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A new approach to diffusion-like relaxation processes

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ABSTRACT

We measured the time decay of the electric field in two dielectric non-crystalline solids and the data were analyzed in the framework of a new theoretical approach. It is observed that the application of this approach allows identifying different regimes in the relaxation process. At long times the dynamics end up by following a power law decay, which suggests the existence of a scale property in the interaction that defines the state of the materials and also the possibility of defining an invariant. We check the stability of this invariant under different time intervals, which means also a low dependence of it on noisy data.

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1. Introduction

Solids (crystalline and non-crystalline) are systems that can be driven out of equilibrium with an external perturbation that changes any of their (measurable) magnitudes. What follows is a variation in time of that magnitude, i.e., a relaxation. The analysis of those relaxations, especially those with long-term decay behavior, is a well defined area of research.

The way under which the relaxation takes place gives clues about fundamental properties of the solid. In fact, the action of the perturbation can be tailored appropriately in order to test specific properties, e.g., magnetic ageing that would point to frustrated and disordered magnetic systems [1]. The problem is that, when facing the analysis, it is usually found that the data can be fitted to several models [2] with a similar degree of confidence, i.e., no conclusion on the physics that underlie the relaxation can be obtained.

We have recently proposed new ideas to identify correctly under which law a relaxation is produced [3]. In this paper, we are studying the dielectric relaxation of two types of non-crystalline solids under this approach, in order to try to infer anything about the nature of the interaction that describes their evolution in time.

2. Materials and methods

The materials studied were polyacrylic acid, a dielectric amorphous material supplied commercially under the name of Carbopol 907, and an acetal-based polymer used as a protection against static charges due to its dielectric properties. The samples were prepared as disks that were polished and painted with a conductive coat of graphite. They were introduced into a capacitor with a 1 mm gap. Then a current of 1 μ A was applied during 20 μ s at room temperature. From the potential created we calculated the electric field on the sample. The variation of this electric field in time was then measured with a sampling frequency of 100 kHz during 10 s for acetal and 1 s for polyacrylic acid; both are long times for these kind of processes.

3. Results

In Fig. 1(a), we show the time decay of the electric field vs. time in acetal. Our way to explore the data is to replot them in the form dE/dlnt vs. E [3]. With this, the whole relaxation process can be viewed in a finite window. This manoeuvre, that yields the graph of Fig. 1(b), is based on the assumption that the evolution of the electric field in time is towards an equilibrium state where it can be described by a function of time that contains parameters that are time-independent. Then, in order to get to a reference point, we hypothesize that the physics governing the evolution are diffusion-like, i.e., the time derivative of the interaction that describes the microscopic state of the sample depends on the spatial-like dependence of that interaction (let us say, dependence on spatial coordinates or other coordinates used for the description of the material). In such a generic case, which covers many of the possible scenarios in solids, we have demonstrated analytically that the decay should end up by following this power law [3]:

$$E = E_0 \left(1 + \frac{t}{\gamma \tau} \right)^{-\gamma},\tag{1}$$



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Fig. 1. (a) Electric field vs. time for acetal. (b) Plot allowing the identification of three relaxation regimes. (c) Values of the invariant Γ , obtained after fittings to a power law, vs. the variation of the time interval used for the fit (t_1 , 10 s). Fits are indicated by solid lines.

where γ and τ are constants dependent on the sample and $E_0 = E(0)$. Note that for γ tending to infinite Eq. (1) would be a exponential law (a Debye process). Our aim with the representation of Fig. 1(b) is to found whether the electric field is decaying as a power law or as one of the more used laws, the stretched exponential:

$$E(t) = E_0 e^{-\left(\frac{t}{\tau}\right)^{\beta}}.$$
(2)

To understand this, let us come back to our representation: we see that this stretched exponential looks like:

$$\frac{\mathrm{d}E}{\mathrm{d}\ln t} = \beta E \ln \frac{E}{E_0},\tag{3}$$

whereas the power law, under the same operation:

$$\frac{\mathrm{d}E}{\mathrm{d}\ln t} = -\gamma E \left[1 - \left(\frac{E}{E_0}\right)^{\frac{1}{\gamma}} \right]. \tag{4}$$

We note now that, in the framework of our plot, the slope of Eq. (3) tends to infinite at very long times, not in the case of Eq. (4). Also, processes described by Eq. (3) would have a minimum at 1/e, not in the case of Eq. (4). Therefore we have arrived to a graphical representation that allows distinguishing which model is present in our case. The strength of the method lies on the possibility of observing globally the whole process from t = 0 to infinity, it is an extensive comparison, instead of the typical local one.

In our case, from Fig. 1(b) we see that there are three recognizable regimes. In this way it is possible to identify them and to observe how, after some time, the electric field follows a power law. This, according to a previous theoretical approach [4], suggests that the interaction that rules the microscopic configuration of the material fulfils a scale property. We also demonstrated that, in the power law regime, a parameter $\Gamma = E_0^{1/\gamma} \tau$ is invariant under changes of time origin. After computer simulations it was also observed that it is not dependent on the initial conditions of the system [4]. We have tested the strength of this parameter against different time intervals used in data fit. In Fig. 1(c), we present the variation of Γ vs. the amplitude of the time interval used in the fit (the considered time intervals start at time t_1 and finish at the last time measured, 10 s in this case). As the decay does not follow the power law for low times, the fit gives changing values of Γ but, once the power law regime is attained, it ends up by keeping quite a constant value. This means also that this parameter is robust against the noise of the data. This can be compared with the same variation observed for the fitted τ 's which shows a higher dependence. For polyacrylic acid the relaxation also ends up by following a power law. Also in this case, the calculation of the



Fig. 2. Values of the invariant Γ , obtained after fittings to a power law, vs. the variation of the time interval used for the fit (t_1 , 1 s). Data of τ , extracted from Fondado et al. [3], are included for comparison.

invariant Γ allows us to observe its relatively small dependence on the amplitude of the time interval used for the fits (Fig. 2).

4. Conclusion

We observe that for both polyacrylic acid and acetal the dielectric relaxation ends up by following a power law, with no ambiguity. This is done after the application of a graphical criterion that allows the identification of different regimes along the process. This, besides evidencing the robustness of the tool, suggests the presence of some scale invariance in the interaction that determines the microscopic state of the material.

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