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Investigation of the possibility of ice film 0 formation on the dielectric surface in a microwave resonator

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Abstract

Purpose: The possibility of detecting ice 0 by deposition of water vapor on quartz glass dielectric plates placed in the cavity of a microwave rectangular resonator near the frequency of 2.8 GHz is investigated.

Experimental: Measurements of the characteristics of the resonator filled with air at atmospheric pressure in the temperature range from 5 to −140 °C have been performed. Variations of the transmittance power of the resonator at the resonance frequency and its quality factor of fit were found with their characteristic change at a temperature of −23 °C. This temperature corresponds to the formation of ice 0 from supercooled water. It is assumed that in the experiment, films of ice 0 are detected in the response of the resonator to the temperature change in the investigated interval.

Conclusions: This result is of interest due to the possible influence of water vapor condensation on the functioning of a variety of technical devices in terrestrial conditions when ice 0 is formed.

Keywords: Ice 0, Conductive films, Microwave range, Resonator measurements

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1. Introduction

The papers [1–3] reported the discovery by computer modeling of a crystalline modification of ice called ice 0. This ice is a ferroelectric, contains 12 water molecules in the unit cell, and forms at a pressure of 0.1 MPa at temperatures below –23 °C. Ice 0 is considered a transitional form from deeply supercooled water to hexagonal ice Ih. In papers [3, 4], such an ice phase was registered during measurements of the extinction of laser radiation in the visible range when radiation was transmitted through plates made of various dielectrics with a layer of nanometer-thick ice condensed from water vapor. The plates consisted of glass, mica, Ih ice and sodium chloride crystals.

The special property of the ferroelectric to form strongly conducting thin, on the order of nanometer, layers at the dielectric boundary was utilized in the experimental studies [5,6]. These layers created a significant absorption (and reflection) of external electromagnetic radiation from the ice film 0 deposited on the dielectric substrate. If the plates with the investigated ice were heated above –23 °C, the extinction effect abruptly disappeared, which served as evidence for the formation of ice 0 on the surface. The electrodynamic model of the structure was presented in the form of island films of ice covered with a layer of high conductivity, in which a resonance of surface plasmon modes appeared [7, 8]. These modes in the case of irregularly shaped conductive particles of nanometer dimensions create scattering and absorption in a wide frequency interval extending from the optical to the radio range. The upper frequency of this interval is determined by the plasma frequency of charge carriers with a sharp maximum of the effect, which is located at the Frelich frequency. At this frequency, for a conducting layer $\varepsilon' = -2$ (ε' is the real part of the relative complex dielectric constant). For example, theory and extinction measurements for small aluminum ellipsoids have shown that the extinction changes by about 5 orders of magnitude as the wavelength increases from its maximum (~0.5 μm) to 1 mm [7]. For spherical particles, the same change is much larger and amounts to 9 orders of magnitude. Thus, it can be expected that extinction should be manifested during the formation of inhomogeneous ice film 0 not only in the optical, but also in the microwave range. It should be noted that in recent years, studies of nanostructures made of water and ice that exhibit unusual physical and chemical properties have been initiated [9].

The purpose of this work was to measure the response of a microwave resonator to a dielectric plate with ice deposited on it in order to establish the possibility of detecting ice 0 when it forms in a cold atmosphere and to determine the degree of influence of such layers on the characteristics of the resonator at microwave frequencies. The detection of this effect is of interest for the development of electromagnetic non-contact techniques for studying the characteristics of boundary layers between ice 0 and various media.

2. Experimental

The scheme of the setup for the study is presented in Fig. 1.

In the experiment, we cooled a rectangular resonator with thin quartz glass plates installed in it. The natural frequency of the resonator is about 2.8 GHz; its linear dimensions are 72×34×77 mm; the resonator material is copper (the quality factor of the loaded resonator at room temperature is ~ 550). The thickness of quartz glass plates was 0.19 mm, their dimensions were 24×24 mm, and their number was 13 pcs. After cooling by cold nitrogen vapor to the minimum temperature, the resonator, in some cases, was pumped with a certain volume of air to introduce an additional amount of water vapor into the cavity. The minimum temperature in the cooling chamber was –140 °C.

