Aging and scaling laws in β -hydroquinone-clathrate

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The dielectric permittivity of the orientational glass methanol(x=0.73)- β -hydroquinone-clathrate has been studied as function of temperature and waiting time using different temperature-time protocols. We study aging, rejuvenation, and memory effects in the glassy phase and discuss similarities and differences to aging in spin glasses. We argue that the diluted methanol-clathrate, although conceptually close to its magnetic pendants, takes an intermediate character between a true spin glass and a pure random-field system.

DOI: 10.1103/PhysRevB.69.144427 PACS number(s): 75.50.Lk, 77.22.Gm, 75.40.Gb, 64.70.Pf

I. INTRODUCTION

The long-time evolution of the response functions of glassy systems has attracted a lot of attention and gave and gives rise to many discussions. 1-3 The basic observation is that the behavior of the physical response functions, like magnetization, polarization, or the corresponding susceptibilities, is time and waiting time dependent. It depends on a waiting time t_{ω} spent before a perturbation (a magnetic or electric field) has been applied or after it has been switched off. A spin glass can even memorize some of the features of the way the system had been prepared. While in the beginning theoretical and experimental work on physical aging mainly concentrated on aging phenomena in polymer glasses, 4 a lot of effort has been made to carry out and understand experiments in other glassy systems, such as spin-glasses,⁵ pure, and disordered ferromagnets,⁶ supercooled organic liquids such as glycerol,⁷ relaxor ferroelectrics⁸ and (crystalline) dipolar glasses. ^{9,10} The differences and the things in common between aging in these systems are still intensively discussed. It is not clear which ingredients on a microscopic scale have to be included to observe aging, rejuvenation, and memory effects. In the present paper we present aging, rejuvenation and memory experiments in a dipolar glass, namely methanol(x= 0.73)- β -hydroquinone-clathrate.

The quinol HO-C₆H₄-OH molecules form a hydrogen bonded $R\bar{3}$ lattice with almost spherical cavities of 4.2 Å diameter. The host lattice tolerates methanol concentrations down to x = 0.35 without collapsing, x being the ratio of filled to available cavities. Whereas the higher concentrated methanol-clathrates undergo a first-order phase transition into an antiferroelectrically ordered low-temperature phase, clathrates with lower concentrations ($x \le 0.76$) freeze into states. 11,12 glass In methanol(x=0.73)- β -hydroquinone-clathrate, the freezing of the dipoles is at least to some part a collective process as the molecules do not only see the crystal field of the cavity but also a contribution from the electric interaction between the dipoles. 11,12 In this sense the methanol(x = 0.73)-clathrate is conceptually close to its magnetic pendants, the spin glasses. Within the

hexagonal basal plane, the dipoles ("pseudospins") are arranged on a triangular lattice. Together with the quasi-antiferroelectric long-ranged dipole-dipole interaction in the basal plane, the dilution leads to strong frustration and to a large number of metastable states. Although there is no clear transition into a glassy phase, we find slow glassy dynamics at low temperatures.

Our first investigation¹³ of aging phenomena in methanol-hydroquinone-clathrates produced qualitative evidence for aging, rejuvenation, and memory effects. In the present paper we investigate the frequency dependence of isothermal aging (Secs. II A and II B) and study rejuvenation and aging in a more quantitative way by following special temperature-time protocols in Sec. II C.

II. EXPERIMENTAL RESULTS

Single crystals of methanol(x = 0.73)- β -hydroquinone-clathrate have been grown from a saturated solution of quinol, methanol, and n-propanol at 313 K. The propanol molecules are not incorporated into the lattice but merely control the percentage of void cavities. The samples were prepared in the form of thin parallel plates ($d \sim 0.5$ mm) with faces perpendicular to the hexagonal c axis. With gold electrodes deposited on these faces they formed capacitors with a capacitance of about 0.4 pF. For the measurement of the frequency-dependent permittivity, a Solartron impedance analyzer FRA 1260 in combination with an interface (Chelsea) has been employed. The samples were placed into a closed-cycle refrigerator with a temperature stability of some mK.

