Effect of temperature on dielectric properties of ice in the range 5–39 GHz

Takeshi Matsuoka,[a] Shuji Fujita, and Shinji Mae
Department of Applied Physics, Faculty of Engineering, Hokkaido University, N13 W8, Sapporo 060, Japan

(Received 14 June 1996; accepted for publication 8 August 1996)

The relative complex dielectric permittivity, $\epsilon^* = \epsilon' - i\epsilon''$, of ice has been measured in the frequency range 5–39 GHz and in the temperature range 190–265 K. The cavity resonator method at 5 and 10 GHz and the open resonator method at 33 and 39 GHz were used to determine the low dielectric loss of ice. The real part of permittivity ϵ' was independent of the frequency. The temperature dependence of ϵ' was observed and is discussed in terms of contributions from anharmonic effects to infrared polarizability. The ϵ'' obtained bridges the gap of previous results between 200 and 258 K. We discuss the frequency and temperature dependence of the effect of the infrared absorption band on ϵ''. The ϵ'' variation with frequency increased as the temperature decreased at 5–39 GHz. It is possible that absorption takes place at frequencies below the infrared region. © *1996 American Institute of Physics.* [S0021-8979(96)01322-9]

I. INTRODUCTION

An understanding of the microwave remote sensing signatures of the polar environment is dependent on a knowledge of the relative complex dielectric permittivity, $\epsilon^* = \epsilon' - i\epsilon''$, of ice. So far, many studies of the dielectric properties of ice have been carried out at microwave frequencies. The results of the investigations have been reviewed by Evans[1] and by Warren.[2] The results indicate that the real part of the permittivity ϵ' is 3.17 ± 0.07, independent of frequency, and the temperature dependence is small at microwave frequencies within the estimated errors. Gough[3] and Johari[4] discussed the small temperature dependence of ϵ' from their MHz frequency data.

Further study was performed by Mätzler and Wegmüller[5] at microwave frequencies. They clarified the slight temperature dependence of ϵ' above 245 K and found that the variation with frequency is less than 0.002 between 2.4 and 9.6 GHz. Recently, the anisotropy of ϵ' was observed by Fujita, Mae, and Matsuoka at 9.7 GHz, giving values of 0.037 ± 0.007 and about $1.2\% \pm 0.2\%$.

The situation with respect to the imaginary part of permittivity ϵ'' for ice is, on the other hand, very unsatisfactory. Warren[2] reported that significant discrepancies exist in the obtained values of the imaginary part of permittivity at microwave frequencies. Generally, the values of ϵ'' are characterized by a high-frequency tail of the Debye relaxation spectrum with a relaxation frequency at kHz range and by a low-frequency tail of the infrared-absorption band. However, the reported values differ by almost half an order of magnitude and it is not even clear what functional form should be used in fitting these data.

Since it is thought that the values of ϵ'' are of order 10^{-4}, measurements with sufficient accuracy are difficult. Moreover, a small amount of impurity in ice can considerably increase ϵ'' (Mätzler and Wegmüller,[5] Fujita, Shiraishi, and Mae,[7] Matsuoka, Fujita, and Mae,[8] and Moore and Fujita[9]). Recently, Mätzler and Wegmüller[5] carried out measurements over a wide range of frequencies between 2 and 100 GHz at 258 and 268 K with the cavity resonator method and radiometric method. Their results seem to be the most reliable ones at present since they used methods suitable for precise measurement in a low-loss medium and they took into account the impurity content. Koh[10] measured ϵ'' of ice at frequencies between 26.5 and 40 GHz at 258 and 270 K with the free space method. At 250 K, Surdyk and Fujita[11] used the open resonator method and obtained ϵ'' at 32 and 39 GHz. Since the temperature range of previous studies was only near the melting point at microwave frequencies, it is necessary to clarify ϵ' and ϵ'' over a wide temperature range in the actual cryosphere (down to about 200 K).

This article reports the dielectric properties of ice using the cavity and the open resonator methods which are the precise measurement methods for low-loss medium in the frequency range 5–39 GHz and the wide temperature range 190–265 K. Then the frequency and temperature dependence of the real and imaginary parts of permittivity are discussed.

II. EXPERIMENTS

A. Cavity resonator method

Two cylindrical cavities made of Invar operating in TE_{01n} mode were used to measure the permittivity of ice at frequencies 5 and 10 GHz. The use of a cavity resonator has been described by Horner *et al.*[12] and by Saito and Kurokawa[13] in detail. The vertical arrangement of the cavity resonator is shown in Fig. 1(a). The cavities operating in the TE_{011} mode for 5 GHz and TE_{012} mode for 10 GHz were of diameters 95 and 50 mm, and silver and gold plated, respectively. The axial length was set to about 50 mm by using a movable plunger plate with micrometer for both cavities. The exciting and pickup loops were located on the opposite side from the movable plunger plate. The values of the unloaded quality factor were more than 10 000 and 20 000 for 5 and 10 GHz, respectively, making measurement of small losses of ice possible.

