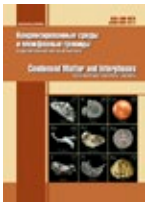

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$1/f^2$ noise as a precursor of structural reconstructions near the melting point of crystalline materials with different types of chemical bonds

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Abstract

Transition phenomena near the melting point (premelting effect) are fundamental processes occurring in materials with different types of chemical bonding. At $T \geq 0.8T_m$, there is a fluctuating heat release. The analysis of fluctuation processes allows obtaining information about dynamic reconstructions in various subsystems and interconnections between them within the system. The purpose of this work was to study the spectral characteristics of heat fluctuations in stationary modes of premelting materials with ionic, covalent, and metallic chemical bonds (KCl, Ge, Cu, and Sb) and to determine the type of the fluctuation process using the Hurst parameter.

The spectral characteristics of heat fluctuations in stationary modes of pre-melting KCl, Ge, Cu, and Sb at $T^* \sim 0.9T_m$ were determined by wavelet analysis. This method allows analyzing the behavior of complex systems at critical points in order to identify certain correlations and development trends in them.

The study showed that in the premelting region, the frequency spectrum of heat fluctuations was characteristic of $1/f^2$ noise or nonlinear Brownian noise, which is a precursor of structural reconstructions during phase transitions. The type of fluctuation processes in the premelting region of KCl, Ge, Cu, and Sb was determined using the Hurst parameter (H). It was shown that in stationary modes of premelting $H > 0.5$. Consequently, the previous trend of the process dynamics was very likely to develop in the same direction. However, with a decrease in the energy of the chemical bond $H \rightarrow 0.5$, which indicated a decrease in the stability of the system and a likely change in the development trend for the structural reconstructions in the premelting transition region.

Thus, near the melting point, there are unstable dynamic states, which are precursors to structural changes in the system, which have certain developmental trends. This should be taken into account when calculating the stability and reliability of materials and systems.

Keywords: Premelting, Fluctuations, Wavelet analysis, $1/f^2$ noise, Hurst parameter, Structural reconstructions

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1. Introduction

There is a large amount of data on $1/f^b$ noise in various complex systems [1–3]. Fluctuations occurring in the transition region during the first order phase transitions are especially interesting since such noise, in contrast to uniform distribution, predetermines certain correlations associated with the occurrence of a spatial or temporal ordering similar to a phase transition. Fluctuating precursors manifest themselves in the form of anomalous behavior of various macroscopic parameters: physical, electrophysical, and structural [4, 5]. This indicates that at certain critical points (bifurcation points) there is a loss of stability of the previous state accompanied by the emergence of dynamic dissipative structures. In this case, the system becomes sensitive to small changes in the initial conditions and fluctuations begin to play an important role. This phenomenon is known as "ordering through fluctuations" [6]. Thus, fluctuating precursors during phase transitions require specifying the degree of spatio-temporal localizations in the system and identifying new information about the behavior of the system in these nonequilibrium conditions.

The purpose of this paper is to study the spectral characteristics of heat fluctuations in stationary modes of pre-melting materials with different types of chemical bonding and to determine the type of the fluctuation process using the Hurst parameter.

2. Experimental

Our earlier studies of melting of materials with ionic, covalent, and metallic chemical bonds conducted by the DTA method at $T \geq 0.8T_m$ revealed temperature-temporal regions of premelting fluctuation phases [7].

The experiments were carried out using materials with different types of chemical bonds without phase transformations before and after the melting point: KCl, Ge, Cu, and Sb. Isothermal endurance (stationary mode) was carried out at $T^* \sim 0.9T_m$. The time of isothermal endurance was 35 minutes. What is more, the fluctuation state remained for a long time with slow relaxation to equilibrium, which is characteristic of dissipative processes.

For the spectral analysis of fluctuation processes and parameterization of the premelting phases of materials with different types of chemical bonding in stationary modes, we used the method of continuous wavelet transform [8]. Spectral analysis of fluctuation signals was carried out using the MatLab software environment with the Symlet8 basic function.

