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Mode coupling, universality, and the glass transition

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We investigate the current status of the mode-coupling theory (MCT) of the glass transition by making a comparison with two recent sets of experiments: the scaling data resulting from the dielectric measurements of Dixon *et al.* [Phys. Rev. Lett. **65**, 1108 (1990)] and the neutron-scattering data. Our main conclusions include the following. (a) Experiments do show the sequence of time scales in the relaxation predicted by MCT. In particular, we believe that the high-frequency tail of the data of Nagel and co-workers is the realization of the so-called von Schweidler relaxation predicted by MCT in the frequency regime. (b) The nature of scaling discovered by Dixon *et al.* is quite different from and more universal than that predicted by MCT, which is basically a time-temperature superposition. Furthermore, the dielectric measurements show that there exists a universal relation between the exponent b governing von Schweidler relaxation and the exponent β governing a stretched exponential: $1+b=\frac{3}{4}(1+\beta)$. (c) Dixon *et al.* find no evidence for the existence of a special temperature T_0 (well above the phenomenological glass-transition temperature T_g) above and below which the relaxation dynamics is substantially different. This constitutes a discrepancy between the two sets of experiments since neutron-scattering experiments have been interpreted as confirming the existence of such a temperature well above T_g. A possible reinterpretation of MCT is suggested so as to reconcile these two sets of experiments and MCT.

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I. INTRODUCTION

Recent theoretical [1] and experimental [2] developments treating the dynamics of the liquid-glass transition have led to considerable clarification of the nature of this "transition." A scaling picture [3] is emerging with a complexity far more elaborate than one would have guessed only a few years ago. From a theoretical point of view there has been considerable progress due to the so-called mode-coupling theory (MCT) of the glass transition. Leutheusser [4] initiated this development by showing that a model obtained from the kinetic theory of dense fluids exhibits an ergodic-nonergodic transition which shares many features with the liquid-glass transition. This model also leads to a viscosity which diverges as $(T-T_0)^{-\gamma}$ as the temperature T approaches the "ideal glass-transition temperature" T_0. The exponent γ was found [5] to take values $\gamma\approx 2$. Das and Mazenko later discovered a cutoff mechanism [6] which rounds off the sharp nature of the ergodic-nonergodic transition, keeping the system ergodic and the viscosity finite for all temperatures. Another, less appreciated, feature of the Leutheusser model is the existence of a sequence of time scales (see Fig. 1) which enter the relaxation on the liquid side of the transition. Recent experiments [7] have given support to this scenario of a sequence of time scales, as carefully elucidated and generalized in MCT by Götze and Sjögren [8], in the relaxation. On the other hand, however, it is also true that there are serious discrepancies between the MCT picture of Götze and Sjögren and the high-precision measurements of Nagel and co-workers [3]. These experiments point to a scaling picture which is considerably more universal than that suggested by MCT [9]. It is also true that Dixon *et al.* [3] find no evidence for the ideal glass-transition temperature T_0 highlighted by Götze and Sjögren [8] in their work.

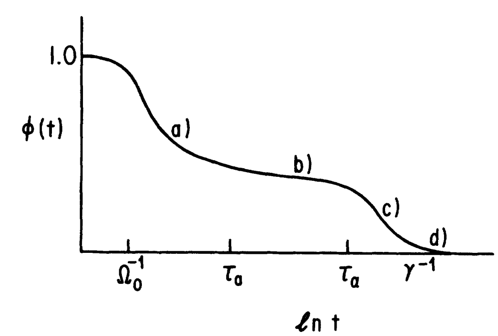

FIG. 1. A schematic plot of the sequence of relaxation mechanisms predicted by MCT; (a) power-law-decay t^{-a} relaxation, (b) von Schweidler relaxation $-Bt^b$, (c) primary relaxation $e^{-(t/\tau)^\beta}$, (d) exponential relaxation $e^{-\gamma t}$.

In this paper we investigate both the considerable agreement between the MCT and experiments and the unresolved discrepancies.

We begin in the next section with a review of the basic theoretical MCT picture as elucidated primarily by Götze and co-workers. This will then serve as a framework in which to discuss some of the important recent experiments.

