
A MODEL OF WATER DIELECTRIC PERMITTIVITY IN MICROWAVE AND TERAHERTZ RANGES

YU.I. MALYSHENKO, V.L. KOSTINA, A.N. ROENKO

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Usikov Institute of Radiophysics and Electronics, Nat. Acad. Sci. of Ukraine
(12, Academician Proskura Str., Kharkiv 61085, Ukraine; e-mail: rk@ire.kharkov.ua)

A model for the calculation of the frequency and temperature dependences of the complex dielectric permittivity (DP) of water within a wide frequency range $f = 0.03 \div 3$ THz ($\lambda = 0.1 \div 10.0$ mm) has been proposed. It reproduces the smooth transition from the Debye mechanism of polarization to the resonance one and ensures a good agreement (with an accuracy of not worse than 5%) between the experimental and theoretical data.

1. Introduction

Water, being the most widespread compound on the Earth and a universal solvent, plays an exclusive role in natural processes, especially in biological ones. The study of its properties, including dielectric ones, was always preferential. But it happened historically that, owing to various technical reasons, experimental researches in the submillimeter (SubMM) wave range were carried on with an appreciable delay, and this circumstance constrained the development of theoretical models. The state of affairs improved within last decades, which assisted to a more intense application of SubMM waves in dielectric spectroscopy, biology, and other branches of science and technology [1]. As a result, there emerged the interest in studying the simple, convenient, and, at the same time, rather exact – with respect to experimental data – models for calculating the complex DP of water in a wide temperature interval from -10 to $+70$ °C and for the frequency range from 0.03 to 3 THz ($\lambda = 0.1 \div 10$ mm). In this work, we propose the model that makes allowance for the main dispersion features of the water DP in the indicated frequency range. It should be noted that the model is iterative and empirical by its character; therefore, in

no case can it be substituted for the results obtained in the framework of the fundamental approach, which has been developed for last years (see, e.g., work [2]). At the same time, our model, owing to its simplicity and, at present time, a higher accuracy of calculations of the complex DP of water in this frequency range, may serve as a convenient supplement to more fundamental ones.

2. General Characterization of the Dispersion of Water Dielectric Permittivity in the Frequency Range of 0.03 ÷ 3 THz

The contemporary experimental data concerning the spectral behavior of the water DP evidence for a smooth dependence of its normal dispersion in the SubMM frequency range without any features (note that the appearance of a weak line near $\tilde{\nu} \approx 60$ cm⁻¹ in the dispersion spectrum of water was reported in work [3]; nevertheless, this fact was not confirmed by other researchers). At the same time, the spectral peculiarities do exist outside the SubMM range, and they influence the frequency dependence of dielectric polarization in it. Thus, on its low-frequency side (in the microwave range), the frequency interval borders upon an intense band of Debye relaxation polarization, with a characteristic maximum located at about $\omega_{\max} = 1/\tau_D$, where τ_D is the temperature-dependent relaxation time (for water, $\tau_D \approx 10$ ps, which corresponds to $\lambda_{\max} = 1.8$ cm at 20 °C). The frequency and temperature dependences of the water DP in the RF and microwave ranges are known to be best described by the Debye equations [4]. For polar substances with the continuous spectrum of relaxation times, the modified Debye equations – Cole–Cole [5] and Cole–Davidson

