Interferometry of spontaneous parametric light scattering

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Abstract. The interference of biphotonic light produced in the process of spontaneous parametric scattering (SPS) in two spatially separated non-centrally symmetric crystals is considered. A scheme of the nonlinear Mach – Zehnder interferometer is realised. It is shown that in this case, the frequency-angular intensity distribution in the far-field zone (the SPS spectrum) contains information not only on non-centrally symmetric crystals, where biphotonic fields are created, but also on the optical properties of the medium located between the scattering crystals. The algorithms for the determination of the dispersion law in this medium are developed.

1. Introduction

The basic properties of biphotonic fields created during spontaneous parametric scattering (SPS) have long been known and were discussed in detail in Ref. [1]. However, the influence of the inhomogeneous spatial distribution of the optical parameters of a scattering object on the SPS spectrum was not in fact considered in Ref. [1]. This problem was studied in detail in Refs [2 – 4]. Nonlinear diffraction and interference of biphotonic fields result in a substantial change in the space and frequency distribution of the field in the far zone. This allows one to use the SPS spectra observed upon scattering from several spatially separated regions for determining not only optical properties of nonlinear scattering crystals [5] but also the dispersion of a substance that fills the space between scattering regions.

All the fundamental aspects of nonlinear (quantum) interference observed upon scattering of light in a spatially inhomogeneous medium have been considered in Ref. [4]. This paper is devoted to the first, in our opinion, practical application of the SPS interferometry, namely, the development of specific algorithms for studying dispersion properties of optical materials in the spectral region where absorption is not too strong (a quantitative criterion for weak absorption is presented below). Note that the main advantage of the method suggested is the expansion of a class of substances that can be studied using the SPS spectroscopy.

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This method can be applied not only to non-centrally symmetrical crystals (with nonzero quadratic susceptibility $\chi^{(2)}$) but also to linear media. The IR absorption related to optical vibrational modes of molecules is not considered in this paper and has been discussed in detail in Ref. [6].

2. SPS interferometry

Fig. 1 shows the principal scheme of a nonlinear Mach – Zehnder interferometer. A laser pump beam is incident on two in-line nonlinear crystals ($\chi^{(2)} \neq 0$) of the same thickness L. A gap between the crystals of width L' is filled with an arbitrary dispersion linear medium. In each of the crystals, SPS occurs, which is often interpreted as the decay of a pump photon with frequency ω_p into a pair of photons in a signal (s) and an idle (i) modes [1]. In the stationary case, the frequencies of the three modes satisfy the condition
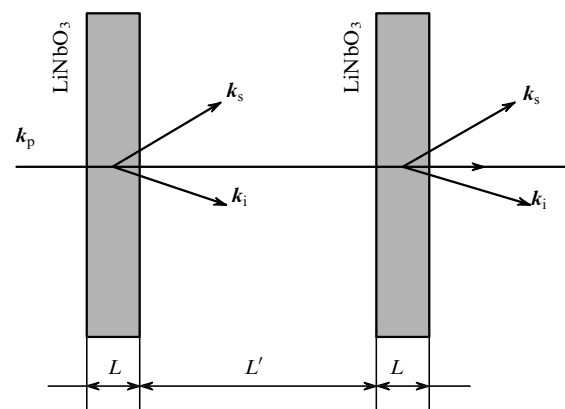
$$\omega_p = \omega_s + \omega_i. \tag{1}$$

Figure 1. Principal scheme of a nonlinear Mach – Zehnder interferometer.

Consider the case when a medium is transparent at the pump (ω_p) and signal (ω_s) frequencies and the absorption coefficient of idle waves is smaller than the inverse thickness of the gap: $\alpha_i < 1/L'$. Therefore, the radiation produced in the first crystal will come to the second crystal, having passed through the intermediate layer. For small scattering angles and (or) thin crystals, the condition of observation of the non-linear (or three-frequency) interference of the fields is satisfied [7] $(2L + L') \tan \vartheta_s \ll a$, where a is the pump beam diameter and ϑ_s is the angle of scattering inside the crystal.

