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Phenomenological model of a broadband optical absorber

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Modelling of the process of electromagnetic wave absorption in a composite layer of nanotubes is carried out on the basis of the phenomenological model of the composite layer of an anisotropic black body. The anisotropic black body model allowed to achieve a small reflection and absorption coefficient of more than 95% in a wide wavelength range, which agrees well with the experimental data.

Keywords: broadband absorbers, carbon nanotubes, black body model.

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Introduction

Ultra-wideband absorbing coatings are used in a wide range of modern optical systems [1–4], including scattered light absorbers to improve the resolution of sensitive spectroscopes, telescopes, microscopes, and optical sensor devices, solar cells [5], for countering visual-optical and optoelectronic reconnaissance [6] and in gravitational interferometers [7], such as LIGO and Virgo, where scattered light can introduce significant noise into the signal at the detector.

The studies devoted to perfect optical absorbers based on metamaterials are of particular interest [8–10]. Most optical absorbers depend on the direction of emission of electromagnetic waves, which greatly limits their use in solar cells, plasmon sensors and thermal emitters, since they can completely absorb light only in narrow angle ranges [1,2]. Therefore, special attention is paid to angle-insensitive absorbers due to their ability to almost completely absorb light at any angle of incidence [11–14]. The Massachusetts Institute of Technology presented a unique material Vantablack [15,16] in 2019 that absorbed up to 99.965% visible light and had a nanoscale average surface roughness of oriented carbon nanotubes (CNT). In the following years, nanostructured absorbing materials based on a forest of vertically-aligned multi-wall carbon nanotubes (VAMCNT) became a key subject in the field of studies of superabsorbing ultra-wideband materials.

When studying diffraction by tin screens or bodies with thin-film coating, it is convenient to use the method of approximate boundary conditions (ABC). For example, two-sided impedance ABCs are obtained in Ref. [17], including for a „black“ coating, which were used to simulate the characteristics of wave scattering by bodies with absorbing coating. For dielectric layers, thin films with curvature, thin metal gratings, etc., two-sided ABC are introduced, in which the thickness of the screen or coating is much less than the wavelength.

Two models were used to theoretically simulate the absorption of electromagnetic waves in the sample.

The first model is a two-dimensional periodic structure of vertical dielectric absorbing cylinders of finite height on a dielectric substrate. The study method is based on the solution of volumetric integro-differential equations for dielectric bodies [18]. Calculations show that it is possible to obtain almost 100%-absorption in a narrow wavelength range. It is possible to obtain a small reflectance in a wider range, but at the same time a large transmission coefficient is obtained. Therefore, this model does not describe the experimental results well. This can be explained by a number of reasons: CNTs are arranged randomly, they have different lengths, they are not perpendicular to the substrate, etc.

The second model is a modification of the Sommerfeld model of black body. From the point of view of electrodynamics, the equality of the characteristic resistances is necessary for the absence of reflection from the interface of media. At normal wave incidence from vacuum (air) the absorber should have

$$\mu = \varepsilon, \ \mathrm{Im}\,\varepsilon \gg \mathrm{Re}\,\varepsilon. \tag{1}$$

This is a well-known blackbody model proposed by Sommerfeld [19].

The purpose of this work is to develop a simple mathematical model of an ultra-wide-band anisotropic absorbing layer and apply it to the theoretical study of the optical characteristics of absorbing coatings, including those based on the CNT forest.

The studied structure is shown in Fig. 1. Absorbing dielectric layer of CNT with complex magnetic μ and dielectric ε permeability, in accordance with the proposed Sommerfeld blackbody model, satisfies the conditions $\mu = \varepsilon$, $\mathrm{Im}\,\varepsilon \gg \mathrm{Re}\,\varepsilon$. Such a model well describes the absorption characteristics at low angles of incidence $\varphi < 20^\circ$. We propose an anisotropic blackbody model to describe the broad-band absorption (see Appendix): the absorbing layer

