AN INFLUENCE OF A SIZE AND OF THE SIZE DISTRIBUTION OF SILVER NANOPARTICLES ON THEIR SURFACE PLASMON RESONANCE

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Abstract. On a basis of the comparative analysis of the references data the correlated dependencies between the optical characteristics of aqueous sols of spherical nanoparticles and their diameter have been discovered. As a result, the empirical dependencies between the values of the square of wave frequency in the adsorption maximum of the surface plasmon resonance and average diameter of the nanoparticles were determined as well as between the values of the adsorption band width on a half of its height and silver nanoparticles distribution per size. Proposed dependencies are described by the linear equations with the correlation coefficients 0,97 and 0,84 correspondingly.

Key words: nanotechnology, silver nanoparticles, aqua sol, surface plasmon resonance, UV/Vis spectra.

1. Introduction

The field of nanoscience has blossomed over the last twenty years and the need for nanotechnology will only increase, since the miniaturization becomes more important in such

areas as computing, sensors and biomedical applications. Advances in this field largely depend on the ability to synthesize nanoparticles of various materials, sizes and shapes as well as on efficiency assemble them into the complex architectures. The early well-known methods to produce suspensions of very small noble–metal particles are still used today and continue to be the standard by which other synthesis methods are compared. The most popular method to synthesize Au suspensions is the so-called Turkevich method, which employs the reduction of chloroauric acid with sodium citrate and produces a narrow size distribution of 10 nm particles [1]. For Ag nanoparticles suspensions a common method is the Lee–Meisel method, which is a variation of the Turkevich method in that $AgNO_3$ is used as the metal source [2], but unlike the Turkevich method, the Lee–Meisel method produces a broad distribution of particle sizes. The most common method for the synthesis of nanosized Ag particles is the reduction of $AgNO_3$ with $NaBH_4$. This method can also be adapted to produce particles of other metals such as Pt, Pd, Cu, Ni, ect. [3–6], although the specific protocols depend on the reduction potential of the source ion. Cu and Ni suspensions, for example, are not very stable since the metal particles are easily oxidized requiring strong capping ligands to prevent the oxidation. Silver nanoparticles (Ag–NPs) are characterized by unique combination of the important physical-chemical properties, namely by excellent optical characteristics, by ability to amplify the signal in spectroscopy of the combination dispersion [7], and also by high antibacterial properties. Among the three metals (Ag, Au, Cu) that display surface plasmon resonances (SPR) in the visible spectrum, exactly Ag exhibits the highest efficiency of the plasmon excitation, that leads to the abnormally high value of the extinction coefficient of Ag–NPs [8]. Moreover, optical excitation of the plasmon resonances in nanosized Ag–NPs is the most efficient mechanism by which light interacts with matter. A single Ag nanoparticle interacts with light more efficiently than a particle of the same dimension composed of any known organic or inorganic chromophore. Silver is also the only material whose plasmon resonance can be tuned to any wave-length in the visible spectrum. Under conditions of modern tendency to the miniaturization and the necessity to improve the

technological processes of the new materials obtaining based on Ag–NPs, there is problem of their identification, which requests the cost equipment and causes a search of the alternative ways of their average size and of their size distribution determination by others methods, in particular, by calculated ones with the use of the empirical equations and dependencies which are based on the property of adsorption of the electromagnetic irradiation in UV/visible diapason by sols of Ag–NPs [9].

We have used the optical properties of silver, namely the dependence of SPR adsorption maximum position on a size of Ag–NPs as the characteristic of their size and the width of the adsorption band on a half of its height as Ag–NPs size distribution.

2. Theoretical Grounds

On Figure 1 the SPR extinction spectra of Ag suspensions by different particle diameters are shown [10]. It is apparent that the dipole maximum rapidly shifts to longer wave–lengths as the particle size increases beyond 70 nm (450 nm spectral maximum) revealing the quadrupole peak at about 420 nm. The observed spectral shift results from the «spreading» of the particle's surface charge over a larger surface area so that the surrounding medium better compensates the restoring force thus slowing the electron oscillations [11].

