Nonlinear diffraction in the parametric scattering of light

A. L. Aleksandrovskiĭ, G. Kh. Kitaeva, S. P. Kulik, and A. N. Penin

University of Moscow

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When the quadratic susceptibility of a medium is spatially modulated, parametric scattering spectra of light convey information about the layered structure. Parametric scattering spectra have been observed for the first time in the presence of nonlinear diffraction. We have computed the period and orientation of the regular domain structure in a polydomain crystal of barium-sodium niobate, as well as the dispersion of the infrared refractive index of the crystal. We discuss the possibility of using parametric scattering spectroscopy to investigate nonlinear layered media.

A layered structure, which results in the periodic modulation of optical and nonlinear optical parameters, changes electromagnetic propagation and scattering processes significantly, as compared with these processes in uniform media. As early as 1962, a description had been given of the effect of periodic spatial modulation of the nonlinear susceptibility of a medium on a parametric process, being manifested by a change in locking direction.[1] Subsequently, this phenomenon, which has been called[2] nonlinear diffraction of light, has been used to ensure spatial coherence in sum- and difference-frequency harmonic generators,[3] as well as to study the layered medium itself and the interface between layers, for example in the case of the domain structure of ferroelectrics.[4] In the present paper, we report on the first experimental observations of three-photon parametric scattering (PS) of light in a layered medium with a periodically modulated quadratic susceptibility.

The spatial distribution of the effective magnitude χ_{eff} of the quadratic susceptibilty (the contraction of the tensor χ with the polarization unit vectors of the waves participating in the process) in a layered medium can be expressed in the form

$$\chi_{eff}(r)=\sum_{m=-\infty}^{\infty}\chi_m\exp(i\mathbf{q}_m\mathbf{r}). \tag{1}$$

Here, the expansion coefficient χ_m (the amplitudes of the spatial harmonics of the quadratic susceptibility) take the form

$$\chi_m\equiv(2l)^{-1}\int_{-l}^{l}\chi(\mathbf{r})\exp(-i\mathbf{q}_m\mathbf{r})\,d\mathbf{r}, \tag{2}$$

where l is the thickness of the layer, $\mathbf{q}_m=\mathbf{n}m\pi/l$ is a wave vector of the reciprocal lattice (superlattice), $\mathbf{n}$ is a unit vector perpendicular to the layers, and m is an integer.

A periodic modulation of the quadratic susceptibility, which determine the scattered radiation intensity, changes the form of the spatial matching condition from $\mathbf{k}_1=\mathbf{k}_3-\mathbf{k}_2$ ($\mathbf{k}_1$, $\mathbf{k}_2$, $\mathbf{k}_3$ are the wave vectors of the signal, polariton, and pump waves respectively) for a homogeneous medium to

$$\mathbf{k}_1=\mathbf{k}_3-\mathbf{k}_2+\mathbf{q}_m \tag{3}$$

for a layered medium. The temporal matching condition $\omega_1=\omega_3-\omega_2$ ($\omega_1,\omega_2,\omega_3$ are the signal, polariton, and pump frequencies, respectively) remains the same. By definition, the signal wave lies in the range $\omega_3/2\leqslant\omega_1<\omega_3$, with the polariton wave in the range $0<\omega_2<\omega_3/2$.

The crystals used in our experiments as a layered medium with periodic modulation of the magnitude of $\chi_{\text{eff}}(\mathbf{r})$ were barium-sodium niobate (BSN) $Ba_2NaNb_5O_{15}$, which was grown in a manner[5] that produced growth layers, forming a structure with periodically varying composition (stoichiometry). The planes of the growth layers were oriented almost perpendicular to the second-order crystallographic Z axis. In the ferroelectric phase ($T<585$ °C), a domain system was formed which reiterated the configuration of the growth layers, the spontaneous polarization of pairs of adjacent domains being oppositely directed. In going from domain to domain, the effective magnitude of the quadratic susceptibility also reversed sign. As a result, χ became a spatially modulated function of the coordinates. In the transition zones between domains, the value of χ passed through zero. Note that the attendant depth of modulation of the refractive index was no more than 10^{-3}–10^{-5}.