3. Results and discussion

The study considered the impact of the chemical bonding on the spectral characteristics of heat fluctuations in the premelting region of KCl, Ge, Cu, and Sb. Heat fluctuations in stationary modes are most interesting in terms of obtaining information on the regularities of the evolution of dynamic transition states during melting since in these premelting modes there is a long-term sequence of correlated fluctuation bursts producing nonlinear Brownian noise. What is more, such spectral parameters as the self-similarity coefficient (β) and the intensity of heat fluctuations provide information on the dynamics of the formation of transient phases.

As an example, we considered the wavelet transform pattern $W(a,b)$ of the premelting effect of Cu in the stationary mode at T^* = 1,221.8 K (Fig. 1). In this pattern, a hierarchical self-similar structure of local extrema was visible at different scales. The branching of the local extrema pattern corresponded to the rescaling of fluctuations and the transition to another level of the process. In general, this pattern of wavelet transform of heat fluctuations in KCl, Ge, and Sb, as well as Cu, is characteristic of signals of nonlinear Brownian noise or $1/f$ noise$^\beta$, which indicates correlations in the premelting transition regions. [9]. This was also confirmed by the statistical, correlation, and Fourier analysis which were carried out in [10]. They showed that fluctuation processes in the premelting region are random processes with normally distributed correlated increments.

The values of the spectrum coefficient β of heat fluctuations of transient processes during KCl, Ge, Cu, and Sb melting at isothermal endurance are given in Table 1. The self-similarity coefficient β ~ 2 indicated that dissipative processes in the transition region during melting were of the same physical nature. It also indicated the generic

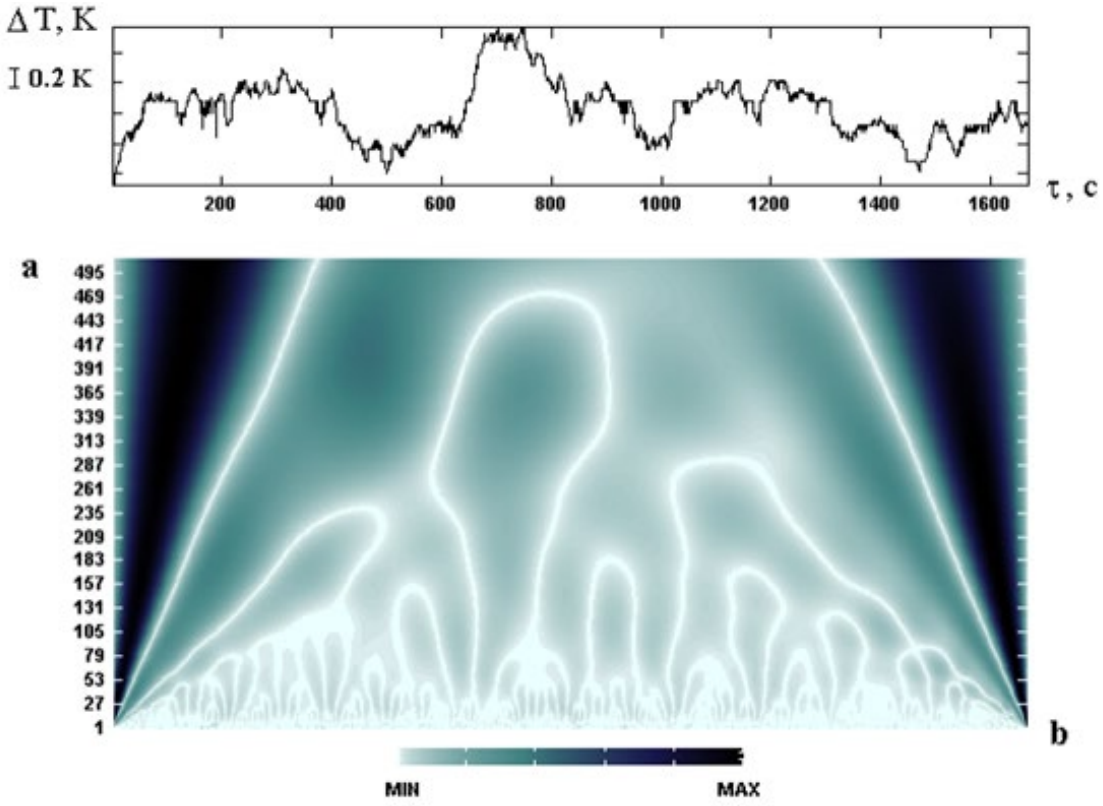

Fig. 1. DTA curve and wavelet transform of the fluctuation process of premelting of Cu (T^* = 1,221.8 K)

