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Two-layered disc quasi-optical dielectric resonators: electrodynamics and application perspectives for complex permittivity measurements of lossy liquids

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Abstract
Electromagnetic properties of novel quasi-optical resonators are studied theoretically and experimentally. The resonators are a radially two-layered dielectric disc sandwiched between conducting endplates. The internal layer can be filled with air or lossy liquid. Whispering gallery modes are excited in such a resonator and the mode energy is concentrated near the inner side of the cylindrical surface of an external layer. The measurement data obtained in the K_a-band are compared with theoretical calculations of eigenfrequencies and quality factors of the Teflon resonator filled with water, ethyl alcohol, benzene and aqueous solutions of ethyl alcohol. A number of 'anomalous' properties of the resonator can be described using Maxwell equations. The experimental data on the complex permittivity of a binary mixture water–ethyl alcohol are compared with the values calculated in terms of Debye's function. An important feature of the proposed technique is that it holds promise for making first principle microwave measurements of the permittivity of lossy liquids.

Keywords: complex permittivity measurements, lossy liquids, quasi-optical dielectric resonators

1. Introduction

Over the past few decades much attention has been paid to studies of the electrodynamic properties of dielectric resonators excited on whispering gallery modes (WGM) (see, for example, [1–3]). WGM resonators are figures of revolution, most often in the form of a cylindrical disc or a sphere [4]. Recently other forms of resonators, such as hemi-disc [5], hemisphere [6], two-hemi-disc [7] and conical ones [8] were studied as well.

Interest in these resonators is explained by the fact that, above all, they allow one to achieve high quality factor values of a resonator with acceptable dimensions in the wavelength range from microwaves up to optical waves (see, for example, [9]). That is important for various applications including filters [10], high stability oscillators [11] and techniques for measuring both complex permittivity of dielectrics [12, 13] and surface impedance of superconductors [14]. In both of the latter cases, a small value of electromagnetic field energy loss occurs in dielectric and conductor (accordingly, the values of loss tangent $\tan\delta = \varepsilon'/\varepsilon''$ or surface resistance R_S are small). In particular, the small values of the losses are really difficult to determine.

Besides, there are dielectric media, which are of great importance for applications and are characterized by a high value of the imaginary part ε'' of complex permittivity $\varepsilon = \varepsilon' - i\varepsilon''$, when $\tan\delta \approx 1$. These main substances are exemplified by water, various spirits, aqueous solutions of spirits, blood, milk and other biological liquids. Because of the high value of tan δ, both eigen electrodynamic characteristics, namely the frequency f and quality factor Q, of resonators containing such a liquid depend on both parts, i.e. ε' and ε'', of the complex permittivity of a liquid. In this case a procedure for calibrating a measuring cell is a difficult matter. Therefore it is desirable to be able to determine ε' and ε'' directly for liquids using the results from f and Q measurements. One can avoid a calibration procedure when an electrodynamic problem on eigenvalues of the resonator has a rigorous solution. At the same time it is worth emphasizing that direct methods of measuring the physical properties of substances are always essential with any values of losses.

Recently a new approach to ε measurement of liquids has been proposed. It allows one to carry out direct measurements of properties of liquids with a high loss in the millimetre wavelength range [15, 16]. The technique is based on the use of dielectric resonators in the form of a layered cylindrical disc excited on WGM. Here a dielectric disc is sandwiched between conducting endplates (CEP) and the inner layer is a liquid under test [15]. Such resonator geometry allows both rigorous solution to an electrodynamic problem using the Maxwell equations and corresponding boundary conditions and is convenient for practical realization in the form of a measuring cell [16]. The dielectric resonators in which WGM are excited are, in fact, quasi-optical devices. Therefore it is natural to call them quasi-optical dielectric resonators (QDR).

The first results of the experimental study of water-filled QDR have shown the distinguishing properties of such a resonator as compared with the QDR containing dielectrics with small or moderate level of microwave losses (see, for example, [17, 18]). Similar features were found in lower-mode cavity and dielectric resonators with lossy substances (for example, see [19–21]). These resonators operated in the classic microwave range, i.e., at frequencies lower than 24 GHz. However, a two-layered QDR as a relatively novel type of measuring cell, which can operate in the millimetre wave range, has not been characterized systematically.

The present work is the first more complete paper, which aims at the theoretical and experimental study of electrodynamic properties of radially two-layered CEP-containing QDRs with lossy liquid and to formulate the prospects of the technique development for the complex permittivity measurement of the above-mentioned liquids by an example of characterization of water–ethyl alcohol solution.

2. Theoretical electrodynamics of radially two-layered CEP-containing QDR

Here the spectral characteristics of resonance oscillations of anisotropic radially two-layered QDR, in which the ends ($z = 0; l$, l is the height of resonator) were limited by ideally conductive planes, are studied (figure 1) by analogy with a study of a single-layered dielectric disc [22].

