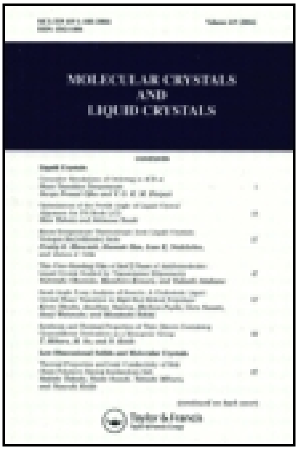

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ANALYTICAL DESCRIPTION OF THE INTENSITY-DEPENDENT OPTICAL PHASE SHIFT DUE TO CASCADED $\chi^{(2)}$ PROCESSES

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Abstract In phase-matchable $\chi^{(2)}$ crystals, the fundamental wave experiences an intensity-dependent phase shift in the vicinity of the phase-matching angle. Its physical origin are two cascaded $\chi^{(2)}$ processes, namely, second harmonic generation and difference frequency mixing between the second-harmonic and the fundamental wave. A new theoretical description of the nonlinear phase shift is presented which is based on the pump depletion of the fundamental beam and on generalized Kramers–Kronig relations. The pump depletion acts as an effective attenuation which, according to Cauchy's integral law, must be related to a corresponding phase shift. The frequency variable in the regular Kramers–Kronig relations is here replaced with the phase mismatch angle. In the limit of small pump depletion, a simple analytical formula is obtained for the phase shift.

INTRODUCTION

Nonlinear-optical phenomena are important for future applications in the field of information transfer and processing. Two main groups of applications are envisioned. Electrooptic modulators, which form the link between electronic and optical processing systems, are based on the variation of an optical phase shift (or index of refraction) with the electric field strength. Such a behavior is described by nonlinear susceptibilities of second order ($\chi^{(2)}$). The dependence of an optical phase shift on the light intensity, on the other hand, which is required for all-optical switching devices, arises from third-order nonlinearities ($\chi^{(3)}$) in most cases. Since $\chi^{(3)}$ processes are of higher order than $\chi^{(2)}$ effects, they are usually much smaller and, hence, less suitable for technical applications.

In the past few years, however, two mechanisms were found which give rise to an intensity-dependent phase shift of a light wave but are based on the combination or cascading of two $\chi^{(2)}$ processes. One of these effects occurs in phase-matchable $\chi^{(2)}$ crystals in the vicinity of the phase-matching angle.[1–3] If a second-harmonic wave is generated in the crystal, an intensity-dependent phase shift of the fundamental wave is observed.[3] The mechanism which was identified as the origin of this behavior,[2,3] consists in the cascading of second-harmonic generation and difference frequency mixing between the second harmonic and the transmitted fundamental wave. The intensity-dependent phase shift has opposite signs on both sides of the phase-matching angle, because it depends on the difference between the phase velocities of the two waves. Exactly at the phase-matching angle, the nonlinear phase shift is zero. By numerically solving the nonlinear propagation equation for the fundamental wave in the crystal, the phase shift could be calculated as a function of the mismatch angle.[2,3] The second mechanism was discovered only very recently;[4] it consits in the combination of optical rectification and the linear electrooptic effect and occurs in all non-centrosymmetric materials.

In the present paper, the first effect connected with phase matching is studied and a new interpretation is given which is based on generalized Kramers–Kronig relations. The pump depletion due to second-harmonic generation gives rise to an effective attenuation of the fundamental wave which must be related to a corresponding phase shift. The calculation shows, however, that for certain mismatch angles a large phase shift can also occur without any attenuation of the fundamental wave.

