

Acousto-optical properties of metamaterials

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Abstract. The possibility of the effective use of metamaterials in acousto-optics is demonstrated. It is shown that photoelastic constants that determine a change in the dielectric constant of a heterogeneous medium under the action of a sound wave can significantly exceed the corresponding constants for conventional crystals. We have analysed the mechanisms of the dielectric constant variation in a heterogeneous medium consisting of nanoparticles in the form of ellipsoids and have found explicitly the values of the photoelastic constants. It is shown that the mechanism of the dielectric constant variation in a longitudinal sound wave is reduced to a change in the local concentration of nanoparticles in the bulk and in a transverse acoustic wave – to a local rotation of space-oriented nanoellipsoids. It is also shown that the use of metamaterials with a nonuniform distribution of nanoparticles provides a unique opportunity for designing qualitatively new instruments and devices that cannot be produced on the basis of conventional crystals. It is noted that metamaterials open ample opportunities for creating devices of the IR region of the spectrum due to the absence of restrictions on the size of such media.

Keywords: acousto-optics, metamaterials, ellipsoidal nanoparticles, photoelastic constant.

A prerequisite for the use of metamaterials for acousto-optics consists in the fact that under the influence of a sound wave the dielectric constant of a medium must change. After the inclusion of small metal (or any other) nanoparticles, an initially homogeneous medium having a low acousto-optical quality becomes a material with a high photoelastic constant.

The mechanism of this change in the dielectric constant under the influence of a sound wave is as follows. If a longitudinal sound wave, which has regions of medium compression and expansion, propagates in a medium, the dielectric constant and the refractive index of the medium will obviously change due to a local variation of the concentration of nanoparticles. This is a fairly common case, which is related to any inclusions as symmetric (e.g., nanospheres) and so asymmetric (e.g., ellipsoids). If a transverse sound wave accompanied by a shear deformation and not changing the volume density of nanoparticles propagates in a medium, the refractive index for randomly arranged nanoparticles will not change. However, for spatially asymmetric nanoparticles, for example ellipsoids, whose symmetry axes are spatially ori-

ented, the shear deformation will result in some rotation of the symmetry axes of ellipsoids, which eventually will cause changes in the dielectric constant and refractive index of the medium.

In this paper, as a heterogeneous medium, we consider a medium formed by an optically transparent material with a dielectric constant ε_m and embedded metal or dielectric inclusions in the form of ellipsoids of revolution with a complex dielectric constant $\varepsilon_p(\lambda) = \varepsilon_p'(\lambda) + i\varepsilon_p''(\lambda)$ (λ is the wavelength of light). It is assumed that the size of the ellipsoids of revolution is substantially smaller than the wavelength of light and they are randomly distributed, but their main axes of rotation are oriented in the same direction. Within the framework of the Maxwell Garnett model, such a medium is described by the averaged dielectric constant ε of the medium, which satisfies the relation [1–3]

$$\frac{\varepsilon - \varepsilon_m}{L(\varepsilon - \varepsilon_m) + \varepsilon_m} - \eta \frac{\varepsilon_p - \varepsilon_m}{L(\varepsilon_p - \varepsilon_m) + \varepsilon_m} = 0, \quad (1)$$

where η is the volume fraction of inclusions, i.e. metal nanoellipsoids of revolution [$\eta(r, t) = \frac{4}{3}\pi ab^2 N(r, t)$]; a is the polar axis of the spheroid; b is the small equatorial semi-axis of the ellipsoid of revolution (spheroid); $N(r, t)$ is the local number of spheroids per unit volume of the medium; and L is the depolarisation factor, which can be expressed as the ratio of semi-axes $\xi = a/b$ for different directions (parallel and perpendicular to the axis of rotation of the spheroid) of the external field:

$$L_{\parallel} = \frac{1}{1 - \xi^2} \left(1 - \xi \frac{\arcsin \sqrt{1 - \xi^2}}{\sqrt{1 - \xi^2}} \right), \quad L_{\perp} = \frac{1}{2}(1 - L_{\parallel}). \quad (2)$$

