Nonlinear optics of field with high angular momentum

S.D. Tvorogov

Institute of Atmospheric Optics,
Siberian Branch of the Russian Academy of Sciences, Tomsk

Received April 20, 2000

One of the main methodical problems of nonlinear optics of a field with high angular momentum, namely, spatial dispersion of the magnetic moment of a unit volume is discussed. It is shown that such a structure appears in ordinary quantum calculation just because of the high angular momentum of field.

1. The density of the angular momentum $\mathbf{m}$ of an electromagnetic field is proportional to the radiation intensity I, and if we assume that the field momentum is created only by I, then, as elementary estimates show, it turns out to be comparable with the angular momentum of a molecule (that is, its value is about the Planck's constant $\hbar$) for fields with the strength exceeding 10^9 V/cm. This sends us to search variants, when the field possesses moderate intensity while having high angular momentum. The solution (in the sense of "existence and uniqueness") is given in Ref. 1 based on analysis of the problem by applying the rules of quantum electrodynamics.

Assuming the field to be set by a preset current, we believe that, according to the results of quantum optics,[2,3] its wave function is a coherent state with the index $\alpha = \alpha(\mathbf{k})$ which is the function of the wave vector $\mathbf{k}$. The basis vectors $\mathbf{k}$ should form a cone with a small angle at its vertex, and $|\partial\psi/\partial\varphi|$ should be large enough. Here φ is the polar angle between $\mathbf{k}$ and the cone axis, and ψ is the phase of the complex parameter $\alpha = |\alpha| \exp(i\psi)$. It is just the large derivative of ψ that leads to high angular momentum of the field, whereas the field intensity is determined only by $|\alpha|$. The possibility of generating such a field was discussed in Refs. 4–6.

Now we can imagine the outline of the electrodynamics of the field with high angular momentum.

First, as such a field interacts with molecules (we mention molecules only for definiteness), the latter acquire (according to the well-known conservation laws) mechanical momentum, whose appearance necessarily implies (see, e.g., Ref. 7) the magnetic moment $\mathbf{M}$ of a unit volume. In the case of optical frequencies it is usually ignored, but now the current density $\mathbf{j}$ in the Maxwell equations should be written as

$$\mathbf{j} = \frac{\partial \mathbf{P}}{\partial t} + c \text{ rot } \mathbf{M} \qquad (1)$$

with the dipole moment $\mathbf{P}$ of the unit volume (c is the speed of light).

Second,

$$\mathbf{m} = [\mathbf{r}(\mathbf{E} \times \mathbf{H})]/(4\pi c) \qquad (2)$$

with the strengths $\mathbf{E}$ and $\mathbf{H}$ of the electric and magnetic fields at the point $\mathbf{r}$, and of course

$$\mathbf{M} = \mathbf{M}(\mathbf{m}). \qquad (3)$$

From Eqs. (1)–(3) it is clear that optics turns out to be nonlinear even at quite moderate I values.

Third, it is clear that Eq. (3) cannot have the structure $\mathbf{M}(\mathbf{r}) \sim \mathbf{m}(\mathbf{r})$, because $|\mathbf{m}| \to \infty$ at $|\mathbf{r}| \to \infty$. It also cannot be assumed that $\mathbf{M}$ is proportional to the total field momentum (integral of Eq. (2) over $\mathbf{r}$), because the Maxwell equations are local. The variant like "spatial dispersion"

$$\mathbf{M}(\mathbf{r}) = \int d\mathbf{r}' \, F(\mathbf{r} - \mathbf{r}') \, \mathbf{m}(\mathbf{r}') \qquad (4)$$

with the corresponding function F (tensor in the general case) ought to be consistent.

2. The construction presented by Eq. (4) is a subject of semi-classical electrodynamics, in which the Hamiltonian of interaction of field particles

$$\hat{H}_{OR} = -\sum_a \frac{e_a}{\mu_a c} \mathbf{p}_a \mathbf{A}(\mathbf{r}_a, t) + \sum_a \frac{e_a^2}{2\mu_a c^2} \mathbf{A}^2(\mathbf{r}_a, t) \qquad (5)$$

appears. In Eq. (5) e_a, μ_a, $\mathbf{r}_a$, and $\mathbf{p}_a = -i\hbar \text{ grad}_{\mathbf{r}_a}$ are the charge, mass, coordinate (as an argument of the wave function), and operator of momentum of a particle with the index a; $\mathbf{A}(\mathbf{r}_a, t)$ is the vector potential of the classic field.

