Metastable dynamics above the glass transition

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The element of metastability is incorporated in the fluctuating nonlinear hydrodynamic description of the mode coupling theory (MCT) of the liquid-glass transition. This is achieved through the introduction of defect density variable n into the set of slow variables with mass density ρ and momentum density $\mathbf{g}$. As a first approximation, we consider the case where motions associated with n are much slower than those associated with ρ. Self-consistently, assuming one is near a critical surface in the MCT sense, we find that the observed slowing down of the dynamics corresponds to a certain limit of a very shallow metastable well and a weak coupling between ρ and n. The metastability parameters, as well as the exponent describing the observed sequence of time relaxations, are given as smooth functions of the temperature without any evidence of a special temperature. We then investigate the case where the defect dynamics is included. We find that the slowing down of the dynamics corresponds to the system arranging itself such that the kinetic coefficient governing the diffusion of the defects approaches from above a small temperature-dependent value.

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I. INTRODUCTION

While there has been considerable progress recently in the development of theories of relaxation near the liquid-glass transition, these theories have ignored one of the fundamental defining qualities of the problem: These systems are metastable. In this paper, we introduce the element of metastability into a framework that connects with the mode coupling theory (MCT) [1]. We find a theoretical picture that is closer to the observed experimental picture than the conventional MCT, since it is freed from the idea of a sharp transition temperature T_0.

The mode coupling theory has been very successful in explaining the very elaborate sequence of time relaxations in supercooled liquids that has been observed in many experiments [2–6]. Despite its success, however, the current status of the MCT is not without questions and controversies. An important question associated with the MCT concerns the temperature dependence of the theory. The conventional MCT [1,7–9] views the glass transition as a sharp ergodic-nonergodic transition as the temperature T approaches the ideal glass transition temperature T_0 well above the calorimetric glass transition temperature T_g. According to the conventional MCT, the relaxation sequence occurs only near $T \sim T_0$, and in particular the so-called von-Schweidler relaxation and the stretching are confined to the region $T > T_0$. This sharp temperature dependence is clearly in contrast with the universal behavior of Dixon *et al.* [6], whose data cover any reasonable choice of T_0. Also, as shown in other recent experiments [10], the existence of a well-defined temperature T_0 well above T_g is very doubtful. In this formulation of the MCT, the exponents governing the time relaxations are temperature independent, just as those involved in a second-order phase transition. This is again in disagreement with recent experiments. In particular, Dixon *et al.* finds the stretching exponent β weakly temperature dependent, which means, by virtue of the universal relationship discussed by Kim and Mazenko [11], that the von-Schweidler exponent b also depends on temperature. since the MCT predicts [9] that the exponent b is related to the so-called critical exponent a through a temperature-independent relation, this also implies that a is a temperature dependent.

There exist recent efforts [12] to reconcile the discrepancies between experiments and the MCT, which include the cutoff effect, discovered by Das and Mazenko [13], into the conventional MCT but still assume a rounded transition around $T \sim T_0$. This attempt, however, ends up by adjusting as many as seven independent parameters to fit experimental data. A simpler interpretation of the situation is that the MCT, despite all the successes, needs to be reformulated at a more fundamental level such that it is not tied to the notion of a sharp transition temperature.

In Ref. [14], we proposed a model where defects and metastability play an important role in the glass transition, in an attempt to reformulate the MCT so that it would be compatible with experiment. Although this theory does not provide any clear explanation for the scaling result of Ref. [6], it does result in a smooth temperature variation without any indication of a special temperature T_0. The various exponents, according to the model, are weak functions of temperature. In this paper, our main focus is to elaborate on the detailed construction of the model, which was omitted in Ref. [14], and to further elaborate on the dynamics of the defects that we treated using a simplified assumption of the previous work.

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The model introduced in Ref. [14] is based on the introduction of the defect variable n into the set of slow variables consisting of mass density ρ and momentum density $\mathbf{g}$, in the fluctuating nonlinear hydrodynamic (FNH) description of the MCT [13]. The introduction of the defect variable would be required in a rigorous hydrodynamic description of crystalline solids, along with the Nambu-Goldstone modes associated with broken translation symmetry [15]. Although there is no broken continuous symmetry involved in the glass transition, we consider the situation where this variable plays a role in the transition [16]. The best analogy to the present case would be the situation where one considers an order parameter in the disordered state. We do not need a microscopic definition for the defect variable here. The only information we need here is that the defect density has the usual Poisson bracket relations of a scalar variable with momentum density $\mathbf{g}$, that they are metastable, and that they interact weakly with the mass density.

One of the key assumptions in the model is that motions associated with defect density n have a very long time scale compared to that of density fluctuation. This is realized in the model via an explicit double-well potential $h(n)$ for the defect variable, with the metastable defect density $\bar{n}$ associated with the minimum of the higher well. A very small diffusion coefficient Γ_v for n results from a rolling around in a shallow metastable well. We find that the coupling between the mass density and the defect variables slows down the defect motion further. The coupling also enables the slow dynamics of defects to influence the density dynamics. Over a significant time period when the defect is trapped within the metastable well, the defect autocorrelation function $\psi(t)$ can be regarded as a constant, while the density autocorrelation function $\phi(t)$ displays the relaxation sequence, leading to the von-Schweidler regime. Under this assumption, we find that the observed stretched dynamics correspond to a self-consistent limit of weak coupling and a low activation barrier for the defect. As will be discussed below, this limit corresponds to the situation where the coupling energy is weak enough not to destroy the metastable defects but still strong enough for the slow dynamics of defects to result in the slowing down of the mass density variable. In this limit, the parameters describing the double-well potential and the coupling, as well as the exponents of the relaxation sequences, are self-consistently determined as smooth functions of temperature.

Since the defect variable is diffusive, the defect autocorrelation function $\psi(t)$ decays, to linear order, as $\sim\exp(-\gamma_v t)$, where in terms of the bare diffusion coefficient Γ_v, $\gamma_v = h''(\bar{n})\Gamma_v q^2$ at wave number q. Self-consistently we find, in the time regime where $\psi(t)$ can be regarded as a constant, that $h''(\bar{n})$ must be small, i.e., the metastable wells become very shallow and broad. As one moves into the later stage of relaxation, however, we expect nonlinear corrections to γ_v become important. Therefore, we need to consider the dynamics of the defects and their coupling to the density fluctuations by including the renormalization of γ_v in the evolution equation for $\psi(t)$. In this paper, we obtain a set of coupled equations for $\phi(t)$ and $\psi(t)$ governing the dynamics of both density and defect variables.

