

On the problem of controlled virtual refractive index

A N Oraevskii, I E Protsenko, A Botero

Abstract. A method for controlling the refractive index that is based on the backward rescattering of the scattered and amplified electromagnetic waves is proposed and analysed. The resulting field is a sum of the initial (probe) wave and the rescattered wave. The effective wave number of the total wave can be varied by varying the gain. This variation in the wave number can be treated as a variation in the refractive index, although the physical properties of the medium do not change. The effective refractive index for the total wave propagating in such a device was called the virtual refractive index.

1. Formulation of the problem

The creation of a medium with a large controlled refractive index would allow new approaches to the construction of optical devices. For example, if the refractive index of the material of a lens could be controlled, the lens would become a zoom lens, allowing the construction of interesting technical devices.

The simplest solution to the problem of controlled refractive index could be based on the use of nonlinear polarisability. By applying a strong external field, one can change the polarisability (and, therefore, the refractive index) of a medium for another, weak field. This straightforward approach has the drawback that the application of a strong controlling field can only reduce the effective polarisability of the medium. Therefore, one cannot count on an appreciable increase in the refractive index, especially in the optical frequency range.

In this work, we discuss the possibility of not only varying but also significantly increasing the effective refractive index of a medium by applying an external perturbation. The method was inspired by Lorentz's notion of the local field [1, 2]. According to Lorentz, each particle of matter is subjected to the action of the local field, which is a sum of the average (Maxwell) field \mathbf{E} and the additional field \mathbf{E}_{aux} produced by the surrounding polarised particles:

$$\mathbf{E}_{\text{loc}} = \mathbf{E} + \mathbf{E}_{\text{aux}}, \quad \mathbf{E}_{\text{aux}} = (4\pi/3)\mathbf{P}. \quad (1)$$

In the linear approximation, $\mathbf{P} = \alpha N \mathbf{E}_{\text{loc}}$, where α is the polarisability of a single particle and N is their concentration. As a result,

$$\mathbf{P} = \chi \mathbf{E}, \quad \chi = \frac{\alpha N}{1 - (4\pi/3)\alpha N}. \quad (2)$$

Formally, expression (2) permits very large values of the refractive index $n = \sqrt{\varepsilon} = (1 + 4\pi\chi)^{1/2}$. Indeed, the polarisability α can be made purely real in atomic gases by using the so-called Λ -scheme of excitation [3–5]. In this case, according to Eqn. (2), we have $\chi \rightarrow \infty$ for $N \rightarrow N_{\text{cr}} \equiv 3/(4\pi\alpha)$.

However, the practical realisation of this idea is problematic due to the fact that the polarisability α ceases to be constant for $N \geq \lambda^{-3}$ (λ is the radiation wavelength) and starts to decrease approximately as $1/N$. Usually, $3/(4\pi\alpha) \gg \lambda^{-3}$, and χ approaches a value near αN with increasing particle concentration.

The method proposed in this work consists in the creation of a sufficiently strong additional field that is proportional to the external field. It can be realised using the scheme shown in Fig. 1. A wave of the external field $\mathbf{E}(\mathbf{r}, t)$ enters the probed sample (p-sample) and is partially scattered in it. The scattered wave is amplified in the amplifier and forms another wave $\mathbf{E}_c(\mathbf{r}, t)$, which undergoes secondary scattering, thus forming the additional field. Hopefully, for sufficiently large gains, the effective polarisability will also be large and controllable by the variation of the amplifier parameters. The following calculations show how justified these hopes are.

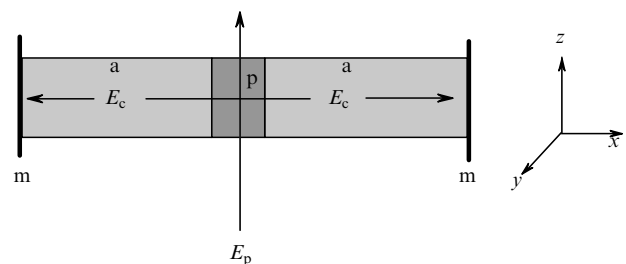


Figure 1. Schematic of the setup for the control of the refractive index: (p) p-sample with controlled refractive index, (a) amplifier medium, (m) mirrors of the amplifier cavity.

