Multifractal phase transitions in the non-Debye relaxation processes

Alexander Leyderman* and Shi-Xian Qu[†]

Department of Physics, University of Puerto Rico, Mayagüez, Puerto Rico 00680

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The multifractal measures of the relaxation-time distributions are analytically obtained for some typical non-Debye dielectric relaxation processes. The characteristics of the corresponding multifractal thermodynamics are discussed. It is shown that the probability of the relaxation times near the poles of their distribution function is fractal scaling. The corresponding Lipschitz-Höld singular exponent is, or can be, determined by the so-called shape parameters in the empirical dielectric relaxation formulas. The relationship to some analytical proofs of the empirical dielectric formulas based upon the fractal models is also analyzed. Some generalized multifractal phase transitions with interesting features are reported in this paper. The recent experiment results on the molten-crystal transition in organic glass systems are also discussed to support our conclusions.

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I. INTRODUCTION

Relaxation processes, deviating from exponential behavior in time domain or from the classical Debye form in frequency domain, occur in many physical systems such as dielectrics, supercooled liquids, viscoelastic solids, liquid crystals, polymer melts and solutions, and porous media. They are usually described by sophisticated mathematical functions, for instance the Cole-Cole formula, the Davidson-Cole formula, the Havriliak-Negami function, and power laws [1-3]. The deep understanding of these dielectric processes and the first principle proof of these empirical relaxation functions are the important topics. Recently, the fractal interpretation of dielectric relaxation phenomena has become a very active research field [4-10]. Nigmatullin and Ryabov [4-6] set up some general relaxation equations based on the fractional order calculus in time structure, and carried out the analytical derivations of the Cole-Cole and the Davidson-Cole dielectric relaxation formulas. Feldman et al. [7] advanced a model to describe the cooperative relaxation in microemulsions near the percolation threshold, in which the excitation transferring via relaxation channels in a fractal cluster of droplets is supposed. Based upon the well-known Zener model, Metzler and his collaborators [8] developed a fractional-order differential equation with respect to time for both strain and stress relaxations, and also obtained the general relaxation-time distribution. A differential equation for non-Debye relaxation and diffusion was built in fractal space-time by Kobelev and his co-workers [9]. The anomalous relaxations were studied by González and Araujo [10] through the random-walk model in disordered structures consisting of regular and fractal lattices.

In Ref. [4], a self-similar relaxation process is assumed for the Davidson-Cole dielectric relaxation process. The authors consider the system in which the relaxation on the macroscopic level may be defined as the relaxation of the enormous elementary components on the microlevel. They believe that the equations describing relaxation of the system on the macrolevel will coincide with the equations describing the relaxation of individual electric dipoles on the microlevel. Then a model with the relaxation times distributing over a fractal set is built. Through the analytic derivation of the fractional-order differential equation for the relaxation function, they obtained the Davidson-Cole formula for the complex susceptibility. They related the exponent in the formula to the fractal dimension of the time structure over which the relaxation time distributes. Their approach provides a reasonable interpretation of the Davidson-Cole formula. In Ref. [11], Gomi and Yonezawa defined a relaxation function by the characteristic function of the fractal random walk. The Fourier-Laplace transformation of this function yields the Cole-Cole formula, in which the breadth exponent is related to the exponent in the scaling of the probability distribution of fractal random jump events.

For the non-Debye relaxation, one often expects the distribution of relaxation times [1]. Analysis of the distribution properties will also shed light on the understanding of the nature of relaxation processes. The multifractal [12] analysis of the distribution property of physics quantities in condensed matters has recently attracted much interest among researchers. The discovery of phase transitions in multifractal analysis is another new development in fractal theory and its applications, and there are still many open questions. In general, a phase transition results from the nonanalytic property of the characteristic thermodynamic functions, and may be accompanied by abrupt changes in the properties of the system. The investigation of phase transitions will provide a way to get a complete knowledge of the system. In analyzing the measure property of the relaxation-time distribution in Davidson-Cole relaxation, we got an analytical description of the corresponding multifractal thermodynamics and found a generalized multifractal phase transition [13]. It is the first evidence of multifractal phase transition in the relaxation processes. In this paper, we will extend the multifractal analysis to the Cole-Cole and the Havriliak-Negami relaxation patterns. The phase transitions in these two processes and the relationship among the phase transitions in different relaxation patterns will be discussed. Our recent experiment results on the molten-crystal transition in organic glass

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^{*}Email address: alex@feynman.upr.clu.edu

[†]Permanent address: Department of Basic Courses, Xi'an Petroleum Institute, Xi'an 710065, China.

systems are also discussed to support the conclusions of this work.