II. MODE-COUPLING THEORY: OVERVIEW

The basic assumption of the mode-coupling theory is that the slowing down observed near the glass transition is governed by density fluctuations. Another, less crucial, assumption is that the glass transition is insensitive to the probing wave number q and it is typically assumed that the various wave-number components can be treated independently. Under these assumptions the theory is formulated in terms of the normalized density-correlation function

$$\phi(t)=\frac{\langle\rho_{\mathbf{q}}(t)\rho_{-\mathbf{q}}(0)\rangle}{\langle\rho_{\mathbf{q}}(0)\rho_{-\mathbf{q}}(0)\rangle} \tag{2.1}$$

where the wave-number dependence of $\phi(t)$ is suppressed. The Laplace transform of $\phi(t)$, defined as

$$\phi(z)=(-i)\int_0^\infty dt\, e^{izt}\phi(t),\quad \mathrm{Im}(z)>0\ , \tag{2.2}$$

has, in the mode-coupling model, the representation [6]

$$\phi(z)=\frac{z+id(z)}{z^2-\Omega_0^2+id(z)[z+i\gamma(z)]}\ . \tag{2.3}$$

The frequency Ω_0 in (2.3) corresponds to a microscopic "phonon" frequency. The renormalized viscosity $d(z)$ in (2.3) is coupled back to the density-correlation function $\phi(t)$ via

$$d(z)=d_0+\Omega_0^2\int_0^\infty dt\, e^{izt}H(\phi(t)) \tag{2.4}$$

where d_0 is the bare viscosity governing the microscopic dynamics. It has typically been assumed that the mode-coupling kernel $H(\phi(t))$ can be truncated at a low finite order in an expansion in $\phi(t)$:

$$H(\phi(t))=\sum_{n=1}^{N}c_n\phi^n(t)\ . \tag{2.5}$$

As discussed further below, the origin of the linear term in (2.5) is not clearly understood from the point of view of fundamental theory. The model where only c_2 is nonzero corresponds to the original Leutheusser model. The more general model given in (2.5), but with $c_1=0$, was introduced in Ref. [10]. The importance of the c_1 term was first pointed out by Götze [11]. In (2.3) all quantities are considered as rather weak functions of wave number, which has been suppressed.

The frequency $\gamma(z)$ in (2.3), introduced by Das and Mazenko [6], serves to cut off the ergodic-nonergodic transition that one finds in its absence. If, as $z\to 0$, the viscosity $d(0)$ becomes very large, then (2.3) reduces to

$$\phi(z)=\frac{1}{z+i\gamma(0)} \tag{2.6}$$

and $\phi(t)$ decays to zero as $e^{-\gamma t}$. If, however, $\gamma=0$, then $\phi(z)\sim z^{-1}$ and $\phi(t)$ does not decay to zero as $t\to\infty$ and the system is not ergodic. It seems crucial, in order to obtain the very slow relaxation observed in experiment, that $\gamma(t)$ remains small for all times and γ^{-1} must be larger than any other time in the problem. Theoretically, at lowest order in perturbation theory, γ is given by

$$\gamma(z)=\gamma_0\int_0^\infty dt\, e^{+izt}[\dot\phi(t)]^2 \tag{2.7}$$

where γ_0 is a constant that is ~ 1. It is easy to see that (2.7) leads to substantial values of $\gamma(0)$ unless one excludes the early-time contributions to γ and writes [12]

$$\gamma(z)=\gamma_0\int_{t_0}^\infty dt\, e^{+izt}[\dot\phi(t)]^2\ . \tag{2.8}$$

For sufficiently large t_0 one can make $\gamma(z)$ rather small. We assume here that $\gamma(0)$ can be made sufficiently small so that it can be neglected in (2.3) except in treating the longest-time scale.