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2. Effective polarisability

We will consider the case of the monochromatic field

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}(\mathbf{r}) \exp(-i\omega t). \quad (3)$$

It follows from the Maxwell equations that

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}) - k_0^2 \mathbf{E}(\mathbf{r}) = 4\pi k_0^2 \mathbf{P}(\mathbf{r}), \quad (4)$$

where $\mathbf{P}(\mathbf{r}) = \chi(\mathbf{r})\mathbf{E}(\mathbf{r})$ and $\chi(\mathbf{r})$ is the dielectric susceptibility. Suppose that

$$\chi = \chi_a + \chi_p + \delta\chi(\mathbf{r}), \quad (5)$$

where χ_a is constant over the amplifier medium and vanishes in the p-sample, whereas $\delta\chi(\mathbf{r})$ is nonzero in the p-sample and vanishes in the amplifier medium. In the following calculations, we will assume that the scattering is induced by the spatial dependence $\delta\chi(\mathbf{r})$ of the dielectric susceptibility of the p-sample. This dependence may be due to density fluctuations or an artificial periodic structure (superlattice). In this work, we consider the scattering by density fluctuations (Rayleigh scattering); the scattering by a superlattice will be considered elsewhere.

To simplify equation (4), we divide the field into two parts:

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) + \delta\mathbf{E}(\mathbf{r}), \quad \nabla \mathbf{E}_0(\mathbf{r}) = 0. \quad (6)$$

If we assume that the quantities $\delta\chi(\mathbf{r})$ and $\delta\mathbf{E}(\mathbf{r})$ are small and neglect their product, it follows from the condition $\nabla[(1+4\pi\chi)\mathbf{E}] = 0$ that

$$\delta\mathbf{E}(\mathbf{r}) = -\frac{4\pi}{\varepsilon} \delta\chi(\mathbf{r})\mathbf{E}_0(\mathbf{r}) + \text{rot}\mathbf{C}, \quad \varepsilon = 1 + 4\pi(\chi_a + \chi_p), \quad (7)$$

where \mathbf{C} is an arbitrary vector, which can be taken equal to zero. Simple transformations using Eqn. (7) finally result in the equation

$$\nabla^2 \mathbf{E}_0(\mathbf{r}) + k_0^2 \varepsilon \mathbf{E}_0(\mathbf{r}) = -\frac{4\pi}{\varepsilon} \nabla \times \nabla \times [\delta\chi(\mathbf{r})\mathbf{E}_0(\mathbf{r})]. \quad (8)$$

2.1. Amplification of the scattered wave

In accordance with the geometry of the problem, we represent the field $\mathbf{E}_0(\mathbf{r})$ as the sum of the probe ($\mathbf{E}_p(\mathbf{r})$) and cavity part ($\mathbf{E}_c(\mathbf{r})$) components

$$\mathbf{E}_0(\mathbf{r}) = \mathbf{E}_p(\mathbf{r}) + \mathbf{E}_c(\mathbf{r}), \quad (9)$$

$$\mathbf{E}_p(\mathbf{r}) = E_p \mathbf{U}_p(\mathbf{r}), \quad \mathbf{E}_c(\mathbf{r}) = E_c \mathbf{U}_c(\mathbf{r}).$$

where $\mathbf{U}_p(\mathbf{r})$ and $\mathbf{U}_c(\mathbf{r})$ are assumed to be orthogonal to each other. To derive the equation for the cavity field, we multiply the both sides of Eqn. (8) by $\mathbf{U}_c(\mathbf{r})$ and integrate over the total volume of the system. In doing so, we neglect the diffraction due to finite dimensions of the system components. This can be done because, in this problem, we are interested in the scattering at large angles, whereas the diffractive scattering occurs at small angles provided that the dimensions of the scatterer are much larger than the wavelength.