[6] ones — can be used, depending on either a symmetric or nonsymmetric distribution in time, respectively. But at higher frequencies in the SubMM range, the Debye equations become invalid [4], although the action of the underlying mechanism of relaxation polarization does not stop immediately. As the frequency increases in the SubMM range from 0.3 to $0.6 \div 0.7$ THz, the contribution of the relaxation polarization gradually falls down, introducing a substantial temperature dependence of the water DP, which is characteristic of the Debye mechanism. For the further frequency increase up to 3 THz (this value confines the SubMM frequency range from above), one should expect that the temperature dependence would inevitably become weaker, because the energy of an electromagnetic (EM) field quantum considerably exceeds here the energy of molecular thermal motion. At frequencies $\nu > 3$ THz ($\tilde{\nu} = \nu/c = 1/\lambda > 100 \text{ cm}^{-1}$) the general behavior of the dielectric permittivity dispersion in the far infra-red (IR) range is governed by a number of overlapping bands of the enhanced dielectric polarization. They are associated with another, distinct from Debye, type of intermolecular (translational) vibrations [7], which are considerably more dynamic than the former ($\tau_{\text{res}} \approx 10^{-2}\tau_{\text{D}}$), are less dependent on temperature, and interact with an external EM field in a resonance manner. These bands of the resonance polarization are so-called Poley lines [8] and possess characteristic maxima at about 62, 17, and 6 μm . The first of them, being nearest to the SubMM range, exerts the most pronounced influence on the polarization in its upper section. But, in its midst, the contribution of the 62- μm line to final polarization decreases (because it is a line of the resonance type), and only the distant wing of this line adds together with the aforementioned Debye component, thus forming the experimentally observable dispersion of the water DP. The 17- μm Poley line is caused by the so-called intermolecular librational vibrations [7]; although being more powerful, it is located farther from the SubMM range and, hence, does not contribute markedly to the DP dispersion.

Such ideas concerning the features and sources of the total polarization in the SubMM frequency range allowed us to develop a model for the calculation of the frequency and temperature dependences of the water DP. But before we begin its description, let us analyze the analytical dependences, which are most suitable for the description of the resonance line shape. One variant can be found in the works of adherents of the biexponential model (the so-called Double Debye Model [9, 10]). These researchers suggest to apply the Debye

expression twice, i.e. not only for the description of the proper Debye component in the expression for the total polarization with the relevant value of τ_{D} in the second term, which is natural, but also for the resonance component in the third term (with a much shorter value of τ_{res} , of course):

$$\varepsilon_{\Sigma}^* = \varepsilon_{\infty} + \frac{\varepsilon_{\text{st}} - \varepsilon_2}{1 + j\omega\tau_{\text{D}}} + \frac{\varepsilon_2 - \varepsilon_{\infty}}{1 + j\omega\tau_{\text{res}}}.$$

Really, proper values of the constants in the third term, which are determined from experimental data, make it possible to reproduce the general trend of the dispersion in this frequency range rather successively [10]. Nevertheless, we believe that, from the physical point of view, it is absolutely inexpedient to extend the validity region of the Debye expression, which earlier was always regarded as belonging to the strictly non-resonance type, onto the resonance range. In this case, eligible may be the application of the Fröhlich's [4] or Lorentz's [11, 12] analytical expressions which were specially designed for using in resonance ranges, in the latter term. In both those cases, as was shown in work [12] and will be shown below (on the basis of work [4]), the application of the resonance line shape factors also helps to reproduce the dispersion dependence in this range well enough, with this procedure being more substantiated. Concerning biexponential models, as well as n -exponential ones, we consider them pertinent too in the range of orientational (Debye) polarization and necessary in relevant cases (polar substances with several relaxation times or their mixtures). Such an example, where the analytical expression with five exponents was used to describe the frequency dependence of the DP of biological tissues, can be found in work [13].

Taking the aforesaid into account, we made our choice in favor of the results of the Fröhlich's careful studies of resonance polarization [4], and, correspondingly, his equations (13.10) and (13.11) from work [4] for $\varepsilon'_{\text{res}}$ and $\varepsilon''_{\text{res}}$, respectively (below, these dependences are quoted in formulas (2)–(4)).

3. Description of the Calculation Model

In the SubMM frequency range, as was pointed out above, a transition from the Debye to resonance polarization takes place, so it is these components that must be taken into account by the calculation model. It will be enough for the adequate reproduction of the total polarization (ε'_{Σ} and ε''_{Σ}). A variant of the equations, which sum the specified components up, was proposed

earlier in work [14] and looks like

$$\begin{aligned}\varepsilon'_{\Sigma} &= \varepsilon'_{\text{res}} + \frac{\varepsilon_{\text{st}} - \varepsilon'_{\text{res}}}{1 + (\lambda_S/\lambda)^2}, \\ \varepsilon''_{\Sigma} &= \varepsilon''_{\text{res}} + \frac{\varepsilon_{\text{st}} - \varepsilon''_{\text{res}}}{1 + (\lambda_S/\lambda)^2} (\lambda_S/\lambda).\end{aligned}\quad (1)$$