In Fig. 1, we present a PS spectrum on the upper polariton branch in a layered BSN crystal. The wavelength of the pump was 457.9 nm, polarized along the crystal Z axis, and the signal and polariton waves were polarized perpendicular to the Z axis. In contrast to the spectrum of a homogeneous crystal, comprising a single closed "ellipse," the spectrum of the layered BSN consists of three closed ellipses. A limited portion of a fourth can also be seen. The falloff in intensity of the scattered radiation at wavelenghts shorter than 500 nm is related to the linearization of the crystal as the polariton frequency approaches the intrinsic vibration frequencies of the lattice.[6] The low-intensity center ellipse, like the ellipse in a homogeneous crystal, is symmetric about the direction of pump propagation (the line of zero scattering angle),

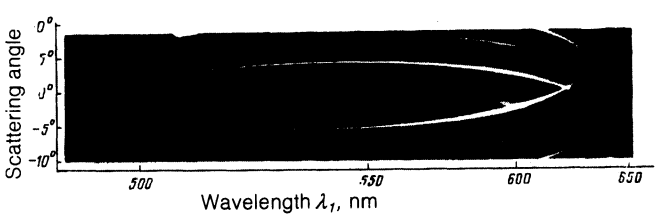

FIG. 1.

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while the two others, characterized by much higher intensity, are shifted in opposing directions.

The spectrum shown in Fig. 1 was obtained under conditions in which there was no linear diffraction of the pump wave, while linear diffraction of the signal wave, for which the Bragg condition was valid in a limited range of frequencies and angles, resulted in the appearance of part of the fourth ellipse.

Nonlinear diffraction is responsible for the appearance of the three closed curves in the spectrum of the layered medium, instead of the one which appears in the PS spectrum of a homogeneous crystal. One indication of this is the consistency of measurements of the thickness and orientation of the layers based on the relative angular displacements of the curves with frequency, assuming that the behavior of the curves follows the spatial locking conditions (3) for $m = -1,0,1$. According to these measurements, the mean value of the thickness l is 6.05 μm, and the variation in thickness measured at different frequencies is less than the measurement error, ± 0.05 μm. The mean value of the angle α between the layers and the front face of the sample is 89.3°, with a variation at different frequencies less than the error of $\pm 0.02°$. Thus, the angular behavior of the unshifted ellipse with frequency satisfies condition (3) if the assumption is made that $\mathbf{q}_m = 0$, and the ellipse is produced by zeroth-order diffraction scattering, while the behavior of the two shifted ellipses satisfies the spatial locking condition (3) for nonlinear diffraction with $m = \pm 1$ (first-order diffraction scattering).

The validity of our interpretation of the observed spectra is also confirmed by measurements of the dispersion of the infrared refractive index of the crystal, $n_2(\omega_2)$. There is good agreement (within the measurement error $\Delta n_2 = \pm 0.002$) among the dispersion curves for $n_2(\omega_2)$ obtained both from the behavior of the unshifted ellipse, and independently from the relative displacement of the first-order diffraction scattering curves (Fig. 2). The spatial locking condition (3) with $m = 0$ is based on calculations of n_2 in terms of the behavior of the unshifted ellipse, and the system of equations (3) with $m = \pm 1$ is based on the behavior of the displaced curves. This system of equations makes it possible simultaneously to determine the parameters of the layered structure, namely the values of the vectors $\mathbf{q}_1$ and $\mathbf{q}_{-1}$ (i.e., the quantities l and α), and the magnitude of the wave vector $\mathbf{k}_2$ (and thus n_2). It should be noted that the period of the superlattice, the orientation of the layers, and their planarity vary over the crystal volume, which results in a variation of the PS spectrum observed in different parts of the sample. Both the position and the shape of the curves due to first-order (and, in principle, higher-order) diffraction change, while the zeroth-order scattering curve only changes in intensity. Figure 2 demonstrates the good agreement between the dispersion curves for $n_2(\omega_2)$ computed from the relative displacement of first-order diffraction scattering in parts of the crystal with differing layered structures.

Analysis of the angular behavior of the curves with frequency, and of the number and mutual displacement of the curves which make up the PS spectrum of the layered medium, makes it possible to determine the spectrum of the reciprocal vectors of the superlattice. The relative scattering intensity in the various diffraction orders at a fixed set of frequencies conveys information about the amplitude of spatial harmonics of χ_m. However, to completely reconstruct the spatial distribution of the quadratic polarizability, information about the signs of the harmonics of χ_m is needed.

Information about the distribution of $\chi(\mathbf{r})$ in the crystal allows us, in turn, to judge the structure of the layers, their thickness and orientation, and the nature of the transition boundaries between domains. The following conclusions can be drawn from the spectrum in Fig. 1. The low zeroth-order scattering intensity and the absence of higher-order (with $|m| > 1$) diffraction scattering (as already noted, the partial fourth curve is due to linear diffraction of the signal wave) are indicative of an expansion of χ in the spectrum and only one harmonic, the first. The almost harmonic behavior of the modulation of χ attests to the large width of the transition region between layers, being comparable to the width of the layers themselves. The low intensity of zeroth-order scattering is indicative of the smallness of the constant component of the effective nonlinear susceptibility χ_0, and the approximate equality of the overall volumes of domains with opposite polarizations (the highly unpolar nature of the sample).

