Degenerate four-wave mixing in transparent two-component medium considering spatial structure of the pump waves

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Abstract. In this paper we investigate spatial selectivity of the degenerate four-wave radiation converter in transparent liquid containing nanoparticles considering spatial structure of the pump waves. The bandwidth of the most efficiently converted spatial frequencies is associated with the rotation and divergence of the pump waves.

1. Introduction
Accounting for spatial structure of the pump waves in the study of radiation (image) conversion quality by four-wave mixing is the necessary condition. The spatial structure of the pump waves fully determines the four-wave converters resolution [1, 2] or it is one of the main factors leading to the deterioration of the image conversion quality [3].

In recent years the ability to use media containing micro- and nanosized particles (colloidal solutions, suspensions, etc.) for realization of the four-wave mixing is actively discussed [4-10]. If liquids containing nanoparticles are used as media in four-wave mixing then in such media electrostriction and Dufour effect influence significantly on the characteristics of four-wave radiation converter filtering high spatial frequencies of the object wave [11, 12], which may be preferable in correction systems of small-scale phase inhomogeneities [13].

The aim of this work is to study influence of the divergence and rotation of the pump waves on spatial selectivity of the degenerate four-wave radiation converter in transparent liquid containing nanoparticles.

2. Relation of the interacting waves spatial spectra
Let us consider a plane layer thickness \( \ell \) of transparent two-component medium (liquid in which nanoparticles are located). In liquid in the Z-axis direction two pump waves with complex amplitudes \( A_1 \) and \( A_2 \) towards each other and signal wave with complex amplitude \( A_3 \) propagate. Diffraction of the second pump wave (\( A_2 \)) on the induced dynamic phase grating in the medium leads to emergence of the object wave with complex amplitude \( A_4 \).

Degenerate four-wave mixing \( \omega + \omega - \omega = \omega \) in transparent two-component medium is described by the Helmholtz equation [1]

\[
\left( \nabla^2 + k^2 + \frac{2k^4}{n_0} \frac{d n}{d T} \frac{dT}{d} \right) (A + A^*) = 0
\]  

(1)
where \( A = \sum_{j=1}^{4} A_j \), \( \delta T \) is a temperature variation, \( k = \frac{\omega n_0}{c} \), \( n_0 \) is the average refractive index.

Equation (1) is complemented by the material equation for the temperature variation [11]

\[
\nabla^2 \delta T = \frac{\gamma D_{12}}{D_{11} D_{22}} \nabla^2 I.
\]

(2)

Here, \( I = A A^* \), \( D_{11} \) is the thermal conductivity coefficient, \( D_{22} \) is the diffusion coefficient, \( D_{12} \) is a coefficient which describes Dufour effect, \( \gamma \) is the electrostriction coefficient.

Four-wave mixing is considered under the following conditions:

- the approximation of the specified field by pump waves is valid \( \left( |A_{1,2}| >> |A_{3,4}| \right) \);
- the reflection (conversion) coefficient is small \( \left( |A_3| << |A_3| \right) \).

The intensity of radiation which propagates in nonlinear medium can be written as follows

\[
I = I_0 + A A_i^* + A_i A_i^*.
\]

Here, \( I_0 = A_0 A_0^* + A_2 A_2^* \).

Then the temperature variation can be expressed as the sum of the quickly \( (\delta T_{31}) \) and slowly \( (\delta T_0) \) varying components which depend on the coordinates

\[
\delta T = \delta T_0 + \delta T_{31},
\]

In view of the foregoing, the Helmholtz equation (1) is divided into four equations

\[
\left( \nabla^2 + k^2 + \frac{2k^2}{n_0} \frac{dn}{dT} \delta T \right) A_{1,2,3} = 0,
\]

\[
\left( \nabla^2 + k^2 + \frac{2k^2}{n_0} \frac{dn}{dT} \delta T_0 \right) A_4 + \frac{2k^2}{n_0} \frac{dn}{dT} \delta T_{31} A_2 = 0
\]

