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Langevin approach to the theory of dielectric relaxation of ice Ih

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HIGHLIGHTS

- A new phenomenological model of the dielectric relaxation of hexagonal ice was developed.
- The origin of the low-temperature crossover of the relaxation time of ice Ih was explained.
- A new expression for the complex dielectric permittivity of ice was obtained.

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ABSTRACT

Within the Langevin approach a new phenomenological model of dielectric relaxation of the ice is developed. This model is based on the concepts of defect migration, which is the main mechanism for dielectric relaxation of the ice. The new model allows to describe the relaxation behavior of the ice over a wide temperature range and to explain its characteristic features: changes in the slope of relaxation time at high and low temperatures ("crossovers"), and the non-monotonic temperature dependence of the broadening of dielectric loss peak parameter. The "crossover" of relaxation time at high temperatures is due to the transition from the predominant motion of the orientational defects of Bjerrum to the preferential motion of ionic defects with decreasing temperature and the weak correlation between them. On the contrary, at low temperatures a strongly correlated motion of ionic and orientational defects arises, which causes the observed low-temperature "crossover".

1. Introduction

Despite the intensive experimental and theoretical studies of electronic properties of the simple hexagonal ice Ih during the last century [1–17], up to now, there has been no unified understanding of physical mechanisms that determine the observed temperature dependence of relaxation behavior. The hexagonal ice is a simple crystalline system with hydrogen bonds. Its structure is well known. However, its dielectric relaxation behavior is quite complex.

The main dielectric loss peak in the hexagonal ice is symmetrically broadened below 240 K, and it can be described by the Cole–Cole expression for the complex dielectric permittivity (CDP) $\varepsilon^*(\omega) = \varepsilon_\infty + (\varepsilon_s - \varepsilon_\infty)/(1 + (i\omega\tau)^\gamma)$, where ε_∞ is the high-frequency permittivity limit, ε_s is static permittivity, τ is characteristic dielectric relaxation time, and γ parameter defines a broadening of dielectric loss peak ($\gamma < 1$). Therefore, a spectrum of relaxation times is observed in the system at the temperature range below 240 K [18–20]. Above the temperature of 240 K, only one relaxation time was detected, and therefore, the basic peak of the imaginary part of the dielectric permittivity has the Debye form ($\gamma = 1$). In

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