If we set $\gamma(z)=0$, we can combine (2.3) and (2.4) to obtain the equation of motion for $\phi(z)$:

$$\frac{-\Omega_0^2\phi(z)}{1-z\phi(z)}=z+id_0+i\Omega_0^2\int_0^\infty dt\, e^{izt}H(\phi(t))\ . \tag{2.9}$$

Equation (2.9) has the form in the time domain

$$\ddot\phi(t)+d_0\dot\phi(t)+\Omega_0^2\phi(t)+\Omega_0^2\int_0^t ds\, H(\phi(t-s))\dot\phi(s)=0 \tag{2.10}$$

with initial conditions $\phi(0)=1$ and $\dot\phi(0)=0$. Equation (2.10) forms the basis of the "mode-coupling model" (MCM). The associated parameters are d_0, Ω_0^2, and the c_n.

The basic assumption in the analysis of (2.10) is that there is, depending on the values of the c_n, an extended intermediate-time regime over which $\phi(t)$ is very slowly varying. Indeed it is assumed that there is a time range over which $\phi(t)$ is approximately time independent and one can write

$$\phi(t)=f+(1-f)\phi_v(t) \tag{2.11}$$

or equivalently, in terms of the Laplace transform,

$$\phi(z)=\frac{f}{z}+(1-f)\phi_v(z)\ , \tag{2.12}$$

where f is a metastable value of $\phi(t)$ yet to be specified. For the frequency region where the inequality

$$|z\phi_v(z)|\ll 1 \tag{2.13}$$

is valid, substituting (2.12) into (2.6) and expanding the left-hand side of (2.9), keeping leading orders up to $O(\phi_v^2(z))$, we obtain, for $\phi_v(z)$,

$$\frac{\sigma_0}{z}+\sigma_1\phi_v(z)-z\phi_v^2(z)+\lambda L(\phi_v^2(t))(z)=0 \tag{2.14}$$

where L stands for the Laplace transform. The coefficients σ_0, σ_1, and λ are defined by

$$\sigma_0=(1-f)V(f)\ , \tag{2.15}$$

$$\sigma_1=(1-f)^2V'(f)\ , \tag{2.16}$$

$$\lambda=\tfrac{1}{2}(1-f)^3H''(f)\ , \tag{2.17}$$

where $V(f)\equiv H(f)-f/(1-f)$ and $V'(f)=\partial V/\partial f$, etc. Equation (2.14) is the fundamental equation for the analysis of the time regime (called the intermediate-time regime here) leading up to and including the early stages of α relaxation. (See Fig. 1.)

The self-consistency of (2.11) and (2.14) is maintained only if the parameters σ_0 and σ_1 are "small." Slow relaxation results for those values of c_n for which σ_0 and σ_1 are small. Götze and Sjögren take this one step further [8] and assume that σ_0 and σ_1 can be made small by adjusting the temperature and density. For reasons developed below we refrain here from this assumption.

An ideal metastable state is achieved if both σ_0 and σ_1 are zero. We are free to determine f by requiring $\sigma_1=0$, and obtain from (2.16) that

$$H'(f)=\frac{1}{(1-f)^2} . \tag{2.18}$$

Equation (2.18) can be solved to give $f=f(c_n), \sigma_0=\sigma_0(c_n)$, and $\lambda=\lambda(c_n)$. From a mathematical point of view the ideal transition state is achieved for the set of c_n^* where $\sigma_0(c_n^*)=0$. For the Leutheusser model [4]

$$H_L=c_2\phi^2 . \tag{2.19}$$

This forces c_2 to take a single critical value $c_2^*=4$. For the Götze model [11]

$$H_G=c_1\phi+c_2\phi^2 \tag{2.20}$$

one has a line of critical values

$$c_1^*=\frac{2\lambda-1}{\lambda^2} , \quad c_2^*=1/\lambda^2 , \tag{2.21}$$

where λ takes values $\frac{1}{2}\leq\lambda<1$.

