Study on frequency dependence of polarized piezoceramics constants in equivalent circuits at weak electric fields (part III)  

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Introduction. It is acknowledged that electroelastic modules do not depend on the amplitude and frequency of oscillations. This approach is reflected in the Russian and foreign standards for determining the complete set of electro-elastic piezoceramics modules. For example, to determine $d_{31}$ piezo-module of a disc-shaped sample, it is required to take measurements in three frequency domains: in the first and second resonances, in the antiresonance region, and at frequencies much below 1 kHz. Accordingly, it is assumed that when determining $d_{31}$, the modules of the ceramic under study in the frequency range from 1 kHz to the second resonance are independent of frequency. The work objective is to study the frequency dependence of electro-elastic ceramic modules. In this case, a disc-shaped sample from LZT (lead zirconate titanate) is used.

Materials and Methods. Techniques of setting and solving problems of the stationary electroelasticity and sections of the electrical engineering basics are applied. To implement the finite element method, the perturbation technique and the ANSYS application package are used. The experimental results are processed in the MATLAB environment.

Research Results. For the LZT piezoelectric ceramics, the frequency dependences of various modules (piezoelectric $d_{31}$, dielectric $\varepsilon_{33}$ and elastic modules of compliance $-s_{11},s_{12},s_{13}$) were investigated. Radial oscillations of a disc-shaped sample with electrodes on the ends were considered. The sample thickness was 1 mm, the diameter was 40 mm, and the oscillation range was up to 700 KHz. First, the frequency dependence was studied for the elastic ceramic modules from the determination of ten resonance frequencies. Then, the frequency dependence of $d_{31}$ and $\varepsilon_{33}$ modules was determined from the measured values of the sample conductivity. For this purpose, we used the expression for the electrical conductivity obtained from the solution of the radial oscillations of the disc considering its thickness.

Discussion. The work objective is to study the frequency dependence of electro-elastic ceramic modules. In this case, a disc-shaped sample from LZT (lead zirconate titanate) is used.

Conclusion. The study is performed within the frame of the independent R&D.

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** Работа выполнена в рамках инициативной НИР.

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Introduction. A significant number of papers study and develop mathematical methods for solving problems of piezoelectric body oscillations. Being a component of the piezoelectric devices, piezoelectric elements serve to excite and register oscillations caused by external effects. The selection of piezoelectric material for measuring transducers and the analysis of their characteristics requires a large amount of information on the parameters of materials. Such information includes:

- a complete set of electroelastic modules [1],
- losses,
- analogous electrical circuits, or equivalent circuits of piezoelectric elements [2].

The reactive dynamic parameters (L, C) of the equivalent circuits are determined by elastic dielectric and piezoelectric modules, as well as by the density of piezoceramics [3]. It is considered that the electroelastic modules - the equivalent circuit parameters are constant; they do not depend on the amplitude (weak electric fields) and on the oscillation frequency. All this is reflected in the Russian [4] and foreign [5, 6] standards for determining a complete set of electroelastic piezoelectric ceramics modules. For example, to determine $d_{31}$ piezo-module of a disk-shaped sample, it is necessary to carry out measurements in three frequency domains: in the first and second resonances, in the antiresonance region, and at frequencies much below 1 kHz. Accordingly, it is assumed that when determining $d_{31}$, the modules of the ceramic under study in the frequency range from 1 kHz to the second resonance are independent of frequency. In the specified range, $e_{33}^T$, $d_{31}$, $k_p$ constants have an insignificant frequency dependence for the considered radial oscillations.

Materials and Methods. Consider a piezoelectric disk with the thickness of 2$h$ and the radius $a$. We introduce a cylindrical coordinate system ($r, \Theta, z$) where $z$ axis coincides with the direction of the polarization axis. The coordinate plane $z = 0$ coincides with the midplane of the disk. Based on the known linear piezoelectric ratios, the equations of the continuum dynamics [1] and Maxwell equations [8], the system of equations for the axisymmetric oscillations of the piezoelectric disk can be written as follows:

\[ \begin{align*}
\partial_1 T_{rr} + \partial_3 T_{rz} + \frac{T_{rz} - T_{\Theta \Theta}}{r} + \rho \omega^2 U &= 0, \\
\partial_1 T_{rz} + \partial_3 T_{zz} + \frac{T_{rz}}{r} + \rho \omega^2 W &= 0, \\
\partial_1 D_r + \partial_3 D_z + \frac{Dr}{r} &= 0. 
\end{align*} \]  

(1)

Henceforward, the following notations and definitions are introduced: $U, W$ are mechanical displacements along $r, z$ coordinate axes, respectively; $\omega$ is circular frequency; $\rho$ is density; $T_{mn}$ are mechanical stresses; $D_r, D_z$ are components of the electric induction vector; $\partial_1$ and $\partial_3$ are operators of derivatives with respect to $r$ and $z$. 