(3)

and material equation (2) is divided into two equations

\[
\nabla^2 \delta T_0 = \frac{\gamma D_{12}}{D_{11} D_{22}} \nabla^2 I_0, \quad \nabla^2 \delta T_{31} = \frac{\gamma D_{12}}{D_{11} D_{22}} \nabla^2 A_i A_i^*.
\]

(4)

We expand the interacting waves in plane waves

\[
A_j(\vec{r}) = \int_{-\infty}^{\infty} \tilde{A}_j(\vec{\kappa}_j, z) \exp(-i\vec{\kappa}_j \cdot \vec{\rho} - i k_{jz} z) d\vec{\kappa}_j, \quad j = 1 - 4
\]

(5)

and expand the quickly varying component of temperature in harmonic gratings

\[
\delta T_{31}(\vec{r}) = \int_{-\infty}^{\infty} \delta \tilde{T}_{31}(\vec{\kappa}_T, z) \exp(-i\vec{\kappa}_T \cdot \vec{\rho}) d\vec{\kappa}_T.
\]

(6)

Here, \( \tilde{A}_j \) is spatial spectrum of \( j \)th wave, \( \delta \tilde{T}_{31} \) is spatial spectrum of the temperature grating, \( \vec{\kappa}_j \) and \( k_{jz} \) are the transverse and longitudinal components of the wave vector \( \vec{k}_j \), \( \vec{k}_T \) is the grating wave vector, \( \vec{\rho} (x, y) \) and \( z \) are the transverse and longitudinal components of the radius vector \( \vec{r} \).

Substituting (5) – (6) in the equations (3) – (4) we obtain the equations describing the changes in spatial spectra of the interacting waves and temperature grating along the \( z \) coordinate.
\[
\begin{aligned}
&\left(\frac{d}{dz} + i\frac{k^2}{k_{1,2,3}, n_0} \frac{dn}{dT} \delta T_0\right) \tilde{A}_{1,2,3}(\tilde{k}_{1,2,3}, z) = 0, \\
&\left(\frac{d}{dz} + i\frac{k^2}{k_{1,2,3}, n_0} \frac{dn}{dT} \delta T_0\right) \tilde{A}_4(\tilde{k}_4, z) = \\
&\quad = -i\frac{k^2}{k_{1,2,3}, n_0} \frac{dn}{dT} \int_{-\infty}^{\infty} \delta T_{31}(\tilde{k}_4 = \tilde{k}_4 - \tilde{k}_2, z) \tilde{A}_2(\tilde{k}_2, z) \exp[-i(k_{2z} - k_{4z})z] d\tilde{k}_2.
\end{aligned}
\]

\[
\begin{aligned}
&\left(\frac{d^2}{dz^2} + \kappa_T^2 - \kappa_T^2\right) \delta T_{31}(\tilde{k}_4, z) = \\
&\quad = \frac{\gamma D_{12}}{D_{11} D_{22}} \int_{-\infty}^{\infty} \tilde{A}_1(\tilde{k}_1, z) \tilde{A}_1(\tilde{k}_1 = \tilde{k}_4 - \tilde{k}_3, z) \exp[-i(k_{1z} - k_{3z})z] \left[\kappa_T^2 + (k_{1z} - k_{3z})^2\right] d\tilde{k}_1.
\end{aligned}
\]

Equations (7) and (8) are written under the condition that \(\tilde{k}_1 = \tilde{k}_1 - \tilde{k}_3 = \tilde{k}_4 - \tilde{k}_2\).