We find, through mainly numerical investigations, that the extended model, including the defect dynamics, provides considerable self-consistent information on the nature of the defects near the glass transition. In particular, we find that the bare value of γ_v is significantly restricted when the observed stretching occurs. In fact, the long time scale of the defect variable corresponds to the case where the system arranges itself such that γ_v is close to some small temperature-dependent value γ_v^c. The system seems to pick out its own defect potential and diffusion coefficient. We find that, when $\gamma_v \simeq \gamma_v^c$, the time scale associated with the defects becomes much longer than that of the density variable, and thus the basic picture obtained using the previous simplifying assumption still holds: The parameters describing the metastable wells and the coupling arrange themselves such that the coupling enables the slow dynamics of the defects to effect the density dynamics without destroying the metastable wells.

We note that the approach we take in this paper is not to explain how the defects bring the system to the observed slowing down starting from the microscopic description of the defects. Instead, we assume that the system arranges itself to be on the critical surface associated with the MCT. We then investigate the conditions that the defect degrees of freedom must satisfy to be on this critical surface.

The cutoff mechanism of Das and Mazenko [13] will eventually influence the very long time dynamics by generating an exponential decay. We note that there exists a mechanism [17,11] that drives the cutoff effect to a small value, so that the observed slowing down is retained. Although in principle we can include this effect in the coupled equations, we consider here the situation where this is neglected. Another simplification made in this analysis is to neglect the wave-number dependences of correlation functions. In terms of the FNH description, one can construct a wave-number dependent model, as shown in Ref. [18]. Since this model is difficult to analyze numerically, we focus here on the wave-number independent model.

In Sec. II we present a detailed formulation of the FNH description of simple fluids, including the defect variable. We then use the well-known field-theoretic techniques to calculate the relevant nonlinear contributions to the glass transition. In Sec. III, the case where the defect autocorrelation is a constant is considered in detail as a first approximation. This will reproduce the results of Ref. [14]. The analysis of the full model, which includes the defect dynamics, is presented in Sec. IV. In Sec. V, we discuss the cutoff mechanism and the temperature dependence of the viscosity in this formulation.

II. FNH WITH THE DEFECT DENSITY VARIABLE

In this section, we formulate the FNH description of compressible fluids in detail, including the defect density n as the additional slow variable. Using well-developed field-theoretic methods, we calculate nonlinear corrections to the density and defect autocorrelation functions systematically.

A. Generalized Langevin equations

Our starting point is the generalized Langevin equation for the set of slow variables $\psi_\alpha=\rho(\mathbf{x}), g_i(\mathbf{x}), n(\mathbf{x})$, where ρ is the mass density and $\mathbf{g}$ is the momentum density. Here α labels the type of the field, the position $\mathbf{x}$, and the vector label i. Following the standard procedure described by Ma and Mazenko [19], we have the equation of motion given by

$$\frac{\partial\psi_\alpha}{\partial t}=\bar{V}_\alpha[\psi]-\sum_\beta \Gamma_{\alpha\beta}\frac{\delta F}{\delta\psi_\beta}+\Theta_\alpha \ , \tag{2.1}$$

where F is the effective Hamiltonian, $\bar{V}_\alpha$ is the streaming velocity governing the reversible dynamics given by

$$\bar{V}_\alpha[\psi]=\sum_\beta \{\psi_\alpha,\psi_\beta\}\frac{\delta F}{\delta\psi_\beta} \ , \tag{2.2}$$

and $\{\psi_\alpha,\psi_\beta\}$ is the Poisson bracket among the slow variables. The dissipative matrix $\Gamma_{\alpha\beta}$ and the Gaussian noise Θ_α satisfy

$$\langle\Theta_\alpha(t)\Theta_\beta(t')\rangle=2k_BT\Gamma_{\alpha\beta}\delta(t-t') \ . \tag{2.3}$$

The effective Hamiltonian for ψ_α is given by

$$F=F_K+F_u[\delta\rho]+F_v[\delta\rho,n] \ , \tag{2.4}$$

where F_K is the kinetic energy:

$$F_K=\int d^3\mathbf{x}\frac{\mathbf{g}^2(\mathbf{x})}{2\rho(\mathbf{x})} \ . \tag{2.5}$$

$F_u[\delta\rho]$ is the potential energy for the density fluctuation; $\delta\rho=\rho-\rho_0$, where ρ_0 is the average density; and $F_v[\delta\rho,n]$ governs the defect density and its coupling to ρ. In general, $F_u[\delta\rho]$ can be any local functional of $\delta\rho$ and the spatial derivatives of $\delta\rho$. In particular, one can study the wave-number dependence of the structure factor by including the spatial derivatives [18]. Recently it has been claimed [20] that by including the wave-number dependences one can effectively generate the von-Schweidler relaxation and the stretching. Analytical treatment in this case, however, is very difficult. In the present case, we consider the simple quadratic form that corresponds to a wave-number independent structure factor,

$$F_u[\delta\rho]=\int d^3\mathbf{x}\frac{A}{2}[\delta\rho(\mathbf{x})]^2 \ , \tag{2.6}$$

where A is the flat inverse susceptibility and wave numbers are restricted to values less than a cutoff Λ. $F_v[\delta\rho,n]$ in Eq. (2.4) is assumed to be of the form

$$F_v[\delta\rho,n]=\int d^3\mathbf{x}[B\delta\rho(\mathbf{x})n(\mathbf{x})+h(n(\mathbf{x}))] \ , \tag{2.7}$$

where we introduced the simple coupling term through the coupling constant B. As discussed in Sec. I, we construct the potential energy $h(n)$ to be a double-well potential [21], with the metastable defect density $\bar{n}$ associated with the higher well We parameterize $h(n)$ such that $h(n)$ has three extrema at $n=0$, $(1-\sigma)\bar{n}$, and $\bar{n}$: $h'(n)=\epsilon n[n-(1-\sigma)\bar{n}](n-\bar{n})$, or upon integration,

$$h(n)=\epsilon\bar{n}^4\left[\frac{1}{4}\left[\frac{n}{\bar{n}}\right]^4-\frac{1}{3}(2-\sigma)\left[\frac{n}{\bar{n}}\right]^3+\frac{1}{2}(1-\sigma)\left[\frac{n}{\bar{n}}\right]^2\right] \ . \tag{2.8}$$

The parameter ϵ gives the correct dimension for $h(n)$ and describes the overall scale of the potential energy. For small positive σ, Eq. (2.8) represents a double-well potential with the global minimum at $n=0$ and the metastable minimum at $\bar{n}$. We note that for small negative σ, $h(n)$ in the above representation is also a double-well potential. In fact, $h(n)$ is invariant under $\sigma\to\sigma'\equiv-\sigma/(1-\sigma)$ and $\bar{n}\to\bar{n}'\equiv(1-\sigma)\bar{n}$. Thus, in the following analysis, only the absolute value of σ will play a role. We also note that, if $\sigma=0$, then $h(n)$ develops an inflection point at $n=\bar{n}$. It is useful to define two dimensionless parameters x and y that characterize the scale of the coupling energy and the potential energy, respectively:

$$x\equiv\frac{B\rho_0\bar{n}}{A\rho_0^2} \ , \quad y\equiv\frac{\epsilon\bar{n}^4}{A\rho_0^2} \ . \tag{2.9}$$

