Dielectric permittivity of deeply supercooled water according to the measurement data at the frequencies 7.6 GHz and 9.7 GHz

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Dielectric permittivity of supercooled volume water has been measured in the range of temperatures from $-20^\circ C$ to $-60^\circ C$ at the frequencies 7.6 GHz and 9.7 GHz. The measurements have been made using microwave resonators and wetted silicate sorbents. From the data obtained, the temperature dependences of the relaxation frequencies were updated for the Debye model with two relaxation frequencies. The updated formulae for dielectric permittivity of cold water may be applied to the frequency range $7\ldots200$ GHz.

Introduction

The knowledge of dielectric permittivity of supercooled volume water (down to $-60^\circ C$) at microwaves is of interest for the study of the physical and chemical properties of water and of water solutions. There are numerous problems relating to the study of hydrosphere, biosphere, and atmosphere, where the knowledge of the microwave characteristics of water and their details is required

However, measuring the complex relative dielectric permittivity ($\varepsilon$), especially at the temperatures ($T$) below $-20^\circ C$, is a complicated task. In [1], it was solved using microemulsion of water, when the researchers were able to cool the water samples down to $-18^\circ C$ and to make measurements of the real ($\varepsilon'$) and imaginary ($\varepsilon''$) parts of $\varepsilon$ near the frequency 9.6 GHz.

In [2, 3], wetted nanoporous silicate materials were used for the purpose, in which water could be supercooled to $-90^\circ C$. Dependences $\varepsilon''$ were found in a broad range of frequencies ($f$) and temperatures. In those works, originally dependence of the attenuation constant for radiation intensity of wetted medium was measured. Further, to calculate $\varepsilon''$, theoretical values $\varepsilon'$ were used, which, however, were not experimentally validated for the water temperature below $-20^\circ C$. The situation was complicated also by the fact that, according to the study results of the dielectric permittivity models [1, 2, 9], which were applied to the temperature range below $-20^\circ C$, it was found that the values of $\varepsilon'$ differed by several times.

To objective of the present study was to measure and to investigate the temperature dependence $\varepsilon'$ for pore water, close for its characteristics to supercooled metastable volume water. The study was conducted for the temperature range $0\ldots-60^\circ C$ near the frequencies 7.6 and 9.7 GHz.

The method used

As shown in [10, 12], water contained in nanometer-size pores of silicate sorbents has properties differing from the volume water only for 1 – 2 layers of water molecules on the pore surface. The other layers are close for their characteristics to metastable volume water. This unique feature of the silicate sorbents was used for measuring $\varepsilon'$ of supercooled water.

It is known that in such media the phase transition temperature ($T_c$) decreases by the value $\Delta T_c = c/(R - t)$, where $c = 62$ degrees-nm, $R$ – the pore radius in nm, $t \approx 0.38$ nm [12]. In the case of pores having the radius 3 nm, $\Delta T_c$ is $\approx 24^\circ C$. In the case of incomplete filling of the pores, additional supercooling is observed, compared to $\Delta T_c$ by approximately $10\ldots15^\circ C$ [13]. In addition, in the cooling – heating process, the freezing – melting temperature hysteresis is formed, at which the freezing temperature is lower than the values found from the formula for $\Delta T_c$. The hysteresis may exceed $10^\circ C$ and is determined by the value of the average water content and the cooling rate [13, 14].

To reach the temperature in the area of the “no man’s land” ($T = -37\ldots-120^\circ C$) [7, 13] low values of wetted sorbent materials with the gravity water content $\sim 3\ldots4\%$ were used. The cooling rate was selected not to exceed $10^\circ C/hr$.