The real part of the permittivity ϵ' was obtained from the measured difference between the resonant axial length of the cavity with and without the disk-shaped sample. The imaginary part of the permittivity ϵ'' was determined by the fre-

---

[a] Electronic mail: matutake@huap.hokudai.ac.jp

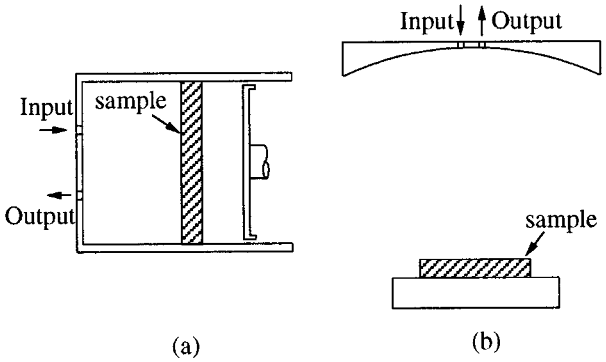

FIG. 1. Vertical arrangements of resonators: (a) cylindrical cavity resonator operating in the TE_{01n} mode at 5 and 10 GHz; (b) hemispherical open resonator operating $TEM_{p,0,q}$ mode at frequencies of 33 and 39 GHz.

quency variation method described in Saito and Kurokawa.[13] The half-power bandwidth of the resonance curves of the cavity were measured with and without the sample to obtain the ϵ'' of the sample. For the dielectric measurement of ice using the cavity resonator method, orientation of (only) the *c* axis parallel to the field is not possible because of the circular direction of the electric field lines in the cavity.

B. Open resonator method

The open resonator method has been described by Jones,[14] Cullen[15] and Komiyama, Kiyokawa, and Matsui.[16] The open resonator, which was made of brass and was gold plated, operating in the $TEM_{p,0,q}$ mode, was of the hemispherical type and used at frequencies between 30 and 40 GHz. The vertical arrangement of the open resonator is shown in Fig. 1(b). It was designed to measure disk-shaped samples with the diameter between 45 and 80 mm. The diameter and the radius of curvature of the concave mirror were, respectively, 175 and 141 mm, and the diameter of the plane mirror was 80 mm. Both mirrors were mounted on an aluminum optical rail and arranged so that the axis of the resonator was vertical. The plane mirror was mounted on a translation stage and was movable in the direction of the resonator axis. Coupling of microwave power into and out of the resonator was done by two 1.5-mm-diam holes with a 5.6 mm center-to-center separation situated symmetrically about the center of the concave mirror. In the fundamental $TEM_{0,0,q}$ mode, *Q* values of the empty resonator were about 90 000. In this study, the frequency variation method was used.

The permittivity of the sample is obtained by measuring the resonant frequency and *Q* value of the resonator with and without the sample. For the measurement, we used modes between $TEM_{0,0,24}$ and $TEM_{0,0,34}$. The corresponding resonant frequencies for the empty resonator were between 28.9 and 40.3 GHz. The intensity of the resonant peaks were different from each other among all 11 modes. Only resonant data which have signal/noise ratio more than 10 dB were used for the analysis of *Q* values. $TEM_{0,0,28}$ mode (at about 33 GHz) and $TEM_{0,0,34}$ mode (at about 39 GHz) always satisfied this condition. Single-crystal ice with the *c* axis perpendicular to the electric field was measured in this open resonator method. Polycrystalline ice was not used in this method, since we cannot treat polycrystalline ice as uniform ice in this frequency range because of scattering loss at the grain boundaries in polycrystalline ice.

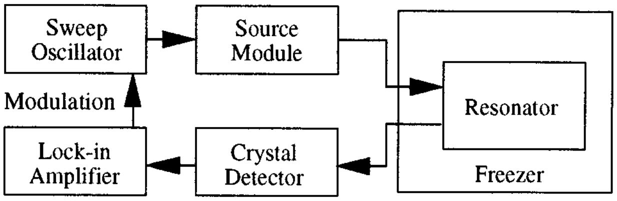

FIG. 2. Experimental setup for measuring the permittivity of ice by resonator methods. The source module is used only for the open resonator measurement.

C. Experimental system and procedure

The experimental setup is illustrated in Fig. 2. Source signals for the cavity resonator measurements were generated using a HP 83623A synthesized sweeper, and a sweeper with HP 83554A millimeter-wave source module was used for the open resonator measurements. The resonant signals were detected by a crystal detector and amplified by NF 5610B lock-in amplifier. All instruments were controlled by a computer. The resonator was set in the freezer, and temperature was measured by a calibrated thermocouple and was accurate to within ±0.2 K. The measured temperature range was between 190 and 268 K for the cavity resonator method, and between 190 and 265 K for the open resonator method. We investigated the temperature coefficient of thermal expansion for the axial length and the diameter or the radius of curvature of the concave mirror of empty resonators before the measurement of ice permittivity.

D. Preparation of ice samples

Polycrystalline ice was made by freezing distilled and de-ionized water in a glass container with diameter of 100 mm. The growth rates of the ice samples were approximately 10 mm per day. The samples were cut from bulk to be free of air bubbles and formed into cylindrical disks. The grain size in the sample was about 5–10 mm and the *c* axes were randomly oriented. The diameters of samples were 95 and 49 mm at 5 and 10 GHz, respectively, equal to the inner diameters of cavities.

Single-crystal ice was collected from Mendenhall Glacier, Alaska. We prepared samples with the *c* axis perpendicular to the cylindrical disk surface. Since the diameters of shaped samples were between 45 and 57 mm, single-crystal samples were used for the cavity resonator at 10 GHz and the open resonator at 33 and 39 GHz.