The QDR layers are made of various uniaxial single crystals, the anisotropic axes of which are parallel to the resonator's longitudinal axis. In this case, the permittivity

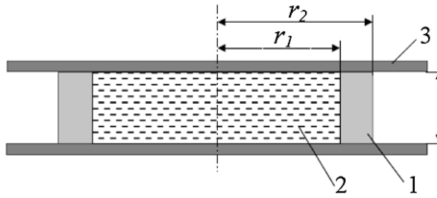

Figure 1. Two-layered quasi-optical dielectric resonator with conducting endplates and liquid: (1) Teflon disc (ring), (2) liquid and (3) conducting endplates.

and permeability tensors of the resonator are written as

$$[\varepsilon_{ij}] = \begin{cases} \begin{pmatrix} \varepsilon_{\perp\nu} & 0 & 0 \\ 0 & \varepsilon_{\perp\nu} & 0 \\ 0 & 0 & \varepsilon_{\parallel\nu} \end{pmatrix}, & \nu = 1, \quad r \leqslant r_1 \\ & \nu = 2, \quad r_1 < r \leqslant r_2, \\ \delta_{ij}, & \nu = 3, \quad r > r_2 \end{cases} \tag{1}$$

$$\mu_\nu = \begin{cases} \mu_1, & \nu = 1, \quad r \leqslant r_1 \\ \mu_2, & \nu = 2, \quad r_1 < r \leqslant r_2, \\ 1, & \nu = 3, \quad r > r_2 \end{cases}$$

where $\varepsilon_{\parallel\nu}$ and $\varepsilon_{\perp\nu}$ are the components of $[\varepsilon_{ij}]$ in parallel and perpendicular directions to the optical axis of a crystal for the ν-layer, r_1 and r_2 are the radii of resonator layers and δ_{ij} is the Kronecker delta.

The modes in such a resonator are described by solutions of a set of Maxwell equations that satisfy the following conditions: (i) the tangential components of electromagnetic field strengths on the medium boundaries $r = r_1$ and $r = r_2$ are continuous; (ii) the tangential components of electrical fields are equal to zero on the resonator ideal CEP; (iii) the electromagnetic fields are finite at $r = 0$; (iv) a damped wave takes a place at $r \to \infty$.

The axial components of electromagnetic fields in radially two-layered QDR have the forms

$$\begin{aligned} E_{z\nu} &= G_{E\nu}(r)\cos(k_z z)\,\mathrm{e}^{\mathrm{i}(n\varphi-\omega t)}; \\ H_{z\nu} &= G_{H\nu}(r)\sin(k_z z)\,\mathrm{e}^{\mathrm{i}(n\varphi-\omega t)}, \end{aligned} \tag{2}$$

where

$$G_{j\nu}(r) = \begin{cases} A_{jn}J_n(q_{j1}r), & \nu = 1, \; r \leqslant r_1 \\ B_{jn}J_n(q_{j2}r) + C_{jn}N_n(q_{j2}r), & \nu = 2, \; r_1 \leqslant r \leqslant r_2 \\ D_{jn}H_n^{(1)}(q_0 r), & \nu = 3, \; r \geqslant r_2 \end{cases}$$

characterizes the distribution of the field for the ν-resonator layer along a radius. Here A_{jn}, B_{jn}, C_{jn}, D_{jn} are the constants determined by the above-mentioned boundary and excitation conditions of electromagnetic modes in QDR; index j accepts a value E or H; $J_n(z)$, $N_n(z)$, $H_n^{(1)}(z)$ are the Bessel, Neumann and first-kind Hankel cylindrical functions; $n = 0, 1, 2, \ldots$ is the azimuth wave number; $\omega = \omega' - \mathrm{i}\omega''$ is the complex cyclical frequency. The radial ($q_{j\nu}$ is for the inside dielectric layers and q_0 is for outside the dielectrics) and axial k_z components of the wave number for the QDR fields have such forms

$$\begin{aligned} q_{H\nu}^2 &= \mu_\nu \varepsilon_{\perp\nu} k_0^2 - k_z^2, & q_{E\nu}^2 &= \frac{\varepsilon_{\parallel\nu}}{\varepsilon_{\perp\nu}} q_{H\nu}^2, \\ q_0^2 &= k_0^2 - k_z^2, & k_z &= \frac{m\pi}{l}, \end{aligned} \tag{3}$$

where $k_0 = \omega/c$, with c being the light velocity, and $m = 0, 1, 2, \ldots$ is the axial index.

The transversal components of the electromagnetic field are expressed in terms of $E_{z\nu}$ and $H_{z\nu}$:

$$q^2 E_{\varphi\nu} = \frac{1}{r}\frac{\partial}{\partial\varphi}\frac{\partial}{\partial z}E_{z\nu} - \mathrm{i}\mu_\nu\omega\frac{\partial}{\partial r}H_{z\nu},$$
$$q^2 E_{r\nu} = \frac{\partial}{\partial r}\frac{\partial}{\partial z}E_{z\nu} + \mathrm{i}\mu_\nu\frac{\omega}{r}\frac{\partial}{\partial\varphi}H_{z\nu}, \qquad (4)$$

$$q^2 H_{\varphi\nu} = \mathrm{i}\varepsilon_{\perp\nu}\omega\frac{\partial}{\partial r}E_{z\nu} + \frac{1}{r}\frac{\partial}{\partial\varphi}\frac{\partial}{\partial z}H_{z\nu},$$
$$q^2 H_{r\nu} = \frac{\partial}{\partial r}\frac{\partial}{\partial z}H_{z\nu} - \mathrm{i}\varepsilon_{\perp\nu}\frac{\omega}{r}\frac{\partial}{\partial\varphi}E_{z\nu}, \qquad (5)$$

where $q^2 = q_{H\nu}^2$ at $\nu = 1, 2$ and $q^2 = q_0^2$ at $\nu = 3$.