THEORY

Kramers–Kronig Relations in Linear Optics

The propagation of light in a transparent medium can be described with a complex transmission function

$$\tilde{\Phi}(\omega) = \phi(\omega) - i\delta(\omega), \tag{1}$$

such that the electric field strength of the light wave behind a sample of thickness L is given by

$$E_L(t) = E_0(t)\exp[-i\tilde{\Phi}(\omega)] = E_0(t)\exp[-i\phi(\omega)]\exp[-\delta(\omega)]. \tag{2}$$

$\tilde{\Phi}$ is a function of the optical frequency ω; its real and imaginary part are related to the index of refraction $n(\omega)$ and to the intensity absorption coefficient $\alpha(\omega)$, respectively. For example, if $\delta(\omega)$ is a Lorentzian absorption profile with amplitude A, full width γ, and center frequency ω_0, the corresponding transmission function reads

$$\tilde{\Phi}(\omega) = -A\frac{\gamma/2}{\omega - \omega_0 - i\gamma/2}. \tag{3}$$

$\tilde{\Phi}(\omega)$ can be extended as an analytical function to the whole complex ω plane. If it has no poles in the lower half-plane, the real and imaginary part are connected by the well-known Kramers–Kronig relations[5]

$$\phi(\omega) = \frac{1}{\pi}\mathrm{PV}\int_{-\infty}^{+\infty}\frac{\delta(\omega')}{\omega'-\omega}d\omega'; \tag{4}$$

$$\delta(\omega) = -\frac{1}{\pi}\mathrm{PV}\int_{-\infty}^{+\infty}\frac{\phi(\omega')}{\omega'-\omega}d\omega', \tag{5}$$

which are based on Cauchy's integral law. PV denotes the Cauchy principal value. The Kramers–Kronig relations are of purely mathematical origin. Their validity is not restricted to a specific physical mechanism. Hence, they hold true for any analytical function which has no poles in the lower complex half-plane and tends to zero for infinite argument. The variable of the function need not necessarily be a frequency.

Application to Phase-Matched Second-Harmonic Generation

In contrast to Beer's law describing absorption phenomena in linear optics [Eq. (2)], the pump depletion of the fundamental wave in phase-matched second-harmonic generation is not governed by an exponential dependence.[6] Nevertheless, in the limit of small pump depletion one can define an effective amplitude attenuation coefficient[7]

$$\delta_\omega(\Theta) = \delta_\omega^{\max} \left[\frac{\sin(\Theta/2)}{\Theta/2}\right]^2 , \tag{6}$$

so that the decrease of the electric light field $E_\omega(t)$ in the crystal is analogous to Eq. (2). The prefactor is $\delta_\omega^{\max} = (1/2)CL^2 I_{\omega,0}$. Here $\Theta = (k_{2\omega} - 2k_\omega)L$ denotes the phase mismatch angle, $I_{\omega,0}$ is the intensity of the incident fundamental beam, and the factor C depends on the $\chi^{(2)}$ coefficient and on the dielectric constants of the crystal. Note that only the first-order term in the Taylor expansion of the factor $\exp[-\delta_\omega(\Theta)]$ is correct. The above description shows immediately, however, that the pump depletion must be connected with an intensity-dependent phase shift $\phi_\omega(\Theta)$ of the fundamental wave. The phase shift can be calculated by inserting Eq. (6) in the first Kramers–Kronig relation [Eq. (4)] with the angle Θ replacing the optical frequency ω. It is possible to solve the integral analytically using standard tables.[8] This has the simple result

$$\phi_\omega(\Theta) = \delta_\omega^{\max} \frac{2}{\Theta}\left(\frac{\sin\Theta}{\Theta} - 1\right). \tag{7}$$

Plots of $\delta_\omega(\Theta)$ and $\phi_\omega(\Theta)$ are shown in Fig. 1(a) and 1(b), respectively. They confirm the results of numerical calculations.[2] From Eqs. (6) and (7) the intensity-dependent complex transmission function is obtained in analogy to Eq. (1)

$$\tilde{\Phi}_\omega(\Theta) = \delta_\omega^{\max} \frac{2i}{\Theta^2}\left(e^{-i\Theta} + i\Theta - 1\right). \tag{8}$$

It has no poles in the whole complex Θ plane, which shows that the Kramers–Kronig relations are applicable.