The thicknesses of all samples were between 2.7 and 5.9 mm. The standard deviation of each thickness of sample was within ±0.2%. The impurity concentration of polycrystalline ice was checked by measuring the conductivity of the melted sample after experimentation. The conductivities of the samples were around 1×10^{-4} S m^{-1}. The amount of impurity ions in ice from Mendenhall Glacier was about 0.1 ppm

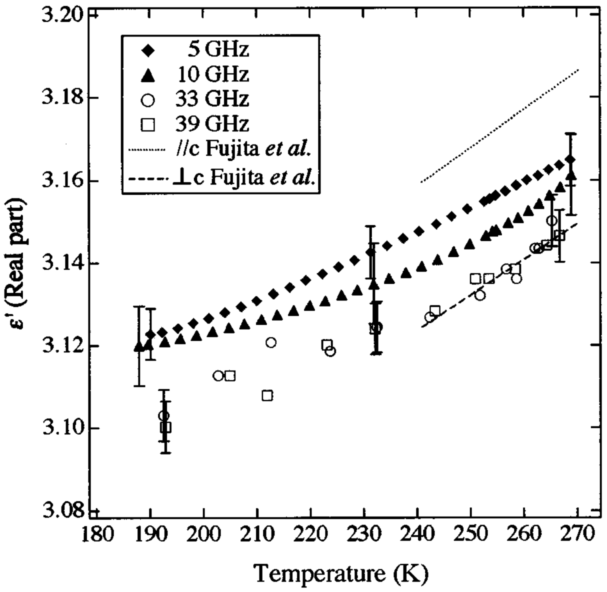

FIG. 3. The temperature dependence of ϵ' of ice at 5–39 GHz. The solid symbols represent the measurements of polycrystalline ice at 5 and 10 GHz and the open symbols represent measurement of single-crystal ice with the *c* axis perpendicular to the electric field at 33 and 39 GHz. The error bars are only shown at three temperature points. The dotted line and dashed line are the ϵ' of parallel and perpendicular to the *c* axis in Fujita and co-workers (Ref. 6), respectively.

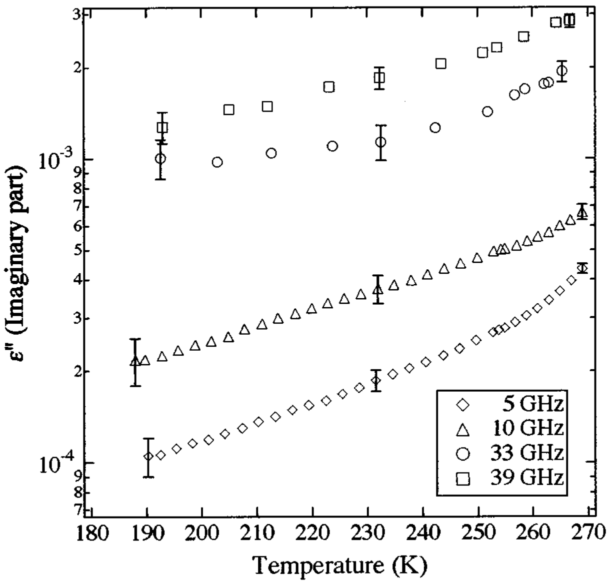

FIG. 4. The temperature dependence of ϵ'' with frequency as a parameter. The estimated errors in ϵ'' of each resonator measurements are also presented. The error bars are only shown at three temperature points.

and mainly consisted of Cl$^-$ ions (Fujita and co-workers[6]). According to the microwave dielectric properties of ice containing salt and acid impurities (Mätzler and Wegmüller[5] and Moore and Fujita[9]), these small amounts of impurities do not significantly affect dielectric properties.

E. Measurement reference

As reference materials, Teflon (PTFE) and crystal quartz were used for precise measurement of ϵ' and ϵ'' at 293 K. The measured ϵ' of the Teflon was 2.060 and the standard deviation was ±0.005 ($\pm0.25\%$) at 5 and 10 GHz. The ϵ' of the crystal quartz with the *c* axis parallel and perpendicular to the electric field were 4.607 and 4.406, respectively, with the open resonator in the range 30–40 GHz. The standard deviation was minimized by using the analysis method in Komiyama and co-workers[16] and found to be ±0.004 ($\pm0.09\%$). The obtained ϵ' of the Teflon and the crystal quartz had good agreement with previous reported values.

The ϵ'' of the Teflon and the crystal quartz are known to be as small as 1×10^{-4} (Jones[14]) and 2.5×10^{-4} (Afsar, Li, and Chi[17]), respectively. In this study, the measured ϵ'' of the Teflon was $(1.5\pm0.1)\times10^{-4}$ with the cavity resonator at 5 and 10 GHz, and ϵ'' of the crystal quartz (perpendicular to the *c* axis) was $(2.1\pm0.4)\times10^{-4}$ with the open resonator at 30–40 GHz. These results show that cavity and open resonator systems are suitable for measuring the permittivity of low-loss samples such as ice.

III. RESULTS

A. The real part of permittivity

In Fig. 3 the real part of permittivity ϵ' of ice is plotted versus temperature with frequency as a parameter. ϵ' was measured over 30 temperature points at 5 and 10 GHz, and over 10 points at 33 and 39 GHz in the range 190–265 K. Each result of the cavity resonator method at 5 and 10 GHz is an average for three polycrystalline ice samples. The standard deviations of the ϵ' values were ±0.006 ($\pm0.2\%$) at 5 GHz and ±0.009 ($\pm0.3\%$) at 10 GHz. The major sources of variability is the small uncertainty of the sample position in the cavity. Anisotropy was not detected by comparison between the ϵ' values of polycrystalline ice and of single crystal ice with the *c* axis perpendicular to the electric field at 10 GHz. Results of the open resonator method at 33 and 39 GHz show the ϵ' values of a single-crystal sample measured perpendicular to the *c* axis. For the open resonator measurement, the estimated errors in ϵ' value were ±0.006 ($\pm0.2\%$) due to decline in the signal/noise ratio of intensity of the resonant peaks at temperature below 230 K.

