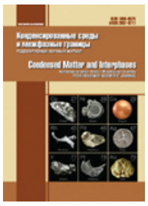

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Entropy features of the PeTa effect during phase transformations of water

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Abstract

The article discusses a hypothesis put forward by V. A. Tatarchenko and M. E. Perelman. According to it, the first order phase transition during vapour condensation or melt crystallisation (PeTa effect) is accompanied by the appearance of nonthermal radiation of the media. The generally accepted point of view is that the latent heat of phase transformation can only be released in the form of heat. When the authors of the hypothesis tried to prove the existence of the effect of nonthermal radiation and considered the facts confirming it, they did not take into account the peculiarities of the initial and final states of the medium (i.e. their entropy). To clarify the physics of the process of liquid crystallisation and to consider the possibility of nonthermal radiation, we studied the peculiarities of water crystallisation and the formation of ice. This is the process the authors referred to in order to prove their hypothesis. It was shown that in various experiments, it is necessary to consider both the state (structure) of the initial water samples and the formed ice, which can consist of various crystalline modifications with chaotic packing. These features of initial and final states, i.e. the entropy of water and ice samples in real experiments and under observed natural phenomena, make it more difficult to assess the characteristics of a possible radiation. The entropy of the initial and final states was determined by the procedure of the system preparation and the peculiarities of the phase transition dynamics. Its values depend on macroscopic parameters, as well as on the microstructure of the media, the determination of which is a very challenging task in each specific case. In addition, in many cases, we have to deal with metastable media, for which it is necessary to take into account the influence of fluctuations on the process of the phase transition. Therefore, the concepts of equilibrium thermodynamics are not applicable to them. However, these are the media where non-heat radiations may occur in accordance with the laws of self-organisation in nonlinear weakly nonequilibrium objects. This work shows a method for preparing low-entropy medium with its subsequent phase transformation into ice. To do so we conducted an experiment which involved freezing concentrated alcohol in order to obtain deeply supercooled water. It appears that to find the characteristics of the PeTa radiation it is necessary to take into account the entropy constraints for each specific case, which will allow assessing the spectrum of possible non-heated radiations and their characteristics.

Keywords: Phase transition, Non-heat radiation, Supercooled water, Entropy

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1. Introduction

It is known that the behaviour of chemical reactions is determined by their energy characteristics and the entropy of the reaction products. In particular, the direction of isothermal processes, close to equilibrium, is determined by the Gibbs energy. Energy and entropy constraints also apply to the phase transitions of matter in isolated and closed systems.

A number of works [1–3] detected non-heat radiation during phase transitions of the first kind. This effect was called the PeTa effect after the names of the researchers who described the phenomenon in detail (M. E. Perelman and V. A. Tatarchenko). Phenomena experimentally similar to the PeTa effect were found to enhance microwave radiation during the deformation of ice crystals [4], during phase transformations in ferromagnetic alloys [5], and during sonoluminescence [6].

For example, in terms of energy relations, during the phase transition of a substance from the liquid state into the solid state, the estimated energy of the radiation quanta for a mole of matter according to [1–3] is calculated from the equality: $h\nu = \frac{\lambda}{N_A}$, where h is the Planck's constant, ν is the frequency of electromagnetic radiation, N_A is Avogadro number, and λ is the specific heat of the phase transition. A multiphoton radiation process is also possible: $h\nu = n\frac{\lambda}{N_A}$, where n is the number of photons in an individual act of radiation. According to the estimates, in most cases the energy quanta are within the infra-red range. Since the explanation of the effect has caused an ambiguous assessment, as was noted in [2], it is of interest to consider other aspects of the issue.

In particular, [1–3] first of all took into account the energy and quantum-mechanical features of the PeTa effect. At the same time, there is a challenging issue regarding the probability of the considered events. It appears that the solution of this issue requires a careful consideration of the initial and final states of the system in terms of changing entropy, i.e. the implementation of the second law of thermodynamics. The entropy of the system is determined by very diverse structural characteristics of the medium and electromagnetic radiation (the volume of the components; the spectrum, polarisation, and the direction of the radiation; the shape of the parts of the system; their mutual arrangement in space, etc.).

The purpose of this work was to use the example of freezing water to consider at a qualitative level the entropy features of the PeTa effect which have not been previously taken into account by researchers.