The spectral characteristics of anisotropic radially two-layered resonators are determined by the solution of the dispersion equation:

$$\begin{aligned}
&\chi_0\chi\left(\gamma_\beta^E - \gamma_\alpha^E\right)\left(\vartheta_\alpha^H - \vartheta_\beta^H\right)Z_J^H Z_N^H \\
&\quad + \left(\chi_0^2 - \gamma_\beta^E\gamma_\beta^H\right)\left(\vartheta_\alpha^E\vartheta_\alpha^H - \chi^2\right)Z_J^E Z_J^H \\
&\quad + \left(\chi_0^2 - \gamma_\alpha^E\gamma_\alpha^H\right)\left(\vartheta_\beta^E\vartheta_\beta^H - \chi^2\right)Z_N^E Z_N^H \\
&\quad + \chi_0\chi\left(\vartheta_\beta^E - \vartheta_\alpha^E\right)\left(\gamma_\alpha^H - \gamma_\beta^H\right)Z_J^E Z_N^E \\
&\quad + \left(\chi_0^2 - \gamma_\alpha^E\gamma_\beta^H\right)\left(\chi^2 - \vartheta_\beta^E\vartheta_\alpha^H\right)Z_N^E Z_J^H \\
&\quad + \left(\chi_0^2 - \gamma_\beta^E\gamma_\alpha^H\right)\left(\chi^2 - \vartheta_\alpha^E\vartheta_\beta^H\right)Z_J^E Z_N^H = 0. \qquad (6)
\end{aligned}$$

In this equation the notation is $\chi_0 = nk_z\left(q_0^{-2} - q_{H2}^{-2}\right)/k_0 r_2^2$, $\chi = nk_z\left(q_{H1}^{-2} - q_{H2}^{-2}\right)/k_0 r_1^2$; $\vartheta_\xi^j = \sigma_2^j\xi_{21}^j - \sigma_1^j\alpha_{11}^j$; $\gamma_\xi^j = \sigma_2^j\xi_{22}^j - \alpha_0$, where ξ denotes α or β; $\sigma_\nu^j = \varepsilon_{z\nu}$ at $j = E$ and $\sigma_\nu^j = \mu_\nu$ at $j = H$; $\alpha_{p\nu}^j = \frac{1}{q_{jp}r_\nu}\frac{J_n'(q_{jp}r_\nu)}{J_n(q_{jp}r_\nu)}$, where $p = 1, 2$; $\alpha_0 = \frac{1}{q_0 r_2}\frac{H_n^{(1)'}(q_0 r_2)}{H_n^{(1)}(q_0 r_2)}$; $\beta_{2\nu}^j = \frac{1}{q_{j2}r_\nu}\frac{N_n'(q_{j2}r_\nu)}{N_n(q_{j2}r_\nu)}$, the prime denotes a derivative with respect to the argument; $Z_R^j = \frac{R_n(q_{j2}r_1)}{R_n(q_{j2}r_2)}$, where R is J or N.

There are independent EH and HE modes in the resonator outside the frequency degeneration area. In the case where the following condition

$$\begin{aligned}
&\left\{\left|\chi^2\left(Z_J^E - Z_N^E\right)\left(\gamma_\beta^H Z_J^H - \gamma_\alpha^H Z_N^H\right) + \left(\vartheta_\alpha^E Z_J^E - \vartheta_\beta^E Z_N^E\right)\right.\right. \\
&\quad \times \left.\left.\left(\gamma_\alpha^H\vartheta_\beta^H Z_N^H - \gamma_\beta^H\vartheta_\alpha^H Z_J^H\right)\right|\right\} \Big/ \left\{\left|\chi_0\left[\left(\vartheta_\alpha^H Z_J^H - \vartheta_\beta^H Z_N^H\right)\right.\right.\right. \\
&\quad \times \left.\left(\vartheta_\beta^E Z_N^E - \vartheta_\alpha^E Z_J^E\right) + \chi^2\left(Z_J^H - Z_N^H\right)\left(Z_J^E - Z_N^E\right)\right] \\
&\quad + \left.\left.\chi\left(\vartheta_\alpha^E - \vartheta_\beta^E\right)\left(\gamma_\alpha^H - \gamma_\beta^H\right)Z_J^E Z_N^E\right|\right\}^{-1} \gg 1 \qquad (7)
\end{aligned}$$

is satisfied, HE modes take a place and, otherwise, EH modes occur.

The total energy losses of the eigen (resonant) electromagnetic mode in the resonator are determined by

$$\frac{1}{Q} = \frac{1}{Q_c} + \frac{2\omega''}{\omega'}. \qquad (8)$$

The first term in (8) determines the energy loss in the CEPs. The second one refers to both loss in a dielectric of the resonator and radiation loss [23]. Theoretical calculations show that the radiation loss is negligible for the given resonator. The QDR eigen frequency $\omega = \omega' - \mathrm{i}\omega''$ is found by solving the characteristic equation (6). The resonator quality factor attributed to losses in conducting endplates is determined by $Q_c = 1/A_S R_S$, whereR_S is the surface resistance of the CEP and A_S is the coefficient which can be calculated on the basis of eigenmode electromagnetic field distribution near the CEPs.

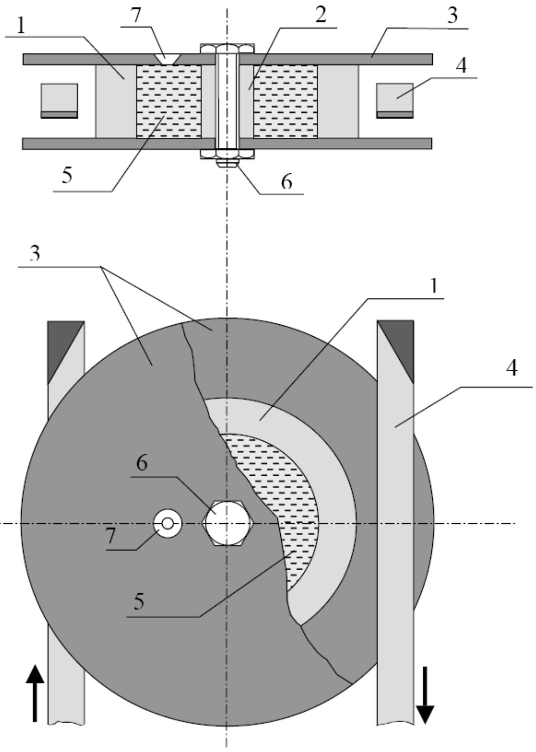

Figure 2. The measuring cell: (1) external Teflon ring, (2) internal Teflon ring, (3) conducting endplates, (4) dielectric waveguides, (5) layer of liquid under test, (6) metal screw and (7) a hole for liquid filling of the resonator with liquid under test.

