

COHERENCE

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Transformation of spatial coherence of pulse laser radiation in a thin layer of a medium with thermal nonlinearity

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Abstract. **Correlation properties of light beams upon self-phase modulation in a thin layer of a medium with thermal nonlinearity are studied. It is found out that the efficiency of spatial coherence transformation of radiation is determined by the ratio of the characteristic coherence radius of the initial radiation to the size of phase inhomogeneities induced in the nonlinear medium.**

Keywords: *spatial coherence, laser beams, nonlinear optics, speckle noise*.

1. Introduction

Transformation of spatial and temporal spectra of coherent light beams in nonlinear media has been studied since the very first publications on nonlinear optics [1]. However, the research activity in this field was predominantly theoretical and was mainly devoted to the physical aspects of these processes. The interest in this field was revived in recent years following the development of high-tech lasers generating a wide range of radiation frequencies and offering bright prospects of their use in various technical applications. Several research groups paid special attention to develop practical methods for transforming the coherence of laser beams by nonlinear methods. Note that both an increase [2 – 4] and decrease [5 – 10] in the laser radiation coherence are important.

The high spatial coherence of radiation is responsible for the appearance of coherent noise (speckle noise), which restricts the resolving power of optical systems and lowers the signal-to-noise ratio in optical communication systems. The coherence of pulsed laser beams can be reduced, for example, by self-phase modulation of radiation upon its interaction with a nonlinear medium [10]. The nonstationary and nonuniform modulation over the light beam cross section, which takes place upon a nonlinear interaction of radiation with the medium, violates spatial coherence in an initially coherent field and makes it possible to transform the coherence in a short period of time with a relatively small broadening of the temporal spectrum and a small beam divergence [7, 11 – 14]. Note, however, that no quantitative experimental investigations have been carried out in this direction so far.

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In this paper we study the efficiency of transformation of spatial coherence of multimode pulsed radiation during self-action in a thin layer of a medium with a reactive cubic (e.g., thermal) nonlinearity. In this case, a high value of the transformed radiation energy can be achieved, which is important when waves in the far UV spectral region are used, for example, in photolithography.

Analysis of pulse coherence transformation is based on the formalism of expansion of light fields in spatially coherent modes [7, 9, 15]. Such an approach provides a simple method for estimating the degree of transformation of spatial coherence of radiation by measuring the contrast of spatial fluctuations of its intensity before and after interaction with a nonlinear medium.

2. Theoretical model

The input radiation transformation in an optically thin layer of a nonlinear medium with a thickness $L < L_{\rm nl} < L_{\rm d}$ (here, $L_{\rm nl}$ and $L_{\rm d}$ are the characteristic lengths of nonlinear refraction and diffraction spread, respectively [12]) is determined by the local characteristics of radiation. Within the framework of the Raman – Nath approximation [16] for a medium with a cubic nonlinearity and a sufficiently slow relaxation ($t \ll \tau_{\rm nl}$, where t, $\tau_{\rm nl}$ are the pulse duration and the nonlinearity relaxation time, respectively), this process is described as a self-phase modulation [7, 11, 12]:

$$E(\boldsymbol{r}, z = L, t) = E_0(\boldsymbol{r}, z = 0, t) \exp\left(\mathrm{i}\eta \int_{-\infty}^{t} I_0(\boldsymbol{r}, t)\mathrm{d}t\right), \quad (1)$$

where $I_0(\boldsymbol{r}, t) = |E_0(\boldsymbol{r}, t)|^2$ is the input radiation intensity and $\eta = \pi\delta L/\lambda n$ is a nonlinear interaction parameter, which depends on the nonlinear variation δ of the permittivity, layer thickness L, refractive index n of the medium, and radiation wavelength λ. The nonstationary and nonuniform self-modulation acts here as a moving phase plate and violates the spatial coherence of radiation [6]. We shall consider the limiting case of inertial interaction with 'infinite memory' because the most efficient transformation of the spatial coherence of a radiation pulse is observed precisely in this case [7].

