Parametric generation of high frequency coherent light in negative index materials and materials with strong anomalous dispersion

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We demonstrate the possibility of generation of coherent radiation with tunable frequencies higher than the frequency of the driving field $\nu_d$ in a nonlinear medium utilizing the difference combination resonance that occurs when $\nu_d$ matches the difference of the frequencies of the two generated fields $\omega_1$ and $\omega_2$. We find that such a resonance can appear in materials which have opposite signs of refractive index at $\omega_1$ and $\omega_2$. It can also occur in positive refractive index materials with strong anomalous dispersion if at one of the generated frequencies the group and phase velocities are opposite to each other. We show that the light amplification mechanism is equivalent to a combination resonance in a system of two coupled parametric oscillators with the opposite sign of masses. Such a mechanism holds promise for a new kind of light source that emits coherent radiation of tunable wavelengths by an optical parametric amplification process with the frequency higher than $\nu_d$.

I. INTRODUCTION

The invention of the laser in 1960’s facilitated exploration of various nonlinear phenomena in optics, such as harmonic generation\textsuperscript{1}, multi photon absorption\textsuperscript{2}, wave mixing\textsuperscript{3}, optical Kerr effect\textsuperscript{4}, stimulated Raman scattering\textsuperscript{5} and so on. Today nonlinear optics plays an important role in the developing of new light sources\textsuperscript{6}, nonlinear spectroscopy\textsuperscript{7}, multiphoton microscopy\textsuperscript{8}, all-optical signal processing\textsuperscript{9}, quantum information\textsuperscript{10} and many other applications\textsuperscript{11}.

Optical parametric oscillation/amplification (OPO/OPA)\textsuperscript{12} which transfers energy from the driving field to the signal and idler waves is an example of parametric processes caused by the nonlinear light-matter interaction. OPA led to development of tunable radiation sources throughout the infrared, visible, and ultraviolet spectral regions\textsuperscript{16}. Such sources yield amplification of coherent radiation at lower-frequencies $\omega_1$ and $\omega_2$ by pumping nonlinear optical crystals with intense laser light at the sum frequency $\nu_d$.

$$\nu_d = \omega_1 + \omega_2.$$  \hspace{1cm} (1)

This process is known as the sum combination resonance. It has been discovered in electronic circuits in 1950’s\textsuperscript{17, 18} and later on found applications in optics\textsuperscript{12, 13}. Similar resonance occurs in the free electron laser in which the low frequency plasma and laser waves are excited by the sum frequency undulator wave in the electron rest frame\textsuperscript{19}. It also appears when electromagnetic waves interact with optical phonons in dielectric crystals\textsuperscript{20, 22}.

In the literature, another type of combination resonance, namely the difference combination resonance, has been discussed in mechanical systems since the 1960’s (see, e.g.,\textsuperscript{23, 26}). It can occur when the driving frequency matches the frequency difference between two normal frequencies

$$\nu_d = \omega_2 - \omega_1$$  \hspace{1cm} (2)

which yields amplification of $\omega_1$ and $\omega_2$. Combination resonances affect dynamic stability of structures and appear in various systems having multiple degrees of freedom.

Recently, the difference combination resonance has been experimentally demonstrated in an electronic circuit in the radio frequency range and proposed as a mechanism of generation of high frequency coherent radiation (e.g. XUV or X-rays) utilizing a low frequency (e.g., Infrared) driving field\textsuperscript{27}. Such a parametric amplifier, which operates by Quantum Amplification by Superradiant Emission of Radiation, is called the QASER for short. In contrast to OPOs, QASER generates light at higher frequencies than $\nu_d$.

Here we study the possibility of generation of high frequency coherent radiation using the difference combination resonance in nonlinear metamaterials. Metamaterials are artificial inhomogeneous structures composed of periodic subwavelength, polarizable elements. They can give rise to negative refractive index, near-zero permittivity or permeability of the macroscopic response in the microwave, terahertz and even optical regions\textsuperscript{28, 30} and have led to a variety of fascinating phenomena such as a perfect lens and invisible cloaks\textsuperscript{31, 33}. Inclusion of nonlinear elements into metamaterials offers unique advantages to enhance nonlinearities by local-field amplification\textsuperscript{34, 35} and to provide particular constructive configurations under the phase-matching condition for highly efficient nonlinear processes\textsuperscript{36, 39}. Anomalous transmission properties of zero-permittivity channels can be used to boost Kerr nonlinearities.
Effect of nonlinearity in combination with the unique linear properties has been recently investigated in connection with new frequencies generation [38, 41, 42], solitary wave propagation [43, 44], self tuning and hysteresis transition [45, 46] and parametric amplification [47–49].

Study of parametric amplification in such materials has demonstrated that if for one of the waves the refractive index is negative (directions of the wave vector and the Poynting vector are opposite) the phase matching leads to a possibility of loss compensation and distributed-feedback parametric oscillation with no cavity [37, 47, 48]. In such system the backward-propagating wave provides an automatic feedback mechanism and yields dynamics analogous to the quasi-phase matched mirror-less OPO [50].