Finally, we obtain the relationship

$$E_c = \frac{1}{\Delta - i/Q} (R_{cc}E_c + R_{pc}E_p), \quad (10)$$

where the following notation is used:

$$\Delta = \left(1 - \frac{\omega_c^2}{\omega^2}\right) \frac{\varepsilon_a V_{ac} + \varepsilon_p V_{pc}}{V_{cc}}; \quad \frac{1}{Q} = \frac{\varepsilon_a'' V_{ac} + \varepsilon_p'' V_{pc}}{V_{cc}}; \quad (11)$$

$$\mathbf{j}_{p,c} = \frac{4\pi}{\varepsilon_p} \nabla \times \nabla \times [\delta\chi(\mathbf{r})\mathbf{U}_{p,c}(\mathbf{r})];$$

$$R_{pc} = \frac{1}{V_{cc}} \int_p \mathbf{j}_p(\mathbf{r})\mathbf{U}_c^*(\mathbf{r})d\mathbf{r}; \quad R_{cc} = \frac{1}{V_{cc}} \int_p \mathbf{j}_c(\mathbf{r})\mathbf{U}_c^*(\mathbf{r})d\mathbf{r}. \quad (12)$$

$$V_{cc} = \int_c |U_c(\mathbf{r})|^2 d\mathbf{r}; \quad V_{pc} = \int_p |U_c(\mathbf{r})|^2 d\mathbf{r};$$

$$V_{ac} = \int_a |U_c(\mathbf{r})|^2 d\mathbf{r}.$$

The subscripts in Eqns (12) denote the integration over the total cavity volume (c), the amplifier volume (a), or the p-sample volume (p). The cavity eigenfrequency ω_c appearing in Eqn. (11) is determined by the equation

$$(c^2 \nabla^2 + \omega_c^2 \varepsilon') U_c(\mathbf{r}) = 0 \quad (13)$$

and the appropriate boundary conditions. We assume that the imaginary part ε_a'' of the permittivity of the amplifier describes the balance between the gain in the active medium and possible losses (for example, due to the finite reflectivity of the cavity mirrors). In Eqn. (10), we did not express E_c explicitly in terms of E_p to simplify the subsequent calculation of average values.

2.2. The equation for the wave vector of the probe field

To determine the effective refractive index, we have to derive the equation for the modulus of the probe field wave vector. To do this, suppose that

$$\langle \delta\chi(\mathbf{r}) \rangle = 0, \quad \langle \delta\chi(\mathbf{r})\delta\chi(\mathbf{r}') \rangle = S\delta(\mathbf{r} - \mathbf{r}'), \quad (14)$$

where the correlation amplitude S is a characteristic of the medium. This is a standard assumption in the studies of Rayleigh scattering [6,7]. Assume also that

$$U_p(\mathbf{r}) = V_p(\mathbf{r}) \exp ik_p z, \quad U_c(\mathbf{r}) = V_c(\mathbf{r}) \cos k_c x, \quad (15)$$

where the functions $V_{p,c}(\mathbf{r})$ vary slowly compared to $\exp ik_p z$ and $\cos k_c x$.

Using these assumptions and carrying out the calculations described in the Appendix, we obtain the following equation for the wave number k_p .

$$k_p^2 - k_0^2 \varepsilon_p = k_p^2 K + ik_0 \varkappa, \quad (16)$$

where \varkappa is the extinction coefficient;

$$K = K' + iK''; \quad K' = G \frac{A\xi - B}{\xi^2 + 1}; \quad K'' = G \frac{A + B\xi}{\xi^2 + 1}; \quad (17)$$

$$G = 8\pi^2 \frac{k_c^2}{k_{20}} \frac{V_{cp}}{V_{cc} V_{pp}} Q; \quad A = \operatorname{Re} \frac{S}{\varepsilon_p^2}; \quad B = \operatorname{Im} \frac{S}{\varepsilon_p^2}. \quad (18)$$

Below, we assume that $(k_c - k_0)k_0 \ll 1$ and, correspondingly, $k_c^2/k_0^2 = 1$.

In a transparent sample, k_p is real. From Eqn. (16), it follows that k_p can be real only if

$$k_p^2 K'' + k_0^2 \varepsilon_p'' + k_0 \varkappa = 0. \quad (19)$$

In this case,

$$k_p^2 = \frac{k_0^2 \varepsilon_p'}{1 - K'}. \quad (20)$$

Formula (20) is similar to relationship (2), with the distinction that K' depends on the gain and can be controlled by varying it. It follows from Eqn. (20) that k_p can be arbitrary large if $K' \rightarrow 1$. However, Eqn. (20) ceases to be valid if $(k_p)^{-1}$ approaches the correlation length r_{cor} of the function $\delta\chi(r)$. In this case, the approximation of the correlation function by a delta function is no longer valid; therefore, k_p cannot exceed $1/r_{\text{cor}}$.