The potential and coupling energies are then completely described by x, y, σ, and $\bar{n}$. We consider the coupling energy term in Eq. (2.7) as a small distortion of the shape of $h(n)$. In particular, $\bar{n}$ is shifted to $n^*[\delta\rho]$, determined by

$$0=\left.\frac{\delta F_v}{\delta n}\right|_{n=n^*[\delta\rho]}=B\delta\rho+h'(n^*[\delta\rho]) \ . \tag{2.10}$$

If we expand in powers of $\delta\rho$,

$$n^*[\delta\rho]=\bar{n}\left[1+a_1\left[\frac{\delta\rho}{\rho_0}\right]+a_2\left[\frac{\delta\rho}{\rho_0}\right]^2+\cdots\right] \ , \tag{2.11}$$

and we can easily calculate the coefficients $a_1,a_2\ldots$ using Eq. (2.10),

$$a_1=-\frac{x}{\sigma y} \ , \quad a_2=-\left[\frac{1+\sigma}{\sigma}\right]\left[\frac{x}{\sigma y}\right]^2,\ldots . \tag{2.12}$$

We will later consider the fluctuation of n around this shifted $n^*[\delta\rho]$: $\delta n=n-n^*[\delta\rho]$. Equation (2.11) indicates that, for small $|\sigma|$, the coupling energy x must be sufficiently small such that

$$|a_1|\ll 1 \ , \quad |a_2|\ll 1 \tag{2.13}$$

in order to have a sensible expansion.

Turning to the reversible streaming velocity terms in the Langevin equations, we assume that the Poisson brackets involving ρ and $\mathbf{g}$ are evaluated in the usual way, as in Ref. [13]. The new variable n is a scalar quantity as is ρ; thus we assume that the Poisson brackets for n have the same structure as those for ρ. The only nonvanishing elements involving n are

$$\{g^i(\mathbf{x}),n(\mathbf{x}')\}=\nabla^i_{\mathbf{x}'}[\delta(\mathbf{x}-\mathbf{x}')n(\mathbf{x})] \ . \tag{2.14}$$

Since there are no Poisson brackets relating ρ to n and

$\Gamma_{\rho\beta}=0$, the Langevin equation for ρ is simply the continuity equation

$$\frac{\partial\rho}{\partial t}=-\nabla\cdot\mathbf{g} \ . \tag{2.15}$$

The Langevin equations for the remaining variables can readily be found. Keeping up to quadratic terms in the fluctuations, $\delta n=n-n^*[\delta\rho]$ and $\delta\rho$, we have

$$\frac{\partial g^i}{\partial t}=-\sum_j \nabla_j(\sigma^u_{ij}+\sigma^v_{ij})-\sum_j \nabla_j\left[\frac{g^i g^j}{\rho}\right] - \sum_j L_{ij}\left[\frac{g^j}{\rho}\right]+\Theta_i \ , \tag{2.16}$$

and

$$\frac{\partial}{\partial t}(\delta n)=-\bar{n}\left[1+\frac{x}{\sigma y}\right]\nabla\cdot\left[\frac{\mathbf{g}}{\rho}\right]-\nabla\cdot\left[(\delta n)\frac{\mathbf{g}}{\rho}\right] +\bar{n}\tilde{\Gamma}_v\nabla^2\left[\sigma y\left[\frac{\delta n}{\bar{n}}\right]-2x\left[\frac{1+\sigma}{\sigma}\right]\left[\frac{\delta\rho}{\rho_0}\right]\left[\frac{\delta n}{\bar{n}}\right] +y(1+\sigma)\left[\frac{\delta n}{\bar{n}}\right]^2\right]+\Xi \ , \tag{2.17}$$

where

$$\sigma^u_{ij}\equiv\delta_{ij}\{c_0^2(\delta\rho)+\tfrac{1}{2}A(\delta\rho)^2\} \tag{2.18}$$

is the usual stress tensor with $c_0^2=A\rho_0$ and

$$\sigma^v_{ij}\equiv\delta_{ij}(A\rho_0^2)\left[(x+\sigma y)\left[\frac{\delta n}{\bar{n}}\right] -2x\left[\frac{1+\sigma}{\sigma}\right]\left[\frac{\delta\rho}{\rho_0}\right]\left[\frac{\delta n}{\bar{n}}\right] +y\left[1+\frac{3\sigma}{2}\right]\left[\frac{\delta n}{\bar{n}}\right]^2\right] . \tag{2.19}$$

In Eqs. (2.16) and (2.17) the dissipative coefficients and the noises satisfy

$$\langle\Theta_i(\mathbf{x},t)\Theta_j(\mathbf{x}',t')\rangle=2k_BTL_{ij}(\mathbf{x})\delta(\mathbf{x}-\mathbf{x}')\delta(t-t') \ , \tag{2.20}$$

$$\langle\Xi(\mathbf{x},t)\Xi(\mathbf{x}',t')\rangle=-2k_BT\Gamma_v\nabla_x^2\delta(\mathbf{x}-\mathbf{x}')\delta(t-t') \ , \tag{2.21}$$

where $L_{ij}(\mathbf{x})=-\eta_0(\frac{1}{3}\nabla_i\nabla_j+\delta_{ij}\nabla^2)-\zeta_0\nabla_i\nabla_j$ with the bare shear and bulk viscosities η_0 and ζ_0, respectively, and $\tilde{\Gamma}_v\equiv(A\rho_0^2/\bar{n}^2)\Gamma_v$. For later use, we define the bare longitudinal viscosity, $\Gamma_0\equiv\zeta_0+\frac{4}{3}\eta_0$.

The Langevin equations [Eqs. (2.15), (2.16), and (2.17)] can be put into a field-theoretical form following standard Martin-Siggia-Rose procedures [22,23]. It essentially amounts to introducing the hatted variable $\hat{\psi}_a$ for each field ψ_α to enforce the equations of motion and integrating out the Gaussian noises to get the quadratic form in $\hat{\psi}_\alpha$. We introduce the local velocity field $\mathbf{V}$, where $\mathbf{g}=\rho\mathbf{V}$, to eliminate the $1/\rho$ nonlinearity in Eqs. (2.16) and (2.17) [24]. In this formalism, one can treat the nonlinearities stemming from the quadratic terms in the equations of motion using standard perturbation theory expansions. The nonlinear corrections to the zeroth-order propagator $G^0_{\alpha\beta}$ between ψ_α and ψ_β are expressed through Dyson's equation,

$$G^{-1}_{\alpha\beta}=[G^0_{\alpha\beta}]^{-1}-\Sigma_{\alpha\beta} \ , \tag{2.22}$$

where $\Sigma_{\alpha\beta}$ is the self-energy.