3. Experimental details

3.1. Measurement technique

The experimental studies were carried out with a QDR, where the external layer was made of Teflon, in the $\mathrm{K_a}$-band ($f = 35$–38 GHz) at room temperature. Teflon was chosen as the first step in the development of the measurement technique because it is easily machined and has acceptable electric properties for applications in microwave and millimetre wave ranges. Its permittivity at $f = 37.5$ GHz is $\varepsilon_2 = 2.07 - \mathrm{i}3.5 \times 10^{-4}$.

The dimensions of the resonator (figure 2) were chosen upon examining its frequency spectrum and quality factor. In the present work an external diameter of $2r_2 = 78$ mm was constant and the internal $2r_1$ changed by the mechanical removal of an internal part of the dielectric disc.

Sufficiently weak distributed coupling of the resonator with image waveguides, also made of Teflon, was controlled so that the measured values of f and Q were eigen ones with a fairly high accuracy. An optimal orientation of waveguides relative to the dielectric disc was chosen for the excitation of axially-homogeneous HE_{n10} modes because only these modes were excited most effectively in CEP-containing QDR with the resonator height $l \approx \lambda$, being convenient for practical purposes (where $\lambda = c/f$). In the present work $l = 7.10$ and 14 mm.

CEPs were made in the form of duraluminium plates of 5 mm thickness. A 100 mm diameter of the plates was chosen to exclude microwave energy diffraction losses due to the plate edges. The whole resonator composed of a Teflon ring and two duraluminium plates was clamped by means of a metal screw. The disposition of a screw in the central part of the resonator did not perturb the electromagnetic field of the WGM because the latter has a surface nature and propagates near the dielectric–air interface in the Teflon ring.

The resonance frequency f and quality factor Q were measured for the resonator without a liquid, i.e. with an air-filled internal layer (f_0, Q_0), and then with a liquid in the internal layer ($f_{\rm liq}$, $Q_{\rm liq}$). Because resonant frequencies depended, to a certain extent, on reassembling of the resonator, the results of frequency measurements were presented as dependences of $\Delta f_{\rm liq} = f_{\rm liq} - f_0$ on thickness $r_2 - r_1$. In contrast, the quality factor was practically independent of reassembling (other things being equal); therefore the values of $Q_{\rm liq}$ were used for the analysis of measurement results.

After finding $\Delta f_{\rm liq}$ and $Q_{\rm liq}$ depending on $r_2 - r_1$ for QDR filled with water (bidistilled), ethyl alcohol and benzene, measurements of dependences $\Delta f_{\rm liq}$ and $Q_{\rm liq}$ on concentration of ethyl alcohol in binary water–ethyl alcohol solution were carried out with a fixed thickness $r_2 - r_1$. These measurements were performed without reassembling the resonator at each given radial thickness $r_2 - r_1$ of the external layer with the purpose of eliminating inaccuracy of measurements.

The measurement error of frequency shift was $<3 \times 10^{-4}\%$. The quality factor was measured using a width of resonance line with an error $<3\%$. A klystron oscillator (when $Q > 10^3$) and a backward tube (when $Q < 10^3$) were used as millimetre wave energy sources. All measurements with liquids were carried out at a temperature of 20 ± 0.3 °C. Variations in permittivity of the dielectric disc and the liquid under test in such a temperature interval were smaller than the accuracy of permittivity measurements using the Teflon resonator.

3.2. Properties of liquid-filled QDR depending on the radial thickness of the dielectric disc

The measurement results of $\Delta f_{\rm liq}$ and $Q_{\rm liq}$ for the resonator filled with bidistilled water, ethyl alcohol and benzene are presented in figures 3 and 4. At the same time the calculation results obtained by solving dispersion equation (1) are shown. Exclusion is a dashed line in figure 4 obtained by averaging the experimental values of the Q-factor of the water-filled resonator. The solid line in figure 4 is the result of $\Delta f_{\rm liq}$ calculation for the water-filled resonator using complex permittivity values [24] assuming that CEPs are perfectly conducting (section 2). We have shown that the conductivity of the CEP does not noticeably affect the value of $\Delta f_{\rm liq}$. Assuming the same, a solid curve for $Q_{\rm liq}$ in figure 4 was obtained for the water-filled resonator

Having obtained the experimental dependence of $Q_{\rm liq}$ on thickness $r_2 - r_1$ for the water-filled resonator and compared it with the calculated dependence, one can find the CEP coefficient A_S as a function of $r_2 - r_1$ (figure 5).