NUMERICAL EXAMPLE

Since the intensity-dependent optical phase shift in phase-matched second-harmonic generation is due to $\chi^{(2)}$ rather than $\chi^{(3)}$ processes, it can reach very high values.[3] The analytical result derived above [Eq. (7)] allows us to calculate its magnitude quantitatively. The prefactor $\delta_\omega^{\max}$ determines both the pump depletion for $\Theta = 0$

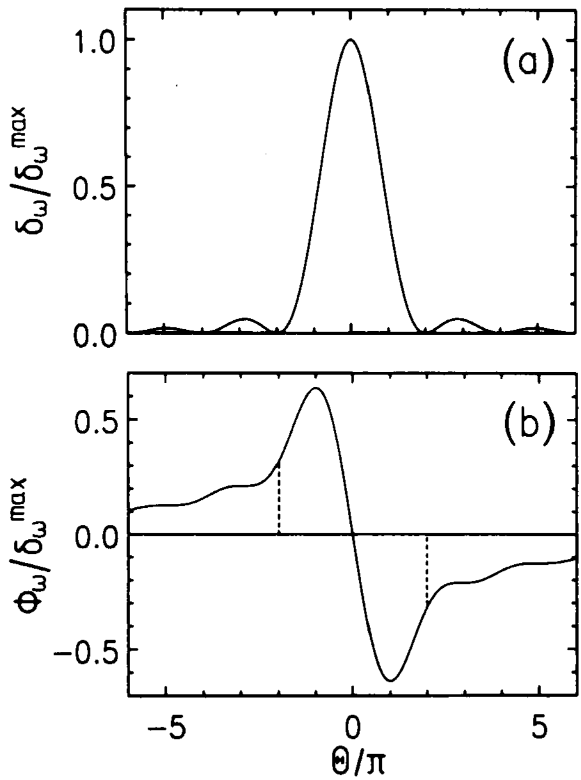

FIGURE 1 Amplitude attenuation coefficient $\delta_\omega(\Theta)$ [part (a); Eq. (6)] and corresponding intensity-dependent phase shift $\phi_\omega(\Theta)$ [part (b); Eq. (7)] of the fundamental wave. At $\Theta = \pm 2\pi$ (indicated by the dashed lines) a large phase shift occurs without any loss.

and the magnitude of the angle-dependent phase shift. Hence, the phase shift is readily obtained when the pump depletion for perfect phase matching and the mismatch angle are known. In this context it is important to note that for certain mismatch angles the fundamental wave experiences a large phase shift without any pump depletion. At the innermost zeroes $\Theta = \pm 2\pi$ of $\delta_\omega(\Theta)$, for instance [indicated by the dashed lines in Fig. 1(b)], the phase shift amounts to $\mp\delta_\omega^{max}/\pi$.

As an example let us assume that at a certain light intensity 10% of the incident light power is converted into the second harmonic for $\Theta = 0$. This corresponds to $\delta_\omega^{max} = 0.05$. Then, if the angle is detuned to $\Theta = \pm 2\pi$, a phase shift of $\mp 0.05\pi^{-1}$ rad or about $\mp 0.9°$ results with respect to zero power and the attenuation of the fundamental wave is zero. Since phase-matchable $\chi^{(2)}$ crystals of excellent optical quality can be fabricated, for instance for intracavity frequency doublers in lasers, this feature may also make them possible candidates for all-optical switching devices.

SUMMARY AND CONCLUSIONS

It was shown that the large intensity-dependent optical phase shift which is observed in $\chi^{(2)}$ crystals in the vicinity of the phase-matching angle, is intimately related to the attenuation of the fundamental wave due to pump depletion. The correlation is similar as in linear optics between absorption and dispersion profiles. Hence, generalized Kramers–Kronig relations can be used for a quantitative calculation of the phase shift. In the limit of small pump depletion, a simple analytical expression is obtained for the phase shift as a function of the phase-mismatch angle. An important finding is the fact that also at those mismatch angles, for which the pump depletion is zero, a large phase shift occurs. This effect has the physical origin that a second-harmonic wave is always generated inside the crystal but is finally transformed into the fundamental wave again for those angles for which $\delta_\omega(\Theta) = 0$. On its way through the crystal the second-harmonic wave participates in difference frequency mixing and, thus, gives rise to the mechanism generating the phase shift.[2,3]

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