Here we investigate parametric amplification in metamaterials caused by the difference combination resonance. We find that when for the idler (frequency $\omega_1$) and the signal (frequency $\omega_2$) waves the refractive index has opposite signs they both can be amplified by a strong driving field with frequency $\nu_d = \omega_2 - \omega_1$ (see Fig. 1). We show that the light amplification mechanism in negative index materials is equivalent to a combination resonance in a system of two coupled parametric oscillators with the opposite sign of masses.

We also find that the difference combination resonance can be achieved in materials with positive refractive index but strong anomalous dispersion. Namely, it can occur if at one of the generated frequencies $\omega_1$ or $\omega_2$ the group velocity is opposite to the phase velocity. This condition is somewhat easier to realize than the negative refractive index. Photonic crystals [51] and fiber Bragg gratings [52] are examples of materials with strong anomalous dispersion. Electromagnetically induced transparency (EIT) is a popular technique to decrease absorption near a resonance in the region of strong dispersion. Using the EIT technique, the group velocity of light as low as meters per second or even completely stopped light was demonstrated in ultracold gases [53, 54], hot gases [55, 56] and in a very cold solid [57]. Negative group velocity can be also obtained in the Kerr medium, which possesses a large nonlinear refractive index and long relaxation time, such as Cr-doped alexandrite, ruby and GdAlO$_3$ [58].

Parametric amplification process due to the difference combination resonance holds promise for a new kind of higher frequency convertor with tunable wavelength that makes use of metamaterials with negative refractive index or strongly dispersive media.

![Diagram](image1)

**FIG. 1:** Generation of lower frequencies $\omega_1$ and $\omega_2$ in a positive index nonlinear material (PIM) by the sum combination resonance (top). Generation of light at higher frequencies in a negative refractive index nonlinear material (NIM) due to the difference combination resonance (bottom). One can physically interpret the latter process as annihilation of the driving field photon with energy $\hbar \nu_d$ and creation of two photons - one with positive energy $\hbar \omega_2$ and one with negative energy $-\hbar \omega_1$.

### II. PARAMETRIC GENERATION OF HIGH FREQUENCY COHERENT RADIATION IN NONLINEAR MATERIALS

#### A. Combination resonances for two coupled parametric oscillators

Generation of high frequencies by a parametric resonance in systems with several normal modes can be understood in a simple example of two classical oscillators with masses $m_1$ and $m_2$ and (positive) frequencies $\omega_1$ and $\omega_2$ that are
weakly coupled with a periodically varying strength. The Hamiltonian of such a system reads

\[ H = \frac{p_1^2}{2m_1} + \frac{1}{2}m_1\omega_1^2x_1^2 + \frac{p_2^2}{2m_2} + \frac{1}{2}m_2\omega_2^2x_2^2 + F(t)x_1x_2. \] (3)

Using the Hamilton equations of motion and rescaling the coordinate \( x_1 \rightarrow \sqrt{|m_2/m_1|}x_1 \) we obtain

\[ \ddot{x}_1 + \omega_1^2x_1 + f_{12}(t)x_2 = 0, \] (4)

\[ \ddot{x}_2 + \omega_2^2x_2 + f_{21}(t)x_1 = 0, \] (5)

where

\[ f_{12}(t) = \frac{F(t)}{\sqrt{|m_1m_2|}}\text{sign}(m_2), \] (6)

and

\[ f_{21} = \text{sign}(m_1m_2)f_{12} \] (7)

describe coupling between oscillators. For harmonic modulation of the coupling strength with frequency \( \nu_d \) we write

\[ f_{12} = \delta \cos(\nu_d t), \] (8)

where \( \delta \) is the modulation amplitude. According to Eq. (7), the coupling between oscillators is symmetric \((f_{21} = f_{12})\) if \( m_1m_2 > 0 \) and antisymmetric \((f_{21} = -f_{12})\) if the masses \( m_1 \) and \( m_2 \) have opposite signs \[59\].

Next we assume that modulation frequency obeys the condition of the combination parametric resonance

\[ \nu_d = \omega_2 \pm \omega_1. \] (9)

Making a slowly varying amplitude approximation, namely writing

\[ x_1(t) = A_1(t)\cos(\omega_1 t), \] (10)

\[ x_2(t) = A_2(t)\sin(\omega_2 t), \] (11)

where \( A_1(t) \) and \( A_2(t) \) are slowly varying envelope functions on a time scale \( 1/|\omega_2 - \omega_1| \) and keeping only the resonant terms, Eqs. (4) and (5) reduce to

\[ \dot{A}_1 \pm \frac{\delta}{4\omega_1}A_2 = 0, \] (12)

\[ \dot{A}_2 + \text{sign}(m_1m_2)\frac{\delta}{4\omega_2}A_1 = 0, \] (13)

which yield the following equation for \( A_1(t) \)

\[ \ddot{A}_1 \mp \text{sign}(m_1m_2)\frac{\delta^2}{16\omega_1\omega_2}A_1 = 0. \] (14)