Figure 3 also shows linear approximations for temperature dependence of ϵ' obtained by Fujita and co-workers[6] at 9.7 GHz in the temperature range 240–268 K. Fujita and co-workers[6] clarified the anisotropy of ϵ' in ice; the reported value was 0.037 ± 0.007 and about $1.2\%\pm0.2\%$. The average ϵ' values of polycrystalline ice in this study are between the ϵ' values parallel and perpendicular to the *c* axis at temperature above 240 K reported by Fujita and co-workers.[6] Furthermore, in this study, as shown in Fig. 3, the ϵ' values of ice and the rate of increase, $(d\epsilon'/dT)$, increase with temperature for all frequencies in the temperature range from 190 to 265 K. The absolute ϵ' values were consistent with each other in the frequency range of this study, considering the condition that we obtained ϵ' values for electric field perpendicular to the *c* axis at 33 and 39 GHz.

B. The imaginary part of permittivity

The ϵ'' of ice versus temperature is shown in Fig. 4 with

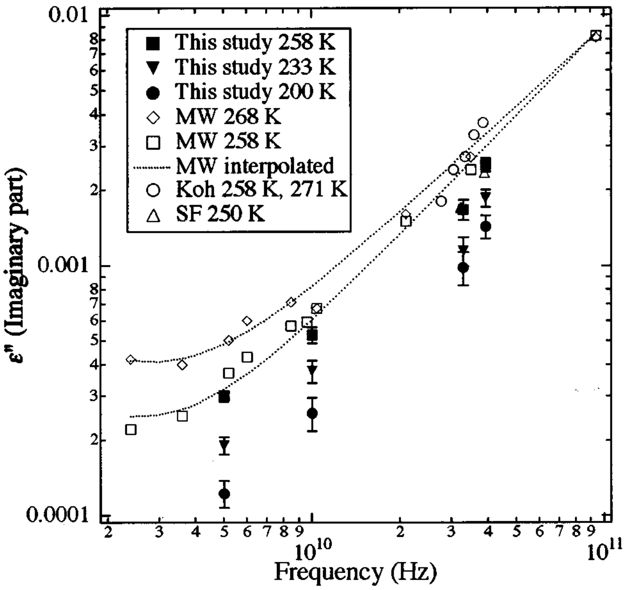

FIG. 5. The ϵ'' variation with frequency with temperature as a parameter. Comparison of ϵ'' observed in this study with previous results [Mätzler and Wegmüller (Ref. 5) (MW), Koh (Ref. 10), and Surdyk and Fujita (Ref. 11) (SF)].

frequency as a parameter. The estimated errors in ϵ'' of the cavity resonator measurement at 5 and 10 GHz were $\pm 1.5\times 10^{-5}$ and $\pm 0.4\times 10^{-4}$, respectively. For the open resonator measurement, the estimated error in ϵ'' at 33 and 39 GHz was $\pm 1.5\times 10^{-4}$. There was no significant difference in the ϵ'' values of the polycrystalline ice and the single-crystal ice measured perpendicular to the c-axis at 10 GHz within the estimated error. Thus, it is suggested that the scattering loss at the grain boundaries in polycrystalline ice is small enough to permit polycrystalline ice to be treated as uniform ice below 10 GHz.

The ϵ'' variations with temperature at each frequency are almost straight lines below 250 K, plotted in semilogarithmic representation, as shown in Fig. 4. At high temperature above 250 K and low frequency such as 5 GHz, it seems that the measured values of ϵ'' are larger than the values of the straight line. It can be explained that the increase is due to the effect of the high-frequency tail of the Debye relaxation spectrum.

Figure 5 shows the ϵ'' variation with frequency of this study with temperature as a parameter. Results are only shown at three temperature points in the measured range 190–265 K. Figure 5 also shows the results of Surdyk and Fujita[11] at 250 K using the same open resonator that is used in this study at 33 and 39 GHz, the measured and interpolated values of Mätzler and Wegmüller[5] using cavity resonator and radiometric measurement in the range 2–100 GHz at 258 and 268 K, and the results of Koh[10] using free-space measurement in the range 26.5–40 GHz at 258 and 271 K. The ϵ'' variation with frequency observed in this study tends to agree well with the previous results. However, at higher frequencies above 33 GHz, the absolute values of ϵ'' in Mätzler and Wegmüller[5] and Koh[10] are larger than in this study at 258 K.

IV. DISCUSSION

A. The absolute value and frequency dependence of ϵ'

As shown in Fig. 3, the ϵ' values of all samples in this study are in the range 3.17 ± 0.07 which is described in the review of Evans.[1] The absolute ϵ' value of the polycrystalline ice at 258 K is 3.154 ± 0.011 in this study, which agrees very well with the results of Cumming[18] at 9.4 GHz, Koh[10] at 26.5–40 GHz, and the estimated value from single-crystal data of Fujita and co-workers[6] at 9.7 GHz. Mätzler and Wegmüller[5] reported approximately 0.02 higher values in their experiments. Since four independent measurements show similar values, we treat our result as the correct absolute ϵ' value at microwave and compare it with the reported MHz range data in the following discussion.