As the proportion of volume water in the medium is $\sim 1\%$, and its value $\varepsilon'$ goes down with the temperature decrease, the resonator method was used, in which complete filling of the resonator with the medium is achieved. Such a method was used, for example, in [15] to measure $\varepsilon'$ of freshwater ice in determining the impact of small concentrations of liquid inclusions in it. In the course of our study, we discovered the ability of wetted disperse media to form macroscopic inhomogeneities due to migration of the liquid in the presence of a temperature gradient and at freezing of free water between the grains. This effect leads to distortion of the resonance curves and to essential reduction of the measurement accuracy. In [4], it was eliminated at measurement of $\varepsilon''$, placing large-size samples in free space, which resulted in reduction of the impact of the medium inhomogeneities and of the spatial dispersion caused by them.

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A similar approach was used in this study for resonator measurements. Resonators were used with higher numbers of the oscillation modes and hence of the resonator volume. Such a technique allows averaging by a large number of inhomogeneities. In addition, data averaging was performed at the frequency range \( \sim 1\% \) of the average frequency.

In our study, we used rectangular transmission-type resonators, connected to waveguide transmission lines with the waves of the \( H_{10} \) type. The resonance frequencies of a rectangular resonator for loss-free medium are shown by the following formula:

\[
f_{mnp} = \left( \frac{c}{\sqrt{\frac{m^2}{a^2} + \frac{n^2}{b^2} + \frac{p^2}{l^2}}} \right) \sqrt{2\varepsilon'}, \tag{1}
\]

where \( m, n, p = 0, 1, 2, \ldots, a, b, l \) — the resonator dimensions along the Cartesian fields of coordinate axes \( (x, y, z) \), \( c \) — the velocity of electromagnetic waves in vacuum. For waves of type \( H_{10} \) in a rectangular waveguide and for radiation propagation along axis \( z \), resonators with the mode of oscillations \( H_{10p} \) are used, where \( p = 1, 2, 3, \ldots \)

In the proposed technique, modes for \( p > 1 \) should be used, i.e., \( H_{102}, H_{103} \) etc. For this case, \( m = 1, n = 0 \) and \( p = 2, 3, \ldots \)

The respective resonance frequencies are:

\[
f_{10p} = \left( \frac{c}{\sqrt{\frac{a^2}{2} + \frac{p^2}{l^2}}} \right) \sqrt{2\varepsilon'}, \tag{2}
\]

From Eq. (2) for a certain mode of oscillations and based on the measurements of the resonance frequencies of an empty \( (f_0) \) and filled resonator, \( \varepsilon'_c \) of wetted medium is found:

\[
\varepsilon'_c = \left( \frac{f_0}{f_{10p}} \right)^2. \tag{3}
\]

In case of losses to the medium, to the resonator walls and losses due to connection with the transmission waveguides, the value of the resonance frequency decreases, and \( \varepsilon'_c \) is found by the formula:

\[
\varepsilon'_c = \left( \frac{f_0}{f_{10p}} \right)^2 \left( 1 - \frac{1}{4Q_c^2} \right), \tag{4}
\]

where \( Q_c \) — the quality factor for the resonator filled with the medium. For \( Q_c \geq 10 \), which was observed in the experiment, the error of estimate \( \varepsilon'_c \) is less than 0.25%. Therefore, the value \( \varepsilon'_c \) for wetted medium filling the resonator was derived from formula (3).

To determine the dielectric parameters of water, a refraction model was used for a mixture of different dielectrics [10], according to which:

\[
\sqrt{\varepsilon_c} = \sqrt{\varepsilon_m V_m} + (1 - V_m) \sqrt{\varepsilon_B}, \tag{5}
\]

where \( \varepsilon_B \) is related to volume water in pores, \( \varepsilon_m \) is related to the structure consisting of solid matrix, empty space in pores and between the material grains, \( V_m \) — the relative volume of the medium, excluding liquid volume water.

