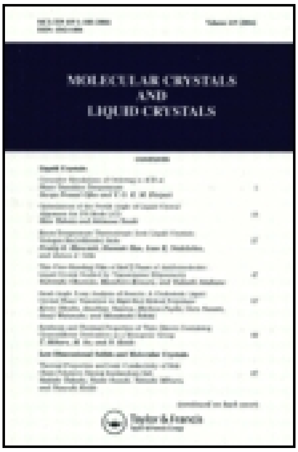

Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

Dielectric Relaxation as a Cooperative Process

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To cite this article: M. Massalska-Arodź (1992): Dielectric Relaxation as a Cooperative Process, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 214:1, 171-178

To link to this article: http://dx.doi.org/10.1080/10587259208037291

Mol. Cryst. Liq. Cryst., 1992, Vol. 214, pp. 171–178
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Printed in the United States of America

Dielectric Relaxation as a Cooperative Process

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(Received September 10, 1990; in final form September 15, 1991)

A comparison of the degree of cooperativity of orientational molecular motions in different kinds of (thermodynamical) phases is presented based on the (literature data of the) Dissado-Hill short and long-range correlation parameters n and m. A possibility of using fractal concepts for an interpretation of the power-law frequency dependence of susceptibility in a disordered medium is pointed out.

Keywords: dielectric relaxation, liquid crystals, plastic crystals, fractal-like scaling, short and long-range correlations

INTRODUCTION

Regarding the relaxation process as a cooperative process seems particularly adequate in the description of the dynamics of molecules of complicated shapes forming rotational disordered phases in all kinds of soft materials. In those cases reorientations of a molecule, especially around the axis with a large moment of inertia, are impossible without some cooperation with the near and more distant neighbours. There are various ways of taking that fact into account.[1,2] Dissado and Hill[3] proposed the following frequency dependence of electric susceptibility:

$$\chi(\omega) = \chi(0)\cdot\chi(1 + i\omega/\omega_{\max})^{1-n}\,{}_2F_1[1 - n, 1 - m; 2 - n; (1 + i\omega/\omega_{\max})^{-1}] \quad (1)$$

for the description of the non-Debye dielectric relaxation process. The ${}_2F_1$ is the hypergeometric Gauss function, $\omega_{\max}$ is the characteristic relaxation rate. In the high and low frequency limits the susceptibility $\chi(\omega)$ has the following power law frequency dependence:

$$\chi''(\omega) \sim [\chi'(\omega) - \chi'(\infty)] \sim (\omega/\omega_{\max})^{n-1} \qquad \text{for } \omega > \omega_{\max}, \quad (2)$$

$$\chi''(\omega) \sim [\chi'(0) - \chi'(\omega)] \sim (\omega/\omega_{\max})^{m} \qquad \text{for } \omega < \omega_{\max} \quad (3)$$

with the values of n and m parameters between 0 and 1. Such a form of "universal response" has been observed by Jonscher[4] in various disordered materials. The discrepancies between the n and m values for the isolated relaxation under study and $n = 0$ and $m = 1$ values characteristic for the Debye model are caused by the existence of some forms of ordering in the molecular system. The n and $1-m$ values are the measures of strength of the short and long range correlations of the molecular reorientations responsible for the relaxation observed. In the fractional power-laws, Equations (2) and (3), the real and imaginary parts of dielectric response are the same functions of frequency and therefore their ratio is frequency independent[5]:

$$\chi''(\omega)/[\chi'(\omega) - \chi'(\infty)] = \cot(n\pi/2) \qquad \text{for } \omega > \omega_{max}, \tag{4}$$

$$\chi''(\omega)/[\chi'(0) - \chi'(\omega)] = \tan(m\pi/2) \qquad \text{for } \omega < \omega_{max}. \tag{5}$$

These rules seem fundamental for governing the energy dependences during the relaxation progress.[3,5]

In the paper the n and m parameters are used to compare the degree of cooperativity of the specific molecular motions in polar materials having some rotational freedom. The attempts are made to consider the influence exerted on dielectric response by various factors ordering molecules in liquid, liquid-crystalline, plastic and glassy phases. The universality of the power-law behaviour of dielectric response is explained by the occurrence of some forms of hierarchy in structural organisation of the medium, in turn forcing the hierarchy of microscopic relaxation processes. The correspondence between the size of correlations in the representatives of soft matter and the extent of their fractal-like behaviour has been pointed out. Based on the theoretical models the spectral and the fractal time dimensionalities have been estimated from the experimental dielectric relaxation data.

