$^{\it 08}$ Experimental detection of bleaching of fresh ice in the optical range near 0°C

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The results of laboratory measurements of the transparency of fresh ice blocks in the optical range in the temperature range from -15° C to 0° C are presented. The studies were performed at two wavelengths in the visible range (at a wavelength of 535 nm) and the ultraviolet range (at a wavelength of 370 nm). We studied the ice of a natural fresh water body with a characteristic predominant spatial orientation of the main optical axis of the crystals. It has been established that when approaching a temperature of 0° , the sample becomes bleaching (decrease in attenuation). The effect occurs in the range $-0.5-0^{\circ}$ C. It is determined by the occurrence of plastic deformation due to thermal stresses caused by the initial stage of the ice-water phase transition. The bleaching in experiments with ice blocks ~ 10 cm thick was 3-25%. The results obtained are of interest for solving remote sensing problems, since melting ice is a widespread object due to its significant heat of phase transition. Previously, the effect of bleaching of fresh ice was also discovered in the microwave range when the temperature of the sample approached the phase transition point.

Keywords: fresh ice, melting temperature, optical range, ice bleaching, plastic deformation.

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Introduction

Electromagnetic properties of ice near 0°C are of interest for practical purposes due to a large value of heat of phase transformation. On the other hand, processes near the phase transitions of water are still understudied. It is believed that near 0°C, depending on the concentration of various inclusions, ice starts melting in some regions of ice structures, thereby resulting in increase in their effective dielectric constant $(\dot{\varepsilon})$. It is defined by higher values of $\dot{\varepsilon}$ of water as compared to clean ice, especially at low frequencies. The closer the medium temperature (T)to 0°C, the higher concentration of liquid inclusions. Due to their structuring, permittivity anisotropy occurs, which can result in polarization specific features of various ice items. For example, the paper [1] has observed polarization difference of radar signals when probing river ice. It is possible that in this case $\dot{\varepsilon}$ of the ice cover in contact with water (in the lower layer, where its temperature is 0° C) had a tensor nature, which is defined by the influence of water flow on the ice structure near the phase transition temperature.

The papers [2,3] have paid attention to special characteristics of fresh ice when the microwave radiation passes through the medium with its heating to 0°C. An effect of "bleaching" has been detected, i.e. a noticeable increase in power of the radiation passing through the ice sample prior to first signs of melting.

The effect is explained in [2,3] by shear plastic deformation of ice crystals occurring near the ice-water phase transition at small mechanical stresses. On the other hand, in terms of the phenomena physics, it can be noted that during melting ice is both at a crystal state and a liquid one, as it exhibits areas with broken hydrogen bonds. This transitional state must result in anomalies of the physical characteristics, which will also depend on structural specific features of the medium (impurities, flaw orientation, etc.).

The above-said works on ice bleaching were conducted in the microwave range only. At the same time, the optical range is one of the most used ranges for remote probing of the Earth. It is applied for atmospheric research and research of an underlying surface of various types. Thus, the paper [4] has studied the areas of surface runoff into Earth's ocean using satellite data in the visible range. The paper [5] has analyzed vegetation characteristics of the earth covers. The [6] has studied atmospheric parameters. The optical range is widely used for research of cryosphere formations [7].

As for ice spectrum's specific features at the temperature near 0°C, they are used in deciphering the data of remote probing of cold cloud formations, frazils, snow and ice covers, frozen soils and grounds. For example, the paper [8] has determined a normalized difference snow index (NDSI) based on using the green and near infrared ranges [9]. In doing so, it is relevant to study ice by specifying its characteristics within the region of the phase transition temperature.

That is why the present research was aimed at studying attenuation of the optical radiation in fresh ice at the temperature near 0° C and to show that in this case it is

necessary to conduct advanced research of absorption and scattering of the electromagnetic waves.

Experiment procedure

The present paper has done research on transmittance of the optical radiation through the block of fresh ice. The measurements were performed in the visible and ultraviolet ranges. The visible range has used the semiconductor laser for the wavelength of (λ) 535 nm with linear polarization. The ultraviolet range has used non-polarized light from the LED emitter for the wavelength of 370 nm. Fig. 1 shows the diagram of the unit for measuring power of the passing radiation (*P*) depending on time (*t*). Accuracy of absolute measurements of the temperature is ~ 0.2°C. The measurement thermocouple was placed in the block at the depth of 1 cm.