From the linear terms in the equations of motion, we can easily read off the elements of the zeroth-order inverse propagator $[G^0_{\alpha\beta}]^{-1}$. By inverting this matrix, we get various correlation and linear response functions. We will be concerned only with the longitudinal parts of those functions and will hereafter use the notation ρ,n instead of $\delta\rho,\delta n$, respectively, for convenience. One obtains a physical interesting situation if

$$\left|\frac{(x+\sigma y)^2}{\sigma y}\right|=|\sigma y(1-a_1)^2|\ll 1 \ . \tag{2.23}$$

Then we have

$$G^0_{n\hat{n}}(\mathbf{q},\omega)\simeq\frac{1}{\omega+i\gamma_v} \tag{2.24}$$

and

$$G^0_{\rho\hat{\rho}}(\mathbf{q},\omega)\simeq\frac{\rho_0\omega+i\Gamma_0q^2}{D_0(\mathbf{q},\omega)} \ , \tag{2.25}$$

where $\gamma_v\equiv\sigma y\tilde{\Gamma}_vq^2$, and $D_0(\mathbf{q},\omega)=\rho_0(\omega^2-q^2c_0^2)+i\omega q^2\Gamma_0$. The assumption in Eq. (2.23) is equivalent to saying that we are considering the small $|\sigma|$ limit, which will be justified self-consistently later when we consider the conditions for the slowing down. Equation (2.25) represents a standard form for the density autocorrelation function. The defect autocorrelation function in Eq. (2.24) has a very slow diffusive mode, $\omega=-i\gamma_v$, since it is assumed that $\gamma_v\ll 1$. This signals a separation of time scales between the density and the defect variables, as noted in Sec. I.

We can achieve a further simplification by looking at the following situation. As one approaches the glass transition, we expect that, since the viscosity is getting extremely large, one eventually reaches a point where $q^2\Gamma_0/\rho_0\gg\omega$. It then follows that

$$G^0_{nn}(\mathbf{q},\omega)\simeq\frac{1}{\sigma y}\left[\frac{\bar{n}}{\rho_0}\right]^2\frac{2\beta^{-1}}{A}\frac{\gamma_v}{\omega^2+\gamma_v^2} \ , \tag{2.26}$$

$$G^0_{\rho\rho}(\mathbf{q},\omega)\simeq\frac{2\beta^{-1}}{A}\frac{\gamma_u}{\omega^2+\gamma_u^2} \ , \tag{2.27}$$

$$G^0_{\rho n}(\mathbf{q},\omega)\simeq\frac{\bar{n}}{\rho_0}\left[\frac{x+\sigma y}{\sigma y}\right]\frac{2\beta^{-1}}{A}\left[-\frac{\gamma_v}{\omega^2+\gamma_v^2}+\frac{\gamma_u}{\omega^2+\gamma_u^2}\right] \simeq-\frac{\rho_0}{\bar{n}}(x+\sigma y)G^0_{nn}(\mathbf{q},\omega) +\frac{\bar{n}}{\rho_0}\left[\frac{x+\sigma y}{\sigma y}\right]G^0_{\rho\rho}(\mathbf{q},\omega) \ , \tag{2.28}$$

where $\gamma_u = A\rho_0^2/\Gamma_0$. Therefore, in this approximation, we have the density-defect correlation function as a linear combination of the density and the defect autocorrelation functions.

Nonlinear corrections to the zeroth-order response and correlation functions are represented in terms of the self-energies through Eq. (2.22). For the case where the separation of time scales observed in the zeroth-order correlation functions remains valid at high orders, one obtains the following renormalized expressions:

$$\psi(\mathbf{q},\omega) \equiv G_{n\hat{n}}(\mathbf{q},\omega) \simeq \frac{1}{\omega + i\gamma_v'(\mathbf{q},\omega)} , \tag{2.29}$$

$$\phi(\mathbf{q},\omega) \equiv G_{\rho\hat{\rho}}(\mathbf{q},\omega) \simeq \frac{\rho(\mathbf{q},\omega)\omega + i\Gamma(\mathbf{q},\omega)q^2}{\rho(\mathbf{q},\omega)[\omega^2 - q^2c^2(\mathbf{q},\omega)] + i\Gamma(\mathbf{q},\omega)\omega q^2} , \tag{2.30}$$

where the renormalizations of parameters are given by

$$\rho(\mathbf{q},\omega) = \rho_0 - i\Sigma^L_{\hat{P}V}(\mathbf{q},\omega) , \tag{2.31a}$$

$$qc^2(\mathbf{q},\omega) = qc_0^2 + \Sigma_{\hat{g}\rho}(\mathbf{q},\omega) , \tag{2.31b}$$

$$q^2\Gamma(\mathbf{q},\omega) = q^2\Gamma_0 + i\Sigma^L_{\hat{g}V}(\mathbf{q},\omega) , \tag{2.31c}$$

$$\gamma_v'(\mathbf{q},\omega) = \gamma_v + i\Sigma_{\hat{n}n}(\mathbf{q},\omega) . \tag{2.31d}$$

Equations (2.29) and (2.30) are the fundamental equations for the dynamics of density and defect fluctuations without the cutoff effect. In the following, we calculate explicitly nonlinear contributions to the renormalized viscosity $\Gamma(\mathbf{q},\omega)$ and the renormalized diffusion coefficient $\gamma_v'(\mathbf{q},\omega)$, which will be reexpressed in terms of $G_{n\hat{n}}(\mathbf{q},\omega)$ and $G_{\rho\hat{\rho}}(\mathbf{q},\omega)$.

B. One-loop evaluation of $\Gamma(\mathbf{q},\omega)$ and $\gamma_v'(\mathbf{q},\omega)$

The density feedback mechanics of the MCT is realized by calculating the nonlinear corrections to the bare viscosity in the density correlation function of the form, Eq. (2.30). In the present case, however, we have two coupled equations, (2.29) and (2.30). Thus we must find both kinetic coefficients, $\Gamma(\mathbf{q},\omega)$ and $\gamma_v'(\mathbf{q},\omega)$, to complete the equations. In this section, we calculate one-loop nonlinear contributions to those quantities.

As noted in Ref. [13], the renormalized longitudinal viscosity in Eq. (2.31c) can also be represented in the hydrodynamic limit as

$$\Gamma(\mathbf{q},\omega) = \Gamma_0 - \frac{\beta}{2q^2}\Sigma^L_{\hat{g}g}(\mathbf{q},\omega) . \tag{2.32}$$

The relevant one-loop diagrams contributing to this self-energy are listed in Fig. 1. As discussed in Ref. [13], the diagrams coming from the convective nonlinearities (ρVV terms in the equations of motion) just renormalize the bare viscosity and are irrelevant to the density feedback mechanism.