Solutions for \( A_{1,2} \) exponentially grow with time, \( A_{1,2} \propto e^{Gt} \) provided \( \pm m_1m_2 > 0 \). Namely, for symmetric coupling \((m_1m_2 > 0)\), the solution grows if \( \nu_d = \omega_2 + \omega_1 \) (the sum combination resonance). For antisymmetric coupling \((m_1m_2 < 0)\) there is exponential growth provided \( \nu_d = \omega_2 - \omega_1 \) (the difference combination resonance). For both resonances the gain per unit time \( G \) is given by

\[ G = \frac{\delta}{4\sqrt{\omega_1\omega_2}}. \] (15)

The sum combination resonance is used for generation of frequencies lower than \( \nu_d \). This is the case for optical parametric oscillators that convert an input laser wave with frequency \( \nu_d \) into two output waves of lower frequency \( \omega_1 \)
FIG. 2: Energy $W_1$ of the first and of the second $W_2$ oscillator [in units of $W_1(0)$] as a function of time for asymmetric coupling $f_{12} = -f_{21} = \delta \cos(\nu_d t)$ and off-resonance modulation $\nu_d = 0.35\omega_2$ obtained by numerical solution of Eqs. (4) and (5) with $\omega_1 = 0.77\omega_2$ and $\delta = 0.02\omega_2$. Initially the first oscillator is excited while the second oscillator is at rest.

FIG. 3: The same as in Fig. 2 but for the resonant modulation $\nu_d = \omega_2 - \omega_1 = 0.23\omega_2$.

and $\omega_2$) by means of the second order nonlinear optical interaction. On the other hand, the difference combination resonance can be used for the generation of higher frequencies. This has been recently demonstrated in nonreciprocally coupled electronic circuits in the radio frequency domain [27].

As an illustration of combination resonances, in Figs. 2-4 we plot energy of the first $W_1$ and the second $W_2$ oscillator as a function of time for off-resonance (Fig. 2) and on-resonance (Figs. 3 and 4) modulation obtained by numerical solution of Eqs. (4) and (5). In simulations we take $\omega_1 = 0.77\omega_2$ and $\delta = 0.02\omega_2$. Initially the first oscillator is excited while the second oscillator is at rest. If $\nu_d$ does not satisfy the resonance condition (9) the oscillators do not exchange their energy, as shown in Fig. 2.

However, if, e.g., the condition of the difference combination resonance is satisfied, $\nu_d = \omega_2 - \omega_1$, the oscillator’s energies exponentially grow for antisymmetric coupling (see Fig. 3). This is the essence of the QASER amplification mechanism [27]. If the coupling is symmetric, the difference combination resonance yields an efficient energy exchange between two oscillators without exponential grow, as illustrated in Fig. 4. This can be used in various applications, e.g., for fast control of propagation of $\gamma$-rays through crystals [60].
In Fig. 4 we plot the gain $G$ as a function of the modulation frequency $\nu_d$ for symmetric ($f_{12} = f_{21}$) and antisymmetric ($f_{12} = -f_{21}$) coupling between oscillators obtained from numerical solution of Eqs. (4), (5) and (8) with $\omega_1 = 0.77\omega_2$ and $\delta = 0.02\omega_2^2$. For symmetric coupling the system has the sum combination resonance at $\nu_d = \omega_2 + \omega_1$, while for the antisymmetric coupling there is the difference combination resonance at $\nu_d = \omega_2 - \omega_1$.

B. Combination resonances in magneto-optical crystals

The possibility of a difference combination resonance in materials with negative refractive index can be demonstrated in the simple example of magneto-optic effect. In a magneto-optic material the presence of an externally applied magnetic field $H_0(t)$ causes a change in the permittivity tensor $\varepsilon_{ik}$. The tensor becomes anisotropic with complex off-diagonal components. The off-diagonal components of $\varepsilon_{ik}$ describe coupling between two perpendicular polarizations of an electromagnetic wave. Such coupling can be periodically modulated if the externally applied magnetic field is a
harmonic function of time $H_0 \propto \cos(\nu_d t)$.

Here we show that the problem of propagation of electromagnetic waves through the gyromagnetic medium can be reduced to the two coupled parametric oscillators discussed in the previous section. Evolution of the electromagnetic field in the medium is described by Maxwell’s equations
\[
\text{curl} \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}, \quad \text{curl} \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t}.
\] (16)

We assume that the wave propagates along the $z$-axis and the electromagnetic field of the wave has only $x$ and $y$ components which depend on $t$ and $z$. Then the Maxwell’s equations (16) reduce to
\[
\frac{\partial E_y}{\partial z} = \frac{\partial B_x}{\partial t}, \quad \frac{\partial E_x}{\partial z} = -\frac{\partial B_y}{\partial t},
\] (17)
\[
\frac{\partial H_y}{\partial z} = -\frac{\partial D_x}{\partial t}, \quad \frac{\partial H_x}{\partial z} = \frac{\partial D_y}{\partial t}.
\] (18)

