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TESTING SOME ALTERNATIVE MODELS FOR COMPLEX LOW-ENERGY EXCITATION RELAXATION IN DENSITY WAVE SYSTEMS[1]

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Dedicated to Professor Boran Leontić on the occasion of his 70th birthday

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We apply the Palmer, Stein, Abrahams and Anderson (PSAA) model of hierarchically constrained dynamics for glassy relaxation to the complex thermal relaxation at very low temperatures in density wave systems. Alternatively, we simulate various experimental conditions in a simple, intuitive model of an electrical RC line and find some relations with the PSAA parameters.

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1. Introduction

Nonexponential relaxation is found in extremely diverse physical systems such as structural glasses, spin glasses, polymers and random systems in general, all of which can be classified as complex [1]. More recently, pinned density-wave (DW) systems with either charge or spin modulation have been added to this list [2]. Although a wide variety of mechanisms can produce nonexponential relaxation patterns, such as stretched-exponential, logarithmic or algebraic ones, it seems that the relaxation in complex systems has a considerable degree of universality

[1] This article is based on the essay "Primjena hijerarhijskog modela na opuštanje u sustavima s valom gustoće spina" (in Croatian) of Andraš Kiš and Domagoj Pavičić, students at the Department of Physics. For the essay, they received the 1997 University of Zagreb Rector's Prize.

and essentially does not depend on the specific microscopic background. Therefore, a more sophisticated global approach is needed.

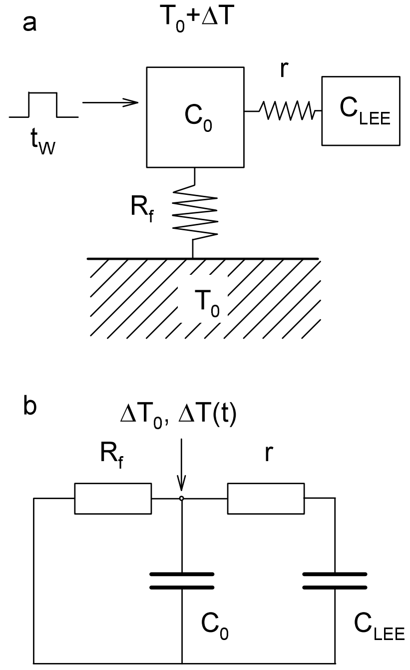
Conceptually, the simplest phenomenological approach is to consider the non-exponential relaxation through a spectrum of relaxations due to non-interacting units each of which relaxes with a different intrinsic time constant. However, this approach is in disagreement with the observation that interactions play an important role in all systems exhibiting nonexponential relaxation, introducing certain hierachy in succesive excitation of various degreees of freedom. We applied two "hierarchical" models to describe the complex thermal relaxation observed at very low temperatures in a spin density wave system.

First, we briefly present the experimental conditions, procedure and basic results that led us to consider some new models. Afterwards, we introduce a model of heat diffusion in our system through voltage relaxation in an equivalent electrical circuit consisting of a series of resistors and capacitors. Finally, the well-known Palmer, Stein, Abrahams and Anderson (PSAA) model of hierarchically constrained dynamics developed for glassy relaxation [3] is applied to the relaxation curves at various temperatures.

2. Description of experimental conditions

A series of heat-pulse experiments made at temperatures below 1 K in a dilution cryostat in CRTBT-CNRS on various density wave systems, including both charge density waves (CDWs) and spin density waves (SDWs) yielded unusual pulse decays [2]. In our experimental technique, the sample is loosely connected through a thermal link (R_f) to the cold sink regulated at some temperature T_0 (Fig. 1.). The sample itself is in the form of a great number of needles (of total 100 mg mass) smoothly pressed between two silicon plates (of an area of 2 cm^2), with a small amount of Apiezon N grease added to improve thermal contact.

