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Method of Determining the Eigenfrequencies of an Ordered System of Nanoobjects

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Abstract—A method is proposed to determine the eigenfrequencies of nanostructures (nanotubes and nanocrystals) by measuring the eigenfrequencies of a "large system" that comprises an array of vertically oriented similar nanotubes or nanocrystals equidistantly grown on a substrate. It is shown that the eigenfrequencies of a single nanoobject can be derived from the eigenfrequency spectra of the large (array–substrate) system and of the substrate. In other words, using experimental data for large systems, one can determine the eigenfrequencies of single nanoobjects, which are difficult to determine otherwise. By way of example, the eigenfrequencies of an array of zinc oxide micro- or nanocrystals on a sapphire substrate are calculated.

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INTRODUCTION

Experimental determination of the mechanical characteristics of nanoobjects is today a challenging problem. One of the most efficient methods to determine elastic moduli in macromechanics is measurement of the eigenfrequencies of an object. However, attempts to apply this approach to nanoobjects sometimes face difficulties. In particular, optical methods used in such measurements usually fail [1].

The optical measuring procedure is as follows: an object under study is mounted on a chassis and illuminated by a laser beam, and another laser beam serves to detect the vibration amplitude at a certain point of the object. The optical signal is converted to an electric one, and the spectrogram thus obtained is analyzed with a spectrometer to determine the eigenfrequencies of the object. The main, but not the only, factor that limits the domain of applicability of this method is a finite (rather than indefinitely small) size of the laser beam: its spot is on the order of the laser wavelength across). If the object is smaller than the laser spot, the results make no sense.

Thus, optical methods fail in measuring the eigenfrequencies of a single nanoobject. However, it seems possible to measure the eigenfrequencies of a regular array of identical nanoobjects on a microsubstrate. Here, two problems lying at the interfaces between mechanics and experimental physics arise. The first one is determination of the elastic moduli of nanoobjects when there is a possibility to measure the frequencies of the microsubstrate–nanoarray system and determine the elastic characteristics of the substrate (for example, from the eigenfrequencies of the free substrate). The second problem is how to extract the eigenfrequencies of nanoobjects from the eigenfrequency spectrum of the substrate–array system. The success in solving both problems directly depends on the experimental conditions, specifically, on the way nanoobjects are mounted on the substrate; on how the substrate with nanoobjects is fixed to the chassis of the measuring device; and on the geometries, weights, and the elastic properties of the nanoobjects and the substrate. Thus, from the mechanical standpoint, one should not only interpret the measured data, but also elaborate upon the design of the experiment.

In this study, we suggest a method of determining the eigenfrequencies of nanostructures (nanotubes and nanocrystals) from the measured eigenfrequencies of a large system comprising a highly ordered array of identical nanotubes or nanocrystals grown on a substrate (Fig. 1) as a result, e.g., of self-organized growth [2, 3]. Nanoobjects constituting such an array are usually of the same size; therefore, one can use the macroscopic

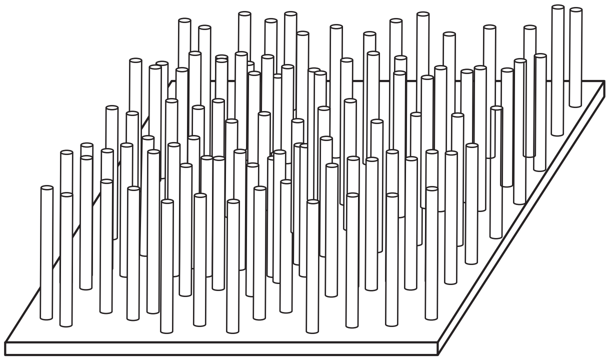

Fig. 1. Large system: an array of nanocrystals or nanotubes on a substrate.

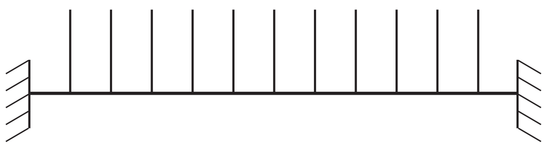

Fig. 2. Rod model.

sizes of the array to study the properties of nanoobjects by finding the first eigenfrequencies of the nanoobjects–substrate system. It is shown that, from the spectrum of the large (nanoobjects–substrate) system, the eigenfrequencies of an individual nanoobject can be derived; that is, the eigenfrequencies of a single nanoobject can be determined from experimental data obtained for the large system.