In this case, the classical Debye equations remain valid, but the constant ε_{∞} in them acquires now, in the SubMM frequency range, a specific physical sense, being coupled with the resonance polarization which is described mathematically by the Fröhlich's equations

$$\begin{aligned}\varepsilon'_{\text{res}} &= 1,7 + \frac{\Delta\varepsilon}{2} \left[\frac{1 + \omega_0(\omega + \omega_0)\tau_{\text{res}}^2}{1 + (\omega + \omega_0)^2\tau_{\text{res}}^2} + \right. \\ &\left. + \frac{1 - \omega_0(\omega - \omega_0)\tau_{\text{res}}^2}{1 + (\omega - \omega_0)^2\tau_{\text{res}}^2} \right],\end{aligned}\quad (2)$$

$$\varepsilon''_{\text{res}} = \frac{\Delta\varepsilon}{2} \left[\frac{\omega\tau_{\text{res}}}{1 + (\omega + \omega_0)^2\tau_{\text{res}}^2} + \frac{\omega\tau_{\text{res}}}{1 + (\omega - \omega_0)^2\tau_{\text{res}}^2} \right].$$

Formulas (1)–(2) contain both the usual constants of the known Debye equations (ε_{st} is the static DP; ε_{∞} is the DP value at the upper frequency limit of the Debye equation validity range (as a rule, at 300 GHz, where $\varepsilon_{\infty} = 4.8$); and $\lambda_S = 2\pi c\tau_D$ is the relaxation wavelength) and the parameters associated with the resonance mechanism of polarization (ω_0 is the resonance frequency; τ_{res} is the relaxation time of resonance vibrations, which is coupled with λ_{res} by the relation $\lambda_{\text{res}} = 2\pi c\tau_{\text{res}}$; $\varepsilon_{\text{opt}} = 1.7$ is the DP value at the optical range edge; and $\Delta\varepsilon = \varepsilon_{\infty} - \varepsilon_{\text{opt}} = 4.8 - 1.7 = 3.1$ is the interval of the DP variation within the resonance frequency range). The frequency ω_0 is connected with the frequency ω_{max} and the amplitude $\varepsilon''_{\text{max}}$ of the maximum in the absorption spectrum by the following relations established by Fröhlich [4],

$$\begin{aligned}\varepsilon''_{\text{max}} &= \frac{\Delta\varepsilon}{2} \omega_{\text{max}} \tau_{\text{res}}, \\ \omega_{\text{max}} &= \frac{1}{\tau_{\text{res}}} \sqrt{1 + (\omega_0 \tau_{\text{res}})^2},\end{aligned}\quad (3)$$

which supplement Eqs. (2).

Equations (1) must satisfy the following boundary conditions:

– at frequencies lower than the middle of the millimeter (MM) range, ε_{res} comes closer to the standard values

$\varepsilon_{\infty} = 4.9 \div 5.5$, and Eqs. (1) transform into the classical Debye ones which are to be used in the microwave range; – in the high-frequency region of the SubMM range, i.e. at the boundary with the far IR range, where the Debye polarization mechanism is impertinent, the second terms in Eqs. (1) disappear, and Eqs. (1) transform into Eqs. (2).

In this work, a particular condition is imposed on Eqs. (2) which can be used, strictly speaking, only in a rather narrow resonance interval. For formulas (1) to be eligible, the scope of the validity of Eqs. (2) is extended to the middle of the SubMM range, i.e. up to its matching with the Debye polarization range. For this purpose, we artificially introduce the frequency-dependent factors q' and q'' . In order to simplify the calculations, we passed to the relative quantities – the frequency $\beta = \tilde{\nu}/\tilde{\nu}_0 = \omega/\omega_0$ and the relaxation time $M = \omega_0\tau_{\text{res}}$ – in Eqs. (2). We recall that the calculation was carried out making use of Eqs. (3) for $\lambda_{\text{max}} = 62 \mu\text{m}$ and $\varepsilon''_{\text{max}} = 1.76$. Therefore, Eqs. (2) read