Measurement of the characteristics of the resonator: its Q-factor (Q) and values of the resonant frequency (f_p) were performed in the mode of slow heating of the refrigerating chamber. The average heating rate of the resonator was ~ 1 °C/min. Heating was carried out when the supply of cold nitrogen vapor was stopped and the temperature in the laboratory room was stable. During this process, water vapor in the resonator condensed on the surface of quartz plates and walls of the resonator to form a thin layer of ice. In the absence of air supply to the resonator, as well as the relative humidity of air ~ 15 % at the initial temperature of 20 °C, the maximum

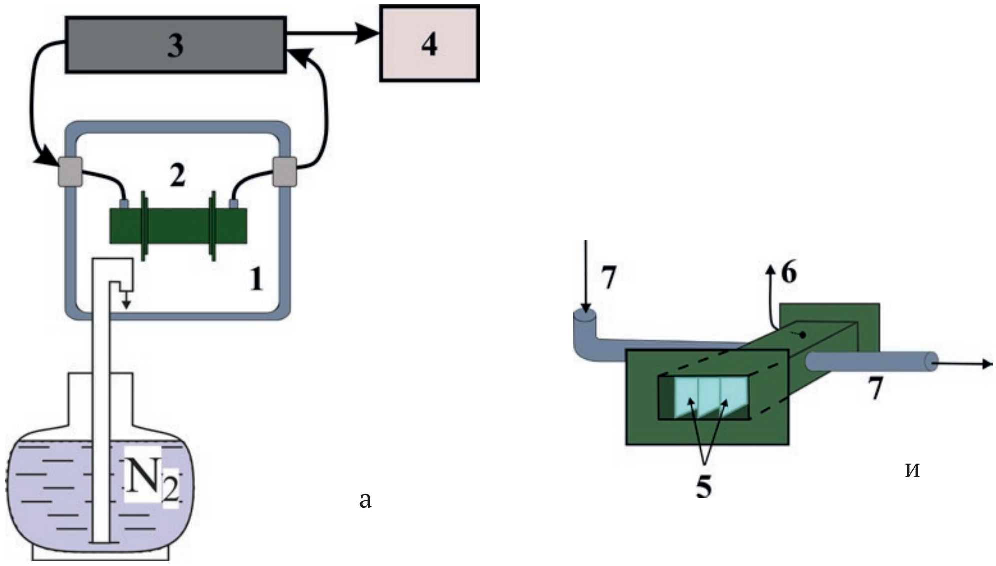


Fig. 1. a) Scheme of the installation for searching for ice films 0 during its formation in a microwave resonator: *1* – refrigerator, *2* – resonator, *3* – scalar analyzer of radio frequency circuits P4-18, *4* – data collection system; b) scheme of the resonator type H_{101}, where *5* – thin plates of quartz glass, *6* – thermocouple outlet, *7* – air pumping tubes

possible calculated thickness of the ice film was ~ 100 nm. Since the plates were cooled through the walls of the resonator, the largest mass of water condensed on them. Therefore, pumping a certain volume of air through the resonator was also used in the experiments. The mass of the incoming water vapor and the formed volume of ice, as well as the known values of the dielectric permittivity of ice Ih show that their amount for the case of hexagonal ice cannot noticeably change the characteristics of the resonator.

From measurements of the resonant response of the resonator, we found f_p, the bandwidth of its passband at the level of 0.5 dB (Δf_p), the passband power at the resonant frequency (P); Q ($Q = f_p/\Delta f_p$) was determined by calculation. One measurement was performed for a time of ~ 1 s. The total time of measurements was ~ 3 hours. Accuracy of absolute measurements of air temperature in the chamber: ~ 1 °C, amplitude of the transmitted power ~ 0.05 dB, Q-factor ~ 3. We also calculated the derivative of the measured quantities by temperature (T): dP/dT and dQ/dT.

3. Measurement results

The results of measurements of dQ/dT and dP/dT as a function of temperature during heating of the resonator from -140 °C are shown in Fig. 2. In this experiment, the deposition of gases from the primary filled and isolated from the atmosphere volume of the resonator cavity was used. The initial value of air temperature was 20 °C and its relative humidity (W) ~ 15 %. The derivatives were found for a slow heating process, which was carried out when the cooling device was switched off. This was done to obtain smooth dependences on temperature and time, since during cooling there were possible some irregularities in the temperature increment, worsening the accuracy of measurements. This procedure also allowed us to obtain a larger value of the thickness of the deposited ice layer in the temperature range of

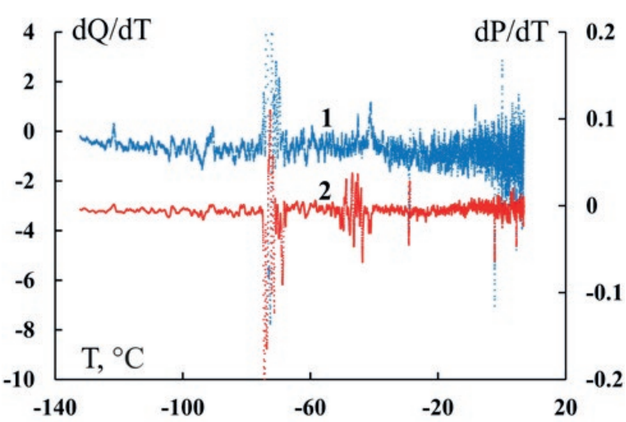


Fig. 2. The dependence of the derivatives of Q-factor (*1*) and power (*2*) of the resonator transmission at the resonant frequency on temperature. Averaging the derivative over 50 points. The total number of measured points is ~ 10^4. (*dP*/*dT* – in relative units)

interest (below -20 °C). Vapor condensation at the beginning of the experiment occurred at about -5 °C. The exact value of the saturated vapor pressure for ice 0 is unknown, but this ice could not have formed at temperatures above -23 °C.