A sufficiently small particle of any conducting material exhibits SPRs, yet its spectral position depends on many factors, most importantly on the material's frequency-dependent complex dielectric function. The wavelength dependence of the real ($\varepsilon_1(\omega)$) and imaginary ($\varepsilon_2(\omega)$) parts of the dielectric function describing polarizability and energy dissipation, respectively, are given on Figure 2 for Ag [12].

An SPR occurs when there is phase-matching between the polarization in the particle and incident field, a condition that is fulfilled for very small particles (<10 nm) when $\varepsilon_1(\omega) = -2\varepsilon_m$, where ε_m is the dielectric constant of the surrounding medium [13] and is satisfied for very small Ag particles suspended in water ($\varepsilon_m = 1,77$) at an excitation wavelength of around 385 nm. The imaginary part of the metal dielectric function, which describes losses, must be small

at the SPR frequency to provide efficient electron oscillations. Several processes can dump the oscillations, such as electron scattering by lattice phonon modes, inelastic electron-electron interactions, scattering of the electrons at the particle surface, and excitation of bound electrons into the conduction band (interband transitions) [14]. Whereas electron-phonon interactions account for a majority of $\varepsilon_2(\omega)$, inelastic electron–electron interactions and surface scattering are less significant, with the latter being important only for <5 nm particles. Interband transitions can cause a substantially decreased efficiency of plasmon excitation as is the case for Au and Cu, where there is significant overlap between the interband adsorption edge and the plasmon resonance. For Ag, however, the adsorption edge is in the UV (320 nm) and has little impact on the SPRs, which appear at wavelengths larger than 370 nm, accounting for the fact that excitation of the SPR in Ag particles is more efficient than for Au and Cu.

For future practical applications of nanoparticles, synthesis techniques capable of producing the highly crystalline particles of many different sizes and narrow distribution are necessary as well as the determination of their size and of their size distribution.
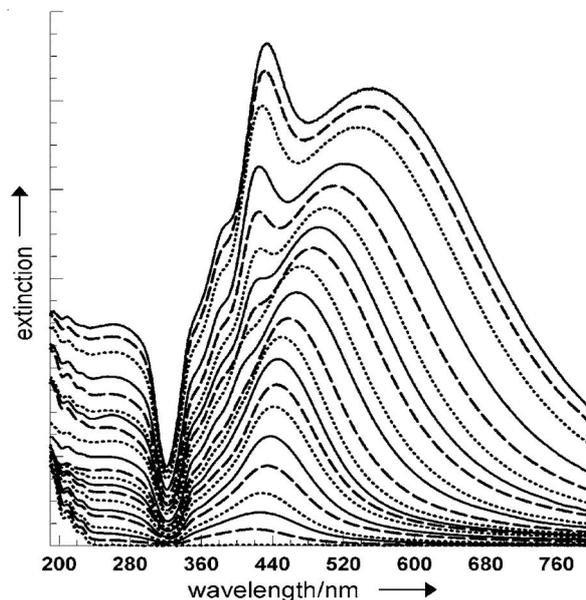
Fig. 1. UV/Vis extinction spectra of silver nanoparticles suspensions for 20 different particle diameters [10]
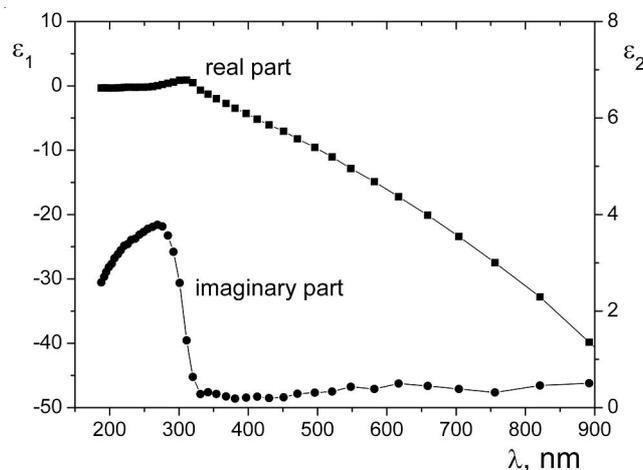
Fig. 2. Real ($\varepsilon_1(\omega)$) and imaginary ($\varepsilon_2(\omega)$) parts of the dielectric function of silver as a function of wavelength. Curves were prepared using values listed in [12]