II. MULTIFRACTAL ANSATZ

The multifractal analysis is based on the standard boxcounting procedure, dividing the system into N_{δ} boxes of linear size δ and determining the probability of some physical property in the *i*th box,

$$P_i(\delta) = \int_{\text{box } i} \rho \, d\Omega, \qquad (1)$$

as a suitable measure, where ρ is the probability density (distribution function). The *q*-order probability moment, i.e., the partition function, is defined by

$$Z(q) = \sum_{i} P_{i}(\delta)^{q}.$$
 (2)

If it is proportional to some power of the box size as $\delta \rightarrow 0$, e.g.,

$$\lim_{\delta \to 0} Z(q) \sim \delta^{(q-1)D_q},\tag{3}$$

then the multifractal behavior or a multifractal set may be derived. D_q is the so-called generalized dimension, which is defined as follows:

$$D_q = \lim_{\delta \to 0} \frac{\ln Z(q)}{(q-1)\ln \delta}.$$
 (4)

The behavior of this multifractal set can be completely described by an infinite number of generalized dimensions. Each generalized dimension has its own meaning, for example, D_0 is the dimension of the support of the measure onto which the elements of the multifractal set distribute, D_1 is the information dimension which determines the scaling of the information with respect to box size, and the correlation dimension D_2 reflects the scaling of density-density correlation function. D_q is often related to the so-called mass exponent [12] (in this paper, the mass exponent is denoted by Ξ),

$$\Xi(q) = (q-1)D_q. \tag{5}$$

While the discussion of the generalized dimension D_q is quite illustrative, the abstract analysis of multifractals is often concerned with the singularity strength of the fractal, given by the Lipschitz-Höld exponent α and the corresponding singularity spectrum $f(\alpha)$. In the *i*th box, the singularity strength α is defined by

$$P_i(\delta) \sim \delta^{\alpha_i}.$$
 (6)

The number of subsets $N(\alpha)$ in which this strength is observed satisfies the following rule:

$$N(\alpha) \sim \delta^{-f(\alpha)},\tag{7}$$

where $f(\alpha)$ is the Hausdorff dimension of these subsets. One can easily get the relationship between $\Xi(q)$ and $f(\alpha)$. According to Eqs. (6) and (7), the partition function could be rewritten as

$$Z(\delta) = \sum_{i} P_{i}(\delta)^{q} = \int d\alpha \,\rho(\alpha) \,\delta^{q\alpha - f(\alpha)}.$$
 (8)

The saddle-point approximation to the integrand in Eq. (8) yields

$$Z(\delta) \sim \delta^{q\,\alpha(q) - f(\alpha)}.\tag{9}$$

Combining the definitions (4) and (5), one gets

$$\Xi(q) = q \alpha(q) - f(\alpha), \quad q = \frac{df(\alpha)}{d\alpha}.$$
 (10)

This equation is just the Legendre transformation, where q is analogous to reciprocal temperature in ordinary thermodynamics. One may immediately obtain the inverse Legendre transformations,

$$f(\alpha) = \alpha(q)q - \Xi(q), \quad \alpha = \frac{d\Xi(q)}{dq}.$$
 (11)

In the multifractal thermodynamics, one also addresses $\Xi(q)$ as "free energy" and $f(\alpha)$ as "entropy." Now, two sets of characteristic functions $\Xi(q)$ and $f(\alpha)$ have been set up to describe the multifractal behavior.