Equation (5) contains complex variables and the unknown value \( V_m \) and \( \varepsilon_m \). To solve equation (5), additional information is introduced. It is assumed based on [7] that at temperature \(-90^\circ\text{C}\) free water freezes out. As its mass constitutes \( \sim 1\% \) of the total mass of the substance, and the dielectric permittivity of the formed ice is close to value \( \varepsilon \) of the silicate material, we find \( \varepsilon_m \) from the resonance frequency measurements at a temperature lower than \(-90^\circ\text{C}\) within the error margin of one percent. The real part \( \varepsilon_m \) is found from its equivalence \( (f_0/f_{p_{\text{max}}})^2 \), where \( f_{p_{\text{max}}} \) — the resonance frequency lower than \(-90^\circ\text{C}\). The imaginary part: \( \varepsilon''_m = \varepsilon'_m (\Delta f_{p_{\text{max}}}/f_{p_{\text{max}}}) \). Here \( \varepsilon'_m \) — the effective loss factor, which takes into account losses to the dry medium and waveguide walls and losses due to connection with the waveguides, \( \Delta f_{p_{\text{max}}} \) — resonance bandwidths at half power transmitting.

To find \( V_m \), a priori information was found of the values of dielectric permittivity of water at \(-18^\circ\text{C}\) from [1].

From (5) it follows:

\[
V_m = \frac{\sqrt{\varepsilon_c} - \sqrt{\varepsilon_B(-18)}}{\sqrt{\varepsilon_c} - \sqrt{\varepsilon_B(-18))}. \tag{6}
\]

\( \varepsilon_c \) is found from measurements at the temperature \(-18^\circ\text{C}\). The real part \( \varepsilon'_c \) is determined from (3). \( \varepsilon''_c \) is found from the measurements of the resonator’s quality factor. Substituting the value \( \varepsilon''_c \) for \(-18^\circ\text{C}\) in (6), we find \( V_m \) and \((1 - V_m)\).

The \( Q_c \) is determined by the sum of two values:

\[
\frac{1}{Q_c} = \frac{1}{Q_a} + (tg\delta)_B, \tag{7}
\]

where \( Q_c = f_{10p}/\Delta f_{10p}, (tg\delta)_B = \varepsilon''_B/\varepsilon'_B \) refers to losses in volume water, \( Q_a \) is the quality factor determined by losses to the resonator walls, connection to the waveguides and dry sorbent material. \( Q_a = f_{p_{\text{max}}}/\Delta f_{p_{\text{max}}} \), determined from the measurements at a temperature lower than \(-90^\circ\text{C}\). From (7) we obtain:

\[
\varepsilon''_c = \varepsilon'_c \left( \frac{1}{Q_c} - \frac{1}{Q_a} \right). \tag{8}
\]

Substituting the required values in (8), we find \( \varepsilon_B \).

The procedure of measurements

The microwave parameters of the resonator were measured using the Micran frequency characteristics analyzer. The scanning time for the entire frequency range
from 6 to 11 GHz was 2 s, and the number of the measured points in the frequency band was 5000. The obtained values were processed on the computer, with dependences smoothed, the resonance curves approximated with bell-like functions and the resonance frequencies and width determined. The temperature of the medium investigated was measured with a thermocouple embedded in the opening of the resonator’s wide wall.

The temperature measurements were made using a climate chamber Espec SU-261, which allowed cooling of the resonator with the medium temperature down to $-60^\circ\text{C}$. To obtain lower temperatures and complete freezing of volume water, the chamber was switched off as the temperature reached $-60^\circ\text{C}$, and the resonator was cooled down with liquid nitrogen, followed by its slow heating. During those changes, hysteresis of the resonator characteristics ($f_{10p}$ and $\Delta f_{10p}$) was observed. With the used cooling mode $\sim 10^\circ\text{C}/\text{hr}$, nanoporous silicate sorbent materials with the pore diameter 3 – 4 nm and their water content 4%, the observed hysteresis of the resonator characteristics in the range of temperatures $-30\ldots-50^\circ\text{C}$ was 30°C. That allowed us to conclude that temperature of the supercooled water in the pores without water crystalization reached $-60^\circ\text{C}$. This conclusion was confirmed by the absence of the increase of $\varepsilon''$ at water cooling below $-23^\circ\text{C}$, at which ferroelectric ice 0 is formed. This crystal modification of ice results in the increase of the loss factor due to emergence of a highly conductive layer at the ice — dielectric boundary [17].