We note that the approximation given by Eq. (2.28) generates two kinds of terms that are proportional to $G_{nn}G_{\rho\rho}$ and $G_{\rho\rho}G_{\rho\rho}$, respectively, in the expression for $\Gamma(\mathbf{q},\omega)$. In fact, one can easily see that the $G_{nn}G_{\rho\rho}$ type terms come from diagrams 1(b)–1(f) and the $G_{\rho\rho}G_{\rho\rho}$ type terms from diagrams 1(a)–1(d). Thus we have

$$\Gamma(\mathbf{q},\omega) = \Gamma_0 + \int_0^\infty dt\, e^{i\omega t} \int \frac{d^3\mathbf{k}}{(2\pi)^3} \{[V^{(1)}(\mathbf{q},\mathbf{k})G_{nn}(\mathbf{k},t) + V^{(2)}(\mathbf{q},\mathbf{k})G_{\rho\rho}(\mathbf{k},t)]G_{\rho\rho}(\mathbf{q}-\mathbf{k},t)\} , \tag{2.33}$$

where $V^{(1)}$ and $V^{(2)}$ are appropriate vertices to be evaluated. In principle, the wave-number dependence of the vertices can be considered [18], for example, by using the spatial derivatives of the density fluctuations in the effective Hamiltonian, Eq. (2.6). This involves, however, very complicated wave-number integrals. In this analysis, we consider the wave-number independent case. A closely related approximation to this is that the correlation functions can be factorized into wave-number and time-dependent parts [25]. We assume here that $G_{nn}(\mathbf{q},t) = T(q)\psi(t)$, $G_{\rho\rho}(\mathbf{q},t) = S(q)\phi(t)$, $T(q)$ and $S(q)$ are the flat structure factors given by

$$T(q) = \left[\frac{\bar{n}}{\rho_0}\right]^2 \frac{1}{\sigma y}\frac{\beta^{-1}}{A} , \quad S(q) = \frac{\beta^{-1}}{A} , \tag{2.34}$$

for $q < \Lambda$ and $T(q) = S(q) = 0$ for $q > \Lambda$, where Λ is the large momentum cutoff. Integrating over wave numbers for the one-loop diagrams in Fig. 1 with the help of Λ, we obtain

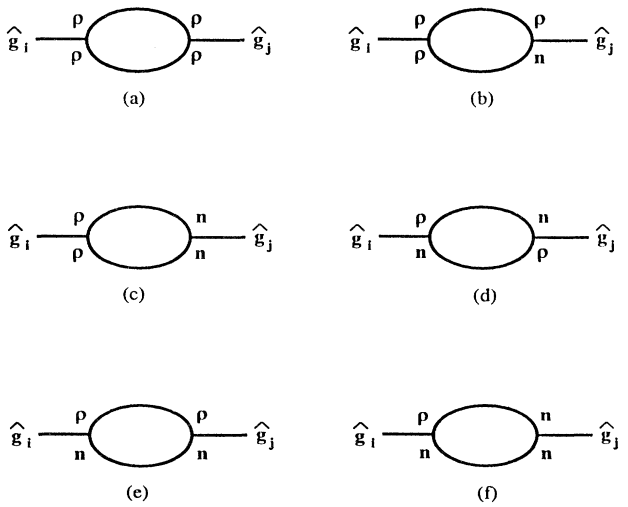

FIG. 1. One-loop diagrams contributing to $\Sigma_{\hat{g}\hat{g}}(\mathbf{q},\omega)$.

$$\Gamma(\omega)=\Gamma_0+k_BT\frac{\Lambda^3}{6\pi^2}\int_0^\infty dt\, e^{i\omega t}[d_1\psi(t)\phi(t)+d_2\phi^2(t)] , \tag{2.35}$$

where the coefficients d_1 and d_2 are functions of x, y, and σ. We find, through detailed calculation of the diagrams, that

$$d_1(x,y,\sigma)=-4x\left[\frac{1+\sigma}{\sigma}\right]^2\left[2+\frac{x}{\sigma y}\right] -4\left[y(2+3\sigma)+2x^2\left[\frac{1+\sigma}{\sigma}\right]^2\right]\left[1+\frac{x}{\sigma y}\right]^2 , \tag{2.36a}$$

$$d_2(x,y,\sigma)=1-4x\left[\frac{1+\sigma}{\sigma}\right]\left[1+\frac{x}{\sigma y}\right] +2\left[y(2+3\sigma)+2x^2\left[\frac{1+\sigma}{\sigma}\right]^2\right]\left[1+\frac{x}{\sigma y}\right]^2 . \tag{2.36b}$$

For later use, we define a dimensionless parameter containing an explicit factor of the temperature,

$$\xi\equiv\frac{k_BT}{A\rho_0^2}\frac{\Lambda^3}{6\pi^2} , \tag{2.37}$$

and $c_i\equiv\xi d_i$, $i=1,2$.

The renormalization of γ_v is given by Eq. (2.31d). As in Eq. (2.32), it is equivalent in the hydrodynamic limit to

$$\gamma_v'(\mathbf{q},\omega)=\gamma_v-\tfrac{1}{2}\sigma y\left[\frac{\rho_0}{\bar{n}}\right]^2\frac{A}{\beta^{-1}}\Sigma_{\hat{n}\hat{n}}(\mathbf{q},\omega) . \tag{2.38}$$

The one-loop diagrams that contribute to $\Sigma_{\hat{n}\hat{n}}(\mathbf{q},\omega)$ and do not have the explicit Γ_v factor, which is assumed to be very small, are listed in Fig. 2. In the small $|\sigma|$ limit, the major contribution to the self-energy comes from diagram (b). Assuming there is no coupling between the density feedback mechanism and the transverse viscosity, which is the case for the flat structure factor case, we finally obtain that

$$\gamma_v'(\mathbf{q},\omega)=\gamma_v-\left[\frac{q}{\Lambda}\right]^2\xi\int_0^\infty dt\, e^{i\omega t}\psi(t)\ddot{\phi}(t) . \tag{2.39}$$

Equations (2.29) and (2.30), together with Eqs. (2.35) and (2.39), complete the specification of our model.

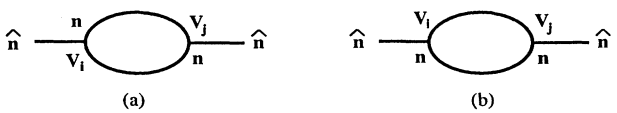

FIG. 2. One-loop diagrams contributing to $\Sigma_{\hat{n}\hat{n}}(\mathbf{q},\omega)$.