Assuming that only phase fluctuations occur in the radiation (this is indeed the case for a laser in which pumping considerably exceeds the lasing threshold), averaging of expression (1) allows us to present the spatial mutual coherence function in the form

$$\Gamma(\boldsymbol{r}_1,\boldsymbol{r}_2,t)=\Gamma_0(\boldsymbol{r}_1,\boldsymbol{r}_2,t)\times\exp\left[\mathrm{i}\eta\int_{-\infty}^{t}(I_0(\boldsymbol{r}_1,t')-I_0(\boldsymbol{r}_2,t'))\mathrm{d}t'\right]. \quad (2)$$

Here $\Gamma(\boldsymbol{r}_1,\boldsymbol{r}_2,t)=\langle E(\boldsymbol{r}_1,t)E^*(\boldsymbol{r}_2,t)\rangle$, and $\Gamma_0(\boldsymbol{r}_1,\boldsymbol{r}_2,t)$ are the correlation functions of radiation emerging from the medium and entering it, respectively. To simplify the subsequent calculations, we also make an additional assumption about the mutual spectral purity of the input radiation [17], when $I_0(\boldsymbol{r},t)=I(\boldsymbol{r})I(t)$.

For inertial interaction of type (1), we use the substitution of variable $t\to\zeta$ [18], where

$$\zeta=\frac{1}{W}\int_{-\infty}^{t}I(t)\mathrm{d}t,\quad \mathrm{d}\zeta=\frac{I(t)}{W}\mathrm{d}t. \quad (3)$$

Here, W is the energy density, and the variable ζ changes from 0 to 1. After the substitution of variables (3), a pulse with an arbitrary envelope becomes equivalent to a rectangular pulse:

$$I'_0(\zeta)=\begin{cases}W, & 0\leqslant\zeta\leqslant 1,\\ 0, & \zeta<0,\ \zeta>1.\end{cases} \quad (4)$$

It follows from (2)–(4) that in case of averaging over the recording time (which coincides with the pulse duration), the spatial correlation function $\Gamma^{(S)}(\boldsymbol{r}_1,\boldsymbol{r}_2)=\int\langle E(\boldsymbol{r}_1,t)E^*(\boldsymbol{r}_2,t)\rangle\mathrm{d}t$ of the radiation emerging from the nonlinear medium has the form

$$\Gamma^{(S)}(\boldsymbol{r}_1,\boldsymbol{r}_2)=\Gamma_0^{(S)}(\boldsymbol{r}_1,\boldsymbol{r}_2)\frac{\mathrm{i}}{\eta W}\times\frac{1-\exp\{\mathrm{i}\eta W[I_0(\boldsymbol{r}_1)-I_0(\boldsymbol{r}_2)]\}}{I_0(\boldsymbol{r}_1)-I_0(\boldsymbol{r}_2)}, \quad (5)$$

where ηW is the pulse-averaged phase shift in the medium.

The main parameter characterising the transformation of coherence of a light beam in the modal approach is the effective number of terms in the expansion

$$N_{\mathrm{eff}}=\frac{\left[\int\Gamma^{(S)}(\boldsymbol{r},\boldsymbol{r})\mathrm{d}^2r\right]^2}{\int\int|\Gamma^{(S)}(\boldsymbol{r}_1,\boldsymbol{r}_2)|^2\mathrm{d}^2r_1\mathrm{d}^2r_2}.$$

The parameter N_{eff} is directly related to the contrast C of the speckle pattern observed after the scattering of radiation from a fine-structure diffuser [15]:

$$C\equiv\frac{\langle I^2\rangle^{1/2}}{\langle I\rangle}\approx\frac{1}{N_{\mathrm{eff}}^{1/2}}. \quad (6)$$

We shall assume that the spatial correlation function of the input field can be represented in the form of a Shell model [17] with a Gaussian correlation coefficient and the input intensity profile

$$\Gamma_0^{(S)}(\boldsymbol{r}_1,\boldsymbol{r}_2)=[I_0(\boldsymbol{r}_1)I_0(\boldsymbol{r}_2)]^{1/2}\exp[-(\boldsymbol{r}_1-\boldsymbol{r}_2)^2/b^2],$$
$$I_0(\boldsymbol{r})=\exp(-r^2/a^2) \quad (7)$$

(the effective number of modes in such a field is $N_0=1+4a^2/b^2$). The Gaussian intensity distribution [19] provides the most efficient coherence destruction for spatially coherent input pulses.