The shape of the SPS line acquires the characteristic modulation structure and is described by the function

$$g(\omega_s) = \left\{ \frac{\sin(\delta/2)}{\delta/2} \cos\left[\frac{\delta + \delta'}{2}\right] \right\}^2, \qquad (2)$$

where $\delta(\omega_s, \vartheta_s) = \Delta L = (k_p - k_s - k_i)L$, $\delta'(\omega_s, \vartheta_s) = \Delta' L' = (k'_p - k'_s - k'_i)L'$ are the wave detunings in crystals and in gap between them, respectively, and k are the projections of wave vectors on the direction perpendicular to the layers.

Expression (2) has a simple physical meaning. The first factor in braces describes scattering in one nonlinear crystal, while the second factor describes the interference of light from both crystals. One can see that spontaneous emission is suppressed or amplified when the phase incursion is $\delta + \delta' = \pi + 2\pi n$ or $\delta + \delta' = 2\pi n$, respectively. Note that diffraction of a plane wave from a screen with two slits is described by a similar expression [8].

A useful corollary of expression (2), from the point of view of spectroscopic applications, is that the properties of the gap material affect only the modulation term. (Note here that the advantage of nonlinear interferometer schemes over the linear ones is that they do not require the adjustment with an accuracy of a few fractions of the wavelength [2].) The general frequency-angle shape of the line remains the same as for one crystal and is determined by the envelope $\text{sinc}^2(\delta/2)$.

The requirement of a weak absorption of the idle waves can be clearly interpreted in terms of correlation functions [6]: if the coherence length of radiation at the idle frequency is smaller than the gap thickness, then the signal fields created at points in space separated by the interval $L' \gg 1/\alpha'_i$ are out of phase, i.e., their intensities are added rather than amplitudes.

To observe spontaneous scattering, we chose congruent crystals of lithium niobate containing 5% of MgO impurity ($MgO : LiNbO_3$). These crystals are transparent in a broad spectral range from 0.4 to 5 μm and are highly resistant to laser irradiation [9]. The optical properties of $LiNbO_3$ and $MgO : LiNbO_3$ are well known [10, 11] and SPS and scattering spectra by polaritons for these materials have been studied in detail depending on various parameters, in particular, temperature and the impurity concentration [12]. All this allow us to focus our attention, in the analysis of the line shape (2), only on the argument of the modulation factor, which depends on the phase incursion δ', and do not take care of the envelope, assuming it to be a known function of the scattering angle and frequency.

The SPS spectrum of a $LiNbO_3$ crystal, which corresponds to scattering on the upper polariton branch, has the form of an 'ellipse' whose axis lies on the line of zero angles θ_s^{out}. The three-photon interference is observed in the Mach–Zehnder scheme at small scattering angles, i.e., near the region of collinear phase matching in the long-wavelength part of the spectrum. Because this region is remote from the absorption line of $LiNbO_3$, the function $\text{sinc}^2\delta$ adequately describes the shape of the scattering line in one crystal. [When idle waves are strongly absorbed in a crystal ($\alpha_i \gg 1/L$), we deal with scattering by polaritons. In this case, the line shape is described by a Lorentzian [1].] The required frequency interval is specified by a choice of the orientation of nonlinear crystals. The exact frequency tuning is usually achieved by slightly tilting crystals with respect to the direction of the pump polarisation.

