Synchronous conversion of radiation frequency under the conditions of the Bragg diffraction

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We determined new synchronisms in the course of second harmonic generation (SHG) under conditions of Bragg diffraction in periodic structures, which yield shortwave radiation with continuously-tunable frequency.

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1. The existing method of nonlinear conversion of radiation frequency in crystals in characterized by a shortwave boundary ($\sim$230 nm) which is determined by insufficient birefringence and transparency of the nonlinear materials in question.[1]

It is known, that in a medium with a periodically-modulated linear susceptibility $\chi(\mathbf{r})$ there occur near the Bragg condition dynamic effects and there exist two values of the effective index of refraction (EIR), each corresponding to a certain type of field.[2] This suggests the possibility of a synchronous wave interaction near the Bragg condition. From example, in the process of SHG, the EIR for the second harmonic field of the first type—somewhat away from the Bragg condition—may be equal to the EIR of the fundamental wave field of the second type. Under this condition, the SHG process should proceed with the highest intensity.

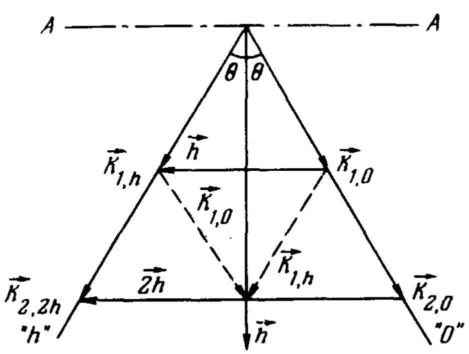

FIG. 1.

2. In a medium with an ideally-periodic structure the linear $\chi(\mathbf{r})$ and quadratic $\beta(\mathbf{r})$ susceptiblities may be expanded in a Fourier series in terms of inverse lattice vectors $\mathbf{h}$.

$$\chi_j(\mathbf{r}) = \sum_{m=0} \chi_{j;mh} e^{im\mathbf{hr}}, \qquad \beta(\mathbf{r}) = \sum_{m=0} \beta_{mh} e^{im\mathbf{hr}}$$

where $j = 1,2$ is the number of a harmonic $\omega_j = j\omega$ and m is an integer.

Let the Bragg condition be satisfied approximately and a two-wave approximation hold for both frequencies; **h** is reflection vector for the fundamental wave and 2**h**—for a harmonic. Deviation from the Bragg condition will be characterized as follows:

$$a = (k_h^2 - k_o^2)c^2/\omega^2 \ ; \ k_o = \bar{n}_o\,\omega/c \ ; \ \mathbf{k}_h = \mathbf{k}_o + \mathbf{h} \ ; \ \bar{n}_o^2 = 1 + (\chi_{1;0} + \chi_{2;0})/2 \ ;$$

$$a = \bar{n}_o^2\,(2\,\delta\theta \sin 2\theta - 4\,\frac{\delta\omega}{\omega}\sin^2\theta),$$

where $\delta\theta = \theta - \theta_B$ and $\delta\omega = \omega - \omega_B$ are the angular and frequency deviation from θ_B and ω_B, respectively; both θ_B and ω_B satisfy the Bragg condition $a\,(\theta_B,\omega_B) = 0$ and are related to each other as follows: $\sin\theta_B = mhc/2\omega_B$. Vector $\mathbf{k}_0$ is directed along "0", i.e., it coincides with a direction of a wave vector of a wave incident on a sample $\mathbf{k}_{1;0}$ (Fig. 1).

It can be shown—on the basis of a solution of Takagi's equations,[2,3] where nonlinear polarization of a medium at the second harmonic frequency is introduced—that under the conditions of Bragg diffraction with respect to a symmetrical Laue scheme (see Fig. 1), there exist, for normal frequency dispersion and $\Delta > |\chi_1 - \chi_2|$ two conditions of synchronism:

$$\text{I.}\quad \gamma_1 + \gamma_2 = \Delta \quad \text{or} \quad a_{\mathrm{CI}}^2 = \left(\Delta + \frac{\chi_1^2 - \chi_2^2}{\Delta}\right)^2 - 4\chi_1^2\,, \quad \Delta \geqslant \chi_1 + \chi_2\,,$$

$$\text{II.}\quad \gamma_2 = \Delta \quad \text{or} \quad a_{\mathrm{CII}}^2 = 4(\Delta^2 - \chi_2^2)\,, \quad \Delta \geqslant \chi_2\,,$$

where $\Delta = \chi_{2;0} - \chi_{1;0}$ is the dispersion of the medium.

The conditions of synchronism I and II may be expressed in terms of EIR in the directions "0" and "h":

$$n_{j;0}^{(\pm)} = (1 + \chi_{j;0} - a/2 \pm \gamma_j)^{1/2}\ ; \quad n_{j;h} = (1 + \chi_{j;0} + a/2 \pm \gamma_j)^{1/2}$$

in the following manner:

$$\text{I.}\ n_2^{(-)} = n_1^{(+)}\ ; \quad \text{II.}\ n_1^{(+)} + n_1^{(-)} = n_2^{(-)}.$$

In the case of synchronism I, the upper dispersion curve (hyperbola) for EIR at a frequency ω intersects the lower hyperbola at a frequency 2ω (Fig. 2).