We measured real and imaginary parts of the dielectric constant, $\varepsilon_c'(\nu)$ and $\varepsilon_c''(\nu)$, along the hexagonal c axis for frequencies from ν =0.2 Hz up to ν =200 Hz. Figure 1 shows the temperature dependence of the imaginary part, $\varepsilon_c''(\nu)$. The relaxation processes are characterized by a rather broad distribution of relaxation times τ and obey an Arrhenius law $\tau^{-1} \sim \tau_0^{-1} \exp(-E_A/T)$ (with the "pseudospin" flip rate τ_0^{-1} =9×10¹⁰ s⁻¹ and an energy barrier E_A =828 K) rather than a Vogel-Fulcher behavior, see inset of Fig. 1. Although there is no clear transition into a dipolar glass state, we find glassy dynamics at low temperatures. The effective

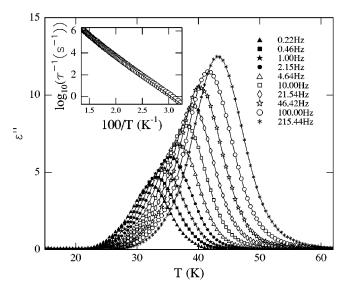


FIG. 1. Temperature dependence of the imaginary part of the dielectric permittivity, $\varepsilon_c''(\nu)$. (From Ref. 13). The inset shows the inverse average relaxation time τ^{-1} as a function of 1/T (determined from the temperature of maximum ε_c'').

energy barrier E_A decomposes into a contribution of the crystal field (E_{loc}) and a contribution of the interaction between the dipoles, $E_A{\approx}E_{loc}{+}4J_c$, J_c being a coupling parameter. In our sample, the freezing is dominated by random interactions $(E_{loc}{\approx}210 \text{ K}, 4J_c{\approx}620 \text{ K})$ and should be close to spin-glass-like. Figure 2 shows the real part of the dielectric constant $\varepsilon_c'(\nu)$ for selected frequencies ν from $\nu=0.1$ Hz to $\nu=1$ MHz. As can be seen from this figure, it is only below about 40 K that the 0.1 Hz and the 1 Hz curves split. Thus the 0.1 Hz curve represents the static dielectric response for temperatures above 40 K. The maximum at about 55 K is a property of the static permittivity that signals the onset of short-range antiferroelectric ordering.

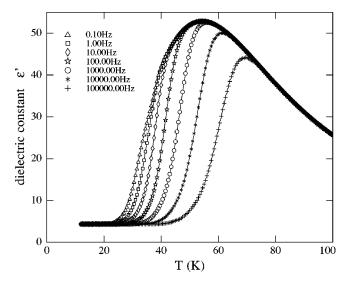


FIG. 2. Temperature dependence of the real part of the dielectric constant, $\varepsilon'_{c}(\nu)$.

A. Isothermal aging

The first experiments refer to isothermal aging and serve to characterizing the isothermal time evolution of the dielectric constant. The system was heated to a high temperature $T=80~\rm K$ to erase a possible thermal history. We then cooled down with a fast rate of $|\Delta T/\Delta t|=7~\rm K/min$ to the measurement temperature T_a . We chose $T_a=30~\rm K$, 32 K, and 34 K, where $\varepsilon_c''(\nu)$ has reasonable high values for the selected frequencies between $\nu=0.2~\rm Hz$ and 200 Hz. The temperature was then kept constant for the aging time $t_a=3~\rm days$. During the aging process, $\varepsilon_c'(\nu)$ and $\varepsilon_c''(\nu)$ were continuously measured for frequencies from $\nu=0.2~\rm Hz$ to $\nu=200~\rm Hz$. From their measurements on the dipolar glass $K_{1-x} \rm Li_x TaO_3$, Alberici $et~al.^{9,10,14,15}$ find that the time evolution of the dielectric constant is well described by a power law,

$$\varepsilon'(\nu,t) = \varepsilon'(\nu,\infty) + \Delta\varepsilon' \left(\frac{t_w + t_0}{t_0}\right)^{-\alpha}.$$
 (1)