III. MODEL WITH A CONSTANT DEFECT AUTOCORRELATION FUNCTION

Metastability plays an important role in this model, since defects spend a considerable amount of time trapped within a well. During this period, we may assume that the defect autocorrelation function can be regarded as a constant,

$$G_{nn}(\mathbf{q},\omega)=2\pi\left[\frac{\bar{n}}{\rho_0}\right]^2\frac{1}{\sigma y}\frac{\beta^{-1}}{A}\delta(\omega) , \quad \text{or } \psi(t)=1 . \tag{3.1}$$

This approximation corresponds to the limit where one takes the bare value of γ_v to zero. In this case, we note that the nonlinear correction to γ_v also vanishes in the long time limit, $\omega\to 0$, since the integrand in Eq. (2.39) is a total derivative when $\psi(t)=1$. Thus the self-consistency of the approximation is maintained.

The coupled equations (2.29) and (2.30) then reduce to a form dependent only on $\phi(t)$, and we can make contact with the standard treatment of the MCT [1]. It is important to note that the MCT coefficients c_1 and c_2 are expressed in terms of the temperature and the parameters describing the metastable potential and the coupling between the density and the defect variables. We now assume that the system organizes itself to be on the critical surface of the MCT, which is described below. This will give relations among the metastability parameters.

The density feedback mechanism associated with the representation of the type Eqs. (2.30) and (2.33) was first studied by Götze [8]. According to Ref. [8], there exists a critical line in the (c_1,c_2) space separating ergodic and nonergodic regions, where the nonergodic phase is characterized by the existence of the limit $f=\lim_{t\to\infty}\phi(t)$, $f>0$. To study the critical condition in terms of the parameters x, y, σ, and ξ, we follow a more general discussion of this model given by Kim and Mazenko [11]. According to Ref. [11], the glass transition can be described by the following three parameters:

$$\sigma_0=(1-f)V(f) , \tag{3.2}$$

$$\sigma_1=(1-f)^2V'(f) , \tag{3.3}$$

$$\lambda=\tfrac{1}{2}(1-f)^3H''(f) , \tag{3.4}$$

where $V(f)=H(f)-f/(1-f)$ and $H(f)=\sum_{i=1}^N c_i f^i$ for a general model containing higher-order terms in the mode coupling integral. In Eq. (3.4), the parameter λ is directly related to the exponents of the sequence of time relaxations [8]:

$$\frac{\Gamma^2(1-a)}{\Gamma(1-2a)}=\lambda=\frac{\Gamma^2(1+b)}{\Gamma(1+2b)} . \tag{3.5}$$

The ideal glass transition is approached when both σ_0 and σ_1 are getting small. Let us consider the situation where $\sigma_1=0$ and the transition is approached by taking $\sigma_0\to 0$. Solving Eqs. (3.2)–(3.4) for c_1 and c_2, we have

$$c_1=\frac{2\lambda-1}{\lambda^2}+\frac{4\sigma_0}{\lambda(1-\lambda)}+O(\sigma_0^2) , \tag{3.6a}$$

$$c_2=\frac{1}{\lambda^2}-\frac{3\sigma_0}{\lambda(1-\lambda)^2}+O(\sigma_0^2)\ . \tag{3.6b}$$

In terms of the parameters x, y, σ, and ξ, the critical condition is given by setting $\sigma_0=0$:

$$d_1(x,y,\sigma)=\frac{2\lambda-1}{\xi\lambda^2}\ , \tag{3.7a}$$

$$d_2(x,y,\sigma)=\frac{1}{\xi\lambda^2}\ ,\quad \tfrac{1}{2}<\lambda<1\ . \tag{3.7b}$$

We note that we are mainly concerned with the region where $|x|\ll 1$ in order to be consistent with the fact that we considered the coupling term as a small perturbation shifting the metastable state as in Eqs. (2.11) and (2.13). We also require the solution be consistent with the assumption, Eq. (2.23). We find that nontrivial solutions to Eq. (3.7) satisfying Eqs. (2.13) and (2.23) exist only if we take $\sigma\to 0$ simultaneously with $x\to 0$, while holding

$$\frac{x}{\sigma^2}\to C\ , \tag{3.8}$$

for some constant C. Let us briefly discuss the physical meaning of Eq. (3.8). The small $|\sigma|$ limit is a sufficient condition for a separation of the time scales between the defect and the density variables, as seen from Eq. (2.23). This indicates that the metastable wells become very shallow. If the coupling energy represented by $|x|$ is stronger than the above limit, i.e., $|x|\sim|\sigma|$, then the metastable state given by Eq. (2.11) does not exist, since the condition Eq. (2.13) is violated. Thus, a strong coupling energy destroys the metastability of defects. On the other hand, if $|x|\sim|\sigma|^3$, we have from Eq. (2.36) $d_1=-8y$, $d_2=1+4y$. Since y is always positive, $d_1<0$, therefore the system never reaches the critical surface given by Eq. (3.7). Thus, if the coupling is weaker than the limit given by Eq. (3.8), the slow dynamics of defects cannot affect the density dynamics so that the density variable does not slow down. Equation (3.8) gives the correct relation between the coupling energy and the barrier size of the defects in the case of observed slowing down. In this limit, Eq. (2.36) reduces to

$$d_1(x,y,\sigma)=-8(y+C)-4\left[\frac{C^2}{y}+3y+8C\right]\sigma+O(\sigma^2)\ , \tag{3.9a}$$

$$d_2(x,y,\sigma)=1+4y+2(3y+2C)\sigma+O(\sigma^2)\ . \tag{3.9b}$$

Thus, from Eqs. (3.7) and (3.9), we have the critical condition given in terms of the metastability parameters by

$$C=\frac{1}{4}\left[1-\frac{2\lambda+1}{2\xi\lambda^2}\right]\ ,\quad y=\frac{1}{4}\left[\frac{1}{\xi\lambda^2}-1\right]\ . \tag{3.10}$$

Furthermore, from Eqs. (3.6) and (3.9), we can easily see that the limit $\sigma\to 0$ can be identified with $\sigma_0\to 0$ if

$$3\left[\frac{C^2}{y}+3y+8C\right]=2(1-\lambda)(3y+2C)\ . \tag{3.11}$$

Therefore, as indicated in Sec. I, we have the situation where the stretched dynamics is associated with the weak coupling ($x\to 0$) and low barrier ($\sigma\to 0$) limit, which is consistent with the separation of the time scales between the density and the defect variables, as can be seen from Eq. (2.23). The condition Eq. (3.11), when we use Eq. (3.10), can be interpreted as a relation between the exponent parameter λ and the temperature represented by the parameter ξ,

$$\xi=\frac{1}{\lambda^2}\left[1-\frac{3(2\lambda-1)}{2[7+2\lambda+\sqrt{4\lambda^2+22\lambda+91}]}\right]\ . \tag{3.12}$$