Discussion and Conclusions. A technique is developed for determining the frequency dependence of LZT piezoelectric ceramic modules. The disc-shaped sample was studied in 15-650 KHz frequency range. It is shown that in the range up to 650 KHz, $s_{11}, s_{12}, s_{13}$ elastic modules with E superscript (it is omitted) or measured at dc field are practically independent of frequency. In the specified range, $e_{33}, d_{31}, k_p$ constants have an insignificant frequency dependence for the considered radial oscillations.

In case of axial polarization, the linear piezoelectric effect equations for weak electric fields in the cylindrical coordinates can be written as follows [1]:

\[
\begin{align*}
T_{rr} &= c_{11} \partial_1 U + c_{12} \frac{\partial U}{r} + c_{13} \partial_3 W + e_{31} \partial_3 \varphi, \\
T_{θθ} &= c_{12} \partial_1 U + c_{11} \frac{\partial U}{r} + c_{13} \partial_3 W + e_{31} \partial_3 \varphi, \\
T_{zz} &= c_{13} \partial_1 U + c_{13} \frac{\partial U}{r} + c_{33} \partial_3 W + e_{33} \partial_3 \varphi, \\
D_z &= e_{33}(\partial_1 U + \frac{\partial U}{r}) + e_{33} \partial_3 W - e_{33} \partial_3 \varphi; D_r &= e_{15}(\partial_3 U + \partial_1 W) - e_{15} \partial_1 \varphi.
\end{align*}
\]

(2)

The following notation is introduced in the relations (2) and further: \(c_{mn}\) are elastic constants in the matrix designation measured on samples with shorted electrodes or at the constant electric field \(E\) (\(E\) upper index of the ceramics constants is omitted here and further); \(e_{num}\) are piezoelectric constants; \(c_{mn}\) are clamped dielectric constants; \(\varphi\) is electric potential where \(E = -\text{grad} \varphi\) [2, 8].

Assume that the boundary conditions are set at the electrode ends and on the lateral surfaces of the disk [1, 2]:

\[
\begin{align*}
at z = \pm h \quad T_{zz} &= 0; \quad T_{rr} = 0; \quad \varphi = 2V, \\
at r = a \quad T_{rr} = 0; \quad T_{zz} = 0; \quad D_r = 0.
\end{align*}
\]

(3)

In (3), \(2V\) value is electric potential difference applied to the ends [2, 8]. Let us introduce dimensionless coordinates and quantities from the formulas:

\[
\xi = \frac{r}{a}; \quad \zeta = \frac{z}{h}; \quad \varepsilon = \frac{h}{a}; \quad \Omega = \omega h \sqrt{\frac{p}{c_{44}}}; \quad \Omega_a = \omega a \sqrt{\frac{p}{c_{44}}}; \quad c_{11} = c_{11} - \frac{c_{33}^2}{c_{33}}.
\]

The solution to the boundary value problem (1-3) consists of the sums of two solutions:
- homogeneous solution at zero boundary conditions at \(z = \pm h\);
- particular solution that satisfies only nonzero conditions at the ends (3).

The constructed system of homogeneous solutions will enable to further satisfy the boundary conditions (4) on the lateral surface (as a rule, using variational methods).