Fig. 2a shows the calculated frequency-angular spectrum of the $LiNbO_3$ – crystalline quartz – $LiNbO_3$ system. The thickness of a quartz plate and crystals was $L' = 1.44$ mm and $L = 0.7$ mm, respectively. Fig. 2b shows the experimental spectrum that was obtained under the same conditions as the calculated one. The spectrum exhibits a distinct interference structure representing a system of alternating inclined maxima and minima. The thickness L of nonlinear crystals is usually constant in experiments, whereas the gap thickness L' can be varied from a few micrometers to a few centimetres.
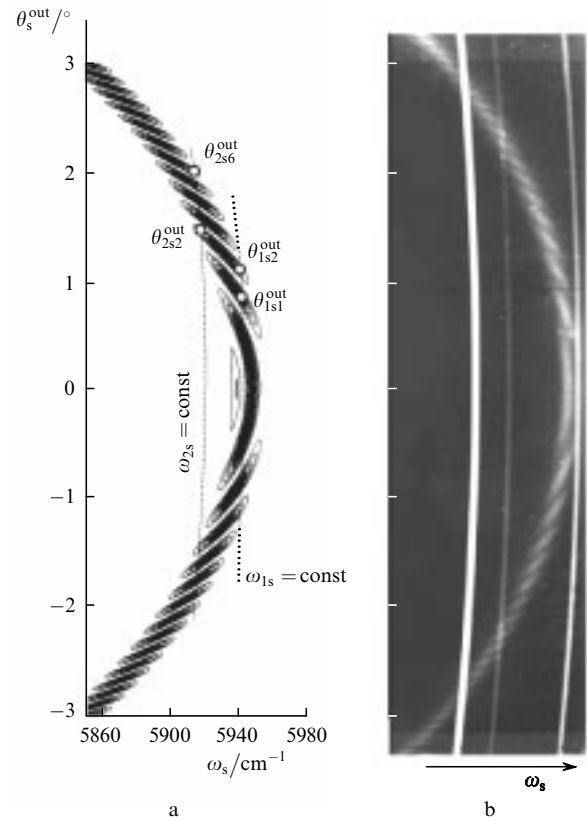
Figure 2. Spectrum calculated in co-ordinates of the outer angle θ_s^{out} and frequency ω_s of the signal wave (a) and experimental spectrum (b) for the thickness of nonlinear crystals $L = 0.7$ mm and the thickness of a linear dispersion medium (crystalline quartz) $L' = 1.44$ mm. Points θ_{isj}^{out} are obtained by the intersection of the constant-frequency line with the number ith (ω_{is}) and the jth order of the interference pattern.

As in the usual SPS spectroscopy, the frequency and the outer scattering angle θ_s^{out} of the signal wave are measured in experiments (the so-called crossed-dispersion scheme [13]). The idle-wave frequency ω_i required for calculations is determined from the condition (1).

Let us express the wave detuning in nonlinear (δ) and linear (δ') media in terms of the measured quantities

$$\delta(\omega_s, \theta_s^{out}) = 2\pi L \left\{ n_p \omega_p - n_s \omega_s \left[1 - \left(\frac{\sin\theta_s^{out}}{n_s}\right)^2\right]^{1/2} - \right.$$
$$\left. - n_i \omega_i \left[1 - \left(\frac{\omega_s \sin\theta_s^{out}}{\omega_i \, n_i}\right)^2\right]^{1/2} \right\}, \qquad (3)$$

$$\delta'(\omega_s, \theta_s^{out}) = 2\pi L' \left\{ n'_p \omega_p - n'_s \omega_s \left[1 - \left(\frac{\sin\theta_s^{out}}{n'_s}\right)^2\right]^{1/2} \right.$$

$$-n'_i\omega_i\left[1-\left(\frac{\omega_s}{\omega_i}\frac{\sin\theta_s^{out}}{n'_i}\right)^2\right]^{1/2}\right\}. \tag{4}$$

Here, the primed quantities are related to a linear medium, and frequencies are expressed in inverse centimetres.

The arguments of the modulation factor $(\delta+\delta')/2$ for two neighbouring maxima (minima) with numbers n and $n-1$ will differ from each other, according to (2), by π:

$$(\delta+\delta')_n-(\delta+\delta')_{n-1}=2\pi. \tag{5}$$

Each point of the experimental spectrum is characterised by quantities ω_s and θ_s^{out} in terms of which the detunings in the nonlinear crystal and the gap are expressed. When the dispersion of the refractive index is taken into account, these points should be chosen at the intersection of the constant-frequency line $\omega_s=const$ (or $\lambda_s=const$) with the intensity maxima (minima), as shown in Fig. 2a. Otherwise, the number of unknowns in the equation or in the system of equations will always exceed the number of equations.

