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Nonlinear Optical Wave Equation for Micro- and Nano-Structured Media and Its Application

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Summary

Major trends recently occurred in the modern physics and applications of nonlinear optics are closely linked with the recent rapid technological advances in material science. These are engineering of material structures of nanometre scale length and novel concepts for tailoring their nonlinear response including substantial enhancement and tuning their nonlinearities. R&D in this subject area called nonlinear nanophotonics. Areas of application for this are enormous. Substantial progress is expected to happen and is also happening in more traditional areas of R&D in nonlinear optics, such as highly efficient nonlinear frequency conversion (harmonics generation, up- and downconversion, supercontinuum generation, etc.) in smaller size devices such as photonics crystal structures, carbon nanotubes and graphene. Evidently all such applications require an adequate theoretical support. The essential feature of the described above structures is their spatial scale length which is smaller than radiation wavelength. This property is however not described by the Bloembergen's iconic nonlinear optical wave equation (NOWE) which is only valid for macroscopically homogeneous media.

In this work a theoretical formulism is developed for optical wave linear and nonlinear propagation and interaction in materials with intrinsic or induced macroscopic spatial inhomogeneity. This formulism has emerged from continuation and further development of the theory of light scattering by Einstein and by Landau and Lifshitz. The obtained optical wave equation (OWE) is applicable to variety linear and nonlinear optical interactions in spatially inhomogeneous media. While it is valid regardless of the spatial scale of these inhomogeneities, it explicitly shows that the strongest contribution is expected from inhomogeneities of a sub-wavelength scale size, typical for nanostructured optical media. Therefore the obtained equation is laying a more rigorous theoretical basis for the rapidly growing activity worldwide in studies of nanophotonic systems.

1. Introduction.

Phenomena of light propagation and interaction in an optically transparent nonmagnetic continuous medium is the subject area of the macroscopic electrodynamics [1,2]. For the majority of these phenomena the medium can be reasonably well approximated as the spatially homogeneous one. Respectively their theoretical description is based on OWE obtained in the approximation of macroscopically homogeneous medium [2,3]. On the other hand there are plenty of phenomena in optics which are inevitably associated with medium's inhomogeneities. Historically first one, Rayleigh scattering, was investigated more than a century ago by Tyndall (1869) and Lord Rayleigh (1899), who attributed the phenomenon to sub-wavelength size particles (macroscopic inhomogeneities of the refractive index) in a visually transparent medium. Later, through the XX century, a variety of linear and nonlinear optical phenomena were discovered and studied, in which macroscopic spatial inhomogeneity of the medium is the integral part of physics behind the phenomenon. Among these are a range of spontaneous and stimulated scattering phenomena, static and dynamic holography (including Bragg gratings), four wave mixing, etc. Most recently, mainly in the XXI century, the attention turned to optical phenomena in micro- and nanostructured media (photonic crystals, randomly and regularly distributed in space nanoscale size objects (particles, rods, rings, etc.)). Our analysis of publications on studies of these phenomena have revealed that their theoretical description is using the OWE for macroscopically homogeneous media, [4], or based on various heuristic approaches. In this work an attempt is made to rigorously consider in OWE the effect of the medium's spatial inhomogeneity.

2. Historical background

2a). Einstein's approach

Probably the first attempt to obtain OWE for macroscopically inhomogeneous media was made by Einstein [5]. Einstein's procedure of obtaining the equation is as follows. If the incident light is a quasi-monochromatic electro-magnetic (EM) wave with the central frequency ω , an optically transparent nonmagnetic ($\mu = 1$) medium can then be characterised by the correspondent to that frequency permittivity ε . The permittivity of an optically isotropic inhomogeneous medium is supposed to be the sum of a background homogeneous part, ε_0 , and of a spatially modulated part $\delta\varepsilon(r)$,

$$\varepsilon(r) = \varepsilon_0 + \delta\varepsilon(r) . \quad (1)$$

Macroscopic electric, \vec{E} , and magnetic, \vec{H} , fields of optical radiation at each point of the medium are governed by the macroscopic Maxwell equations,

$$\nabla \times \vec{H} = \frac{1}{c} \frac{\partial \vec{D}}{\partial t} , \quad (2)$$

