Influence of the size, shape and concentration of magnetic particles on the optical properties of nano nickel films**

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In general, the optical properties of nanodispersed structures are very different from those of the corresponding bulk materials and depend on the structural parameters: the occupancy q of the volume of the matrix with the dispersed nanoparticles, the size f and shape of the particles, the degree of order in the arrangement of the particles, the properties of the medium, and the presence of vicinal nanoparticles manifested in the dielectric constant ε_m. In the present paper, using discontinuous Ni films as examples, we consider theoretically and experimentally the influence of these structural parameters on the optical properties. The optical spectra strongly depend on composition and dielectric constants of particles and matrix; in their turn the dielectric constants are functions of the structural and electronic parameters and can differ from those of the corresponding bulk materials. Thus, optical spectra investigations can give very useful information about the structural parameters of ultrafine structures. In this work the optical properties of such structures are derived from the theoretical Maxwell Garnett model. The optical spectra of thin Ni films was explained within the framework of the effective medium approximation in two cases: $q < 0.5$ and $0.5 < q < 1$. In this approach an effective refractive index ($n + ik$) of the nanostructures can be calculated as a function of ε_m, q and particle shape. The results were in good agreement with experimental data.

Keywords: effective medium approximation, nanoparticles, optical spectra

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1. Introduction

Nowadays, it is difficult to name any field of technology where nanomaterials in some form are not used. The advance of technology requires materials with new properties. This is why intensive experimental and theoretical researches of magnetic substances are taking place worldwide with a view to creating cutting-edge materials; exploration of the mutual influence of microstructure and structural content on magnetic, magneto-optical and optical properties of nanoheterostructures has become the focus of intensive research. It is known that the qualities of already-explored materials change in the process of their transformation into nanocrystals. This was, however, predictable because these structures contain from some to thousands of atoms and occupy an intermediate position between atoms and massive substances; consequently they have properties different from both of them, acquiring a set of particular qualities such as giant magnetoresistance, giant magnetoimpedance, anomalous Hall effect, anomalous optical and magneto-optical effects [1–3] etc.

The optical spectra strongly depend on composition and dielectric constants of particles and matrix. In their turn the dielectric constants are functions of the structural and electronic parameters and can differ from those of the corresponding bulk materials [4–7]. Thus, optical spectra can give very useful information about the structural parameters of the ultrafine structures (meaning an ensemble of particles with sizes smaller than 100 nm).

In this paper we present theoretically and experimentally the influence of the structural parameters on the optical properties of nanodispersed nickel. We interpret the obtained optical spectra in the framework of the effective medium approximation [8].

2. Theory

The Maxwell Garnett theory was developed for optically isotropic materials having scalar dielectric constants [9]. The theory deals with a heterogeneous medium consisting of random cells of more than two kinds of materials, considering it as a homogeneous effective medium having an effective dielectric tensor in a specified wavelength region. In other words, using electromagnetic radiation of a much longer wavelength than the size of each component, we cannot identify individual components embedded in the medium. In the effective medium theory a heterogeneous structure or a composite is replaced by a suitable effective medium having an effective dielectric constant with a smooth surface parallel to an arbitrarily chosen plane.

We have modified the Maxwell Garnett effective medium theory to study a nanostructured medium in which optically anisotropic nanoparticles (ellipsoids) are dispersed in a matrix. After generalization of the Maxwell Garnett theory for such a nanodispersed structure composed of particles with different dielectric permittivities $\varepsilon_i (i = 1, 2, 3, ..., n)$ and a matrix with dielectric permittivity ε_m we arrived at a formula for calculating the diagonal elements of the dielectric tensor for nonspherical ultrafine particles:

$$\frac{\varepsilon_{ef} - \varepsilon_m}{\varepsilon_m + f(\varepsilon_{ef} - \varepsilon_m)} = \sum_i q_i \cdot \frac{\varepsilon_i - \varepsilon_m}{\varepsilon_m + f(\varepsilon_i - \varepsilon_m)} \quad (1)$$

where ε_{ef} is the effective dielectric permittivity of the nanodispersed structure, q is the ratio of the volume occupied by the particles to the total volume of the medium, and f is a shape factor of the ultrafine particles.