3. Synchronous SHG occurs colinearly and concurrently along the directions "0" and "h" and the inverse lattice vector does not enter, as a rule, in the synchronism condition. Calculations show that in a region $a > 0(k_h > k_0)$, an effective harmonic wave is generated in the "h" direction, and in a region $a < 0(k_h < k_0)$—along "0". If $\chi_1 = 0$, $\chi_2 \neq 0$, i.e., only the second harmonic radiation is Bragg-diffracted while the fundamental is not, the effectiveness of synchronism II becomes trivial and it fails to manifest itself; synchronism I occurs only at $a_{\mathrm{CI}} = (\Delta^2 - \chi_2^2)/\Delta > 0$.

If the modulation of linear susceptibility is very small $\chi_j \ll \Delta$, the effective nonlinearity of all the synchronisms is also small, with the exception of the synchronism I ($\alpha_{CI} = \Delta$) in the direction "h" whose effective nonlinearity in this case is β_{2h}. Moreover, nonlinear diffraction synchronism is achieved $\mathbf{k}_{2;2h} = 2\mathbf{k}_{1;0} + 2\mathbf{h}$.

In principle, the case $0 < \Delta < |\chi_2 - \chi_1|$ is possible, for which other synchronisms occur: III. $\gamma_1 - \gamma_2 = \Delta$, $n_1^{(+)} = n_2^{(+)}$ for $\Delta < \chi_1 - \chi_2$; and IV. $\gamma_2 = -\Delta$, $n_1^{(+)} + n_1^{(-)} = 2n_2^{(+)}$ for $\Delta < \chi_2 - \chi_1$.

In the case of anomalous dispersion $\Delta < 0$, the following synchronisms are also possible:

$$\text{V. } \gamma_1 + \gamma_2 = -\Delta, \; n_1^{(-)} = n_2^{(+)}; \quad \text{VI. } \gamma_2 = -\Delta, \; n_1^{(+)} + n_1^{(-)} = 2n_2^{(+)}.$$

In the case of Bragg diffraction, we may write out similar conditions for synchronism: $\alpha_{CI} = (\chi_1^2 - \chi_2^2)/\Delta$; $\alpha_{CII} = \Delta \pm 2\chi_2$, the latter corresponding to the boundaries of a region of reflection of a harmonic.

In the interval $\theta = \pi/2$, $\chi_1 = 0$, the Bragg synchronism I degenerates into a single-dimensional synchronism,[5,6] and the effectiveness of synchronism II becomes trivial. A critical transition to the single-dimensional case does not occur for the Laue case.

The new form of synchronism may be called "Fourier-synchronism." In contrast to sychronism based on the anisotropy, "Fourier-synchronism" may also be satisfied at high frequencies in the uv and x-ray regions. It may also be achieved for the parametric amplification of waves.

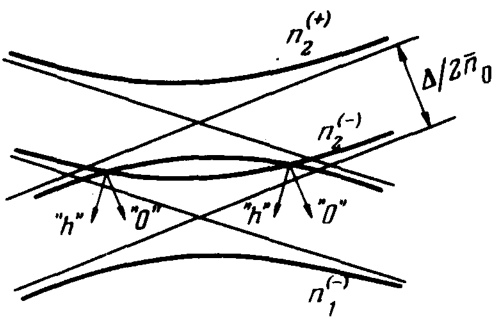

FIG. 2.

4. The "Fourier-synchronism" method described here yields high-frequency radition with a continuously-tunable frequency. Frequency tuning is based on the fact that fulfillment of the synchronism condition is contigent on the parameter a which, in turn, is a function of two variables: angular detuning $\delta\theta$ and frequency detuning $\delta\omega$. The magnitude of a for the synchronism is a_c and is determined by the values of χ_j and dispersion Δ which are constant for a given item. A tunable laser may be used as a source of fundamental radiation. By varying the angle of incidence, the frequency ω at which condition of synchronism is satisfied, may be changed. Variation of the angle θ in the $\Delta\theta$ interval results in a variation of the output frequency in the interval

$\Delta\omega/\omega \sim \Delta\theta \cot\theta$ that should correspond to the interval in which the frequency of the fundamental radiation source in retuned.

5. In order to implement the proposed method experimentally, periodic structures obtained by the molecular epitaxy method, may be used, with a period $d \sim 10^3$ Å. Among these are zeolite crystals with $d \sim 10$ Å and liquid crystals with $d \sim 100$–1000 Å.

6. "Fourier-synchronism" may be used for the conversion of frequency in any wavelength interval in an isotropic medium with a periodic structure ($\lambda \sim d$); for example, it may be used for the IR-region in the staggered layers of AlAs-GaAs crystals.[7]

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