

# The low-frequency conductivity of snow near the melting temperature

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**ABSTRACT.** Dielectric measurements of snow were carried out in the temperature range  $-15^{\circ}$  to  $0^{\circ}\text{C}$  and in the frequency range 50 Hz to 5 MHz. The snow samples (about  $400\text{ kg m}^{-3}$  density) used were stored snow (average particle size: 2 mm) and hoar-frost (particle size:  $<1$  to 5 mm). The frequency characteristics of dielectric parameters showed a dielectric dispersion (Davidson–Cole type) around 30 kHz and a low-frequency dielectric dispersion (Cole–Cole circular law type). The a.c. conductivity showed a dielectric dispersion around 30 kHz and two characteristic constant values in the frequency ranges above 1 MHz and below 100 Hz (the high-frequency conductivity  $\sigma_{\infty}$  and the low-frequency conductivity  $\sigma_{\text{LOW}}$ ). The low-frequency conductivity  $\sigma_{\text{LOW}}$  showed a peak at about  $-2^{\circ}\text{C}$ . This behavior has never been noted by previous researchers. The  $\sigma_{\text{LOW}}$  showed an activation energy of about 1 eV below  $-5^{\circ}\text{C}$ . This means that the  $\sigma_{\text{LOW}}$  is mainly caused by a surface conduction. The activation energy increased with increasing temperature above  $-5^{\circ}\text{C}$ . This means that the  $\sigma_{\text{LOW}}$  in this temperature range is affected by the quasi-liquid layer on ice surfaces. The  $\sigma_{\text{LOW}}$  above  $-2^{\circ}\text{C}$  decreased with increasing temperature. The apparently curious behavior near the melting temperature is attributed to the numerous free ice surfaces within the porous snow. This conclusion was reached because our measurements without the free ice surfaces showed no such conductivity peaks for solid polycrystalline ice samples and for snow samples soaked with kerosene in the cooling process.

## INTRODUCTION

There have been many investigations of static permittivity in relation to the texture of dry snow as modeled in a mixed dielectric medium (ice and air) (Kuroiwa, 1962; Denoth, 1985), of high-frequency permittivity for estimating water content in wet snow (Denoth and others, 1984; Tiuri and others, 1984), of high-frequency permittivity for estimating the density of dry snow (Glen and Paren, 1975) and of dielectric dispersion for revealing the basic properties of snow (Yosida and others, 1958; Keeler, 1969; Maeno, 1978; Paren and Glen, 1978). However, natural snow shows metamorphic processes after falling and depositing (Colbeck, 1982), which involve changes of snow particle shapes, bonds between particles, and density. These proceed by the movement of water molecules on surfaces of particles and through pores between particles and by rearrangement of the snow particles. The dielectric properties of snow thus change with time (Kuroiwa, 1962; Paren and Glen, 1978; Fujino and others, 1985), though the separation and explanation of these changes can be realized only by careful measurements and analyses.

In this paper we report an apparently curious behavior observed in the temperature dependence of a low-frequency dielectric conductivity of snow near the melting temperature. The behavior has never been noted by previous researchers, but we believe that it indicates important changes of the texture of snow near the melting temperature.

## EXPERIMENTAL

For our measurements, we used samples of natural snow stored in a freezer at  $-20^{\circ}\text{C}$  after collection, and artificial hoar-frost grown in a freezer. Additional measurements were made with samples of polycrystalline ice and snow soaked with kerosene. We prepared samples by loosening a snow block and packing it into a sample cell (100 mm diameter and 18 mm depth, made of a polystyrene Petri dish). The sample cell has a main electrode 54 mm in diameter, a guard ring 20 mm wide and a counter electrode 96 mm in diameter. The electrodes of the sample cell were plated with gold. Samples prepared were 100 mm in diameter and about 10 mm thick, and were annealed near the melting temperature ( $-2^{\circ}$  or  $-1^{\circ}\text{C}$ ) for 1 or 2 weeks.