The coefficient $A_S = (G_S)^{-1}$, where G_S is usually referred to as a geometric factor, determines the loss contribution of the CEP metal to the quality factor of the resonator (see, for example, [15]):

$$Q_{\rm liq}^{-1} = k_1 \tan\delta_1 + k_2 \tan\delta_2 + A_S R_S, \qquad (9)$$

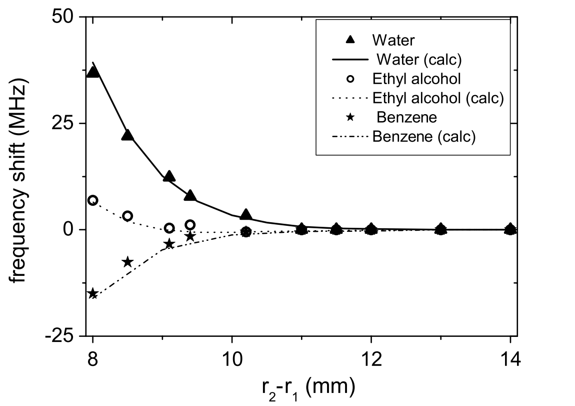

Figure 3. Frequency shift $\Delta f = f_{\rm liq} - f_a$ of QDR as a function of radial thickness $r_2 - r_1$ at a constant external diameter $2r_2$. The solid line represents calculated data for the resonator with water and perfect conducting endplates. The dotted and dash-dotted lines are calculated for the resonator with ethyl alcohol and benzene, respectively; experimental results: (▲) water, (○) ethyl alcohol, (★) benzene.

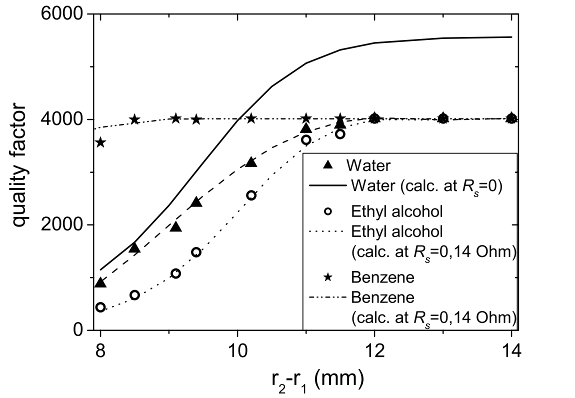

Figure 4. Quality factor of QDR as a function of radial thickness $r_2 - r_1$ at a constant external diameter $2r_2$. The solid line represents calculated data for the resonator with water and perfect conducting endplates. The dashed line is a guide to the eye for experimental points (▲) obtained for water. The dotted line is calculated for ethyl alcohol taking into account microwave loss in conducting endplates, (○) experimental points for ethyl alcohol, (★) experimental points for benzene.

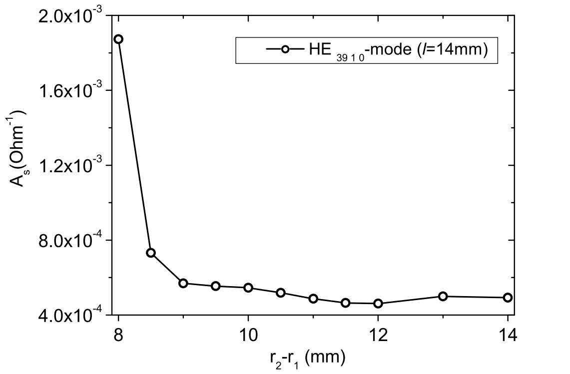

Figure 5. Coefficient A_S against radial thickness $r_2 - r_1$. The solid line is a guide to the eye.

where $k_1 = k_{\rm liq}$ and k_2 are the electric field filling factors of the liquid under test (or air) and the dielectric which the resonator

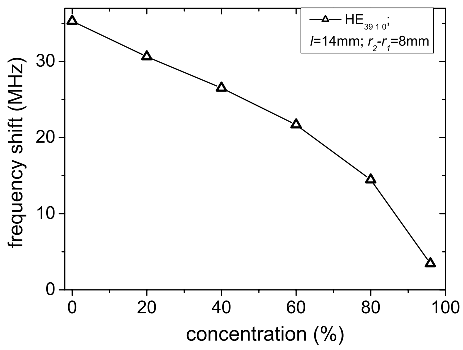

Figure 6. Frequency shift of QDR with binary water–ethyl alcohol mixture as a function of ethyl alcohol concentration in water, mode $HE_{39\,1\,0}$, $l = 14$ mm, $r_2 - r_1 = 8$ mm.

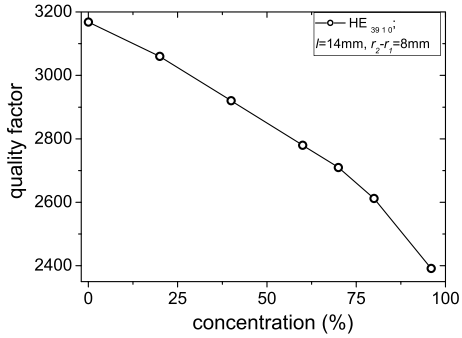

Figure 7. Quality factor of QDR with binary water–ethyl alcohol mixture as a function of ethyl alcohol concentration in water, mode $HE_{39\,1\,0}$, $l = 14$ mm, $r_2 - r_1 = 8$ mm.

ring was made of, $\tan\delta_1$ and $\tan\delta_2$ are the corresponding loss tangents and R_S is the surface resistance of CEP metal. As seen from (9), it is not difficult to find A_S with known R_S. Here all of the coefficients k_1, k_2 and A_S depend on thickness r_2-r_1 and can be calculated using the known distribution of electromagnetic fields in the resonator. Strictly speaking, these coefficients also depend on the properties of a liquid but their dependence on $r_2 - r_1$ is much stronger. This conclusion is confirmed by the comparison between the calculated curves (dashed lines in figures 3 and 4 excluding the dashed line for the water-filled resonator in figure 4) and the experimental results for ethyl alcohol and benzene where values of A_S, shown in figure 5, were used.