Figure 3 shows the spectrum of a sample with a domain layered structure different from the one previously considered. The angular displacement of the individual ellipses in this spectrum is much smaller, and the thickness of the crystal layers is consequently larger. The larger number of ellipses, or in other words, the scattering in more diffraction orders, indicates that the spectrum of spatial harmonics χ_m is richer than in the first case, while the spatial modulation function for $\chi(\mathbf{r})$ is closer to a random walk. The transition regions in this case must be considerably more narrow than the layers.

The fact that the superlattice is nonideal, which leads to variations in the thickness, direction, and polarity of the lay-

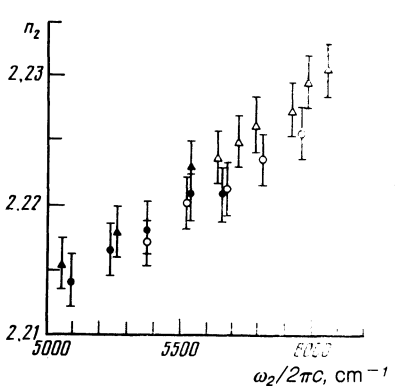

FIG. 2. Dispersion of the refractive index of a BSN crystal as determined from the PS spectrum of Fig. 1 (points marked ● come from the angular behavior of the center ellipse with frequency, and ○ denotes the two oppositely-displaced ellipses; the parameters of the layered structure are $l = 6.05$ μm, $\alpha = 89.3°$), and from the PS spectrum in part of a crystal with layered structure parameters $l = 9.1$ μm, $\alpha = 87.7°$ (where ▲ denotes the central ellipse and △ the two displaced ellipses).

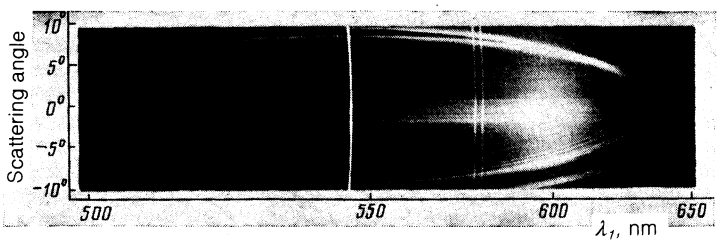

FIG. 3.

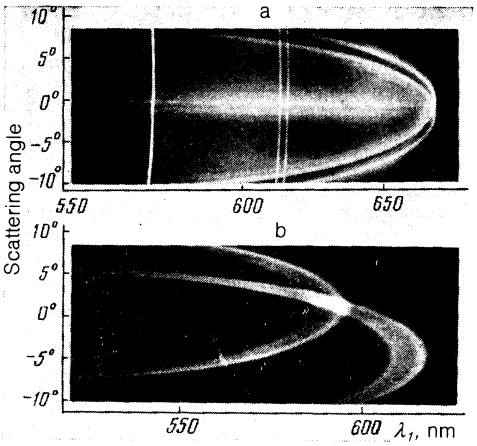

FIG. 4.

ers over small distances comparable to the size of the regions producing the measured scattering, results in an increase in the spectral and angular width of the curves. Here the various types of non-ideality of the superlattice variously affect the variation in the frequency behavior of the angular scattering, which enables one to assess the nature of the imperfections on the basis of the PS spectra. Figure 4a shows the PS spectrum of a sample with layers of variable thickness, which keep their planarity and overall direction. The constancy of the orientation of the layers as the thickness changes results in a stratification of the curves by angle if the pump wave propagates along the layers. The spectrum in Fig. 4b cannot be accounted for by a one-dimensional layered model. The major feature of that spectrum is the broadening of the curves in frequency, which is apparently related to the two-dimensionality of the superlattice.

The results we have described, based on observation of parametric scattering of light in a medium with periodically modulated nonlinear susceptibility, enable us to verify the following. Parametric scattering spectra under nonlinear diffraction conditions possess high sensitivity to the structure of the scattering medium and to the form of the modulating function. The large size of the spectral region taken in by parametric scattering of light and the high sensitivity to changes in spatial locking conditions make PS a simple and informative method of determining the parameters of nonlinear superlattices. It is possible to realize conditions in the PS process in which the wavelength of one of the participating waves is comparable to or larger than the characteristic size of the periodic inhomogeneity in the crystal. We also note the possibility of using PS to investigate the effects of stratification of the structure on the intrinsic vibrations of the crystalline lattice (optical phonons) and on all processes in which they interact with photons or with each other.

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