Problem statement. When considering the PeTa effect, a number of issues arise. 1. It is not clear what proportion of energy can be converted into monochromatic radiation or other types of non-thermal radiation and what their characteristics are (degree of coherence, spectrum, etc.). 2. In what type of processes PeTa and PeTa related radiations can occur (for example, during cavitation, supercooling with a subsequent phase transition, pressure pulses, etc.). It seems that to answer these questions, it is necessary to take into account entropy constraints alongside with the law of conservation of energy and the concept of quantum mechanics about the probability of radiative transitions. It is known that in isolated systems, entropy cannot decrease spontaneously (i.e. the energy of the system cannot be effectively transformed into the energy of monochromatic radiation, which has a lower entropy).

2. Theoretical consideration

As follows from [3], A. D Sakharov (in a private communication) drew attention to the process of freezing supercooled water, which can be accompanied by an energy discharge from the sample by means of radiation resulting in non-thermal radiation. Its registration could serve as an experimental confirmation of the existence of the PeTa effect. This follows from the fact that the relatively fast transition from the liquid to the solid state of bulk water observed in some experiments is impossible since it is not provided for by the process of thermal conductivity.

Let us consider this process of phase transformation in terms of the second law of thermodynamics. Let us denote S_0 as the entropy of the initial state (for a mole of liquid water), S_L as the entropy of the formed ice, and S_E as the entropy of the released energy equal to the latent heat of the phase transition. Then, S_E will be represented as the sum of two components: $S_E = S'_E + S''_E$, where S'_E is associated with thermal energy, and S''_E is associated with non-thermal radiation. Then, from the second law of thermodynamics:

$$S'_E + S''_E + S_L \geq S_0\text{, or } (S'_E + S''_E) \geq (S_0 - S_L) \qquad (1).$$

If the entire process of phase transition is accompanied by a release of heat (in quasi-static processes), it means that for one mole of a substance $(S_0 - S_L) = \dfrac{\lambda}{T_0}$, where T_0 is the temperature of the phase transition (it is assumed that liquid water is initially at T_0, the same is true for the formed solid fraction).

In case of the PeTa effect and a release of part of the energy (E'') in the form of radiation during transformations with one mole of water, relation (1) is represented as:

$$(S_0 - S_L) \leq \left(\frac{\alpha\lambda}{T_0} + S''_E\right), \qquad (2),$$

where $\alpha = (0\ldots1)$ characterises the share of thermal energy released during the phase transformation. Full energy: $E = \alpha\lambda + E'' = \alpha\lambda + (1-\alpha)\lambda$. The relation $E''/E = (1 - \alpha)$ characterises the energy output of the PeTa effect.

For the equilibrium phase transition, the value of the difference between S_0 and S_L equals $\dfrac{\lambda}{T_0}$. Then, it follows from (2) that if $\alpha < 1$ (i.e. in the event of radiation), S''_E given per single unit of energy should not be lower than for the process of heat transfer by contact. This can be verified by using the equal sign in (2) and $(S_0 - S_L) = \dfrac{\lambda}{T_0}$, then the minimum $S''_E = \dfrac{(1-\alpha)\lambda}{T_0}$. Hence, the conclusion that there is a thermodynamic exclusion of the PeTa effect or more precisely that the radiation entropy for the equilibrium phase transition cannot be lower than the entropy accompanying the energy released in the form of heat.

This conclusion is quite obvious, since electromagnetic radiation with a narrow band has a low entropy due to its “ordering”. $S''_E \sim \Delta\nu\Delta\varphi$ [7], where $\Delta\nu$ is the frequency bandwidth for the electromagnetic radiation and $\Delta\varphi$ is the width of the solid angle of the radiation beam. For the case when $\Delta\nu$ or $\Delta\varphi$ tend to zero, S''_E also tends to zero, and relation (2) is not satisfied if α is less than one (for fixed values of S_0 and S_L).

At the same time, in real-life physical systems, the difference $(S_0 - S_L)$ presented in formulas (1, 2) can be reduced by preparing a special initial state leading to a decrease in this difference. To do this, S_0 should be reduced and S_L should be increased. Let us consider both cases.

2.1. Increasing S_L. This is possible if the structure of the formed ice is broken, for example, part of the ice is in an amorphous state or there are mixed structures with a chaotic arrangement of the ice crystals Ih and Ic. In recent works, the discovery of transitional forms during the phase transition of water [8, 9] in the form of the so-called stacking-disordered ice I_{SD} was reported. This is a mixture of Ih (hexagonal) ice and Ic (cubic) ice with a complex random structure. It is believed that previous reports of the experimental determination of cubic ice actually referred to stacking-disordered ice.