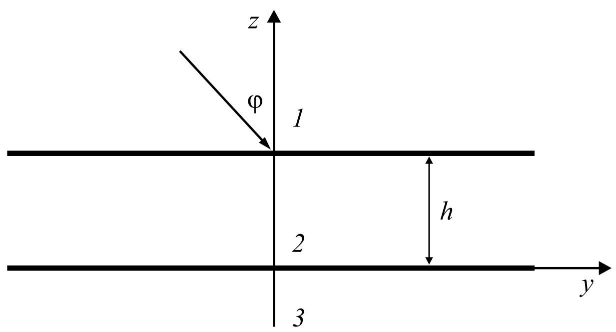

Figure 1. A two-dimensional model of the heterostructure of an anisotropic dielectric layer on a substrate.

is replaced by an anisotropic dielectric with complex $\hat{\mu}$ and $\hat{\varepsilon}$, $\hat{\mu} = \hat{\varepsilon}$:

$$\hat{\varepsilon} = \begin{pmatrix} \varepsilon_{\parallel} & 0 & 0 \\ 0 & \varepsilon_{\parallel} & 0 \\ 0 & 0 & \varepsilon_{\perp} \end{pmatrix}, \quad \hat{\mu} = \begin{pmatrix} \mu_{\parallel} & 0 & 0 \\ 0 & \mu_{\parallel} & 0 \\ 0 & 0 & \mu_{\perp} \end{pmatrix}, \tag{2}$$

symbols „$\parallel$“ and „$\perp$“ denote tensor components parallel and perpendicular to the substrate plane.

The anisotropic model corresponds to the spatial anisotropy of the CNT coating.

The introduction of anisotropy (2) has almost no effect on the results at small angles of incidence φ, however, it results in a large (by an order of magnitude) reduction of the reflectance at $\varphi > 30°$. Let's consider the reflectance from the interface of a vacuum–semi-infinite anisotropic dielectric to explain these results. The reflectance will look as follows for s-polarization:

$$R_s = \frac{1-\eta}{1+\eta}, \quad \eta = \frac{1}{\sqrt{1-\sin^2\varphi}} \sqrt{\frac{\varepsilon_{\parallel}}{\mu_{\parallel}} - \frac{1}{\mu_{\perp}\mu_{\parallel}} \sin^2\varphi}. \tag{3}$$

At small angles φ, it is sufficient to achieve the condition $\eta \approx 1$ to get $R_s \approx 0$, if we put

$$\mu_{\parallel} = \varepsilon_{\parallel}, \tag{4}$$

$\mu_{\parallel}$ and $\varepsilon_{\parallel}$ should be complex for the wave to attenuate in the dielectric. The ideal case $R_s = 0$ at any angles φ is theoretically possible if the following condition is met:

$$\mu_{\perp}\mu_{\parallel} = 1. \tag{5}$$

The condition (5) is impossible for complex μ, but, as shown by calculations it is possible to obtain a small reflectance at large angles of incidence if the following condition is met

$$\mathrm{Re}\,\mu_{\perp}\mathrm{Re}\,\mu_{\parallel} \approx 1. \tag{6}$$

It is necessary to replace μ with ε in expressions (2)–(6) to obtain expressions for p-polarization.

Therefore, it is sufficient to enter one parameter $\varepsilon_{\parallel}$ to describe the absorbing coating. It should be emphasized that $\mu_{\parallel}$, $\varepsilon_{\parallel}$ are just model parameters, not values of CNT dielectric and magnetic permeability. Therefore, $\mu \neq 1$ does not mean that CNTs have magnetic properties.

Figure 2 shows the results for a layer with $\varepsilon_{\parallel} = 15 - 2.9i$. The remaining parameters are determined by the formulas (1), (6). The dielectric constant of a semi-infinite substrate $\varepsilon_3 = 9$. The calculations for the isotropic layer $\varepsilon_{\parallel} = \mu_{\parallel} = 15 - 2.9i$, $\varepsilon_{\perp} = \mu_{\perp} = 1$ are also provided. The reflectance is an order of magnitude less at large angles of incidence.

The proposed model is in good agreement with the results of measurements of the parameters of commercial absorbent coatings [20,21] (Fig. 3–5). Fig. 2–4 show that the anisotropic blackbody model allows achieving a low reflectance and high absorption over over 95% in a wide range of wavelengths — 8 : 1. It should be noted that the coefficient of reflection from a thin layer is determined by the product $h\varepsilon''$ (Fig. 5).