We can determine the refractive index of the intermediate layer at one of the two frequencies, ω_s or ω_i, from equation (5). In this case, the refractive index at another frequency is considered unknown. For example, using the data on the dispersion in the visible region, we can find the dispersion law in the IR region. According to (3) and (4), the solution of (5) $n'_s=n'(\omega_s)$ will have the form

$$\frac{1}{n'_s}=\frac{1-A-B}{(L'/2)\omega_s(\sin^2\theta_{s4}^{out}-\sin^2\theta_{s3}^{out})}-\frac{1}{n'_i}\left(\frac{\omega_s}{\omega_i}\right), \tag{6}$$

where second subscripts at θ^{out} are the numbers n and $n-1$ of maxima (minima). The refractive index $n'_i=n'(\omega_i)$ at the idle frequency has the similar form

$$\frac{1}{n'_i}=\frac{1-A-B}{(L'/2)\omega_i(\omega_s/\omega_i)^2(\sin^2\theta_{s4}^{out}-\sin^2\theta_{s3}^{out})}-\frac{1}{n'_s}\left(\frac{\omega_i}{\omega_s}\right), \tag{7}$$

where

$$A=L\omega_s n_s\left[\left(1-\frac{\sin^2\theta_{s3}^{out}}{n_s^2}\right)^{1/2}-\left(1-\frac{\sin^2\theta_{s4}^{out}}{n_s^2}\right)^{1/2}\right], \tag{8}$$

$$B=L\omega_i n_i\left\{\left[1-\left(\frac{\omega_s}{\omega_i}\frac{\sin\theta_{s3}^{out}}{n_i}\right)^2\right]^{1/2}\right.$$
$$\left.-\left[1-\left(\frac{\omega_s}{\omega_i}\frac{\sin\theta_{s4}^{out}}{n_i}\right)^2\right]^{1/2}\right\} \tag{9}$$

are the functions of wave detunings at the signal and idle frequencies, respectively.

Expressions (6) and (7) were derived using the only approximation — to reduce expression (6) to a linear form, square roots in expressions (3) and (4) were expanded in Taylor series. The order of smallness of discarded terms is determined by the expression

$$\left(\frac{\sin\theta_s^{out}}{n}\right)^4. \tag{10}$$

This approximation is well justified because the scattering angles of the signal and idle waves are within few degrees

(1 – 2°) and the discarded quantity is four orders of magnitude smaller than other quantities entering the equation.

To determine refractive indices in the gap simultaneously at the frequencies of signal and idle waves, it is necessary to solve a system of two equations of the type (5). It is possible to show that such a system will have analytic solutions if the following two conditions are satisfied: (i) the wave detunings for each of the equations are calculated for two different constant-frequency lines: $\omega_1=const$ (for the first equation) and $\omega_2=const$ (for the second equation) and (ii) the refractive index of the linear medium is constant within the interval $\Delta\omega=\omega_2-\omega_1$. For a sufficiently small value of $\Delta\omega$ (which corresponds to $\Delta\lambda$ of the order of 1 nm), the requirement that n'_s and n'_i should be constant within this interval is well justified if the spectral range under study lies outside the region of resonances of the linear medium.

The system of equations for two frequencies has the form

$$(\delta_1+\delta'_1)_{n+1}-(\delta_1+\delta'_1)_n=2\pi,$$
$$(\delta_2+\delta'_2)_{n+1}-(\delta_2+\delta'_2)_n=2\pi. \tag{11}$$

The subscripts of the wave detunings correspond to frequencies ω_1 and ω_2 and the subscripts n and $n+1$ denote the number of the interference-pattern order in which the corresponding wave detuning are calculated.

The determinant of the system (11) is calculated from the expression

$$Det=\omega_{1s}\omega_{2i}\left(\frac{\omega_{2s}}{\omega_{2p}}\right)^2-\omega_{2s}\omega_{1i}\left(\frac{\omega_{1s}}{\omega_{1i}}\right)^2. \tag{12}$$

The required refractive indices n'_s and n'_i can be found from the system (11)

$$\frac{1}{n'_s}=\frac{D_1\omega_{2i}(\omega_{2s}/\omega_{2i})^2-D_2\omega_{1i}(\omega_{1s}/\omega_{1i})}{Det}, \tag{13}$$

$$\frac{1}{n'_i}=\frac{D_1\omega_{2s}-D_2\omega_{1i}}{Det}. \tag{14}$$

Here, the coefficients D_1 and D_2 contain the components of wave detunings at frequencies ω_1 and ω_2, respectively:

$$D_1=\frac{1-D_{1s}-D_{1i}}{(L'/2)(\sin^2\theta_{1s4}^{out}-\sin^2\theta_{1s3}^{out})}, \tag{15}$$