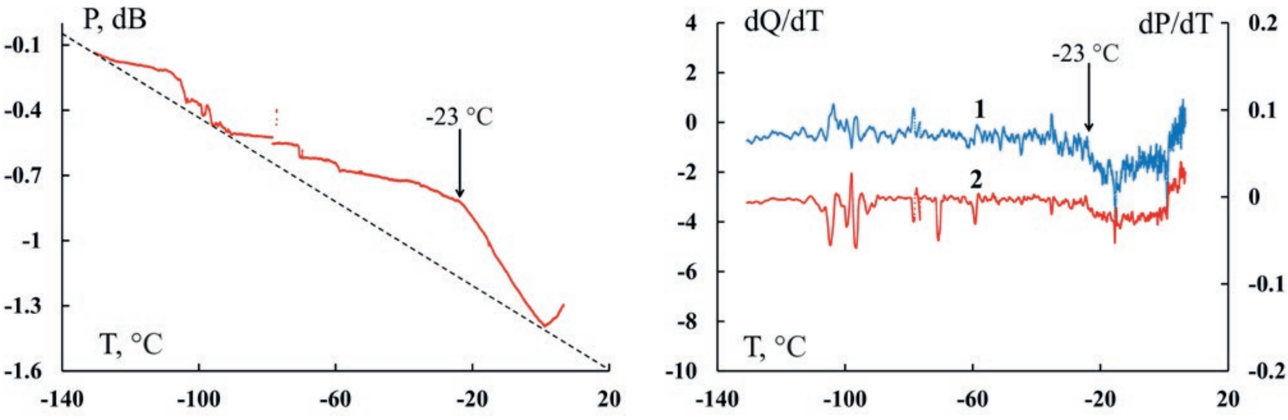
The total surface area of the metal and 13 quartz glass plates used in the experiment allowed us to obtain the maximum possible ice thickness at a water vapor mass of 5.5×10^{-4} g of about tens of nanometers with its uniform distribution on the surface of the plates.

Since, as noted above, it was expected that thinner films were deposited on the quartz plates than on the metal of the resonator walls, the experiments also included an additional air supply to the cavity. Fig. 3 shows the data for this case. At a relative humidity of 16 % (air temperature 20 °C), 3 liters of air were injected into the cavity using a compressor for a time of 1.5 min. The addition of water vapor was 0.0083 g. Additional water vapor was introduced at a temperature in the resonator of about -150 °C. The maximum calculated thickness of the ice layer on the surface was 900 nm. The calculations were performed for the known values of the mass of water vapor, the area of quartz plates and the walls of the resonator. The real value of this layer on the plates is significantly lower, since some vapors could condense in the supply tube and also escape from the waveguide cavity before settling on the plates.

To compare the data in the analysis, we measured the parameters of the resonator with plates when it was filled with nitrogen. In this case, a uniform variation of the transmittance power with temperature was observed. The approximation of $P(T)$ for the case of no gases in the resonator is shown in Fig. 3a with a dashed line.

4. Results and discussion

When performing the experiments, it was assumed that the formed ice layer 0 creates a conductive layer of nanometer thickness at the dielectric boundary [3, 4]. This will lead to a sufficiently pronounced effect of changing the goodness of the resonator and, consequently, the transmittance power even in the low-frequency region of the microwave range. In this case, the absorption effect is associated with an increase in the loss factor due to the resonance of surface plasmons in island conducting films. The thickness of about 10 nm is necessary for the occurrence of such a state of the films [7]. Therefore, it is sufficient to deposit on the surface of quartz plates layers of ice 0 with a thickness of ~ 10...100 nm for appreciable changes in the Q-factor. Since it is difficult to obtain ice 0 in the experiment, we used a technique with initial cooling of the whole system (vapor and substrate), in which some initial vapor deposition in the form



a b

Fig. 3. a) The dependence of the power transmission through the resonator on the temperature when it is heated after purging the resonator with air with an initial humidity of 16% (at an initial temperature of 20 °C). Power in decibels from the conditional level. The dashed line is the same for the case of approximating the absence of gases in the resonator; b) The dependence of the derivatives of Q (*1*) and power (*2*) of the resonator transmission at the resonant frequency on the temperature after purging the resonator with air with an initial humidity of 16 % (at an initial temperature of 20 °C). Averaging the derivative over 100 points. The total number of measured points is ~ 104. (*dP*/*dT* – in relative units)

of ice Ih in the temperature range from -5 °C and below was possible. However, ice 0 can also form on the surface of ice Ih, which was shown in our earlier experiment [10]. It can also be formed in the volume of ice Ih in the form of individual clusters, which was shown in [11].