The measuring device consists of an outer vessel (180 mm diameter and 180 mm high, stainless steel) and an inner vessel (120 mm diameter and 120 mm high, stainless steel). The sample cell is put into the inner vessel which is wound with a heater wire for temperature controls. The device is set in a freezer controlled at  $-15 \pm 1^{\circ}\text{C}$ . The temperature of the inner vessel is controlled to  $\pm 0.01^{\circ}\text{C}$ . The sample's temperature depends on the temperature of the inner vessel. The measuring system consists of an LCR meter (Hioki, 3531) for dielectric measurements, and a digital voltmeter (Hewlett-Packard, 3478A) for temperature measurements connected to a computer (NEC, PC9801) for controlling and recording using a General-Purpose Interface Bus. Measurements were carried out for frequencies between 50 Hz and 5 MHz and temperatures between  $-15^{\circ}$  and  $0^{\circ}\text{C}$ .

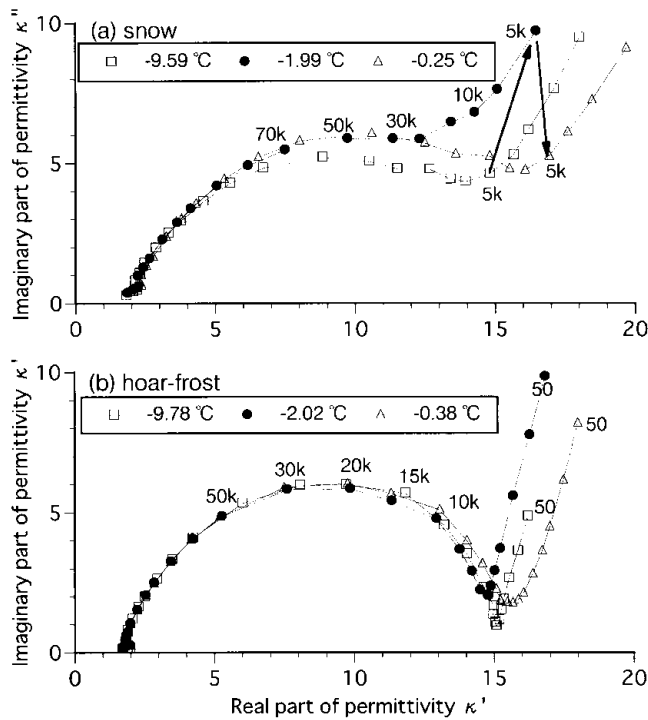


Fig. 1. Cole–Cole plots of (a) snow at  $-9.59^\circ$ ,  $-1.99^\circ$  and  $-0.25^\circ\text{C}$  and (b) hoar-frost at  $-9.78^\circ$ ,  $-2.02^\circ$  and  $-0.38^\circ\text{C}$ . Numbers of the data points are the measuring frequencies (Hz). Arrows in (a) indicate the dependence of  $\kappa''$  on temperature from  $-9.59^\circ$  to  $-0.25^\circ\text{C}$  at 5 kHz.

The error of the LCR meter is estimated to be less than  $\pm 2\%$  in the frequency range 100 Hz to 1 MHz;  $\pm 3.5\%$  below 100 Hz; and  $\pm 7\%$  above 1 MHz. Because the samples are large (100 mm diameter, 10 mm thick), it took them 30 min to reach equilibrium at a new temperature. The setting temperature of the cell was changed every 2 hours in both heating and cooling processes, and the dielectric measurements were performed just before the temperature changed. Each dielectric measurement took a few minutes and was made every 10 min.

## RESULTS

The dielectric properties of samples change with time after preparation, due to changes of the texture of the samples with time. For the purpose of stable measurements, our snow sample ( $400\text{ kg m}^{-3}$  density, 2 mm average particle size) was kept at  $-2^\circ\text{C}$  for 9 days, and the hoar-frost sample ( $390\text{ kg m}^{-3}$ , <1 to 5 mm) at  $-1.2^\circ\text{C}$  for 7 days. The relaxation time of the dielectric dispersion around 30 kHz tended to be shorter, and the dispersion strength (static dielectric constant) to increase with time. The low-frequency part of permittivity changed remarkably with time. The aging effects of snow are explained by the development of bonding bridges between ice particles and between the snow and the electrodes (Kuroiwa, 1962; Paren and Glen, 1978).