It is not difficult to construct the particular solution \(D_{r0} = \text{const}\) and \(D_{z0} \equiv 0\), which satisfies automatically the third equation from the system (1) and the boundary conditions at the ends (3). According to the first two equations of the system (1), the mechanical and electrical components of the particular solution are equal:

\[
\begin{align*}
T_{rr}^0 &= T_{θθ}^0 = A(e_{33} + \frac{e_{33}^2 c_{13}}{e_{33}}) \sin(\chi \zeta) + e_{31} K; \quad U^0 = 0; \quad T_{zz}^0 = 0; \\
φ^0 &= Kh \zeta + Ah \sin(\chi \zeta); \quad D_z^0 = -e_{33} K; \quad T_{zz}^0 = e_{33} K + e_{33}^p A \zeta \cos(\chi \zeta); \quad A = \frac{V}{\frac{h e_{33}^p \zeta \cos(\chi \zeta) - k^2 \sin(\chi \zeta)}}; \quad K = \frac{V}{h} \frac{1}{1 - k^2 \tan(\chi \zeta)}.
\end{align*}
\]

(5)

The following notation is introduced in (5):

\[
\chi = \Omega \sqrt{\frac{c_{44}}{c_{33}}}; \quad e_{33}^p = e_{33} + \frac{e_{33}^2 c_{13}}{c_{33}}; \quad k^2 = 1 - \frac{c_{33}^2}{c_{33}}.
\]

If the vector of external forces and the electric potential are zero on the end surfaces, then the construction of homogeneous solutions is associated with the definition of the dispersion equation roots [9]. For symmetric oscillations, the dispersion equation has the following form:

\[
a_n M_n \tan^{-1}(β_n) = 0, \quad (n = 1, 2, 3).
\]

(6)

The following notation is introduced in (6):

\[
\begin{align*}
a_n &= a^2 c_{13} k_{1n} + c_{33} k_{2n} + e_{33} k_{3n}; \quad b_n = k_{1n} β_n + k_{2n}; \\
M_1 &= b_2 k_{32} - b_3 k_{32}; \quad M_2 = b_3 k_{31} - b_3 k_{33}; \quad M_3 = b_1 k_{32} - b_2 k_{31}.
\end{align*}
\]

Here, \(k_{mn}\) are algebraic complements to the elements of the third determinant row of the system for symmetric oscillations (1); \(β_n\) is binary cubic root from [9], \(a\) is non-dimensional wave number of oscillations along \(r\) axis.

To find the roots (\(a\)) at the given values of \(Ω\), it is necessary to solve the dispersion equation (6) in combination with the binary cubic. A detailed analysis of the dispersion equation roots of symmetric lossless oscillations for the piezoelectric layer is given in [9], considering losses – in [10]. It is the sum of the particular and homogeneous solutions that will allow satisfying the boundary conditions both at the ends and on the lateral surfaces of the disk.

With an arbitrary ratio of disk sizes, the inverse problem of its forced oscillations (1–4) is very difficult to analyze and has a finite analytical solution only in some special cases (for example, oscillations of a thin disk or without considering its thickness when \(ε \ll 1\)). Therefore, when determining the modules of ceramics, it is more convenient to
solve the inverse boundary problem using approximate methods with allowance for thickness corrections for low-frequency radial oscillations of the disk. In this case, we can get an analytical solution in the form of finite formulas. In this paper, we seek the solution in the form of expansion with respect to $\varepsilon$ small parameter:

$$a^2 = \varepsilon^4 \omega_0^2 + \varepsilon^4 \Omega_1^2 + \varepsilon^6 \Omega_2^2 + \varepsilon^8 \Omega_3^2 + \varepsilon^{10} \Omega_4^2 + \ldots$$

Here, $\mathbf{U}_n$ is vector function with $\mathbf{U}(U, W, \varphi)$ components; $n$ is the order of the constructed approximate theory of symmetric disk oscillations considered in the paper; $\gamma, \eta$ are unknown constants which depend on the modules of piezoelectric ceramics, they are determined from (1, 2) and satisfy the homogeneous (zero) boundary conditions (3).