Usually, the spatial dispersion is caused by interaction of molecules from different elementary volumes, while here, the non-local spatial coupling (4) occurs due to high momentum of the field. However, formal consequences are the same: we should refuse from the long-wave approximation in Eq. (5), by using instead its "phase transformation" following known (with only insignificant variations) scheme.[8,9] Then the following expression is to appear:

0235-6880/00/05 423-04 $02.00

$$\hat{H}_{0R} = -\sum_a e_a \mathbf{r}_a \mathbf{E}(\mathbf{r}_a, t) - \\ -\frac{1}{2c}\sum_a \frac{e_a}{\mu_a}\left(\mathbf{r}_a \times \mathbf{H}(\mathbf{r}_a, t)\right)\mathbf{p}_a . \quad (6)$$

The terms in Eq. (6) are classified as description of the field interaction with the electric and magnetic dipoles. The electric quadrupole has the same order of magnitude as the second term in Eq. (6), but its influence can be neglected. Actually, it does not contribute to Eq. (3) since its product with the "magnetic" terms does not exceed the accuracy of Eq. (6), and the electric dipole approximation is quite sufficient for calculating $\mathbf{P} = \mathbf{P}(\mathbf{E})$.

Calculations are further continued according to typical (see, e.g., Refs. 9 and 10) procedure of quantum mechanics for nonlinear optics. The term proportional to $\mathbf{H}$ appears in the first order of the perturbation theory in terms of the operator (6). It is just this term that determines the dependence of $\mathbf{j}$ on $\mathbf{H}$ for the "ordinary" field, and therefore it is insignificant. We need to follow up how the combination (2), in the form (4), appears; for that we need the second order of the perturbation theory. Then (accurate to a constant factor) the parameter (3) is

$$\left(\frac{1}{i\hbar}\right)^2 \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \,\mathrm{Sp}\,\hat{\rho}\,[\hat{\mathbf{M}}'(t), \hat{H}'_{OR}(t_1)\,\hat{H}'_{OR}(t_2)] = \\ = \left(\frac{1}{i\hbar}\right)^2 \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \,\mathrm{Sp}\,\hat{\rho} \times \\ \times\Big[\mathrm{e}^{-\frac{t}{i\hbar}\hat{H}_0}\hat{\mathbf{M}}\mathrm{e}^{\frac{t}{i\hbar}\hat{H}_0}, \underline{\mathrm{e}^{-\frac{t_1}{i\hbar}\hat{H}_0}\hat{H}_{OR}(t_1)\mathrm{e}^{\frac{t_1-t_2}{i\hbar}\hat{H}_0}}\,\hat{H}_{OR}(t_2)\underline{\underline{\mathrm{e}^{\frac{t_2}{i\hbar}\hat{H}_0}}}\Big]. \quad (7)$$

Here t is time; $\hat{\rho}$ is the Gibbs density matrix of the system of particles unperturbed by the field; the prime denotes the transition to the interaction representation

$$\hat{K}'(t) = \mathrm{e}^{-\frac{t}{i\hbar}\hat{H}_0}\,\hat{K}\,\mathrm{e}^{\frac{t}{i\hbar}\hat{H}_0}$$

for the operator $\hat{K}$ with the Hamiltonian of the system of particles $\hat{H}_0 = \hat{T} + V$ ($\hat{T}$ is the operator of kinetic energy; V is the potential energy); finally we have that

$$\hat{\mathbf{M}} = \sum_a \delta(\mathbf{r} - \mathbf{r}_a)\frac{e_a}{\mu_a}(\mathbf{r}_a \cdot \mathbf{p}_a) \quad (8)$$

is the density operator of the magnetic moment (δ is the delta function).

It is clear that, after substitution of Eq. (6) into Eq. (7) the subject of analysis are the terms including the products of $\mathbf{E}$ and $\mathbf{H}$. Expressions with $\mathbf{E}^2$ and $\mathbf{H}^2$ are the second-order corrections of the perturbation theory in Eq. (1), which are insignificant at low intensity. Let us recall here once more that it is the angular momentum of the field, which has high value, just because of the assumptions on the field structure.

A prerequisite for the following simplification of Eq. (7) is the almost obvious statement that interaction of the field having high angular momentum and a molecule results in an intense rotation of the latter.

