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Nonlinear wave interactions in modern photonics

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One of the main factors impeding further progress in the field of application of met materials is significant energy losses due to the physical nature of exploited plasmon resonance, and their compensation is the most urgent problem to be addressed by the modern science of met materials. That is why this paper studies the parametric interaction of electromagnetic waves and, in particular, the process of generation and amplification of the second harmonic generation in met materials with the negative refractive index. It is found that the fundamental waves in the process of the second harmonic generation cannot exchange energy through the second harmonic wave at the non-collinear phase matching, and, thus, further consideration is required of optical rectification of the field in the nonlinear met materials.

Key words: met material, fundamental wave, second harmonic generation.
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1 Introduction

Recent advances in the technology of structured materials made it possible to create new materials with unusual physical properties, which are not encountered in nature. The best known example of such materials is the so-called met materials that are nanocomposites with the negative refractive index [1,2].Such unique physical characteristics can be obtained by using structured materials, which include those based on metal-insulator [3,4], metal-organic [5,6] biological [7], and other media.

On the one hand the linear optical properties of met materials are well studied at the moment. On the other hand the invention of powerful sources of coherent radiation, associated with the creation of lasers, led to a new field of optics, nonlinear optics, which gave a direct impetus to the development of optoelectronic devices and information technologies. At present the formation of nonlinear optics of met materials is under way, which, along with effects similar to those of the classical nonlinear optics, discloses a number of phenomena that are unique for met materials. It should be noted that interest in the theory of nonlinear optical phenomena in met materials is heated by the problems of both fundamental science and potential applications in technology. Possible applications of met materials are still hampered by essential energy losses due to the plasmon resonance. Compensation of those losses is the most urgent problem to be addressed by the modern science of met materials. In this regard, this paper studies the parametric interaction of electromagnetic waves and, in particular, the process of generation and amplification of the second harmonic wave in met materials with the negative refractive index, with further focus on the effectiveness of frequency conversion and compensation of energy losses in met materials.

Second harmonic generation is a nonlinear optical process, in which electromagnetic waves with the same frequency interact with a nonlinear material to effectively generate electromagnetic wave with the doubled frequency. In the classical case, the second harmonic generation occurs in strongly nonlinear crystals with quadratic nonlinearity χ^2 [8,9]. It is well known that for an effective second-harmonic generation in ordinary matter the perfect phase matching $\Delta k = 2k_1 - k_2 = 0$ must be satisfied. In case of its violation a periodic exchange of energy between the fundamental wave and the second harmonic wave is observed.

2 Non-collinear second harmonic generation in met materials

Consider the process of the second harmonic generation in a met material with a negative refractive index. The phase matching is achieved by the interaction of two collinear waves of the fundamental frequency and the second harmonic wave, so that $\mathbf{k}_1^+ + \mathbf{k}_1^- + \mathbf{k}_2 = 0$, where $\mathbf{k}_1^\pm, \mathbf{k}_2$ denote the wave vectors corresponding to the pump waves and the second harmonic, see Fig.1. This situation is a generalization of the case of collinear second harmonic generation studied in [10].

In case of the non-collinear second-harmonic generation in a met material the pump wave and the second harmonic wave can be represented as follows:

$$E_1(t,\mathbf{r}) = e_1^{(+)}(t,x,z)\exp\left[-i\omega t + i\mathbf{k}_1\cdot\mathbf{r}\right] + \\ + e_1^{(-)}(t,x,z)\exp\left[-i\omega t + i\mathbf{k}_2\cdot\mathbf{r}\right], \\ E_2(t,\mathbf{r}) = e_2(t,x,z)\exp\left[-2i\omega t + i\mathbf{k}_3\cdot\mathbf{r}\right]. \quad (1)$$