The absolute ϵ' values for polycrystalline ice of Johari and Charette[19] at 35 and 60 MHz and Johari[4] at 100 MHz are 0.010–0.035 higher than that for polycrystalline ice at 5 and 10 GHz in this study. Johari[4] suggested that a minor relaxation process exists at MHz frequencies and is due to the orientation of water molecules at the grain boundaries or at the imperfection sites which required less energy for reorientation. In this sense a decrease in ϵ' value with frequency can be explained. The difference of ϵ' value between 35 MHz and 5 GHz is 0.030 ± 0.009 in the temperature range 250–265 K.

B. The temperature dependence of ϵ'

Johari and Jones[20] discussed the temperature dependence of ϵ' of the polycrystalline ice in terms of contributions from anharmonic effects to molecular polarizability α. They treated the polycrystalline ice of isotropically polarizable material. Then α is given by the Clausius–Mosotti relationship

$$\frac{N_i}{3}\,\alpha=\left(\frac{\epsilon'-1}{\epsilon'+2}\right), \qquad (1)$$

where N_i is number of molecules per unit volume calculated for a density of 0.9169 $\mathrm{Mg\,m^{-3}}$ at 270 K, taking into consideration the regression line of average expansivity derived from previous work. α is the sum of the electronic polarizability α_{el} and the infrared polarizability α_{IR} caused by the displacement, or vibration, of molecules within the lattice. Since $\alpha_{\mathrm{el}}(=18.85\ \text{Å}^3)$ is independent of the temperature, it appears that α_{IR} increases with temperature. Johari and Jones[20] have found that α_{IR} in polycrystalline ice increases linearly with the square of temperature, as is shown in Fig. 6, according to the equation

$$\alpha_{\mathrm{IR}}=\alpha_{0,\mathrm{IR}}+aT^2, \qquad (2)$$

where $\alpha_{0,\mathrm{IR}}$ is the infrared polarizability at 0 K and a is an empirical constant. The values of $\alpha_{0,\mathrm{IR}}$ and a obtained by Johari and Jones[20] were 20.61 Å^3 and $25.51\times 10^{-6}\ \text{Å}^3\,\mathrm{K}^{-2}$, respectively. The calculated α_{IR} values from ϵ' in this study at 5 and 10 GHz are also plotted against the square of temperature in Fig. 6. The derived $\alpha_{0,\mathrm{IR}}$ and a were 20.46 Å^3 and $20.94\times 10^{-6}\ \text{Å}^3\,\mathrm{K}^{-2}$ for 5 GHz and 20.47 Å^3 and $19.44\times 10^{-6}\ \text{Å}^3\,\mathrm{K}^{-2}$ for 10 GHz. There are good agreements

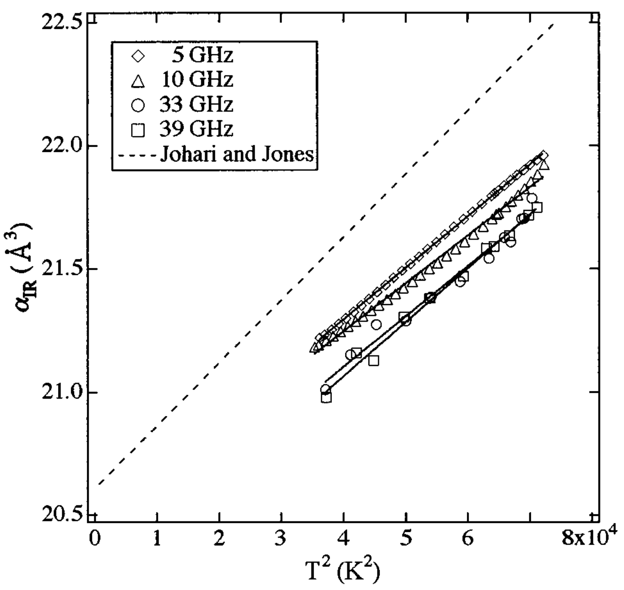

FIG. 6. The infrared polarizability α_{IR} of ice plotted against the square of temperature. The dashed line is fitted line of Johari and Jones (Ref. 20) and the solid lines represent fitted lines at 5, 10, 33, and 39 GHz in this study.

of $\alpha_{0,IR}$ and a values between 5 and 10 GHz within the standard deviation ($\pm 0.5\%$ for $\alpha_{0,IR}$, $\pm 5.6\%$ for a). In addition, Fig. 6, which represents the results for single-crystal ice at 33 and 39 GHz, shows that a single crystal can be treated as isotropic ice as far as the temperature change is concerned. The a values were 20.43×10^{-6} and 21.86×10^{-6} Å^3 K^{-2} at 33 and 39 GHz, respectively. The average a value is 20.67×10^{-6} in the range 5–39 GHz and about 20% lower than that for the MHz range in Johari and Jones.[20] Since the increase in a value is due to the effects of the orientation polarization at high temperature in MHz frequency, it seems that the a values of microwave data represent more accurate infrared effects than MHz data.