Dielectric measurements at each temperature were carried out after the annealing process. Figure 1 shows Cole–Cole plots of snow and hoar-frost at  $-10^\circ$ ,  $-2^\circ$  and  $-0.3^\circ\text{C}$ . The high-frequency part of the semicircle (dielectric dispersion around 30 kHz) in Cole–Cole plots is rather depressed. This asymmetric shape is more clearly recognized for the hoar-frost sample (Fig. 1b) than for the snow sample (Fig.

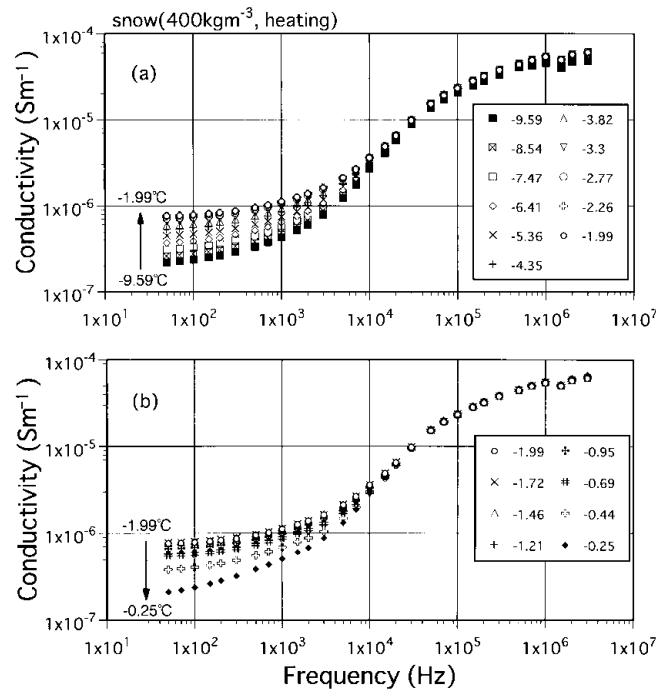


Fig. 2. Frequency dependence of the a.c. conductivity of a snow sample in a temperature range between  $-9.59^\circ$  and  $-0.25^\circ\text{C}$ . (a) From  $-9.59^\circ$  to  $-1.99^\circ\text{C}$ , the a.c. conductivity increases with increasing temperature. (b) From  $-1.99^\circ$  to  $-0.25^\circ\text{C}$ , the a.c. conductivity in the low-frequency range decreases with increasing temperature.

1a). Our result agrees with those of Paren and Glen (1978), but not with those of Kuroiwa (1962), Keeler (1969) and Maeno (1978), who reported a dielectric dispersion of snow with a simple semicircle. We do not know the reason for this discrepancy. The dispersion in the Cole–Cole plots can be analyzed successfully by assuming that it is composed of two dispersions (Davidson–Cole type and Cole–Cole circular law type) and a d.c. conductivity (Cole and Cole, 1941; Davidson and Cole, 1951). The dielectric dispersion around 30 kHz (dispersion 1, Davidson–Cole type) is caused by a reorientation of water molecules. The relaxation times (e.g.  $8.92 \times 10^{-6}\text{ s}$  for snow at  $-0.39^\circ\text{C}$ ) of the snow samples are shorter than those of ice ( $\tau = 2.2 \times 10^{-5}\text{ s}$  at  $-0.1^\circ\text{C}$ ; Auty and Cole, 1952). This means that the reorientation of water molecules is faster in snow than in solid ice. Dispersion 1 shows that the relaxation is shifted toward higher frequencies. The easier reorientation reflects the behavior of water molecules near the surface of ice particles in porous snow. Water molecules near the surface may move more easily than in bulk ice. The low-frequency dispersion (dispersion 2) is caused by a space-charge effect near the electrodes because the migration of ions through the sample is blocked at the electrodes.