Therefore, the results of modeling the amplitude-frequency characteristics of electromagnetic wave absorption in a composite layer of nanotubes performed by the composite layer of an anisotropic blackbody are provided. The frequency dependences of the power reflection coefficient were obtained for the anisotropic blackbody model at different angles of incidence of s- and p-polarized radiation. It was found that the anisotropic blackbody model makes it possible to achieve a low reflectance and absorption coefficient of more than 95% in a wide range of wavelengths, which is in good agreement with data from other sources.

Results of theoretical modeling of optical characteristics of ultra-wideband absorbing coatings based on wood CNTs can be used to develop a wide range of devices containing perfect absorbers that are insensitive to the absorption angle.

Appendix. Anisotropic blackbody model

For solving the problem of absorption of electromagnetic waves in a composite layer, as shown in Figure 1, let's consider an incident wave with a semi-infinite region *1* of free space onto an absorbing anisotropic layer *2* with a thickness of h, described by the complex permittivity tensor (1) and lying on a semi-infinite substrate *3*. The layers *1* and *2* of isotropic materials have magnetic permeability $\mu_1 = \mu_3 = 1$ and dielectric permittivity of the real ε_1 and ε_3, respectively.

Let's consider the incidence of s-polarized wave $\mathbf{E}_0(E_0, 0, 0)$ from the region *1*, where $E_0 = \exp(ik_y x - ik_z y)$, $k_y = k\sin\varphi$, $k_z = k\cos\varphi$, k — wave number, φ — the angle of incidence of the wave, measured relative to the normal to the surface of the layer *2*.

Using Maxwell's equations, it is possible to obtain the wave equation [22] in the layer *2*

$$\frac{\mu_{\parallel}}{\mu_{\perp}} \frac{\partial^2 E(y,z)}{\partial y^2} + \frac{\partial^2 E(y,z)}{\partial z^2} + k^2\mu_{\parallel}\varepsilon_{\parallel}E(y,z) = 0 \tag{A1}$$