III. RELAXATION-TIME DISTRIBUTIONS AND THE MULTIFRACTAL PARTITION FUNCTIONS

In Debye's dielectric relaxation theory, the dipoles in dielectric media relax with a single relaxation time τ_0 , and the frequency dependence of the complex permittivity reads

$$\boldsymbol{\epsilon}^*(\boldsymbol{\omega}) = \boldsymbol{\epsilon}_{\infty} + \frac{\Delta \boldsymbol{\epsilon}}{1 + i\,\boldsymbol{\omega}\,\tau_0},\tag{12}$$

where $\Delta \epsilon = (\epsilon_0 - \epsilon_\infty)$ is the dielectric increment; τ_0 is reciprocally proportional to the peak frequency of the dielectric loss. For most relaxation processes this single relaxation time model should be modified to secure agreement between theory and experiment. A straightforward modification is to introduce a relaxation-time distribution. Let $G(\tau)d\tau$ be the contribution to the permittivity of the group of dipoles having individual relaxation time in a range $d\tau$ near τ ; the contribution of the various groups may be linearly superposed if the interaction between dipoles can be neglected. The total complex permittivity is then expressed as

$$\boldsymbol{\epsilon}^{*}(\boldsymbol{\omega}) = \boldsymbol{\epsilon}_{\infty} + \Delta \boldsymbol{\epsilon} \int \frac{d\tau G(\tau)}{1 + i\boldsymbol{\omega}\tau}.$$
 (13)

Here $G(\tau)$ is the relaxation-time distribution function. Introducing reduced relaxation time (RRT) $t = \tau/\tau_0$, $G(\tau)$ is replaced by $\rho(t)$, and Eq. (13) changes into

$$\boldsymbol{\epsilon}^{*}(\boldsymbol{\omega}) = \boldsymbol{\epsilon}_{\infty} + \Delta \boldsymbol{\epsilon} \int \frac{dt \, \boldsymbol{\rho}(t)}{1 + i \, \boldsymbol{\omega} t \, \tau_{0}}.$$
 (14)

The above two equations are just the Laplace transformations of the distribution function. Therefore, the distribution of relaxation times will be uniquely determined through Eq. (14) if any analytic function that can represent the relaxation data is known.

As is well known, three expressions for the dielectric permittivity allow one to describe a wide range of the experimental relaxation data that cannot be described by the Debye formula. They are the Cole-Cole formula

$$\boldsymbol{\epsilon}^{*}(\boldsymbol{\omega}) = \boldsymbol{\epsilon}_{\infty} + \frac{\Delta \boldsymbol{\epsilon}}{1 + (i \,\boldsymbol{\omega} \, \tau_{0})^{\mu}}, \qquad (15)$$

the Davidson-Cole formula

$$\boldsymbol{\epsilon}^{*}(\boldsymbol{\omega}) = \boldsymbol{\epsilon}_{\boldsymbol{\omega}} + \frac{\Delta \boldsymbol{\epsilon}}{(1 + i\,\boldsymbol{\omega}\,\boldsymbol{\tau}_{0})^{\nu}},\tag{16}$$

and the Havriliak-Negami formula

$$\boldsymbol{\epsilon}^{*}(\boldsymbol{\omega}) = \boldsymbol{\epsilon}_{\boldsymbol{\omega}} + \frac{\Delta \boldsymbol{\epsilon}}{\left[1 + (i\boldsymbol{\omega}\tau_{0})^{\mu}\right]^{\nu}},\tag{17}$$

where $0 < \mu < 1$ and $0 < \nu < 1$ are parameters, determining the characteristics of the dielectric relaxation uniquely. The parameter μ appears to represent the breadth while ν represents the skewness of the distribution of relaxation times [2].