2.2. Decreasing S_0. S_0 can be decreased by the supercooling of water. Supercooling in the natural environment was observed for cloudy aerosol down to −37.5 °C [10]. In this case, the entropy of supercooled metastable water S_{0S} is below the initial state S_0 by the value of $\Delta S = \displaystyle\int_{T_0}^{T_x} \frac{\delta Q}{T}$, where δQ is a variation of thermal energy and T_x is the supercooling temperature. The lower T_x is, the smaller the new value of S_{0S} is. In addition, a reduced entropy can occur due to its decrease determined by the geometry of parts of the system if the water volume is divided into separate small fragments (in the case of hydrosol or films on the surface of pores or small particles).

The integral for ΔS significantly depends on the value of T_x. However, there are additional complications in the case of water. According to numerous studies, the heat capacity of supercooled water at constant pressure tends to soar when approaching −45 °C (at normal atmospheric pressure) [11]. This feature is determined by the influence of the second critical point of water and the appearance of the Widom line, the locus of increased density and entropy fluctuations of liquid bulk water [11, 12]. Although reaching –45 °C without water crystallisation is very difficult in practice, a decrease in entropy during supercooling is possible, for example, in porous media with nanosized pores, where the supercooling of bulk water to –70 °C was observed [13]. Due to an increase in heat capacity during the supercooling of liquid water, the entropy decreases nonlinearly with the temperature and it plummets near –45 °C. It might be for this reason that the temperature of homogeneous nucleation (i.e. below which there is only crystalline ice) is considered to be approximately –41 °C, i.e. it corresponds to a higher value than the temperature on the Widom line.

In graphical form, the section of the process where the PeTa radiation might occur during the supercooling of water is shown in Fig. 1. The graph has a starting point A. Point 1 corresponds to temperature $T_{0,}$ at which ice formation for bulk water is observed experimentally. Up to point 2, under certain conditions, there is a possibility of liquid water supercooling with the formation of metastable water with reduced entropy. Region 2–2′ is the area where the PeTa effect might appear, where a nonequilibrium phase transition is observed. 2′–3 is the area of “normal”, i.e. equilibrium phase transformation of a liquid into a crystalline body with a release of heat. In other words, compared to area 2′–3, in region 2–2′, more thermal energy is released per unit mass of the formed ice, since the medium is heated from T_x to T_0. Moreover, the heat takes more entropy than for section 2′–3 where the temperature is constant. However, some of the energy may have reduced entropy.

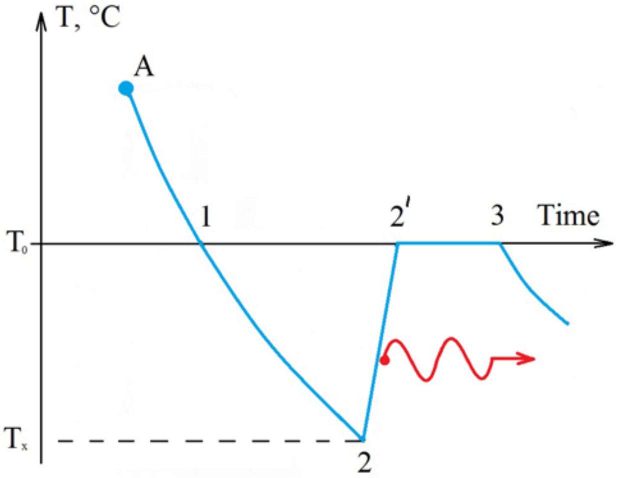

Fig. 1 Graphical representation of water cooling over time and the area where there is a possibility of PeTa radiation during the process of supercooling of a certain volume of water. The wavy arrow marks radiation from region 2–2′ on the temperature versus time graph (non-thermal radiation)

3. Experimental

We studied the freezing of an associated liquid (concentrated ethyl alcohol containing a small amount of water) as an example explicitly demonstrating the peculiarities of the initial state. In this experiment, it was expected to achieve extreme supercooling of water released as a result of the decomposition of the associates during the phase transition of alcohol and, thus, its special initial state with a low entropy. This technique was used for the supercooling of water for the first time. To register the moment of the phase transition and its features, a non-contact method was used. It recorded the electromagnetic radiation passing through the sample in the microwave range. The change in the transmitted power depending on the medium temperature allowed determining the onset of the phase transformation, the rate of the thermal energy release, and the variations of the sample temperature.