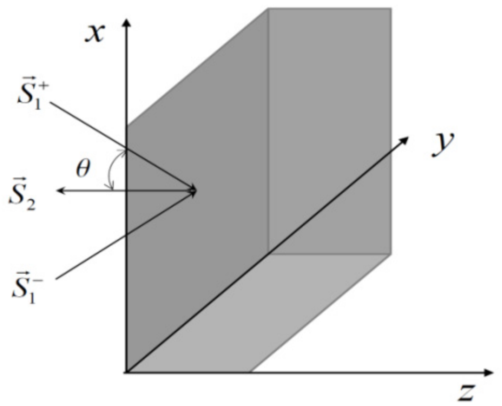


Figure 1 – Non-collinear SHG process. Two pump waves are incident at a certain angle to the normal of the met material surface. The directions of the respective energy fluxes are characterized by Poynting vectors $\mathbf{S}_1^\pm$. and $\mathbf{S}_2$, which is oppositely directed to the normal

It is assumed that at the fundamental frequency ω the refractive index of the metamaterial is negative, and it is positive at the second harmonic frequency 2ω. The second harmonic wave propagates oppositely to the normal of the metamaterial surface. The direction of the Poynting vector $\mathbf{S}_2$ of the second harmonic is opposite to the sum of the Poynting vector $\mathbf{S}_1^\pm$ of the pump waves, which is directed along the normal (see Fig. 1). That is, the second harmonic wave vector $\mathbf{k}_3$ is directed along the axis Z and the wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ of the incident waves lie in the XZ plane, so that

$$\mathbf{k}_1 = k_1\begin{pmatrix}\eta_x \\ -\eta_z\end{pmatrix},\ \mathbf{k}_2 = k_2\begin{pmatrix}-\eta_x \\ -\eta_z\end{pmatrix}, \quad (2)$$

where $\eta_x = \sin\theta$, $\eta_z = \cos\theta$, θ refer to the angle between vectors $\mathbf{k}_1$ and $\mathbf{k}_3$.

The set of equations describing the non-collinear synchronization at the second harmonic generation is written as follows:

$$(-\eta_z\partial_z + \eta_x\partial_x + \upsilon_1^{-1}\partial_t)E_1^{(+)} = i\gamma_1 E_1^{(-)*}\varepsilon_2 e^{i\Delta\mathbf{k}\cdot\mathbf{r}}, \\ (-\eta_z\partial_z - \eta_x\partial_x + \upsilon_1^{-1}\partial_t)E_1^{(-)} = i\gamma_1 E_1^{(+)*}E_2 e^{i\Delta\mathbf{k}\cdot\mathbf{r}}, \\ (\partial_z + \upsilon_2^{-1}\partial_t)E_2 = i\gamma_2 E_1^{(-)}E_1^{(+)}e^{-i\Delta\mathbf{k}\cdot\mathbf{r}}. \quad (3)$$

Here $E_1^{(\pm)}$ and E_2 designate the complex envelopes of the electric fields of the pump and second harmonic waves with the corresponding group velocities $\upsilon_{1,2}$, $\gamma_{1,2}$ stand for the coefficients of the nonlinear interaction.

The mismatch in the vector synchronism $\Delta\mathbf{k}$ is then obtained in the form:

$$\Delta k = k_z(2\omega) + 2k(\omega)\eta_z. \quad (4)$$

If $E_1^{(\pm)}$ and E_2 change slowly, they can be expressed as

$$E_1^{(\pm)}(x,z,t) = \Phi_1^{(\pm)}(x)A_1^{(\pm)}(z,t), \\ E_2(x,z,t) = \Phi_2(x)A_2(z,t), \quad (5)$$

and wave equations (3) can be rewritten as follows:

$$\Phi_2(x)\left(\frac{\partial}{\partial z} + \frac{1}{\upsilon_2}\frac{\partial}{\partial t}\right)A_2(z,t) = i\gamma_2\Phi_1^{(-)}(x)\Phi_1^{(+)}(x)A_1^{(-)}(z,t)A_1^{(+)}(z,t)e^{-i\Delta kz}. \quad (6)$$