Using the approximation $I_0(\boldsymbol{r})\approx 1-\ 2r^2/a^2$, which is valid for the axial region of a light beam with $r\sim a$ [20], we arrive, after certain cumbersome transformations, at the analytic dependence of the effective number of modes emerging from a nonlinear medium on the other parameters of the problem:

$$N_{\mathrm{eff}}=\frac{\eta^2W^2}{4}\left[\frac{\eta W}{\sqrt{N_0}}\arctan\left(\frac{\eta W}{2\sqrt{N_0}}\right)-\ln\left(1+\frac{\eta^2W^2}{4N_0}\right)\right]^{-1}. \quad (8)$$

For $\eta W\gg\sqrt{N_0}$, expression (8) has an especially simple form:

$$\frac{N_{\mathrm{eff}}}{N_0}=\frac{\eta W}{2\pi\sqrt{N_0}}. \quad (9)$$

Thus, a decrease in the overall degree of coherence $\mu=1/N_0$ of the input pulse leads to a decrease in the nonlinear transformation efficiency by a factor of N_{eff}/N_0.

3. Experimental investigation of coherence transformation

Figure 1 shows the optical scheme of the setup used for studying the destruction of the spatial coherence of radiation upon interaction with a nonlinear medium. 266-nm radiation (*1*) from a single-pulse laser is focused by lens (*2*) into cell (*3*) filled with a nonlinear liquid CCl_4. The emerging radiation falls on ground quartz plate (*4*) placed right behind the cell. The extent of spatial coherence destruction of radiation is estimated from the change in the contrast of the speckle pattern recorded by linear CCD detector (*5*) before and after interaction with the medium in the far-field zone of the light field scattered from the ground quartz plate. The cell thickness was chosen equal to the smaller of the lengths L_{nl}, and L_{d} defined for a Gaussian distribution of light beam intensity by the relations [12]

$$L_{\mathrm{nl}}=a\left(\frac{n}{2\Delta n}\right)^{1/2},\quad L_{\mathrm{d}}=\frac{\pi a^2}{\lambda\sqrt{N_0}}.$$

Here, $2a$ is the diameter of the focal spot and $\Delta n=\delta W/n$ is the nonlinear correction to the refractive index of the medium, which depends on the parameters of the medium and the radiation energy density. The layer thickness L in our experiments was ~ 250 μm, and $2a$ was ~ 60 μm.

We studied the efficiency of transformation of spatial

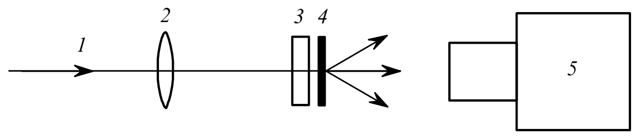

Figure 1. Optical scheme of the experimental setup: (*1*) input radiation; (*2*) lens; (*3*) cell; (*4*) ground quartz plate; (*5*) CCD detector.

coherence of radiation, which is defined as the ratio of the effective number of spatial modes entering the medium to the number of modes emerging from it. Transverse laser mode selection with the help of diaphragms of various diameters, inserted in the laser cavity, was used to change the degree of radiation coherence at the input to the medium.

Figure 2 shows the dependences of the efficiency of transformation of spatial radiation coherence on the initial radiation coherence, which were calculated by using expression (8) and the measurements of the speckle pattern contrast. Calculations were made under the assumption that the thermal mechanism of interaction predominates [21, 22], i.e.,

$$\delta = \frac{\partial n}{\partial T}\frac{\alpha n}{\rho c_p}, \qquad (10)$$

where $\partial n/\partial T$ is the temperature coefficient of the refractive index; α is the absorption coefficient; c_p is the specific heat at a constant pressure; and ρ is the density of the nonlinear medium.

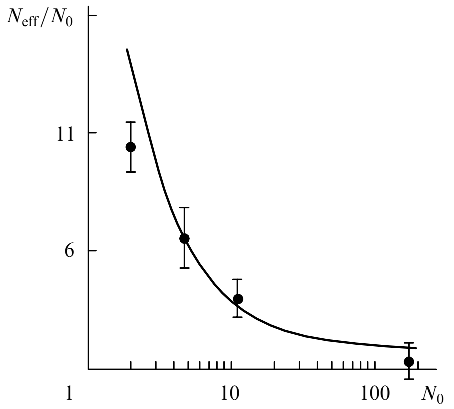

Figure 2. Calculated (solid curve) and experimental (circles) dependences of the transformation efficiency of spatial coherence of radiation on its initial coherence.