The effective dielectric permittivity $\varepsilon_{ef} = \varepsilon_{1ef} - i\varepsilon_{2ef}$ is connected to the refractive index n_{ef} and the absorption index k_{ef} of a nanodispersed medium by the formula

$$\varepsilon_{ef} = (n_{ef} + ik_{ef})^2 . \tag{2}$$

If the nanodispersed medium consists of ellipsoidal particles with dielectric permittivity $\varepsilon_i = \varepsilon$, eqn (1) can be expressed as

$$\frac{\varepsilon_{ef} - \varepsilon_m}{\varepsilon_m + f(\varepsilon_{ef} - \varepsilon_m)} = q\frac{\varepsilon - \varepsilon_m}{\varepsilon_m + f(\varepsilon - \varepsilon_m)}. \tag{3}$$

For spherical particles $f = 1/3$, hence from eqn (3) we can obtain the Maxwell Garnett formula

$$\frac{\varepsilon_{ef} - \varepsilon_m}{\varepsilon_{ef} + 2\varepsilon_m} = q\frac{\varepsilon - \varepsilon_m}{\varepsilon + 2\varepsilon_m}. \tag{4}$$

We presume that eqn (3) is valid for $q < 0.5$. If $q \geq 0.5$ the structure can be represented as a metal matrix with dielectric inclusions. In this case eqn (3) can be written as:

$$\frac{\varepsilon_{ef} - \varepsilon}{\varepsilon + f'(\varepsilon_{ef} - \varepsilon_m)} = (1-q)\frac{\varepsilon_m - \varepsilon}{\varepsilon + f'(\varepsilon_m - \varepsilon)} \tag{5}$$

where q is replaced by $(1 - q)$, ε is replaced by ε_m, and ε_m is replaced by ε; f' is the shape factor for the dielectric insertions.

3. Experimental

In this work we investigate the optical properties of discontinuous nickel films, the "weight thickness" d (or effective thickness; $d = m/\rho S$, where m is film mass, ρ is metal density and S is film area) of which falls within the range 48–300 Å. Discontinuous films were obtained by evaporation in a 10^{-5} Torr vacuum on glass substrates with a rate of 1–5 Å/s. The optical constants were determined using the Avery method [10], according to which it is sufficient to define the ratio of reflexion coefficients R_p/R_s of p and s polarized light reflected from a sample at two angles of incidence. Fig. 1 shows the block scheme of the measurement.

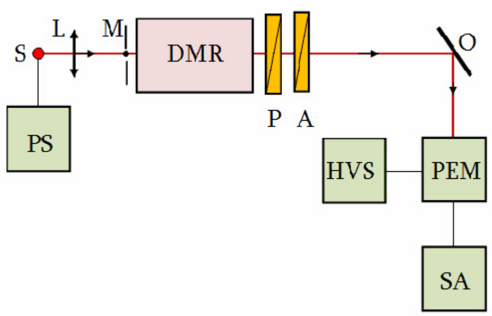

Figure 1. Schematic diagram of the method of measuring the optical constants. S, light source; L, collecting lens; PS, power supply; M, chopper; DMR, monochromator; P, polarizer; A, analyser; O, sample; PEM, photoreceiver; SA, selective amplifier; HVS, high-voltage rectifier.

The light source is placed at the focus of lens L. The light is modulated by the chopper. After going through the monochromator, polarizer and analyser it is reflected from the sample and its intensity measured. The sample is glued to a special holder able to rotate around the vertical axis thus changing the angle of the incident light. Light was polarized at an angle of 45°. The analyser separates the p and s polarized light, the intensities of which in the incident light will be equal, and subsequently the ratio of their coefficients of reflexion (R_p, R_s) will be equal to the ratio of the recorded photocurrents I_p, I_s from the reflected light, $R_p/R_s = I_p/I_s$.

4. Results and discussion

Fig. 2 shows the frequency dependences of the optical constants n and k for discontinuous nickel films with different thicknesses d, compared with those for bulk nickel. It can be seen that the character of the frequency dependences of the optical constants for the films is significantly different from that for bulk nickel and depends on d. We have applied our modified effective-medium theory (eqn 3) to explain these spectra: Fig. 3 gives the calculated dependences on $\hbar\omega$, of the effective optical constants for nanodispersed nickel having different q. Comparing these experimental and theoretical results, we observe that the experimental spectra for $d = 4.8$–30 nm are similar to the computed spectra for $q = 0.1$–0.85.