Thus the exponents a and b are given as smooth functions of temperature (see Fig. 3). The temperature dependence of the parameters y and C that describe the potential $h(n)$ and its coupling to ρ is given by Eq. (3.10). All the temperature dependences certainly do not give any indication of a special temperature. In the conventional MCT [7,9], the control parameter σ_0 is assumed to have a temperature dependence as $\sigma_0\sim T_0-T$, which is the origin of the sharp temperature dependences. In this case, however, σ_0 is proportional to σ with the weakly temperature-dependent coefficient $f(\xi)$,

$$\sigma_0=f(\xi)\sigma\equiv-\tfrac{2}{3}[3y+2C]\xi\lambda(1-\lambda)^2\sigma\ . \tag{3.13}$$

We note that $f(\xi)$ vanishes at the lower and upper bounds of ξ that correspond to $\lambda=1$ and $\frac{1}{2}$, respectively. Therefore, the above analysis is not applicable to the region near the two end points of ξ. Our basic picture of the observed slowing down of the dynamics is that the parameters describing the metastable wells and the coupling, while displaying a smooth temperature dependence, achieve the critical limit given by Eq. (3.8). In this model, the transition is actually controlled by the parameter σ, which is the size of barrier in the metastable potential.

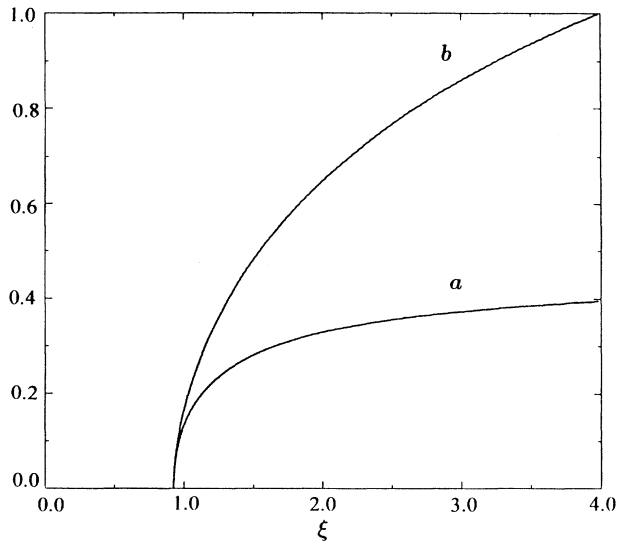

FIG. 3. Exponents a and b as functions of temperature represented by ξ.

IV. MODEL INCLUDING THE DYNAMICS OF DEFECTS

The approximation, Eq. (3.1), made in the preceding section, enabled us to treat the model analytically and to obtain the critical condition between the shallow metastable wells and the weak coupling. However, when nonlinear corrections to the bare diffusion coefficient come into play, the constraint Eq. (3.1) must be relaxed and the dynamics of defects should be taken into account. In this section, we analyze the full coupled equations, (2.29) and (2.30), with (2.35) and (2.39). Since the nonlinear couplings in these equations are highly nontrivial, an analytical treatment is very difficult. In the time domain, these equations are coupled integro-differential equations:

$$\ddot{\phi}(t)+d_0\dot{\phi}(t)+\Omega_0^2\phi(t)+\Omega_0^2\int_0^t ds\, H(t-s)\dot{\phi}(s)=0\ , \quad (4.1)$$

$$\dot{\psi}(t)+\gamma_v\psi(t)+\int_0^t ds\, G(t-s)\psi(s)=0\ , \quad (4.2)$$

where

$$H(t)=c_1\psi(t)\phi(t)+c_2\phi^2(t)\ , \quad G(t)=-\tilde{\xi}\ddot{\phi}(t)\psi(t)\ , \quad (4.3)$$

with the initial conditions $\phi(0)=\psi(0)=1$, $\dot{\phi}(0)=0$, and the definition $\tilde{\xi}\equiv(q/\Lambda)^2\xi$. In the following, we integrate them numerically. We fix $d_0=\Omega_0^2=1$ and study the relaxation of the system at fixed wave number $q/\Lambda=0.1$ as γ_v and the MCT coefficients c_1,c_2 change. We use the parametrization, Eq. (3.6), for c_1,c_2 and the value of ξ determined from Eq. (3.12).

We find that the system depends in a crucial way on the value of γ_v. When the value of γ_v is large enough, we expect that the defects quickly diffuse away and only the density fluctuations are important. It is clearly seen in the numerical integration of Eqs. (4.1) and (4.2) for fixed c_1,c_2 and ξ in Fig. 4 The defect autocorrelation function $\psi(t)$ decays exponentially for large γ_v. Thus, in the large γ_v limit, the model essentially reduces to the one originally considered by Leutheusser [7], where only the c_2 term is present. For smaller values of γ_v, we expect that the system goes into the regime where the result of the preceding section applies such that the slow dynamics of defects and the coupling between defects and density fluctuations play an important role in stretched dynamics. We can see from Figs. 4 and 5 that, as the value of γ_v is decreased, $\psi(t)$ and $\phi(t)$ are more and more stretched. We cannot, however, take γ_v to zero, since for small enough γ_v such that $\gamma_v<\gamma_v^c$ for some γ_v^c, $\psi(t)$ starts to increase with time and consequently the model becomes unphysical. This surprising result restricts the value of the parameter γ_v to $\gamma_v>\gamma_v^c$. It is clear that $\psi(t)$ and $\phi(t)$ are stretched most when γ_v is very close to but still larger than γ_v^c. In fact, the time scale for $\psi(t)$ become extremely large as $\gamma_v\to\gamma_v^c$ so that it seems to approach some plateau value g as $t\to\infty$. We will discuss later that only when $\gamma_v\simeq\gamma_v^c$ is the time scale of $\psi(t)$ much greater than that of $\phi(t)$, $\psi(t)$ more stretched than $\phi(t)$, and the picture consistent with the basic picture obtained in the preceding section.