 $\varepsilon'(\nu,\infty)$ is the asymptotic limit at infinite time, $\Delta\varepsilon'$ measures the magnitude of the time-dependent part and α characterizes the decay. Because cooling, heating, and stabilizing the temperature always takes some time, the start of an aging experiment is not well defined. The time $t_0(>1)$ takes into account a possible delay of the equilibrium of the temperature across the sample and the sample cell. The evolution of both imaginary and real part of the permittivity for all frequencies and all temperatures is well described by Eq. (1). See Table I for the fit parameters and Fig. 3 for the ν =0.2 Hz data on the real and imaginary parts of the permittivity measured at $T_a = 30,32,34$ K. For each combination of T and ν we obtain about the same values of t_0 and α from fits to $\varepsilon'(t)$ and $\varepsilon''(t)$. On the other hand, t_0 and α depend strongly on T (and slightly on ν). t_0 varies from about 900 s at 34 K to 5000 s at 30 K. The temperature equilibration time of our setup is of the order of some minutes. t_0 is clearly larger than that. Obviously, the equilibration between the thermal bath and the polar degrees of freedom of the sample takes somewhat longer. The exponent α of the power law decay increases with decreasing T. A fit of a linear T dependence,

$$\alpha(T) = A \left| (T_c - T)/T \right| \tag{2}$$

(with a parameter A=-0.05), to the data (Fig. 4) suggests a characteristic temperature T_c of about 38 K above which aging no longer occurs. Also experimentally we find a temperature of about T=38 K above which we do not observe aging behavior. This is confirmed by direct observation: Fig. 5 shows an aging experiment at T=40 K for frequencies ν of $\nu=2$ Hz, 20 Hz, and 200 Hz where the dielectric loss remains constant in time. Because the effect of aging is more pronounced in the imaginary part $\varepsilon_c''(\nu)$, we will in the following experiments concentrate on this.

B. Scaling experiments

In spin glasses an ωt_w scaling is observed, i.e., the aging curves for different frequencies fall on a single master curve if the waiting time t_w is multiplied by the applied angular

TABLE I. Fit parameter for the frequencies $\nu = 0.2$ Hz, 2 Hz, and 20 Hz for temperatures $T = 30$ K, 32 K,
and 34 K. The prime values indicate results for the real part, and the double prime values for the imaginary
part.

T(K)	$\nu(\mathrm{Hz})$	$arepsilon_{\infty}'$	$\Delta arepsilon'$	α'	t_0'	ϵ_∞''	$\Delta \varepsilon''$	lpha''	t_0''
30	0.2	12.37	0.81	0.37	4800	3.37	0.53	0.40	4800
	2	7.95	0.25	0.41	4750	2.28	0.24	0.42	5250
	20	5.67	0.05	0.35	5250	1.24	0.08	0.37	5250
32	0.2	18.40	1.28	0.24	1400	3.83	0.49	0.28	1400
	2	11.8	0.50	0.22	1600	3.97	0.39	0.27	1600
	20	7.22	0.12	0.24	1600	2.27	0.15	0.23	1500
34	0.2	24.96	1.56	0.19	900	2.79	0.29	0.22	900
	2	18.09	1.01	0.15	900	5.36	0.47	0.20	900
	20	10.31	0.26	0.19	900	4.01	0.30	0.16	700

frequency ω ($\omega = 2\pi\nu$). In orientational glasses there often is a deviation from ωt_w scaling, indicating that microscopic time scales are still relevant for the aging process. In Fig. 6, $\varepsilon'' - \varepsilon''(\nu, \infty)$ is plotted versus $\omega(t_w + t_0)$ in a linear-logarithmic scale. The values for $\varepsilon''(\nu, \infty)$ and t_0 are the ones obtained from the fits of Eq. (1) to the data. For $T_a = 32$ and 34 K the data taken at lower frequencies still do show aging but no longer obey ωt_w scaling, the deviations increase with decreasing ν . The Arrhenius law of Fig. 1 translates into a boundary $\nu_0(T)$ in the T, ν plane separating the regimes in which the dipoles are mobile or frozen in the sense that they can or cannot follow the oscillations of the applied electric field (Fig. 7). ν_0 is the most probable relaxation rate at the chosen temperature. Since the distribution of relaxation rates