The case $\xi < 1$ corresponds to a prolate ellipsoid of revolution, $\xi > 1$ – to a flattened ellipsoid, and $\xi = 1$ – to a sphere (in this case, $L_{\parallel} = L_{\perp} = 1/3$). Solving equation (1) with respect to ε , we find the explicit form of the dielectric constant of the metamaterial:

$$\varepsilon_{\parallel, \perp} = \frac{\varepsilon_m [(-1 + L_{\parallel, \perp})(-1 + \eta)\varepsilon_m + (L_{\parallel, \perp} + \eta - L_{\parallel, \perp}\eta)\varepsilon_p(\lambda)]}{[1 + L_{\parallel, \perp}(-1 + \eta)]\varepsilon_m + L_{\parallel, \perp}(1 - \eta)\varepsilon_p(\lambda)} \quad (3)$$

for the electric field along and across the polar axis of the ellipsoid of rotation, respectively. It follows from expressions (3) that the dielectric constant of the medium is a diagonal tensor with the components $\varepsilon_{11} = \varepsilon_{22} = \varepsilon_{\perp}$ and $\varepsilon_{33} = \varepsilon_{\parallel}$, other components being equal to zero. The condition of the applicability of the Maxwell Garnett model suggests that the filling factor lies in the range $1/3 < \eta < 2/3$ [4].

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Expressions (3) make it possible to find the change in the dielectric constant of the medium under the action of a sound wave. We consider the cases of longitudinal and transverse sound waves separately, because the mechanisms of the dielectric constant variation differ from one another.

1. Let the longitudinal sound wave propagate along any axis of symmetry, such as the x axis: $\mathbf{u}(x, t) = \frac{1}{2}\mathbf{u}_0 \exp(i\Omega t - i\mathbf{q}\mathbf{x}) + \text{c.c.}$, where $\mathbf{u}(x, t)$ is the amplitude of the medium displacement in the acoustic wave; Ω is the frequency; and \mathbf{q} is the wave vector of the sound wave. In the longitudinal sound wave the directions of the displacement vector $\mathbf{u}(x, t)$ and the wave vector \mathbf{q} coincide. The acoustic properties of the medium are considered to be isotropic. The latter means that the moduli of elasticity of asymmetric inclusions in question, unlike the moduli of a matrix medium, are such that under the action of the elastic wave they are not deformed. In the longitudinal sound wave, the medium is compressed (stretched), and therefore the local concentration of nanoparticles (spheres) changes according to the law

$$\eta(x, t) = \eta_0 + \eta_{\perp}(x, t) = \frac{4\pi}{3}ab^2(N_0 + N_{\perp}(x, t)). \quad (4)$$

Here, $N_{\perp}(x, t)$ is the change in the concentration under the action of a sound wave; and N_0, η_0 are the volume-averaged values. From the equation of continuity, for a matrix medium we can obtain

$$\eta_{\perp}(x, t) = \frac{i2\pi}{3}a^2bN_0\frac{u_0\Omega}{v_s}\exp(i\Omega t - i\mathbf{q}\mathbf{x}) + \text{c.c.}, \quad (5)$$

where $v_s = \Omega/q$ is the phase velocity of the sound wave in a metamedium. Possible dispersion properties of the elastic medium, associated with different elastic properties of the matrix and nanoinclusions, are not considered here. We assume that because the size of the nanoparticles is much smaller than the wavelength of sound, they cannot lead to a significant dispersion. The corresponding additions to the components of the dielectric constant tensor of a metamedium, conditioned by the longitudinal sound wave, have the form

$$\Delta\varepsilon_{\parallel, \perp} = \frac{\varepsilon_m[\varepsilon_m - \varepsilon_p(\lambda)][\varepsilon_m(L_{\parallel, \perp} - 1) - L_{\parallel, \perp}\varepsilon_p(\lambda)]}{\{\varepsilon_m[1 + L_{\parallel, \perp}(\eta_0 - 1)] - L_{\parallel, \perp}\varepsilon_p(\lambda)(\eta_0 - 1)\}^2}\eta_{\parallel}(x, t). \quad (6)$$

In the chosen coordinate system, the z axis coincides with the direction of the main (polar) axis of the spheroid, and the x and y axes lie in a perpendicular plane. Using the generally accepted definition of photoelastic constants [5] as additions to the impermeability tensor of the medium, it is possible to find their explicit form for the longitudinal sound wave for a given direction of propagation of light and sound:

$$p_{36} = [\varepsilon_m - \varepsilon_p(\lambda)][(-1 + L_{\parallel})\varepsilon - L_{\parallel}\varepsilon_p(\lambda)] \\ \times \sqrt{\frac{\varepsilon_m[(-1 + L_{\parallel})\varepsilon_m(-1 + \eta_0) + \varepsilon_p(\lambda)(L_{\parallel} + \eta_0 - L_{\parallel}\eta_0)]}{\varepsilon_m - L_{\parallel}\varepsilon_m + L_{\parallel}\varepsilon_p + L_{\parallel}[\varepsilon_m - \varepsilon_p(\lambda)]\eta_0}} \\ \times \{2\varepsilon_m[(-1 + L_{\parallel})\varepsilon_m(-1 + \eta_0) + \varepsilon_p(\lambda)(L_{\parallel} + \eta_0 - L_{\parallel}\eta_0)]^2\}^{-1}. \quad (7)$$