The low-frequency part of the Cole–Cole plots shows a general increase as temperature increases from  $-10^\circ$  to  $-2^\circ\text{C}$  (see points at 5 kHz marked with an arrow in Figure 1a). The imaginary part  $\kappa''$  at 5 kHz decreases as temperature increases from  $-2^\circ$  to  $-0.3^\circ\text{C}$ . Such a temperature dependence above  $-2^\circ\text{C}$  is curious and difficult to explain by the usual dielectric mechanism.

Figure 2 shows a.c. conductivity of the snow at frequencies from 50 Hz to 5 MHz. The a.c. conductivity corresponds to the imaginary part  $\kappa''$  of permittivity multiplied by the angular frequency  $\omega$  and the vacuum's permittivity  $\epsilon_0$ ,  $\sigma = \epsilon_0 \omega \kappa''$ .

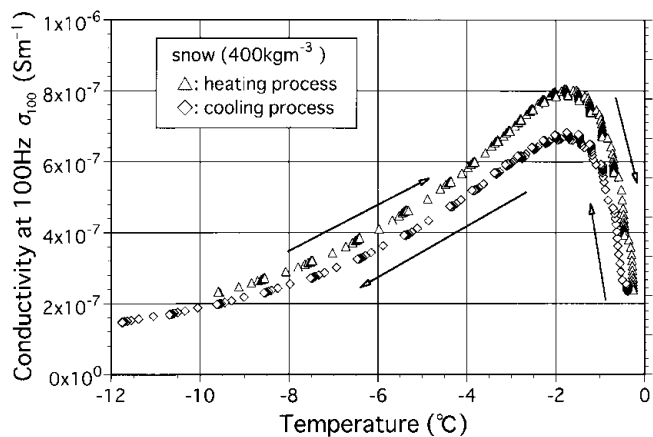


Fig. 3. Temperature dependence of the low-frequency conductivity (the a.c. conductivity at 100 Hz) for snow under the heating and cooling processes. A peak around  $-2^{\circ}\text{C}$  is recognized in both the heating and cooling processes.

In the frequency ranges above 1 MHz and below 100 Hz, the a.c. conductivity shows two characteristic constant values: the high-frequency conductivity  $\sigma_{\infty}$  and the low-frequency conductivity  $\sigma_{\text{LOW}}$ . The value of  $\sigma_{\infty}$  depends on the behavior of charge carriers showing a dispersion at around 30 kHz, and is given by  $\sigma_{\infty} = \sum e_i n_i \mu_i$ . This relation is well known as Jaccard's theory for electrical properties of ice (Jaccard, 1959). Here  $e_i$ ,  $n_i$  and  $\mu_i$  are the effective charge, the concentration and the mobility of defect  $i$ , respectively. The suffix  $i$  indicates ionic defects (+ or -) or Bjerrum defects (D or L). The latter defects reflect a behavior of the reorientation of water molecules. The d.c. conductivity ( $\sigma_0$ ) of ice is caused by the long-distance migration of the ionic defects (and

impurity ions). The value of  $\sigma_{\text{LOW}}$  reflects the d.c. conduction of the snow samples.

Figure 2 shows the increase of low-frequency conductivity  $\sigma_{\text{LOW}}$  as temperature increases from  $-10^{\circ}$  to  $-2^{\circ}\text{C}$  (Fig. 2a), and its decrease as temperature increases from  $-2^{\circ}\text{C}$  to the melting temperature (Fig. 2b). This is a curious behavior because d.c. conductivity for ice generally increases with increasing temperature.

Figure 3 shows the temperature dependence of the a.c. conductivity ( $\sigma_{100}$ ) at 100 Hz for snow. The 100 Hz conductivity  $\sigma_{100}$  reflects the low-frequency conductivity  $\sigma_{\text{LOW}}$  and also the d.c. conduction of the sample. The conductivity  $\sigma_{100}$  shows a peak around  $-2^{\circ}\text{C}$ . The peak in conductivity is recognized in both the heating and the cooling process. The peak temperature of the heating process seems to be higher than that of the cooling process.