During the experiment, a sample of a nanoporous sorbent saturated with alcohol was exposed to radiation in the microwave range at a frequency of 34 GHz and was placed in a waveguide section. The experimental installation included a low-power Gunn diode generator and a crystal detector. The waveguide in which the sample was placed had a rectangular section of 3.4 mm by 7.2 mm. A sample in a 5 mm thick plate-like cuvette with sorbent powder was placed in the waveguide at an angle of ~ 45 degrees to the waveguide axis to eliminate interference effects. The power of the transmitted radiation and the temperature of the medium were measured as a function of time while cooling the sample with cold nitrogen vapour. To prepare the sample, ethyl alcohol and silica gel KSKG were kept in a desiccator to saturate the sorbent. The average size of the KSKG pores was 8 nm.

Ethyl alcohol with a concentration of 95% was used in the experiment. The alcohol contained 5% of water. As a result, its freezing point was about −114 °C. At the moment of the phase transition, a release of deeply supercooled water was expected. Its freezing proceeded according to a scheme similar to that shown in Fig. 1 for area 2–2′. The temperature of the medium was measured using a thermocouple. The temperature accuracy was within several degrees. The time constant of the thermocouple was about 0.1 s. Signals were recorded by an Agilent data acquisition system at a rate of two measurements per second. The measurement results of the transmitted power of microwave radiation depending on the temperature of the medium are shown in Fig. 2.

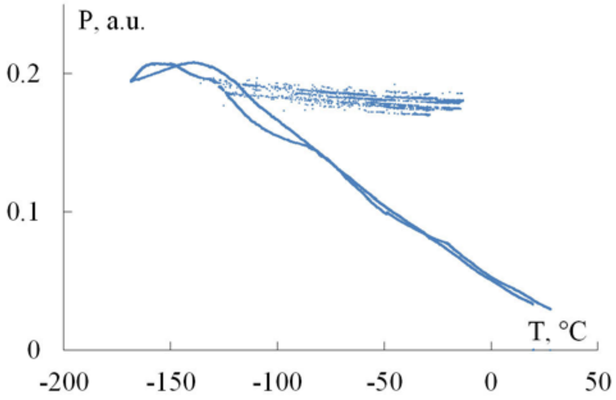

Fig. 2 A change in the power of microwave radiation (P) (in relative units) transmitted through the waveguide section with silica gel saturated with 95% ethyl alcohol depending on the temperature of the medium

When the sample was cooled, multiple temperature jumps were observed during the alcohol crystallisation within a certain time interval when the temperature was below −114 °C. This was accompanied by a rapid heating of the medium. This phenomenon is well known in cryology and is explained by the freezing of nonequilibrium supercooled water. Multiple jumps can be explained by the inhomogeneity of the temperature distribution in the sample volume and by the variations in the parameters of the sorbent with alcohol and the pore space. Since the measurement time between individual points was 0.5 s, it was possible to use points of the graphs to estimate the time of a temperature jump when the released volumes of water were frozen. It was about one minute. The temperature increment during the jump approached 0 °C (the graphs showed a value equal to approximately −10 °C, this can be explained by the thermal inertia of the medium and the thermocouple, as well as the spatial separation of the thermocouple and active regions where the heat was released). It is obvious that the heat emissions were accompanied by the formation of ice from the state of water with a reduced entropy. Currently, there are no data regarding the entropy of the formed stacking-disordered ice that would allow estimating this value. It was reported that the proportion of cubic ice in the I_{SD} ice can reach 70% [8].

The obtained results confirmed the hypothesis regarding the release of heat by supercooled metastable water with a low initial entropy at temperatures well below the point of the equilibrium phase transition, which creates the preconditions for the appearance of nonthermal radiation.

4. Discussion

There is an additional challenge with regard to the discussed process owing to a hindered emission of quanta into the external medium due to their absorption in the bulk of the liquid and solid phases. Therefore, the PeTa effect can only be observed on the surface layer with the thickness of a skin layer of water. For the thermal infra-red region, the skin layer value for water and ice is about 10 μm. As a result, the equal sign will not work in relation (1). Thus, only sign “>” remains, which complicates the analysis of the system. In this case, nonthermal radiation may not be registered in the experiment.