Using the boundary conditions on spatial spectra of the interacting waves \(\tilde{A}_{1,3}(\tilde{k}_{1,3}, z = 0) = \tilde{A}_{30}(\tilde{k}_{1,3}), \tilde{A}_4(\tilde{k}_2, z = \ell) = \tilde{A}_{30}(\tilde{k}_2)\), \(\tilde{A}_4(\tilde{k}_2, z = \ell) = 0\) and the condition that the temperature is invariable at the nonlinear layer edges \((\delta T_{31}(z = 0) = \delta T_{31}(z = \ell) = 0)\) we find changes in spatial spectra of the pump waves and the signal wave by nonlinear layer thickness

\[
\tilde{A}_{1,3}(\tilde{k}_{1,3}, z) = \tilde{A}_{30}(\tilde{k}_{1,3}) \exp[-P(z)], \tilde{A}_2(\tilde{k}_{1,3}, z) = \tilde{A}_{20}(\tilde{k}_{1,3}) \exp[-P(\ell) + P(z)]
\]

then the spatial spectrum of the temperature grating and the spatial spectrum of the object wave on the front edge of the nonlinear layer

\[
\begin{aligned}
\delta T_{31}(\tilde{k}_4, z) &= \frac{\gamma D_{12}}{D_{11} D_{22}} \int_{-\infty}^{\infty} d\tilde{k}_4 \tilde{A}_1(\tilde{k}_1) \tilde{A}_1(\tilde{k}_1 = \tilde{k}_4 - \tilde{k}_3) \left\{\frac{1}{\sinh \kappa_T \ell} \left[\exp[-i(k_{1z} - k_{3z})\ell] \sinh \kappa_T z - \sinh \kappa_T z \sinh \kappa_T \ell \right]\right\} \\
\tilde{A}_4(\tilde{k}_4, z = 0) &= -i \frac{k y D_{12}}{n_0 D_{11} D_{22}} \exp[-P(\ell)] \int_{-\infty}^{\infty} d\tilde{k}_2 d\tilde{k}_4 \tilde{A}_1(\tilde{k}_1) \tilde{A}_2(\tilde{k}_2) \tilde{A}_3(\tilde{k}_3) \tilde{A}_4(\tilde{k}_4 = \tilde{k}_4 - \tilde{k}_3) \times \\
&\quad \times \left[\frac{1}{2 \sinh |\tilde{k}_4 - \tilde{k}_2| \ell} \left(\exp[-i(k_{2z} - k_{4z})\ell] - \exp(-|\tilde{k}_4 - \tilde{k}_2|\ell)\right) \times \exp\left[\left|\tilde{k}_4 - \tilde{k}_2\right| - i(k_{2z} - k_{4z})\ell\right]\right]^{-1} \times \\
&\quad \times \exp\left[\left|-\left|\tilde{k}_4 - \tilde{k}_2\right| + i(k_{2z} - k_{4z})\ell\right|\ell\right]^{-1} - 1, \Delta \right\}.
\end{aligned}
\]

Here, \(P(z) = i k \frac{dn}{dT} \int_{-\infty}^{\infty} \delta T_0(z_i) dz_i\), \(\Delta = \left(\tilde{k}_1 + \tilde{k}_2 - \tilde{k}_3 - \tilde{k}_4\right)\) is the projection of the wave mismatch on the Z-axis. Expression (10) is written at quasicollinear propagation of waves \((k_k^{1,2,3} \pm k_{k,k,k}^{1,2,3} \pm 1)\) and fully describes the relation of the interacting waves spatial spectra in transparent two-component medium.
3. Discussion of the results

We consider the signal wave emanating from a point source located on the front edge of the nonlinear layer \( \hat{A}_{30}(\hat{k}_{3}) = 1 \). Let the first pump wave spatial spectrum changes according to the Gaussian law and the second pump wave is plane

\[
\hat{A}_{0}(\hat{k}_{1}) = \exp\left(-\frac{\kappa_{0}^{2}}{\kappa_{0}^{2}}\right), \quad \hat{A}_{20}(\hat{k}_{2}) = \delta(\hat{k}_{2} - \hat{k}_{20})
\]

(11)

where \( \kappa_{0} \) is a parameter which characterizes the first pump wave divergence, \( \hat{k}_{20} \) is a vector determining the propagation direction of the second pump wave.