$$\nabla \times \vec{E} = -\frac{1}{c} \frac{\partial \vec{H}}{\partial t} , \quad (3)$$

$$\nabla \cdot \vec{D} = 0 , \quad (4)$$

$$\nabla \cdot \vec{H} = 0 . \quad (5)$$

where c is the velocity of light and \vec{D} is the electric induction which is coupled with the electric field, \vec{E} , through the relation

$$\vec{D} = \varepsilon \vec{E} . \quad (6)$$

Using the standard procedure of exclusion of the magnetic field, \vec{H} , one can convert Eqs (2) and (3) to the wave equation for the electric field,

$$\nabla \times \nabla \times \vec{E} + \frac{1}{c^2} \frac{\partial^2 \vec{D}}{\partial t^2} = 0, \quad (7)$$

which, along with Eqs(4) and (6), describe propagation of an EM wave in a dielectric medium.

In an optically inhomogeneous medium the electric field of the optical wave, \vec{E}_p , which is propagating in the direction of the incident to that medium optical wave, differs from the electric field of the incident wave, \vec{E} , by a field of the scattered wave, \vec{E}_s , so, the total electric field in the medium is

$$\vec{E} = \vec{E}_p + \vec{E}_s. \quad (8)$$

When $\delta\varepsilon(r)$ and \vec{E}_s are infinitesimal compared to ε_0 and \vec{E}_p , Eq.(7) can be split to two coupled equations for \vec{E}_p and for \vec{E}_s , the latter of which is

$$\nabla \times \nabla \times \vec{E}_s + \frac{1}{c^2} \frac{\partial^2 \vec{D}_s}{\partial t^2} = 0, \quad (9)$$

with

$$\vec{D}_s = \varepsilon_0 \vec{E}_s + \delta\varepsilon \vec{E}_p, \quad (10)$$

and the equation for \vec{E}_p is

$$\nabla \times \nabla \times \vec{E}_p + \frac{1}{c^2} \frac{\partial^2 \vec{D}_p}{\partial t^2} = 0, \quad (11)$$

with

$$\vec{D}_p = \varepsilon_0 \vec{E}_p. \quad (12)$$

Small terms of the second order were neglected in obtaining Eqs (9)-(12). This in particular means that propagation of the pump field, \vec{E}_p , is governed by the optical wave equation, Eq.(11), with a constant background permittivity, ε_0 .

Substituting Eq.(10) into Eqs (4) and (9) and taking into account that $\nabla \times \nabla \times \vec{F} = \nabla(\nabla \cdot \vec{F}) - \nabla^2 \vec{F}$ converts the equations for the scattered field to the form,

$$\nabla^2 \vec{E}_s - \frac{\varepsilon_0}{c^2} \frac{\partial^2 \vec{E}_s}{\partial t^2} = \nabla(\nabla \cdot \vec{E}_s) + \frac{\delta\varepsilon}{c^2} \frac{\partial^2 \vec{E}_p}{\partial t^2}, \quad (13)$$

$$\nabla \cdot \vec{D}_s = \nabla \cdot (\varepsilon_0 \vec{E}_s + \delta\varepsilon \vec{E}_p) = 0. \quad (14)$$

Using Eq.(14) and taking into account that $\nabla(\Phi \vec{F}) = \Phi \nabla \vec{F} + \vec{F} \nabla \Phi$, [6], $\nabla \cdot \vec{E}_p = 0$ and $\nabla \cdot \varepsilon_0 = 0$ Einstein obtained the expression for $\nabla \cdot \vec{E}_s$,

$$\nabla \cdot \vec{E}_s = -\frac{1}{\varepsilon_0} \vec{E}_p \cdot \nabla \delta\varepsilon, \quad (15)$$

substitution of which into Eq.(13) has given his working OWE for the scattered field,

$$\nabla^2 \vec{E}_s - \frac{\epsilon_0}{c^2} \frac{\partial^2 \vec{E}_s}{\partial t^2} = -\frac{1}{\epsilon_0} \nabla(\vec{E}_p \nabla \delta\epsilon) + \frac{\delta\epsilon}{c^2} \frac{\partial^2 \vec{E}_p}{\partial t^2}, \quad (16)$$

in which the first term on the RHS is proportional to $\nabla \delta\epsilon$, that is it explicitly accounts the effect of spatial variation of $\delta\epsilon$, while the second term is proportional to the magnitude of $\delta\epsilon$ showing that it is independent of the rate of its spatial variation.