The mesurements are performed by applying short heat pulses to the sample and the heat capacity is calculated from the time dependence of the sample temperature during and after the pulse. This procedure is simple as long as the relaxation is exponential (Fig. 2a), but for temperatures below 1 K, the heat pulse decay of DW systems becomes non-exponential, indicating a new, very slow contribution, as shown in Fig. 2b. Moreover, the underlying dynamics [2], and consequently specific heat [5], strongly depend on the time duration of the heat pulse, i.e. the heating time, as shown in Fig. 3, and this feature becomes more pronounced as the temperature is lowered (see Fig. 4). These new properties are attributed to some kind of low-energy excitations (LEEs) represented with an additional $C_{\rm LEE}$ (see Fig. 1b). To account for the observed phenomena, there should be a substantial "lag" between the temperature that we measure (which is in fact that of fast relaxing degrees of freedom, or phonons, see Fig. 1) and the "temperature" (considered as a measure of excitation) of LEE subsystem. Therefore, it seems that the "slow" LEE subsystems are, contrary to other glassy systems, only weakly coupled to the fast phonon subsystem.



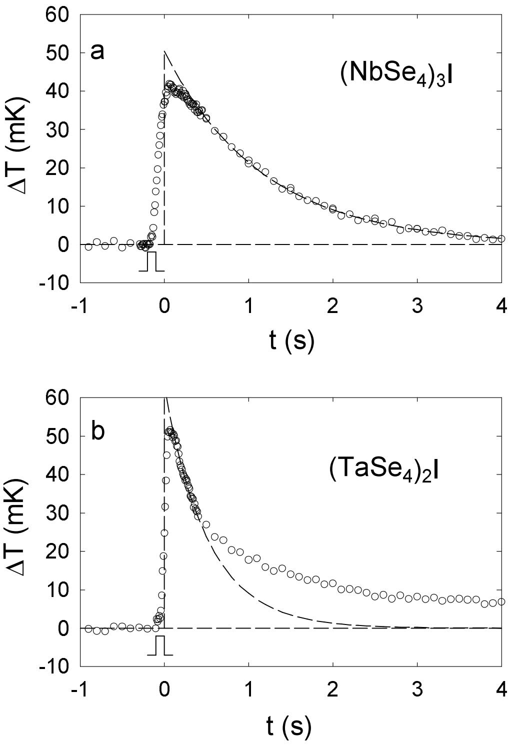
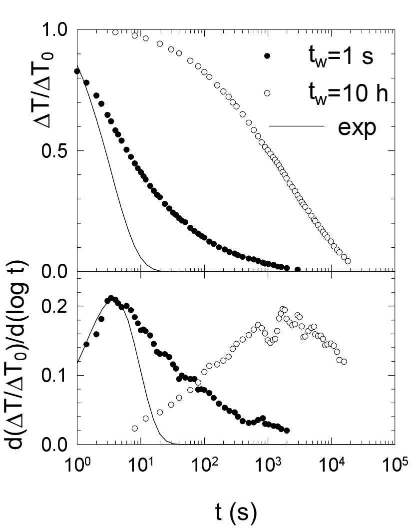


Fig. 1. Schematic representation of the method: a) R_f is the thermal link of the sample to the thermal bath, C_0 is the regular part of specific heat, weakly coupled through internal thermal link r, to the LEEs. Their specific heat contribution $C_{\rm LEE}$ shows strong time dependence. b) Simplest equivalent electrical circuit where R_f represents the main thermal link, C_0 the fast modes (phonons and addenda) and $C_{\rm LEE}$ the slow modes, long-living metastable states (taken from Ref. [2]).

Fig. 2 (right). Thermal transients after a short pulse at $T = 0.11$ K in stucturally similar compounds: a) $(\mathrm{NbSe_4})_3\mathrm{I}$ without CDW metastable states as referent system with a simple exponential decay (dashed lines). b) $(\mathrm{TaSe_4})_2\mathrm{I}$ in glassy CDW ground state exhibiting long-time nonexponential decay due to LEEs. (taken from Ref. [4]).