For higher-order models one obtains critical surfaces. The key assumption associated with the existence of slow dynamics is that σ_0 is small. This requires that the system chooses the set of c_n such that σ_0 is small. This does not require that σ_0 possess a zero at some definite values of density and temperature. Equation (2.14) can then be written as

$$\frac{\sigma_0}{z}-z\phi_v^2(z)+\lambda L(\phi_v^2(t))(z)=0 . \tag{2.22}$$

For the higher-frequency region where the first term can be neglected compared with other terms in (2.22),

$$(|\sigma_0|)^{1/2}\ll|z\phi_v(z)| , \tag{2.23}$$

Eq. (2.22) reduces to

$$-z\phi_v^2(z)+\lambda L(\phi_v^2(t))(z)=0 . \tag{2.24}$$

Equation (2.24) is satisfied by

$$\phi_v(t)=A(t\Omega_0)^{-a} \tag{2.25}$$

where the constant A is determined from the microscopic details which are already eliminated from (2.14). The exponent a is given in terms of the parameter λ by

$$\frac{\Gamma^2(1-a)}{\Gamma(1-2a)}=\lambda=\tfrac{1}{2}(1-f)^3H''(f) \tag{2.26}$$

where Γ is the gamma function. The inequality (2.23) and the solution (2.25) imply that the above solution holds only in the time region

$$\Omega_0^{-1}\ll t\ll\tau_a\equiv|\sigma_0|^{-1/2a} . \tag{2.27}$$

The time scale τ_a diverges at the ideal metastable state where $\sigma_0=0$. Therefore $\phi(t)$ decays algebraically toward the specified metastable value f of $\phi(t)$.

For the time region where $t\gg\tau_a$, the dynamics is quite different for the cases $\sigma_0>0$ and $\sigma_0<0$. For $\sigma_0>0$, (2.22) has the solution $\phi_v(z)\sim(\sigma_0)^{1/2}/z$ as $z\rightarrow0$ and

$$f=f_0+c(\sigma_0)^{1/2}+O(\sigma_0) \tag{2.28}$$

where f_0 is the value of f at the ideal transition point and c is a constant. The dynamics are more interesting for the case where $\sigma_0<0$. In this case the solution, which is more singular than $1/z$ as $z\rightarrow0$, has the form in the time regime

$$\phi_v(t)=-B(t/\tau_\alpha)^b , \tag{2.29}$$

where B is a positive constant determined from the microscopic details and where the exponent b satisfies

$$\frac{\Gamma^2(1+b)}{\Gamma(1+2b)}=\lambda . \tag{2.30}$$

Equation (2.29) is known as the von Schweidler relaxation law. Since the von Schweidler relaxation eventually violates the inequality (2.13), this decay mechanism is valid in the time regime

$$\tau_a\ll t\ll\tau_\alpha \tag{2.31}$$

where τ_α is given by

$$\tau_\alpha=|\sigma_0|^{-(1/2a+1/2b)} . \tag{2.32}$$

For $t\geq\tau_\alpha$, the system enters into the α-relaxation regime [13]. In this regime the equation of motion (2.22) cannot be used since the inequality (2.13) breaks down and one has to consider the original equation of motion (2.9). While the analytic solution for (2.9) is not known, the numerical solution for (2.9), or equivalently (2.10), for certain sets of parameters in the α-relaxation regime is well fit by a stretched exponential. Therefore, by assuming that the stretched exponential of the form

$$\phi(t)=fe^{-(t/\tau_\alpha)^\beta} \tag{2.33}$$

is a solution of (2.10) and looking at the small-frequency limit, we obtain an approximate expression for the exponent β in terms of the parameters c_n;

$$\sum_{n=1}^{N}c_nfn^{-1/\beta}=1 . \tag{2.34}$$

As an example, for the Leutheusser model (2.13), (2.34) can be rewritten as

$$\beta_L=\frac{\ln2}{\ln(c_2f)} . \tag{2.35}$$

Equation (2.35) gives β close to 1 near the ideal metastable state where $c_2=4$ and $f=\frac{1}{2}$ and hence the relaxation is exponential, not showing a stretching behavior. Götze [11] pointed out that by adding a phenomenological linear term $c_1\phi$ to H_L, the model shows a stretching. For Götze's model (2.20) β can be written as

$$\beta_G=\frac{\ln2}{\ln[c_2f/(1-c_1)]}=-\frac{\ln2}{\ln(1-\lambda)} . \tag{2.36}$$

In this model, for example, with $\lambda=0.6$, (2.36) yields $\beta=0.76$.