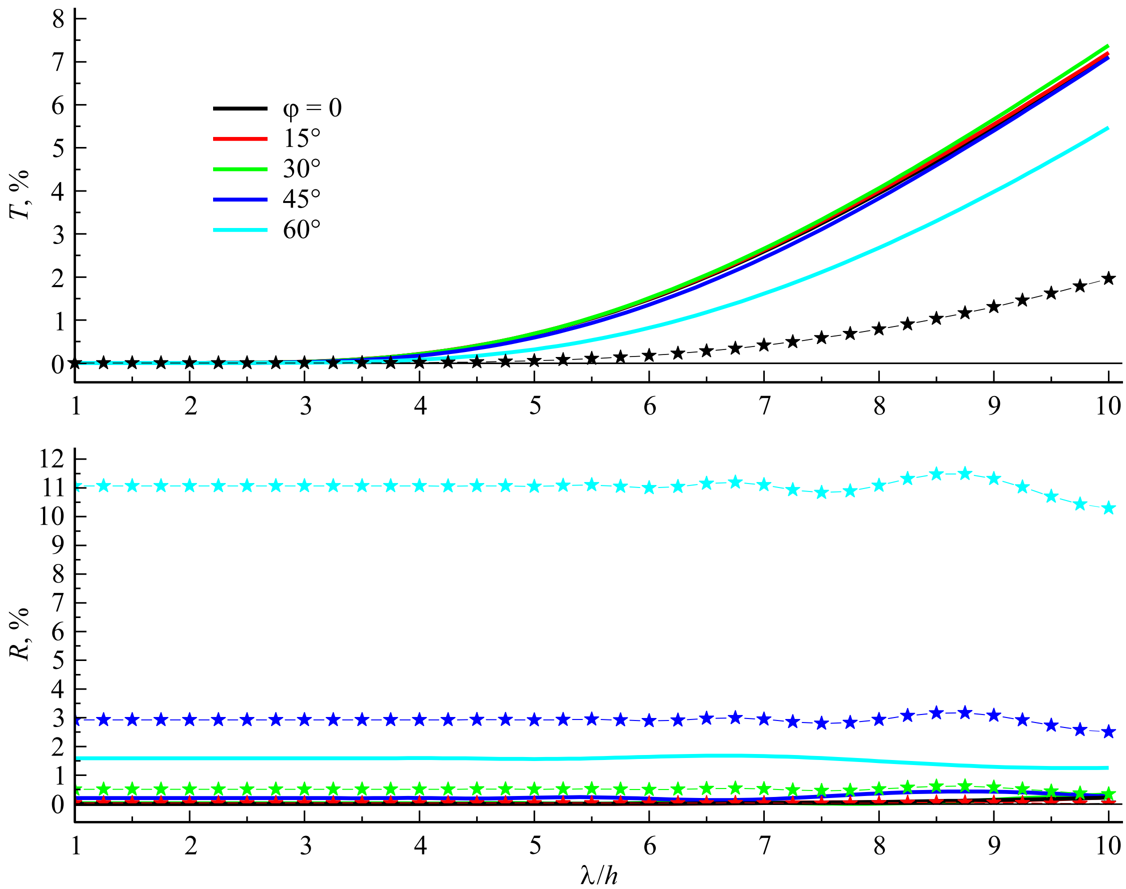

Figure 2. Coefficients of transmission T and reflection R in power from isotropic (lines with markers) and anisotropic layers with permittivity $\varepsilon_{\|} = 15 - 2.9i$ lying on a semi-infinite substrate with permittivity $\varepsilon_3 = 9$.

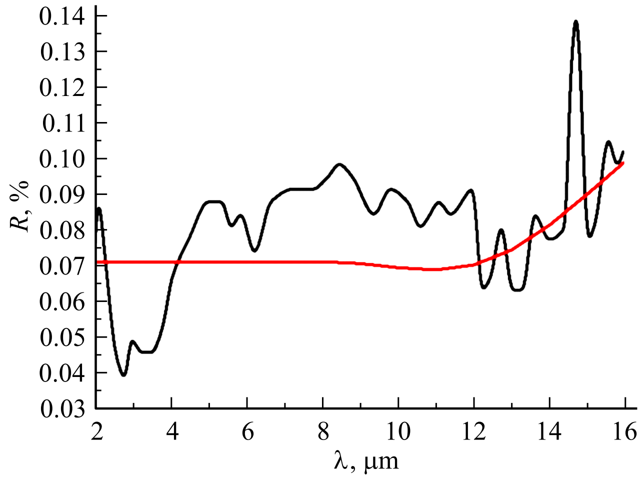

Figure 3. Frequency characteristics of the absorbing coating [20]. Black curve — experiment from the article [20], red curve — calculation at $\varepsilon_{\|} = 3 - i2.5$, $\mu_{\|} = 2.6 - i2.5$, $\varepsilon_{\perp} = \mu_{\perp} = 0.33$, $h = 14\,\mu$m.

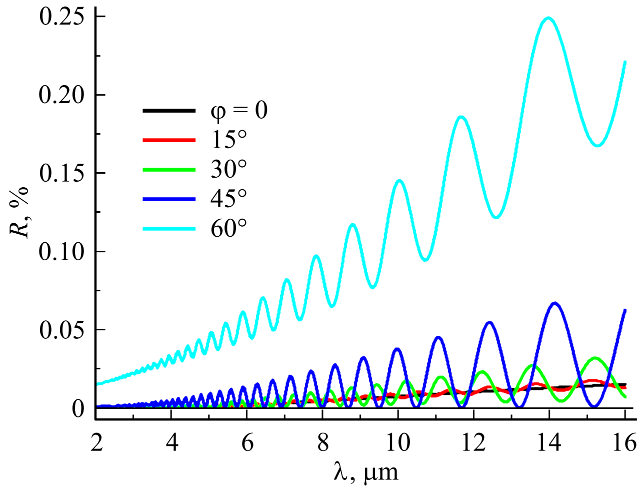

Figure 4. Another approximation of the results [20] by an anisotropic layer with a high dielectric constant. $h = 2.4\,\mu$m. $\varepsilon = 30 - i\varepsilon''$. ε'' varies linearly from 0.5 at $\lambda = 2\,\mu$m to 2 at $\lambda = 25\,\mu$m.

and the magnetic field strength

$$H_y = \frac{1}{i\omega\mu_{\|}}\frac{\partial E}{\partial z}, \quad H_z = -\frac{1}{i\omega\mu_{\perp}}\frac{\partial E}{\partial y}.$$