The LEEs in DW systems at low tempertures naturally appear in the dynamics of pinned topological defects of DWs, such as solitons or dislocation loops. Interaction with random impurities produces a number of metastable states very close in energy, but far apart in phase space. Therefore, the transiton between different states is energetically feasible, but takes a substantial time, which accounts for the observed phenomena. Recently, a microscopic model of the dynamics of LEEs in DW systems at very low temperature has been developed [7]. It has been shown that a single DW soliton interacting with an impurity can be in two (meta)stable states and the whole problem is treated as a distribution of two-level systems. Unfortunately, it does not account for the hierarchical relaxation we are considering here.



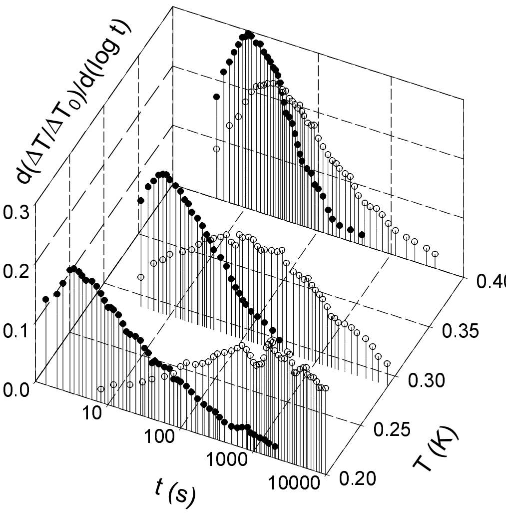


Fig. 3. a) Relaxation of sample temperature after the heat perturbation with a short pulse ($\leq$ 1 s) and 10 hours long heating with $\Delta T/T_0 \leq 10\%$. The sample is SDW system $(TMTSF)_2PF_6$. *Solid line indicates exponential relaxation with the same mean relaxation time (see b). b) The relaxation rate is obtained as the logarithmic derivative of the relaxation curve and reflects the form of a relaxation time distribution [5]. The position of the maximum gives the mean relaxation time. It shifts to longer time for longer duration of the heat flow and gets wider.*

Fig. 4 (right). Time dependence of the relaxation rate gets more pronounced at lower temperatures (the same sample and conditions as in Fig. 3) for three different temperatures.

3. Simulation by the electrical analogue

We have presented in Fig. 1 an equivalent RC circuit of our experimental setup for the heat-capacity measurements. Heat is accumulated in heat capacitors and relaxes through thermal resistors. To account for the time dependence of heating (as evidenced in Fig. 4), the capacitor representing LEEs should be time-dependent. However, similar properties can be obtained by introducing a more complex, but time-independent RC scheme. One simple way to describe the non-exponential decay would be to consider the relaxing system as an assembly of exponentially relaxing units having different sizes and relaxation times τ. The relaxation function can then be expressed as the summation over all units,

$$\mathcal{R}\,(t) = \int w(\tau)\exp(-t/\tau)d\tau \tag{1}$$

where $w(\tau)$ is a suitable weight function. This approach assumes that units are independent, relaxing in *parallel*. The general notions concerning disordered systems with many metastable states show that a proper approach should take into account the hierachy in succesive excitation of various degreees of freedom ([3] and references therein). Therefore, the corresponding equivalent RC circuit should be in series, and this is the scheme we adopt in this work, as presented in Fig. 5.

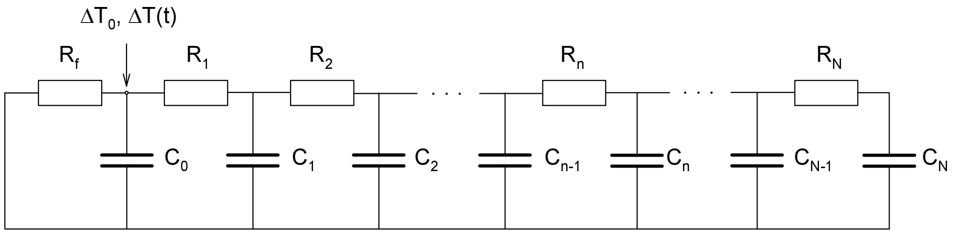


Fig. 5. Schematic presentation of the infinite RC chain (a linear generalisation of the scheme in Fig. 1b for a serial arrangement).