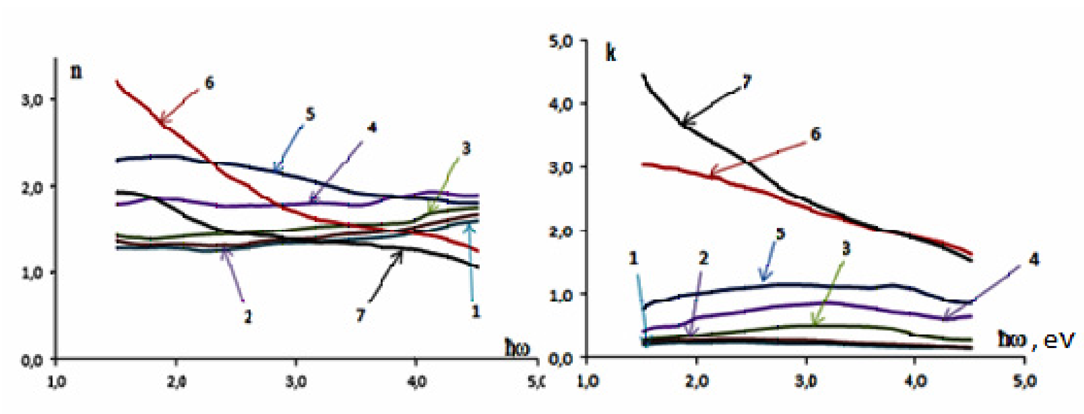

Figure 2. Experimental dependences of the optical constants n (left) and k (right) on the quantum energy of incident light $\hbar\omega$ for discontinuous Ni films with weight thicknesses $d = 3.2$ nm (curve 1), 4.8 nm (2), 6.4 nm (3), 9.0 nm (4), 14.4 nm (5), 30 nm (6) and bulk polycrystalline nickel (7).

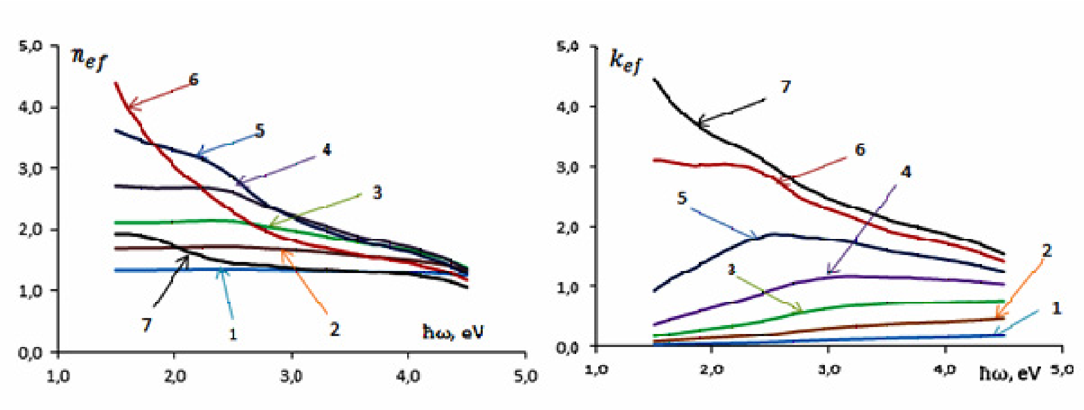

Figure 3. Theoretical dependences of the optical constants n_{ef} and k_{ef} on the quantum energy of incident light $\hbar\omega$ for ultrafine nickel, calculated by eqns (2) and (3) with $q = 0.1$ (curve 1), 0.25 (2), 0.4 (3), 0.55 (4), 0.7 (5), 0.85 (6) and 1.0 (7), with the refractive index of the matrix $n_m = 1.15$ and $f = 1/3$.

Fig. 4 gives the dependences of optical constants n_{ef} and k_{ef} on the quantum energy of incident light $\hbar\omega$, calculated by formulas (5) and (2) for nickel with different dielectric inclusions. The character of their spectral dependences is similar to that of polycrystalline bulk nickel. Hence we can conclude that optical properties of nanostructures significantly depend on the volume fraction q occupied by metal particles.