Omit the cumbersome manipulations. We confine ourselves to the terms with $\varepsilon^6$ to determine $a^2$ wave number, the terms with $\varepsilon^8$ for $U$ vector function in (7), and we give the final result of the considered boundary value problem with the boundary conditions (3) at the ends [11]:

$$\gamma = \frac{c_{13}^2}{3c_{33}^2}; \quad \nu = \frac{s_{12}}{s_{11}}; \quad k_p^2 = \frac{2d_{31}^2}{\varepsilon_33(s_{11} + s_{12})}; \quad \eta = \varepsilon (\frac{2}{15} + \frac{2cp_{11}}{5c_{33}} + \gamma) + \text{piezo;}
$$

$$\text{piezo} = \frac{\varepsilon^6}{45c_1^3c_{33}^3(1 - k_p^2)}[(\frac{d_{31}}{s_{11} + s_{12}} - c_{11}^2(2td_{31} + d_{33}))^2].$$

Here, $S_{mn}$ are flexibility modules with $E$ constant; $d_{31}$ is piezo-module; $k_p$ is planar coupling coefficient; $\nu$ is planar Poisson ratio; $\varepsilon$ is free dielectric constant of the disk.

Next, we introduce the following definitions and notation: $\alpha_0^2 = \varepsilon^2 \Omega_0^2$ is approximate zero-order wave number; $\alpha_2^2 = \varepsilon^2 \Omega_2^2 + \varepsilon^4 \Omega_4^2$ is approximate wave quadratic number; $\alpha_4^2 = \varepsilon^2 \Omega_4^2 + \varepsilon^4 \Omega_6^2 + \varepsilon^6 \Omega_8^2$ is approximate quartic wave number; $C$ is disk capacity.

Table 1 presents the results of the exact solution of the wave number ($\alpha$) of the PZT4 ceramics depending on the frequency from the dispersion equation (6) and an approximate calculation from (7) at $\varepsilon = 0.033$ for various approximations of $\alpha_m$.

**Table 1**

An example of calculating wave number ($\alpha$) at various frequencies for the piezoceramic disk under study

<table>
<thead>
<tr>
<th>$f$, kHz</th>
<th>$\Omega$</th>
<th>$\alpha$ from (6)</th>
<th>$\alpha_0$</th>
<th>$\alpha_2$</th>
<th>$\alpha_4$ piezo = 0</th>
<th>$\alpha_4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>0.085</td>
<td>0.045055</td>
<td>0.045049</td>
<td>0.045055</td>
<td>0.045055</td>
<td>0.045055</td>
</tr>
<tr>
<td>250</td>
<td>0.425</td>
<td>0.2262</td>
<td>0.2252</td>
<td>0.22603</td>
<td>0.22605</td>
<td>0.2261</td>
</tr>
<tr>
<td>500</td>
<td>0.85</td>
<td>0.4578</td>
<td>0.4505</td>
<td>0.4568</td>
<td>0.4574</td>
<td>0.4575</td>
</tr>
<tr>
<td>700</td>
<td>1.1903</td>
<td>0.6522</td>
<td>0.6307</td>
<td>0.6477</td>
<td>0.6514</td>
<td>0.6518</td>
</tr>
</tbody>
</table>

The results given in Table 1 show that for the disk 1 mm thick and with frequencies up to 700 KHz, the calculation of the wave numbers from the dispersion equation (6) and the approximate calculation for $\alpha_4$ almost coincide, and the piezo correction can be neglected. In this case, the decomposition (7) for $a^2$ depends only on the disk geometry, density, and moduli of flexibility $- s_{11}, s_{12}, s_{13}$.

Omitting the relatively cumbersome manipulations, we give the expression of $Y$ conductivity for the piezoelectric disk. It is obtained from the approximate solution (7) for various $\varepsilon^2$.

$Y000$ is zero approximation of $\varepsilon = 0$, or the known equation of radial oscillations of a zero-thickness disk:

$$x = \frac{\alpha_0}{\varepsilon}; \quad Y000 = \omega C \left(1 - k_p^2 + k_p^2(1 + \nu) \frac{J_1(x)}{xJ_0(x) - (1 - \nu)J_1(x)}\right).$$

$Y040$ is approximate quartic wave number:

$$x = \frac{\alpha_4}{\varepsilon}; \quad Y040 = \omega C \left(1 - k_p^2 + k_p^2(1 + \nu) \frac{J_1(x)}{xJ_0(x) - (1 - \nu)J_1(x)}\right).$$

$Y042$ is approximate quartic wave number, quadratic particular solution:

$$Y042 = \omega C \left(1 - k_p^2 + c_{44} \frac{\Omega^2 \varepsilon_{13}^2}{3\varepsilon_{33} c_{33}} + k_p^2(1 + \nu) \frac{J_1(x)}{xJ_0(x) - (1 - \nu)J_1(x)}\right).$$