We keep thermal link R_f and capacity of fast degrees of freedom C_0 and represent the LEE's contribution through a (R_n, C_n) chain. In this approach, C_n would correspond to the size of different subsystems at different levels. The rate of energy transfer between different levels would be regulated via R_n. We can easily set charging and discharging equations for such a system. For the n-th capacitor, we have

$$I_n = \frac{U_{n-1} - U_n}{R_n}, \quad I_n - I_{n+1} = \frac{dQ_n}{\mathrm{d}t}, \quad C_n = \frac{Q_n}{U_n}\,, \tag{2}$$

where I_n, U_n and Q_n represent the flow through the resistor R_n, temperature difference accross the condensor C_n and heat stored in condensor C_n, respectively. In this way we get a system of N coupled linear differential equations of the first order which can be numerically solved for eigenvalues and eigenvectors. Once these are known for a given (R_n, C_n) set, the heat stored during constant temperature application (setting T_0 to a desired value), as well as heat released after the heating is switched off (letting T_0 free with the capacitors already charged), can be calculated.

In order to avoid the "size effect", we treated the cases with $N > 50$ capacitors in series. We have tried various monotonous functions for R_n and C_n such as linear, exponential and logaritmic. We got relatively good results for the simple case of $R_n = nR$ and $C_n = C$ (except for R_f and C_0 which are varied separetely). This configuration corresponds to the equivalent subsystems of equal weights, with a linear increase of the "constraint" for the heat transfer between the levels. The

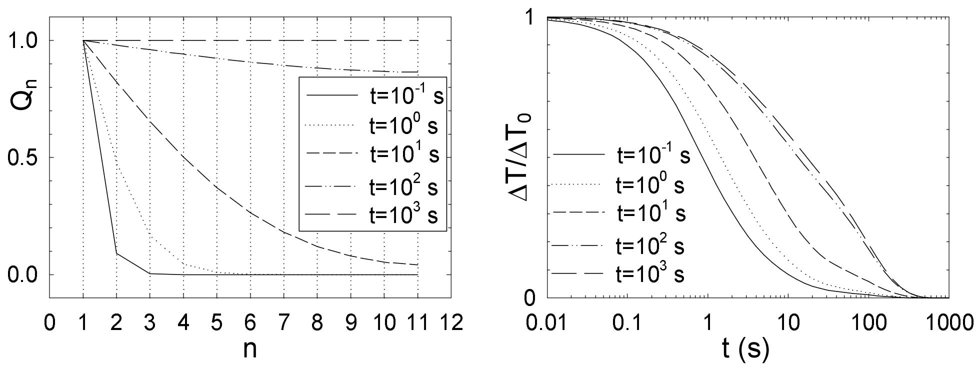


Fig. 6. a) The dependence of the normalized charge Q_n of respective capacitors on heating time. b) The corresponding decay of the stored heat resembles the relaxations observed experimentally (Figs. 3 and 8). The following parameters are used: $R_f = 5$, $C_0 = 1$, $R = 1$ ($R_n = nR$) and $C = 1$ ($C_n = C$).