Taking into account Eqn. (20), we can rewrite Eqn. (19) as

$$K'' + \frac{1}{\varepsilon_p'} \left(\varepsilon_p'' + \frac{\varkappa}{k_0} \right) (1 - K') = 0. \quad (21)$$

If K' is close to unity, Eqn. (21) reduces to the relationship $K'' \approx 0$. According to Eqns (18), we then have

$$A = -B\xi, \quad K' = -GB. \quad (22)$$

Thus, we have reached the important conclusion that B must be negative.

The crucial question is whether K' can be made arbitrary close to unity. Since K' is proportional to the quality factor Q and coefficient B , we have to estimate these quantities.

2.3. The attainable Q factor

Seemingly, the Q factor can be made arbitrary large by increasing the gain. However, as the gain approaches the lasing threshold, the risk appears that the fluctuation of the amplifier parameters (first of all, the pumping power) will make it slip to the lasing regime. The passage to the lasing regime disrupts the linear relation between E_c and E_p (10) and thereby the entire considered scheme of the refractive index control. Therefore, the attainable Q factor is limited by fluctuations. This means that the Q factor defined by equation (11) cannot exceed

$$Q_{\text{max}} = \left[\left\langle (\delta\varepsilon_a'')^2 \right\rangle^{1/2} \frac{V_a}{V_{cc}} \right]^{-1}. \quad (23)$$

Suppose that $|\varepsilon_a''| = \beta_a N_a$, where N_a is the concentration of the active particles created by the pumping. If the pumping fluctuations follow Poisson statistics, then

$$\begin{aligned} \left\langle (\delta\varepsilon_a'')^2 \right\rangle^{1/2} \frac{V_a}{V_{cc}} &= \beta_a \frac{1}{V_{cc}} \left\langle (\delta N_a V_a)^2 \right\rangle^{1/2} \\ &= \beta_a \frac{1}{V_{cc}} \left\langle N_a V_a \right\rangle^{1/2}. \end{aligned} \quad (24)$$

If the amplifier is near the self-excitation mode and most of the losses occur in medium of the p-sample, we have

$$\beta_a \langle N_a \rangle V_a \approx \chi_p'' V_{pc}. \quad (25)$$

Inserting Eqns (24) and (25) in formula (23), we obtain

$$Q_{\text{max}} = \frac{V_{cc}}{(\beta_a \varepsilon_p'' V_{pc})^{1/2}}. \quad (26)$$

To give an example, we estimate Q_{max} of a semiconductor amplifier. For GaAs, we have $\beta_a \approx 2 \times 10^{-19} \text{ cm}^3$; in a metal, $\varepsilon_p'' \approx 0.5 - 1$, $V_{cc} \approx 1 \text{ cm}^3$, and $V_{pc} \approx 10^{-3} \text{ cm}^3$. This finally leads to $Q_{\text{max}} \approx 10^{12}$. For obvious reasons, the allowed spectral range of the probe signal is limited by the ratio ω/Q .

2.4. Coefficients A and B

This specific form of coefficients A and B depends on the material of the p-sample. As an example of the p-sample, we will consider the suspension of dielectric or metallic nanospheres in a liquid or a solid. It is implied that the diameter of the nanospheres is much smaller than the wavelength.

The Rayleigh scattering of radiation by nanospheres was studied in Ref. [8]. It was shown that the correlation amplitude S is given by the equations

$$S = (\delta\alpha)^2 N_b, \quad \delta\alpha = b^3 \frac{\varepsilon_b - \varepsilon_s}{\varepsilon_b + 2\varepsilon_s}, \quad (27)$$

where b is the nanosphere radius; ε_b and ε_s are the dielectric susceptibilities of the nanospheres and the suspender, respectively; and N_b is the nanosphere concentration. Suppose that $b \approx 10 \text{ nm}$ and $N_b \approx 10^{17} \text{ cm}^{-3}$. In this case, nanospheres occupy 40% of the suspension volume and, according to the Clausius - Mossotti relationship (Ref. [9], p. 373), $(\varepsilon_p - 1)/(\varepsilon_p + 2) = 0.4(\varepsilon_b - 1)/(\varepsilon_b + 2) + 0.6(\varepsilon_s - 1)/(\varepsilon_s + 2)$.