Analysing this equation Einstein has however noted that the first term on the RHS of Eq.(16) is actually generating a longitudinal electric field, which cannot contribute to the scattered field, propagation of which is described by the LHS of this equation. Moreover he has found that its contribution is compensated by the longitudinal component from the second term, [5]. Consequently the wave equation for the scattered field acquires the form,

$$\nabla^2 \vec{E}_s - \frac{\epsilon_0}{c^2} \frac{\partial^2 \vec{E}_s}{\partial t^2} = \left(\frac{\delta\epsilon}{c^2} \frac{\partial^2 \vec{E}_p}{\partial t^2} \right)_{\perp}, \quad (17)$$

where the symbol \perp on the RHS bears witness to the fact that only transverse components of the expression in the brackets are contributing to the field \vec{E}_s on the LHS.

Evidently the RHS of Eq.(17) does not have terms accounting the effect of any kind of spatial variation of $\delta\epsilon$. As such in its very essence it describes the scattered signal as a secondary emission of the deviations $\delta\epsilon(r)$ regardless of the rate of spatial variation of these deviations, that is the scattering in spatially homogeneous media too. Obviously this is in direct contradiction with the basic physics of the Rayleigh scattering phenomenon introduced/considered by Tyndall (1869) and Lord Rayleigh (1899) which explicitly demonstrate dependence of scattering characteristics on the size of inhomogeneities.

The form Eq.(17) is fully consistent with the iconic NOWE, which was obtained by Bloembergen with colleagues for the case of optically homogeneous media [3]. The only difference is that in NOWE $\delta\epsilon$ is dependent of the optical field(s).

Obviously an alternative approach is required to obtain OWE for describing the effect(s) in optically inhomogeneous media. Since we are interested in accounting for the spatial effect, it can be done through the term in the wave equation for the scattered field, Eq.(9), with spatial derivatives only.

2b). Landau's approach

The way to do this is actually described in [1]. Instead of replacing \vec{D}_s in Eq.(9), as it was done by Einstein, Landau and Lifshitz (L&L) replaced \vec{E}_s under $(\nabla \times \nabla \times)$ by

$$\vec{E}_s = \frac{\vec{D}_s - \delta\epsilon \vec{E}_p}{\epsilon_0}, \quad (18)$$

which follows from Eq.(10). Then in the approximation of monochromatic incident and scattered waves they got the equation for the induction of the scattered radiation, \vec{D}_s ,

$$\nabla^2 \vec{D}_s + \frac{\epsilon_0}{c^2} \omega_s^2 \vec{D}_s = -\nabla \times \nabla \times [\delta\epsilon \vec{E}_p]. \quad (19)$$

Equation (19) was solved and analysed in [1] to elucidate various properties of the intensity, $I_s = |E_s|^2$, of scattered radiation in various media.

Interestingly but in spite of essential difference between the RHSs of Eqs (17) and (19), the calculated by both Einstein in [5] and L&L in [1] the integral extinction coefficient due to Rayleigh scattering coincide. Probably because of this coincidence the difference between the two approaches was not considered so far.

3. OWE for spatially inhomogeneous media

To understand, clarify and appreciate the actual value of L&L approach and its difference from Einstein's approach we firstly must consider the cases in similar approximations. We consider the interaction of quasi-monochromatic quasi-plane incident and scattered waves as a more general approximation compared to the case of interaction of monochromatic plane waves considered in [1].

In this approximation by substitution of \vec{E}_s into the first term on the LHS of Eq.(9) we shall get the equation for the induction of the scattered radiation, \vec{D}_s , in the form

$$\nabla \times \nabla \times \vec{D}_s + \frac{\varepsilon_0}{c^2} \frac{\partial^2 \vec{D}_s}{\partial t^2} = \nabla \times \nabla \times [\delta\varepsilon \vec{E}_p]. \quad (20)$$

Since the electric field is scattered radiation is must be transverse and it is conventionally detected at large enough distance from the scattering region, \vec{D}_s is related to \vec{E}_s there as $\vec{D}_s = \varepsilon_0 \vec{E}_s$, and Eq.(20) transforms to

$$\nabla^2 \vec{E}_s - \frac{\varepsilon_0}{c^2} \frac{\partial^2 \vec{E}_s}{\partial t^2} = -\frac{1}{\varepsilon_0} \nabla \times \nabla \times [\delta\varepsilon \vec{E}_p]. \quad (21)$$

Here again on the RHS we should consider the contributions from its transverse components only.