Figure 4 shows the temperature dependences of conductivity at 100 Hz and at 1 MHz ( $\sigma_{100}$  and  $\sigma_{1\text{M}}$ ) for snow and hoar-frost. These samples show that the low-frequency conductivity around 100 Hz, which reflects the d.c. conduction, has a peak near  $-2^{\circ}\text{C}$ , whereas the high-frequency conductivity around 1 MHz does not show a peak.

### DISCUSSION

We use a.c. conductivities at 100 Hz and at 1 MHz ( $\sigma_{100}$  and  $\sigma_{1\text{M}}$ ) to investigate the temperature dependence of the low-frequency and the high-frequency conductivity ( $\sigma_{\text{LOW}}$  and  $\sigma_{\infty}$ ) of snow. Figure 5 gives the Arrhenius plots of the conductivities  $\sigma_{100}$  and  $\sigma_{1\text{M}}$  for snow and hoar-frost. The activation energy of the conductivity  $\sigma_{1\text{M}}$  below  $-2^{\circ}\text{C}$  is 0.15 eV for snow and 0.26 eV for hoar-frost. The conductivity  $\sigma_{100}$  below  $-5^{\circ}\text{C}$

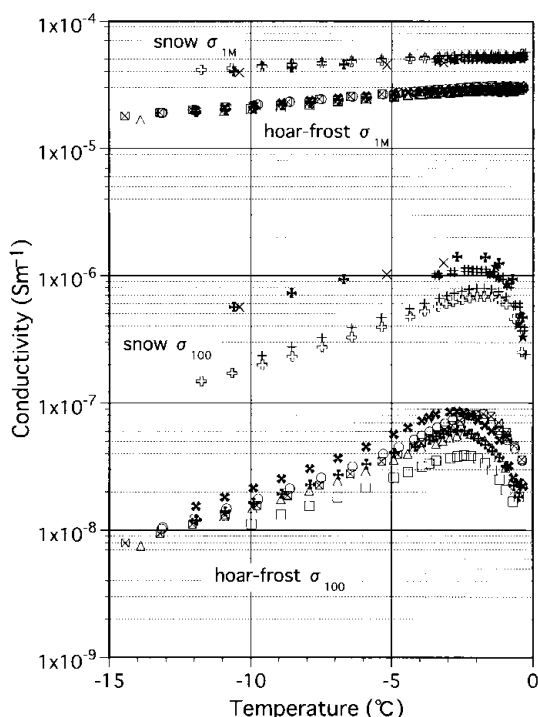


Fig. 4. Temperature dependence of the a.c. conductivity at 100 Hz ( $\sigma_{100}$ ) and the a.c. conductivity at 1 MHz ( $\sigma_{1\text{M}}$ ) for snow and hoar-frost samples. The low-frequency conductivity ( $\sigma_{100}$ ) shows a peak around  $-2^{\circ}\text{C}$  and decreases with increasing temperature above  $-2^{\circ}\text{C}$ , but the high-frequency conductivity ( $\sigma_{1\text{M}}$ ) does not show a peak.