In the experiment, a transmission resonator was used, with the dimensions $a = 23$ mm, $b = 10$ mm, $l = 43$ mm. Diaphragms with holes 5 mm in diameter was placed in the plane $x, y$, where the cross section of the transmission waveguides was $23 \times 10$ mm$^2$. The measured resonance frequencies of the empty resonators were found to be equal to: $f_{101} = 7.420$ GHz, $f_{102} = 9.805$ GHz, $f_{103} = 12.23$ GHz.

### Measurement results

The above technique was applied to measurements in the range of frequencies 7.6…9.7 GHz at temperatures from $0^\circ\text{C}$ to $-60^\circ\text{C}$. Their choice was connected with the experimental work [1], where at the frequency 9.6 GHz water was supercooled to $-18^\circ\text{C}$. The data from this study presented in [1] were used to update our measurements in determining the volume concentration of metastable water.

In the experiment, KSKG silica gel made in Hong Kong with the average pore size 8 nm and Acros silica gel manufactured in Belgium with the average pore size 6 and 9 nm were used. Measurements with a rectangular resonator were conducted simultaneously at two frequencies for the modes $H_{102}$ and $H_{103}$. For KSKG with the gravity water content 4%, resonances for the modes $H_{102}$ and $H_{103}$ were near the frequencies 7.6 GHz and 9.7 GHz.

The results of $\varepsilon$ measurements after computer process-

| $i$ | $a_1$ | $a_2$ |
|-----|-------|-------|
| 0   | 5.6500E00 | 6.3143E00 |
| 1   | 1.6960E-02 | 2.8841E-02 |
| 2   | -1.4810E-04 | 3.9205E-01 |
| 3   | 1.4627E01 | -3.2094E-03 |
| 4   | -4.2926E-01 | 7.6578E-04 |
| 5   | 7.5714E-03 |

Table I: Coefficients in formulae (10)

The formulae presented for the two-frequency Debye relaxation model look as follows for dielectric permittivity of pure water:

$$\varepsilon(T) = \frac{\varepsilon_s(T) - \varepsilon_1(T)}{1 + i f / f_1(T)} + \frac{\varepsilon_1(T) - \varepsilon_\infty(T)}{1 + i f / f_2(T)} + \varepsilon_\infty(T), \quad (9)$$

where $\varepsilon_1$ is the intermolecular constant of dielectric permittivity, $f$ is frequency, $\varepsilon_s$ — static dielectric constant, $f_1$ and $f_2$ — the first and second relaxation frequencies. For $\varepsilon_s$, $\varepsilon_1$, $\varepsilon_\infty$, $f_1$, $f_2$, there are the following formulae [1]:

$$\varepsilon_s(T) = \frac{3.70886 \cdot 10^4 - 8.2168 \cdot 10^3 T}{4.21854 \cdot 10^2 + T}$$

$$\varepsilon_1(T) = \frac{A + T}{a_3 + a_4 T + a_5 T^2}$$

$$\varepsilon_\infty(T) = \frac{A + T}{a_8 + a_9 T + a_{10} T^2},$$

where $f$ is expressed in GHz, $T$ — in degrees Celsius.

For the purpose of their use in the region of negative temperatures, dependences of two relaxation frequencies $f_{1,2}(T)$ were specified. In [1], assuming the impact of the Widom line of water for the temperature of $-45^\circ\text{C}$ [13, 17, 18], parameter $A$ was chosen to be equal to 45. As $T$ was expressed in degrees Celsius, at that relaxation temperatures turned into zero. In our measurements, we observed an electromagnetic response at lower temperatures, too.

