Russian Technological Journal. 2024;12(3):55–64 ISSN 2500-316X (Online)

Micro- and nanoelectronics. Condensed matter physics

Микро- и наноэлектроника. Физика конденсированного состояния

UDC 537.632
https://doi.org/10.32362/2500-316X-2024-12-3-55-64
EDN SWVVUI

RESEARCH ARTICLE

Modeling of the magnetorefractive effect in Co-Al_2O_3 nanocomposites in the framework of the Bruggeman approximation

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Abstract
Objectives. To investigate the magnetorefractive effect (MRE) in nanocomposites, which consists in changing the reflection, transmittance and light absorption coefficients of samples with large magnetoresistance (MR) upon their magnetization. Materials offering high magneto-optical activity and significant MR include magnetic nanocomposites. These materials are based on a polymer matrix, which includes inorganic magnetic particles, fibers or layered particles, whose nanometer sizes range from 1 to 100 nm in at least one dimension. The main purpose of creating such nanocomposites is to combine the special properties of several components in one material. The presence in such materials of gigantic, colossal and tunneling MR, as well as the giant anomalous Hall effect, is of practical interest. Uses range from magnetic recording, light modulation, and receivers for thermal radiation, while the MRE itself is a promising method for the non-destructive testing of any nanostructures, e.g., measuring MR.
Methods. The use of effective medium theory to describe the optics and magneto-optics of dispersed media provides a means to determine the complex permittivity of a medium through the permittivity of its constituent components or vice versa. The present work considers the example of a Co-Al_2O_3 nanocomposite with a concentration of ferromagnetic metal Co 0.4 near the percolation threshold. This particular case was considered for study, since all the properties of nanocomposites change dramatically near the percolation threshold.
Results. Using the Bruggeman effective medium approximation (EMA) to describe the optical and magneto-optical properties of nanocomposites on the example of Co-Al_2O_3, the characteristics of MRE are obtained, namely, the change in MRE for reflection and transmission of light at normal incidence and at the angle of incidence near the Brewster angle (below the percolation threshold) or the main angle of incidence for metals (above the percolation threshold), which enhances MRE. The advantage of the EMA is the ability to study magneto-optical spectra in the range of average volume concentrations of the metal component.
Conclusions. The obtained values correspond well to the known experimental data. Moreover, the described approach can be used to study any nanostructures.

Keywords: magnetorefractive effect, nanocomposites, magnetoresistance, dielectric permittivity tensor, Bruggeman approximation

• **Submitted:** 16.05.2023 • **Revised:** 16.10.2023 • **Accepted:** 08.04.2024

For citation: Mukhutdinova M.A., Yurasov A.N. Modeling of the magnetorefractive effect in Co-Al_2O_3 nanocomposites in the framework of the Bruggeman approximation. *Russ. Technol. J.* 2024;12(3):55–64. https://doi.org/10.32362/2500-316X-2024-12-3-55-64

Financial disclosure: The authors have no a financial or property interest in any material or method mentioned.

The authors declare no conflicts of interest.

НАУЧНАЯ СТАТЬЯ

Моделирование магниторефрактивного эффекта в нанокомпозитах Co-Al_2O_3 в рамках приближения Бруггемана

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Резюме

Цели. Цель работы – изучить магниторефрактивный эффект (МРЭ) в нанокомпозитах, заключающийся в изменении коэффициентов отражения, пропускания и поглощения света образцов с большим магнитосопротивлением (МС) при их намагничивании. Существует ряд материалов, обладающих большой магнитооптической активностью и значительным МС. К таким материалам относятся магнитные нанокомпозиты. Они представляют из себя материалы на основе полимерной матрицы, в которую включены неорганические магнитные частицы, волокна или слоистые частицы, с нанометровыми размерами от 1 до 100 нм хотя бы в одном измерении. Главной целью создания таких нанокомпозитов является совмещение нескольких компонентов с их особыми свойствами в одном материале. Наличие в таких материалах гигантского, колоссального и туннельного МС, гигантского аномального эффекта Холла представляет практический интерес. Данные материалы применяют для магнитной записи, модуляции света, как приемники теплового излучения, а сам МРЭ является перспективным методом неразрушающего контроля любых наноструктур, например, для измерения МС.