We will assume that the dielectric susceptibility of the suspender is purely real. Then, according to formula (18), coefficients A and B of the suspension depend on the parameters ε_b' , ε_b'' and ε_s , resulting in a great number of different variants of these coefficients. Figs 2 and 3 show some of these variants for the coefficient B . First, one can see that the dependence of B on the medium parameters is not monotonous. Second, it turns out that there are intervals of parameter values where the coefficient B is negative, as required. Third, one can see that the coefficient B can be appreciable for negative values of ε_b' .

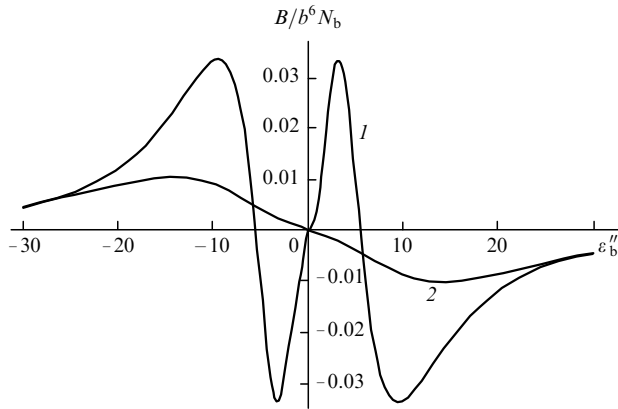


Figure 2. Quantity $B/b^6 N_b$ as a function of ε_b'' for $\varepsilon_s = 2$ and $\varepsilon_b' = 10$ (1) and 2 (2).

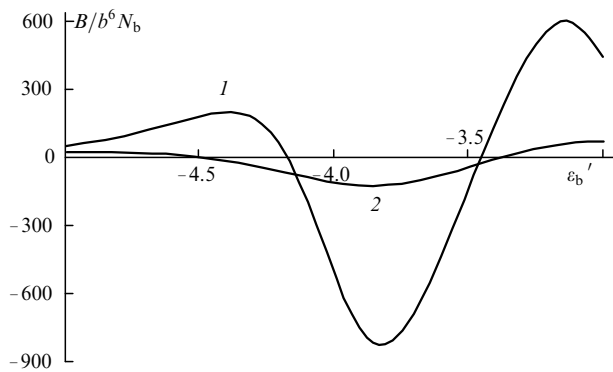


Figure 3. Quantity $B/b^6 N_b$ as a function of ε_b' for $\varepsilon_s = 2$ and $\varepsilon_b'' = 0.5$ (1) and 1 (2).

It is known that, for the optical frequency range in metals, $\varepsilon_b' < 0$ [10]; therefore, suspensions of metallic nanospheres are promising for our purposes. The nanosphere dimensions should be lower than the thickness of the skin layer at the mentioned frequencies, which agrees with the value of the nanosphere radius introduced above. One should therefore use nanospheres with the radius of the order of 10 nm. For a nanosphere concentration of $N_b \approx 10^{17} \text{ cm}^{-3}$, we then have $B \approx 10^{-17} - 10^{-16}$. Given an attainable Q factor of the order of 10^{12} , we can count on $K' \approx 10^{-3}$, which is rather far from the desired unity.

There is a possibility to increase significantly the Q factor and, therefore, the quantity K by modulating the gain in such a way that it periodically passes through the threshold value. In this case, as the gain approaches the threshold from below, the system may be in a mode with a large refractive index. However, this approach requires that the system operate in a pulsed mode.

The coefficient K can be drastically increased by changing the system geometry. The geometry of the amplifier shown in Fig. 1 allows using only a small fraction of the scattered light. A significantly greater part of the scattered light can be used in a system with the cylindrical or spherical geometry (Fig. 4). In the case of the cylindrical geometry, the usable fraction of the scattered light increases by a factor of $2\pi R/\lambda$, where R is the radius of the cylindrical p-sample. In the case of the spher-