The main problem arising in measuring the frequencies of nanoobjects mounted on an elastic substrate is that the eigenfrequencies of the system as a whole are distributed among its components—a phenomenon well known in mechanics [4]. The shift of the spectrum strongly depends on the parameters of the substrate and object. It is known that, in many-body systems with distributed parameters, the vibration of one body may be dynamically damped at the partial frequency of another body (antiresonance). It will be shown below that systems of highly ordered arrays of identical nanotubes or nanocrystals grown on a substrate (Fig. 1) may also exhibit antiresonance and that this effect can be used to extract the eigenfrequencies of the nanoobjects from the spectrum of the large system. The normal modes of the system shown in Fig. 1 can hardly be analyzed in terms of the 3D elasticity theory. Therefore, we will first consider a rod model of the large system where a horizontal rod represents a substrate and vertical rods simulate nanoobjects (Fig. 2). In terms of this model, the normal modes of an array of nanocrystals are analyzed and the feasibility of extracting their eigenfrequency spectrum from the respective spectrum of the large system is demonstrated.

The array of nanocrystals assumed in this work is to a great extent similar to micro- and nanocrystals of semiconducting zinc oxide. Owing to their high mechanical and physical performance, such crystals are of considerable interest for nanomechanics and nanophotonics and can be fabricated by different techniques, pulsed laser evaporation among them [5–7]. Numerical analysis of the dynamics of such an array is the second stage of this study. The eigenfrequencies of an array of zinc oxide micro- or nanocrystals on a sapphire substrate are calculated in terms of the 2D problem of the elasticity theory. The calculation results also indicate the feasibility of extracting the spectrum of nanoobjects from the spectrum of the large system.

ANALYSIS OF THE MODEL PROBLEM

Let us apply the rod model to a large system consisting of a horizontal rod of length L depicting a substrate and N vertical rods of length H depicting nanoobjects (Fig. 2). The lower ends of the vertical rods are rigidly fixed to the horizontal rod and are spaced l apart, so that $L = (N + 1)l$. The upper ends of the vertical rods are free, the horizontal rod is rigidly fixed at both ends. In what follows, the problem of free vibrations is solved in two, discrete and continuum, statements.

Discrete Model

Assume that the horizontal rod comprises $N + 1$ small rods of length l rigidly attached to one another. The dynamics of such a system is described by the equations of the classical rod theory,

$$Cu_n^{IV} + \rho_1\ddot{u}_n = 0, \quad Dv_n^{IV} + \rho_2\ddot{v}_n = 0, \tag{1}$$

where u_n and v_n are the vertical and horizontal displacements of nth horizontal and vertical rods, respectively; C and D are the flexural rigidities of the horizontal and vertical rods, respectively; and ρ_1 and ρ_2 are the linear mass densities. The other values characterizing the state of stress of the rods are given by

$$\begin{aligned} \varphi_n &= u_n', \quad M_n = Cu_n'', \quad T_n = -Cu_n''', \\ \psi_n &= -v_n', \quad L_n = -Dv_n'', \quad N_n = -Dv_n'''. \end{aligned} \tag{2}$$

Here, φ_n and ψ_n are the rotation angles of the rod's cross sections, M_n and L_n are the bending moments, and T_n and N_n are the shear forces. The vertical motion of the vertical rods is described by the equations

$$w_n' = 0, \quad F_n' = \rho_2\ddot{w}_n, \tag{3}$$

where w_n are the vertical displacements of an nth vertical rod and F_n are longitudinal forces. The kinematic conditions at the joints between the rods are

$$\begin{aligned} u_n\big|_{x=nl} = u_{n+1}\big|_{x=nl}, \ v_n\big|_{y=0} = 0, \ w_n\big|_{y=0} = u_n\big|_{x=nl}, \\ \varphi_n\big|_{x=nl} = \varphi_{n+1}\big|_{x=nl}, \quad \psi_n\big|_{y=0} = \varphi_n\big|_{x=nl}. \end{aligned} \tag{4}$$

The force balance at the joints has the form

$$T_{n+1}|_{x=nl} - T_n|_{x=nl} + F_n|_{y=0} = 0,$$
$$M_{n+1}|_{x=nl} - M_n|_{x=nl} + L_n|_{y=0} = 0. \quad (5)$$

The boundary conditions for the system are given by

$$u_1|_{x=0} = 0, \quad \varphi_1|_{x=0} = 0,$$
$$u_{N=1}|_{x=L} = 0, \quad \varphi_{N=1}|_{x=L} = 0, \quad (6)$$
$$N_n|_{y=H} = 0, \quad F_n|_{y=H} = 0, \quad L_n|_{y=H} = 0.$$