This allows us, first, to ignore the commutator Eq. (8) and $\hat{H}_0$ and, consequently, assume $\mathbf{M}'$ and $\exp\left(\pm(t/i\hbar)\hat{H}_0\right)$ to be permutative (nevertheless, the need in the approximation $\hat{\mathbf{M}}' \cong \hat{\mathbf{M}}$ does not follow herefrom, since it proves to be too rough for the considered equation). Actually,

$$[\delta(\mathbf{r} - \mathbf{r}_a), \hat{H}_0] = [\delta(\mathbf{r} - \mathbf{r}_a), \hat{T} + V] = [\delta(\mathbf{r} - \mathbf{r}_a), \hat{T}] .$$

However, the flux of particle density through the boundary of an elementary volume, which is obviously zero at only rotation of particles, is connected with the commutator $[\delta(\mathbf{r} - \mathbf{r}_a), \hat{H}_0]$ (as it is proved in statistical physics[11]). Then, permutability of $(\mathbf{r}_a \cdot \mathbf{p}_a)$ and $\hat{T}$ is checked directly, and $V = \mathrm{const}$ is a good approximation for description of rotation.

Now we have the possibility of placing the exponential operator underlined by one line in Eq. (7) outside the square brackets [ ] sign, use its exact commutation with $\hat{\rho}$, and place it outside the [ ] sign in the expression under the Sp sign. Then the combination appears

$$\hat{H}_{OR}(t_1)\,\mathrm{e}^{-\frac{t_1-t_2}{i\hbar}\hat{H}_0}\,\hat{H}_{OR}(t_2)\,\mathrm{e}^{-\frac{t_1-t_2}{i\hbar}\hat{H}_0}.$$

Similarly, but in the reverse order, we can proceed with the exponential operator underlined by two lines. Thus, we have the expression

$$\mathrm{e}^{-\frac{t_1-t_2}{i\hbar}\hat{H}_0}\,\hat{H}_{OR}(t_1)\,\mathrm{e}^{-\frac{t_1-t_2}{i\hbar}\hat{H}_0}\,\hat{H}_{OR}(t_2) .$$

The first or second variants are used to remove the exponential factors from the operator $(\mathbf{r}_a\,\mathbf{E}(\mathbf{r}_a, t))$ in the case that this expression (in the product with $\mathbf{H}(\mathbf{r}_a, t)$) is on the left or right-hand side. In the other factor, after a simple vector transformation, we derive $\hat{\mathbf{M}}_a$ – the term from Eq. (8):

$$\mathrm{e}^{-\frac{t}{i\hbar}\hat{H}_0}\,\mathbf{H}(\mathbf{r}_a, t)\,\hat{\mathbf{M}}_a\,\mathrm{e}^{-\frac{t}{i\hbar}\hat{H}_0} = \mathbf{H}(\mathbf{r}'_a(t), t)\,\hat{\mathbf{M}}'_a.$$

At a sufficiently intense rotation it can be treated as a classic one with the initial value $\mathbf{r}_a$, and the "trajectory" $\mathbf{r}'_a(t)$ can be presented as $\mathbf{r}_a$ + "something"; this "something" = 0 (size of a molecule), if we deal with rotation. However, the spatial coordinate enters into $\mathbf{H}$ as $\mathbf{k} \cdot \mathbf{r}$ with $k \sim 1/$(wavelength). Therefore, the value $\mathbf{k}$ multiplied by "something" can be ignored, and $\mathbf{H}(\mathbf{r}'_a) \to \mathbf{H}(\mathbf{r}_a)$ in the last expression.

The meaning of such a transformation is rather simple. The consequence of Eq. (2) is $\mathbf{m} = (\mathbf{r}\,\mathbf{E})\,\mathbf{H} - (\mathbf{r}\,\mathbf{H})\,\mathbf{E}$, and $\mathbf{M}$ having oriented by $\mathbf{H}$ is connected

only with the first term. However, it is just it that appears because of the above actions.