The last equation can be transformed to

$$\frac{\left(\partial_z+(1/\upsilon_2)\partial_t\right)A_2(z,t)}{i\gamma_2 A_1^{(-)}(z,t)A_1^{(+)}(z,t)e^{-i\Delta kz}}=\frac{\Phi_1^{(-)}(x)\Phi_1^{(+)}(x)}{\Phi_2(x)}, \quad (7)$$

in which the right side of equation (7) depends on z, whereas the left function singly depends on x. This means that the left hand side of (7) is constant, that is:

$$\frac{\Phi_1^{(-)}(x)\Phi_1^{(+)}(x)}{\Phi_2(x)}=const. \quad (8)$$

The equations for the incident waves can be rewritten as follows:

$$\Phi_1^{(\pm)}(x)\left(-\eta_z\frac{\partial}{\partial z}+\frac{1}{\upsilon_1}\frac{\partial}{\partial t}\right)A_1^{(\pm)}(z,t)\pm$$
$$\pm\eta_x\frac{\partial\Phi_1^{(\pm)}}{\partial x}A_1^{(\pm)}(z,t)= \quad (9)$$
$$=i\gamma_1\Phi_1^{(\mp)*}(x)\Phi_2(x)A_2(z,t)A_1^{(\mp)*}(z,t)e^{+i\Delta kz}.$$

Dividing this expression by $\Phi_1^{(\pm)}$ gives rise to

$$\frac{\Phi_2(x)\Phi_1^{(\pm)*}(x)}{\Phi_1^{(\mp)}(x)}=m\left|\Phi_1^{(\mp)}(x)\right|^2. \quad (10)$$

Assume that $\left|\Phi_1^{(\mp)}(x)\right|^2=1$. Then, the equation for the incident wave is written as

$$\left(-\eta_z\frac{\partial}{\partial z}+\frac{1}{\upsilon_1}\frac{\partial}{\partial t}\right)A_1^{(\pm)}(z,t)\pm$$
$$\pm\eta_x\frac{1}{\Phi_1^{(\pm)}}\frac{\partial\Phi_1^{(\pm)}}{\partial x}A_1^{(\pm)}(z,t)= \quad (11)$$
$$=i\gamma_1 mA_2(z,t)A_1^{(\mp)*}(z,t)e^{+i\Delta kz}.$$

Separating the variables yields

$$\frac{1}{\Phi_1^{(\pm)}}\frac{\partial\Phi_1^{(\pm)}}{\partial x}=const^{(\pm)}, \quad (12)$$

which allows one to conclude that $\Phi_1^{(\pm)}=C^{(\pm)}e^{\pm const^{(\pm)}x}$.

Taking into account that $\left|\Phi_1^{(\mp)}(x)\right|^2=1$, one finds that $\Phi_1^{(\pm)}=e^{\pm il_1x}$, where l_1 are arbitrary constant.

Finally, taking into account all the substitutions, equation (11) can be rewritten as:

$$\left(-\eta_z\frac{\partial}{\partial z}+\frac{1}{\upsilon_1}\frac{\partial}{\partial t}\right)A_1^{(\pm)}(z,t)\pm$$
$$\pm il_1\eta_x A_1^{(\pm)}(z,t)= \quad (13)$$
$$=i\gamma_1 mA_2(z,t)A_1^{(\mp)*}(z,t)e^{+i\Delta kz}.$$

Using the phase shift in the substitution $A_1^{(\pm)}=A_1^{(\pm)}\exp(il_1\eta_x/\eta_z)$ the left hand side of equation (13) can be eliminated. Thus, the set of equations describing the interaction of three non-collinear waves in the nanocomposite medium, i.e. a metamaterial with the negative refractive index, is found as:

$$\left(-\eta_z\frac{\partial}{\partial z}+\frac{1}{\upsilon_1}\frac{\partial}{\partial t}\right)A_1^{(+)}(z,t)=i\gamma_1A_2(z,t)A_1^{(-)*}(z,t)e^{+i\Delta kz},$$
$$\left(-\eta_z\frac{\partial}{\partial z}+\frac{1}{\upsilon_1}\frac{\partial}{\partial t}\right)A_1^{(-)}(z,t)=i\gamma_1A_2(z,t)A_1^{(+)*}(z,t)e^{+i\Delta kz},$$
$$\left(\frac{\partial}{\partial z}+\frac{1}{\upsilon_2}\frac{\partial}{\partial t}\right)A_2(z,t)=i\gamma_2A_1^{(+)}(z,t)A_1^{(-)}(z,t)e^{-i\Delta kz}.$$
$$(14)$$