3.3. Properties of QDR with water–ethyl alcohol solution

Now we present the results of measuring Δf_{liq} and Q_{liq} of the resonator filled with a binary mixture of water and ethyl alcohol depending on the volumetric part C_e of alcohol over the total interval of variation in alcohol concentration. There are a number of works on the study of binary mixtures of this type (see, for example, [25–28]). The experiments are important for establishing the dispersion of substance properties in the microwave and millimetre wavelength ranges [29]. In this connection it is important to find an opportunity to measure the concentration dependence of the complex permittivity of solutions without using the calibration procedure.

Figures 6 and 7 show the dependence of Δf_{liq} and Q_{liq} on C_e in bidistilled water with the fixed radial thickness $r_2 - r_1 = 8$ mm. Such a thickness allows one to achieve a good sensitivity of a frequency shift Δf_{liq} to variation in the alcohol concentration C_e. We have an opportunity to obtain a stronger dependence of Q_{liq} on concentration with larger thickness $r_2 - r_1$. However, at the same time, the dependence of Δf_{liq} on C_e will be much weaker. Evidently, a certain thickness $r_2 - r_1$ exists, which is optimum in achieving the maximum accuracy of measuring ε' and ε''.

Using a specially designed program for calculations of QDR eigen frequencies and quality factor, the real ($\varepsilon'_{\text{liq}}$) and imaginary ($\varepsilon''_{\text{liq}}$) parts of the complex permittivity of the mixture were determined by fitting the calculated values of Δf_{liq} and Q_{liq} and experimental data. These data are presented in figures 8 and 9. Here the solid lines show the results obtained independently on the basis of Debye's model

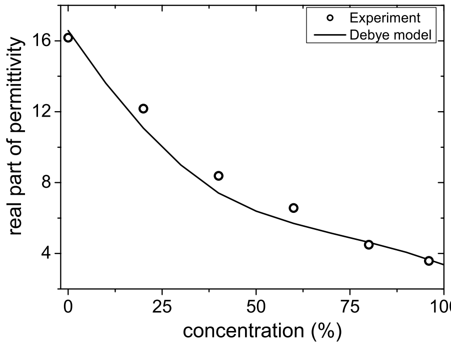

Figure 8. Real part of the permittivity of the binary water–ethyl alcohol mixture as a function of ethyl alcohol concentration. The solid line is the result of calculation using Debye's formula (see the appendix).

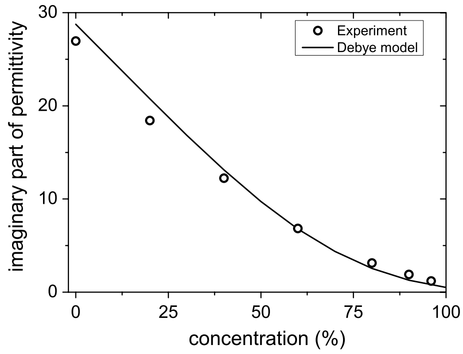

Figure 9. Imaginary part of permittivity of binary water–ethyl alcohol mixture as a function of ethyl alcohol concentration. The solid line is the result of calculation using Debye's formula (see the appendix).

using recently published constants in Debye's formula for the complex permittivity of a binary mixture (see the appendix, where $\varepsilon'_{\text{liq}} = \varepsilon'_m$ and $\varepsilon''_{\text{liq}} = \varepsilon''_m$).

4. Discussion of the results. Justification of a new technique for the complex permittivity measurement of lossy liquids

A number of 'anomalies' found in studying lossy liquid-filled QDR are of certain interest. The 'anomalies' do not manifest themselves for resonators filled with dielectrics having

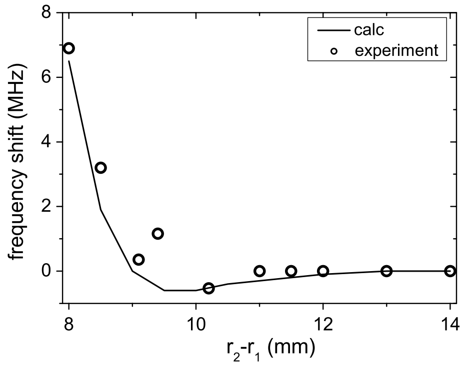

Figure 10. Frequency shift $\Delta f = f_{\text{liq}} - f_a$ of the resonator filled with ethyl alcohol as a function of radial thickness $r_2 - r_1$.

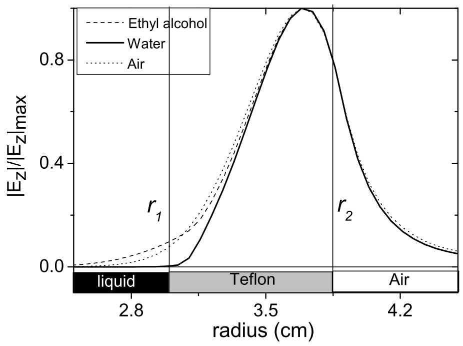

Figure 11. Distribution of the reduced z-component of the electric field along a radius of the dielectric resonator, when it is filled with ethyl alcohol, water or air.

$\tan\delta \ll 1$. They are as follows: (i) a positive sign of the frequency shift Δf_{liq} for water and ethyl alcohol is observed (see figure 3); (ii) the quality factor Q_{liq} of the water-filled resonator is higher than Q_{liq} with ethyl alcohol (see figure 4), although $\tan\delta$ of ethyl alcohol is lower than $\tan\delta$ of water; (iii) the sign of Δf_{liq} for ethyl alcohol can vary depending on radial thickness $r_2 - r_1$ (figure 10).