$$\begin{aligned}\varepsilon'_{\text{res}} &= 1,7 + \frac{\Delta\varepsilon}{2} \left[\frac{1 + (1 + \beta)M^2}{1 + (1 + \beta)^2M^2} + \right. \\ &\left. + \frac{1 + (1 - \beta)M^2}{1 + (1 - \beta)^2M^2} \right] q',\end{aligned}\quad (4)$$

$$\varepsilon''_{\text{res}} = \frac{\Delta\varepsilon}{2} \left[\frac{\beta M}{1 + (1 + \beta)^2M^2} + \frac{\beta M}{1 + (1 - \beta)^2M^2} \right] q'',$$

where

$$q' = 0.5371\beta + \frac{0.8853}{2.0346\beta + 0.6210}$$

and

$$\begin{aligned}q'' &= \frac{M(1 + \beta)}{\beta} \left[0.3773 + 0.4036(0.4036\beta + 1) \times \right. \\ &\left. \times \left(\frac{\beta - 0.6934}{\beta + 0.6934} \right)^2 \right].\end{aligned}$$

The advantage of the chosen variant of the summation of the aforementioned components in the framework of the model developed consists in that the form of Eqs. (1) can be preserved untouched in a very wide frequency range: almost from RF range up to IR one (practically, to frequencies where the next Poley line becomes active).

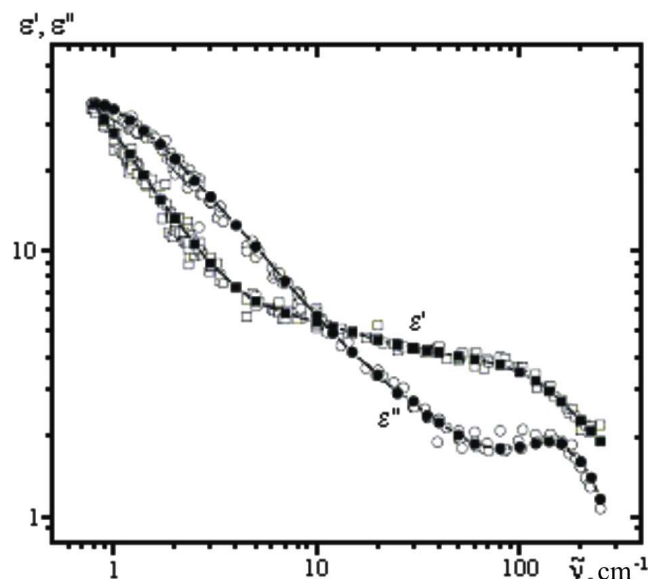


Fig. 1. Experimental data for the complex dielectric permittivity of water in the frequency range $0.03 \div 3$ THz at 25°C and the corresponding averaged curves

Such an approach to the development of a calculation model, but in a more simplified form, was proposed earlier in work [14]. The sufficient accuracy was not achieved at that time, mainly owing to the fact that the then available experimental data for the SubMM frequency range (see, e.g., works [15, 16]) had lower accuracy in comparison with that already achieved in the MM range [17–23]. Nevertheless, for the lack of other calculation opportunities, the equation given in work [14] came into use while predicting the influence of rain droplets on the SubMM radiation propagating through the rain [24], while studying clouds [25], and in other works. That variant of the calculation model was presented most completely in [25, 26].

Since then, new experimental data have been accumulated in both the MM [27–39] and SubMM [10, 40–50] frequency ranges; in the latter case, to a large extent, owing to the implementation of the Fourier-spectroscopy method and the application of femtosecond laser pulses. The results of measurements from all cited experimental works are summarized in Fig. 1. The careful analysis and the generalization of those data evidence for their good coincidence, although they have been obtained by various authors working in various countries and at various laboratories. It allows one to talk about the validity and reliability of the averaged empirical dependences plotted in this figure and about the eligibility of their application as the reference ones while developing the calculation model.