Consider the stationary case in which the set of equations (14) can be rewritten as follows:

$$-\eta_z\frac{\partial}{\partial z}A_1^{(+)}(z)=i\gamma_1a_2(z)A_1^{(-)*}(z),$$
$$-\eta_z\frac{\partial}{\partial z}A_1^{(-)}(z)=i\gamma_1a_2(z)A_1^{(+)*}(z), \quad (15)$$
$$\frac{\partial}{\partial z}a_2(z)-i\Delta ka_2(z)=i\gamma_2A_1^{(+)}(z)A_1^{(-)}(z).$$

On substituting $A_2(z)=a_2\exp(-i\Delta kz)$, $z/z_0=x$, $\Delta k=\Delta$, $\eta_z=\alpha$ gives rise to the following set of equations in dimensionless form:

$$-\frac{\partial}{\partial x}e_1^{(+)}(x)=ie_2(x)e_1^{(-)*}(x),$$
$$-\frac{\partial}{\partial x}e_1^{(-)}(x)=ie_2(x)e_1^{(+)*}(x), \quad (16)$$
$$\frac{\partial}{\partial x}e_2(x)-i\delta e_2(x)=ie_1^{(+)}(x)e_1^{(-)}(x).$$

The normalization of the amplitude of the interacting waves are defined herein as $A_1^{(\pm)}=(1/z_0)\sqrt{\alpha/\gamma_1\gamma_2}e_1^{(\pm)}$, $a_2=(\alpha\Delta/\gamma_1 z_0)e_2$ and $\delta=\Delta z_0$ .If the length of the sample is denoted as l, the boundary conditions in this case can be written as:

$$e_1^{(\pm)}(0)=e_{10}^{(\pm)}\exp(i\phi_{10}),\ e_2(l)=0. \quad (17)$$

Thus, the set of differential equations (16), describing non-collinear wave interaction at the SHG, should be solved together with boundary conditions (17), set at the opposite ends of the sample.

The set of equations (16) can be rewritten in terms of amplitudes and phases, by setting $e_1^{(\pm)}=u^{(\pm)}\exp(i\varphi_1^{(\pm)}),\ e_2=v\exp(i\varphi_2)$ and subsequent separation of the imaginary and real parts yields:

$$\partial_z u^+=vu^-\sin\theta,$$
$$\partial_z u^-=vu^+\sin\theta,$$
$$\partial_z v=u^+u^-\sin\theta, \quad (18)$$
$$\partial_z\theta=\left(vu^-/u^+ +vu^+/u^- +u^+u^-/v\right)\cos\theta-\delta,$$
$$u^\pm(0)=u_0^\pm,\ \text{v}(l)\text{=0},\ \theta(l)\text{=-}\pi/2,$$

where $\theta=\varphi_2-\varphi_1^+-\varphi_1^-$, $\varphi_1^\pm$ and φ_2 denote the phases of the pump and second harmonic waves, respectively.

The presence of a common factor $\sin\theta$ in the equations above means that the exchange of energy between the harmonics takes place in such a way that the fundamental waves both lose or gain some energy at the same time, that is the energy exchange between the fundamental waves u^- and u^+ through the second harmonic is impossible.

The set of equations (18) has three first integrals, among which are the Manley-Rowe relations:

$$\left(u^+\right)^2-v^2=m_1^2,$$
$$\left(u^-\right)^2-v^2=m_2^2, \quad (19)$$
$$\left(u^+\right)^2-\left(u^-\right)^2=m_1^2-m_2^2,$$

where $m_{1,2}=u_1^\pm(l)$.

The last equation in (18) can be easily integrated to give:

$$u^+u^+v\cos\theta-v^2\delta/2=m_3, \quad (20)$$

where m_3 is a constant .