Based on the assumption that in cold water its vitrification occurs at the temperature of approximately $-130^\circ\text{C}$ [18], parameter $A$ was selected to be equal to 130. After adjustment of the coefficients in formulae (10) to achieve compliance of $\varepsilon'$ and $\varepsilon''$ with the experimental values at the temperatures below 0°C, the following coefficient values were found: the table I contains coefficients in formulae (10).

Shown in Fig. 1 are dependences $\varepsilon'$ and $\varepsilon''$ on temperature for frequency 9.7 GHz. To compare, the results of computations using formulae [1] are provided. At the
temperture below $-45^\circ$C, in [1] the values $\varepsilon''$ are not determined, therefore, they were taken to be equal to zero.

Frequency dependences $\varepsilon'$ and $\varepsilon''$ for different temperatures are shown in Figs. 2, 3.

The obtained values of $\varepsilon'$ were used for adjusting the results for $\varepsilon''$ at the frequencies of the millimeter band by the measurements of the attenuation coefficient provided in [8]. Fig. 4 demonstrates the temperature dependences $\varepsilon''$ from [8] for the frequency of 125 GHz.

**Discussion of results**

The main difficulty in conducting measurements of $\varepsilon$ consists in eliminating the distortions of resonance curves in the form of oscillations on the curve slopes. The distortions are related to inhomogeneities emerging in wetted micro disperse medium. In wetted medium with the water content above 4%, it is practically impossible to make
measurements. This feature made computer calculations more difficult due to the difficulty of determining the precise values of the resonance frequency and of the width of the resonance curve. In smoothing of the measured resonance curves: 1 — by averaging of curves by neighboring points in the band of frequencies; 2 — by Gaussian curves; 3 — by the Lorentz dependence, variations of the final results were observed \( (\varepsilon') \sim \pm 25\% \) from the mean value. The smoothest dependences \( \varepsilon'(T) \) were obtained at averaging in the bandwidths 70 MHz (at 100 points), followed by approximation by the Gaussian function.

To obtain dependences \( \dot{\varepsilon} \) on frequency, formulae from [1] were specified for the two-frequency Debye model for pure volume water. The main idea was in correct selection of the temperature dependence of relaxation frequencies on temperature. The application of the two-frequency model has certain reasons due to development of a two-structure model of water, which considers the fluctuating clusters of water of high-density (HDL) and of low-density (LDL) [13]. These structures are in complex interaction. Their concentration is determined by temperature, and at cooling below \(-50^\circ\text{C}\) LDL begins to prevail. At temperature around \(-130^\circ\text{C}\) the relaxation time becomes equal to 100 s (vitrification begins), hence, in Debye model near this temperature we can consider the relaxation frequencies to turn into zero.

Validation of formulae (9, 10) for the available measurement data of \( \varepsilon'' \) at millimeter frequencies [6, 8] demonstrates good compliance of the experimental results obtained by using two different methods. A number of factors influence the accuracy of the results, apart from the measurement error. It is known that water density decreases as it is cooled from \(+4^\circ\text{C}\) to \(-70^\circ\text{C}\) [19]. This effect is compensated to a certain degree by selection of coefficients in the temperature dependence of the relaxation times. A more complicated issue consists in compliance of the parameters of pore water and of volume water (for macro volumes), as well as the error introduced by strongly bound water. For strongly bound water, the relaxation frequencies are in the range of 0.1 GHz, but this water may influence the value of \( \varepsilon' \) at the frequencies of the gigahertz order.

In certain cases, when crystallization begins at temperatures below \(-23^\circ\text{C}\), not ice Ih or Ic but ice 0 may be formed in wetted medium [20, 21]. The peculiar feature of this ice is that it is ferroelectric and has strong influence on the microwave characteristics of the medium [21]. Ice 0 may be formed on the grain surface or in the space in-between the sorbent grains. It is likely that it caused the failure of the experiments in wetted media with the water content higher than \(3 - 4\%\).