Here, $p_{16} = p_{61}$ is the photoelastic constant for the longitudinal sound wave propagating along the minor optical axis of the ellipsoid of revolution when the light wave propagates in the same direction with the polarisation along the z axis. Similarly, from (7) we can obtain the photoelastic constant p_{16} by a formal replacement $L_{\parallel} \rightarrow L_{\perp}$. The metamedium under consideration is equivalent to a uniaxial crystal, and therefore there are only two independent components of the matrix of photoelastic constants. (Notations p_{16}, p_{26} correspond to standard six-dimensional representations.)

Equation (7) shows that the inclusion of nanoparticles with other dielectric constants into the matrix medium changes both optical and acousto-optical properties of the medium. It is seen that they will change most of all with the introduction of nanoparticles having a negative dielectric constant, such as metals. Since the resonance conditions for the dielectric constant and for photoelastic constants coincide, the results of the study of optical resonance properties of heterogeneous media can be fully utilised in relation to photoelasticity properties [1, 2, 6]. However, resonances for photoelastic constants turn to be sharper because of the square in the denominator.

2. Consider now the case of a transverse acoustic wave in which the amplitude of the displacement vector $\mathbf{u}(x, t) = \frac{1}{2}\mathbf{u}_0 \exp(i\Omega t - i\mathbf{q}\mathbf{x}) + \text{c.c.}$ is perpendicular to the wave propagation direction. The sound wave still propagates along the x axis so that the shear deformations are directed along the y axis. It is easy to see that these deformations lead to a slight rotation of the ellipsoids around the direction perpendicular to the displacement vector. This small change in the angle of rotation $\delta\alpha$ can be expressed in terms of the strain tensor component: $\delta\alpha \approx \partial u_y / \partial x$. To determine the photoelasticity constants it is necessary to perform a series of operations with the initial tensor of the dielectric constant of a composite medium. One should carry out the rotation by a certain angle α around the x axis (the choice of the x axis is dictated by the given direction of propagation of the transverse sound wave and its polarisation along the y axis). The rotation of the tensor is produced by a known transformation (see Ref. [7], Ch. 7). A preliminary rotation by some angle is needed to preserve the possibility of choosing the optimal direction of propagation of the light wave, for which the photoelastic interaction constants would be the greatest.

As a result of rotation around the x axis the dielectric constant tensor will change and there will appear off-diagonal components responsible for the interaction of the light waves propagating along the x axis, polarisation vectors of which are directed along the z and y axes. After a formal substitution $\alpha \rightarrow \alpha + \partial u_y(x, t) / \partial x$, we obtain additions to the dielectric constant tensor, proportional to the deformation. For the explicit definition of the components of the photoelasticity tensor it is necessary to find the inverse dielectric constant tensor of the medium and separate out the components that are proportional to the strain tensor, which, by definition, will be the components of the tensor of the photoelastic constants of the metamedium. As a result, for the transverse sound wave we obtain:

$$p_{46} = \frac{\varepsilon_{\parallel} - \varepsilon_{\perp}}{2\varepsilon_{\perp}\varepsilon_{\parallel}} \sin(2\alpha) - \frac{\varepsilon_{\parallel} - \varepsilon_{\perp}}{2\varepsilon_{\perp}\varepsilon_{\parallel}} \cos(2\alpha), \\ p_{26} = \frac{\varepsilon_{\perp} - \varepsilon_{\parallel}}{\varepsilon_{\perp}\varepsilon_{\parallel}} \sin(2\alpha), \quad (8)$$

$$p_{36} = \frac{\varepsilon_{\perp} - \varepsilon_{\parallel}}{\varepsilon_{\perp} \varepsilon_{\parallel}} \cos(2\alpha).$$

For the transverse acoustic wave polarised along the z axis, nonzero will be the following photoelastic constants: $p_{64} = p_{46}$, $p_{62} = p_{26}$ and $p_{63} = p_{36}$. The resulting expressions for the photoelastic constants depend on the concentration of nanoparticles (in this case, ellipsoids of revolution), on their geometry and dielectric properties of the material of which they are made. It is also seen that formula (8) makes it possible to find such an angle α for which the constant will have the greatest value. For example, for p_{46} $\alpha = [(2k + 1)/4 - 1/8]\pi$, $k = 0, \pm 1, \pm 2, \dots$. For metal nanoparticles, the dielectric constant ε_{\perp} is a complex quantity, and therefore the photoelasticity constant is also a complex quantity and, as the dielectric constant of a composite medium, allows the resonances to appear at certain wavelengths of light [1]. (It is advisable to investigate this practically important problem for a particular composite medium.)