3. Results & Disscussion

Generally, for the theoretical description of the SPR phenomenon of the metallic little particles and for the Ag–NPs, in particular, the solvings of the Maxell's equations are used, which in 1908 have been proposed by Gustav Mie [15]. Starting from the macroscopic Maxell equations, Gustav Mie calculated the extinction, scattering and absorption cross-sections of Au nanoparticles and showed how the spectra of the suspensions evolve as a function of particle size. The results of these calculations also allowed him to sketch scattering diagrams for different particle sizes and diagrams depicting the electric and magnetic fields of the dipole, quadrupole, octupole and sextupole components of the resonance. Now, it is well known that the optical resonances in noble-metal nanoparticles are the collective oscillations of conduction electrons termed "plasmons".

The extinction coefficient (C_{ext}) of the spherical nanoparticles in accordance with Mie's theory is described by the equation:

$$C_{ext} = \frac{24\pi^2 r \varepsilon_M^{3/2}}{\lambda} \frac{\varepsilon_2}{\left(\varepsilon_1 + 2\varepsilon_M\right)^2 + \varepsilon_2^2} \qquad (1)$$

where r is the radius of a particle, λ is a length of a wave of the electromagnetic irradiation, ε_M is the dielectric transmissivity of the solvent, ε_1 is a real part of the value of dielectric transmissivity of a part of the metal, ε_2 is the imaginary part of the value of dielectric transmissivity of a part of the metal.

It is known [9; 10], that the position of SPR maximum adsorption depends on a size of the Ag–NPs. Such phenomenon is explained by dependence of real and imaginary parts of the dielectric permeability of silver on size of the nanoparticle. In accordance with Drude's model [16], ε_1 and ε_2 can be described by the expressions:

$$\varepsilon_1 = \varepsilon'_{bulk} + \frac{\omega_p^2}{\omega^2 + \omega_d^2} - \frac{\omega_p^2}{\omega^2 + \omega_r^2} \qquad (2)$$

$$\varepsilon_2 = \varepsilon''_{bulk} + \frac{i\omega_p^2 \omega_r}{\omega\left(\omega^2 + \omega_r^2\right)} - \frac{i\omega_p^2 \omega_d}{\omega\left(\omega^2 + \omega_d^2\right)} \qquad (3)$$

$$\omega_r = \omega_d + \frac{v_F}{r} \qquad (4)$$

where ε''_{bulk} and ε'_{bulk} are values of the real and of the imagined parts of dielectric permeability of silver mass, ω, ω_p and ω_d are correspondingly the frequency of the electromagnetic irradiation, plasmon frequency of the metal and decrement of electron gas extinction in the mass metal, v_F is the Fermi rate.

However, calculated accordingly to such expressions adsorption spectra of aqueous sols of spherical Ag–NPs are differed from the experimental ones, that can be explained by different reasons, in particular: firstly, in presented example of the calculations it was not taken into account the distribution of Ag–NPs per sizes, that has an influence on a value of the SPR adsorption band width on a half of its height and, secondly, in classical Drude's model the adsorbed stabilizer on the surface doesn't take into account; in turn, such stabilizer can influence on the value of the wave length in adsorption maximum of the Ag–NPs sol.

In order to determine the dependencies between the optical characteristics and size of the nanoparticles we have done an analysis of the great data of references [10; 17–51] concerning to the synthesis and the investigations of Ag–NPs.

It was determined (see Fig. 3), that a square of the wave frequency in adsorption maximum of SPR (ω_2) linearly depends on a value of the average diameter (d) Ag–NPs. Such dependence is described by the expression:

$$\omega^2 = \left(6,14 \pm 0,05\right) \cdot 10^{29} - \left(2,45 \pm 0,08\right) \cdot 10^{27} \quad (5)$$

with the correlation coefficient 0,97.