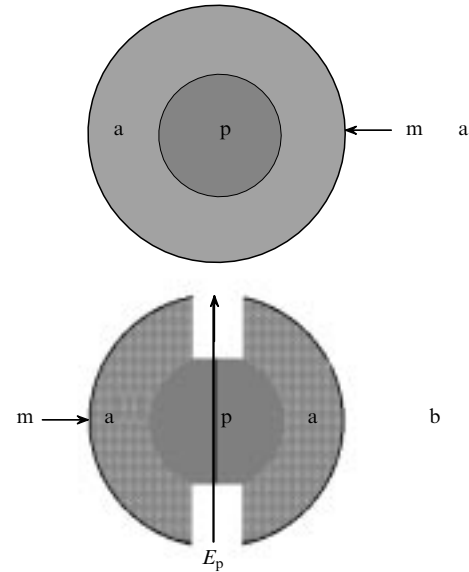


Figure 4. (a) Cylindrical variant of the system: the cross section of the setup by the plane perpendicular to the propagation direction of the probe wave. (b) Spherical variant of the system: the cross section of the setup by the great-circle plane parallel to the propagation direction of the probe wave. The notation is the same as in Fig. 1.

rical geometry, the enhancement factor equals $(2\pi R/\lambda)^2$, where R is the radius of the spherical p-sample. For $R \approx 0.1 \text{ cm}$ and $\lambda \approx 1 - 0.5 \text{ }\mu\text{m}$, the enhancement factor of the cylindrical geometry is 10^4 , which is enough to reach the required values of coefficient K near unity.

The field distribution in cavities with the cylindrical or spherical geometry is inhomogeneous along the radius. This fact calls for a special analysis of the systems with a cylindrical or spherical shape.

Apart from the nanosphere suspension, we studied various resonance media: atomic gases, atoms embedded in a matrix, and quantum dots. The two-level approximation was used in the calculations. All mentioned media produce noticeably worse results than the nanosphere suspension. In the resonance media, saturation of the resonance transition can be a harmful factor that strongly limits the intensity of the input (probe) wave. In gaseous media, the attainable Q factor is also limited by the fast dissipation of fluctuations under the influence of sound waves.

3. Conclusions

The performed analysis shows that the proposed method for the control of the refractive index is physically grounded. However, further investigation is needed to solve this problem. First, the systems with the cylindrical or spherical geometry require further analysis. Second, one should consider the possibility of manufacturing the p-sample from the material that can be kept at the temperature of a phase transition and then utilising the critical opalescence [6, 7]. Materials with the fractal structure can also be expected to produce strong scattering. Third, in the case of a sufficiently strong field E_c , four-wave mixing can play an important role that was neglected in this discussion.

An obvious modification of the proposed method is the use of a periodic structure, such as a superlattice or a photonic crystal, as the p-sample. Unlike the Rayleigh scattering, the scattering by regular structures is coherent and

therefore more intense. However, in the case of scattering by a periodic structure, the effective wave number k_p is rather rigidly fixed to the structure period, which limits the possibilities for its control. One should also keep in mind that, in periodic structures, there exist forbidden bands of the wave vector. These circumstances call for a special treatment of the problem of utilisation of periodic structures in the systems with the controlled refractive index.

The choice of the active medium of the amplifier is also an important problem. Here, the main problem is how to suppress the amplification of the spontaneous emission (superluminescence). It can be solved by using a suitable two-component medium. One of the components should be amplifying, whereas the other should be an absorber with a low concentration of absorbing particles and a large absorption cross section. This would provide sufficiently strong absorption and a weak intensity of the saturating field. Such a medium has a threshold with respect to the initial intensity of the amplified field [11, 12]. This threshold should be chosen so as to prevent the amplification of the spontaneous emission, but it should not hinder the amplification of the scattered radiation. If the gain medium is solid, one can manufacture a layered sample with periodically interchanging amplifying and absorbing layers. The material of the absorbing layers should satisfy the conditions mentioned above.

A high quality factor Q of the amplifier allows the efficient control of the group velocity, which was successfully realised in Refs [13, 14]. For $Q \approx 10^{11} - 10^{12}$, the group velocity can be reduced to a few centimetres per second.

The system considered in this paper is based on the use of an amplifier and therefore can be termed as an ‘active system’. Passive slowing systems are widely used in microwave devices (see Ref. [9], p. 45, and Ref. [15]). Similar systems for the optical range are also possible.

One of such systems can be a string of dielectric microspheres threaded by a dielectric waveguide. When a wave propagates in such a waveguide, electromagnetic oscillations are induced in the microspheres by the near-surface fields of the waveguide and the microspheres. The whispering-gallery modes of microspheres have high Q factors. By retaining the energy of the propagating wave, they slow it down. The supercrystals composed of microspheres should possess similar properties.