If the absorption losses can be neglected, the permittivity tensor is a Hermitian matrix [61] and we can write
\[
\begin{pmatrix}
E_x \\
E_y
\end{pmatrix} =
\begin{pmatrix}
\tilde{\varepsilon}_x & ig(t) \\
-ig(t) & \tilde{\varepsilon}_y
\end{pmatrix}
\begin{pmatrix}
D_x \\
D_y
\end{pmatrix},
\] (19)

\[
\begin{pmatrix}
H_x \\
H_y
\end{pmatrix} =
\begin{pmatrix}
\tilde{\mu}_x & 0 \\
0 & \tilde{\mu}_y
\end{pmatrix}
\begin{pmatrix}
B_x \\
B_y
\end{pmatrix},
\] (20)

where the off-diagonal component $g(t)$, to first order, is proportional to the applied external magnetic field $H_0(t)$ that lies along the $z$-axis [62]. Periodic variation of $H_0(t)$ with time yields periodic modulation of $g$, $g(t) = g_0 \cos(\nu_d t)$. Taking time derivative from Eqs. (18) and using Eqs. (17), (19) and (20) we obtain
\[
\frac{\partial^2 D_x}{\partial t^2} - \tilde{\mu}_x \tilde{\varepsilon}_x \frac{\partial^2 D_x}{\partial z^2} - ig(t)\tilde{\mu}_y \frac{\partial^2 D_y}{\partial z^2} = 0,
\] (21)
\[
\frac{\partial^2 D_y}{\partial t^2} - \tilde{\mu}_x \tilde{\varepsilon}_y \frac{\partial^2 D_y}{\partial z^2} + ig(t)\tilde{\mu}_y \frac{\partial^2 D_x}{\partial z^2} = 0.
\] (22)

Change of functions
\[
D_x = i\sqrt{|\tilde{\mu}_y|} \tilde{D}_z, \quad D_y = \sqrt{|\tilde{\mu}_x|} \tilde{D}_y
\] (23)
yields
\[
\frac{\partial^2 \tilde{D}_z}{\partial t^2} - \tilde{\mu}_x \tilde{\varepsilon}_x \frac{\partial^2 \tilde{D}_z}{\partial z^2} - \text{sign}(\tilde{\mu}_y)\sqrt{|\tilde{\mu}_x\tilde{\mu}_y|} g(t) \frac{\partial^2 \tilde{D}_y}{\partial z^2} = 0,
\] (24)
\[
\frac{\partial^2 \tilde{D}_y}{\partial t^2} - \tilde{\mu}_x \tilde{\varepsilon}_y \frac{\partial^2 \tilde{D}_y}{\partial z^2} - \text{sign}(\tilde{\mu}_x)\sqrt{|\tilde{\mu}_x\tilde{\mu}_y|} g(t) \frac{\partial^2 \tilde{D}_z}{\partial z^2} = 0.
\] (25)

One can look for a solution of these equations in the form
\[
\tilde{D}_{x,y}(t,z) = \tilde{D}_{x,y}(t)e^{ikz}
\] (26)
which results in the equations of two coupled harmonic oscillators with time dependent coupling
\[
\frac{\partial^2 \tilde{D}_x}{\partial t^2} + \omega_1^2 \tilde{D}_x + f_{12}(t)\tilde{D}_y = 0,
\] (27)
\[
\frac{\partial^2 \tilde{D}_y}{\partial t^2} + \omega_2^2 \tilde{D}_y + f_{21}(t)\tilde{D}_x = 0,
\] (28)
where

$$\omega_1^2 = \tilde{\mu}_y \tilde{\varepsilon}_x k^2, \quad \omega_2^2 = \tilde{\mu}_x \tilde{\varepsilon}_y k^2,$$

$$f_{12}(t) = \sqrt{|\tilde{\mu}_x \tilde{\mu}_y|} |k^2| \text{sign}(\tilde{\mu}_y) g(t) \propto \cos(\nu_d t),$$

and

$$f_{21}(t) = \text{sign}(\tilde{\mu}_x \tilde{\mu}_y) f_{12}(t).$$

Eqs. (27) and (28) are identical to the equations of the coupled parametric oscillators (4) and (5) investigated in the previous section and, hence, they display the same combination resonances when the modulation frequency satisfies the condition $\nu_d = \omega_2 \pm \omega_1$. Namely, if $\tilde{\mu}_x \tilde{\mu}_y > 0$ the coupling between the two field polarizations $x$ and $y$ is symmetric and there is the sum combination resonance at $\nu_d = \omega_2 + \omega_1$ which yields generation of lower frequencies $\omega_1$ and $\omega_2$. This is the case for the positive index materials and is an operation mechanism of the optical parametric amplifiers.

The coupling between the two field polarizations is antisymmetric if $\tilde{\mu}_x$ and $\tilde{\mu}_y$ have opposite signs. To make frequencies in Eq. (29) real the corresponding components of the permittivity tensor also should have opposite signs. Namely, if $\tilde{\mu}_x < 0$ and $\tilde{\mu}_y > 0$ then we must have $\tilde{\varepsilon}_y < 0$ and $\tilde{\varepsilon}_x > 0$. Under these conditions there is the difference combination resonance at $\nu_d = \omega_2 - \omega_1$ which yields generation of higher frequencies $\omega_1$ and $\omega_2$. This case is similar to the two coupled oscillators with the opposite sign of masses discussed in the previous section.

According to Eq. (20), generated waves at frequencies $\omega_1$ and $\omega_2$ have the same wave vector $k$, but orthogonal polarizations. The values of $\omega_1$ and $\omega_2$ differ due to the anisotropy of the medium ($\tilde{\mu}_y \tilde{\varepsilon}_x \neq \tilde{\mu}_x \tilde{\varepsilon}_y$). Using combination resonance one can also generate waves with different wave vectors $k_1$ and $k_2$ if the driving field wave vector $k_d$ compensates for the momentum mismatch $k_d = k_2 \pm k_1$.