Методы. Для описания оптики и магнитооптики дисперсных сред рассмотрена теория эффективной среды, благодаря которой можно решить задачу определения комплексной диэлектрической проницаемости среды через диэлектрические проницаемости составляющих ее компонент или наоборот. В статье этот подход рассматривался на примере нанокомпозита Co-Al_2O_3 с концентрацией ферромагнитного металла Co, равной 0.4, вблизи порога перколяции. Для изучения рассмотрен именно этот случай, т.к. вблизи порога перколяции кардинально меняются все свойства нанокомпозитов.

Результаты. Используя приближение Бруггемана (effective medium approximation, EMA) для описания оптических и магнитооптических свойств нанокомпозитов на примере Co-Al_2O_3, авторы получили характеристики МРЭ, а именно: изменение МРЭ на отражение и пропускание света при нормальном падении и при угле падения вблизи угла Брюстера (ниже порога перколяции) или главного угла падения для металлов (выше порога перколяции), что усиливает МРЭ. Преимущество EMA заключается в возможности исследовать магнитооптические спектры в диапазоне средних объемных концентраций металлической компоненты.

Выводы. Полученные значения хорошо соответствуют известным экспериментальным данным. Важно отметить, что данный подход позволяет исследовать любые наноструктуры.

Ключевые слова: магниторефрактивный эффект, нанокомпозиты, магнитосопротивление, тензор диэлектрической проницаемости, приближение Бруггемана

• **Поступила:** 16.05.2023 • **Доработана:** 16.10.2023 • **Принята к опубликованию:** 08.04.2024

Для цитирования: Мухутдинова М.А., Юрасов А.Н. Моделирование магниторефрактивного эффекта в нанокомпозитах Co-Al_2O_3 в рамках приближения Бруггемана. *Russ. Technol. J.* 2024;12(3):55–64. https://doi.org/10.32362/2500-316X-2024-12-3-55-64

Прозрачность финансовой деятельности: Авторы не имеют финансовой заинтересованности в представленных материалах или методах.

Авторы заявляют об отсутствии конфликта интересов.

INTRODUCTION

The magnetorefractive effect (MRE) describes changes in the reflection $R\left(\frac{\Delta R}{R}\right)$, transmission $T\left(\frac{\Delta T}{T}\right)$ and absorption A coefficients of light in samples with large magnetoresistance (MR) when they are magnetized [1].

Nanocomposites are promising multiphase materials obtained by introducing nanoparticles having geometric particle sizes ranging from 1 to 100 nm into the matrix of a base material [1]. Nanocomposites have a significant MR $\frac{\Delta\rho}{\rho}$, where ρ is the electrical resistance. Significant opportunities are opened by the application of nanocomposites in the field of magneto-optics (MO), which studies the phenomena arising in the magnetic field as a result of the interaction of optical radiation with matter. For example, nanocomposites are used to measure MR. Magnetic nanocomposites, representing heterogeneous magnetics in which ferromagnetic particles are placed in a metal or dielectric matrix, are also used for magnetic recording, light modulation, and as receivers of thermal radiation [1–7]. In turn, MO methods are methods of nondestructive testing of any nanostructures [1, 2].

The purpose of the present work is to investigate MREs in nanocomposites near the percolation threshold.

CALCULATION METHODOLOGY

Under the influence of magnetic field, the dispersion curves of refractive index n and absorption coefficient k, leading to the appearance or change of optical anisotropy of the medium. When MR changes, the value of reflection and transmittance coefficients changes. The reflection coefficient (Fresnel formulae) at normal incidence has the form:

$$R = \frac{(1-n)^2 + k^2}{(1+n)^2 + k^2}, \quad (1)$$

where n is the refraction coefficient, and k is the extinction coefficient of the nanocomposite [1].

Formula (1) is used to calculate the value of $\frac{\Delta R}{R}$ [2]:

$$\frac{\Delta R}{R} = -(1-R)\cdot\frac{\Delta\rho}{\rho}\cdot k^2 \times \times \frac{3n^2 - k^2 - 1}{(n^2+k^2)((1-n)^2+k^2)}\cdot 100\%. \quad (2)$$

At an angle of incidence different from zero, the reflection coefficient for a semi-infinite medium is calculated by the following formulas:

$$R = |r_{12}|^2, \quad (3)$$

$$r_{12} = \frac{g_1\eta_2^2 - g_2 n_1^2}{g_1\eta_2^2 + g_2 n_1^2}, \quad (4)$$

where $g_1 = \sqrt{n_1 - n_1(\sin\varphi_0)^2}$, $g_2 = \sqrt{\eta_2 - n_2(\sin\varphi_0)^2}$, φ_0 is the angle of incidence of light on the nanocomposite surface, $\eta_2 = n_2 - k_2 i$ is the complex refractive index of the nanocomposite, and n_1 is the refractive index of the medium from which the light is incident [2].