Table 1. Parameters of transient processes of premelting of KCl, Ge, Cu, and Sb in a stationary mode and the energy of the chemical bond

	T^*, K	β	H	ΔE, kJ/mol
KCl	940	2.1	0.58	423.2
Ge	1180	2.2	0.56	327.6
Cu	1220	2	0.55	306.7
Sb	814	2.1	0.52	165.8

nature of transient processes of premelting of crystalline materials with different types of chemical bonding. This indirectly reflected the occurrence of structural reconstructions in the materials in the premelting phase.

The energy spectrum $E_w(a)$ of heat fluctuations of transient processes of premelting KCl, Ge, Cu, and Sb in stationary modes is shown in Fig. 2. The analysis of the energy spectrum showed that with a decrease in the energy of the chemical bond from KCl to Sb, there was a decrease in the intensity of the fluctuation process in the pretransition region. The change in the intensity of heat fluctuations reflected the varying degree of correlation in the system. For example, a low value of the energy of chemical bond (ΔE) of Sb apparently resulted in emergence of long-time correlation bonds in the dynamic system. However, KCl, which had the largest energy of chemical bond among the studied group of materials, was characterized by the occurrence of short-lived correlation states.

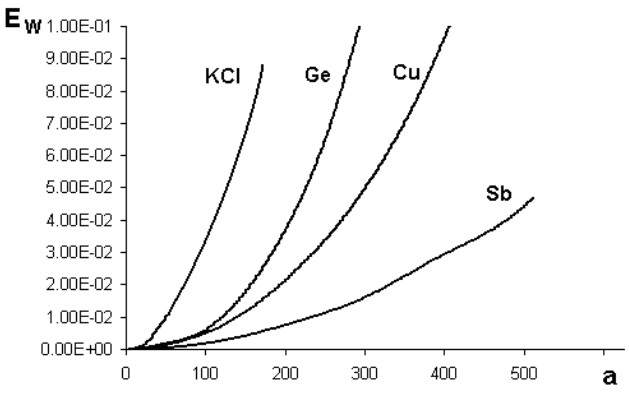

Fig. 2. Energy spectra of heat fluctuations in the premelting phase of KCl, Ge, Cu, and Sb in a stationary mode

Evaluation of the Hurst parameter allowed identifying certain hidden patterns or so-called trends in the fluctuation process in the premelting region. At a value of $H > 0.5$, the previous trend for the system transformation was highly likely to continue in the same direction. If $H < 0.5$, the direction of the trend was to change, while at $H = 0.5$ it was difficult to determine the direction of the trend [11, 12]. The Hurst parameter was calculated using RS-analysis.

For all types of the studied materials, the Hurst parameter was $H > 0.5$ (Table 1). In our case, the value of $H > 0.5$ indicated a persistent behavior of correlations in the system, i.e. the trend for the character of changes of the process persisted. However, a decrease in the energy of the chemical bond led to a decrease in the stability of the system and an approximation to uncertainty. This correlated with the change in the intensity of the fluctuation process within the premelting region in the series KCl, Ge, Cu, and Sb.

4. Conclusions

Regardless of the type and energy of the chemical bond, the pattern of the wavelet transform of the premelting heat fluctuations of Ge, Sb, and Cu in stationary modes is that characteristic of $1/f^2$ noise signals (non-linear Brownian noise). Thus, it can be stated that the processes that occur during nonequilibrium phase transitions in materials with ionic, covalent, and metallic chemical bonds have identical energy patterns.

With a decrease in the energy of the chemical bond in the series from KCl to Sb, the intensity of the fluctuation process in the pre-melting phase decreases, which indicates the determining influence of the energy of the chemical bond on the nature of formation and the fluctuation parameters of the premelting transient processes of various crystalline materials.

Evaluation of the chaotic state of fluctuation processes using a stochastic parameter, the Hurst (H) parameter, showed that a previous trend for structural reconstructions is not likely to change. Therefore, the analysis of the spectral characteristics of fluctuation processes near the melting point can provide information not only about the state of the system, but also can be used in methods aimed at predicting its evolution.

Conflict of interests

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

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