The equation (N1) will have the following form if we consider the axis z directed along the normal to the surface

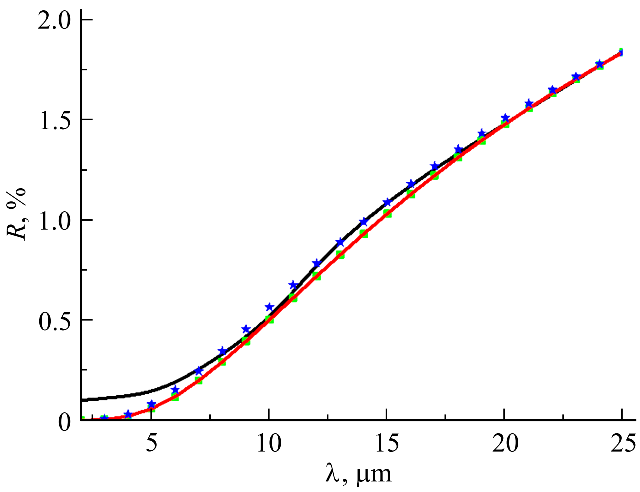

Figure 5. Frequency characteristics of the absorbing coating [21]. Black curve — experiment from the article [18], red curve — ($h = 0.1\,\mu$m, $\varepsilon' = 10$, $\varepsilon'' = 26$) with a square symbols — (0.2, 5,13), with asterisks — (0.4, 2.5, 6.5).

of the layer *2* and $E(y, z) = \bar{E}(z)\exp(ik_y y)$

$$\frac{d^2\bar{E}(z)}{dz^2} + \gamma_2^2\bar{E}(z) = 0, \qquad \text{(A2)}$$

where

$$\gamma_2^2 = k^2\mu_{\|}\varepsilon_{\|} - \frac{\mu_{\|}}{\mu_{\perp}}k_y^2.$$

The wave equation will have a similar (N2) form for areas *1* and *3* if γ_2^2 is replaced by $\gamma_n^2 = k^2\varepsilon_n - k_y^2$, $n = 1, 3$.

The solutions of the wave equations (N2) for layers *1–3* will have the following form

$$\bar{E}(z) = \begin{cases} \exp(-i\gamma_1(h-z)) + R\exp(i\gamma_1(h-z)), & z \geq h, \\ \frac{A\sin(\gamma_2 z) + B\sin(\gamma_2(h-z))}{\sin(\gamma_2 h)}, & 0 \leq z \leq h, \\ T\exp(i\gamma_3 z), & z \leq 0, \end{cases}$$

where R — reflectance, T — transmission coefficient and coefficients $A = 1 + R$, $B = T$, which can be determined from the condition of continuity of $\bar{E}(z)$ at interface.

It follows from the condition of continuity of $H(y, z)$ at the interface that:

$$\frac{\gamma_2}{\mu_{\|}}\frac{(1+R)\cos(\gamma_2 h) - T}{\sin(\gamma_2 h)} = i\gamma_1(1 - R),$$

$$\frac{\gamma_2}{\mu_{\|}}\frac{(1+R) - T\cos(\gamma_2 h)}{\sin(\gamma_2 h)} = i\gamma_3 T.$$

Solving this system of equations with respect to R and T, we obtain an expression for calculating the power reflection coefficient for s-polarized wave:

$$R_s = \frac{\left(\frac{\gamma_2^2}{\mu_{\|}^2} - \gamma_1\gamma_3\right)\sin(\gamma_2 h) + i\,\frac{\gamma_2}{\mu_{\|}}(\gamma_1 - \gamma_3)\cos(\gamma_2 h)}{-\left(\frac{\gamma_2^2}{\mu_{\|}^2} + \gamma_1\gamma_3\right)\sin(\gamma_2 h) + i\,\frac{\gamma_2}{\mu_{\|}}(\gamma_1 + \gamma_3)\cos(\gamma_2 h)} \qquad \text{(A3)}$$

It is necessary to replace $H \leftrightarrow E$ and $\varepsilon \leftrightarrow -\mu$for p-of the polarized wave in all formulas, including (N3). Since for a completely black body $\varepsilon = \mu$, the reflectances for both polarizations will be the same.