For the transmittance at normal incidence of light we have [2]:

$$T = |t_{12}|^2, \quad (5)$$

$$t_{12} = \frac{g_1 2\eta_2}{g_1\eta_2^2 + g_2 n_1^2}, \quad (6)$$

$$\frac{\Delta T}{T} = \frac{1}{2}\cdot\frac{\Delta\rho}{\rho}\cdot Tk^2\cdot\left(\frac{2n^2+n}{n^2+k^2}\right)\cdot 100\%. \quad (7)$$

The effective dielectric permittivity tensor (DPT) is considered to describe the electrical, optical and magnetic properties of are complex nanocomposite materials:

$$\tilde{\varepsilon}^{\text{eff}} = \begin{pmatrix} \varepsilon_{xx}^{\text{eff}} & i\gamma^{\text{eff}} & 0 \\ -i\gamma^{\text{eff}} & \varepsilon_{xx}^{\text{eff}} & 0 \\ 0 & 0 & \varepsilon_{xx}^{\text{eff}} \end{pmatrix}, \quad (8)$$

where diagonal components $\varepsilon_{xx}^{\text{eff}} = (\varepsilon_{xx}^{\text{eff}})' - i(\varepsilon_{xx}^{\text{eff}})''$ are the optical constituent, while the non-diagonal components $\gamma^{\text{eff}} = (\gamma^{\text{eff}})' - i(\gamma^{\text{eff}})''$ are the MO constituent of the DPT [4–8].

The most interesting case arises when the nanocomposite is near the percolation threshold, since a significant enhancement of MO effects occurs in this region. In this connection, all calculations were performed when the volume concentration X = 0.4, which corresponds to the proximity to the percolation threshold.

Let us use the Bruggeman effective medium approximation (EMA) at $0.3 < X < 0.7$, which works well for describing nanocomposites at an average concentration of magnetic (metallic) component [7–10].

In order to find ε^{eff} and γ^{eff}, we use the following equations:

$$X\frac{(\varepsilon_1-\varepsilon^{\text{EMA}})}{\varepsilon^{\text{EMA}}+(\varepsilon_1-\varepsilon^{\text{EMA}})L_{xx}}+(1-X)\frac{(\varepsilon_0-\varepsilon^{\text{EMA}})}{\varepsilon^{\text{EMA}}+(\varepsilon_0-\varepsilon^{\text{EMA}})L_{xx}}=0,$$

$$X\frac{(\gamma^{\text{EMA}}-\gamma)}{\left[\varepsilon^{\text{EMA}}+(\varepsilon_1-\varepsilon^{\text{EMA}})L_{xx}\right]^2}+(1-X)\frac{\gamma^{\text{EMA}}}{\left[\varepsilon^{\text{EMA}}+(\varepsilon_0-\varepsilon^{\text{EMA}})L_{xx}\right]^2}=0, \quad (9)$$

where $\varepsilon^{\text{EMA}} = \varepsilon^{\text{eff}}$, $\gamma^{\text{EMA}} = \gamma^{\text{eff}}$, $L_{xx} = \frac{1-L}{2} = \frac{1-\frac{1}{3}}{2} = \frac{1}{3}$ is the particle form factor $\left(L = \frac{1}{3} \text{ for spherical particles}\right)$, $\varepsilon_0 = \varepsilon_0' - i\varepsilon_0''$ is the dielectric permittivity of the nonmetallic (non-ferromagnetic) component, $\varepsilon_1 = \varepsilon_1' - i\varepsilon_1''$ is the dielectric permittivity for the ferromagnetic component (the diagonal part of the corresponding DPT), $\gamma = \gamma' - i\gamma''$ is the non-diagonal component of the DPT of the ferromagnetic component [2].