In view of (11) in the paraxial approximation the expression for the spatial spectrum of the object wave (10) can be written as follows

\[
\hat{A}_{4}(\hat{k}_{4}) = -i \frac{n_{0}D_{1}D_{2}}{2\pi k} \int \frac{1}{\sinh(\kappa_{4} - \kappa_{20})} \left\{ \exp \left[ -\frac{1}{2k} \left( \kappa_{4} - \kappa_{20} \right)^{2} - \frac{\kappa_{0}^{2}}{4k^{2}} \right] \right\}
\]

\[
- \exp\left(-\frac{1}{2k} \left( \kappa_{4} - \kappa_{20} \right)^{2} \right) - \exp\left(-\frac{1}{2k} \left( \kappa_{4} - \kappa_{20} \right)^{2} \right)
\]

\[
\times \left\{ \text{erf} \left[ \frac{\kappa_{4} - \kappa_{20}}{2k} \right] - i \frac{\kappa_{20}(\kappa_{4} - \kappa_{20})}{\kappa_{4} - \kappa_{20} \kappa_{0}} + \text{erf} \left[ \frac{\kappa_{20}(\kappa_{4} - \kappa_{20})}{\kappa_{4} - \kappa_{20} \kappa_{0}} \right] \right\}
\]

(12)

where \( \text{erf}(x) = \frac{2}{\sqrt{\pi}} \int_{0}^{x} \exp(-t^{2}) dt \) is the error function.

In figure 1 we provide characteristics plots of the normalized module of the spatial spectrum of the object wave \( \left( A_{4}^{\ast}(\hat{k}_{4}) = \hat{A}_{4}(\hat{k}_{4}) \right) \) obtained using the expression (12).

If the pump waves propagating accurately toward each other are plane then the spatial spectrum module of the object wave with the growth of spatial frequency \( (\hat{k}_{4}) \) increases and it reaches a constant value. However, accounting pump waves divergence reduces the wave conversion efficiency at high spatial frequencies without changing the form of the spatial spectrum on the low spatial frequencies. In the pump wave flat \( (\hat{k}_{4} \hat{k}_{20} = \kappa_{4} \kappa_{20}) \) the second pump wave rotation shifts the band of spatial frequencies cut out by four-wave radiation converter on rotation value and narrows the spatial spectrum of the object wave. In the flat that is perpendicular to the pump waves flat \( (\hat{k}_{4} \hat{k}_{20} = 0) \) the pump wave rotation does not influence the spatial selectivity of four-wave radiation converter.
Figure 1. Spatial spectra modules of the object wave at $k\ell = 5 \times 10^3$, $\kappa_{20z} = 0$, $\kappa_{20z} k^{-1} = 2 \times 10^{-2}$, $\kappa_0 k^{-1} = 5 \times 10^{-2}$ (a), $1 \times 10^{-8}$ (b).

Four-wave radiation converter with the rotated second pump wave filters low spatial frequencies along with the high spatial frequencies due to the presence of the non-zero wave mismatch projection included explicitly in the last term of the expression (10). A similar behaviour of the spatial spectrum module form with changing of the wave mismatch is observed for nondegenerate four-wave radiation converter in transparent two-component medium [12].

When the second pump wave is plane and propagates accurately along the $Z$-axis the spatial spectrum of the object wave is symmetrical about an axis passing through the zero spatial frequency and depends on $\kappa_0 = |\kappa_0|$ (figure 2). To compare in the same figure we present the spatial spectrum module of the object wave of four-wave radiation converter on the Kerr nonlinearity calculated using the expression [1]

$$\tilde{A}_4(\tilde{\kappa}_4) = iG \text{erf} \left( \frac{\kappa_0 \kappa_0}{2k} \ell \right).$$

(13)

Figure 2. Dependence of the spatial spectra modules from the spatial frequency value at $k\ell = 5 \times 10^3$, $\tilde{\kappa}_{20z} = 0$, $\kappa_0 k^{-1} = 5 \times 10^{-2}$ (1, 3), $1 \times 10^{-8}$ (2), transparent two-component medium (1, 2), medium with Kerr nonlinearity (3).
Here, $G$ is the parameter that determines the efficiency of four-wave radiation converter.