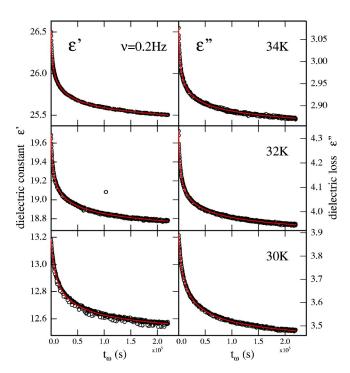


FIG. 3. (Color online) Isothermal time evolution of the imaginary part of the 0.2 Hz permittivity after quenching the sample from 80 K to T_a = 30 K, 32 K, and 34 K, respectively. Solid lines are fits after Eq. (1).

extends over several decades, ¹² the crossover from mobile to frozen states is rather gradual and we still observe aging for ν , T combinations well below the $\nu_0(T)$ boundary, which is deep in the mobile regime. It is only at temperatures above about 38 K that aging effects can no longer be resolved in the frequency range of the present study. See Fig. 5 for T = 40 K. For the ωt_w scaling, however, the distinction of frozen and mobile states given by the $\nu_0(T)$ boundary appears to be quite reliable. At 30 K all measuring frequencies chosen fall into the frozen regime, $\nu > \nu_0(T)$, thus the ωt_w scaling holds for all measuring frequencies used. For 32 and 34 K only data with $\nu > \nu_0(T)$ obey scaling.

C. Rejuvenation and memory

If the system has aged at a temperature T_1 in the frozen regime and the temperature is changed thereafter to a new value T_2 , it may forget that it has aged at T_1 . This is rejuvenation. For perfect rejuvenation the values of the quantity

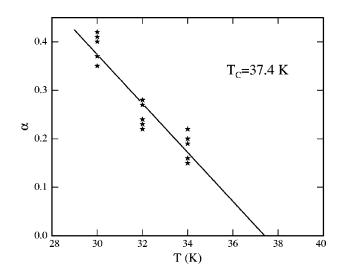


FIG. 4. Temperature dependence of the exponent α of the power law for isothermal aging [Eq. (1)]. The symbols (\star) indicate the values fitted for α' und α'' from Table I for the different frequencies for the respective temperature. The linear fit points to a critical temperature of T_c =37.4 K.

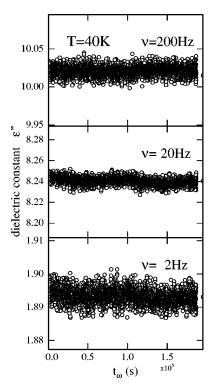


FIG. 5. Time evolution of the dielectric loss ε'' at T=40 K for $\nu=2$ Hz, 20 Hz, and 200 Hz. ε'' stays constant in time and there is no aging behavior.

of interest (here ε'') at T_2 are the same with and without aging at T_1 . Returning to T_1 the system may remember that it has aged at this temperature before. In case of perfect memory the value of ε'' after returning to T_2 is identical to the final value of the previous aging at this temperature. If T_2 is well in the mobile regime, rejuvenation and the loss of memory is complete and on returning to T_1 the system is

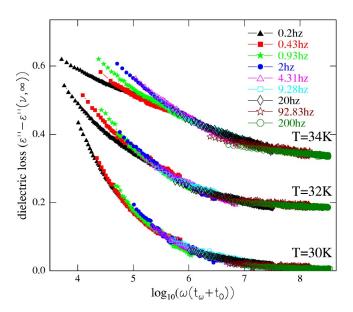


FIG. 6. (Color online) $\varepsilon'' - \varepsilon''(\nu, \infty)$ vs $\omega(t_w + t_0)$ in a linear-logarithmic scale (ωt_w scaling) for the temperatures $T_a = 30$ K, 32 K, and 34 K and frequencies from $\nu = 0.2$ Hz to $\nu = 200$ Hz.