COOPERATIVENESS OF MOLECULAR MOTION IN VARIOUS PHASES

Generally, both the absorption $\chi''(\omega)$ and the dispersion $\chi'(\omega)$ curves presented in the double logarithmic coordinates have the same shapes for all temperatures of the thermodynamic phase. Thus they can be scaled with respect to the temperature and the relaxation process can be described by a pair of single $\log\chi''(\log\omega)$ and single $\log\chi'(\log\omega)$ master curves in one chosen scaling temperature. The slopes of the linear wings characteristic of the absorption curve $\lg\chi''(\lg\omega)$ give the $n-1$ value for the frequency region $\omega > \omega_{max}$ and the m value for $\omega < \omega_{max}$. The n and m correlation parameters seem to be a proper tool not only for the estimation but also for the comparison of the degrees of cooperativity of specific molecular motions in different materials.[6] Table I lists the literature data of n and $1-m$ evaluated for substances with various shapes of molecules in liquid, liquid crystalline, plastic and glassy phases. The subsequent examples of dielectric relaxations have been chosen in such a way as to illustrate a gradual increase of the cooperativity of reorienting molecules on local and long-range levels when passing from sample No. 1 towards sample No. 6. Almost each case is representative of a wider class

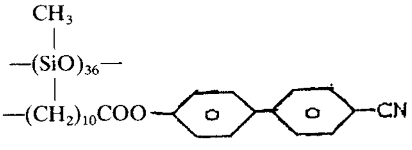
TABLE I

Correlation parameters for some polar substances

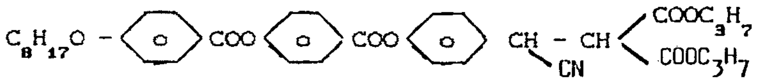
No.	Substance	Phase temperature [K]	Correlation parameters n ± 0.1	 (1−m) ± 0.1	d_s	D_t	$1-k$
1	C_8O/OC_7[a]	S_A, S_C 335–361	0.1	0.04	1.8	0.9	0.93
2	glycerol	supercooled liquid 186.5–221.5	0.43	0.09	1.14	0.57	0.63
3	cyclohexanol	plastic I 267–298	0.4	0.39	1.2	0.6	0.98
4	polymeric[b]	smectic A 317–443	0.4	0.45	1.2	0.6	1.09
5	swallow-tail[c]	S_A, nematic 333–404	0.5	0.21	1.0	0.4	0.51
6	MBBA	glassy 120–190	0.8	0.4	0.4	0.2	0.33

[a] 4-n-Heptyloxyphenyl 4′-n-octyloxybenzoate.

[b] Siloxane derivative.



[c] Substituted benzylidene malonate.



of dielectric materials. The case No. 1 represents many typical weakly polar liquid crystals where MHz-relaxation connected with reorientations of elongated molecules around the short axes is of nearly Debye type.[7,8] The same geometry of motion is characteristic of the swallow-tail liquid-crystalline molecules[9] having extra CN-side group and two alkyloxy chains instead of one. The strong electric interactions as well as evident steric hindrances result in essential enlargement of the n parameter while the long range spatial disorder seems to remain similar to that in conventional mesophase: the 1 − m values estimated for the first seven members of PAA homologous series,[8] for the examined members of the alkyloxyphenyl alkyloxybenzoate series[7] and for some others with rod-like molecules[3,6,7] ranged from 0.02 to 0.23. In polymeric molecules in the smectic phase the elongated strongly polar side-chains reorient around the main chain of the SiO segments[10,11] giving the kHz-relaxation. Large deviations of both n and 1 − m parameters from the Debye case come from the fact that each reorienting CN dipole is coupled to the near neighbours due to the electric interactions and to far distant cyano biphenyls due to long polymer chain and to spacer $(CH_2)_{10}$ groups adapting to all the rearrangements. In glycerol and cyclohexanol representing hydrogen-bonded structures (in the supercooled liquid and in the plastic phases) the molecules perform overall rotations.[12,13] The large local cooperativity of reorienting molecules is justified by inter and intra molecular OH bonds. The additional long range effect is essential only for plastic phase with well defined crystalline lattice. In the glassy phase of MBBA molecules with tangling butyl chains the extremal deviations from the Debye-type relaxation on both local and long range levels[6,14] is produced by a tight packing of disordered non-globular molecules at low temperatures.

The samples whose correlation parameters are collected in Table I all represent disordered materials. However some forms of order are introduced there by such factors as structural anisotropy, hydrogen bonding, crystalline lattice, steric hindrances, rigidity of the polymeric main chains or contraction of free volume. In the Dissado and Hill approach the above ordering factors affect the correlation parameters and the n values are sensitive to those ordering on the local scale, whereas the $1-m$ values are sensitive on the long-range scale.