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Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] N.I. Landy, S. Sajuyigbe, J.J. Mock, D.R. Smith, W.J. Padilla. Phys. Rev. Lett., **100**, 207402 (2008). DOI: 10.1103/PhysRevLett.100.207402
[2] Z. Li, S. Butun, K. Aydin. ACS Photonics, **2**, 183 (2015). DOI: 10.1021/ph500410u
[3] D. Zhao, L. Meng, H. Gong, X. Chen, Y. Chen, M. Yan, Q. Li, M. Qiu. Appl. Phys. Lett., **104**, 221107 (2014). DOI: 10.1063/1.4881267
[4] P. Yu, L.V. Besteiro, Y. Huang, J. Wu, L. Fu, H.H. Tan, C. Jagadish, G.P. Wiederrecht, A.O. Govorov, Z. Wang. Adv. Opt. Mater., **7** (3), 1800995 (2019). DOI: 10.1002/adom.201800995
[5] A. Shevlyagin, V. Il'yaschenko, A. Kuchmizhak, E. Mitsai, A. Sergeev, A. Amosov, A. Gerasimenko, A. Gutakovskii. Appl. Surf. Sci., **602** 154321 (2022). DOI: 10.1016/j.apsusc.2022.154321
[6] T.V. Borbotko, H.N. Wang. BGUIR Reports, **5**, 23 (2005) (in Russian).
[7] A.F. Brooks, G. Vajente, H. Yamamoto, R. Abbott, C. Adams, R.X. Adhikari, A. Ananyeva, S. Appert, K. Arai, J.S. Areeda, Y. Asali, S.M. Aston, C. Austin, A.M. Baer, M. Ball, S.W. Ballmer, Sh. Banagiri, D. Barker, L. Barsotti, J. Bartlett, B.K. Berger, J. Betzwieser, D. Bhattacharjee, G. Billingsley, S. Biscans, C.D. Blair, R.M. Blair, N. Bode, Ph. Booker, R. Bork, A. Bramley, D.D. Brown, A. Buikema, C. Cahillane, K.C. Cannon, H.T. Cao, X. Chen, A.A. Ciobanu, F. Clara, C. Compton, S.J. Cooper, K.R. Corley, S.T. Countryman, P.B. Covas, D.C. Coyne, L.E. Datrier, D. Davis, Ch.D. Difronzo, K.L. Dooley, J.C. Driggers, P. Dupej, Sh.E. Dwyer, A. Effler, T. Etzel, M. Evans, T.M. Evans, J. Feicht, A. Fernandez-Galiana, P. Fritschel, V.V. Frolov, P. Fulda, M. Fyffe, J.A. Giaime, D.D. Giardina, P. Godwin, E. Goetz, S. Gras, C. Gray, R. Gray, A.C. Green, A. Gupta, E.K. Gustafson, D. Gustafson, E. Hall, J. Hanks, J. Hanson, T. Hardwick, R.K. Hasskew, M.C. Heintze, A.F. Helmling-Cornell, N.A. Holland, J.D. Jones, Sh. Kandhasamy, S. Karki, M. Kasprzack, K. Kawabe, N. Kijbunchoo, P.J. King, J.S. Kissel, R. Kumar, M. Landry, B.B. Lane, B. Lantz, M. Laxen, Ya.K. Lecoeuche, J. Leviton, L. Jian, M. Lormand, A.P. Lundgren, R. Macas, M. Macinnis, D.M. Macleod, G.L. Mansell, S. Marka, Z. Marka, D.V. Martynov, K. Mason, T.J. Massinger, F. Matichard, N. Mavalvala, R. McCarthy,

