An analytically based computer model for evaluating effective elastic properties of nitride nanowire-polymer compositions using laser-generated surface acoustic waves

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Abstract. The investigation of physical and mechanical properties of nitride nanowires (NWs) is a topical field of modern nanotechnological research due to a wide range of promising applications. Normally, the NWs are embedded in a host polymer (e.g., hydrogen silsesquioxane – HSQ), and it becomes necessary to determine the effective properties of polymer-NW compositions rather than of separate wires. The central idea of the present research is the evaluation of the effective elastic moduli of the anisotropic composite HSQ-NW material via the minimization of the goal function that specifies the discrepancy between the measured and calculated characteristics of laser-generated surface acoustic waves. The experimental data are obtained on the basis of transient grating spectroscopy while the theoretical dispersion curves are calculated using the analytically based computer model for elastic guided wave excitation and propagation in multilayered elastic anisotropic half-spaces. The numerical examples illustrate the validation of the developed computer model against the experimental data acquired for sandwich microstructures with known elastic moduli as well as demonstrate the restoration of the latter in the case of HSQ-NW compositions with unknown mechanical properties.

1. Introduction
For many years the materials of the III-Nitrides group (GaN, AlN, InGaN, and others) have attracted the attention of researchers due to their exceptional properties that are very suitable for modern electronic and optoelectronic applications. From the early 2000s, the use of these materials in the form of nitride nanowires (NWs) has been intensively studied [1,2] due to their remarkable piezoelectric properties, which are very suitable for novel ultra-compact and efficient piezoelectric generators providing energy harvesting and autonomous operation of microelectronic devices. A review of research from single III-nitride nanowires to piezoelectric generators can be found in Ref. [3].

The electric output of a single piezoelectric NW depends on the law of its deformation in response to mechanical loading. Therefore, to maximize the output and optimize the design, a study of such laws is highly desirable. Meanwhile, the NWs are normally used in the form of arrays embedded in a host polymer (e.g., in hydrogen silsesquioxane – HSQ [4]), and the shape of their deformation is determined by the elastic properties of the polymer-NW composition
as a whole. This fact was the incentive for the undertaken research aimed at developing non-destructive methods for evaluating elastic moduli of anisotropic HSQ-NW composites via the measurement of laser-generated surface acoustic waves (SAWs).

The preparation of the samples, the measurement procedure, and the experimental results are described in separate paper [5] submitted to the METANANO 2018 in parallel with this article. Here we present the mathematical and computer model used to restore sample parameters based on the measured data. To simulate the SAWs, we have used the integral transform technique and the Green’s matrix approach yielding explicit integral and asymptotic representations for the wave fields generated in anisotropic multilayered structures by a given surface load [6,7]. Earlier, the evaluation of effective elastic properties has been performed for layered composite fiber-reinforced plastic plates using piezoelectrically induced guided waves and laser Doppler vibrometry [8].

2. Physical and mathematical framework

The experimental samples are sandwich compositions with an HSQ-NW interlayer of about micron thickness deposited on a silicon (Si[111]) substrate (half-space) and covered by a thin metallic layer which provides an electrical contact. The nanowires embedded into the HSQ polymer are vertically oriented across the interlayer, closing the circuit between the coating and the substrate. Thus, the effective elastic properties of the HSQ-NW composition can be simulated by a transversally isotropic medium with the horizontal plane of isotropy (x; y) and the substrate. Therefore, the effective elastic properties of the HSQ-NW composition can be simulated by a transversally isotropic medium with the horizontal plane of isotropy (x, y). Such materials are specified by five independent elastic moduli $C_{ij}$ and density $\rho$.

To simulate the frequency spectra of the laser-generated SAWs, the boundary value problem (BVP) is formulated with respect to the complex displacement amplitude $\mathbf{u} = (u_1, u_2, u_3)$ of the time-harmonic oscillation $\mathbf{u}(\mathbf{x})e^{-i\omega t}$; $\omega = 2\pi f$ is angular frequency; $f$ is frequency, $\mathbf{x} = (x, y, z)$ is a point in the Cartesian coordinate system; plane $(x, y)$ and axis $z$ are aligned with the sample’s surface and the outward normal.