The accuracy of determining the values \( \dot{\varepsilon} \) by formulae (10) for volume water at temperatures below \(-20^\circ\text{C}\) is approximately equal to 30%. In order to raise the degree of this accuracy, measurements are required to be made in a broader range of frequencies and the special relaxation and absorption mechanisms have to be investigated. For example, in [6, 8] the increase of \( \varepsilon'' \) near \(-45^\circ\text{C}\) was investigated due to the impact of the specific behavior of water on the Widom line [18]. The Widom line is related to the existence of the second critical point of water. At it, fluctuations of water density and entropy drastically rise, and a number of the thermodynamic characteristics of water change (specific heat capacity at constant pressure, coefficients of volume expansion and of isothermal compressibility).

It is to be noted that the obtained results proved to be applicable, based on the comparison results, to the temperature \(20^\circ\text{C}\) and the frequency 200 GHz.

**Conclusions**

1. A method is proposed for measuring relative dielectric permittivity of supercooled water in the microwave range for temperatures from \(-20^\circ\text{C}\) to \(-60^\circ\text{C}\). It is based on the special properties of water contained in nanoporous silicate materials, in which water is close to metastable volume water for its parameters. In this technique, in order to eliminate the impact of inhomogeneities resulting from emergence of clusters at migration of water due to temperature gradients and phase transitions, measurements are conducted in resonators which have relatively large volumes and a low degree of wetting of the materials. In this case, the impact of the inhomogeneities is averaged, and distortions in the measured electromagnetic characteristics of the media under study become eliminated. The values \( \varepsilon' \) and \( \varepsilon'' \) near the frequencies 7.6 GHz and 9.6 GHz were measured, and analytical dependence \( \dot{\varepsilon}(T) \) in the range of \(-20\ldots-60^\circ\text{C}\) was obtained. The obtained dependence \( \dot{\varepsilon}(T) \) may be spread to the range of frequencies \(7 \ldots 200\) GHz and temperatures \(-90\ldots+20^\circ\text{C}\), which the comparison results showed.

2. The technique proposed may ensure approximately 30% accuracy of measuring dielectric permittivity of supercooled water. However, a question remains regarding correspondence of the parameters of water contained in the pores of a material to those of the ideal volume water. The accuracy of determining the dielectric characteristics of water depends on a number of parameters: the proportion of bound water and the breakdown of its characteristics by the water layers adjacent to the surfaces of the medium boundaries; the specific details of the sample cooling techniques; the physical-chemical characteristics of the sorbent matrix; the values of electric conductivity of the layers at the medium interfaces and certain other parameters. Therefore, the expected accuracy of determining the values of dielectric permittivity in relation to ideal volume water may significantly vary in any method of measurement. This relates also to the previously conducted measurements using microemulsions of water [4]. In our experiments involving two different types of silica gels, the difference between the values of \( \varepsilon' \) reached 25%.

3. The measurements performed refer to cold water in the region of "no man’s land". This region is interesting due to the most conspicuous anomaly of water, decrease
of its density when temperature changes from $+4^\circ C$ to $-70^\circ C$ \cite{19}. In its turn, this anomaly is related to the structural features of liquid water: interaction and mutual transformations of clusters LDL and HDL, and the existence of the second critical point. The specific features of the dielectric permittivity of supercooled water in porous media may be used in microwave spectroscopy of the processes of water freezing (melting) in closed space. For example, it has been found that at freezing of water below $-23^\circ C$, ferroelectric ice 0 is formed. Its appearance results in the rise of electromagnetic losses, which may be used for the study of the chemical processes in cooling of natural disperse media due to the change of the chemical potential of the inclusions. Equally interesting is the issue of investigating the microwave characteristics of water near the Widom line using the proposed technique (at the temperatures $-45 \ldots -53^\circ C$ and the pressure $0.1 \ldots 100$ MPa). At present, there are little experimental results on this issue.