D.E. McClelland, S. McCormick, L. McCuller, J. McIver, T. McRae, G. Mendell, K. Merfeld, E.L. Merilh, F. Meylahn, T. Mistry, R. Mittleman, G. Moreno, C.M. Mow-Lowry, S. Mozzon, A. Mullavey, T.J. Nelson, P. Nguyen, L.K. Nuttall, J. Oberling, R.J. Oram, C. Osthelder, D.J. Ottaway, H. Overmier, J.R. Palamos, W. Parker, E. Payne, A. Pele, R. Penhorwood, C.J. Perez, M. Pirello, H. Radkins, K.E. Ramirez, J.W. Richardson, K. Riles, N.A. Robertson, J.G. Rollins, Ch.L. Romel, J.H. Romie, M.P. Ross, K. Ryan, T. Sadecki, E.J. Sanchez, L.E. Sanchez, S.R. Tiruppatturrajamanikkam, R.L. Savage, D. Schaetzl, R. Schnabel, R.M. Schofield, E. Schwartz, D. Sellers, Th. Shaffer, D. Sigg, B.J. Slagmolen, J.R. Smith, S. Soni, B. Sorazu, A.P. Spencer, K.A. Strain, L. Sun, M.J. Szczepanczyk, M. Thomas, P. Thomas, K.A. Thorne, K. Toland, C.I. Torrie, G. Traylor, M. Tse, A.L. Urban, G. Valdes, D.C. Vander-Hyde, P.J. Veitch, K. Venkateswara, G. Venugopalan, A.D. Viets, Th. Vo, Ch. Vorvick, M. Wade, R.L. Ward, J. Warner, B. Weaver, R. Weiss, Ch. Whittle, B. Willke, Ch.C. Wipf, L. Xiao, H. Yu, H. Yu, L. Zhang, M.E. Zucker, J. Zweizig. Appl. Opt., **60**, 4047 (2021). DOI: 10.1364/AO.419689

[8] Y. Fan, F. Zhang, Q. Zhao, Z. Wei, H. Li. Opt. Lett., **39**, 6269 (2014). DOI: 10.1364/OL.39.006269

[9] Y. Fan, N. Shen, F. Zhang, Q. Zhao, H. Wu, Q. Fu, Z. Wei, H. Li, C.M. Soukoulis. Adv. Opt. Mater., **7**, 1800537 (2019). DOI: 10.1002/adom.201800537

[10] H. Li, M. Qin, L. Wang, X. Zhai, R. Ren, J. Hu. Opt. Express, **25**, 31612 (2017). DOI: 10.1364/OE.25.031612

[11] G. Lu, F. Wu, M. Zheng, C. Chen, X. Zhou, C. Diao, F. Liu, G. Du, C. Xue, H. Jiang, H. Chen. Opt. Express, **27**, 5326 (2019). DOI: 10.1364/OE.27.005326

[12] F. Ding, J. Yi, B. Li, H. Cheng, L. Mo, S. He. Laser Photonics Rev., **8**, 946 (2014). DOI: 10.1002/lpor.201400157

[13] A.S. Hall, M. Faryad, G.D. Barber, L. Liu, S. Erten, T.S. Mayer, A. Lakhtakia, T.E. Mallouk. ACS Nano, **7** (6), 4995 (2013). DOI: 10.1021/nn4003488

[14] P. Yu, L.V. Besteiro, J. Wu, Y. Huang, Y. Wang, A.O. Govorov, Z. Wang. Opt. Express, **26** (16), 20471 (2018). DOI: 10.1364/OE.26.020471

[15] V. Ghai, H. Singh, P.K. Agnihotri. ACS Appl. Nano Mater., **2**, 7951 (2019). DOI: 10.1021/acsanm.9b01950

[16] J. Lehman, C. Yung, N. Tomlin, D. Conklin, M. Stephens. Appl. Phys. Rev., **5**, 011103 (2018). DOI: 10.1063/1.5009190

[17] A.M. Lehrer, V.V. Makhno, V.I. Kravchenko. ZhTF, **93** (4), 438 (2023) (in Russian). DOI: 10.21883/0000000000

[18] A.M. Lerer. J. Commun. Technol. Electron., **57** (11), 1151 (2012). DOI: 10.1134/S106422691210004X

[19] L.N. Zakhar'yev, A.A. Limanskiy *Rasseyaniye voln chernymi telami* (M.: Sov. radio, 1972) (in Russian).

[20] B.D. Wood. J. Appl. Phys., **118**, 013106 (2015). DOI: 10.1063/1.4923390

[21] *Vantablack® S-VIS/S-IR. Opticheskie svojstva i dannye kosmicheskilkh kvalifikacionnykh ispytanij* (Surrey NanoSystems 2023)

[22] F.I. Fedorov. *Optika anizotropnykh sred* (Editorial URSS, M., 2004), p. 379 (in Russian).

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