3a). OWE for scattering from a scalar type spatial inhomogeneities

To compare Eqs (17) and (21) consider scattering of a quasi-monochromatic plane wave, $\vec{E}_p(r, t) = \vec{E}_p(z, t) e^{-i(\omega_p t - k_p z)}$, (here $\vec{E}_p(z, t)$ is the slowly varying in space and time amplitude of that wave) as an incident field in a medium where $\delta\varepsilon(r)$ is an independent of time scalar function of spatial coordinates. In such case, by using the properties of the *del* operator, $\nabla \cdot (\Phi \vec{F}) = \Phi (\nabla \cdot \vec{F}) + \nabla \Phi \cdot \vec{F}$, $\nabla \times (\vec{F} \times \vec{G}) = (\vec{G} \cdot \nabla) \vec{F} - (\vec{F} \cdot \nabla) \vec{G} + \vec{F} (\nabla \cdot \vec{G}) - \vec{G} (\nabla \cdot \vec{F})$ and $\nabla \times (\nabla \Phi) = 0$ (see Section 5.5 in [6]), we shall get

$$\nabla \times \nabla \times [\delta\varepsilon \vec{E}_p] = \delta\varepsilon (\nabla \times \nabla \times \vec{E}_p) + \nabla \delta\varepsilon \times (\nabla \times \vec{E}_p) - \vec{E}_p \nabla^2 \delta\varepsilon + (\vec{E}_p \cdot \nabla) \nabla \delta\varepsilon - (\nabla \delta\varepsilon \cdot \nabla) \vec{E}_p. \quad (22)$$

By taking into account that \vec{E}_p is a transverse electric field of the incident radiation, for which according to Eq.(11) we have

$$\nabla \times \nabla \times \vec{E}_p = -\nabla^2 \vec{E}_p = -\frac{\varepsilon_0}{c^2} \frac{\partial^2 \vec{E}_p}{\partial t^2}, \quad (23)$$

Eq.(22) can be rewritten as

$$\begin{aligned} \nabla \times \nabla \times [\delta\varepsilon \vec{E}_p] &= -\delta\varepsilon \nabla^2 \vec{E}_p + \nabla \delta\varepsilon \times (\nabla \times \vec{E}_p) - \vec{E}_p \nabla^2 \delta\varepsilon + (\vec{E}_p \cdot \nabla) \nabla \delta\varepsilon - (\nabla \delta\varepsilon \cdot \nabla) \vec{E}_p = \\ &= -\frac{\varepsilon_0}{c^2} \delta\varepsilon \frac{\partial^2 \vec{E}_p}{\partial t^2} + \nabla \delta\varepsilon \times (\nabla \times \vec{E}_p) - \vec{E}_p \nabla^2 \delta\varepsilon + (\vec{E}_p \cdot \nabla) \nabla \delta\varepsilon - (\nabla \delta\varepsilon \cdot \nabla) \vec{E}_p. \end{aligned} \quad (24)$$

Substituting Eq.(24) into Eq.(21) we obtain the equation for the scattered field,

$$\nabla^2 \vec{E}_s - \frac{\varepsilon_0}{c^2} \frac{\partial^2 \vec{E}_s}{\partial t^2} = \frac{\delta\varepsilon}{c^2} \frac{\partial^2 \vec{E}_p}{\partial t^2} + \frac{1}{\varepsilon_0} [\vec{E}_p \nabla^2 \delta\varepsilon - \nabla \delta\varepsilon \times (\nabla \times \vec{E}_p) - (\vec{E}_p \cdot \nabla) \nabla \delta\varepsilon + (\nabla \delta\varepsilon \cdot \nabla) \vec{E}_p]. \quad (25)$$

It's easy to see that the first term on the RHS of Eq.(25) coincides with the RHS of Einstein's equation, Eq.(17). The remaining chain of four terms on the RHS of Eq.(25) is new to the theory of wave propagation in optical media, and these are representing a specific contribution of a medium's macroscopic spatial inhomogeneity.