$$D_2=\frac{1-D_{2s}-D_{2i}}{(L'/2)(\sin^2\theta_{2s4}^{out}-\sin^2\theta_{2s3}^{out})}, \tag{16}$$

where

$$D_{1s}=L\omega_{1s}n(\omega_{1s})\left\{\left[1-\frac{\sin^2\theta_{1s3}^{out}}{n^2(\omega_{1s})}\right]^{1/2}-\left[1-\frac{\sin^2\theta_{1s4}^{out}}{n^2(\omega_{1s})}\right]^{1/2}\right\}; \tag{17}$$

$$D_{1i}=L\omega_{1i}n(\omega_{1i})\left\{\left[1-\left(\frac{\omega_{1s}}{\omega_{1i}}\frac{\sin\theta_{1s3}^{out}}{n(\omega_{1i})}\right)^2\right]^{1/2}\right.$$
$$\left.-\left[1-\left(\frac{\omega_{1s}}{\omega_{1i}}\frac{\sin\theta_{1s4}^{out}}{n(\omega_{1i})}\right)^2\right]^{1/2}\right\}; \tag{18}$$

$$D_{2s} = L\omega_{2s}n(\omega_{2s})\left\{\left[1 - \frac{\sin^2\theta_{2s3}^{\text{out}}}{n^2(\omega_{2s})}\right]^{1/2}\right.$$

$$\left. - \left[1 - \frac{\sin^2\theta_{2s4}^{\text{out}}}{n^2(\omega_{2s})}\right]^{1/2}\right\}; \quad (19)$$

$$D_{2i} = L\omega_{2i}n(\omega_{2i})\left\{\left[1 - \left(\frac{\omega_{2s}}{\omega_{2i}}\frac{\sin\theta_{2s3}^{\text{out}}}{n(\omega_{2i})}\right)^2\right]^{1/2}\right.$$

$$\left. - \left[1 - \left(\frac{\omega_{2s}}{\omega_{2i}}\frac{\sin\theta_{2s4}^{\text{out}}}{n(\omega_{2i})}\right)^2\right]^{1/2}\right\}; \quad (20)$$

angles $\theta_{isj}^{\text{out}}$ correspond to the ordinate θ_s^{out} of the point in Fig. 2a; $n(\omega)$ is the refractive index of the nonlinear medium at the corresponding frequency; and the subscript $i = 1, 2$ numbers the constant-frequency lines.

3. Experiment

Fig. 3 shows the scheme of experimental observation of three-photon interference. We used two variants of this scheme. In the first variant, two $LiNbO_3$ crystals of thickness 0.7 mm were placed in tandem with respect to the pump wave vector. In the second variant, one $LiNbO_3$ crystal of thickness 0.7 mm was used. The image of the first crystal in a broadband metal mirror played the role of the second crystal. The pump beam reflected from the mirror (together with the signal and idle beams) was incident on the crystal a second time. The resulting frequency-angular distribution of the scattering radiation intensity at the output of this system was determined by phase incursions at frequencies ω_p, ω_s, and ω_i in the gap between the crystal and mirror, according to expressions (3)–(5).
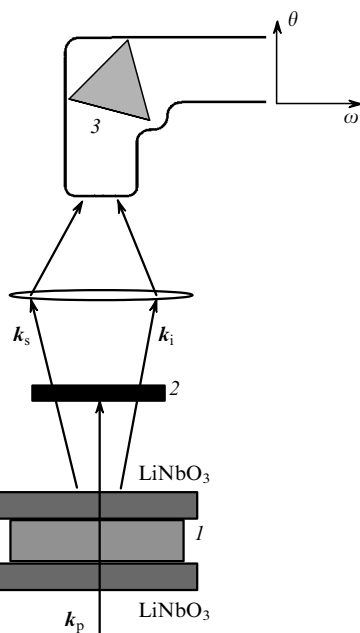

Figure 3. Scheme of the experimental observation of three-photon interference: (1) linear dispersion gap; (2) interference filter; (3) spectrograph.

Because reflection of light from a mirror is accompanied by the phase shift by π, the system consisting of the crystal and mirror separated by a layer of the dispersion medium of width $L'/2$ is equivalent to two nonlinear crystals with oppositely oriented polar axes separated by the gap L' [3, 4].