$Y242$ is approximate quartic wave number, quadratic homogeneous and particular solution:

$$Y242 = \omega C \left(1 - k_p^2 + c_{44} \frac{\Omega^2 \varepsilon_{13}^2}{3\varepsilon_{33} c_{33}} + k_p^2(1 + \nu) \frac{J_1(x)}{z\gamma(1 - c_{44} \frac{\Omega^2 \varepsilon_{13} c_{13}}{3\varepsilon_{31} c_{33}})}\right);$$
\[ z_n = [xf_n(x) - (1 - \nu)f_1(x)] \left[ 1 + \varepsilon^2 a_1^2 \left( \frac{1}{3} + \varepsilon \right) \right] - \varepsilon^2 a_1^2 y x f_0(x); \] (9)

Table 2 shows the numerical analytic calculation results. The conductivities of the exact solution (5, 6) and approximate solutions (8, 9) with respect to \( \varepsilon^2 \) were found using the MATLAB program [12]. For the disk made of PZT4 [13] with the thickness of 1 mm and the diameter of 30 mm, numerical calculations were carried out in the ANSYS and ACELAN systems [14]. The latter software package was developed by the employees of the Southern Federal University (SFU) and focused on the calculations of piezoelectric devices.

<table>
<thead>
<tr>
<th>Conductance calculation for various frequencies</th>
</tr>
</thead>
<tbody>
<tr>
<td>Frequency, kHz</td>
</tr>
<tr>
<td>----------------</td>
</tr>
<tr>
<td>350</td>
</tr>
<tr>
<td>554.4</td>
</tr>
<tr>
<td>700</td>
</tr>
</tbody>
</table>

Table 2 shows that in the construction of approximate theories of the type (8, 9), it is required to use the decomposition (7) to calculate the wave number at least of the second order.

**Research Results**

1. **Study of the dependence of the elastic ceramic moduli on frequency.** For a sufficiently well-studied PZT piezoceramics, we investigate the frequency dependence of the following modules: piezoelectric \( d_{31} \), dielectric \( e_{33}^T \) and flexibility modules - \( s_{11}, s_{12}, s_{13} \). Consider low-frequency radial oscillations of a disc-shaped sample with electrodes on the ends. The sample thickness is 1 mm, the diameter is 40 mm, and the oscillation range is up to 700 KHz.

First, we study the frequency dependence for elastic modules. To do this, according to [15], we precheck the first three resonant low frequencies \( f_r \) (the major resonance and its two overtones). Elastic constants are determined from the solution of the frequency equation (9) of the radial oscillations of the disk considering \( \varepsilon \) relative thickness: three equations for three unknown variables. It is \( \varepsilon \) relative thickness that distinguishes the above frequency equation (9) for radial oscillations of the finite thickness disk from the known frequency equation for radial oscillations of the disk with “zero thickness” [15]:

\[ R J_d(R) = (1 - \nu) J_1(R). \]

The introduction of \( \varepsilon \) thickness correction into the solution of the known equation of radial oscillations of a disc-shaped sample improves the accuracy and measurement informativeness of the elastic modules.

The elastic compliances determined by [11, 15] through the technique of three resonances for the considered ceramics turned out to be equal:

\[ s_{11} = 12.29e - 12, s_{12} = -4.05e - 12, s_{13} = -5.28e - 12. \]

Table 3 shows the first ten resonant frequencies for the PZT19 ceramics. In the “Experiment” stock, the frequencies measured on the \( \Psi K \) 6500B conductivity measuring device at the Institute of High Technologies and Piezotechnics at the Southern Federal University are given. In the “Analysis” line there are \( s_{11}, s_{12}, s_{13} \), calculated by the formula (9) for the frequency-independent elastic modules defined by [11, 15]. Measurement errors did not exceed the values recommended by the standard [4].

<table>
<thead>
<tr>
<th>First ten resonant frequencies for PZT19 ceramics</th>
</tr>
</thead>
<tbody>
<tr>
<td>Resonant frequencies ( f_r ), KHz</td>
</tr>
<tr>
<td>Experiment</td>
</tr>
<tr>
<td>Analysis</td>
</tr>
</tbody>
</table>

Table 3 shows that in the frequency range up to 600 KHz, the difference between the calculated and experimental data with constant elastic moduli does not exceed 1%, therefore the following conclusion can be made: “For the ceramics considered, \( s_{11}, s_{12}, s_{13} \) elastic modules with \( \Psi \) superscript (it is omitted) or measured at a constant electric field are practically independent of the frequency in the range up to 650 KHz.”