It follows from the last expression that $m_3=0$, $F=\cos\theta=v\delta\Big/\left[2\sqrt{(v^2+m_1^2)(v^2+m_2^2)}\right]$. The maximum value of this function is achieved at the point $v_0=\sqrt{m_1m_2}$. Taking into account the inequality, $|\cos\Phi|\le 1$ allows one to conclude that, like in the collinear case, there are two regimes of the second harmonic generation. In case of $|\delta|\le\delta_{cr}=2(m_1+m_2)$, v can take any value, increasing indefinitely along the axis z . This in turn means that for the entire range energy $0\le z\le L$ the energy is transferred from the fundamental wave to the second harmonic. In case of $|\Delta|>\Delta_{cr}$ the domain of allowed values of v lies in the interval $0\le v\le v_0$, where

$$v_0=\sqrt{K-\sqrt{K^2-64m_1^2m_2^2}}\Big/2\sqrt{2},$$

$K=\delta^2-4(m_1^2+m_2^2)$. Thus, the exchange of energy between the fundamental wave and the second harmonic takes place along the sample, leading to spatial oscillations of the amplitudes of the interacting waves. In Figure 2, the curves of the function $F=\cos\theta$ are plotted for different values of the amplitude of the second harmonic which corresponds to the solid line with $|\delta|=2(m_1+m_2)$. For the lower curve $|\delta|<2(m_1+m_2)$ and in this case the amplitude of the second harmonic wave can take arbitrary values. If $|\delta| > 2(m_1 + m_2)$,there are forbidden bands for the values of the amplitudes of the second harmonic corresponding to the upper curves. This is the so-called supercritical regime of the second harmonic generation.

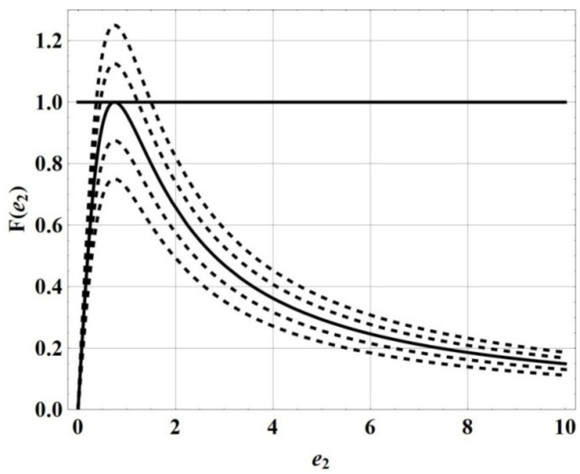


Figure 2 – The dependence of the function $F = cos\theta$ on the amplitude of the second harmonic at m_1 – 0.8, m_2 0.7

Using the conservation laws (19) together with (18) the following equation for the intensity of the second harmonic is derived for $m_3 = 0$:

$$\frac{d}{dz}v^2 = -2\sqrt{v^2(m_1^2+v^2)(m_2^2+v^2)-(\frac{v^2}{2}\delta)^2}. \quad (21)$$

Let the intensity of the second harmonic be such that $v^2 = P$ and, then, the last equation is rewritten in the form

$$\frac{\partial P}{\partial z} = -\sqrt{4P^3 + (4(m_1^2+m_2^2)-\delta^2)P^2 + 4m_1^2m_2^2P}, \quad (22)$$

and its solution is found as:

$$-\int\frac{dP}{\sqrt{4P^3 + (4(m_1^2+m_2^2)-\delta^2)P^2 + 4m_1^2m_2^2P}} = \int dz. \quad (23)$$

The integral on the left side is elliptical, and the solution of (23) can be expressed by using the Weierstrass function. Thus, the solution of (21) can be written as:

$$z - l = -\int\frac{ds}{\sqrt{4s^3 - g_2 s - g_3}}. \quad (24)$$

Here $P = P_0 + 6f'(P_0)/(24s - f''(P_0))$, where P_0 is one of the roots of the polynomial $f(P) = 4P^3 + (-K)P^2 + 4m_1^2m_2^2P$, $K = \delta^2 - 4(m_1^2 + m_2^2)$.