The solution to the BVP is obtained in terms of the Green’s matrix $k(\mathbf{x})$ and the vector of load $\mathbf{q}$ applied to a surface area $\Omega$, or equivalently, via their Fourier symbols $K(\alpha_1, \alpha_2, z) = \mathcal{F}_{xy}[k]$ and $Q(\alpha_1, \alpha_2) = \mathcal{F}_{xy}[\mathbf{q}]$ [7-9]:

$$u(x) = \int_{\Omega} k(x-\xi, y-\eta, z)q(\xi, \eta)d\xi d\eta = \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} K(\alpha_1, \alpha_2, z)Q(\alpha_1, \alpha_2) e^{-i(\alpha_1 x + \alpha_2 y)} d\alpha_1 d\alpha_2.$$  

Here $\mathcal{F}_{xy}$ is the Fourier transform with respect to $x$ and $y$ variables; the Fourier parameters $\alpha_1$ and $\alpha_2$ play the role of wavenumbers for the waves propagating along the $(x, y)$ surface. The residues from the poles $\zeta_n$ of the matrix $K$ elements describe traveling SAWs generated by the source load $\mathbf{q}$ [6,7].

3. Validation

To verify the applicability of the theoretical model developed, first, the samples with known material parameters have been chosen for the theory-to-experiment comparisons. Figure 1 gives an example of such comparisons for the test sample Ni/HSQ/Si with a rather thin nickel coating (the thickness $h_1 = 0.015$ $\mu$m) and an HSQ interlayer without NWs of thickness $h_2 = 1$ $\mu$m. The material parameters for all examples are collected in Table 1. Here and below we use the experimental data obtained by the authors of adjoined contribution [5]. In all figures, the experimental results are shown by circle markers.

The coloured level-line plot in Fig. 1, left depicts a surface plot of $|K_{33}|$ as a function of the wavelength $\lambda = 2\pi/\alpha_1$ in the $x$ direction ($\alpha_2 = 0$) and frequency $f$. The element $K_{33}$ of the matrix $K$ describes the vertical displacement response of the multilayered substructure on a normal elementary loading (it is enough for laser generation and measurement). The areas of
$K_{33}$ maximal amplitudes correspond to the parameters of dominate SAWs. They yield peaks in the frequency spectra detected in experimental measurements for fixed wavelengths $\lambda$.

The ridges of $K_{33}$ amplitude go along the lines specified by its real and nearly real poles $\alpha_1 = \zeta_0(f)$ that determine the dispersion characteristics of the traveling SAWs and leaky guided waves. The right subplot of Fig. 1 shows the corresponding dispersion curves in the frequency-slowness plane. The slowness $s = \alpha_1/\omega = 1/(\lambda f)$ is convenient for the analysis since its magnitude varies in a limited range specified by the frequency-independent body-wave slownesses of layer materials shown in the figures by thin horizontal lines.

4. Preliminary results
In real applications, a platinum coating is more suitable. However, the transition from the test sample to a Pt/HSQ/Si specimens with ten times thicker coatings ($h_1 = 0.15 \mu m$) considerably changed both theoretical curves and experimental markers that cease to be in agreement (e.g., Fig. 2, left). Nevertheless, via the variation by the HSQ polymer thickness $h_2$, which also could not be precisely controlled in the course of sample fabrication, it was achieved a match with the same material parameters as in the previous example and unexpectedly small thickness $h_2 = 0.25 \mu m$ (Fig. 2, right).

The presence of nanowires complicates elastic properties of HSQ-NW composites. It turned out hard to select them manually based on seemingly realistic guesses, even accounting for the
Table 1. Elastic moduli $C_{ij}$ [GPa] and density $\rho$ [$10^3$ kg/m$^3$]

|     | $C_{11}$ | $C_{33}$ | $C_{12}$ | $C_{13}$ | $C_{44}$ | $C_{66}$ | $\rho$ |
|-----|----------|----------|----------|----------|----------|----------|-------|
| Ni  | 247      | 247      | 153      | 153      | 122      | 122      | 8.9   |
| Pt  | 315      | 315      | 193      | 193      | 61       | 61       | 21.5  |
| HSQ | 11.89    | 11.89    | 5.95     | 5.95     | 2.97     | 2.97     | 1.0   |
| HSQ-NW, guess | 7.37 | 27.39 | 3.58 | 3.61 | 1.90 | 1.9 |
| HSQ-NW, restored | 20.77 | 105.50 | 10.08 | 0. | 10.41 | 5.35 | 1.77 |
| Si[111] | 195 | 205 | 54.3 | 44.7 | 60.7 | 70.3 | 2.331 |

Figure 3. Nanowire specimen Pt/HSQ-NW/Si: guessed HSQ-NW parameters (left) and the adjusted ones (right).

known moduli of the NW material (GaN in the case, Fig. 3, left). To fit the curves to the experimental markers, we have minimized the goal function, which specifies their deviation, by varying the five independent elastic moduli $C_{ij}$, the density $\rho$, and the thickness $h_2$ of the HSQ-NW interlayer. Figure 3, right gives an example of such a fit reached at the set of HSQ-NW parameters shown in Table 1; the guessed and adjusted thicknesses are $h_2 = 1.1$ $\mu$m and $h_2 = 0.89$ $\mu$m, respectively.

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5. References
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