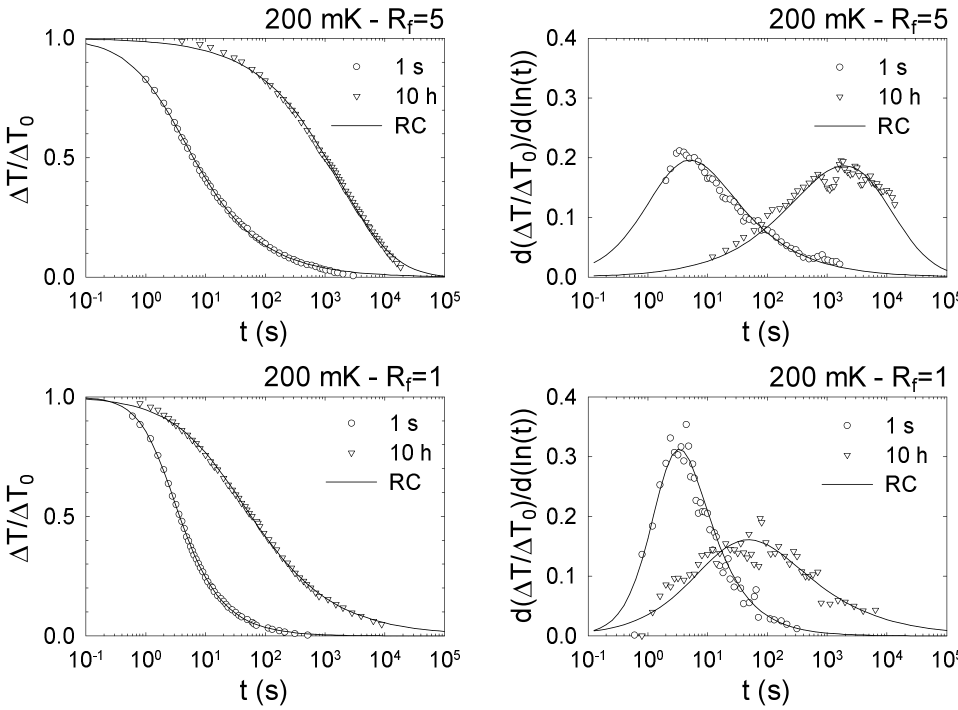


Fig. 7. Our experimental results compared with the simulations for different durations of heating (charging), with configuration $R_n = nR$ and $C_n = C$. Upper row is for the value of the thermal link 5 times higher, $R_f^1 = 5R_f^2$. Real experimental values at $T = 0.2$ K have been $R_f^1 = 5\,10^8$ K/W and $R_f^2 = 10^8$ K/W.

amount of heat stored in the respective capacitors depends strongly on the heating time, and so does the temperature decay, as can be seen in Fig. 6.

Figure 7 shows the simulation of our results for two different thermal links used in two different experimental runs at the same temperature. Actual values of the parameters used are $R_f^1 = 40$, $R_f^2 = 8$, $C_0 = 2$, $R = 1.1$ and $C = 1.4$. The decay dynamics depends only on the relative value of resistors and capacitors, i.e., if we multiply all the values of resitors by a number, and divide the values of the capacitors by the same number, we get the same relaxation curves.

We stress that the values of C, R and C_0 are kept constant between two simulations, and only the value of R_f is changed. Therefore, we have been able to account for the complex heat relaxation, including the heating time dependence and R_f influence, by a relatively simple RC circuit described by only four independent parameters.

4. Application of hierarchically constrained PSAA model

The successful application modeling of nonexponetial relaxation with the RC series led us to consider in some detail a more general approach which naturally incorporates this principle. In their paper [3], Palmer, Stein, Abrahams and Anderson (PSAA) have developed a hierarchical model that gives a universal mechanism controlling complex dynamics which does not depend on microscopic details. Their theory involves hierarchy of degrees of freedom, so that faster degrees of freedom successively constrain slower ones, as would happen in a system with a strong interaction.

The PSAA model develops expressions for a process of relaxation of a multilevel system of N Ising spins (or pseudospins) S_i. It is completely abstract and does not specify any model for the levels of the system, except the requirement that spins in one level constrain spins in the level immediately above it. Nevertheless, at high temperatures, the constraints must disappear, and a single relaxation time is recovered.

The number of spins in level n is N_n. Each spin in level $n+1$ is only free to change its state if μ_n spins in level-n attain one particular state of their 2^n possible ones. The relaxation times are related by

$$\tau_{n+1} = 2^{\mu_n}\tau_n \tag{3}$$

leading to

$$\tau_n = \tau_0 \exp\left(\sum_{k=0}^{n-1} \widetilde{\mu}_k\right) \tag{4}$$

with $\widetilde{\mu}_k = \mu_k \ln 2$. Total spin at level n is $N_n \exp(-t/\tau_n)$ and the relaxation function is

$$\mathcal{R}(t) = \sum_{n=0}^{\infty} w_n e^{-t/\tau_n} \tag{5}$$

with $w_n = N_n/N$ and $N = \sum_{n=0}^{\infty} N_n$. To avoid factors of 2, the probability per unit time of flipping a level-n spin is taken as $2/\tau_n$.