In summary, the sequence of time scales predicted by MCT follows from the behavior of the parameter σ_0 (see Fig. 1). For $\sigma_0<0$, we have first the power-law decay, followed by the von Schweidler relaxation, the stretched exponential relaxation, and finally the cutoff γ enters and one obtains, finally, exponential decay. For $\sigma_0=0$, the power-law decay is extended to the longest-time scale until the cutoff γ becomes relevant. For $\sigma_0>0$, the system, after the initial power-law decay, freezes in a state of structural arrest until finally the cutoff γ enters and returns the system to ergodicity.

III. EXPERIMENTAL RESULTS

A. Scattering experiments

We now turn our attention to recent experiments and discuss in detail the extent to which MCT is compatible with these experiments. Consider first the neutron-scattering measurements of the normalized density-correlation function $\phi_q(t)$ of the ionic liquid [14] $[Ca_{0.4}K_{0.6}(NO_3)_{1.4}]$ and polymeric liquid (polybutadiene) [15] at $q=q_0$, the wave number associated with the first maximum in the structure factor. These experiments reveal that $\phi_{q_0}(t)$ has two well-separated relaxation regimes. The earlier regime, probed by neutron time-of-flight measurements [16] with a time window extended to 10^{-3} sec shows the power-law decay t^{-a} predicted by MCT. The intensity spectrum in the recent light-scattering experiments by Tao, Li, and Cummins [17] exhibits an $\omega^{-(1-a)}$ frequency dependence in the intermediate-time region, supporting the existence of the power-law decay. Neutron spin-echo measurements [14], with a time window of 10^{-8}–10^{-11} sec, shows a slowing down as the temperature is lowered. The relaxation is well fit by the stretched exponential form (2.33) with $f\sim0.84$, $\beta\sim0.58$ for the ionic liquid and $f\sim0.90$, $\beta\sim0.45$ for the polymeric liquid. It was concluded that for these systems, f and β in (2.33) are temperature independent and the relaxation time τ alone carries the temperature dependence. In this case the scaling takes the form of "time-temperature superposition." This means that data at any temperature can be fit by (2.33) by adjusting the temperature-dependent relaxation time τ. Hence, (2.33) is indeed a scaling function of the relaxation, that is, the time-temperature superposition holds.

Another main result of the neutron-scattering experiments is the measurement of the temperature dependence of the plateau value f. In (2.28), if it is assumed that $\sigma_0(T)\propto T_0-T$, T_0 being the temperature at which an ideal metastable state is achieved, then we have

$$f(T)=\begin{cases} f(T_0)+c(T_0-T)^{1/2}, & T<T_0 \\ f(T_0), & T>T_0 \end{cases} . \tag{3.1}$$

The above cusplike temperature dependence has been observed in a polymeric liquid [18] where they found $T_0=216$ K, about 35 K above the phenomenological glass-transition temperature T_g. This observation has been viewed as important evidence in favor of MCT. The important implication is that above T_g there exists a specific temperature T_0, above and below which the dynamics is quite different.

There are a number of other experiments [19] and numerical simulations [20] which find results similar to those described above.

B. Dielectric measurements

Nagel and his collaborators [3] recently discovered a sophisticated form of scaling in the α relaxation of supercooled liquids. From the measurements of the dielectric susceptibility of a variety of glass-forming liquids they observed that the data for *all* sample liquids studied can be scaled so that they fall on top of one another over 13 decades of frequency (10^{-3} Hz $<\omega/2\pi<10^{10}$ Hz). The scaling curve is described by two parameters, the peak frequency ω_p and the normalized width W (with respect to the Debye width $W_d=2\log_{10}(2+\sqrt{3})=1.14\ldots$) of the imaginary part of the dielectric susceptibility $\chi''(\omega)$. While the observed scaling curve can be well fit by a stretched exponential in the low-frequency region, it significantly deviates from the scaling curve for the stretched exponential in the high-frequency tail. We believe that this high-frequency tail is the realization of the von Schweidler relaxation (2.29) in the frequency regime.