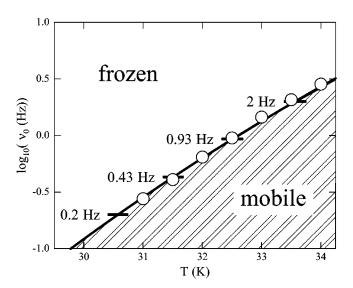


FIG. 7. Freezing line that separates frozen from mobile states. The solid line is the Arrhenius law after Fig. 1, the points are corresponding experimental data. Measurement frequencies are marked.

reinitialized. Thus the effects of rejuvenation and memory are typically probed by a temperature-time protocol consisting of isothermal aging over time periods t_1 , t_2 , t_3 at temperatures T_1 , T_2 , T_1 with $T_2 = T_1 + dT$. In the clathrate the ν ,T regime in which the dipole system is frozen but where on the other hand $\varepsilon''(T,\nu)$ is sufficiently large such that the slight changes of ε'' brought forth by aging can be studied with a reasonable resolution is rather narrow. We have chosen ν =0.2 Hz, T_1 =30 K, and dT=+2,+1,-1,-2 K in combination with periods t_1 = t_2 = t_3 =30 h [and realize that for dT=2 K, T_2 =32 K, the system is already mobile if one strictly refers to the $\nu_0(T)$ boundary of Fig. 7]. The results are shown in Fig. 8, including a reference curve where aging

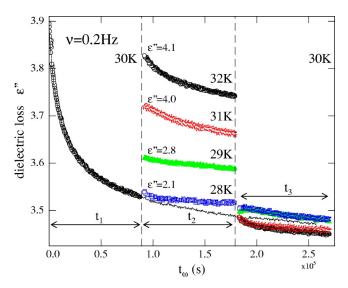


FIG. 8. (Color online) Temperature-cycling experiment for $\nu = 0.2$ Hz and starting temperature T = 30 K. The curves in interval t_2 are shifted by arbitrary values of ε'' , the original values are given in the figure.

has not been interrupted by a T excursion, that is with dT=0. The initial state at the beginning of the first period has been approached by rapid cooling out of the mobile regime at 80 K by cooling down with a rate of 7 K/min. For the changes from T_1 to T_2 and back to T_1 the rate was 1 K/min. The t_2 section of the curves has been shifted vertically by arbitrary amounts in order to present the results on a sufficiently expanded scale, but the absolute values of $\epsilon^{\prime\prime}$ in this interval can be recovered since the start values are indicated in the figures. The results on the t_1 interval are of course independent on the subsequent value of the temperature excursion dT and reproduce what has been shown already in Fig. 3. The start values of the t_2 interval are close but not quite identical to what is obtained by measuring the dielectric response on continuous heating or cooling. ε'' at T = 32 K for ν = 0.2 Hz may serve as an example. Without previous aging a value of about 4.3 is obtained as shown in Fig. 3. After aging at 30 K for 30 h, ε'' recovers from a value of about 3.5 at the end of 30 K aging t_1 period to 4.1 at 32 K. Thus there is rejuvenation, but rejuvenation is not complete, meaning that the age of the system at the temperature T_2 depends somewhat on the former aging at the temperature T_1 . As shown already in Ref. 13 complete rejuvenation is only observed for T changes dT large enough. Bringing the sample back to $T_1 = 30$ K in Fig. 8 after the excursion to T_2 the response is not reinitialized but the system remembers that it has aged at this temperature before. In fact the aging curves of the t_3 interval are quite close to the reference curve for which aging has not been interrupted by a temporary T excursion. This statement holds independent of whether dTand the change $d\varepsilon''$ of ε'' induced by changing the temperature from T_1 to T_2 is positive or negative. A closer inspection shows, however, that the curves of the t_3 interval are slightly shifted with respect to the reference curve, upward for dT $<0, d\varepsilon<0$ and downward for $dT>0, d\varepsilon>0$. Note that the sign of shift is opposite to that of $d\varepsilon$. Thus there is memory not only of the t_1 period but also a residual one of the t_2 period. Surprisingly, it is not the sign of $d\varepsilon''$ what the system remembers. We propose to refer instead to the age at the temperature T_1 being modified by the excursion to the temperature T_2 . For this purpose the curves of the t_3 interval are shifted horizontally until they coincide with the reference curve. For dT < 0 this shift is about 0.5×10^5 s to the left. Thus intermediate aging at temperatures somewhat lower than T_1 contributes to the age at T_1 but is only about half as effective as the aging at T_1 . For dT>0 aging at T_2 is more effective than aging at T_1 for the same time span, the effective age is increased by $t = 6.5 \times 10^4$ s for dT = 1 K and by 1.1×10^5 for dT = 2 K.