It has been shown that the corresponding RRT distribution functions for the three relaxation patterns are as follows [1,2], i.e., the Cole-Cole pattern

$$\rho(t) = \frac{t^{-(1-\mu)}\sin\mu\pi/\pi}{1+t^{2\mu}+2t^{\mu}\cos\pi\mu},$$
(18)

the Davidson-Cole pattern

$$\rho(t) = \begin{cases} t^{-(1-\nu)}(1-t)^{-\nu} \sin \nu \pi/\pi, & t \leq 1 \\ 0, & t > 1, \end{cases}$$
(19)

and the Havriliak-Negami pattern

$$\rho(t) = \frac{t^{-(1-\mu\nu)}\sin\theta\pi/\pi}{(1+t^{2\mu}+2t^{\mu}\cos\pi\mu)^{\nu/2}},$$
(20)

where

$$\theta = \arctan \frac{\sin \mu \pi}{t^{\mu} + \cos \mu \pi}$$

These distribution functions are in fact the probability density, i.e., $\rho(t)dt$ is the probability of the reduced relaxation time between t and t+dt. In order to conduct multifractal analysis on this system, the time domain $[t_{\min}, t_{\max}]$, over which the relaxation times distribute, is divided into $N (N \rightarrow \infty)$ equal intervals with length $\delta \rightarrow 0$. One may note that there are some poles in the distribution functions in Eqs. (18)–(20). Thus, one should treat the boxes near the poles separately when computing the partition functions. For the Davidson-Cole relaxation, there are two poles at $t=t_{\min}=0$ and $t=t_{\text{max}}=1$, respectively. We have gotten some very interesting results in Ref. [13]. There is only one pole at $t=t_{\text{min}}=0$ in the distribution functions of the other two kinds of relaxation processes, and $t_{\text{max}}=\infty$. Therefore, the corresponding partition function can be obtained in the following way:

$$Z(q) = \sum_{i=1}^{N} P_i^q = \left[\int_0^{\delta} \rho(t) dt \right]^q + \delta^{q-1} \int_{\delta}^{\infty} \rho(t)^q dt.$$
(21)

Simple calculation gives the partition functions for the three kinds of relaxation patterns (Z_{DC} has been calculated in Ref. [13] by the authors),

$$Z_{\rm CC}(q) = g(\mu, \mu) \,\delta^{\mu q} + C_1 \,\delta^{q-1}, \tag{22}$$

$$Z_{\rm DC}(q) = g(\nu, \nu) \,\delta^{\nu q} + g(1 - \nu, \nu) \,\delta^{(1 - \nu)q} + C_2 \,\delta^{q - 1},$$
(23)

$$Z_{\rm HN}(q) = g(\mu\nu,\theta)\,\delta^{\mu\nu q} + C_3\,\delta^{q-1},\qquad(24)$$

where

$$C_{1} = h(\mu) \int_{0}^{\infty} t^{-q(1-\mu)} (1 + t^{2\mu} + 2t^{\mu} \cos \mu \pi)^{-q} dt,$$

$$C_{2} = h(\nu) B(1 - \nu q, 1 + q - \nu q),$$

$$C_{3} = h(\theta) \int_{0}^{\infty} t^{-q(1-\mu\nu)} (1 + t^{2\mu} + 2t^{\mu} \cos \mu \pi)^{-\nu q/2} dt,$$

$$g(\xi, \eta) = [\xi^{-q} - (1 + \xi q - q)^{-1}]h(\eta),$$

$$h(\eta) = (\sin \eta \pi / \pi)^{q}.$$

Here, $B(z_1, z_2)$ is the beta function. Z_{CC} , Z_{DC} , and Z_{HN} are the partition functions for the RRT distributions of Cole-Cole, Davidson-Cole, and Havriliak-Negami relaxation patterns, respectively.

IV. RESULTS AND DISCUSSION

One may note that there is more than one term on the right-hand side of Eqs. (22)–(24) in Sec. III. They are the origination of the phase transitions in the multifractal thermodynamics of the RRT distributions for different relaxation processes. The contribution of each term to the corresponding partition function is determined by parameters μ or ν , or μ and ν . The competing between each pair of terms results in different behaviors of the partition functions with respect to the variation of q. For some specific q value the two terms are equally competing, and the critical condition occurs. We denote the critical value by q_c , which separates different thermodynamic states. One can easily get the equations of these critical values of q by setting the exponents in the corresponding two competing terms to be equal. They are the critical line for the Cole-Cole process,

$$q_c = \frac{1}{1-\mu}, \quad 0 < \mu < 1,$$
 (25)

the critical line for the Davidson-Cole process,

TABLE I. Generalized dimensions for different relaxation patterns.