CALCULATION RESULTS

We calculated the absolute values of the reflection $\frac{\Delta R}{R}$ and transmission $\frac{\Delta T}{T}$ MRE at incidence angles of $\varphi_1 = 0°$ and $\varphi_2 = 70°$, as well as the effective DPT at different wavelengths of the infrared (IR) range (from 0.8 to 10 μm) incidence on the surface of Co-Al_2O_3. If the nanocomposite is in a state below the percolation threshold, the choice of the angle $\varphi_2 = 70°$ is conditioned by its proximity to the Brewster angle, while, if the nanocomposite is in a state above the percolation threshold, the angle is determined by its proximity to the main angle of incidence for metals, which also enhances optical effects.

The results of calculations are given in Tables 1–6. Here E is the energy of the incident electromagnetic wave.

Table 1. Absolute values of reflection coefficients $\frac{\Delta R}{R}$ at normal light incidence $\frac{\Delta\rho}{\rho} = 1\%$

E, eV	λ, μm	$n_{1(Al_2O_3)}$	$n_{2(Co)}$	$k_{2(Co)}$	$\frac{\Delta R}{R}$, %		
					$\varphi_1 = 0°$		
					$\frac{\Delta\rho}{\rho} = 1\%$	$\frac{\Delta\rho}{\rho} = 5\%$	$\frac{\Delta\rho}{\rho} = 10\%$
1.550	0.8	1.76	1.90	4.95	0.117	0.583	1.167
1.240	1	1.76	2.20	5.50	0.099	0.495	0.989
0.620	2	1.74	5.15	7.00	−0.069	−0.344	−0.687
0.413	3	1.71	4.90	8.45	0.0006	0.003	0.006
0.310	4	1.68	4.70	11.00	0.043	0.214	0.429
0.248	5	1.62	4.70	14.70	0.045	0.225	0.450
0.207	6	1.56	5.00	17.50	0.039	0.195	0.389
0.177	7	1.46	5.40	20.90	0.033	0.163	0.325
0.155	8	1.32	5.80	24.00	0.028	0.140	0.280
0.138	9	1.15	6.56	27.20	0.025	0.123	0.247
0.124	10	0.85	7.10	29.50	0.023	0.114	0.228

Table 2. Absolute values of reflection coefficients $\frac{\Delta R}{R}$ at light incidence at the angle of 70°

E, eV	λ, μm	$n_{1(Al_2O_3)}$	$n_{2(Co)}$	$k_{2(Co)}$	$\frac{\Delta R}{R}$, %		
					$\varphi_2 = 70°$		
					$\frac{\Delta\rho}{\rho} = 1\%$	$\frac{\Delta\rho}{\rho} = 5\%$	$\frac{\Delta\rho}{\rho} = 10\%$
1.550	0.8	1.76	1.90	4.95	−0.277	−1.383	−2.767
1.240	1	1.76	2.20	5.50	−0.281	−1.406	−2.811
0.620	2	1.74	5.15	7.00	−0.351	−1.756	−3.512
0.413	3	1.71	4.90	8.45	−0.300	−1.500	−3.001
0.310	4	1.68	4.70	11.00	−0.226	−1.131	−2.262
0.248	5	1.62	4.70	14.70	−0.154	−0.771	−1.541
0.207	6	1.56	5.00	17.50	−0.122	−0.612	−1.224
0.177	7	1.46	5.40	20.90	−0.095	−0.473	−0.947
0.155	8	1.32	5.80	24.00	−0.076	−0.379	−0.758
0.138	9	1.15	6.56	27.20	−0.064	−0.322	−0.645
0.124	10	0.85	7.10	29.50	−0.059	−0.294	−0.588

Table 3. Absolute values of transmittance coefficients $\frac{\Delta T}{T}$ at normal light incidence

E, eV	λ, μm	$n_{1(Al_2O_3)}$	$n_{2(Co)}$	$k_{2(Co)}$	$\frac{\Delta T}{T}$, %		
					$\varphi_1 = 0°$		
					$\frac{\Delta\rho}{\rho} = 1\%$	$\frac{\Delta\rho}{\rho} = 5\%$	$\frac{\Delta\rho}{\rho} = 10\%$
1.550	0.8	1.76	1.90	4.95	1.378	6.888	13.777
1.240	1	1.76	2.20	5.50	1.513	7.567	15.134
0.620	2	1.74	5.15	7.00	1.763	8.817	17.635
0.413	3	1.71	4.90	8.45	2.023	10.114	20.228
0.310	4	1.68	4.70	11.00	2.211	11.053	22.107
0.248	5	1.62	4.70	14.70	2.243	11.213	22.427
0.207	6	1.56	5.00	17.50	2.269	11.347	22.694
0.177	7	1.46	5.40	20.90	2.298	11.491	22.982
0.155	8	1.32	5.80	24.00	2.331	11.653	23.306
0.138	9	1.15	6.56	27.20	2.442	12.208	24.416
0.124	10	0.85	7.10	29.50	2.518	12.591	25.182