The polynomial under the square root of equation (24) has the following roots:

$$P_a = 0, \quad P_{b,c} = \frac{1}{8}(K \mp \sqrt{(K^2 - 64m_1^2m_2^2}). \quad (25)$$

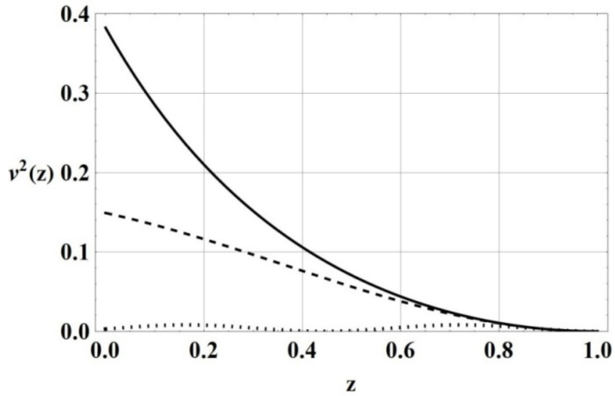
The invariants of the Weierstrass function g_2 and g_3 are, thus, equal to:

$$g_2 = \frac{1}{12}(-K)^2 - 4m_1^2m_2^2, \quad g_3 = \frac{1}{3}m_1^2m_2^2(-K) - \frac{1}{216}(-K)^3. \quad (26)$$

A special feature of this case is that at perfect phase matching of $\delta = 0$, the solution is expressed in terms of Jacobi elliptic functions :

$$P = v^2 = -m_1^2 sn^2[im_2(z-l), \frac{m_1^2}{m_2^2}]. \quad (27)$$

Solutions for the intensities of the remaining two pump waves are easily found using expression (19).It should be noted that in the collinear interaction of waves solutions are expressed in terms of hyperbolic functions at $\delta = 0$.In the case of $|\delta| > \delta_{cr} = 2(m_1 + m_2)$ the second harmonic wave $v$starts to exchange energy with the pumping waves u^+ and u^-. Figure 3 shows the intensity of the second harmonic wave from as a function of the coordinate z. The solid curve corresponds to the perfect phase matching at which the energy is permanently transferred to the second harmonic along the entire sample. The dashed curve corresponds to the critical value of the phase mismatch when $\delta > \delta_{cr}$ and the energy exchange between the pump waves and second harmonic takes place along the sample, i.e. the amplitude of the second harmonic turns a periodic function of the coordinate. Thus, in contrast to the classical case, the effective frequency conversion in metamaterials is possible for the whole range of the phase mismatches both in collinear or non-collinear matching which is quite an attractive feature in sense of possible applications.



$u_{10}^{+} = 1$, $u_{10}^{-} = 0.9$, $l = 1$

Solid line – $\delta < \delta_{cr}$, dashed line – $\delta < \delta_{cr}$, dotted line – $\delta < \delta_{cr}$

Figure 3 – Spatial profiles of the second harmonic wave along the sample at different values of δ

In the ideal phase matching solution (18) for the wave intensity is written as follows:

$$v(z)^2 = -m_1^2 \,\mathrm{sn}^2\left[i m_2 (z - l), \frac{m_1^2}{m_2^2} \right],$$

$$\left(u^{+}(z)\right)^2 = v(z)^2 + m_1^2,\ \left(u^{-}(z)\right)^2 = = v(z)^2 + m_2^2. \quad (28)$$

Note that expressions (28) are implicit solutions of (18), since they contain undefined parameters such as m_1 and m_2, which are corresponding values of the amplitudes of the pump waves at the right end of the sample $u^{+}(l)$, $u^{-}(l)$. To determine the dependence of $m_{1,2}$ on the amplitude of the input value of $u^{\pm}(0)$ the following transcendental equations must be solved:

$$\left(u^{\pm}(0)\right)^2 = v(0)^2 + m_{1,2}^2. \quad (29)$$

The results of the numerical solution of transcendental equations (29) are shown in Figures 4-5. Figure 4 corresponds to the case of ideal phase matching, in which the curve of the amplitude of the incident pump wave at the right end of the sample as a function of its value at the left end has several branches. The physical meaning can only be prescribed to the lower branch, whereas for the parameters of the upper branches the fields within the sample can take infinite values. It follows from the analysis of the behavior of the lower branch that the second harmonic reaches saturation, which corresponds to the total transformation of the energy of the fundamental waves into the energy of the second harmonic.

