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The origin of the "Excess Wing" and $\beta\mbox{-relaxation}$ phenomena in glass-forming materials

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ABSTRACT

We suggest a model that allows to describe the β -relaxation and Excess Wing phenomena from the unified positions based on the hypothesis related to the self-similar (fractal) character of dielectric relaxation. It has been shown that these phenomena have similar nature but with different organization of relaxation channels. Additional arguments proving that additive combination of the well-known empirical functions forming an expression for the complex dielectric permittivity in frequency domain is not suitable for description of experimental data are given. In the frame of the model suggested we derive new functions for the complex dielectric permittivity that describe well the experimental data where two phenomena mentioned above (β -relaxation and Excess Wing) take place.

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1. Introduction

In various extended reviews devoted to description of the broad band dielectric spectroscopy specialists give a picture (Fig. 1) from the review [1] showing different types of relaxation processes that are observed in glass-forming materials. It is necessary to note that this behavior of relaxation processes is typical for many another complex systems [2]. In this paper we concentrate our analysis of the imaginary part of the complex dielectric permittivity (CDP) in the frequency range $10^{-2} \div 10^{9}$ Hz because many dielectric measurements are realized in this range. As it follows from Fig. 1 in this frequency range two types of spectra are differentiated: (A,a) – when imaginary part of the total dielectric response has only one peak (usually defined as α – peak) and its high-frequency branch (located on the right-hand side) decreases more slowly in comparison with lowfrequency wing of this peak. This phenomenon in literature is usually defined as Excess Wing phenomenon (EWP) [10,17,18]; (B,b) when in the frequency dependence of the imaginary part of the CDP more than one loss peak is appeared. It is accepted to mark these loss peaks by Greek letters e.g. β , γ , δ -relaxation peaks (sometimes to β – relaxation peaks the prefix "slow" is added in order to differentiate it from the "fast β -relaxation process," which is essential in high-frequency range). It is commonly assumed that spectra of A-type and B-type describe quite different phenomena [3,4].

The dielectric loss spectra of type B are observed frequently in various polymers and low-molecular glasses [5,6]. Many researches describe these spectra by an additive combination of empirical functions

* Corresponding author. E-mail address: ii.popov@yandex.ru (I.I. Popov). as Cole–Cole (CC), Cole–Davidson (CD) or Havriliak–Negami (HN) expressions. Usually, this linear combination is formed from HN functions (see, for example [7–9]). Mathematically, any dielectric response is presented in the form

$$\varepsilon(\omega) = \varepsilon_{\infty}^{HN} + \sum_{k} \frac{\Delta \varepsilon_{k}^{HN}}{\left[1 + (i\omega\tau_{k}^{HN})^{\alpha_{k}^{HN}}\right]^{\beta_{k}^{HN}}}.$$
 (1)

This presentation is related to the fact that the HN function contains a sufficient number of the fitting parameters and as a partial case includes in itself more simple CC and CD functions. Definitely, linear combination (1) enables to fit a dielectric response of any complexity because a strict criterion that can limit the number of HN functions in this combination is absent. As one can see from (1) any new HN function figuring in combination (1) adds four additional fitting parameters (α_k^{HN} , β_k^{HN} , τ_k^{HN} , $\Delta \varepsilon_k^{HN}$). Such kind of "logic" came to dielectric spectroscopy from the resonance spectroscopy, where all peaks in the most cases are well-resolved from each other. But in dielectric spectroscopy the situation is different because the imaginary part of the CDP is formed by wide peaks that are not well-resolved relatively each other. So, this "conventional logic" needs essential corrections.

The A-type relaxation spectra are reminded in early Cole and Davidson papers [10] and are observed frequently in glass-forming materials [3,4,11–14]. Many researches try to describe these A-type spectra by linear combination of two HN functions or with inclusion of its partial cases representing the CC and CD functions (see, for example, [2,15]). Other attempts are based on combination of the Kohlrausch-Williams–Watts (KWW) functions (see, for example, [16]). Also some

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