C. The frequency and temperature dependence ϵ''

Although minor relaxations which exist between Debye relaxation and the infrared-absorption band have been reported by several investigators (Gough and Davidson,[21] Gough,[3] von Hippel, Knoll, and Westphal,[22] Johari,[4] and Johari and Whalley[23]), the observed relaxation frequencies were contradictory to each other without clear detectability. Therefore, the simple suitable model for the frequency dependence of ϵ'' at microwave frequencies can be expressed by the sum of two components. One is inversely proportional to frequency for the high-frequency tail of the Debye relaxation spectrum and the other is proportional to the frequency for the low-frequency tail of the infrared-absorption band. Mätzler and Wegmüller[5] obtained the equation from fitting of their experimental data, written as follows:

$$\epsilon'' = (A/f) + Bf^{C}, \tag{3}$$

where the coefficients A, B and C are temperature-dependent empirical constants and f is the frequency in GHz. The first term represents the effect of Debye relaxation and the second term corresponds to the effect of infrared absorption.

TABLE I. The coefficient A calculated only from the component of the main Debye relaxation (D) and from the data of Johari and Charette[a] (JC) that contain the possible additional component.

Temperature (K)	$A_D\times10^4$	$A_{JC}\times10^4$
190	0.005	
200	0.010	
220	0.031	
240	0.268	
248	0.635	1.870
253	1.059	2.222
258	1.728	3.091
263	2.769	4.591
265	3.326	5.693[b]

[a] Reference 19.
[b] Interpolated value between 263 and 268 K.

The temperature dependence of ϵ'' was not discussed in detail by previous investigators. Walford[24] suggested that the infrared-absorption band showed no temperature dependence. Mätzler and Wegmüller[5] derived the coefficients A, B, and C in Eq. (3) only at temperatures of 258 and 268 K. The results of our measurements over a wide temperature range show precisely the ϵ'' change with temperature in the range 190–265 K, as shown in Fig. 4.

Based on Eq. (3), we discuss the frequency and temperature dependence of the effect of infrared-absorption band in ϵ'' considering the effect of the Debye relaxation spectrum reported by previous investigators. First we derive the coefficient A considering only the component of the main Debye dispersion, and then we investigate the possible effect of the minor dispersion which was detected by Johari and Charette.[19] As a result, we can assess whether the latter component is important or not.

Possible effects of the orientational polarization are the main Debye dispersion in the kHz range and minor dispersion in the MHz range. The temperature dependence of the Debye relaxation spectrum below the kHz range has been discussed in several papers (Wörz and Cole,[25] Gough and Davidson,[21] Kawada,[26] Johari and Jones,[27] and Johari and Whalley[23]). Therefore, the temperature dependence of A in Eq. (3), extrapolated from the Debye equation, is represented as follows:

$$A(T) = (\epsilon_0' - \epsilon_\infty')f_r, \tag{4}$$

TABLE II. The fitted values of B and C and their standard deviations obtained from our results of ϵ'', taking into account the coefficient A shown in Table I and considering the weighting of standard deviation of the ϵ'' values at each frequency.

Temperature (K)	$B_D\times10^5$	$B_{JC}\times10^5$	C_D	C_{JC}
190	1.537±0.391		1.175±0.086	
200	1.747±0.396		1.168±0.077	
220	2.469±0.429		1.129±0.060	
240	3.495±0.468		1.088±0.047	
248	4.006±0.483	3.330±0.433	1.073±0.043	1.125±0.045
253	4.380±0.494	3.725±0.448	1.062±0.040	1.108±0.042
258	4.696±0.500	3.924±0.449	1.056±0.038	1.107±0.040
263	5.277±0.520	4.220±0.455	1.038±0.036	1.101±0.038
265	5.646±0.535	4.396±0.460	1.024±0.035	1.096±0.037