In the theory of diffraction of light by sound waves the photoelasticity constant complexity does not introduce significant changes because the intensity of the diffracted light always includes the squared modulus of this constant [6, 7]. Note also that for conventional crystalline media the photoelasticity constants obtained in experiments are always considered to be purely real quantities. It follows from expression (8) that for symmetric nanoparticles, such as nanospheres, this constant is identically zero, and therefore the diffraction on the transverse acoustic waves is impossible.

3. Consider the case of a longitudinal sound wave in the case of the inclined position of the ellipsoids of revolution. We rotate tensor (3) around the y and z axes by the angle β and γ , respectively (note that the operation of rotation does not have the properties of commutativity and therefore sequence of their implementation is important). Then, we find the inverse value of the resulting dielectric constant tensor of the medium. These components of impermeability tensor have the form

$$\begin{aligned} \vartheta_{xx} &= \left(\frac{\cos^2 \beta}{\varepsilon_{\perp}} + \frac{\sin^2 \beta}{\varepsilon_{\parallel}} \right) \cos^2 \gamma + \frac{\sin^2 \gamma}{\varepsilon_{\parallel}}, \\ \vartheta_{xy} = \vartheta_{yx} &= \frac{\sin(2\gamma)}{2\varepsilon_{\perp}} - \frac{1}{2} \left(\frac{\cos^2 \beta}{\varepsilon_{\perp}} + \frac{\sin^2 \beta}{\varepsilon_{\parallel}} \right) \sin(2\gamma), \\ \vartheta_{xz} = \vartheta_{zx} &= \frac{\varepsilon_{\perp} - \varepsilon_{\parallel}}{\varepsilon_{\perp} \varepsilon_{\parallel}} \sin(2\beta) \cos \gamma, \\ \vartheta_{yy} &= \left(\frac{\cos^2 \beta}{\varepsilon_{\perp}} + \frac{\sin^2 \beta}{\varepsilon_{\parallel}} \right) \sin^2 \gamma + \frac{\cos^2 \gamma}{\varepsilon_{\perp}}, \\ \vartheta_{zz} &= \frac{\cos^2 \beta}{\varepsilon_{\parallel}} + \frac{\sin^2 \beta}{\varepsilon_{\perp}}, \end{aligned} \quad (9)$$

where x, y, z is the new, rotated coordinate system. Using expressions (9), it is easy to find the explicit form of the photoelasticity tensor components for this spatial orientation of the metamedium. We will present only the most important photoelasticity component, responsible for the coupling of the light waves propagating in the same direction, i.e., the component that provides collinear diffraction:

$$p_{14} = p_{41} = \frac{4\pi}{3} a^2 b N_0 \frac{\partial}{\partial \eta} \left(\frac{\varepsilon_{\perp} - \varepsilon_{\parallel}}{\varepsilon_{\perp} \varepsilon_{\parallel}} \sin(2\beta) \cos \gamma \right) \Big|_{\eta = \eta_0}. \quad (10)$$

Next, the optimum conditions can be found under which the interaction of collinear light waves is the strongest.

4. Note peculiarities of diffraction of light waves by sound in apodized composite structures. Anisotropic composite structures, in contrast to conventional crystals, allow any spatial distribution of the nanoparticles. This opens up new possibilities for the implementation of specific regimes of diffraction, which are prohibited in crystals with uniform properties. An analysis of the phenomenon of diffraction by an inhomogeneous sound wave shows that in certain cases it is possible to create conditions when the intensities of side diffraction maxima are significantly attenuated or absent. It is known that the reason for the appearance of side maxima, such as in the collinear diffraction of light by sound waves, consists in the presence of boundaries during the propagation of light, or, in other words, in the finiteness of the sound beam along the propagation direction of light.