Examination of the results of Fig. 2, which correspond to an ice layer on ~10 nm quartz glasses, reveals two features in the region slightly below –78 °C and in the –45 °C region. No clear traces associated with ice 0 are observed in this experiment. In Fig. 3, where films on the order of one hundred nanometers thick were deposited, in contrast, the temperature of –23 °C clearly stands out, where an extremum of increasing *P* compared to the calibration value was observed (dashed line for the case of no gases in the resonator). In addition, a power jump was observed at –78 °C and some increase in the –110 °C region.

The deviations of the plots from the mean value near –78 °C can be related to the solid-gas phase transition process for carbon dioxide, which is also in air and appears in the mixture with ice. The phase transition of solid carbon dioxide into gas and leads to some fluctuations of values in a small temperature range. These temperature values appear more sharply in the plots of the dQ/dT and dP/dT derivatives, Fig. 3b. This figure shows a sharp change in the slope of the derivative plots as the temperature rises above –23 °C, which corresponds to the transition of ferroelectric ice 0 to hexagonal ice Ih with the disappearance of conductive films of nanometer thickness at the boundaries of ice 0 with other dielectrics.

It is interesting to note the appearance of the resonator response for the case presented in Fig. 3, at a temperature of –105 °C (approximately –95 to –115 °C). Such a feature was also observed in the extinction plots of laser radiation when passing through ice films deposited on a dielectric, previously found in [3, 4], which, however, was not investigated.

It also turned out that in the subsequent work [12], in the study of flexoelectric phenomena – charge formation in areas with anisotropy of mechanical strain, a phase transition near –110 °C was detected in ice. It was concluded that electric polarization induced by strain anisotropy can be an effective method to detect phase transitions in surface layers; especially for ferroelectric. Our results support this conclusion.

It is worth mentioning the physical features of thin layers during gas condensation which were utilized in the technique of the performed measurements. The most important one is the detection of highly conductive layers several nanometers thick. According to papers [5, 6], such layers arise at the contacts of dielectric and ferroelectric. Consequently, using the proposed technique it is possible to detect both ultra-thin (nanometer) and thicker (micron) layers of ferroelectric materials. The temperature dependences of the resonator response determine the ferroelectric phase transition.

The phase transition near –105 °C can be associated with the formation of ice XI. This ice is formed at temperatures below –200 °C, but its nuclei remain when the samples are heated to a temperature of ~ –120 °C, as presented in the paper [13]. Since the concentration of ice XI is small at high temperatures, it has not been previously recorded by standard methods of matter structure measurements. Another important feature manifested in the method we used is the increase in the goodness of the resonator, i.e., the decrease in its losses, during the formation of ferroelectric ices. This can be explained by plasmon effects during the growth of nanometer island films and the emergence of highly conductive contact films. Such phenomena associated with increasing electromagnetic fields are studied in plasmonics and are associated with resonances of plasmonic modes [7, 14]. An example is giant Raman scattering, when the amplification of scattered fields by groups of molecules near specially fabricated structures can increase 10^{10} times [8]. The details of this phenomenon require a special study. For example, in the paper [15] they investigated the effect of percolation transition on ultra-thin island metal films and found significant changes in the resonance of surface plasmon-polaritons and, consequently, in their electrophysical properties.

4. Conclusions

It is shown that the emergence of a conducting layer of nanometer thickness on the dielectric surface is a consequence of the formation of a film of ferroelectric ice 0 on it. The occurrence of resonance of plasmon modes in the microwave range in ice films deposited in the resonator was confirmed by the change of its parameters.

In the performed experiment with a microwave resonator near the resonant frequency of 2.8 GHz, in the temperature range of 5...–140 °C, filled with quartz plates, the changes in the transmitted power at the resonant frequency and its goodness of fit were observed. The measurements revealed characteristic changes of these parameters at a temperature of –23 °C, which corresponds to the value at which the ferroelectric ice 0 is formed.

The used microwave technique for recording the formation of ferroelectric films during gas condensation is applicable to the study of the characteristics of superfine layers when their electrical conductivity changes.

Contribution of the authors

The authors contributed equally to this article.

Conflict of interests

The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

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