According to the method used, radiation was passed through the parallelepiped sample and it was assumed that the reflectance (R) slightly changed during the experiment. It follows from the fact that at the wavelengths used, the real part of the refraction index of ice (n) at 0°C is 1.311 $(\lambda = 535 \text{ nm}), 1.334 \ (\lambda = 370 \text{ nm}) \ [10,11].$ Hence, the reflectance value $R \sim 0.018 - 0.02$, which is determined by the formula $R = [(n-1)/(n+1)]^2$. If for some reason n changes by 0.001, then the increment ΔR will be 0.0001 or $\sim 0.5\%$. This change is possible, if layers of liquid water occur at the boundary. If the layer thickness is of an order of the wavelength, then it is necessary to take into account transmittance coefficient of such a threelayer medium. For water n = 1.333 ($\lambda = 535$ nm), 1.334 $(\lambda = 370 \text{ nm})$. These values are higher than those for ice, so when the water films occur at the initial stage of melting and interference appears, the passing coefficient should decrease (i.e. radiation attenuation shall increase) [12]. This specific feature can be used for determining a moment when the liquid starts occurring at the ice surface (when analyzing the influence of the boundary layer on radiation transfer). The imaginary part of the refraction index can be omitted as its value is of many orders less than the real part.

The ice samples for the research were extracted from the ice cover of the fresh water lake Arakhley (Zabaykalsky



Figure 1. Diagram of the unit for measuring the optical-range radiation passing through ice near the phase transition temperature. I — emitter, 2 — sample — block of ice, 3 — thermocouple, 4 — data acquisition and recording system, D — photodetector.

Krai) in February–March. At this time of the year concentration of the salts in ice is about 1 mg/l [3] in the middle part of the ice cover along its height.

During the experiment, the sample was placed to direct the vector of the electric field of the electromagnetic waves either in parallel or perpendicular to a basal plane of the ice crystals. As established in a number of the papers, during formation of lake ice, after the initial stage of cover formation (when subglacial flows disappear) 90-100% of the crystals in the ice cover are oriented with its main optical axis "C" in perpendicular to an ice-air interface surface and can deviate from the normal within several degrees. In accordance with the classification of N.V. Cherepanov, it corresponds to the A1-type ice, which is formed in small lakes at the temperature gradients 1-4°C/m in the sub-ice layer of water [13].

Previously, the paper [2] has marked the effect of bleaching of the ice samples in the microwave range depending on a position of the basal planes of the crystals in relation to the vector of the electric field. This specific feature was considered in the present research.

The ice samples were stored in a freezer at the temperature of -15° C. Then, they were place in the unit and heated at the room temperature to 0°C. After the ice surface was humid, i.e. after the signs of melting appeared, the experiment was finished. The receiver and source of radiation were at the constant temperature.

The signals from the thermocouple and the photodetector were recorded to the data acquisition system. The signals were registered at the interval of ~ 0.3 s.

Results obtained and discussion thereof

Fig. 2 shows the results of measurement of power of transmittance of laser radiation in the visible range depending on time. This experiment had the vector of the electric field of the wave placed in the basal plane of the ice crystals. The results are characterized by the noticeable short-time increase in the power of the registered signal by approximately 25% when ice approaches the phase transition temperature.

Fig. 3 shows the results of the similar measurements for the case when the vector of the electric field of the waves is perpendicular to the basal plane of the ice crystals. Here, the increment approaches 5%, but it was observed during melting and was complex depending on time. When the temperature was 0° C, the short-time decrease in the signal was observed.

Fig. 4 shows the results of the measurements of passing of radiation in the ultraviolet range for the non-polarized radiation, but with the vector **E** in the basal plane. Fig. 4, a — the graphs of the ice temperature and the passing power of radiation, 4, b — the position of the block with distinguished predominant orientation of the basal plane of the fresh ice crystals. The graph for the registered power is characterized by: its smooth change within the region of the



Figure 2. (*a*) Ice temperature (a dashed line) and power of the linearly-polarized visible radiation passing through the sample, depending on time at the wavelength of 535 nm. Registered power in units of voltage of the photodetector (U). (*b*) Diagram of the position of the basal plane (B) and of orientation of the vector of the electric field (E).



Figure 3. (a) Results of measurement of power of the passing visible radiation; (b) Diagram of the position of the ice block; the designations are similar to Fig. 2.

temperatures $-12--8^\circ C$ and the abrupt increment near $0^\circ C$ at $\sim 23\%.$

For comparison, Fig. 5 shows the results of measurements when the vector of the electric field was chaotic to the basal planes of the ice crystals. The graph of the transmitted power in Fig. 5 differs from the graph of Fig. 4 in oscillations of the signal power, which begin with decrease in its value. The amplitude of the oscillations is $\sim 3\%$.

Thus, the experiments for measurement of attenuation of optical-range radiation at the wavelengths of 535 and 370 nm have resulted in detecting the specific features in the form of substantial changes of passing radiation's power prior to the start of melting of the sample, whose structure contains the crystals with distinguished spatial orientation of the main optical axis. The maximum effect was observed for the position of the vector **E** in the basal plane. The distinguished orientation of the crystals is typical for naturally formed ice. In accordance with the Curie principle, growing ice follows the symmetry of the ambient space [14,15]. In case of the ice cover, from which the sample was extracted, it is a cone symmetry at which the axis C^{*} is oriented in perpendicular to the ice-water interface surface.