Table 4. Absolute values of transmittance coefficients $\frac{\Delta T}{T}$ at light incidence at the angle of 70°

E, eV	λ, μm	$n_{1(Al_2O_3)}$	$n_{2(Co)}$	$k_{2(Co)}$	$\frac{\Delta T}{T}$, %		
					$\varphi_2 = 70°$		
					$\frac{\Delta\rho}{\rho} = 1\%$	$\frac{\Delta\rho}{\rho} = 5\%$	$\frac{\Delta\rho}{\rho} = 10\%$
1.550	0.8	1.76	1.90	4.95	−0.026	−0.128	−0.255
1.240	1	1.76	2.20	5.50	−0.023	−0.114	−0.228
0.620	2	1.74	5.15	7.00	−0.013	−0.064	−0.129
0.413	3	1.71	4.90	8.45	−0.012	−0.058	−0.117
0.310	4	1.68	4.70	11.00	−0.009	−0.047	−0.093
0.248	5	1.62	4.70	14.70	−0.007	−0.033	−0.065
0.207	6	1.56	5.00	17.50	−0.005	−0.025	−0.050
0.177	7	1.46	5.40	20.90	−0.004	−0.019	−0.038
0.155	8	1.32	5.80	24.00	−0.003	−0.015	−0.030
0.138	9	1.15	6.56	27.20	−0.002	−0.012	−0.024
0.124	10	0.85	7.10	29.50	−0.002	−0.010	−0.021

Table 5. Spectral values of ε^{EMA} at the different values of λ calculated via the coefficients *n* and *k*

E, eV	λ, μm	$n_{1(Al_2O_3)}$	$n_{2(Co)}$	$k_{2(Co)}$	ε_1	ε_0	ε^{eff}
0.539	2.30	1.73	5.15	7.27	−26.330 − 74.881*i*	2.993	5.018 − 13.031*i*
0.729	1.70	1.74	4.6	6.70	−23.730 − 61.640*i*	3.028	4.666 − 11.622*i*
1.000	1.24	1.75	3.2	6.10	−26.970 − 39.040*i*	3.063	2.975 − 9.599*i*
1.253	0.99	1.76	2.94	5.50	−21.606 − 32.340*i*	3.098	3.033 − 8.558*i*
1.494	0.83	1.76	2.53	4.95	−18.102 − 25.047*i*	3.098	2.762 − 7.487*i*
1.797	0.69	1.76	2.31	4.45	−14.466 − 20.559*i*	3.098	2.712 − 6.634*i*
2.000	0.62	1.77	2.19	4.11	−12.096 − 18.002*i*	3.133	2.729 − 6.108*i*
2.296	0.54	1.77	2.05	3.81	−10.314 − 15.621*i*	3.133	2.661 − 5.606*i*
2.480	0.50	1.77	1.88	3.55	−9.068 − 13.348*i*	3.133	2.524 − 5.158*i*
2.696	0.46	1.78	1.78	3.30	−7.722 − 11.748*i*	3.168	2.498 − 4.782*i*
3.024	0.41	1.78	1.61	3.05	−6.710 − 9.821*i*	3.168	2.343 − 4.361*i*
3.263	0.38	1.79	1.53	2.82	−5.612 − 8.629*i*	3.204	2.325 − 4.025*i*

Table 6. Spectral values of ε^{EMA} and γ^{EMA} at the different values of λ with tabulated values of ε and γ