Relaxation patterns $D_q (q \ge q_c)$ $D_q (q < q_c)$ q_c Cole-Cole $q \mu/(q-1)$ 1 $1/(1-\mu)$ Davidson-Cole $q \min\{\nu, 1-\nu\}/(q-1)$ 1 $1/\max\{\nu, 1-\nu\}$				
Cole-Cole $q\mu/(q-1)$ 1 $1/(1-\mu)$ Davidson-Cole $q\min\{\nu, 1-\nu\}/(q-1)$ 1 $1/(\max\{\nu, 1-\nu\})$	Relaxation patterns	$D_q \; (q \ge q_c)$	$D_q (q < q_c)$	q_c
Davidson-Cole $q\min\{\nu, 1-\nu\}/(q-1)$ 1 $1/\max\{\nu, 1-\nu\}$	Cole-Cole	$q\mu/(q-1)$	1	$1/(1-\mu)$
	Davidson-Cole	$q\min\{\nu, 1-\nu\}/(q-1)$	1	$1/\max\{\nu, 1-\nu\}$
Havriliak-Negami $q \mu \nu/(q-1)$ 1 $1/(1-\mu)$	Havriliak-Negami	$q\mu\nu/(q-1)$	1	$1/(1 - \mu \nu)$

$$q_c = \frac{1}{\max\{\nu, 1 - \nu\}}, \quad 0 < \nu < 1, \tag{26}$$

and the critical surface for the Havriliak-Negami process,

$$q_c = \frac{1}{1 - \mu \nu}, \quad 0 < \mu, \ \nu < 1.$$
 (27)

As we have already shown in Sec. II, the partition function is scaling when $\delta \rightarrow 0$. The generalized fractal dimensions for the three non-Debye relaxation processes can be easily obtained by comparing Eqs. (22)–(24) with Eq. (3). The results are shown in Table I and Fig. 1. Consequently, the corresponding thermodynamic functions were obtained through the Legendre transformations (10) and (11). The results are listed in Table II.

All these results reveal that phase transitions will be observed at the corresponding q_c 's. The generalized dimensions and the free energies for different relaxation patterns are continuous, but their first order derivatives are discontinuous when $q \rightarrow q_c$ (see Fig. 1 and Table II). Thus the phase transitions are of the first order. These multifractal phase transitions are very similar to the one observed in the multifractal thermodynamics of the logistic map [14], but have some new features. In terms of the terminology in Ref. [14], we may also address the phase transitions observed in the current work as the transformation between "hyperbolic" and "nonhyperbolic" phases. A detailed analysis of their properties for different relaxation patterns will be given in the following section.

One may have already noted that the results for the Cole-Cole and the Havriliak-Negami patterns in Tables I and II are nearly the same except replacing μ by $\mu\nu$. This can be easily understood because their RRT distribution functions (18) and (20) are simply related by a power-law relation with a positive order, i.e., $\rho_{\rm HN}(t) \sim [\rho_{\rm CC}(t)]^{\nu}$. This implies that the topological properties for both of them are the same, i.e., they are topologically conjugated. Thus they have the same measure properties. For these two relaxation patterns, there is only one pole at t=0, where the distribution functions go to infinity. The hyperbolic contribution [the second terms in Eqs. (22) and (24) to the partition function arises from the bulk of the RRT intervals away from the singular point. One may see from Table II and Eq. (6) that the probability of the RRT in this kind of box satisfies $P_i \sim \delta$. However, the nonhyperbolic contribution [the first terms in Eqs. (22) and (24)], depending on μ , or on μ and ν , arises from the singularity of the distribution functions at t=0. The probability of the relaxation times in the intervals very close to this pole is scaled by $P_i \sim \delta^{\xi}$, which is much bigger than the probability for those in the intervals away from the pole. Here, ξ = μ corresponds to the Cole-Cole pattern, and $\xi = \mu \nu$ corresponds to the Havriliak-Negami pattern. In the multifractal thermodynamics [12], big q favors the events with high probability and small q favors the events with low probability. Therefore, the relaxation processes with much shorter RRT, or the fast relaxation processes, dominate the multifractal property when $q \ge q_c$. The phase diagram is shown in Fig. 2(a).