\begin{thebibliography}{21}

\bibitem{1} T. Meissner and F. J. Wentz, IEEE Trans. Geosci. Remote Sens. \textbf{42}, 1836 (2004).
\bibitem{2} P. W. Rosenkranz, IEEE Trans. Geosci. Remote Sens. \textbf{53}, 1387 (2015).
\bibitem{3} D. D. Turner, S. Kneifel, and M. P. Cadeddu, J. Atmos. Oceanic Technol. \textbf{33}, 33 (2016).
\bibitem{4} D. Bertolini, M. Cassettari, and G. Salvetti, J. Chem. Phys. \textbf{76}, 3285 (1982).
\bibitem{5} G. S. Bordonskii, A. O. Orlov, and S. D. Krylov, Journal of Communications Technology and Electronics \textbf{64}, 375 (2019).
\bibitem{6} G. S. Bordonskiy and A. O. Orlov, Preprint arXiv: 1901.03979 [cond-mat.soft], 8 (2019).
\bibitem{7} D. T. Limmer and D. Chandler, J. Chem. Phys. \textbf{137}, 044509/1 (2012).
\bibitem{8} G. S. Bordonskii and A. O. Orlov, Izvestiya, Atmospheric and Oceanic Physics \textbf{55}, 1005 (2019).
\bibitem{9} W. J. Ellison, J. Chem. Phys. Ref. Data \textbf{36}, 1 (2007).
\bibitem{10} L. I. Menshikov, P. L. Menshikov, and P. O. Fedichev, Journal of Experimental and Theoretical Physics \textbf{125}, 1173 (2017).
\bibitem{11} S. R.-V. Castrillon, N. Giovambattista, I. A. Arsay, and P. G. Debenedetti, J. Phys. Chem. C \textbf{115}, 4624 (2011).
\bibitem{12} E. G. Solveyra, E. Llave, D. A. Scherlis, and V. Molinero, J. Phys. Chem. B \textbf{115}, 14196 (2011).
\bibitem{13} P. Gallo, K. Amann-Winkel, C. A. Angell, M. A. Anisimov, F. Caupin, C. Chakravarty, E. Lascaris, T. Loerting, A. Z. Panagiotopoulos, J. Russo, J. A. Sellberg, H. E. Stanley, H. Tanaka, C. Vega, L. Xu, and L. G. M. Pettersson, Chem. Rev. \textbf{116}, 7463 (2016).
\bibitem{14} J. B. W. Webber, Progress in Nuclear Magnetic Resonance Spectroscopy \textbf{56}, 78 (2010).
\bibitem{15} C. Mätzler and U. Wegmuller, J. Phys. D: Appl. Phys. \textbf{20}, 1623 (1987).
\bibitem{16} J. R. Birchak, L. G. Gardner, J. W. Hipp, and J. M. Victor, Proc. IEEE \textbf{62}, 93 (1974).
\bibitem{17} B. Widom, J. Chem. Phys. \textbf{39}, 2808 (1963).
\bibitem{18} V. Holten, C. E. Bertrand, M. A. Anisimov, and J. V. Sengers, J. Chem. Phys. \textbf{136}, 094507 (2012).
\bibitem{19} F. Mallamace, C. Branca, M. Broccio, C. Corsaro, C.-Y. Mou, and S.-H. Chen, Proceedings of the National Academy of Sciences \textbf{104}, 18387 (2007).
\bibitem{20} D. Quigley, D. Alfè, and B. Slater, J. Chem. Phys. \textbf{141}, 161102/1 (2014).
\bibitem{21} G. S. Bordonskiy and A. O. Orlov, JETP Letters \textbf{105}, 492 (2017).
\end{thebibliography}