The scaling discovered in the dielectric measurements corresponds to plotting $(1/W)\log_{10}\phi(\omega)$ versus $(1/W)(1+1/W)\log_{10}(\omega/\omega_p)$ where $\phi(\omega)$ is the Fourier transform of the normalized density-correlation function and is related to the susceptibility via the fluctuation-dissipation theorem: $\phi(\omega)=(\chi''(\omega)/\chi)(\omega_p/\omega)$, χ being the static susceptibility. Both W and ω_p are found to be strong functions of temperature. One of the intriguing aspects of the scaling is the presence of $1/W$ and $1+1/W$ factors. The longest-time regime can be fit to the stretched exponential form (2.33) where τ and β are strong functions of temperature and one finds the empirical [21] relation $1/W\sim\beta$.

We briefly discuss here why the stretched exponential form satisfies (approximately) the scaling discussed by Dixon *et al.* [3]. It is easy to show for (2.33) that

$$\ln\phi(\omega)=\begin{cases} A_L(\beta), & \dfrac{\omega}{\omega_p}\ll 1 \\ A_H(\beta)-(1+\beta)\ln(\omega/\omega_p), & \dfrac{\omega}{\omega_p}\gg 1 \end{cases} \tag{3.2}$$

where

$$A_L(\beta)=\ln\left[\Gamma\left[1+\frac{1}{\beta}\right]\omega_p\tau\right] \tag{3.3}$$

and

$$A_H(\beta)=\ln\left[\Gamma(1+\beta)\sin\left[\frac{B\pi}{2}\right](\omega_p\tau)^{-\beta}\right] . \tag{3.4}$$

We see that dividing by $1+1/W=1+\beta$ renders the high-frequency slope temperature independent. Then, the $1/W$ factor in the scaling makes $(1/W)A_{L,H}(\beta)$

weaker functions of β than $A_{L,H}(\beta)$, hence, approximate scaling holds. Since, for example, $(1/W^2)A_{L,H}(\beta)$ are even weaker functions of β, one finds that the plot $(1/W^2)\log_{10}\phi(\omega)$ versus $(1/W^2)(1+1/W)\log_{10}(\omega/\omega_p)$ leads to improved scaling. It is important to note that this is no longer true for actual data in which the high-frequency region significantly differs from the stretched exponential form and the $1/W$ factor is preferred. Therefore the factors of $1/W$ and $1+1/W$ are essential in order to obtain the scaling for the experimental data.

MCT predicts [13] that the high-frequency tail of the α relaxation is characterized by the frequency spectrum of the von Schweidler relaxation (2.29). Using (2.29), we can then obtain an approximate expression for the high-frequency representation for the data as

$$\log_{10}\phi(\omega)=A_0(\beta)-(1+b)\log_{10}\left[\frac{\omega}{\omega_p}\right], \quad \frac{\omega}{\omega_p}\gg 1 \tag{3.5}$$

where

$$A_0(\beta)=\log_{10}\left[B_0\Gamma(1+b)\sin\left[\frac{b\pi}{2}\right](\omega_p\tau)^{-b}\right]$$

with B_0 being an amplitude factor governing the crossover from the stretched exponential relaxation to the von Schweidler relaxation. Equation (3.5) can be written in the scaling form

$$\frac{1}{W}\log_{10}\phi(\omega)=\beta A_0(\beta)-\frac{1+b}{(1+\beta)W}\left[1+\frac{1}{W}\right] \times\log_{10}\left[\frac{\omega}{\omega_p}\right], \quad \frac{\omega}{\omega_p}\gg 1 , \tag{3.6}$$

where it is assumed that $1+1/W=1+\beta$. Thus the high-frequency slope in the scaling plot is given by $(1+b)/(1+\beta)$ and the scaling implies that this slope is independent of temperature. Experiment gives the result

$$\frac{1+b}{1+\beta}=\frac{3}{4}, \tag{3.7}$$

leading to the linear relation between the two exponents b and β. Note that according to the linear relation (3.7), the condition $b>0$ leads to a constraint on β, that is, $\beta>\frac{1}{3}$. This means [22] that the width cannot be larger than 3. Likewise, the condition $\beta\leq 1$ leads to $b\leq\frac{1}{2}$.

C. Comparisons

There does seem to be agreement all around that for temperatures $T>T_0$ $(\sigma_0<0)$ there is a relaxation sequence which can be roughly described by von Schweidler's law followed by stretched exponential behavior. The experiments of Dixon *et al.* [3] go further, and the scaling they find implies a universal relation between the exponents b and β. Indeed they find a complete universal curve.