The increment of the radiation power approached 25%. In some experiments, the detected effect looks like "bleaching" of the medium during surface application, e.g., of a quarterwave layer with an intermediate value of the real part of the relative dielectric constant. However, in this case the physical mechanism of bleaching seems to be different. First of all, it is known that shear deformation induces the nonlinear dependence of stress and strain in the ice crystal. At the same time, the region of plastic flow exhibits negative differential viscosity [16]. Generally, it can be assumed that here, due to available electric moments, the water molecules of the layer with flowing can exhibit electric instability and originate amplification of the electromagnetic radiation. These layers are nanometer-thick and they are separated by flat crystal plates having a thickness of several dozens nanometers [13]. The more specific mechanism of increase in strength of the electric field can be related to surface plasmons appearing on the flat conductive film or island films [17]. The conductivity at the boundary of a



Figure 4. (a) Graph of the passing power of UV radiation (in units of voltage at the detector output) (the solid line) and the temperature depending on time (the dashed line); (b) Position of the ice block, the basal plane of the crystals in the sample is marked; the designations are similar to Fig. 2.



Figure 5. (a) Graph of the passing power at the wavelength of 370 nm (in units of voltage at the detector output) (the solid line), the temperature graph (the dashed line) depending on time; (b) Position of the ice block; the designations are similar to Fig. 2.

layer for the flat crystal layer flowing therealong can occur due to ferroelectric ordering of the medium dipoles. It can be assumed that the broken hydrogen bonds result in orientation of the dipoles of the water molecules in a plane of media contact. In turn, as shown in [18], the contact of the ferroelectric and the dielectric exhibits higher conductivity (by many orders of the magnitude). To confirm such a possible mechanism of field amplification, we can indicate discovery of gigantic amplification of Raman scattering of radiation in the presence of metal periodic or percolation nanostructures [19].

Besides, another specific feature has exhibited in the form of non-monotonic modification of attenuation at the negative temperatures of ice, when the block was heated while being placed into the unit (for example, it is seen on Fig. 4, *a* at the temperatures from -15° C to 0° C). This specific feature can be also explained by plastic deformation starting due to the temperature gradient in the sample.

As noted above, the bleaching effect was previously observed within the microwave range [2]. At the same time, the paper [20] has established a special state of surface layers of ice. It was studied both when illuminating the sample and in the resonant cavity as well. It was found that at the stage of increase in the radiation's passing power there was no liquid water in the sample as the resonance frequency of the resonant cavity did not change noticeably. In case of occurring of liquid water, the substantial decrease in the resonance frequency would be observed, so would increase in the resonant cavity losses. The paper [20] has assumed that the detected specific feature near the melting start was related to formation of the conductive percolation films or change of the properties of clean ice when the crystals start breaking in the phase transition point.

For the case of Fig. 5, when the vector of the electric field did not have the distinguished direction in relation to the basal plane, the intensity of the passing radiation had a time-oscillating nature. However, the starting point of ice destruction of this experiment has seen the decrease in the passing power, which can be associated to the influence of the liquid layer appearing on the sample surface. It should be noted that it is difficult to determine the moment of melting start due to a small temperature interval for the

bleaching effect appearing therein. Nevertheless, by a set of experimental data, it is possible to distinguish an effect of wave field's interaction with the layers on the basal planes (bleaching) and an effect of liquid films on the sample surface (increase in absorption).

At the same time, the question needs to be further studied due to possible incomplete orientation of the crystal axes in a certain distinguished direction in the research object, difference of values of the temperature gradients in various experiments and influence of impurities and inclusions (the concentration of salts, chemical specific features, etc.). Generally, ice can flow not only along the basal planes of the crystal, but mechanical stresses by a higher order are required for this [13].

Conclusion

1. The present paper has detected the effect of ice bleaching prior to the start of its melting, when its mechanical strength is the least at the wavelengths of 535 and 370 nm. In our tests, the power increment in the sample of the thickness of 10 cm approached 25%. The similar effect had been previously detected in the microwave range, too [2].

2. The bleaching effect shall become the most apparent, if the vector of the electric field of the wave in ice coincided with the basal plane of the ice crystal. It was established during the research of natural ice extracted from the ice cover of the fresh water bodies.

3. It was assumed that the medium bleaching could be related to the formation of the conductive nanostructures on the crystals' planes for plastic deformation thereon. The plastic flow occurs due to thermal stresses, which are the most conspicuous near the phase transition temperature. It is possible that significant variations of attenuation can be observed at any mechanical impact, if there is ice flowing.

4. The effect of change of the electromagnetic characteristics of the ice formations under mechanical stresses can be used in polarization measurements of processes in the cryosphere objects both in the optical and microwave ranges.

Conflict of interest

The authors declare that they have no conflict of interest.

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