The analysis of the problem revealed two groups of solutions. The first one corresponds to the situation when the vertical rods move as cantilever beams. In this case, the vibration eigenfrequencies of the system can be determined from the equation

$$1 + \cos(\mu H)\cosh(\mu H) = 0, \quad \mu = \sqrt[4]{\frac{\rho_2}{D}}\sqrt{\omega}. \quad (7)$$

The vibration amplitudes of the horizontal rods are small compared to those of the vertical rods. The ratio of these amplitudes is proportional to a small parameter,

$$\sqrt{\frac{\rho_2 D}{\rho_1 C}} \sim \left(\frac{h_2}{h_1}\right)^3, \quad (8)$$

where h_1 and h_2 are the characteristic transverse sizes of the horizontal and vertical rods, respectively, with $h_2/h_1 \ll 1$.

The second group of solutions meets the situation when the system vibrates with frequencies close to those in the system without the vertical rods. In this case, the equations from which the eigenfrequencies of the system and the form of horizontal rod vibrations are found contain two small parameters,

$$\sqrt{\frac{\rho_2 D}{\rho_1 C}}\frac{H}{L} \sim \left(\frac{h_2}{h_1}\right)^3\frac{H}{L}, \quad \frac{\rho_2 H}{\rho_1 L} \sim \left(\frac{h_2}{h_1}\right)^2\frac{H}{L}, \quad (9)$$

which distinguish the frequencies and forms of vibration of the complete system from those of the system without the vertical rods. The vibration amplitudes of the vertical rods are small compared to the vibration amplitudes of the horizontal rods and their ratio is proportional to a small parameter,

$$\sqrt[4]{\frac{\rho_1 D}{\rho_2 C}} \sim \left(\frac{h_2}{h_1}\right)^{1/2}. \quad (10)$$

Comparing λL and μH (here, $\lambda = \sqrt[4]{\frac{\rho_1}{C}}\sqrt{\omega}$), one can determine the mutual position of the spectra of the substrate and nanoobjects. The first eigenfrequencies of the substrate correspond to $\lambda L \sim 1$; the first eigenfrequencies of the nanoobjects, to $\mu H \sim 1$. If $\mu H/\lambda L \ll 1$, the first eigenfrequencies of the nanoobjects are considerably lower than those of the substrate; if $\mu H/\lambda L \gg 1$, the situation is reverse. If $\mu H/\lambda L \sim 1$, the first eigenfrequencies of the system include the eigenfrequencies of both the substrate and nanoobjects. The following estimate is valid:

$$\frac{\mu H}{\lambda L} \sim \left(\frac{h_2}{h_1}\right)^{1/2}\frac{H}{L}. \quad (11)$$

All the above asymptotic estimates are made on the assumption that Young's moduli and the volume mass densities of the nanoobjects and substrate are of the same order of magnitude.

Continuum Model

Let us now assume that the horizontal rod is a single whole with the vertical rods rigidly mounted on it. Then, the kinematic conditions at the joints appear as

$$v_n|_{y=0} = 0, \quad w_n|_{y=0} = u|_{x=nl}, \quad \psi_n|_{y=0} = \varphi|_{x=nl}. \quad (12)$$

Solving Eqs. (1) and (3) of motion for the vertical rods subject to boundary conditions (6) at their free ends yields a relationship between the forces and displacements at the lower points of the rods,

$$F_n|_{y=0} = -\rho_2 H\ddot{w}_n|_{y=0}, \quad L_n|_{y=0} = \frac{D\mu}{g(\mu H)}\psi_n|_{y=0}, \quad (13)$$

where parameter $g(\mu H)$ is given by

$$g(\mu H) = \frac{1 + \cos(\mu H)\cosh(\mu H)}{\sin(\mu H)\cosh(\mu H) - \cos(\mu H)\sinh(\mu H)}. \quad (14)$$

The equations of motion for the horizontal rod have the form

$$T' + \sum_{n=1}^{N} F_n|_{y=0}\delta(x - nl) = \rho_1\ddot{u},$$
$$M' + T + \sum_{n=1}^{N} L_n|_{y=0}\delta(x - nl) = 0. \quad (15)$$

Eliminating shear force T and using the relationship of elasticity $M = Cu''$, one can reduce Eqs. (15) to a single differential equation,

$$Cu^{IV} + \rho_1\ddot{u} = \sum_{n=1}^{N}[F_n|_{y=0}\delta(x - nl) - L_n|_{y=0}\delta'(x - nl)]. \quad (16)$$