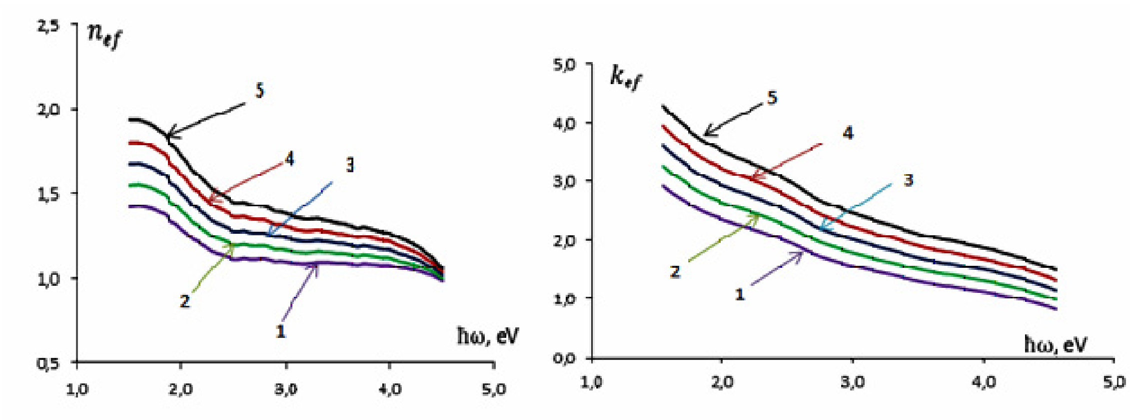

Figure 4. Dependences of n_{ef} and k_{ef} on the quantum energy of incident light $\hbar\omega$, calculated by eqns (5) and (2) for nickel with different dielectric inclusions: $(1 - q) = 1 - 0.6$ (curve 1), $1 - 0.7$ (2), $1 - 0.8$ (3), $1 - 0.9$ (4) and polycrystalline nickel (5) with $n = 1.15$ and $f = 1/3$.

We have also calculated the dependences of n_{ef} and k_{ef} on the quantum energy of incident light $\hbar\omega$, for nano-dispersive nickel with different n_m. In our calculation we have used eqn (3) ($q < 0.5$). The results of these calculations are shown in Fig. 5

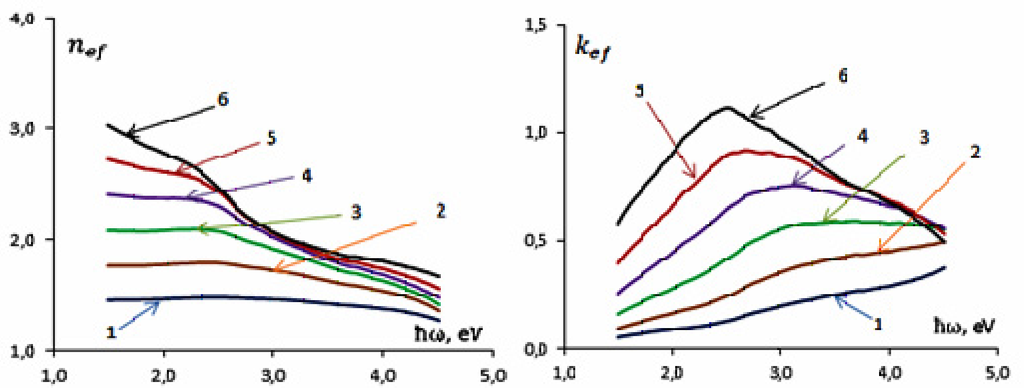

Figure 5. Dependences of n_{ef} and k_{ef} on the quantum energy of incident light $\hbar\omega$, for nano-dispersive nickel calculated by eqns (3) and (2) with different n_m, viz. 1.0 (curve 1), 1.2 (2), 1.4 (3), 1.6 (4), 1.8 (5), 2.0 (6) with $f = 1/3$ and $q = 0.25$.

5. Summary and conclusions

We have modified the Maxwell Garnett effective medium theory to study a nanodispersed medium with optically anisotropic particles (ellipsoids) dispersed in a matrix with dielectric contstant ε_m. To ascertain the suitability of the modified formula, we have considered theoretically and experimentally the influence of the structural parameters on the optical properties of nanodispersed nickel films. Their optical spectra were considered within the framework of the effective medium approximation in two cases: volume fraction $q < 0.5$ and $0.5 < q < 1$. The effective refractive index $(n + ik)$ of the nanodispersed structures has been calculated as a function of ε_m, q and particle shapes. The experimental data are well compatible with the above-discussed theory. It is evident from the results that the optical spectra of nanodispersed metals strongly depend on the volume fraction of the particles, their shape and on the dielectric constant of the matrix.

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