Moreover, if we take into account frequency dispersion the expression for the gain $G$ is modified. Straightforward calculations similar to those we present in the next section yield

$$G^2 = \pm \frac{\omega_1 \omega_2 g_0^2}{16 \tilde{\varepsilon}_x (\omega_1) \tilde{\varepsilon}_y (\omega_2)} \frac{V_{g1} V_{g2}}{V_{p1} V_{p2}},$$

where $V_{g1,2}$ and $V_{p1,2}$ are the group and phase velocities of the waves with frequencies $\omega_{1,2}$. Eq. (32) shows that the difference combination resonance can also occur in materials with positive refractive index if at one of the generated frequencies the group and the phase velocities are opposite to each other. This requires medium with strong anomalous dispersion. We discuss these questions in details in the next section in which we consider combination resonances in nonlinear dispersive medium assuming that waves have the same linear polarization.

C. Combination resonances in nonlinear dispersive medium

Using nonlinear crystals the optical parametric oscillators generate light at frequencies $\omega_1$ and $\omega_2$ satisfying the condition of the sum combination resonance $\nu_d = \omega_1 + \omega_2$, where $\nu_d$ is the frequency of the strong driving (pump) field $E_d$. Here we investigate the possibility of difference combination resonance in such materials. In the present model we assume that medium is spatially uniform, isotropic and polarization is a nonlinear function of the electric field, while magnetization is a linear function of the magnetic field. We also assume that the driving field and generated waves propagate along the $z$-axis and have the same polarization. Under these conditions the Maxwell’s equations reduce to

$$\frac{\partial E}{\partial z} = \frac{\partial B}{\partial t}, \quad \frac{\partial H}{\partial z} = \frac{\partial D}{\partial t}.$$  \hspace{1cm} (33)

We write the total electric field as

$$E = E_d + E_1 + E_2,$$

where the strong driving field is assumed to be fixed

$$E_d = A_d \cos (\nu_d t - k_d z)$$  \hspace{1cm} (35)
and $E_{1,2} \ll E_d$. In nonlinear isotropic medium the total displacement field in the absence of dispersion can be written as

$$D(t, z) = \varepsilon_0 E + \chi \varepsilon_0 E^2 \approx \varepsilon_0 (E_d + E_1 + E_2) + \chi \varepsilon_0 E_d^n$$

$$+ 2 \chi \varepsilon_0 A_d \cos(\nu_d t - k_d z) + 2 \chi \varepsilon_0 E_2 A_d \cos(\nu_d t - k_d z),$$

where we omitted small terms of the second order in $E_{1,2}$. $\varepsilon$ is the linear permittivity of the material and $\chi$ is the nonlinear susceptibility. If there is frequency dispersion it is convenient to write the Maxwell’s equations (33) and the constitutive relations in terms of the field Fourier components, namely

$$kE(\omega, k) = -\omega B(\omega, k), \quad kH(\omega, k) = -\omega D(\omega, k),$$

$$B(\omega, k) = \mu(\omega) \mu_0 H(\omega, k),$$

$$D(\omega, k) = \varepsilon(\omega) \varepsilon_0 E(\omega, k) +$$

$$\chi \varepsilon_0 A_d [E_1(\omega - \nu_d, k - k_d) + E_1(\omega + \nu_d, k + k_d) + E_2(\omega - \nu_d, k + k_d) + E_2(\omega + \nu_d, k + k_d)],$$

where we disregarded irrelevant term proportional to $E_d^n$, took into account that Fourier transform of $e^{-\nu_d t + ik_d z} f(t, z)$ gives $f(\omega - \nu_d, k - k_d)$ and for simplicity assumed that the nonlinear susceptibility $\chi$ has no dispersion. In our considerations the wavenumber $k$ is real while frequency $\omega$ has small imaginary part $G$ which determines the wave amplification or attenuation per unit time. Plug Eqs. (37) and (38) into Eq. (36) yields the following equation for $E_1$ and $E_2$

$$\left[c^2 k^2 - \omega^2 \varepsilon(\omega) \mu(\omega)\right] (E_1(\omega, k) + E_2(\omega, k))$$

$$- \omega^2 \chi(\mu) A_d [E_1(\omega - \nu_d, k - k_d) + E_1(\omega + \nu_d, k + k_d) + E_2(\omega - \nu_d, k + k_d) + E_2(\omega + \nu_d, k + k_d)] = 0.$$ (39)

Next we assume that

$$\nu_d = \omega_2 \pm \omega_1,$$ (40)

$$k_d = k_2 \pm k_1,$$ (41)

where $k_{1,2}$ and $\omega_{1,2}$ obey the relations

$$\varepsilon^2 k_1^2 = \omega_1^2 \varepsilon(\omega_1) \mu(\omega_1),$$ (42)

$$\varepsilon^2 k_2^2 = \omega_2^2 \varepsilon(\omega_2) \mu(\omega_2).$$ (43)

Since the driving field propagates through the same medium its frequency $\nu_d$ and the wavenumber $k_d$ satisfy the similar equation

$$\varepsilon^2 k_d^2 = \nu_d^2 \varepsilon(\nu_d) \mu(\nu_d).$$ (44)

Eqs. (40) and (41) are consistent with Eqs. (42)-(44) provided

$$\omega_2 \left(1 - \frac{\varepsilon \mu_2}{\varepsilon_d \mu_d}\right) = \mp \omega_1 \left(1 - \frac{\varepsilon \mu_1}{\varepsilon_d \mu_d}\right),$$ (45)

where $\varepsilon_{1,2}$ and $\varepsilon_d$ stands for $\varepsilon(\omega_{1,2})$ and $\varepsilon(\nu_d)$, etc.