With regard to expressions (12) and (13), Eq. (16) takes the form

$$Cu^{IV} + \rho_1\ddot{u} = -\sum_{n=1}^{N}\left[\rho_2 H\ddot{u}\delta(x - nl) + \frac{D\mu}{g(\mu H)}u'\delta'(x - nl)\right]. \quad (17)$$

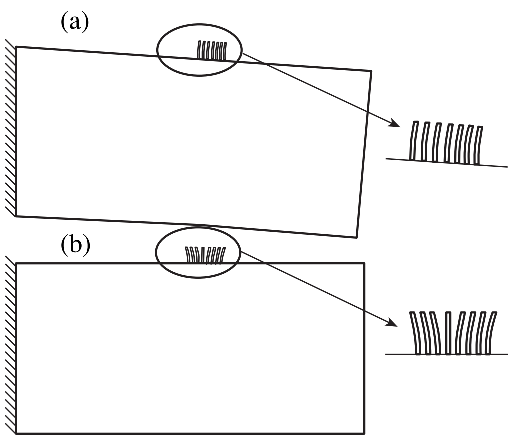

Fig. 3. Eigenmodes (a) due to the substrate and (b) localized in the nanocrystal array.

If the number of vertical rods is sufficiently large, they may be assumed to be continuously distributed over the length of the horizontal rod. Averaging the right-hand side of Eq. (17), we simplify the problem, reducing it to the equation

$$u^{IV} - \frac{ND\mu}{Cg(\mu H)L}u'' + \frac{\rho_1}{C}\left(1 + N\frac{\rho_2 H}{\rho_1 L}\right)\ddot{u} = 0. \quad (18)$$

The analysis of the continuum model showed that respective solutions can be divided into two groups, as in the case of the discrete model. The first type includes the vibrations at the frequencies defined by (7). In this case, according to (12), the condition $\psi_n|_{y=0} = 0$ is met; the vertical rods move as cantilever beams; and the vibration amplitudes of the horizontal rod are much smaller than those of the vertical rods. The second group of solutions embraces vibrations with frequencies close to the eigenfrequencies of the horizontal rod. Here, the vertical rods vibrate with amplitudes much smaller than those of the horizontal rod.

From the above considerations, it follows that, when the large system is treated in terms of the rod model, the eigenfrequencies of an individual nanoobject can be extracted from the spectrum of the whole system.

DYNAMICS OF THE NANOCRYSTAL ARRAY: NUMERICAL ANALYSIS

As an example of studying a real nanostructure, let us determine the eigenfrequencies of an array comprising ZnO micro- and nanocrystals. Owing to their high optical and mechanical properties, piezoelectric ZnO single crystals hold much promise for nanoelectronics and nanophotonics, in particular, for the production of UV lasers, chemical and biological sensors, solar cells, UV detectors, and other devices. Currently, ZnO single nanocrystals are obtained by thermal evaporation, chemical vapor deposition, pulsed laser deposition, etc. Nanocrystals thus grown are 1.5–3.0 μm high and 30–100 nm in diameter, and microcrystals are 20–100 μm high and 1–3 μm in diameter.

From the standpoint of continuum mechanics, an array of nanocrystals fixed on a substrate represents a composite piezoelectric body. In the electrostatic approximation and in the absence of mass forces, the basic equations of the electroelasticity theory have the form [8–10]

$$\rho\ddot{\mathbf{u}} = \nabla\boldsymbol{\sigma}, \quad \nabla\cdot\mathbf{D} = 0, \quad (19)$$

$$\boldsymbol{\sigma} = \mathbf{C}\boldsymbol{\varepsilon} - \mathbf{e}\mathbf{E}, \quad \mathbf{D} = \mathbf{e}\boldsymbol{\varepsilon} + \epsilon\mathbf{E}, \quad (20)$$

$$\boldsymbol{\varepsilon} = \frac{1}{2}(\nabla\mathbf{u} + \nabla\mathbf{u}^{\mathrm{T}}), \quad \mathbf{E} = \nabla\varphi. \quad (21)$$

Here, $\mathbf{u}$ is the displacement vector, $\mathbf{E}$ is the electric field strength expressed in terms of potential φ, $\boldsymbol{\sigma}$ is the stress tensor, $\mathbf{D}$ is the electric induction vector, $\boldsymbol{\varepsilon}$ is the strain tensor, ∇ is the gradient operator, ρ is the density, $\mathbf{C}$ is the rigidity matrix, and $\mathbf{e}$ and ϵ are the piezoelectric and dielectric constants.