Figure 9 shows analogous data for a slightly higher base temperature T_1 of 32 K. Most temperatures of relevance for this experiment are already in the mobile regime of Fig. 7. Nevertheless, the results are quite similar to those shown in Fig. 8, except that the starting value of the t_3 interval after an excursion to 34 K is higher than the final value of the t_1 interval. Obviously an excursion to such a high temperature has partially erased the memory and has lead to first traces of reinitialization.

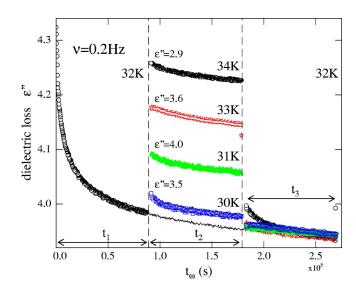


FIG. 9. (Color online) Analogous to Fig. 8 with a base temperature of $T_1 = 32 \, \text{K}$.

III. DISCUSSION

We find aging, rejuvenation, and memory in methanol-(x=0.73)- β -hydroquinone-clathrate similar to the effects in spin glasses. Although there is no experimental evidence for a sharp glass *transition* in our system, we observe a strong increase in the relaxation time with decreasing temperature. Below 32 K our system is out of equilibrium for all relevant experimental time scales and the dynamics has pronounced glassy features: In temperature cycle experiments we observed rejuvenation as well as memory effects. Since our system is randomly diluted, the dipolar interactions between the moments lead to the presence of a strong frustration due to a random mixture of ferroelectric and antiferroelectric interactions.

Two different theoretical explanations for the aging phenomena were put forward in the context of spin glasses: In the droplet theory¹⁶ rejuvenation is explained by the assumption of a finite overlap length beyond which two equilibrium configuration at different temperatures are decorrelated—a scenario that also goes under the name *chaos in spin glasses*. ^{16,17} Strong rejuvenation effects necessitate in this simplified picture a very small overlap length. If it would be much larger than the length scales that have been equilibrated during a particular waiting time, the chaotic rearrangements due to the temperature shift become invisible. However, there still might be some effect due to dangerously irrelevant droplets whose free energy gaps are quite small, as discussed in Refs. 18–21.

An alternative picture is the one presented in Refs. 2,22,23. Here rejuvenation after a negative temperature shift comes from fast modes equilibrated at T_1 , but fall out of equilibrium and are slow at T_2 . Therefore, one should expect to see this phenomenon if the equilibrated regions [on length scales $\leq L_{T_1}(t_w)$] are sufficiently different at the two temperatures. This mechanism is obviously qualitatively different from the interpretation involving the notion of temperature chaos (see above), which implies that length scale smaller than the overlap length are essentially unaffected by

the temperature shift, while larger length scales are completely reshuffled by the shift.

Moreover, within this picture the memory effect that we observe in our temperature-cycle experiments is a simple consequence of the separation of time and length scales. When the system is at $T_2 < T_1$, rejuvenation involves very small length scales as compared to the length scales involved in the aging at T_1 . Thus when the temperature is shifted back to T_1 , the correlation of length scale $L_{T_2}(t_s')$ nearly instantaneously reequilibrate at T_1 . The memory is just stored in the intermediate length scales, between $L_{T_2}(t_s')$ and $L_{T_1}(t_s)$.