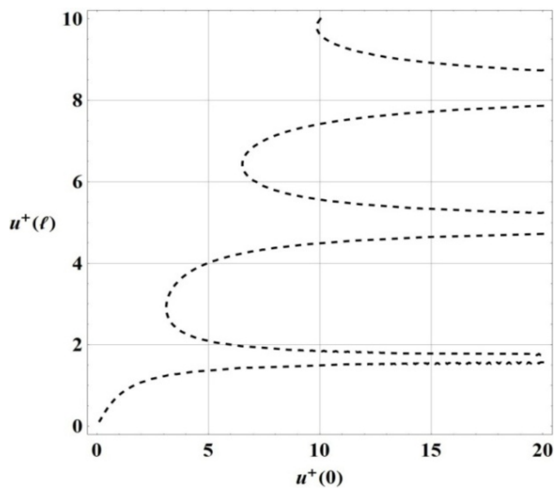
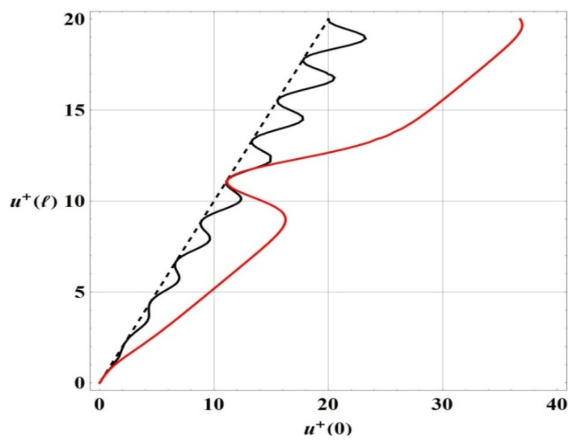


Figure 4 – The dependence of the amplitude of the pump wave $u^{+}(l) = m_1$ on $u^{+}(0)$



Red curve: $|\delta| \sim \delta_{cr} = (m_1 + m_2)$,
black curve: $|\delta| \sim 2.2(m_1 + m_2)$,
dashed curve: $|\delta| \sim 20(m_1 + m_2)$

Figure 5– The dependence of the amplitude of the pump wave $u^{+}(l) = m_1$ on $u^{+}(0)$

3 Conclusions

The curves in Figure 5 show the intensity of the fundamental wave at the left end of the sample $u^+(0)$ as a function of its value $u^+(l)$ at the right end in the supercritical regime of SHG. The red curve corresponds to the case of $|\delta| \sim \delta_{cr} = 2(m_1 + m_2)$, the black curve is drawn for $|\delta| \sim 2.2(m_1 + m_2)$, and for the dashed curve $|\delta| \sim 20(m_1 + m_2)$. For quite large values of the phase mismatch the dependence is almost linear, which actually prevents the SHG since the energy transfer from the fundamental wave to the second harmonic does not take place, and the intensity of the incident fundamental waves remains unchanged while passing along the sample. For values of the phase mismatch not higher than the critical one, rapid oscillations occur due to the periodic exchange of energy between the fundamental wave and the second harmonic.

In a case of non-collinear phase matching it is found for the second-harmonic generation in a metamaterial with the negative refractive index that the fundamental waves simultaneously either lose or gain energy, i.e. the energy exchange between them through the second harmonic turns impossible. Nevertheless, the exchange of energy between the pump waves is possible the constant field appearing due to the optical rectification, which is always present in the process of the second harmonic generation.

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