E, eV	λ, μm	*X* = 0.4			ε^{eff}	γ^{eff}
		ε_1	ε_0	γ		
0.539	2.30	2.993	−23.835 − 57.178*i*	1.529 − 3.008*i*	4.381 − 11.119*i*	0.179 + 0.017*i*
0.729	1.70	3.028	−18.091 − 44.063*i*	0.748 − 2.051*i*	4.213 − 9.450*i*	0.134 + 0.004*i*
1.000	1.24	3.063	−13.307 − 31.657*i*	0.203 − 1.241*i*	3.857 − 7.761*i*	0.098 + 0.008*i*
1.253	0.99	3.098	−11.358 − 24.914*i*	−0.009 − 0.895*i*	3.520 − 6.829*i*	0.083 − 0.007*i*
1.494	0.83	3.098	−9.474 − 17.882*i*	−0.135 − 0.530*i*	3.018 − 5.762*i*	0.060 − 0.017*i*
1.797	0.69	3.098	−8.295 − 14.346*i*	−0.160 − 0.380*i*	2.739 − 5.160*i*	0.051 − 0.024*i*
2.000	0.62	3.133	−7.613 − 12.339*i*	−0.162 − 0.299*i*	2.580 − 4.823*i*	0.045 − 0.028*i*
2.296	0.54	3.133	−6.507 − 9.779*i*	−0.144 − 0.199*i*	2.349 − 4.301*i*	0.033 − 0.028*i*
2.480	0.50	3.133	−5.945 − 8.626*i*	−0.130 − 0.159*i*	2.240 − 4.042*i*	0.029 − 0.029*i*
2.696	0.46	3.168	−5.217 − 7.378*i*	−0.111 − 0.118*i*	2.144 − 3.745*i*	0.023 − 0.029*i*
3.024	0.41	3.168	−4.407 − 6.281*i*	−0.088 − 0.084*i*	2.059 − 3.415*i*	0.018 − 0.026*i*
3.263	0.38	3.204	−3.504 − 5.468*i*	−0.066 − 0.059*i*	2.058 − 3.103*i*	0.013 − 0.023*i*

From analyzing the values in the tables, the calculated values of the effective DPT can be seen to coincide well with the literature data, such as those given in [4].

On the basis of the performed calculations, plots of the dependence of MRE on reflection and transmission at normal light incidence and angle of incidence of 70° were constructed (Figs. 1–4).