To this end, one may conclude that the relaxation processes in these two patterns consist of the ones with the probabilities obeying both the conventional and fractal scaling rulers. However, the fractal relaxation processes make the leading contribution, and therefore represent the nature of the systems. This conclusion supports the fractal time random-walk model in which a fractal scaling of the probability density with a single pole at t=0 is proposed [11], and the self-similar relaxation model in terms of the fractional order calculus [4–6], based upon which the analytical derivations of the Cole-Cole formula were carried out. We expect the analytical derivation of the Havriliak-Negami for-



FIG. 1. The generalized dimension spectra. (a) $\xi = \mu$, for the Cole-Cole type relaxation, and $\xi = \mu \nu$, for the Havriliak-Negami type relaxation; (b) for the Davidson-Cole type relaxation.

	$\Xi(q)$		$\alpha(q)$	
Relaxation patterns	$(q \ge q_c)$	$(q < q_c)$	$(q \ge q_c)$	$(q < q_c)$
Cole-Cole	$q\mu$	q - 1	μ	1
Davidson-Cole	$q\min\{\nu,1-\nu\}$	q-1	$\min\{\nu, 1-\nu\}$	1
Havriliak-Negami	$q \mu \nu$	q-1	$\mu \nu$	1

TABLE II. Free energies and Lipschitz-Höld singular exponents.

mula based on the fractal model.

As we have discussed in [13], the RRT distribution in the Davidson-Cole relaxation process has two poles at t=0 and t=1, respectively. The nonhyperbolic contribution (when q $\geq q_c$) to the partition function comes from the competition between two kinds of characteristic relaxation processes, the ones with RRT in the intervals near the pole at t=0 and t =1, respectively. It introduces interesting new features to the multifractal thermodynamics of the system. They are equally competitive when $\nu = \frac{1}{2}$, and the corresponding phase transition has no intrinsic difference from the ones observed in the two relaxation patterns mentioned above. If $\nu < \frac{1}{2}$, however, the relaxation processes with RRT very close to the pole at t=0 dominate the contribution. We address this kind of process as the fast relaxation process. When $\nu > \frac{1}{2}$, the relaxation processes with RRT very close to the pole at t= 1 dominate the contribution. We refer to them as the slowrelaxation processes. One can immediately get a conclusion that the nonhyperbolic phase will bifurcate, at $\nu = \frac{1}{2}$, into two subphases, the fast-relaxation phase and the slow-relaxation phase, respectively. Therefore, the critical line between the hyperbolic and the nonhyperbolic phases decomposes into two critical lines, i.e., the one between the hyperbolic and the fast-relaxation phases, and the other one between the hyperbolic and the slow-relaxation phases. It is worthwhile to emphasize that the first order derivatives of the multifractal thermodynamic functions are discontinuous at $\nu = \frac{1}{2}$, which also shows a phase transition (a new feature). As shown in Fig. 2(b), there is a triple point at $(\nu = \frac{1}{2}, q = 2)$ in the phase diagram, at which the relaxation processes with the RRT in the three kinds of intervals mentioned previously are equally competing.

Clearly, the two poles of the distribution function represent the intrinsic features of the Davidson-Cole relaxation process. Because the probability of the relaxation processes with RRTs near the poles is scaled by $P_i \sim \delta^{\min\{\nu,1-\nu\}}$, which is much bigger than that away from the poles, they make the leading contribution to the multifractal thermodynamic functions, and thus to the overall relaxation mechanism. It is the additional pole at t=1 that makes the multifractal behavior in this relaxation pattern much different from that in the other two patterns. All the results evoke the analytical derivation of this empirical dielectric relaxation formula by employing the fractal random-walk model advanced by Gomi and Yonezawa [11], but introducing another singular point, at t=1, into the probability density of the random-walk events. This will be discussed elsewhere.