3b). OWE for backscattering from a periodic spatial inhomogeneity

Consider then the case when the incident radiation is a plane wave of the frequency ω_p with the wavevector k_p , which is propagating along $+z$,

$$\vec{E}_p(r,t) = \vec{E}_p e^{-i(\omega_p t - k_p z)}, \quad (26)$$

and $\delta\epsilon(r)$ is a periodic grating with the wavevector q also along $+z$,

$$\delta\epsilon(z) = \beta(z) e^{iqz}. \quad (27)$$

In such case the scattered radiation is also a plane wave of the frequency $\omega_s = \omega_p$ with the wavevector $k_s = k_p + q$, the last two terms on the RHS of Eq.(26) are zeros and the equation transforms to

$$\frac{\partial^2}{\partial z^2} \vec{E}_s + k_s^2 \vec{E}_s = -\frac{\delta\epsilon}{\epsilon_0} \vec{E}_p [k_p^2 + q^2 + qk_p]. \quad (28)$$

This to be compared with the result of substitution Eqs (26) and (27) into Eq.(17). Obviously the first term on the RHS of this equation is zero, and therefore equation for the scattered field reduces to

$$\frac{\partial^2}{\partial z^2} \vec{E}_s + k_s^2 \vec{E}_s = -\frac{\delta\epsilon}{\epsilon_0} \vec{E}_p k_p^2. \quad (29)$$

The difference is clear: the RHS of Eq.(28) has two terms which explicitly depend on the rate of spatial variation of $\delta\epsilon$, that is on q . It reduces to Einstein's equation for the scattered field when the medium is macroscopically homogeneous that is when the characteristic size of inhomogeneities $a \propto q^{-1} \rightarrow \infty$ ($q \rightarrow 0$) or when $\delta\epsilon(r)$ is a slowly enough varying on r function, $a \gg \lambda_p$ ($\lambda_p = 2\pi m/k_p$ is the wavelength of the incident radiation and n is the refractive index). When $q \leq$ or $\geq k_p$ the contribution of the last two terms on the RHS of Eq.(28) can be not small. In particular when $q = 2k_p$ (this case is a typical for the Rayleigh and Brillouin backscattering and for the Bragg grating backreflection) the contribution from the last two terms is 6 times bigger than form the first one.

Clearly the difference is much more pronounced when $a \ll \lambda_p$. That case is typical for nanostructured media, where a are conventionally in the range from ~ 100 nm to ~ 1000 nm. This circumstance in particular would allow account for a substantial inconsistency between experimental observations of some phenomena in nanostructured materials and their theoretical description in frames of the existing theory [7]. In such media the second terms on the RHS of Eq.(28) and of Eq.(25) are dominating over all other. An important result of this is essential simplification of the equations,

$$\frac{\partial^2}{\partial z^2} \vec{E}_s + k_s^2 \vec{E}_s = -\frac{\delta\epsilon}{\epsilon_0} \vec{E}_p q^2, \quad (30)$$

and

$$\nabla^2 \vec{E}_s - \frac{\epsilon_0}{c^2} \frac{\partial^2 \vec{E}_s}{\partial t^2} = \frac{1}{\epsilon_0} \vec{E}_p \nabla^2 \delta\epsilon \quad (31)$$

which is obviously useful for practice.

3c). Generalisation of the new OWE

Obviously $\delta\varepsilon$ in a medium can vary in both space and time, $\delta\varepsilon(r,t)$. Then scattering of an incident EM wave in an inhomogeneous medium is the appearance in it of EM waves, not only directions but also frequencies of which not coincide with these of the original incident wave [1]. Then numerous, mostly nonlinear, optical phenomena, which conventionally were not considered being scattering, can be understood and treated as scattering. Harmonics generation, self- and cross-phase modulation, various parametric processes, four-wave mixing, etc. are among such phenomena.