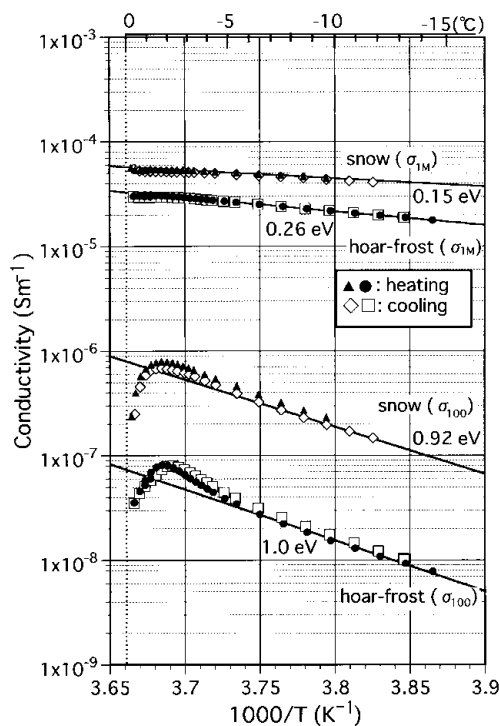


Fig. 5. Temperature dependence of the a.c. conductivity at 100 Hz ( $\sigma_{100}$ ) and the a.c. conductivity at 1 MHz ( $\sigma_{1\text{M}}$ ) for snow and hoar-frost (logarithm of  $\sigma_{100}$  and  $\sigma_{1\text{M}}$  vs reciprocal of temperature). The low-frequency conductivity ( $\sigma_{100}$ ) shows a peak around  $-2^{\circ}\text{C}$ . The high-frequency conductivity ( $\sigma_{1\text{M}}$ ) does not show a peak.

is 0.92 eV for snow and 1.0 eV for hoar-frost. The activation energy of  $\sigma_{100}$  from  $-5^\circ$  to  $-2^\circ\text{C}$  tends to increase.

It is well known that the activation energy of the high-frequency conductivity  $\sigma_\infty$  of ice around  $-10^\circ\text{C}$  is 0.56 eV for undoped ice (Auty and Cole, 1952), 0.24–0.60 eV for granular snow and 0.18–0.42 eV for dry snow samples (a.c. conductivity at 20 kHz; Traub and Gribbon, 1978), with a lower value of around 0.2 eV for doped ice with HCl or HF (review by Petrenko, 1993). Therefore we can conclude that our samples have some impurity ions. But the conductivity  $\sigma_{100}$  of hoar-frost with a low impurity content also shows a peak around  $-2^\circ\text{C}$ , similar to that for the sample of stored snow.

On the other hand, it is known that the activation energy of the d.c. conductivity  $\sigma_0$  of ice around  $-10^\circ\text{C}$  shows values of 0–0.6 eV for undoped ice (review by Petrenko, 1993; Petrenko and Whitworth, 1999), and higher values of the activation energy of the surface conductivity of monocrystalline ice are reported, of 1.1–1.4 eV below about  $-6^\circ\text{C}$  (Maeno and Nishimura, 1978). The activation energy of the electrical d.c. conductivity for different kinds of snow reported by Kopp (1962) showed values of 0.7–1.1 eV between  $-8^\circ$  and  $-50^\circ\text{C}$ . Since the low-frequency conductivity ( $\sigma_{100}$ ) of our porous snow samples also shows large values for the activation energy, of about 1 eV below  $-5^\circ\text{C}$ , the low-frequency conductivity is mainly caused by surface conduction. This conclusion is supported by the fact that our samples as porous materials have large specific surfaces and many bonding bridges between ice particles.

Kopp (1962) showed that the activation energy of the d.c. conductivity of snow increases with increasing temperature above  $-8^\circ\text{C}$ . Maeno and Nishimura (1978) reported that the activation energy of the surface conductivity of ice tends to increase with increasing temperature above  $-6^\circ\text{C}$ . They explained this result in terms of the appearance and development of the quasi-liquid layer on the ice surface. Our samples showed that the activation energy of the conductivity  $\sigma_{100}$  increases as temperature increases from  $-5^\circ$  to  $-2^\circ\text{C}$ . This means that the low-frequency conduction of snow above  $-5^\circ\text{C}$  may depend on the formation of the quasi-liquid layer on surfaces of ice particles. But the low-frequency conductivity ( $\sigma_{100}$ ) above  $-2^\circ\text{C}$  decreases as temperature increases toward the melting temperature, and the activation energy becomes negative (Fig. 5).