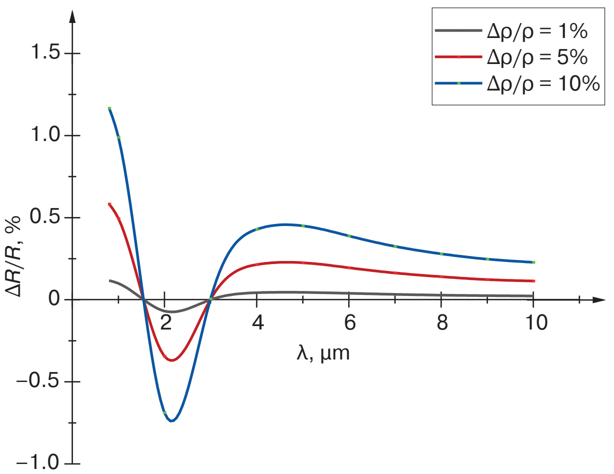

Fig. 1. Spectral dependence of MRE on reflection at normal light incidence

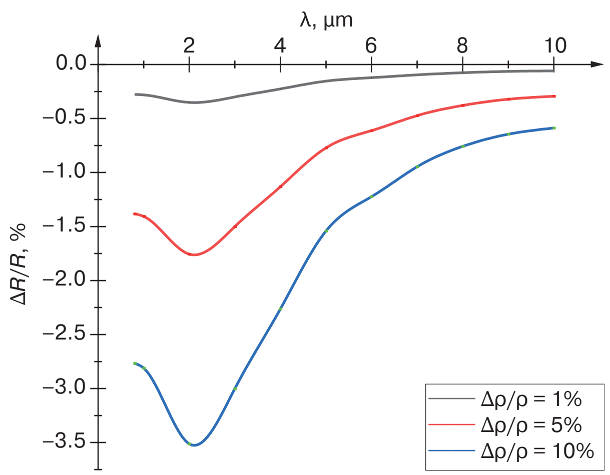

Fig. 2. Spectral dependence of MRE on reflection at $\varphi_2 = 70°$

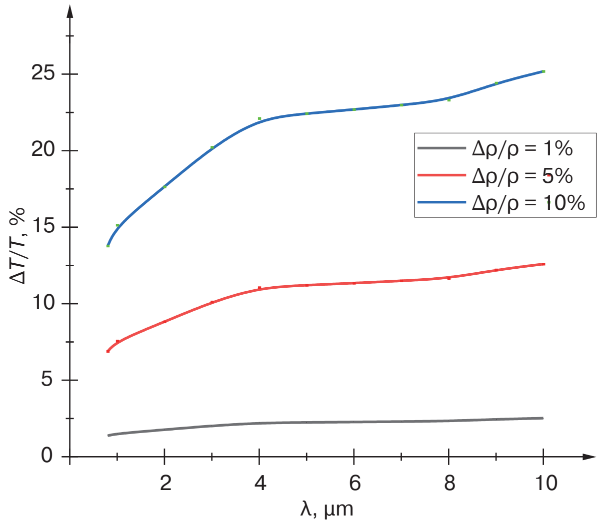

Fig. 3. Spectral dependence of MRE on transmittance at normal light incidence

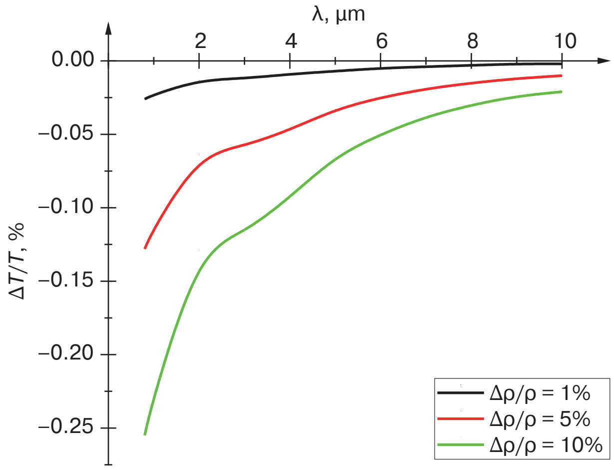

Fig. 4. Spectral dependence of MRE on transmittance at $\varphi_2 = 70^{\circ}$

When analyzing these graphs, we can draw the main conclusion: the spectral dependencies $\frac{\Delta R}{R}$ and $\frac{\Delta T}{T}$ in the IR range are linearly correlated with MR values $\frac{\Delta\rho}{\rho}$, and the dependence is directly proportional. The higher the MR value, the stronger the changes in the values $\frac{\Delta R}{R}$ and $\frac{\Delta T}{T}$.

It can be seen from Fig. 1 that in the near-IR region of the spectrum the MRE on reflection varies strongly, which is due to interzone transitions that significantly affect the optical characteristics of the material in the IR range. In Fig. 2, we can see a large absolute value of the parameter $\frac{\Delta R}{R}$. This can be explained by the significant role played in this region by intraband transitions, in addition to interband transitions, and the angle of incidence close to the Brewster angle (the main angle of incidence for metals) [3]. Figure 3 shows a smooth increase of the MRE on transmittance $\frac{\Delta T}{T}$, while Fig. 4 shows that as the wavelength increases (i.e., E decreases), the transmission MRE values $\frac{\Delta T}{T}$ first increase sharply and then change smoothly. By varying the wavelength λ and the real part of the refractive index n for a given film thickness d of the nanocomposite, it is possible to obtain the interference conditions under which the MRE value increases significantly [11–15].

CONCLUSIONS

The theoretical study of optical and MO spectra of Co-Al_2O_3 nanocomposites presented in this work was carried out in the framework of the MRE theory. The spectra were calculated using formulas (2)–(7). The spectra of the effective DPT were calculated in the Bruggeman approximation, which provides a good description of the nanocomposite properties at average volume concentrations of the metallic component ($X = 0.4$). As a non-contact MR measurement method for nondestructive control of any nanostructures, MRE represents a promising tool for the study of nanomaterials. The approaches described in this work are valid for the study of a wide class of nanostructures.

ACKNOWLEDGMENTS

The work was supported by the Ministry of Education and Science of the Russian Federation (State task for universities No. FGFZ-2023-0005).

Authors' contributions

M.A. Muhutdinova—literature review, computer simulation, discussion of results, and writing the text of the article.

A.N. Yurasov—computer simulation, discussion of results, writing and editing the article.

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Translated from Russian into English by Lyudmila O. Bychkova
Edited for English language and spelling by Thomas A. Beavitt