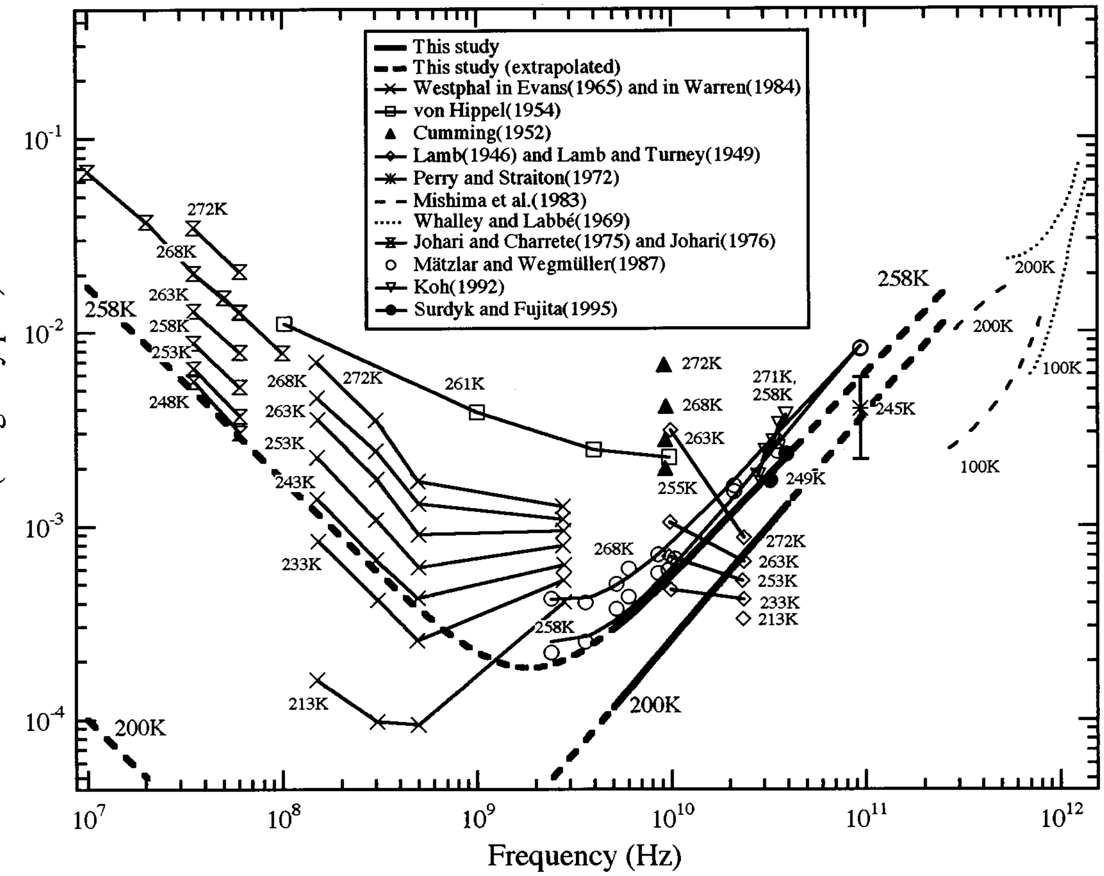

FIG. 7. The ϵ'' plotted logarithmically against frequency with temperature as a parameter. The bold lines represent fitted lines in this study with the parameters $B=1.747\times10^{-5}$, $C=1.168$ and $B=4.696\times10^{-5}$, $C=1.056$ at 200 and 258 K, respectively. The previous reported values are from Warren (Ref. 2).

where $(\epsilon_0' - \epsilon_\infty')$ is the difference of the static and high-frequency permittivity and is expressed as

$$(\epsilon_0' - \epsilon_\infty') = \beta/(T - T_0). \tag{5}$$

Based on the data in Kawada[26] over a wide temperature range from 123 to 273 K, the constant β was 23 700 and the Curie–Weiss temperature T_0, was 15 K (approximate value estimated from the permittivities parallel and perpendicular to the c axis). f_r is the relaxation frequency, expressed as

$$f_r = [2\pi\tau_0 \exp(-E/RT)]^{-1}, \tag{6}$$

with $\tau_0 = 5.3\times10^{-16}$ s. The activation energies E above and below 223 K were, respectively, 55.3 kJ mol^{-1} (0.57 eV) and 22.6 kJ mol^{-1} (0.23 eV). R is the gas constant. The derived values of A at integral temperatures between 190 and 265 K are given in Table I as A_{D}. The extrapolated A values from experimental results of Johari and Charette[19] and Johari[4] at MHz frequencies are larger than the values derived from Eq. (4) (the results of Westphal cited in Evans[1] and in Warren[2] also showed a similar tendency, but the detailed data were unpublished). Therefore, the A values estimated from Johari and Charette[19] at 35 and 60 MHz in the temperature range 248–265 K are also given in Table I as A_{JC}.

The effects of the infrared-absorption band are obtained from the A values described above. The temperature dependence of the low-frequency tail of the infrared-absorption band, B and C, is determined from the difference between the ϵ'' obtained in this study and A_{D} in the temperature range 190–265 K for the main Debye relaxation, or A_{JC} in the range 248–265 K for the minor relaxation reported in Johari and Charette.[19] In Table II the fitted B and C values are calculated from four frequency points considering the weighting of standard deviation of the ϵ'' values at each frequency. In the frequency range 5–39 GHz, significant differences exist between B_{D} and B_{JC} above 263 K. Therefore the minor relaxation in the MHz range must be considered to determine the coefficients B and C at high temperature above 263 K. However, B_{D} and C_{D} agree well with B_{JC} and C_{JC}, respectively, below 263 K, within the standard deviations.

The coefficients B and C, were obtained by Mätzler and Wegmüller[5] at 258 and 268 K. B values obtained by Mätzler and Wegmüller[5] were 3.6×10^{-5} and 6.5×10^{-5}, and C values were 1.20 and 1.07 at 258 and 268 K, respectively. At 258 K B values obtained by Mätzler and Wegmüller[5] agree with B_{JC}. C obtained by Mätzler and Wegmüller[5] is larger than the results obtained in this study. Moreover, C obtained by Koh[10] is larger than C obtained by Mätzler and Wegmüller.[5] Considering that Mätzler and Wegmüller and Koh used polycrystalline ice and not single crystals, it is plausible that the difference (large C values) is due to the scattering loss effect at grain boundaries in the polycrystalline ice samples, qualitatively. However, additional experiments are necessary to clarify the frequency dependence of the scattering loss effect.

The measurement of the far-infrared tail of the absorption band was performed by Mishima, Klug, and Whalley[28] in the temperature range 80–200 K. At 200 K, B derived from difference band theory by Mishima and co-workers[28] is 1.669×10^{-5} and agrees well with B_D. Although the C values in Table II increase as temperature decreases and the value is 1.168 at 200 K at microwave frequencies, the C value derived in Mishima and co-workers[28] was independent of temperature and was unity ($C=1$). Although it is not clear why C values increase as temperature decreases from this study, it might be considered that absorption takes place at frequencies below the infrared region.