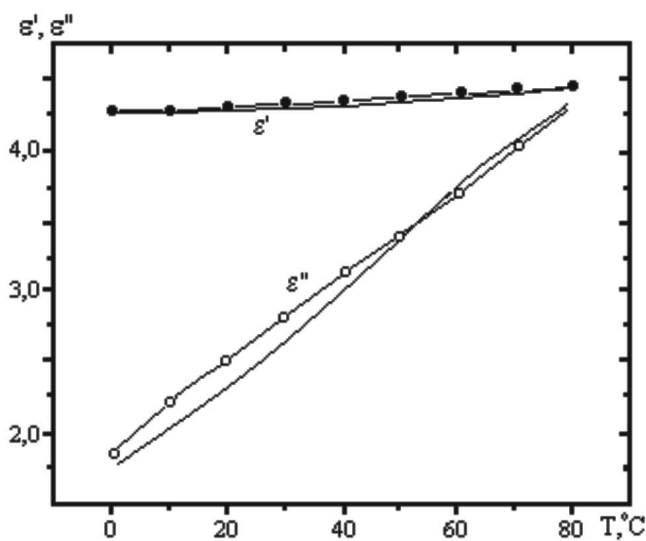


Fig. 2. Temperature dependences of ε' and ε'' for water at a wavelength of $337\ \mu\text{m}$; the data were taken from works [15] (solid circles) and [40] (hollow circles)

For this purpose, at the first stage, the constants in Eq. (3) were determined more accurately; at the second stage, the accuracy of the reproduction of the averaged frequency dependences shown in Fig. 1 was increased by introducing the factors q' and q'' . The optimal values of those quantities were found using a computer code which selected them iteratively following the criterion of minimal root-mean-square deviation (RMSD) of calculated values from experimental ones in the whole SubMM range; the theoretical formulas of Fröhlich (2) were subjected to a minimal correction also.

At the last stage, one more normalization to modern experimental data was made in Eqs. (1) and (2) for the temperature dependence of water DP in the SubMM range, which confirmed that here the dependence is really weaker than that in the MM range. For this purpose, using the results of measurements in the temperature interval from 0 to $+70^\circ\text{C}$ and at the wavelength of $337\ \mu\text{m}$ [15, 40], in the temperature interval from -2 to $+94^\circ\text{C}$ and in the frequency range $0.1 \div 2$ THz [10], and in the temperature interval from -5.6 to $+81.4^\circ\text{C}$ [46], the theoretical temperature dependences were fitted to experimental ones by choosing iteratively the parameter $M = \omega_0 \tau_{\text{res}}(t)$, which enters into Eqs. (4); the fitting was carried out first for a wavelength of $337\ \mu\text{m}$ (see Fig. 2) and then in the whole SubMM range.