Analysis of the conditions of interaction of radiation with a nonlinear medium shows that for the used input pulse duration of 15 – 20 ns and a scale of the induced phase inhomogeneity $d = 2\pi/|q|$ (where $|q| = \Theta k$ is the modulus of the wave vector of scattering and Θ is the radiation scattering angle), sound propagates a distance d over a time equal to the pulse duration. Accordingly, thermal expansion of the substance may take place as well as the formation of a pressure wave accompanied by a considerable change in the permittivity of the medium [23]. Hence, in order to estimate the efficiency of transformation of the number of radiation modes during self-phase modulation in a nonlinear medium, we can take the maximal temperature variation $(\partial n/\partial T)_p$ of the refractive index of the medium, which is equal to -5.5×10^{-4} K^{-1} for CCl_4 (p is the steady-state pressure in the medium) [24]. The following values of the parameters of CCl_4 were used for theoretical estimates [25, 26]: $c_\rho = 0.84$ J g^{-1} K^{-1} is the specific heat of CCl_4 for a constant density $\rho = 1.63$ g cm^{-3}, the average laser radiation energy $E = 1.5$ mJ, $\alpha = 1.23$ cm^{-1}, $\Theta \sim 7\times 10^{-4}$ rad, and the spectral width $\Delta\omega \sim 0.3$ cm^{-1}.

4. Conclusions

The results presented in Fig. 2 [see also expressions (8) and (9)] show that the transformation of spatial coherence of radiation with a given energy in a nonlinear medium is more efficient for a high initial coherence of radiation. These results differ from those obtained earlier by the delay line method [9], when the efficiency of coherence suppression is independent of the correlation properties of the initial pulse. In particular, for the scheme described in [9], the following expression is valid for partial beams of equal intensity in the absence of a correlation between them:

$$N_{\rm eff} = N_0 P,$$

where P is the number of partial waves in the scheme. In other words, the setup proposed by us earlier serves as an 'ideal filter' for spatial coherence of radiation.

A comparative analysis of the transformation of spatial coherence of radiation in a nonlinear medium and using a nonstationary phase screen with a phase inhomogeneity dispersion $\sigma_{\rm ph} \gg 1$ [20] shows that the transformation efficiency $N_{\rm eff}/N_0$ in both cases is determined by the ratio of the coherence radius of the initial radiation to the characteristic size of phase inhomogeneities of the converter. However, the transformation rules are slightly different in these cases. Indeed, for $N_0 \gg 1$, expression (9) can be written in the form

$$\left(\frac{N_{\rm eff}}{N_0}\right)_{\rm nl} \approx \frac{b}{b_0}, \qquad (11)$$

where b is the coherence radius of the input radiation and $b_0 = 4\pi a/\eta W$ is the radius of the induced phase inhomogeneity of the nonlinear medium.

For a nonstationary phase screen, we have

$$\left(\frac{N_{\rm eff}}{N_0}\right)_{\rm ph\,s} \approx 2\sigma_{\rm ph}^2\frac{b^2}{s^2}, \qquad (12)$$

where s is the correlation radius of phase inhomogeneities of the screen. We assume that the beam diameter is much larger than the size of inhomogeneities of the screen.

It follows from expressions (11) and (12) that for partially coherent input radiation, the reduction of the spatial coherence by using nonlinear methods is less efficient than its 'mechanical' transformation. Apparently, this is due to inertial nonlinear phase modulation effects.