ϵ'' is calculated as a function of frequency from 10^7 to 2×10^{11} Hz and temperature from 190 to 265 K using Eq. (3) and the coefficients A_D, B_D, and C_D. The derived ϵ'' only at 200 and 258 K are shown in Fig. 7, together with the previous measured values. The fitted curves derived in this study are useful for analysis of microwave remote sensing data of ice and snow in the temperature range 190–265 K.

V. CONCLUSIONS

The temperature dependence of the relative complex permittivity of ice has been measured over a wide temperature range from 190 to 265 K using cavity resonators at 5 and 10 GHz and an open resonator at 33 and 39 GHz. The real part of permittivity ϵ' in this study was independent of frequency and the values agree with those of Cumming,[18] Koh,[10] and the precise measurement of Fujita and co-workers[6] within their values of anisotropy. The absolute values of ϵ' at microwave frequencies were smaller than MHz data, suggesting the existence of minor relaxation at MHz frequencies. The temperature dependence of ϵ' was observed and discussed in terms of contributions from anharmonic effects on infrared polarizability.

The frequency dependence of the imaginary part of permittivity ϵ'' in this study tended to agree with previous results; however, at frequency above 33 GHz, the existence of scattering loss should be investigated for polycrystalline ice samples. In the frequency range 5–39 GHz, the frequency dependence of ϵ'' was mainly characterized as the low-frequency tail of the infrared-absorption band due to the lattice vibrations of ice. The temperature dependence of ϵ'' was observed and the ϵ'' variation with frequency increased as temperature decreased at 5–39 GHz.

ϵ'' is described as a function of frequency and temperature. The effect of the infrared-absorption band, which is frequency and temperature dependent, has been compared to the results of Mätzler and Wegmüller[5] at 258 K and the extrapolated values of far-infrared measurements in Mishima and co-workers[28] at 200 K. In the frequency range 5–39 GHz the ϵ'' values in this study bridged the gap of previous results between 200 and 258 K. Therefore, the derived fitted curves for frequency and temperature dependence of ϵ'' are useful for analysis of the microwave remote sensing data of the cryosphere. Additional experiments are necessary to clarify whether other effects of the infrared absorption band exist below far-infrared frequencies.

ACKNOWLEDGMENTS

This work has been supported by a Grant-in-Aid for Scientific Research and a Grant-in-Aid for JSPS (Japan Society for the Promotion of Science) fellows.

[1] S. Evans, J. Glaciol. **5**, 773 (1965).
[2] S. G. Warren, Appl. Opt. **23**, 1206 (1984).
[3] S. R. Gough, Can. J. Chem. **50**, 3046 (1972).
[4] G. P. Johari, J. Chem. Phys. **64**, 3998 (1976).
[5] C. Mätzler and U. Wegmüller, J. Phys. D **20**, 1623 (1987); **21**, 1660(E) (1988)
[6] S. Fujita, S. Mae, and T. Matsuoka, Ann. Glaciol. **17**, 276 (1993).
[7] S. Fujita, M. Shiraishi, and S. Mae, IEEE Trans. Geosci. Remote Sens. **GRS-30**, 799 (1992).
[8] T. Matsuoka, S. Fujita, and S. Mae, Proc. NIPR Symp. Polar Meteorol. Glaciol. **7**, 33 (1993).
[9] J. Moore and S. Fujita, J. Geophys. Res. **98**, 9769 (1993).
[10] G. Koh, J. Appl. Phys. **71**, 5119 (1992).
[11] S. Surdyk and S. Fujita, Geophys. Res. Lett. **22**, 965 (1995).
[12] F. Horner, T. A. Taylor, R. Dunsmuir, J. Lamb, and W. Jackson, Proc. IEE (London) **93**, part III, 53 (1946).
[13] S. Saito and K. Kurokawa, Proc. IRE **44**, 35 (1956).
[14] R. G. Jones, Proc. IEE **123**, 285 (1976).
[15] A. L. Cullen, *Infrared and Millimeter Waves* (Academic, New York, 1983), Vol. 10, p. 233.
[16] B. Komiyama, M. Kiyokawa, and T. Matsui, IEEE Trans. Microwave Theory Tech. **MTT-39**, 1792 (1991).
[17] M. N. Afsar, X. Li, and H. Chi, IEEE Trans. Microwave Theory Tech. **MTT-38**, 1845 (1990).
[18] W. A. Cumming, J. Appl. Phys. **23**, 768 (1975).
[19] G. P. Johari and P. A. Charette, J. Glaciol. **14**, 293 (1975).
[20] G. P. Johari and S. J. Jones, Nature (London) **263**, 672 (1976).
[21] S. R. Gough and D. W. Davidson, J. Chem. Phys. **52**, 5442 (1970).
[22] A. von Hippel, D. B. Knoll, and W. B. Westphal, J. Chem. Phys. **54**, 134 (1971).
[23] G. P. Johari and E. Whalley, J. Chem. Phys. **75**, 1333 (1981).
[24] M. E. R. Walford, J. Glaciol. **7**, 89 (1968).
[25] O. Wörz and R. H. Cole, J. Chem. Phys. **51**, 1546 (1969).
[26] S. Kawada, J. Phys. Soc. Jpn. **44**, 1881 (1978).
[27] G. P. Johari and S. J. Jones, J. Glaciol. **21**, 259 (1978).
[28] O. Mishima, D. D. Klug, and E. Whalley, J. Chem. Phys. **78**, 6399 (1983).