Does MCT provide an explanation for the linear relation (3.7)? MCT gives a relation between b and β via (2.30) and (2.34), and this relation is in general highly nonlinear. Note that Götze's model, (2.20), does not satisfy (3.7) since in the asymptotic region the model always yields $b>\beta$. There are choices of the parameters $\{c_n\}$ which do lead to (3.7). We find [23] that the simplest schematic model yielding the linear relation (3.7) is

$$H(\phi)=c_1\phi+c_2\phi^2+c_3\phi^3+c_4\phi^4 . \tag{3.8}$$

There is, as yet, no principle for choosing such models except the desire to fit the experiments.

This last point is indicative of the main weakness at present of the MCT. One does not know how to determine the coupling parameters $c_n(T,q)$ for a given physical system. Indeed the parameter $c_1(T,q)$, introduced phenomenologically by Götze is known to be important in obtaining the stretching in the α relaxation but is not generated in the kinetic-theory approach or in the nonlinear fluctuating hydrodynamic approach. Thus work remains to establish a principle for its existence [4].

There appears to be a discrepancy between MCT and the results of the scattering experiments, which indicate the existence of a special temperature T_0, and the scaling of Nagel and co-workers shows no temperature sensitivity for temperatures near T_0. It appears, if these two sets of experiments are to be reconciled with each other and MCT that the MCT must be reinterpreted. The experiments of Nagel and co-workers are clearly carried out at a temperature range which includes any reasonable estimate for T_0. This can be seen from an analysis of the temperature dependence of the peak frequency ω_p. The high-temperature data for ω_p^{-1} can be fit to the form $(T-T_0)^2$ and one finds, as shown in Fig. 2, for salol, $T_0=270$ K which is well placed in the middle of the temperature range studied. The simplest interpretation reconciling MCT and the data in Ref. [3] is that $-\sigma_0\sim\omega_p^{-1}$ and σ_0 is activated at low temperatures. This assumption is not in agreement with (3.1). However, it may be that the T_0 measured using (3.1) reflects the ki-

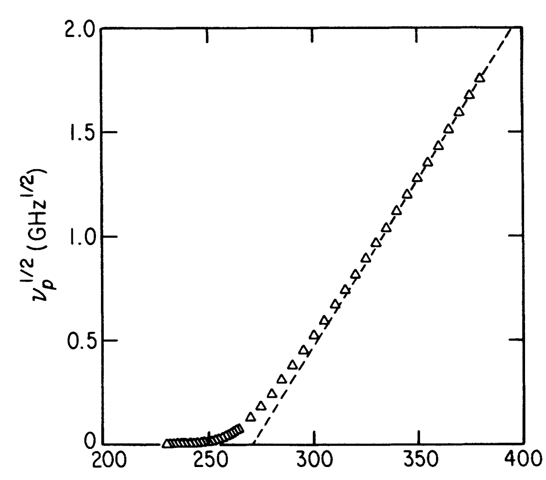

FIG. 2. $\nu_p^{1/2}$ (GHz$^{1/2}$) temperature T (K) for salol, which is taken from Ref. [3]. The dotted line is a linear fit to the last five points of the data. The crossover temperature is estimated to be $T_0=270$ K, which lies in the temperature range studied in Ref. [3].

netic effect that as T is lowered and τ grows there will be a temperature where τ passes beyond the time window sampled by a particular experiment. This temperature will have a rough correspondence to T_0 [25].

It seems appropriate to conclude that there is a temperature range aroung T_0 where relaxation times cross over from power-law to activated temperature dependence. This process is shown in Fig. 2. Once this temperature dependence is absorbed in ω_p then there is no residual temperature dependence near T_0 in the scaling function.

IV. CONCLUSIONS

While the MCT has led to considerable progress in understanding the time sequence associated with relaxation near the glass transition there remain significant questions which require study. One must find some method for determining the parameters c_n appropriate to a given physical system and their variation with temperature, density, and wave number. If one is to obtain the universal results of Nagel and co-workers, then there must be some important and universal principle which influences the physically selected c_n.

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