The latter model accounts well for the aging effects observed in spin glasses. Strong random fields presumably destroy the spin-glass character. In orientational glasses, in which the dipole dynamics is usually strongly influenced by local crystal fields, which are randomly distributed from site to site ("random fields"), aging effects might be more appropriately described by a slow domain growth, where the domain wall movement is impeded by the inhomogeneous crystal fields. In this case the objects that are subject to reconformations due to temperature changes are only the domain walls whereas in spin glasses one expects that it is the whole bulk of the domains.

In the present case local random fields arise from the partial occupation. The neighborhood of statistically empty and filled cavities leads to a random distortion of the crystal field in the cages. Considering this special feature of dipolar glasses, aging in the dipolar glass $K_{1-x}Li_xTaO_3$ has been explained by Alberici-Kious *et al.*^{9,10} by a domain model. They assume the existence of polarization domains, i.e., regions of strongly coupled dipoles (even though the interaction is random and the domains will not be comparable to ordered domains as, e.g., in a ferromagnet). During the process of domain growth the domain walls have to overcome energy barriers via thermal activation, in close analogy what is supposed to happen in random-field systems²⁴ as well as in the droplet theory for spin glasses. 16 Only molecules in the domain walls contribute to the dielectric response, molecules from inside the domains are already frozen. The dielectric response ε'' is inversely proportional to the domain size R, $\varepsilon'' \propto 1/R(t)$, as the volume fraction of wall molecules reduces when domains grow. For each temperature T there is an equilibrium domain size $\xi(T)$. The lower the temperature, the larger the ξ ; $\xi(T) \propto 1/\sqrt{T}$. All domains grow towards their equilibrium size; the evolution stops when they have reached $\xi(T)$. When the temperature is increased the equilibrium correlation length decreases and it might happen that some of the domains have grown beyond this length scale and have to adjust, i.e., shrink. For domains with size R(t,T) larger than $\xi(T)$ shrinking would be a very slow process because they

have to overcome large energy barriers. A more efficient scenario would be the nucleation of small domains (with sizes of the order of the lattice spacing) within the larger domains and a rapid initial growth. In this way nucleation would generate new domain walls giving rise to the overshoot of the dielectric response in time interval t_3 of the temperature cycling experiments in Fig. 8.

Matsuo and Suga²⁵ find for β -hydroquinone-clathrates with various polar guest molecules a proportionality between the transition temperature, where the dipole system orders, and the square of the dipole moment, i.e., between the thermal energy and the energy of the dipolar interaction between the guest molecules. This suggests that the dipole-dipole interaction between the guest molecules plays the leading role for the dielectric behavior. From measurements of the methanol(x = 0.97), (x = 0.84), and (x = 0.79) samples, ¹¹ the (hypothetical) transition temperature of the methanol(x= 0.73)- β -hydroquinone-clathrate can be estimated to about T=38 K, where molecular reorientations should slow down drastically. The critical temperature T_c that arises from the dielectric experiments agrees quite well with the experimental finding suggesting that the collective dipolar interactions lead to aging, memory, and rejuvenation.

IV. CONCLUSIONS

We present dielectric measurements with different temperature-time protocols to study aging effects in the dipolar glass methanol(x = 0.73)- β -hydroquinone-clathrate. We find aging, rejuvenation, and memory effects, although there is no experimental evidence for a sharp glass transition. The diluted methanol clathrate is conceptually close to its magnetic pendants, the spin glasses. The electrical behavior is dominated by the dipole-dipole interactions of the guest molecules but the coupling of the pseudospins to random fields is not negligible. We argue that the diluted methanolclathrate takes a somehow intermediate character between a true spin glass and a pure random-field system. The influence of random fields cannot be neglected as the effects are much broader than the corresponding effects in spin glasses (e.g., in CdCr_{1.7}In_{0.3}S₄, see Ref. 5), and the overshoot in the time interval t_3 is well explained by the domain model. Furthermore, memory is preserved at moderate temperature increase opposed to the observations in spin glasses. On the other hand the ωt_w scaling at low temperatures and the temperature dependence of the fitting parameter α resemble the effects found in spin glasses and suggest a random bond dominated system. Aging, rejuvenation, and memory effects can just be observed below a temperature at which the dipolar motions are expected to slow down and collective dipolar interactions become dominant.

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