The aforementioned peculiarities remain valid for water–ethyl alcohol mixtures as well. All specific characteristics of the mixtures are reproduced theoretically by using Maxwell equations and can be explained by the features of the field distribution in a two-layered resonator filled with lossy liquid. Figure 11 shows that the microwave field damps more weakly in alcohol than in water because of a smaller loss in alcohol. This means that the field penetration depth for alcohol is larger than for water and results in a higher value of Q_{liq} in the case of the water-filled resonator. The high values of $\tan\delta_{\text{liq}}$ and $\varepsilon'_{\text{liq}}$ also yield a positive sign Δf_{liq} of the resonator with water and alcohol. The calculations presented in figures 3 and 4 show a good mutual coincidence of all experimental and calculated data. Thus, the ascertained 'anomalies' are not such, despite their seeming uncommonness.

As indicated in the introduction, analogous features were found in lossy substance-containing cavity and dielectric resonators excited on lower modes [19–21]. Thus, the features are qualitatively independent in the type of resonators and frequency range. These factors determine only the quantitative level of the above features.

A very good coincidence of the results of independent measurement of $\varepsilon_{\text{liq}} = \varepsilon'_{\text{liq}} - \mathrm{i}\varepsilon''_{\text{liq}}$ for water–ethyl alcohol mixture using QDR on the one hand, and calculation of the same characteristic with Debye's model on the other (see figures 8 and 9), indicates that the mentioned model describing the binary mixture properties is valid. This coincidence can be considered also as additional justification of the applicability of the proposed technique for microwave characterization of lossy liquids. The proposed approach has one remarkable feature, namely, in principle the resonator allows one to perform direct measurements, i.e. measurements without a calibration procedure using reference samples, because the given resonator makes it possible to solve the electrodynamic problem by means of Maxwell equations. The obtained field components allow us to calculate the coefficients k_{liq}, k_2 and A_s and thus determine $\tan\delta_{\text{liq}}$ by a measured value Q_{liq}. In the present work a coefficient A_s was calculated only for large thickness of $r_2 - r_1$ (see, for example, [14]). It was determined over the entire interval of $r_2 - r_1$ variation using the known properties of water and by comparison of the experimental dependence of Q_{liq} on $r_2 - r_1$ and the calculated dependence of Q_{liq} for the perfect CEP-containing resonator. In future, the coefficient A_s will be calculated through field components of the corresponding mode of the resonator intended for use as a measuring cell.

The dependence of measured values Δf_{liq} and Q_{liq} on both parts, ε' and ε'', of complex permittivity

$$\Delta f_{\text{liq}} = \Delta f_{\text{liq}}\left(\varepsilon'_{\text{liq}}, \varepsilon''_{\text{liq}}\right), \qquad Q_{\text{liq}} = Q_{\text{liq}}\left(\varepsilon'_{\text{liq}}, \varepsilon''_{\text{liq}}\right) \tag{10}$$

complicates the analysis of errors of $\varepsilon'_{\text{liq}}$ $\varepsilon''_{\text{liq}}$ measurements by means of the proposed technique. However, the most probable error (MPE) of the permittivity measurement can be evaluated:

$$\delta\varepsilon'_{\text{liq}} = \left\{\left[\frac{\partial\varepsilon'_{\text{liq}}}{\partial\Delta f_{\text{liq}}}\delta(\Delta f_{\text{liq}})\right]^2 + \left[\frac{\partial\varepsilon'_{\text{liq}}}{\partial Q_{\text{liq}}}\delta Q_{\text{liq}}\right]^2\right\}^{1/2} \tag{11}$$

$$\delta\varepsilon''_{\text{liq}} = \left\{\left[\frac{\partial\varepsilon''_{\text{liq}}}{\partial\Delta f_{\text{liq}}}\delta(\Delta f_{\text{liq}})\right]^2 + \left[\frac{\partial\varepsilon''_{\text{liq}}}{\partial Q_{\text{liq}}}\delta Q_{\text{liq}}\right]^2\right\}^{1/2}. \tag{12}$$

Values of $\delta(\Delta f_{\text{liq}})$ and δQ_{liq} in (11), (12) are the measurement errors of the frequency shift Δf_{liq} and the quality factor Q_{liq} and they give a technical description of frequency-meter and Q-meter, respectively. Derivatives in (11), (12) depend on the properties of the measuring cell, i.e., of the resonator. The above-mentioned derivatives depend strongly on thickness $r_2 - r_1$ and correlation of dielectric constants of liquids and material of the external layer of a two-layered QDR. When using the QDR with Teflon external layer, the errors of $\varepsilon'_{\text{liq}}$ and $\varepsilon''_{\text{liq}}$ measurements of a water–ethyl alcohol mixture can account for tens of per cent. This is due to the fact that the value of $\varepsilon'_{\text{liq}}$ of water is much greater than ε' of Teflon and, as a result, values of derivatives in (11), (12) become rather substantially reduced ($\approx$10%) when using water as a reference liquid. At the same time values of $\varepsilon'_{\text{liq}}$ and $\varepsilon''_{\text{liq}}$ measured by Δf_{liq} and Q_{liq}, with three values of thickness $r_2 - r_1$ were averaged. In addition, the measurements were repeated twice or thrice.

The calculations show that the accuracy of $\varepsilon'_{\text{liq}}$ and $\varepsilon''_{\text{liq}}$ measurements increases sharply when $\varepsilon'_{\text{liq}} < \varepsilon'_2$. However, a complete analysis of $\varepsilon'_{\text{liq}}$ and $\varepsilon''_{\text{liq}}$ error measurements proceeding from improved measurement accuracy goes beyond the scope of the present work. This problem along with the calculation of the coefficient A_S and the development of a measuring cell made of a new material with a larger dielectric constant will be the subject of further studies.