At the same time, it was not discovered the direct dependence between the width of the adsorption band of Ag–NPs on a half of its height ($\Delta\lambda$) and nanoparticles distribution per size (Δd). Evidently, it is connected with the nonmonotonic change of the adsorption band of Ag–NPs at their size increasing [16]. However, the all analyzed data are satisfactory described by the linear equation:

$$log(d \cdot \Delta\lambda) = \left(0,2 \pm 0,1\right) + \left(0,89 \pm 0,06\right) \cdot log(\Delta d \cdot \lambda_{max}) \quad (6)$$

with the correlation coefficient 0,84 (see Fig. 4). Here λ_{max} is a value of the wave length in a maximum of the SPR.

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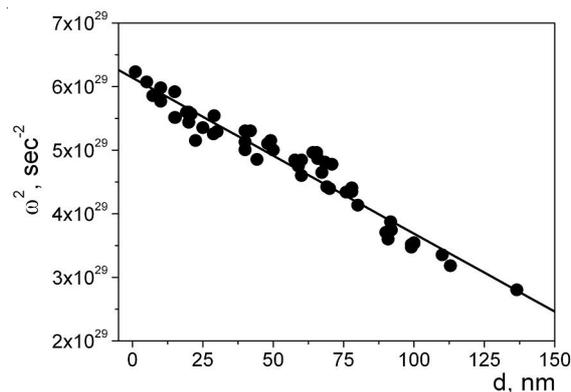
Fig. 3. Dependence between the square of the wave frequency in adsorption maximum of SPR and diameter of Ag–NPs
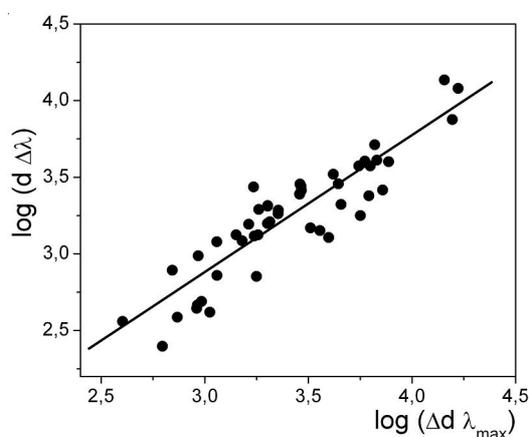
Fig. 4. Dependence between the logarithms of compositions $\log(d \cdot \Delta\lambda)$ and $\log(\Delta d \cdot \lambda_{max})$ for Ag–NPs

It is necessary to notify, that in processed data Ag–NPs were obtained in aqueous solution with the use of different upon nature stabilizers of the surface and precursors. However, in spite of this fact, discovered by us dependencies are good described with the respective correlation coefficients. It is clear, that for the explanation of nature for such dependencies the advanced theoretical analysis of the electron gas interaction with the electromagnetic irradiation is necessary, however, at the presented stage, such empirical dependencies can be used as the rapid method of the synthesized by different methods of Ag–NPs identification in laboratory and industrial conditions without the application of complicated, cost and often absent in Ukraine devices for their identification.

4. Conclusions

Empirical dependencies between the dimensional and optical characteristics of silver nanoparticles were determined. Such dependencies can be used for estimation of value of the average diameter and distribution per size of Ag–NPs without application of complicated equipment.

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ВЛИЯНИЕ РАЗМЕРА И РАСПРЕДЕЛЕНИЯ РАЗМЕРА СЕРЕБРЯНЫХ НАНОЧАСТИЦ НА ИХ ПОВЕРХНОСТНЫЙ РЕЗОНАНС ПЛАЗМЕННЫХ ВОЛН

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Аннотация. На основе сравнительного анализа справочных данных были обнаружены корреляционные зависимости между оптическими особенностями водных солей сферических наночастиц и их диаметра. В результате получены эмпирические зависимости между значениями квадрата частоты волны в адсорбционном максимуме поверхностного резонанса плазменных волн и средним диаметром наночастиц, а также между значениями адсорбционной ширины группы на половине из ее высоты и распределения размеров серебряных наночастиц. Предложенные зависимости описаны линейными уравнениями с коэффициентами корреляции 0,97 и 0,84 соответственно.

Ключевые слова: нанотехнология, серебряные наночастицы, водные соли, поверхностный резонанс плазменных волн, спектры UV/Vis.