References

1. Akhmanov S.A., Sukhrukov A.P., Khokhlov R.V. *Usp. Fiz. Nauk*, **93**, 19 (1967).
2. Klyukach I.L., Sokolovskii R.I. *Zh. Eksp. Teor. Fiz.*, **71**, 424 (1976).
3. Abdulin U.A., Gorshkov V.A., Klyukach I.L., Lyakhov G.A., Orlov R.Yu., Telegin L.S. *Kvantovaya Elektron.*, **2**, 967 (1975) [*Sov. J. Quantum Electron.*, **5**, 526 (1975)].
4. Kotov V.M., Shkerdin G.N., Shkerdin D.G. *Opt. Spekrosk.*, **93**, 313 (2002).
5. Partlo W.N., Oldham W.G. *J. Vac. Sci. Technol. B*, **9**, 3126 (1991).
6. Wang L., Tschudi T., Halldórsson T., Pétursson P.R. *Appl. Opt.*, **37**, 1770 (1998).
7. Karelin N.V., Lazaruk A.M. *Opt., Spekrosk.*, **83**, 403 (1997).

8. Kotov V.M., Shkerdin G.N., Shkerdin D.G., Bulyuk A.N., Tikhomirov S.A. *Kvantovaya Elektron.*, **31**, 839 (2001) [*Quantum Electron.*, **31**, 839 (2001)].
9. Ivakin E.V., Kitsak A.I., Karelin N.V., Lazaruk A.M., Rubanov A.S. *Kvantovaya Elektron.*, **33**, 255 (2003) [*Quantum Electron.*, **33**, 255 (2003)].
10. Kohler D., Seitz W.L., Loree T.R., Gardner S.D. *Opt. Commun.*, **12**, 24 (1974).
11. Akhmanov S.A., Vysloukh V.A., Chirkin A.S. *Optika femtosekundnykh lazernykh impulsov* (Optics of Femtosecond Laser Pulses) (Moscow: Nauka, 1988).
12. Aleshkevich V.A., Kozhoridze G.D., Matveev A.N. *Kvantovaya Elektron.*, **15**, 829 (1988) [*Sov. J. Quantum Electron.*, **18**, 529 (1988)].
13. Karelin M.U., Lazaruk A.M. *Proc. SPIE Int. Soc. Opt. Eng.*, **4243**, 17 (2001).
14. Ivakin E.V., Kitsak A.I., Karelin N.V., Lazaruk A.M., Rubanov A.S. *Proc. SPIE Int. Soc. Opt. Eng.*, **4751**, 34 (2002).
15. Leshchev A.A. *Opt. Spekrosk.*, **55**, 988 (1983).
16. Lazaruk A.M. *Opt. Spekrosk.*, **53**, 1057 (1982).
17. Mandel L., Wolf E. *Optical Coherence and Quantum Optics* (Cambridge: Univer. Press, 1995; Moscow: Fizmatgiz, 2000).
18. Lazaruk A.M., Karelin N.V., in *Proceedings of the 3rd Conference on Laser Physics and Spectroscopy* (Grodno, 1997) Vol. 2, pp 70–73.
19. Karelin M., Lazaruk A. *Proc. SPIE Int. Soc. Opt. Eng.*, **3573**, 71 (1998).
20. Akhmanov S.A., D'yakov Yu.E., Chirkin A.S. *Vvedenie v statisticheskuyu radiofiziku i optiku* (Introduction to Statistical Radiophysics and Optics) (Moscow: Nauka, 1981).
21. Bespalov V.I., Litvak A.G., Talanov V.I., in *Proceedings of the 2nd All-Union Seminar on Nonlinear Optics* (Novosibirsk: Nauka, 1968) pp 428–463.
22. Petrishchev V.A., Talanov V.I. *Kvantovaya Elektron.*, (6), 35 (1971) [*Sov. J. Quantum Electron.*, **1** (6), 587 (1971)].
23. Bespalov V.I., Pasmanik G.A. *Nelineinaya optika i adaptivnye lazernye sistemy* (Nonlinear Optics and Adaptive Laser Systems) (Moscow: Nauka, 1986).
24. Ravdel' A.A., Ponomareva A.M. (Eds) *Kratkii spravochnik fiziko-khimicheskikh velichin* (Handbook of Physical and Chemical Quantities) (Leningrad: Khimiya, 1983).
25. Ioffe B.V. *Refraktometricheskie metody khimii* (Refractometric Methods in Chemistry) (Leningrad: Khimiya, 1983).
26. Nikol'skii B.P. (Ed.) *Spravochnik khimika* (Handbook of Chemistry) (Leningrad–Moscow: GNTI khim. lit., 1962) Vol. 1.