Analysis of the diffraction conditions on a nonuniform sound wave, carried out in [8–11], showed that a suitable apodization of the sound beam can result in a significant suppression of side diffraction maxima. The condition for the required apodization is to be reduced to the fact that the amplitude of the sound wave and its derivatives at the boundaries of the interaction region should be small or zero (see papers [8, 11]). To ensure these conditions with the help of sound waves introduced from the outside is quite difficult; therefore, this diffraction regime has not been implemented in practice.

A completely different situation is possible in composite media, when, by changing the spatial distribution of the embedded nanoparticles, one can obtain any prescribed distribution of the photoelasticity constant responsible for the interaction of light with the sound wave. As an example, we consider the reflection of light from an isotropic heterogeneous composite medium when the addition to the dielectric constant tensor (3) can be represented in its simplest form:

$$\Delta\varepsilon(x) \equiv \left\{ \sin \left[2\pi \left(\frac{x}{l} - \frac{1}{2} \right) \right] \left[2\pi \left(\frac{x}{l} - \frac{1}{2} \right) \right]^{-1} \right\}^2$$

(l is the size or length of the composite region along the propagation direction of the light wave). The above-described spatial distribution can be realised with the help of a travelling sound wave only for a very short time when the apodized sound wave fills a part of the volume of the crystal, in which light interacts with sound, and so the diffraction without side maxima can exist for a relatively short time. A completely different situation occurs in a metamaterial, when such a nonuniform distribution can be realised due to an appropriate distribution of the nanoparticles. For example, if the exact phase-matching condition is fulfilled, the medium with the above distribution becomes fully transparent [11]. This result, of course, refers to the waves satisfying the exact phase-matching conditions. The nonuniform distribution of the nanoparticles finally leads to some violation of the phase-matching conditions, and therefore, such a structure will not be entirely transparent. However, by selecting the appropriate distribution of the nanoparticles and the value of $\Delta\varepsilon(x)$, the transparency of such a medium can be controllably high. It should be noted that this result relates to the media without absorption, i.e. when $\text{Im}\varepsilon_m = \text{Im}\varepsilon_p = 0$.

Introduction of nanoparticles whose density exceeds the density of the metamedium obviously changes the speed of acoustic waves. If the distribution of the nanoparticles is not uniform, the phase-matching condition for diffraction of the

light waves will depend on the coordinates and, therefore, will be fulfilled for a frequency-modulated light pulse in accordance with the spatial phase-matching condition.

Expression (3) derived for the dielectric constant tensor refers only to one kind of the nanoparticles, such as spheroids or spheres, but it is clear that the Maxwell Garnett model allows generalisation to the case of two or more different inclusions. To do this, by a matrix medium in formula (1) is meant the dielectric constant defined by expression (3), and by ϵ_p – the dielectric constant of a new kinds of the particles, whose concentration per unit volume will be η_1 (η is the previous concentration of the particles). Of course, it is now needed to satisfy the condition $\eta + \eta_1 \ll 1$.

The inclusion of the nanoparticles into the medium results in an increase of the absorption and scattering of the incident radiation, i.e. to extinction in such a heterogeneous medium. However, if the condition on the size of the nanoparticles is met: $a_{\text{lim}} \ll l/\sqrt{|\epsilon_{\parallel}|}$, $a_{\text{lim}} \ll l/\sqrt{|\epsilon_{\perp}|}$, where a_{lim} is the characteristic size of the nanoparticles (e.g., its diameter), extinction is very small (see paper [12], which gives the scattering, absorption and extinction cross sections for gold particles of various size, as in [1]). It can be seen that at $\alpha \sim 2-50$ nm the absorption and scattering cross sections of light at a wavelength of ~ 800 nm on one particle will not exceed $0.01\pi a_{\text{lim}}^2$ and will decrease with increasing wavelength. Of course, the given restriction imposes certain conditions on the concentration of the particles per unit volume of the medium; however, with increasing wavelength, i.e. for IR and THz wavelength ranges, these restrictions are sharply reduced.

We have considered above metamedia with inclusions, in which the permeability is equal to unity. Nevertheless, it is clear that the situation is completely analogous to the case of magnetic inclusions and inclusions of more complex geometric shapes. The use of the inclusions of the nanoparticles having complex shape (for example, different spirals [12]) and special magnetic, dielectric and conductive properties will allow one to produce qualitatively new acousto-optical devices with unique properties. Unlike conventional crystals, for metamedia there are no size restrictions, which make it possible to design devices for the IR region of the spectrum; as is known, in using conventional crystals it causes certain difficulties.

An explicit determination of the photoelastic constants for metamaterials is apparently an only example, when the values of photoelasticity can be found theoretically, since, as a rule, they are found experimentally.

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