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Appendix

Suppose that the fields E_p and E_c are polarised along the y -axis. Then, introducing the unit vectors e_j , $j = x, y, z$, we recast the right-hand side of equation (8) to the form

$$\begin{aligned} \nabla \times \nabla \times [\delta\chi(\mathbf{r})\langle E(\mathbf{r}) \rangle] &= \left\{ -\frac{\partial^2}{\partial x \partial y} [E(\mathbf{r})\delta\chi(\mathbf{r})] \right\} e_x \\ &+ \left\{ -\frac{\partial^2}{\partial x^2} [E(\mathbf{r})\delta\chi(\mathbf{r})] - \frac{\partial^2}{\partial z^2} [E(\mathbf{r})\delta\chi(\mathbf{r})] \right\} e_y \\ &+ \left\{ -\frac{\partial^2}{\partial z \partial y} [E(\mathbf{r})\delta\chi(\mathbf{r})] \right\} e_z. \end{aligned} \quad (\text{A1})$$

The projections of the vector j on the axes x and z can be neglected in equation (A1) since the fields are slowly varying along the y -axis. Thus,

$$\begin{aligned} \nabla \times \nabla \times [\delta\chi(\mathbf{r})\langle E(\mathbf{r}) \rangle] &\approx \left\{ -\frac{\partial^2}{\partial x^2} [E(\mathbf{r})\delta\chi(\mathbf{r})] \right. \\ &\left. - \frac{\partial^2}{\partial z^2} [E(\mathbf{r})\delta\chi(\mathbf{r})] \right\} e_y. \end{aligned} \quad (\text{A2})$$

Next, a series of routine operations is required: replace E_c in Eqn. (A2) by its expression through E_p (10); introduce the resulting relationship into equation (8); multiply the both sides of the new equation by $U_p(\mathbf{r}) = V_p(\mathbf{r}) \exp k_p z$ and integrate the products over the total system volume, taking into account the orthogonality of $U_p(\mathbf{r})$ and $U_c(\mathbf{r})$; and carry out the statistical averaging. The statistical averaging has to do with averages of the following type

$$\left\langle \frac{\partial^\mu}{\eta^\mu} \delta\chi(\mathbf{r}) \frac{\partial^\nu}{\eta^\nu} \delta\chi(\mathbf{r}') \right\rangle, \quad (\text{A3})$$

where η stands for x or z ; $\mu, \nu = 0, 1, 2$. To calculate these quantities, we use the relationship

$$\begin{aligned} \int dx' \int dx'' \frac{\partial^{\mu+\nu} \delta(x-x')}{\partial x^\mu \partial x'^\nu} f(x, x') \\ = (-1)^{\mu+\nu} \int dx' \int dx'' \delta(x-x') \frac{\partial^{\mu+\nu} f(x, x')}{\partial x^\mu \partial x'^\nu}, \end{aligned} \quad (\text{A4})$$

which can be derived using $\delta\chi(\mathbf{r})$:

$$\begin{aligned} \delta\chi(\mathbf{r}) &= \int \delta\chi_q \exp(i\mathbf{q}\mathbf{r}) d\mathbf{q}, \\ \frac{\partial^\mu}{\partial z^\mu} \delta\chi(\mathbf{r}) &= \int (iq_z)^\mu \delta\chi_q \exp(i\mathbf{q}\mathbf{r}) d\mathbf{q}. \end{aligned} \quad (\text{A5})$$

Note that, $\langle \delta\chi(\mathbf{r})\delta\chi(\mathbf{r}') \rangle = S\delta(\mathbf{r}-\mathbf{r}')$, then $\langle \delta\chi_q \delta\chi_{q'}(\mathbf{r}') \rangle = [S/(2\pi)^3] \delta(\mathbf{q}+\mathbf{q}')$. In the calculations, we assumed that $E_p = \langle E_p \rangle + \delta E_p$, and $E_c = \langle E_c \rangle + \delta E_c$, and neglected the third-order terms, such as $\langle \delta E_{p,c} \delta\chi(\mathbf{r})\delta\chi(\mathbf{r}') \rangle$. As a result of these calculations, we obtained equation (17) for the wave number k_p .

In the derivation of equation (17), the term $\langle E_p \mathbf{j}_p(\mathbf{r}) \rangle$, which describes extinction [6, 7], appears. It is taken into account by introducing the extinction coefficient κ .

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