We look for a solution of Eq. (39) in the form

$$E_1(\omega, k) \approx E_1 \delta(\omega \pm \omega_1 - iG) \delta(k \pm k_1), \quad E_2(\omega, k) \approx E_2 \delta(\omega - \omega_2 - iG) \delta(k - k_2),$$ (46)
where $G$ is the gain per unit time. Keeping the resonant terms we obtain

\[
\left[(\varepsilon k^2 - \omega^2 \mu(\omega)) E_1 - \omega^2 \mu(\omega) A_d E_2 \right] \delta(\omega \pm \omega_1 - iG) \delta(k \pm k_1) = 0, \tag{47}
\]

\[
\left[(\varepsilon k^2 - \omega^2 \mu(\omega)) E_2 - \omega^2 \mu(\omega) A_d E_1 \right] \delta(\omega - \omega_2 - iG) \delta(k - k_2) = 0. \tag{48}
\]

The factor in front of the delta functions in Eq. (47) must be calculated at $k = \pm k_1$ and $\omega = \mp \omega_1 + iG$, while the factor in Eq. (48) should be estimated at $k = k_2$ and $\omega = \omega_2 + iG$. Making a Taylor expansion one can write

\[
(c^2 k^2 - \omega^2 \mu(\omega)) \bigg|_{k=\mp k_1, \omega=\mp \omega_1 + iG} \approx \pm iG \frac{\partial}{\partial \omega_1} \left[ \varepsilon k^2 (\omega_1) \mu(\omega_1) \right] = \pm iG \frac{2c \omega_1 n_1}{V_{g1}}, \tag{49}
\]

\[
(c^2 k^2 - \omega^2 \mu(\omega)) \bigg|_{k=k_2, \omega=\omega_2 + iG} \approx -iG \frac{\partial}{\partial \omega_2} \left[ \varepsilon k^2 (\omega_2) \mu(\omega_2) \right] = -iG \frac{2c \omega_2 n_2}{V_{g2}}, \tag{50}
\]

where $|n(\omega)| = \sqrt{\varepsilon(\omega) / \mu(\omega)}$ is the refractive index ($n$ is positive for $\varepsilon, \mu > 0$ and negative for $\varepsilon, \mu < 0$),

\[
V_g(\omega) = \frac{\partial \omega}{\partial k} = \frac{2cmn(\omega)}{\mu(\omega) n_0}.
\]

is the group velocity of the wave at frequency $\omega$ and $c = 1/\sqrt{\varepsilon_0 \mu_0}$ is the speed of light in vacuum. For brevity we introduced notations $n_{1,2} = n(\omega_{1,2})$ and $V_{g1,2} = V_g(\omega_{1,2})$. Taking into account Eqs. (49) and (50), and keeping the leading terms, Eqs. (47) and (48) reduce to

\[
\pm iG \frac{2c \omega_1 n_1}{V_{g1}} E_1 - \omega_1 \chi \mu_1 A_d E_2 = 0, \tag{52}
\]

\[
iG \frac{2c \omega_2 n_2}{V_{g2}} E_2 + \omega_2 \chi \mu_2 A_d E_1 = 0. \tag{53}
\]

Eqs. (52) and (53) give the following expression for the gain

\[
G^2 = \pm \mu_1 \mu_2 \omega_1 \omega_2 \chi^2 A_d^2 \frac{V_{g1} V_{p1} V_{g2} V_{p2}}{4c^4}, \tag{54}
\]

where $V_{p1,2} = c/n_{1,2}$ is the phase velocity at frequency $\omega_{1,2}$. In the time domain solution (49) reads

\[
E_1(t, z) = E_1 e^{+i \omega_1 t + ik_1 z + Gt}, \quad E_2(t, z) = E_2 e^{-i \omega_2 t + ik_2 z + Gt}. \tag{55}
\]

This solution exponentially grows with time if $G$ is real (and positive). Thus, in order to have amplification the right hand side of Eq. (53) must be positive. For materials with $\mu_{1,2} > 0$ there is gain for the upper sign, that is when $\nu_d = \omega_2 - \omega_1$. Such materials yield the sum combination resonance and are used in conventional optical parametric amplifiers. In order to achieve light amplification at the difference combination resonance $\nu_d = \omega_2 - \omega_1$ (the lower sign in Eq. (54)) we must have

\[
\mu_1 \mu_2 < 0. \tag{56}
\]

Conditions (56) and (55) can be fulfilled, e.g., if the crystal has a negative refractive index at the frequency $\omega_1$ ($\varepsilon_1 < 0$ and $\mu_1 < 0$), but a positive refractive index at the frequency $\omega_2$ ($\varepsilon_2 > 0$ and $\mu_2 > 0$). In addition, Eq. (54) shows that the difference combination resonance can also occur in nonlinear materials with positive refractive index if at one of the generated frequencies $\omega_1$ or $\omega_2$ the group velocity $V_g$ is opposite to the phase velocity $V_p$. This condition is somewhat easier to achieve than the negative refractive index. It requires strong anomalous dispersion, however, material does not need to have both magnetic and electric responses.