2. **Study of the dependence on the frequency of \( d_{31} \) and \( e_{33}^T \) modules.** We use the expression (8) for \( \Psi 040 \) conductance from the solution of the radial disk oscillations considering its thickness, as well as the values of imaginary parts of the complex conductivity of the piezoceramics under study measured at room temperature. In this case, it is possible to investigate the dependence of \( d_{31} \) and \( e_{33}^T \) modules on the frequency in the range from 10 KHz to 600 KHz.

To determine two unknown \( d_{31} \) and \( e_{33}^T \), measure \( Y \) at two frequencies \( f_1, f_2 \). The difference between \( d_{31} \) and \( e_{33}^T \) is selected so that we can neglect the dependence of \( d_{31} \) and \( e_{33}^T \) modules on the frequency in \( f_1 - f_2 \) range.
In this paper, two conditions are introduced:
1) \( f_2 - f_1 = 200 \) Hz;
2) the conductivities are calculated for an ideal piezoelectric body or without considering losses, therefore the frequencies of resonances or in the neighborhood of resonances are not taken into account.

As a result, we obtain an easily solvable system of two linear equations with respect to two unknown неизвестных \( k_p^2 \) and \( \varepsilon_{33}^T \).

Fig. 1 shows \( k_p^2 \), \( d_{31} \) and \( \varepsilon_{33}^T/\varepsilon_0 \) dependences on 10-650 kHz frequencies.

Here, \( \varepsilon_0 \) is the dielectric constant of the vacuum. In the range from 15 to 650 KHz, \( k_p^2 \) coupling coefficient first increases with frequency growth from 0.28 to 0.34, and then decreases to 0.26. \( d_{31} \) piezo-module has a similar frequency dependence: at 15 KHz, from \(-1.50 \cdot 10^{-10}\) to maximum of \(1.661 \cdot 10^{-10}\), and then decreases to \(1.31 \cdot 10^{-10}\). The relative dielectric constant decreases monotonically with increasing frequency from 1766 to 1455.

Fig. 2 shows frequency dependences in the range of 1–10 KHz measured and calculated from the formula (9) of the ceramics capacity with the constant modules defined according to OST [4, 15].

There is sharp difference between two PZT ceramics capacities for low frequencies. This probably explains the dependence of the ceramics parameters on the frequency below 15 KHz (see Fig. 1). This implies a higher degree of dispersion — first of all, of \( d_{31} \) piezo-module d31. This issue will be considered in greater detail in the next paper (\( \varepsilon_{33}^T, d_{31}, k_p \) constants are investigated in the low-frequency range).

Discussion and Conclusions. A technique has been developed for determining frequency dependence of the PZT piezoelectric ceramic modules. The disk-shaped sample was studied within the frequency range of 15–700 KHz. It is shown that in the range up to 650 KHz, \( s_{11}, s_{12}, s_{13} \) elastic modules with \( E \) superscript (it is omitted) or measured at
a constant electric field are practically independent of frequency. In the indicated range, \( \varepsilon_{33}^T, d_{31}, k_p \) constants for the radial oscillations considered have an insignificant frequency dependence.

Only the frequency dependence of real part of the ceramic modules was studied, the losses were not considered. Therefore, the experimentally obtained frequency dependences of the imaginary part of the ceramics conductivities were measured far from resonances, where the effect of losses was absolutely null. Even so, in the future, both the real and imaginary parts of the modules must be considered. This means that losses are included. This problem is supposed to be studied considering the frequency dependence of the complex modules of the ceramics under study.

In the present paper, the radial oscillations of the sample are considered to obtain more complete information when measuring a larger set of constants (except for the piezoelectric and dielectric modules, \( s_{11}, s_{12}, s_{13} \) elastic modules are measured). This technique can be extended to other forms of samples (rods, plates, etc.), since in these one-dimensional problems, there is a simple replacement of Bessel functions by trigonometric functions.

References


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