One could also conclude that the Davidson-Cole formula may describe the relaxation systems including two kinds of typical relaxation processes with relatively shorter and longer relaxation times, respectively, such as the dielectric relaxation in the plastic crystal [15] during the glass transition and that in some organic glasses during the moltencrystal transitions [16]. In our recent work on the relaxation dynamic study of the organic glasses 2-cyclo-octylamino-5nitropyridine (COANP), (S)-2-N- α -(methylbenzylamino)-5-nitropyridine, 2-(N-prolinol)-5-nitropyridine, and N-(4-nitrophenyl)-(L)-prolinol (NPP), it was found that their dielectric relaxation spectra could be perfectly described by the Davidson-Cole formula [16]. Furthermore, the temporal dependence measurement, of the dielectric spectra of COANP glass in the isothermal condition near the moltencrystal transition point, shows that the peak of the dielectric loss spectrum decreases with time but the position remains unchanged. The skewness parameter ν decreases from 0.71



FIG. 2. The phase diagrams. (a) The curve represents the critical line for the Cole-Cole pattern when $\xi = \mu$, but represents the critical surface for the Havriliak-Negami pattern when $\xi = \mu \nu$. (b) The critical lines for the Davidson-Cole pattern.

to 0.38 in 50 min. The same phenomenon was also observed in NPP glass, and ν decreases from 0.55 to 0.25 in 8 h [17]. These results imply that the crystallization does not induce the slow-down of the relaxation process. During the crystallization processes, the COANP and NPP glasses can be regarded as the two-phase systems consisting of the glasslike and crystal-like parts, which coincides with the two-orderparameter model for the supercooled liquids [18]. The glasslike parts relax with longer characteristic relaxation times while their crystal-like counterparts relax with the shorter ones. The dielectric loss could be described by a weighted superposition of the two limiting spectra [15], i.e., $\epsilon''(\omega,t)$ = $[1-N(t)]\epsilon''_{glass}(\omega)+N(t)\epsilon''_{crystal}(\omega)$, where N(t) is the crystallinity. At the very beginning of the crystallization, the glasslike parts dominate the composition and make the principal contribution to the overall dielectric loss. So the parameter ν is greater than 0.5. As the process goes on, the weight of the crystal-like parts becomes higher than that of their glasslike counterparts, and hence ν decreases. The overall relaxation time exhibits no obvious change because the characteristic times of both parts do not change significantly. This agrees with the discussions in the preceding paragraph, keeping the characteristic relaxation time τ_0 unchanged, but varying the parameter ν may only result in the variation of the relaxation-time distribution. Therefore, one may understand the fact that the relaxation dynamics of these organic glasses obeys the Davidson-Cole formula. Finally, we want to point out that the changing of the parameter ν from above to below $\frac{1}{2}$ implies that there is a multifractal phase transition associated with the molten-crystal transition process.

V. SUMMARY

We have set up the multifractal measures and analytically got the multifractal thermodynamic functions of the RRT distributions for Cole-Cole, Havriliak-Negami, and Davidson-Cole relaxation processes, which provides some deep understanding of the distribution properties of the relaxation processes. The phase transitions observed in these systems reveal the competing of the relaxation processes with the relaxation times in different time scales. The relaxation processes with the relaxation time very near the poles of distribution functions make the leading contribution to the overall relaxation mechanism, which represents the intrinsic features of the relaxation systems. The probability of the RRT in this time scale is fractal scaling. The corresponding exponent is the Lipschitz-Höld singular exponent which is, or can be, determined by the so-called shape parameter in the empirical dielectric relaxation formulas. This conclusion supports the analytic proofs of these empirical dielectric formulas in terms of the fractal concept. To the authors' knowledge, the multifractal analysis on these relaxation processes and the multifractal phase transitions observed in these systems are reported in this paper for the first time. These transitions, different from the conventional multifractal phase transition, are dominated not only by q but also by the shape parameters of the relaxation curves.

The multifractal analysis method employed in this work can be applied to any other relaxation pattern if one may somehow get the relaxation-time distribution function analytically or numerically. Different multifractal phase transitions will be expected when there are different kinds of singularities in the distribution functions occurring somewhere in the time domain or even in the space domain. We believe that the multifractal analysis of the dielectric relaxation or other relaxation phenomena would be another active area in the application of fractal theory, and meanwhile, as we have already shown in this paper, will provide some deep understanding of the relaxation mechanism.

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