In our analysis we have assumed that medium is spatially uniform. An interesting question appears in this connection. Namely, if a material is nonuniform can this eliminate the requirement of having a negative refractive index to achieve the difference combination resonance in medium with weak dispersion? In Appendix A we investigate this question for materials described by the local constitutive relations $D = \varepsilon(\mathbf{r}) \varepsilon_0 \mathbf{E}$ and $B = \mu(\mathbf{r}) \mu_0 \mathbf{H}$. Based on a general consideration we show that the difference combination resonance in nonlinear weakly dispersive materials is possible only if for one of the generated frequencies both the magnetic permeability $\mu$ and dielectric permittivity $\varepsilon$ are negative in some region of space.
III. SUMMARY

In this paper we consider parametric generation of coherent radiation in nonlinear medium produced by a strong driving field. In conventional nonlinear crystals with a positive refractive index such parametric mechanism yields generation of smaller frequencies due to the sum combination resonance (see Eq. (1)) which is the operating principle of optical parametric amplifiers. Here we find that in left-handed materials the difference combination resonance described by Eq. (2) can occur if for one of the generated frequencies the refractive index is negative. Such a resonance yields excitation of frequencies higher than $\nu_d$.

We show that this mechanism is similar to a combination resonance in a system of two coupled parametric oscillators with periodically modulated coupling strength. The difference combination resonance occurs when the coupling between oscillators in the equation of motion is antisymmetric [see Eqs. (4) and (5)]. Such antisymmetric coupling is realized in the Hamiltonian system if the two oscillators have the opposite sign of masses. If coupling is symmetric (both masses are positive) the system has the sum (rather than the difference) combination resonance.

We also find that generation of higher frequencies by means of the difference combination resonance is possible in nonlinear materials with positive refractive index but strong anomalous dispersion. In such a medium the resonance can occur if at one of the generated frequencies the group and phase velocities are opposite to each other. Anomalous dispersion is easier to realize than the negative refractive index. Thus, highly dispersive nonlinear optical systems are an attractive tool for applications in generation of high frequency coherent radiation.

The sum combination resonance is a robust phenomenon which occurs in various areas of physics. In contrast, the difference combination resonance is hard to achieve because it requires special conditions, e.g., nonreciprocal coupling. Recently such a resonance has been experimentally demonstrated in nonreciprocally coupled resonant RLC circuits at radio frequencies [27]. It has also been proposed for generation of high frequency coherent radiation in atomic medium by driving the atomic ensemble with a much smaller frequency [27]. The proposed new kind of light amplifier (called the QASER), contrary to a laser, does not need any population of atoms in the excited state. The amplification mechanism of the QASER is governed by the difference combination resonance which occurs when the driving field frequency matches the frequency difference between two normal modes of the coupled light atom system.

The present paper links the QASER amplification mechanism and parametric processes that can occur in negative index nonlinear materials and strongly dispersive media. The difference combination resonance in such systems opens a perspective for development of new kind of light sources that, contrary to conventional OPOs, emit coherent radiation of tunable wavelengths with the frequency higher than the input frequency.

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Appendix A: Combination resonance in nonuniform medium

Here we investigate conditions of the combination resonance in nonlinear medium assuming that magnetic permeability $\mu(r)$ and dielectric permittivity $\varepsilon(r)$ depend on coordinates. This allows us to include nonuniform dielectric structures into consideration. For simplicity we assume that dispersion is weak which allows us to do calculations in the time domain. Maxwell’s equations (16) yield

$$\text{curl} \left( \frac{1}{\mu} \text{curl} \mathbf{E} \right) + \frac{1}{c^2 \varepsilon_0} \frac{\partial^2 \mathbf{D}}{\partial t^2} = 0. \quad (A1)$$

We write the total electric field as

$$\mathbf{E} = \mathbf{E}_d(t, r) + \mathbf{E}_1(t, r) + \mathbf{E}_2(t, r), \quad (A2)$$

where

$$\mathbf{E}_d(t, r) = A_d(r)e^{i\nu_d t} + c.c., \quad (A3)$$

$$\mathbf{E}_1(t, r) = A_1(r)e^{i(\omega_1 t + G t)}, \quad (A4)$$
Taking imaginary part of Eq. (A12), we then obtain

\[ \mathbf{E}_2(t, \mathbf{r}) = \mathbf{A}_2(\mathbf{r}) e^{i\omega_2 t + G t}, \] (A5)

and \( G \) is a small gain. We assume that the driving field frequency \( \nu_d \) satisfies the condition of the combination resonance

\[ \nu_d = \omega_2 \pm \omega_1. \] (A6)