In the most general case $\delta\varepsilon$ in a medium is a tensor, $\delta\hat{\varepsilon}(r,t)$. In this case $[\delta\varepsilon\vec{E}_p]$ in Eqs (21) and (25) has to be a vector whose components are $[\delta\varepsilon_{ik}(r,t)E_k]$, where $\delta\varepsilon_{ik}(r,t)$ and E_k are the components of the medium's permittivity tensor and of the pump field [1]. A variation of permittivity can be imprinted (linear scattering) or induced by the incident optical radiation (nonlinear scattering) in the medium, i.e.

$$\delta\hat{\varepsilon}(r,t) = \delta\hat{\varepsilon}_L(r,t) + \delta\hat{\varepsilon}_{NL}(r,t), \quad (32)$$

where $\delta\hat{\varepsilon}_L(r,t)$ and $\delta\hat{\varepsilon}_{NL}(r,t)$ are the linear and nonlinear parts of the medium's permittivity. When $\delta\varepsilon_{NL}(r,t)$ is negligible Eq.(25) is describing linear light scattering and Bragg reflection without and/or with (when $\delta\varepsilon_L(r,t)$ is dependant of time) change of the frequency spectrum (broadening and/or shift). When $\delta\varepsilon_{NL}(r,t)$ is not small Eq.(25) describes a range of nonlinear optical phenomena when the medium is induced to be macroscopically inhomogeneous (for example stimulated scattering phenomena). Thus Eq.(25) with Eq.(32) is the generalization of the iconic NOWE obtained in [3], for light-matter interactions in macroscopically inhomogeneous media.

4. Conclusions

A novel optical wave equation is developed for optical wave propagation and interaction in materials with intrinsic or induced *macroscopic spatial inhomogeneity*. It shows that the effect of spatial inhomogeneity is most pronounced for sizes of inhomogeneities on a sub-wavelength scale, that is in materials commonly referred to as nanophotonic materials. Respectively the new equation provides a rigorous theoretical template for emerging experimental activity worldwide in the area of linear and nonlinear nanophotonics.

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List of Symbols, Abbreviations, and Acronyms

- r and t - the space(position) and time variables
 ω - the frequency of optical radiation
 ε - the medium's permittivity
 μ - the medium's permeability
 ε_0 - the background homogeneous part of the medium's permittivity
 $\delta\varepsilon(r)$ - spatially modulated part of the medium's permittivity
 \vec{E} - the macroscopic electric field of an optical radiation wave
 \vec{H} - the macroscopic magnetic field of an optical radiation wave
 \vec{D} - the electric induction
 ∇ - the del operator
 \vec{E}_p - the field of the optical wave propagating in the direction of the incident radiation
 \vec{E}_S - the field of the scattered wave
 \vec{D}_p - the electric induction of the wave propagating in the direction of the incident radiation
 \vec{D}_S - the electric induction of the scattered wave
 \vec{F} - a vector function of the position
 Φ - a scalar function of the position
 \perp - the symbol denoting the transverse components
 I_S - the intensity of scattered radiation
 $\vec{E}_p(z,t)$ - the slowly varying in space and time amplitude of \vec{E}_p
 ω_p - the frequency of the field \vec{E}_p
 k_p - the wavevector of the field \vec{E}_p
 $+z$ - the direction of the incident radiation propagation
 $\beta(z)$ - the slowly varying along z amplitude of $\delta\varepsilon(z)$
 q - the wavevector of the grating of $\delta\varepsilon(z)$
 ω_S - the frequency of the scattered radiation
 k_S - the wavevector of the scattered radiation
 a - the characteristic size of inhomogeneities
 λ_p - the wavelength of the incident radiation
 n - the refractive index
 $\delta\hat{\varepsilon}(r,t)$ - the tensor of medium's permittivity.
 $\delta\hat{\varepsilon}_L(r,t)$ and $\delta\hat{\varepsilon}_{NL}(r,t)$ - the linear and nonlinear parts of the medium's permittivity tensor
 $\delta\varepsilon_{ik}(r,t)$ and E_k - the components of the medium's permittivity tensor and of the incident field
OWE - optical wave equation
R&D - research and development
NOWE - nonlinear optical wave equation
EM wave - electro-magnetic wave
RHS - right hand side
LHS - left hand side
L&L - Landau and Lifshitz