It is possible that when using porous, weakly moistened sorbents, the radiation emerging from the medium in the IR region may not be significantly attenuated if the matrix material is sufficiently transparent within this wavelength range.

The experiment with concentrated ethyl alcohol showed the possibility of deep supercooling of water that is formed as a result of alcohol freezing and that at the initial moment has a temperature of its phase transformation of −114 °C. In the case of such deep supercooling, water has an extremely low entropy. It is interesting to note that below this temperature, a glass transition of water occurs (at approximately

–130 °C), which is presented in a form of amorphous (disordered) ice.

To perform a similar experiment in order to detect non-thermal electromagnetic radiation, sufficiently fast measuring instruments are required to register electromagnetic fields. In the performed experiment that involved alcohol freezing in a porous medium, temperature jumps were observed over a time of about tens of seconds and the intensity of microwave radiation did not change significantly. This is owing to the thermal inertia of the sorbent matrix and the applied measuring devices: a microwave detector and a thermocouple. The proposed technique can also be used to search lower frequency radiations that penetrate well the medium than radiations in the IR range considered in [1–3], for example, at frequencies below 1 GHz.

As an example of using the entropy approach, let us estimate the energy of nonthermal radiation during the crystallisation of supercooled water in a cloudy aerosol, for example, at a temperature of -10 °C. Let us accept a number of assumptions that simplify the assessment. Let us assume that no stacking-disordered ice is formed and let us not take into account the change in the entropy of water upon its crushing into droplets in aerosol. We also assume that the value of the water heat capacity is invariable at constant pressure (C_{P}) within the range of 0 to –10 °C. During the process of water supercooling, its entropy decreases by $\Delta S = C_p \int_{T_0}^{T_x} \frac{dT}{T}$, where T_0 = 273 °C, T_{X} = 263 °C, C_{P} = 4.2 J/(g deg). As a result of calculations, we get ΔS = 0.15 J/(g deg). For an equilibrium process for 1 gram of water, this change will be $S' = \lambda/\mathrm{T}_0 \approx 1.2$ J/(g deg), here λ = 333 J/g. The entropy difference ($S_0 - S_{\mathrm{L}}$) is provided by the outflow of thermal energy. If this difference decreases with the same outflow of energy, part of the energy (i.e. E'') may have zero entropy in the limiting case. This means the PeTa effect is possible.

Relative fraction of non-thermal radiation energy is $E''/\lambda = \Delta S/(S_0 - S_{\mathrm{L}})$, hence $E'' = T_0\Delta S$. The ratio, i.e. the fraction of the radiation energy per unit mass of matter, will be 0.12 in this example. However, it should be taken into account that in the process of phase transformation, the temperature of the medium rises and its effective temperature will be above –10 °C. If this process is considered to be linear, the effective ΔS will be twice as low. Consequently, the final estimate of the fraction of the energy of nonthermal radiation in the overall balance of the released energy will be 0.06.

It should be noted that low-frequency electromagnetic fields during water freezing at frequencies below 1 MHz were previously recorded, for example in [14], however, their occurrence was interpreted as associated with the relaxation of spatial or surface charges. In addition, at the beginning of the supercooled water freezing, there is a sharp increase in the entropy production (entropy derivative with respect to time) due to a rapid release of energy. According to the thermodynamics of irreversible processes, in this case, ordered structures might appear [15], therefore, the electromagnetic radiation can be studied using these concepts. The same applies to the study of radiation during cavitation [2] and other weakly nonequilibrium processes [6].

5. Conclusions

1. The example of water crystallisation was used to show that the PeTa effect associated with the appearance of nonthermal radiation during the first-order phase transition is only possible in specially prepared systems. Due to a lowered value of entropy (and in the case of decreased energy barrier for the formation of solid phase nuclei), such systems should be in a metastable state. Upon the initiation of a phase transition, a nonequilibrium phase transition will occur in the system, during which nonthermal radiation may appear.

2. It was shown that in the specific case of the phase transition of deeply supercooled water, it is necessary to carefully study the issue regarding the generation of electromagnetic nonthermal radiations, whose characteristics depend on the entropy of the initial and final states. The same applies to any other media with their own unique properties. The entropy of the initial and final states is determined by the sample preparation procedure, the features of the phase transition, and the macro and micro structure of the medium in the initial and final states. These

features will determine a wide range of radiation characteristics (intensity, frequency spectrum, coherence, direction, and polarisation).

Conflict of interests

The author declares that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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