After the above simplifications and calculation of Sp in Eq. (7), the subject of analysis become expressions of the form

$$\sum_{n_1 \ldots n_4} \rho_{n1}\, e^{i\omega_{n_1 n_2} t}\, B_{n_1 n_2}\, C_{n_2 n_3}(\omega')\, e^{-i\omega'(t-\tau_1)}\, D_{n_3 n_4}(\omega'') \times \times e^{-i\omega''(t-\tau_1-\tau_2)}\, G_{n_4 n_1}\, e^{i\omega_{n_5 n_1}\tau_2}\, e^{-i\omega t}. \quad (9)$$

As to Eq. (9), the detailed explanation should be given (we wrote down here only one term arising in the commutator from Eq. (7)).

The subscript n numbers the eigenfunctions of $\hat{H}_0$, ρ_n is the diagonal matrix element of $\hat{\rho}$; $\omega_{nn'} = (\varepsilon_n - \varepsilon_{n'})/\hbar$ with ε_n – eigenvalues of $\hat{H}_0$; exponents with $\omega_{nn'}$ follow from the interaction representation. In Eq. (7) the replacement of the variables $t_2 = t_1 - \tau_2$ and $t_1 = t - \tau_1$ were made in turn, then $\mathbf{E}(t)\ \mathbf{H}(t)$ was expanded into the Fourier integral (with the components $\mathbf{E}(\omega')$ and $\mathbf{H}(\omega'')$ and integration over the frequencies ω' and ω''). Thus appear the exponents with ω' and ω'' in Eq. (9) and the Fourier transform of $\mathbf{M}(t)$ itself with the frequency ω is considered. This is demonstrated by $\exp(i\omega t)$. The designations $B_{nn'}$, $C_{nn'}$, $D_{nn'}$, and $G_{nn'}$ are used for the matrix elements of the operator (8), $(\mathbf{r}_a\ \mathbf{E}(\mathbf{r}_a, \omega'))$, one of the vector components of $\mathbf{H}(\mathbf{r}_a, \omega'')$, and the vector $(\mathbf{r}_a \times \mathbf{p}_a)$.

Certainly, Eq. (9) is under the sign $\int_{-\infty}^{+\infty} dt \int_0^{\infty} d\tau_1 \int_0^{\infty} d\tau_2$, what, as usually, leads to singular functions giving rise to restrictions $\omega'' = -\omega'$, $\omega + \omega_{n_1 n_2} = 0$, and $\omega'' = \omega_{n_4 n_1}$ on summation in Eq. (9) and integration $\int_{-\infty}^{+\infty} d\omega'\, d\omega''$ arising after transition to Fourier transform. Now $\overline{\mathbf{E}(\omega'')}$ should be written instead of $\mathbf{E}(\omega')$, where the bar denotes complex conjugation. The appearance of combination

$$\sum_{n_1 n_2 n_4} (CD)_{n_2 n_4}\, G_{n_4 n_1}\, \rho_{n_1}\, B_{n_1 n_2} \equiv \equiv \sum_{n_2 n_4} (CD)_{n_2 n_4}\, \Pi_{n_4 n_2} = S_p\, \hat{\Pi}\, (\hat{C}\hat{D}) \quad (10)$$

is also clear.

It is a matter of course that, $\hat{\Pi}$ from Eq. (10) possesses, according to its definition and "rotational" origination, all the attributes of a second-rank tensor, and for isotropic media it is naturally diagonal. Therefore, $\hat{C}\hat{D}$ transforms into the expected combination $(\mathbf{r}\ \mathbf{E})\ \mathbf{H}$.

Besides, we should take into account summation over particles in Eq. (6). So, the result of previous transformation is

$$\sum_{a,a'} (\mathbf{r}_a\ \mathbf{E}(\mathbf{r}_a))\ \mathbf{H}(\mathbf{r}_{a'})\ \Pi_{a'}, \quad (11)$$

where $\Pi_{a'}$ is the term of the matrix element of $\hat{\Pi}$ from Eq. (10) corresponding to the subscript a. We should assure that the preference in Eq. (11) should be given to the terms with $a' = a$.

Let us denote by x and y the coordinates of particles with the indices a and a'; $X(x, y)$ are the wave functions used in calculation of matrix elements; f and g are the parameters entering into Eq. (10). We need to compare the integrals

$$\int dx dy\ \bar{X}(x, y)\ f(x)\ g(x)\ X(x, y)$$

and

$$\int dx dy\ \bar{X}(x, y)\ f(x)\ g(y)\ X(x, y)$$

corresponding to the variants $a = a'$ and $a \neq a'$.