Equations (19)–(21) should be complemented by boundary conditions. Let surface Γ of the body consist of two parts, $\Gamma = \Gamma_1 \cup \Gamma_2$, with $\Gamma_1 \cap \Gamma_2 = \varnothing$. Let displacements $\mathbf{u}_0$ are set on part Γ_1 and forces $\mathbf{f}$ on part Γ_2. In this case, the boundary conditions are given by

$$\mathbf{u}|_{\Gamma_1} = \mathbf{u}_0, \quad \mathbf{n}\boldsymbol{\sigma}|_{\Gamma_2} = \mathbf{f}. \quad (22)$$

For a piezoelectric system, along with mechanical boundary conditions (22), one should specify the electrical boundary conditions. Let $\Gamma = \Gamma_3 \cup \Gamma_4$ ($\Gamma_3 \cap \Gamma_4 = \varnothing$), with electric potential φ_0 and surface charge q set on Γ_3 and Γ_4, respectively. Then, we have

$$\varphi|_{\Gamma_3} = \varphi_0, \quad \mathbf{n}\mathbf{D}|_{\Gamma_4} = q. \quad (23)$$

For nonstationary processes, the boundary-value problem stated by (19)–(23) should also include initial conditions for the displacement field.

We will tackle a plane problem. Modal analysis of Eqs. (19)–(23) with appropriate boundary conditions for ZnO crystals on a sapphire substrate was performed with the use of the ACELAN finite-element package [11–13]. Consider the finite-element approximations of the ACELAN packet for field equations (19), relationships (20) and (21), and different types of boundary conditions (22) and (23).

The finite-element approximation of the problem of acoustoelectric elasticity leads to a set of ordinary differential equations [11],

$$\mathbf{M}\ddot{\mathbf{a}} + \mathbf{K}\mathbf{a} = \mathbf{F},$$

$$\mathbf{M} = \begin{pmatrix} \mathbf{M}_{uu} & 0 \\ 0 & 0 \end{pmatrix}, \quad \mathbf{K} = \begin{pmatrix} \mathbf{K}_{uu} & \mathbf{K}_{u\varphi} \\ \mathbf{K}^*_{u\varphi} & -\mathbf{K}_{\varphi\varphi} \end{pmatrix}, \quad (24)$$

$$\mathbf{F} = (\mathbf{F}_u, \mathbf{F}_\varphi)^T, \quad \mathbf{a} = (\mathbf{U}, \Phi)^{\mathrm{T}},$$

$$\mathbf{u}(x, t) = \mathbf{N}_u^{\mathrm{T}}\mathbf{U}(t), \quad \varphi(x, t) = N_\varphi^{\mathrm{T}}\Phi(t),$$

where $\mathbf{a}$ is the vector of nodal degrees of freedom, $\mathbf{M}$ and $\mathbf{K}$ are the mass and rigidity matrices, and $\mathbf{N}_u$ and N_φ are shape functions. Matrix $\mathbf{M}_{uu}$ represents the inertial properties of the medium, and matrices $\mathbf{K}_{uu}$, $\mathbf{K}_{u\varphi}$, and $\mathbf{K}_{\varphi\varphi}$ characterize its elastic, piezoelectric, and dielectric properties, respectively. Vectors $\mathbf{F}_u$ and $\mathbf{F}_\varphi$ take into account mechanical and electrical actions at the domain boundary.

After substitution of $\mathbf{a} = \mathbf{A}e^{i\omega t}$ into Eq. (24), the modal analysis of boundary problem (19)–(23) for the ZnO crystal array on the sapphire substrate in terms of the ACELAN package is reduced to a generalized equation in eigenvalues [11],

$$-\omega^2\mathbf{MA} + \mathbf{KA} = 0.$$

From a numerical experiment, we obtained the eigenfrequencies of a cantilever-mounted ZnO nanocrystal 1 μm in height (height-to-diameter ratio h/d = 10). The parameters of the ZnO single crystals and sapphire substrate were taken from [14]. The results of calculation are listed in Table 1.