Taking into account that nonlinear susceptibility \( \chi \) is small and disregarding the fast oscillating terms we obtain the following equations for \( \mathbf{A}_1(\mathbf{r}) \) and \( \mathbf{A}_2(\mathbf{r}) \)

\[
- \text{curl} \left( \frac{1}{\mu_1} \text{curl} \mathbf{A}_1 \right) + \frac{\varepsilon_1(\omega_1 \pm iG)^2}{\omega_1^2 c^2} \mathbf{A}_1 + \chi^* \frac{\omega_1^2}{c^2} \mathbf{A}_2 = 0, \]

(A7)

\[
- \text{curl} \left( \frac{1}{\mu_2} \text{curl} \mathbf{A}_2 \right) + \frac{\varepsilon_2(\omega_2 - iG)^2}{\omega_2^2 c^2} \mathbf{A}_2 + \chi^* \frac{\omega_2^2}{c^2} \mathbf{A}_1 = 0, \]

(A8)

where \( \varepsilon_1, \mu_1 \) and \( \varepsilon_2, \mu_2 \) are the values of \( \varepsilon \) and \( \mu \) at the frequencies \( \omega_1 \) and \( \omega_2 \) respectively. Multiplying Eq. (A7) by \( A_1^* \) and Eq. (A8) by \( A_2^* \) yields

\[
- \frac{1}{\omega_1^2} A_1^* \cdot \text{curl} \left( \frac{1}{\mu_1} \text{curl} \mathbf{A}_1 \right) + \frac{\varepsilon_1(\omega_1 \pm iG)^2}{\omega_1^2 c^2} |A_1|^2 + \chi^* \frac{\omega_1^2}{c^2} A_1^* A_2 = 0, \]

(A9)

\[
- \frac{1}{\omega_2^2} A_2^* \cdot \text{curl} \left( \frac{1}{\mu_2} \text{curl} \mathbf{A}_2 \right) + \frac{\varepsilon_2(\omega_2 + iG)^2}{\omega_2^2 c^2} |A_2|^2 + \chi^* \frac{\omega_2^2}{c^2} A_2^* A_1 = 0. \]

(A10)

Subtracting these equations from each other, taking into account that

\[
A^* \cdot \text{curl} \left( \frac{1}{\mu} \text{curl} \mathbf{A} \right) = \frac{1}{\mu} |\text{curl} \mathbf{A}|^2 + \text{div} \left( \frac{1}{\mu} \text{curl} \mathbf{A} \times A^* \right), \]

(A11)

and integrating over space we find

\[
\int d\mathbf{r} \left[ \frac{1}{\omega_2^2 \mu_2} |\text{curl} \mathbf{A}_2|^2 - \frac{1}{\omega_1^2 \mu_1} |\text{curl} \mathbf{A}_1|^2 + \frac{\varepsilon_1(\omega_1 \pm iG)^2}{\omega_1^2 c^2} |A_1|^2 - \frac{\varepsilon_2(\omega_2 + iG)^2}{\omega_2^2 c^2} |A_2|^2 \right] = 0. \]

(A12)

Taking imaginary part of Eq. (A12), we then obtain

\[
\int d\mathbf{r} \left[ \frac{\varepsilon_1}{\omega_1} |A_1|^2 + \frac{\varepsilon_2}{\omega_2} |A_2|^2 \right] = 0. \]

(A13)

Here the upper (lower) sign corresponds to the sum (difference) combination resonance.

On the other hand, taking real part of Eqs. (A9) and (A10) and disregarding the small terms caused by nonlinearity we find after integration over space

\[
\int d\mathbf{r} \varepsilon_1 |A_1|^2 = \frac{c^2}{\omega_1^2} \int d\mathbf{r} \frac{1}{\mu_1} |\text{curl} \mathbf{A}_1|^2, \]

(A14)

\[
\int d\mathbf{r} \varepsilon_2 |A_2|^2 = \frac{c^2}{\omega_2^2} \int d\mathbf{r} \frac{1}{\mu_2} |\text{curl} \mathbf{A}_2|^2. \]

(A15)

Eq. (A13) implies that for the difference combination resonance (lower sign “−”) the integrals \( \int d\mathbf{r} \varepsilon_1 |A_1|^2 \) and \( \int d\mathbf{r} \varepsilon_2 |A_2|^2 \) must have opposite signs. If, e.g., \( \int d\mathbf{r} \varepsilon_1 |A_1|^2 < 0 \) then Eq. (A14) yields that \( \mu_1 < 0 \) in some region of
space. Thus, we obtain that the difference combination resonance in nonlinear weakly dispersive materials is possible only if for one of the generated frequencies both the magnetic permeability \( \mu \) and dielectric permittivity \( \varepsilon \) are negative in some spatial volumes.

One should note that calculations presented here assume local (in time and space) constitutive relations \( \mathbf{D} = \varepsilon(\mathbf{r})\varepsilon_0 \mathbf{E} \) and \( \mathbf{B} = \mu(\mathbf{r})\mu_0 \mathbf{H} \). Thus, the results obtained are valid if one can disregard frequency and spatial dispersion.
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