The theory contains two unspecified functions, μ_n and w_n (or N_n). So far we have used a stretched-exponential function (or Kohlrausch phenomenological law [8]) with the power-law pre-exponential factor (as it has been widely used in spin glasses [9]) to describe non-exponential decay $\Delta T(t)$ [2,4]

$$\frac{\Delta T(t)}{\Delta T_0} = A \left(\frac{t}{\tau}\right)^{-\alpha} \mathrm{e}^{-(t/\tau)^{\beta}} . \tag{6}$$

Therefore, we have chosen a set of μ_n and w_n that, according to authors of Ref. [3], results in a relaxation function barely distinguishable from the exact Kohlrausch form, i.e., the power law form $\mu_n = \mu_0 \, (n+1)^{-p}$ and $N_{n+1} = N_n/\lambda$, where λ is a constraint greater than 1 (or $w_n = \lambda^{-n}$). So, the relaxation function which we used to fit our results (by simplex Nedler-Mead method) is

$$\mathcal{R}(t) = w_0 \sum_{n=0}^{\infty} \lambda^{-n} \exp\left(-t/\left(\tau_0 \exp\left[\widetilde{\mu}_0 \sum_{k=1}^{n} k^{-p}\right]\right)\right) \tag{7}$$

with five fitting parameters, w_0, λ, τ_0, $\widetilde{\mu}_0$ and p.

We show in Fig. 8 our experimental data for the heat release at six temperatures and for different durations of the heat input in $(\mathrm{TMTSF})_2\mathrm{PF}_6$, which is a SDW prototype system. The main characteristics of the measured relaxations are non-exponentiality and a strong shift towards longer times with the increased duration of the heat input. Both effects progressively disappear at higher temperatures at which a dominant role is taken over by phonons.

Our fits are in very good agreement with the data, what is not surprising since one uses five parameters. Though it is our first probing of PSAA model, it gives some important information about the temperature and time dependence of parameters (Fig. 9), which is a good starting point for further searching for appropriate functions.

λ is a relative weight of the hierarchic level and shows no dependence on the duration of the heat pulse, but changes abruptly above 400 mK. This indicates that at higher temperature higher hierarchical levels do not influence the relaxation or, stated differently, their number is reduced as the interaction between neighbouring levels disappears.

One can not easily understand the behaviour of τ_0, the parameter representing the characteristic relaxation time, as it is partly given by experimental arrangement and depends on the thermal link we used. This point has been discussed in the previous section, where we have simulated two different conditions. Least we can say here is that this influence is less pronounced at the lowest temperature, where the characteristic intrinsic relaxation time is much longer than the time corresponding to the external thermal link, and scales with duration of the heat input (real effect of "aging" [2,9]).