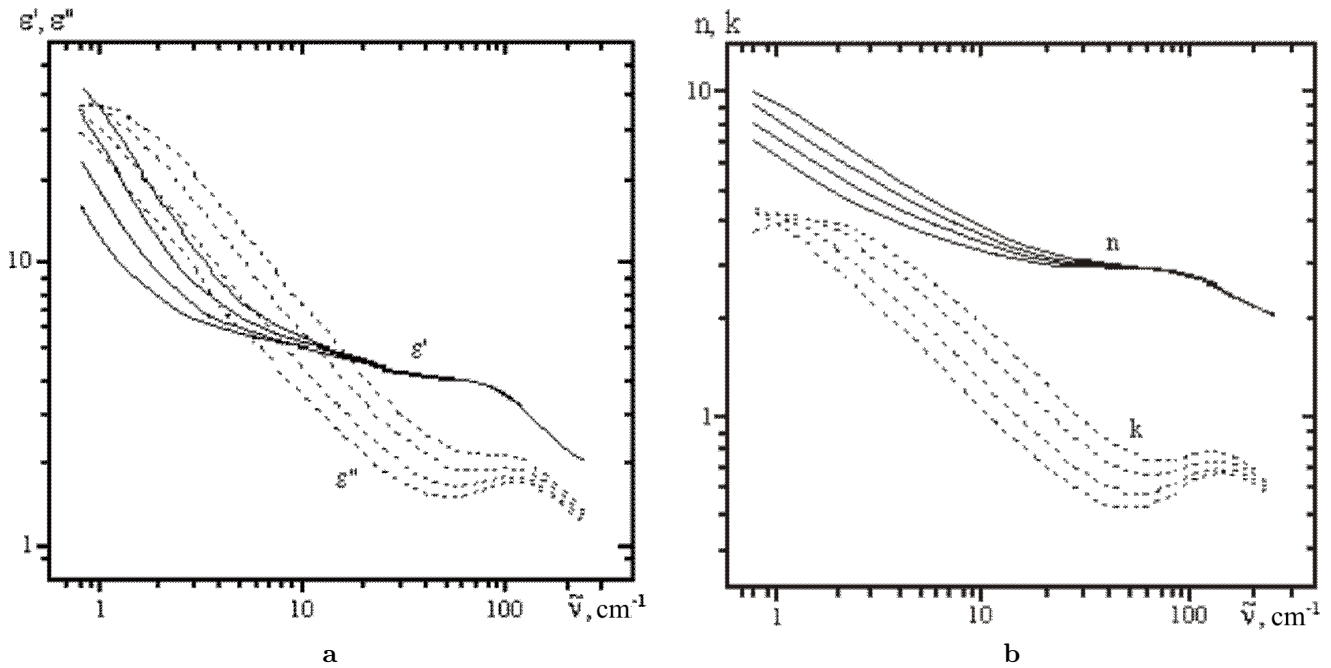


Fig. 3. Final calculated dependences of the complex DP ϵ^* (a) and the complex refractive index $m^* = \sqrt{\epsilon^*} = n+ik$ (b) of water on the frequency at various temperatures 0, 10, 25, and 40 °C (from bottom to top)

The resulting optimal temperature dependence $\tau_{\text{res}}(t)$, where t is measured in centigrade degrees, looks like

$$\tau_{\text{res}}(t, \text{ }^\circ\text{C}) = 6.4423 \cdot 10^{-14} + 2.9144 \cdot 10^{-18}t.$$

In the course of calculations, the temperature dependence of the conventional Debye parameters ϵ_{st} and λ_S was calculated by the formulas [52]

$$\epsilon_{\text{st}}(t, \text{ }^\circ\text{C}) = 0.00081t^2 - 0.40885t + 88.2;$$

$$\lambda_S(t, \text{ }^\circ\text{C}) = 1.4662e^{-0.0634t} + 0.000136t^2 -$$

$$-0.027296t + 1.8735116.$$

In a wider temperature interval, the frequency dependences of the water DP and its refractive index $m^* = (\epsilon^*)^{1/2} = n+ik$ in the MM and SubMM frequency ranges are given in Figs. 3,a and b which can be recommended for practical use.

As was said above, the temperature dependence of the real part of the complex DP of water in the SubMM range is weak (for example, at a wavelength of 337 μm , the tangent of its slope angle falls within the interval 0.002 \div 0.003 [10, 15, 40]) and close to linear; it is also

true for the nearest section of the MM range. This gives a convenient practical opportunity to calculate the required quantities at one temperature using their values at another one, with the help of equations of the type

$$\epsilon'_{\Sigma}(t_2) = \epsilon'_{\Sigma}(t_1) + k'_t(t_2 - t_1),$$

$$\epsilon''_{\Sigma}(t_2) = \epsilon''_{\Sigma}(t_1) + k''_t(t_2 - t_1). \quad (5)$$

The plots of the frequency dependences of the temperature factors k' and k'' in the MM and SubMM ranges, useful for practical application, are depicted in Fig. 4. In the MM range, they transform into the well-approximated Debye dependences. On the right-hand side of the frequency interval, two features should be pointed out in their behavior. First, according to the results of qualitative measurements [10, 46], these factors are minimal in the frequency interval 110 \div 180 cm^{-1} , i.e. just where ϵ'' is maximal, and the value of k' changes its sign (eventually, it becomes negative, as it is in optics). Secondly, the coefficients k' and k'' also become close to the optical ones by magnitude. These agree with the results of measurements [52] carried out at a wavelength of 25 μm (400 cm^{-1}), i.e. near the optical range, $k' = 0.0018$ and $k'' = 0.0022$.