In constructing graphs we equated the spatial spectra modules of the object waves of two four-wave radiation converters at spatial frequency $\kappa_0 = 5\kappa_0$. As seen, the spatial selectivity of four-wave radiation converters in transparent liquid containing nanoparticles and medium with Kerr nonlinearity is the same at high spatial frequencies.

In the area of spatial frequencies cut out by four-wave radiation converter the object wave amplitude is independent on the divergence and pump wave rotation.

To characterize the spatial selectivity of four-wave radiation converter in the pump waves flat we use the bandwidth of the most efficiently converted spatial frequencies defined as difference between spatial frequencies at which the maximum value of spatial spectrum module of the object wave is reduced by half (figure 2) [12].

Without the second pump wave rotation the reduction of the bandwidth of the most efficiently converted spatial frequencies with increasing of the first pump wave divergence (figure 3) is well described by the expression of the form

$$\frac{\Delta \kappa}{k} = a \left( \frac{\kappa_0}{k} \right) - b.$$

Here, $a$ and $b$ are the parameters which are inversely proportional to the multiplication of the wave number on nonlinear layer thickness. As for the values $k\ell = 1.5 \times 10^4$, $5 \times 10^4$, $1.5 \times 10^5$ parameters in the expression (14) take the values $a = 2.4 \times 10^{-3}$, $7.2 \times 10^{-4}$, $2.4 \times 10^{-4}$ and $b = 5 \times 10^{-3}$, $2 \times 10^{-3}$, $5 \times 10^{-4}$.

![Figure 3](image1.png)  
**Figure 3.** Dependence of the bandwidth of spatial frequencies from the pump wave divergence at $\kappa_{20} = 0$, $k\ell = 1.5 \times 10^3$ (1), $5 \times 10^3$ (2), $1.5 \times 10^4$ (3).

![Figure 4](image2.png)  
**Figure 4.** Dependence of the bandwidth of spatial frequencies from the pump wave rotation at $k\ell = 5 \times 10^4$, $\kappa_j k^{-1} = 5 \times 10^{-2}$ (1), $1 \times 10^{-2}$ (2).

Within the bandwidth of spatial frequencies $\Delta \kappa$ accounting the pump waves divergence $\left( \kappa_0 / k < 5 \times 10^{-2} \right)$ insignificantly changes the spatial spectrum phase of the object wave.

When $\kappa_0 \neq 0$ the second pump wave rotation leads to additional reduction of the wave conversion efficiency at high spatial frequencies in the pump waves flat. With the growth of the second pump wave rotation the bandwidth of spatial frequencies reduces monotonically (figure 4). When $\kappa_{20} \leq \kappa_0$ the dependence of bandwidth of the most efficiently converted spatial frequencies from the second pump wave rotation is well described by a Gaussian law

$$\Delta \kappa = \Delta \kappa_0 \exp \left( -b \frac{\kappa_{20}^2}{k^2} \right).$$
Here, $\Delta k_0$ is the bandwidth of the most efficiently converted spatial frequencies without the second pump wave rotation, $b$ is the coefficient that depends on the value of the first pump wave divergence. When the first pump wave divergence $\kappa_1 k^{-1} = 1 \times 10^{-2}$ and $5 \times 10^{-2}$ coefficient $b$ takes the values 50 and 10 respectively.

When $\kappa_2 > 2\kappa_0$, the bandwidth of spatial frequencies most efficiently converted at four-wave mixing is independent on the first pump wave divergence and is fully determined by the second pump wave rotation.

4. Conclusion
As a result of the paper, shown that in the area of spatial frequencies cut out by the degenerate four-wave radiation converter the object wave amplitude is independent on the spatial structure of the pump waves. The divergence and pump waves rotation reduce the wave conversion efficiency at high spatial frequencies without changing the spatial spectrum form of the object wave at low spatial frequencies. We obtained the expressions relating the bandwidth of spatial frequencies most efficiently converted at degenerate four-wave mixing with the divergence and pump waves rotation.

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