The examples of dielectric relaxation systematized in Table I are chosen from a limited number of systems where the experimental data of n and m parameters have been available. Therefore only cautionary statements could be formulated. The values of correlation parameters presented in the Table are a good confirmation of the opinion that in condensed matter the Debye process is an exceptional one rather than a rule.[1,3–6,10] In view of the results it seems that the size of reorientational cooperativity on local scale in the system depends mostly on the electrical structure of molecules and also on the free volume magnitude and the details of the sterical situation. The latter two seem to be important factors determining the cooperativity on long-range scale also.

FRACTAL CONCEPTS FOR DIELECTRIC RELAXATION

The n and m parameters bring information about the microstructure of the sample. Thus Equations (2) and (3) dependences show a direct relation between the observed temporal evolution of the system and its spatial structure. The frequency dependences of the electric susceptibility have the fractional power-laws form regardless of the chemical bonding and physical properties of the system.[3] This independence of microscopic details means universality which points to the importance of looking for a general mechanism in the understanding of the relaxation phenomena. Therefore in the interpretation of the results presented in Table I we will not make use of the theoretical approaches describing the non-Debye relaxation in terms of the relaxation time distribution,[1] but select the approaches where the relation between response observed and a morphology of the medium is the main goal.

In some materials with spatial randomness a kind of vague similarity i.e., the scale invariance on certain interval of scale changes is reported.[15–17] Such features are understandable for the materials which have long, flexible chain-like structural elements at short length scales. They can arrange themselves into networks with arbitrarily self-similar connectivity.[16] At the length scales larger than the connectivity length the medium appears to be homogeneous.[17] Convincing results have been found for polymeric, glassy and hydrogen bonded materials and for liquid crystals.[2,18–23] For these cases departure from the exponential decay of polarization can be understood as a signature of the fractal-like i.e. statistically self-similar structures where physical properties behave as powers of the relevant length scale with non-integer exponents.[24]

It seems obvious that for condensed matter the scale invariance in the space is connected with the temporal one, as the spatial and temporal correlations cannot exist independently.[24] Thus the fractional power-law dependence of an electric susceptibility on frequency could be understood as the macroscopic manifestation of some kind of self-similarity on the microscopic level of the system.[3] In a wide range of materials self-similarity seems to be a hidden symmetry intermediate between translational symmetry and the lack of it.[25] These systems are hierarchically composed: large units/blocks/clusters in original scale are composed of similar subelements in smaller scale and so on.[3] In many cases, fractal like nature originates from the ramified structures of chemical bonding rather than from the relevant mass distribution so their density can be even homogeneous to the near atomic scales.[13,26,27]

Since 1977 physicists have started to believe that seemingly structureless materials can satisfactorily be quantitatively described with help of scaling exponents/fractal dimensionalities.[15,21,28] The arrangement of objects (say clusters of molecules) of various sizes creating the network is defined by fractal dimensionality d_f introduced to show the mass M dependence of the distance L: $M(L) \sim L^{d_f}$. The d_f value depends on the degree of disorder present in the system.[28] In order to measure the connectivity in the network[16] the spectral (fracton) dimensionality d_s is used. It has been introduced[21,28] to describe the classical diffusion on fractals. The above dimensionalities are ordered as follows $d > d_f > d_s$[21] where d defines the Euclidean space integer dimensionality. For the typical fractal construction the Sierpiński gasket embedded in three dimensional space $d_f = 2$ and $d_s = 1.55$.

It seems reasonable to expect the existence of the relationship between the correlation parameters describing the relaxation spectra and the scaling exponents d_f and d_s.[8,29] Many phenomena leading to macroscopic scaling behaviour with fractional exponents of some observables can be related to the diffusion of random walk[19] which does not go on in the same way as in the Euclidean space. The mean-square displacement $\langle R^2(t)\rangle$ grows more slowly than linearly with time and scales with time as follows $\langle R^2\rangle \sim t^{d_s/d_f}$. Essential for this anomalous behaviour is the presence of disorder on various length scales of the sample.[29–32] It is possible to relate the diffusion problem and the resistor network problem.[33] Theoretical description of the a.c. conductivity have been performed on a model where irreducible circuits of single capacitor and resistor are the elements arranged to give the self-similar network construction.[34,35] The final circuit is a hierarchical combination of many subcircuits having random number of the smaller circuits. Such constructions are an analogy of the disordered dielectric media. The deterministic model being a gross simplification of any real system can be solved analytically. The relations in Equations (4) and (5) observed for real dielectric samples have been confirmed by analytical derivation for the above circuit model constructions. The calculations done on the final circuit having the Sierpiński gasket geometry[35] allows one to formulate the correspondence between n and d_s: $d_s = 2 - 2n$.[36] The d_s values for the examples of soft materials are collected in Table I.