Since we deal with finite rotation, which is a sort of motion, we can refer to the oscillation theorem.[12] In the first case $\int dy\, \bar{X}(x, y)\, X(x, y)$ refers to one particle and has, say, N oscillations with N being large and rotation is rather intense. However, in the second case the number of oscillations increases significantly, what certainly leads to significant decrease of the integral.

Further, it should be explained why does Π_a in Eq. (11), where now $a' = a$, have the argument $\mathbf{r} - \mathbf{r}_a$. The corresponding arguments although look as only qualitative, but are rather standard. First, $\mathbf{r} - \mathbf{r}_a$ enters into the definition (8). Second, at only rotation of molecules (which, recall, is firmly associated with the high angular momentum of the field) translational invariance should necessarily take place. Finally, the same rotation of molecules allows us to use classic language for its description, in which Sp in Eq. (10) transforms into $\int d\mathbf{r}_a$ ($\int d\mathbf{r}'$ after the appropriate replacement of variables).

Let us return to Eq. (10) and pay attention to the fact that $\hat{\Pi}$ depends on ω and ω'' (or ω'), and they enter it not independently. As a consequence of the well-known reasoning on the stationary state of statistical processes [Sp in Eq. (10) (!)] is the statement that the corresponding argument is finally $(\omega - \omega')$.

The result of the above procedure – derivation of the parameter (3) – can now be represented by the equations

$$\mathbf{M}(\mathbf{r}, t) = \int_{-\infty}^{+\infty} \mathbf{M}(\mathbf{r}, \omega)\, e^{-i\omega t}\, d\omega,$$

$$\mathbf{M}(\mathbf{r}, \omega) = \quad (12)$$

$$= \int_{-\infty}^{+\infty} d\omega' \int_{-\infty}^{+\infty} d\mathbf{r}'\, F(\omega - \omega', \mathbf{r} - \mathbf{r}')\, (\mathbf{r}'\ \mathbf{E}(\mathbf{r}', \omega'))\, \mathbf{H}(\mathbf{r}', \omega'),$$

which are just those that correspond to the structure of Eq. (4). Of course, it is now simple to represent F in

terms of numerous matrix elements and other quantum characteristics, but such equations are useless for computation. Usually, parameters similar to F are declared empirical, when the form of equations they enter in is already found.

So, along with the Maxwell equations (in which **P** is linearly related to **E**) Eqs. (1) and (12) form nonlinear optics for the field possessing high angular momentum. It is characterized by not necessarily high intensity, the presence of the magnetic moment of a unit volume, and of spatial dispersion. The latter owes its origin to peculiarities of interaction between a medium and a field with high momentum.

References

1. S.D. Tvorogov, Izv. Vyssh. Uchebn. Zaved., Ser. Fiz., No. 10, 93–103 (1996).
2. R. Glauber, in: *Quantum Optics and Quantum Radiophysics* [Russian translation] (Mir, Moscow, 1966), 452 pp.
3. G. Clauder and E. Sudarshan, *Principles of Quantum Optics* [Russian translation] (Mir, Moscow, 1970), 428 pp.
4. A.V. El'shin and S.D. Tvorogov, Dokl. Akad. Nauk SSSR **314**, No. 3, 524–527 (1989).
5. A.V. El'shin and S.D. Tvorogov, Atm. Opt. **2**, No. 5, 373–377 (1989).
6. V.P. Lopasov, Izv. Vyssh. Uchebn. Zaved., Ser. Fiz., No. 8, 119–123 (1999).
7. D.I. Blokhintsev, *Principles of Quantum Mechanics* (Nauka, Moscow, 1976), 664 pp.
8. F. Arecci, M. Scally, G. Haken, and V. Vaidlih, *Quantum Fluctuation of Laser Radiation* [Russian translation] (Mir, Moscow, 1974), 236 pp.
9. M. Goeppert-Mayer, Ann. Phys. **9**, 273 (1931), Ref. as in N. Blombergen, *Nonlinear Optics* [Russian translation] (Mir, Moscow, 1966), 424 pp.
10. I.R. Shen, *Principles of Nonlinear Optics* [Russian translation] (Mir, Moscow, 1980), 558 pp.
11. D.N. Zubarev, *Nonequilibrium Statistical Thermodynamics* (Nauka, Moscow, 1971), 415 pp.
12. L.D. Landau and E.M. Lifshitz, *Quantum Mechanics* (Pergamon Press, Oxford, 1974).