In order to check the role of the surface and bulk conduction, we used samples without inner airspace. Figure 6 shows the temperature dependence of the conductivities (at 100 Hz and 1 MHz) of polycrystalline ice samples 3 and 5 mm thick. The samples were prepared by freezing mixtures of snow and water. The water was degassed before freezing, but some small bubbles remained around the center of the samples. The polycrystalline ice samples consisted of grains of about 2 mm. Since the polycrystalline ice shows large aging effects, the results shown in Figure 6 correspond to values after a sufficiently long annealing process. The polycrystalline ice samples did not show a peak of the low-frequency conductivity ( $\sigma_{100}$ ). The bulk conduction of ice therefore does not contribute to the peak phenomenon. Activation energy of the conductivity  $\sigma_{100}$  is near 1 eV (0.70 eV, 0.85 eV) below  $-5^\circ\text{C}$  and shows a higher value above  $-5^\circ\text{C}$ . The polycrystalline ice samples have many interfaces between ice grains, and the conductivity  $\sigma_{100}$  is expected to show an electric conduction along the interfaces. Since the activation energy of the conductivity  $\sigma_{100}$  increases above  $-5^\circ\text{C}$ , the conduction of the interfaces may be affected by the quasi-liquid layer or a disorder layer of ice lattice in the interfaces.

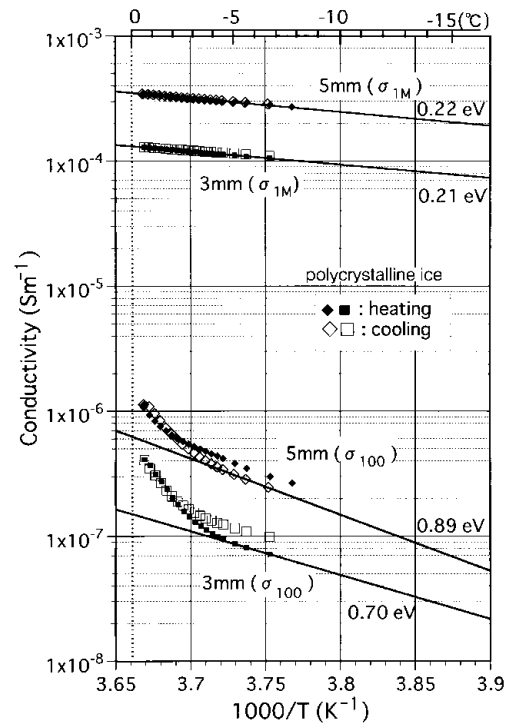


Fig. 6. Temperature dependence of the a.c. conductivities at 100 Hz ( $\sigma_{100}$ ) and 1 MHz ( $\sigma_{1M}$ ) for polycrystalline ice samples with 3 and 5 mm thickness (logarithm of  $\sigma_{100}$  and  $\sigma_{1M}$  vs reciprocal of temperature). The low-frequency conductivity ( $\sigma_{100}$ ) does not show a peak.

The absence of peaks of conductivity  $\sigma_{100}$  for the polycrystalline ice samples means that the presence of the free ice interface between water vapor and ice is more important. In order to check the role of the free ice interface and the inner airspace, we measured another type of sample without inner airspace: a snow sample soaked with kerosene. The sample ( $318 \text{ kg m}^{-3}$ ) was made of hoar-frost (2–7 mm grain-size) and annealed at  $-1.3^\circ\text{C}$  for 10 days. The sample showed a peak around  $-2^\circ\text{C}$ . This sample was soaked with kerosene, and the same dielectric measurement was carried out. Figure 7 shows the low-frequency conductivity ( $\sigma_{100}$ ) in the heating and cooling processes. The conductivity  $\sigma_{100}$  showed a peak around  $-2^\circ\text{C}$  in the heating process, but not in the cooling process. This phenomenon is reproducible: the peak in the heating process and no peak in the cooling process were also recognized after the first cooling process. This means that the peak phenomenon in the cooling process depends on the air-ice interface in the pores. After the experiment the conductivity  $\sigma_{100}$  below  $-5^\circ\text{C}$  showed a larger value than after the first stage of the experiment. Generally our samples were hardened by the experiment, but the sample with kerosene was fragile.