The pumping was performed with a 3-W, 488-nm argon laser (the laser beam divergence was 2×10^{-4} rad). The scattered emission was detected photographically. Both variants of measuring the refractive index were tested using the simplest linear media whose dispersion dependences are known with high accuracy and which do not absorb in the spectral regions of interest for us (air and crystalline quartz).

The refractive index of the linear medium (n_i' or n_s') was calculated from experimental interference spectra by expressions (6) and (7) assuming that the refractive index at another frequency is known. The refractive indices n_i' and n_s' obtained in this way differed from their tabulated values in the second decimal place.

The refractive indices at the frequencies of signal and idle waves were determined simultaneously from the same spectra using expressions (13) and (14). The measurements were performed in accordance with the variant given by expressions (13) and (14) on two constant-frequency lines in the region of the signal radiation with frequencies corresponding to $\lambda_{1s} = 585.2$ nm and $\lambda_{2s} = 588.2$ nm. The tabulated refractive indices of crystalline quartz for the ordinary and extraordinary waves at these frequencies differ in the fourth decimal place ($n_{1s}^o = 1.54440$ and $n_{2s}^o = 1.54427$). Therefore, the above assumption that refraction indices of the linear medium are constant within the spectral region under study is well justified. The calculated values of n_i' and n_s' differed from their tabulated values in the second decimal place.

The accuracy of measurements in both variants was limited by the error in measurements of the angular co-ordinates of the maxima (minima) of the interference pattern. This error was determined first of all by the angular resolution of the optical system, which was $0.025°$ ($1.5'$) in our experiments.

Note that, by varying the angular co-ordinates of the points being calculated within the measurement error, one can substantially improve the accuracy of measurements of refractive indices up to the fourth decimal place. Here, we mean that the measurement error is determined not only by the error in direct measurements of angular distances but also by a finite angular width of the maxima and minima of the interference pattern. One of the possible ways of reducing the errors related to angular measurements can be photoelectronic detection of the spectra.

In conclusion, we consider the consecutive procedure of measuring refractive indices of unknown substances. Liquids and gases are more convenient for such measurements because they do not require a special optical treatment and can be directly studied in cells whose input and output windows represent nonlinear crystals. Solids should be prepared in the form of polished plane-parallel plates a few millimetres in thickness. The visual analysis of the SPS spectrum immediately provides the answer to the question whether a substance under study is transparent in the spectral region of interest to us (we mean here the transparency region of a $LiNbO_3$ crystal between 0.4 and 5 μm). If the interference pattern is observed, then the substance is transparent at all three frequencies ω_s, ω_p, and ω_i. Weak absorption, as shown in Ref. [6], does not affect the frequency-angular dependence of the maxima (minima) of the interference pattern and only

broadens them, thereby reducing the accuracy of measurements.

Note that intense and narrow resonances of the dielectric constant are directly manifested as 'hooks' in the interference spectra[*] [3]. The use of the second algorithm near these spectral regions is limited because the refractive index is no longer constant at small frequency shifts. However, the second algorithm can be applied where the frequency-angular dependence of the interference maxima (minima) is monotonic. This algorithm allows one to estimate the refractive index of the intermediate layer simultaneously for different pairs of frequencies (ω_s, ω_i) and, hence, to obtain the dispersion law over the entire transparency region of the nonlinear LiNbO$_3$ crystal. The obvious advantage of this method is that it does not require *a priori* information on the optical properties of the substance under study.

If the dispersion of the refractive index of the substance under study is known in the visible range, the first algorithm can be used. In this case, the accuracy of measuring the dispersion of the refractive index in the IR region will be somewhat better than for the second algorithm.

We would like to emphasise that the methods for determining the dispersion law discussed above should not be considered as precision. We have proposed them first of all to demonstrate that the fundamental phenomenon of quantum interference, which forms the basis of the both algorithms, also has some applied spectroscopic aspects. At the same time, it is possible that these methods, being quite simple and requiring only minimal initial information, will be used along with other known methods.

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[*]D S Rozhdestvenskii has observed similar intensity distributions in a linear interferometer, which was later called by his name [14].