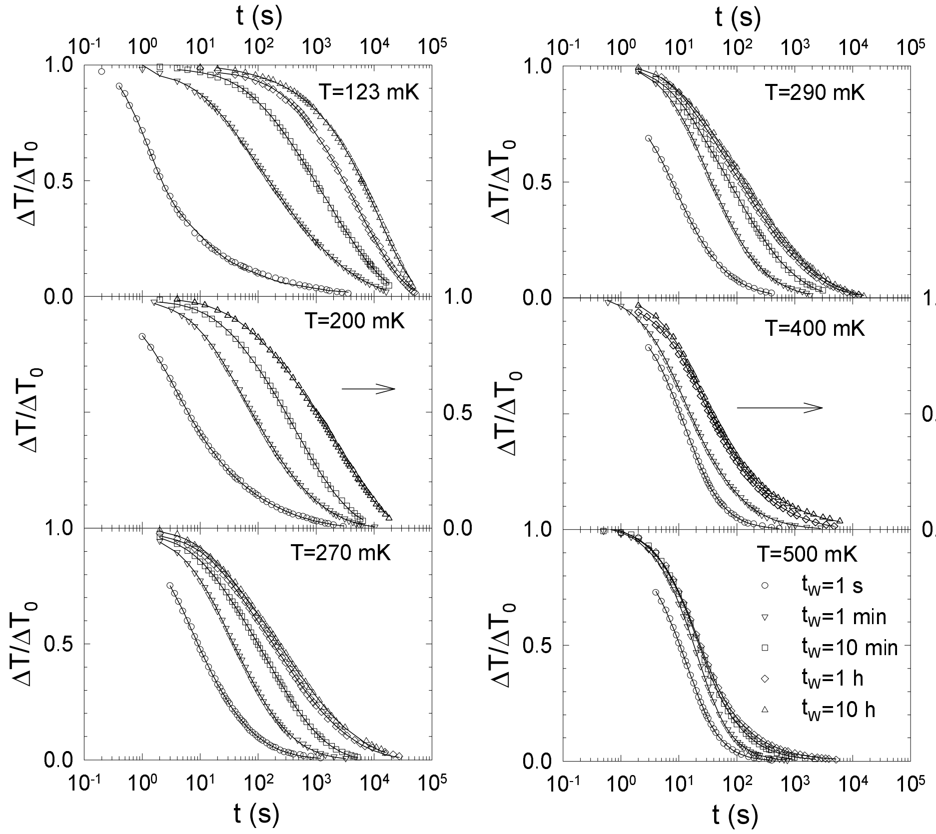


Fig. 8. Heat relaxation at very low temperatures for SDW prototype system $(TMTSF)_2PF_6$ *for different durations of the heat input. Corresponding PSAA fits show a very good agreement with our experimental results.*

Parameter μ_n is related to the number of events at the n-th level preceding the relaxation at the $n+1$ level, and our first choice was $\mu_n = \mu_0(n+1)^{-p}$. As μ_0 is almost constant (close to 1), which is normal for the normalizing factor, p is the parameter which controls the temperature and time dependence. It indicates that something intrinsically changes in this system at $T \simeq 300$ mK. Obviously, further investigation of the most appropriate functional forms is needed. However, as the experimental conditions influence the dynamics, first we should be able to separate the intrinsic dynamics from the apparent one.