The peak of the conductivity  $\sigma_{100}$  around  $-2^\circ\text{C}$  means that the d.c. conduction (the surface conduction) decreases with increasing temperature above  $-2^\circ\text{C}$ , so the migration of electric charges is restricted by something caused by increasing temperature. The electric conduction of bulk ice medium is increased by heating (Fig. 6). Therefore the decrease of conduction should be caused by properties of the surface of ice particles or the intergranular structure of the snow samples. The physical properties of the surface near the melting temperature depend on the formation of the quasi-liquid layer and impurities, etc. Since it is not yet clear how the quasi-liquid layer or impurities affect physical properties, no conclusions can be drawn for their influence



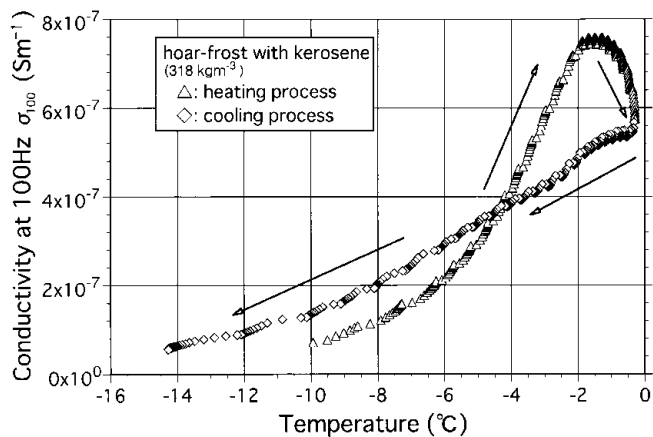


Fig. 7. Temperature dependence of the low-frequency conductivity (the a.c. conductivity at 100 Hz) for the snow sample ( $318 \text{ kg m}^{-3}$ , made of hoar-frost 2–7 mm grain-size) soaked with kerosene, in the heating and cooling processes. A peak around  $-2^\circ \text{C}$  is recognized in the heating process, but there is no peak in the cooling process.

on conduction. We know that the sublimation of ice particles (in other words, the vapor diffusion) and the surface diffusion of water molecules is active in snow near the melting temperature (Maeno and Ebinuma, 1983). Such a transfer of water molecules can change the microstructure of snow. Thus we may discuss an effect of the microstructure of snow on the peak phenomenon. If bonds between ice particles in snow disappear due to the vapor diffusion and/or the surface diffusion, these broken bonds may contribute to (or cause) the decrease of d.c. conduction. But bonds (with a negative curvature of the surface) of the ice particles generally grow due to the vapor diffusion and the surface diffusion near the melting temperature.

Now we discuss the possibility of disappearance of contact between the ice particles. We prepared samples by loosening a snow block and packing it into a sample cell. Our samples consist of many ice particles. Although the samples were annealed for 1 week (or more) near the melting temperature, we expect that a perfect network of ice could not be constructed and that there were tight bonds, imperfect weak bonds and simple contacts between ice particles in the samples. We can expect that some weak bonds and parts of the contacts (with a positive curvature of the surface) may disappear due to such an active transfer of water molecules, and that the d.c. conduction decreases in the heating process. In the cooling process, contacts may be reconstructed by the condensation process at sites of the broken bonds and contacts. Tight bonds (and some weak bonds and contacts) between ice particles grow due to the vapor diffusion and the surface diffusion near the melting temperature. But neck growth of the tight bonds may have only a minor effect on surface conduction compared to the disappearance of weak bonds and contacts. Even one disappeared contact can effectively restrict a long-range migration of ions. This process could explain the results for the sample soaked with kerosene. The peak in the heating process is caused by destruction of weak bonds and contacts by surface diffusion of water molecules, and the absence of a peak in the cooling process is caused by the suppression of the reconstruction by condensation of new contacts by the kerosene. The increase of conductivity at low temperatures after the experiment is explained

by the neck growing of tight bonds near the melting temperature. If the disappearance and reconstruction of the weak bonds and contacts is in fact the main reason for the peak of the d.c. conduction, this finding may be important for pre-melting processes in snow.

The above discussion with many assumptions is one possible way of explaining our experimental results. The quasi-liquid layer or impurities on the ice surface may also be involved in the peak phenomenon. Additional investigations of the physical properties (mechanical properties, quasi-liquid layer and effects of impurities) of snow near the melting temperature are needed to reveal this phenomenon.

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