In the model, the substrate was taken in the form of a rectangular sapphire microcrystal (10 × 20 μm) fixed on its large side. The results of the modal analysis of this crystal are presented in Table 2 (columns 2 and 4). The large system comprised the sapphire crystal with eight identical nanocrystals mounted on its free large side. The results of the modal analysis for such a large system are also listed in Table 2 (columns 1 and 3). Some of the eigenmodes are shown in Fig. 3. Figure 3a depicts the waveform of the first flexural mode. The kinematic analysis of the waveforms corresponding to the next eight eigenfrequencies indicates that the motion of the large system is localized in a nanocrystal-line “brush” (Fig. 3b) and the vibration waveforms of the nanocrystals meet the first eigenfrequency presented in Table 1. The vibration frequencies of the set of nanocrystals differ from the frequency of a single nanocrystal by less than 4%. Near the second eigenfrequency of an individual nanocrystal (Table 2), the large system behaves in a similar way, as illustrated in Fig. 4, where the plateaus of the curve correspond to the eigenfrequencies of a nanocrystal.

Thus, the eigenfrequency spectrum of the large system can be approximated by a combination of the eigenfrequencies of the substrate and those generated by one nanocrystal. This conclusion is consistent with analytical calculations in terms of the rod model.

Table 1

No.	Eigenfrequencies of individual nanocrystal, GHz	Modes
1	0.10797	First flexural mode
2	0.67763	Second flexural mode
3	1.92039	Third flexural mode
4	3.48340	First longitudinal mode (extension–compression)
5	3.83993	Fourth flexural mode

Table 2

Eigenfrequencies of large system, GHz	Eigenfrequencies of substrate, GHz	Eigenfrequencies of large system, GHz	Eigenfrequencies of substrate, GHz
0.036494	0.036594	0.611800	0.614194
0.103909		0.636879	
0.103971		0.650301	
0.104039		0.650491	
0.104106		0.650885	
0.104226		0.651087	
0.104322		0.651771	
0.104467		0.652115	
0.104612		0.653619	
0.134652	0.134973	0.673730	0.675596
0.136246	0.136017	0.693176	0.689642
0.280004	0.280137	0.714337	0.713020
0.350831	0.352308	0.715752	0.715594
0.399614	0.400228	0.752689	0.750820
0.458963	0.461709	0.796596	0.797104
0.468378	0.469073	0.825551	0.828275
0.510692	0.512042	0.833135	0.833899
0.533656	0.535745	0.895234	0.905771
0.554328	0.560235	0.927530	0.927179
0.581777	0.586277	0.938998	0.940679

DISCUSSION

The method proposed enables an accurate experimental determination of the first eigenfrequencies of a single nanoobject from the spectrum of the nanoarray–substrate system and the spectrum of the substrate alone. The method is especially efficient in the case when the first eigenfrequencies of nanoobjects are comparable to the first eigenfrequencies of the substrate (as they are in the example considered in this study). The main factor limiting the applicability of the method is the frequency range of measuring devices: the frequencies of nanoobjects may be too high to be detected.

Along with further refinement of this method, which determines the mechanical characteristics of nanocrystals by comparing the spectrum of the substrate with the spectrum of the nanoarray–substrate system, one may also take advantage of the fact that the eigenfrequencies of such a system are highly sensitive to its mechanical

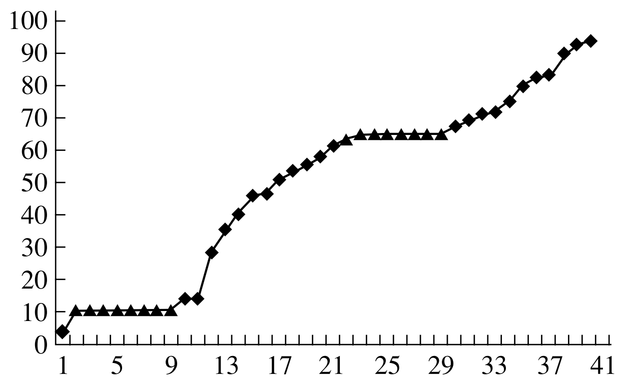

Fig. 4. Distribution of the eigenfrequencies of the large system: the eigenfrequencies of (▲) an individual nanocrystal and (◆) the substrate.

parameters and devise a nanosensor that will detect resonance changes in the nanoarray–substrate system, for example, a pollution nanosensor. It should be noted that, in this study, we disregarded the semiconducting properties of ZnO single crystals. It is known [15] that an electric field applied to a semiconductor generates an electric current, which may amplify or attenuate an acoustic signal due to the piezoelectric effect. An array consisting of many ZnO semiconducting nanocrystals can basically be used as an amplifier of weak acoustic signals that is similar to those considered in [15–17].

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