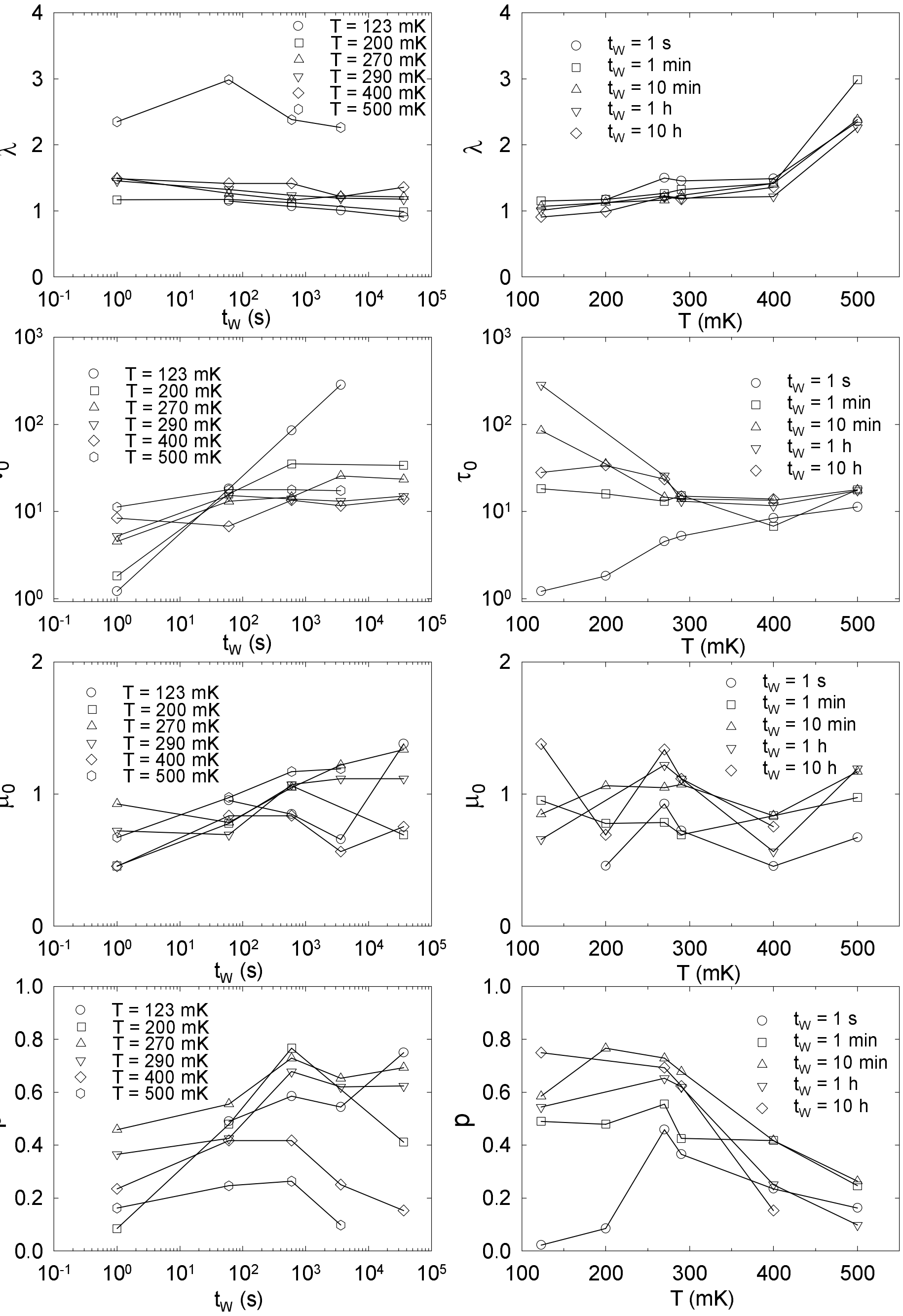


Fig. 9. PSAA fitting parameters as a function of the temperature and duration of the heat input.

5. *Conclusion*

We have demonstrated the first application of the well-known PSAA model of hierarchically constrained dynamics for glassy relaxation to our data of a very unusual heat release below 1 K in a SDW system. There is a good agreement between our results and the obtained fits.

However, the complex, long-time relaxation caused by LEEs of the DW is somehow coupled to the normal (fast) degrees of freedom, as regular phonons and the experimental thermal link. Consequently, the obtained parameters have hidden intrinsic properties of the underlying excitations. Our simple simulation by the electrical circuit, equivalent to the experimental heat transfer through the system, bears very good results, especially when the influence of the major thermal link on the overall relaxation of the system is considered. Further investigation is needed for the separation of the slow (complex, disordered) part from the fast (normal) part of our system, what should make possible a proper characterization of the LEE thermal response function. It gives a good basis for approaching a more specific microscopic model governing this exceptional physical phenomenon.

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ALTERNATIVNI MODELI ZA SLOŽENO OPUŠTANJE NISKOENERGIJSKIH POBUĐENJA U SUSTAVIMA S VALOVIMA GUSTOĆE

Primjenjujemo model Palmera, Steina, Abrahamsa i Andersona (PSAA) za hijerarhijski zapriječenu dinamiku opuštanja u staklima na kompleksno opuštanje topline na vrlo niskim temperaturama u sistemima s valovima gustoće. Jednako tako, pomoću jednostavnog modela slijeda električnih RC (otpor – kapacitet) krugova oponašamo različite eksperimentalne uvjete i nalazimo neke odnose s dobivenim PSAA parametrima.