Figure 4 was used to recalculate the numerous experimental data of the aforementioned authors to a

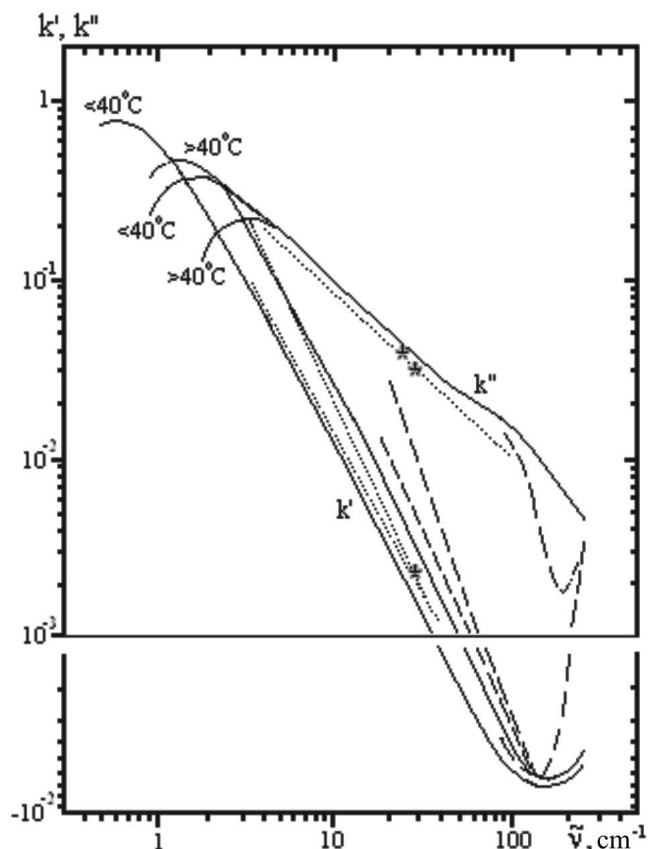


Fig. 4. Calculated frequency dependences of the temperature coefficients k' and k'' in Eqs. (5). For comparison, the approximations of the experimental data from works [10] (dotted) and [46] (dashed curves), as well as the results of measurements from works [15, 40] (asterisks), are plotted

unified temperature. It can be recommended for the application in all similar cases.

4. Conclusions

A model for the calculation of the complex dielectric permittivity of water in the frequency range $0.03 \div 3$ THz and a wide temperature interval from -10 to $+70$ °C has been proposed. It is based on the idea that the frequency dependence of the dielectric permittivity of water in this range is affected by the bands of the enhanced dielectric polarization which are located near to this range, namely, the known Debye relaxation band on the microwave-range side and a number of resonance polarization bands (primarily, by the $62\text{-}\mu\text{m}$ Poley line, which is nearest to the SubMM range) on the far-IR-range side; the phenomenon of resonance polarization was considered by Fröhlich in the general

case. The model proposed provides the coincidence between the calculated frequency and temperature dependences of the water dielectric permittivity and the corresponding experimental data to be not worse than 4–5%.

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МОДЕЛЬ ДІЕЛЕКТРИЧНОЇ ПРОНИКНОСТІ ВОДИ У МІКРОХВИЛЬОВОМУ ТА ТЕРАГЕРЦОВОМУ ДІАПАЗОНАХ ХВИЛЬ

Ю.І. Малишенко, В.Л. Костіна, О.М. Роецько

Резюме

Запропоновано розрахункову модель для обчислення частотної та температурної залежностей діелектричної проникності води у широкому частотному діапазоні $f = 0,03 \div 3$ ТГц ($\lambda = 10,0 \div 0,1$ мм), яка відтворює плавний перехід від дебайвського механізму поляризації до резонансного та забезпечує збіг з експериментальними даними з похибкою в межах 5%.