5. Conclusion

Thus, theoretical and experimental investigations into the electrodynamic properties of cylindrical radially two-layered quasi-optical dielectric resonators with conducting endplates have been made. Whispering gallery modes are excited in such resonators. The mode energy is concentrated near the inner side of the cylindrical surface of an external layer. Liquid having an arbitrary value of complex permittivity including liquids with $\tan\delta_{\text{liq}} > 1$ is an internal layer. A number of peculiarities of electrodynamic properties observed experimentally, when the resonator is filled with the lossy liquid, is explained theoretically by peculiarities of the field distribution in the resonator containing such liquids. They are quite similar to the features of other types of resonators with lossy substances irrespective of the frequency range, as should be so from Maxwell equations.

The practical significance of the proposed and studied resonator consists in that it can be applied to direct measurements of complex permittivity of substances under test including liquids with a large microwave energy loss. The QDR-based measurement technique may be promising for application in biology, medicine and biotechnologies.

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Appendix. Concentration dependence of the complex permittivity of a water–ethyl alcohol mixture

The dependences of $\varepsilon'_{\text{liq}}$ $\varepsilon''_{\text{liq}}$ on alcohol concentration in a mixture with water, shown in figures 8 and 9, were calculated using the known Debye's function [24]:

$$\varepsilon_{\text{m}} = \varepsilon'_m - \text{i}\varepsilon''_m = \varepsilon^m_\infty + \frac{\varepsilon^m_0 - \varepsilon^m_\infty}{1 + \text{i}\omega\tau_m}, \tag{A.1}$$

where ε^m_∞ ε^m_0 are the high-frequency and static dielectric constants of mixtures respectively, τ_{m} is the relaxation time in the mixture and $\omega = 2\pi f$ is the cyclic frequency.

Calculations were made for frequency $f = 37.5$ GHz at room temperature (20 °C). The value of ε^m_0 can be calculated on the basis of the model given in [30]. The calculation results presented in [31] are in good agreement with the measured values of ε^m_0 and are used in the present work.

Values ε^m_0 and ε^m_∞ can be connected by means of the relation that follows from the expanded theory by Frölich for static permittivity, where the effective dipole orientation factor

Table 1. Values of ρ_m, $g = g^m_{\text{eff}}$, ε^m_0, ε^m_∞ and τ_m depending on ethyl alcohol concentration in water.

C_e	X_e	ρ_m [33]	g_m [33]	ε^m_0 [30, 31]	ε^m_∞ [32]	τ^m [29]
0	0	0.998	1.185	81.603	3.863	9.6
0.1	0.042	0.985	1.181	76.907	3.936	10.856
0.2	0.089	0.97	1.173	72.107	4.025	12.488
0.3	0.144	0.953	1.161	67.173	4.128	14.669
0.4	0.207	0.935	1.139	62.107	4.264	17.682
0.5	0.281	0.915	1.078	56.86	4.513	22.022
0.6	0.37	0.893	1.045	51.403	4.689	28.608
0.7	0.477	0.87	1.071	45.673	4.675	39.286
0.8	0.61	0.844	1.166	39.563	4.443	58.17
0.9	0.779	0.817	1.341	33.037	4.014	95.735
1	1	0.789	1.607	25.38	3.357	184

g^m_{eff} is introduced [32]:

$$\frac{(\varepsilon^m_0 - \varepsilon^m_\infty)(2\varepsilon^m_0 + 2\varepsilon^m_\infty)}{\varepsilon^m_0(\varepsilon^m_\infty + 2)^2} = \frac{N_{\text{a}}}{9\varepsilon_0 k_{\text{B}} T} g^m_{\text{eff}} \rho_m \left(\frac{X_e\mu_e + X_w\mu_W}{X_e M_e + X_w M_W}\right). \tag{A.2}$$

Here X_e and X_w are the mole fractions of ethyl alcohol (ethanol) and water respectively, M_e and M_w are the mole masses of the mixture components, μ_e and μ_w are the dipole moments of ethanol and water ($\mu_e = 1.68$ D $= 1.68 \times 3.335\,64 \times 10^{-30}$ K m, $\mu_w = 1.84$ D $= 1.68 \times 3.335\,64 \times 10^{-30}$ K m), N_{a} is Avogadro's number, k_{B} is the Boltzmann constant, $\varepsilon_0 = 1/4\pi \times 9 \times 10^9$ F m^{-1} and ρ_m is the density of the mixture.

Data on ρ_m of ethanol–water mixtures are presented in a wide temperature interval in [33]. In the same work, the concentration dependence of g^m_{eff} has been obtained. Values of g^m_{eff} for each component of the mixture equal Kirkwood's orientation correlation factors g_w and g_e.

The temperature dependence of relaxation time is given by the Eyring equation [34]

$$\tau = \frac{h}{k_{\text{B}}} \exp\left(\frac{\Delta G}{RT}\right), \tag{A.3}$$

where h is Planck's constant, T is the absolute temperature, R is the gas constant and ΔG is given by [29]

$$\Delta G_m = X_e \Delta G_e + (1 - X_e)\,\Delta G_w, \tag{A.4}$$

where ΔG_e and ΔG_w are the activation free energies for ethanol and water, respectively. The relaxation time τ_{m} for the mixture is then expressed as

$$\tau_m = \frac{h}{k_{\text{B}} T} \exp\left(\frac{\Delta G_m}{RT}\right) = \tau_e^{X_e} \cdot \tau_w^{(1-X_e)}, \tag{A.5}$$

where τ_e and τ_w are the relaxation times of the mixture components.

The obtained values of ρ_m, $g_m = g^m_{\text{eff}}$, ε^m_0, ε^m_∞ and τ_m depending on the content of ethyl alcohol in water are listed in table 1. The values $\tau_e = 184$ ps and $\tau_w = 9.6$ ps have been drawn from [33] and [35], respectively.

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