The striking feature of dipolar relaxation is not only the power type of dielectric response but also the existence of two distinctly separate time scales. The Jonscher concept of "screened hopping" dipoles[5] relates it to the rapid changes of orientations of dipoles followed by gradual adjustments of the interacting dipoles in the surrounding. The "chain" responses are stretched both in time and in space beyond the initiating event.[37] The rapid reorientation occurs after a period called waiting time which is different for each dipole. The waiting time distribution corresponds to the disorder of the time scale characterizing the system of dipoles. Taking the whole experimental time of relaxation as a (0.1) segment and cutting out shorter and shorter waiting time gaps can eventually give a fractal sequence of events in the form of inhomogeneous Cantor set with fractal time dimensionality D_t. This inhomogeneous distribution of waiting times is caused by the long-range interactions.[38] Such a mathematical model proposed by Weron,[37] in which the decay of the polarization follows the *k*th order rate equation, allows us to obtain the final dielectric response in form Equations (2) and (3) with $n = 1-D_t$ and $m = D_t/(1-k)$. The $1-k$, taking values near unity, seems to be connected with the strength of the interactions in the medium. The D_t values and $1-k$ values calculated from the n and m parameters are collected in Table I. The D_t values cover the interval (0.1). The smaller D_t the more pronounced factal-like behaviour.

The d_s data collected in the Table can be compared with rather few experimental results. They have been obtained for silica aerogeles.[21] Due to different microstructures of the samples prepared in various conditions the d_s have ranged the values from 0.9 to 1.85. The experimental techniques used are the small-angle neutron scattering, the Raman scattering and the low-temperature specific heat measurements. There are also d_s results from the exciton annihilation for low temperature glassy and polymeric samples[23] giving d_s values between 0.6 and 1.8. The d_s results in the Table obtained based on fractal-like circuit model[35] correspond with D_t data describing the problem in terms of temporal randomness based on the Weron's model.[37] Moreover, for the cases where $1-k$ values are near 1 i.e., for cyclohexanol and for polymeric and C_8O/OC_7 liquid crystals, the D_t values are equal to the m values as has been proposed by Niklasson.[39] For all other cases the $1-k$ values are lower than 1, which means that D_t values are lower than results from mere m values.

There appeared other theoretical models[40–42] taking into account the influence of some forms of self-similarity on the evolution from a disturbed state of the sample towards equilibrium. In approaches with random fractal relief of the energy barriers[40,41] temperature dependence of the scaling exponents is derived. Niklasson[39] and Dissado and Hill[43] have presented the review of fractal concepts in the description of dielectric response in disordered materials. Up till now some problems concerning the materials that exhibit fractal-like geometrical regularities have not been satisfactorily explained. There still are controversies as to the uniform presentation of correlation parameters in terms of fractal dimensionalities. Anyway there are advantages of using fractal concepts for the description of the relaxation phenomena. The models existing for systems with fractal-like elements allow us to compare the analytical solutions of some problems with the respective behaviour in real media. Results concerning the influence of the various forms of disorder (spatial, temporal, energetical) on the macroscopic response seem promising.[26,40,41] The fractal philosophy provides a better tool for uniformly defining all amorphous media. This language occurred to be adequate also to describe dynamical features of the system resulting from the nature of the reorientations of the dipoles and/or the geometry of the system. Basing on the temperature independence of the correlation parameters in the temperature range under study we can conclude that the geometrical factor dominates the temporal one.[43]

For each kind of substance the analysis of the microscopic reasons justifying fractal concepts for interpretation of the power-law $\chi(\omega)$ behaviour should always precede their application. In liquid crystalline phases[20] the structure of the focal conic, schlieren and dendritic-type textures reveals self-similarity at least for a certain range of scales. The transition from isotropic to smectic A phase is accompanied by creation of the disclination lines network distributing well large anisotropic interfacal energy. In dielectric measurements the bulk samples are used which have such defects even when uniformly aligned. For substances with the molecules with O—H groups the existence of the irregular network of the intramolecular and intermolecular hydrogen bonds has been reported.[13,19] The dynamical behaviour of glass phases seems to originate from the hierarchical structure of the free energy surface.[22,40,41] The liquid-glass phase transition is usually connected with the percolation problem.[18] The fractal nature of polymer systems investigated in various experiments seems the most transparent due to a random shape of molecules.[3,16,44]

Acknowledgment

I wish to express my gratitude to Professor Jerzy A. Janik for his interest and valuable discussions.

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