_{31} \sin \theta - d_{22} \cos \theta \sin 3\varphi.
\]

Consequently,

\[
\frac{d_{\text{experimental}}}{d_{\text{theoretical}}} = \frac{5}{8}.
\]

The quantum efficiency varies as \(d^2\), therefore the experimental value of efficiency is less than the theoretical value by a factor of 0.391 due to deficient nonlinear properties.

### APPENDIX B

**Effect of crystal absorption on conversion efficiency**

For perfect phase matching the upconverted power \(P_s\) which includes effects of crystal absorption is given by (Aggarwal & Lax, 1977)

\[
P_s = P_{os} \exp(-\alpha_s L) \left\{ 1 + \exp \left[ -(\alpha_p + \alpha_{ir} - \alpha_s) L \right] \right. \\
- \left. 2 \exp \left[ -\frac{1}{2}(\alpha_p + \alpha_{ir} - \alpha_s) L \right] \right\} \\
\times \left[ (L/2)(\alpha_p + \alpha_{ir} - \alpha_s)^2 \right]^{-1},
\]

where \(P_{os}\) represents upconverted power when absorption is negligible and \(\alpha_p\), \(\alpha_{ir}\), and \(\alpha_s\) are power absorption coefficients at pump, infrared, and sum frequencies, respectively. For our case, \(\alpha_p = 0.271\) cm\(^{-1}\), \(\alpha_{ir} = 0.362\) cm\(^{-1}\), and \(\alpha_s = 0.571\) cm\(^{-1}\) (the absorption coefficients of the crystal were evaluated experimentally), consequently

\[
P_s = 0.328 P_{os}.
\]

Conversion efficiency is directly proportional to upconverted signal power, hence the absorption of the crystal reduces the efficiency by 0.328.

### References