It is well known that the later stage of relaxation of $\phi(t)$ is well fit by the stretched exponential. As $\gamma_v\to\gamma_v^c$, we find that the later stage of the relaxation of $\psi(t)$ is also well fit by a stretched exponential. We now use two stretched exponential forms,

$$\phi(t)=f\exp[-(t/\tau)^\beta]\ , \quad \psi(t)=g\exp[-(t/\tau')^{\beta'}] \quad (4.4)$$

in Eqs. (4.1) and (4.2). Since, for γ_v near γ_v^c, the time scale τ' for $\psi(t)$ is much greater than τ for $\phi(t)$, we can write

$$\exp[-(t/\tau')^{\beta'}]\simeq 1-\left[\frac{\tau}{\tau'}\right]^{\beta'}\left[\frac{t}{\tau}\right]^{\beta'}\ . \quad (4.5)$$

We find the following qualitative relations among these parameters from Eqs. (4.1) and (4.2):

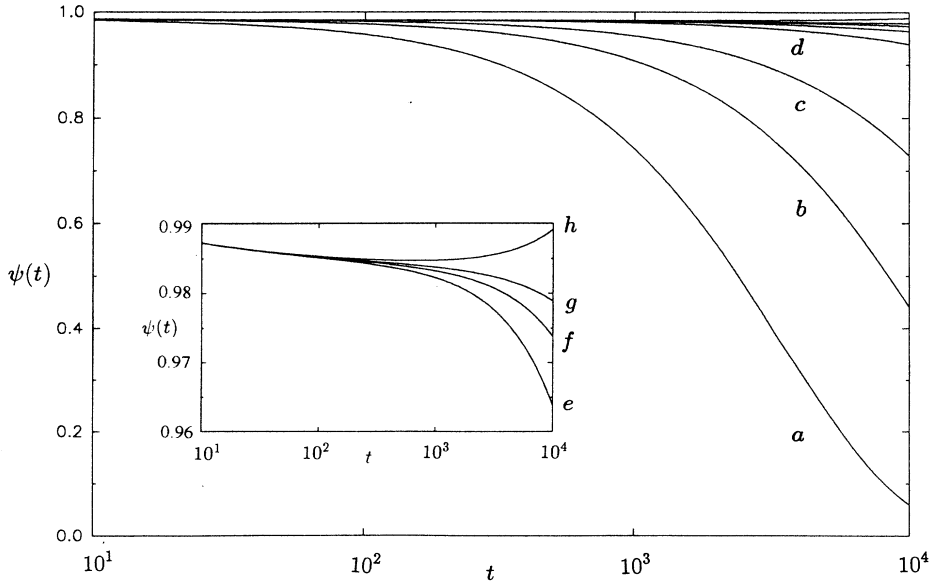

FIG. 4. Defect autocorrelation function $\psi(t)$ for fixed $(c_1,c_2)=(0.556,2.778)$ and for various γ_v: curve a, 2.0×10^{-3} curve b, 1.8×10^{-3}, curve c, 1.75×10^{-3}, curve d, 1.725×10^{-3}, curve e, 1.7225×10^{-3}, curve f, 1.7215×10^{-3}, curve g, 1.721×10^{-3}, and curve h, 1.72×10^{-3}. We can estimate the value of γ_v^c as $1.72\times10^{-3}<\gamma_v^c<1.721\times10^{-3}$.

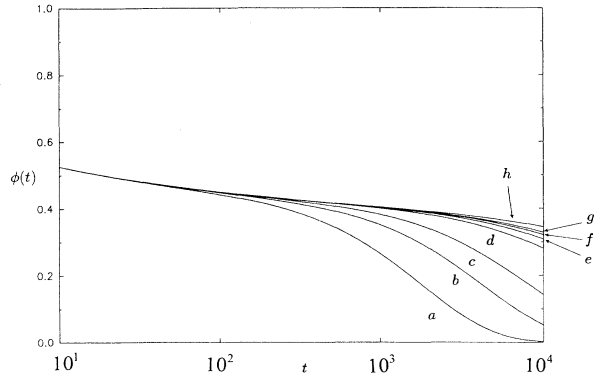

FIG. 5. Density autocorrelation function $\phi(t)$ for fixed $(c_1,c_2)=(0.556,2.778)$ and for various γ_v: curve a, 2.0×10^{-3}, curve b, 1.8×10^{-3}, curve c, 1.75×10^{-3}, curve d, 1.725×10^{-3}, curve e, 1.7225×10^{-3}, curve f, 1.7215×10^{-3}, curve g, 1.721×10^{-3}, and curve h, 1.72×10^{-3}.

$$2^{-1/\beta}=\frac{1-c_1g}{c_2f}+\frac{c_1g}{c_2f}\frac{\Gamma((1+\beta')/\beta)}{\Gamma(1/\beta)}\left[\frac{\tau}{\tau'}\right]^{\beta'}, \quad (4.6)$$

$$(\gamma_v-\gamma_v^c)\tau=\frac{1}{g\Gamma(1+1/\beta')}\left[\frac{\tau}{\tau'}\right]+fg\beta'\tilde{\xi}\Gamma\left[\frac{\beta+\beta'-1}{\beta}\right]\left[\frac{\tau}{\tau'}\right]^{\beta'}. \quad (4.7)$$

Numerically, we find that the second term in the right-hand side of Eq. (4.7) is much smaller than the first term. Thus, we can rewrite Eq. (4.7) as

$$(\gamma_v-\gamma_v^c)\tau'\simeq\frac{1}{g\Gamma(1+1/\beta')}, \quad (4.8)$$

which means that τ' diverges as $(\gamma_v-\gamma_v^c)^{-1}$ when $\gamma_v\to\gamma_v^c$. By obtaining the parameters β, β', τ, and τ' from explicit curve fits of $\phi(t)$ and $\psi(t)$, using Eq. (4.4) for various values of γ_v near γ_v^c, we find some evidence for this relation. Without the second term in the right-hand side of Eq. (4.6), it gives the expression for β of the conventional MCT with c_1 and $c_2(g=1)$: $\beta=-\ln(2)/\ln((1-c_1)/c_2f)$. However, in the presence of defect dynamics, Eq. (4.6) represents a more complicated expression for β that depends on β' as well as $\gamma_v-\gamma_v^c$. We find again evidence for this relation from our numerical data.

It is clear from these results that the separation of time scales between the defect and the density fluctuations occurs only when γ_v is very close to γ_v^c. We find that, as a function of ξ, the value of γ_v^c shows a smooth linear temperature dependence. Under the previous assumption that the slowing down corresponds to the $\sigma\to0$ (or $\gamma_v\to0$) limit, we were able to determine the temperature dependence of the metastability parameters, except for that of σ itself. This was done by assuming that the system arranges itself to be on the critical surface The present analysis of the model, including the defect dynamics, determines the temperature dependence of the parameter $\gamma_v\equiv\sigma y\tilde{\Gamma}_v q^2$ when the system is on the critical surface.

V. DISCUSSION

It was discovered by Das and Mazenko [13] that the nonhydrodynamic correction due to $\Sigma_{\hat{V}_\rho}(\mathbf{q},\omega)$ included in a representation like Eq. (2.30) cuts off the sharp nature of the ergodic-nonergodic transition. In the presence of the new variable n, however, we have a very complicated expression for the density response function. In order to see the cutoff effect, let the viscosity Γ become arbitrarily large. Then the expression reduces to

$$G_{\rho\hat{\rho}}(\mathbf{q},\omega)\simeq\frac{\omega}{\omega[\omega+iq\Sigma_{\hat{V}_\rho}(\mathbf{q},\omega)]+iq\Sigma_{\hat{V}n}(\mathbf{q},\omega)\Sigma_{\hat{n}\rho}(\mathbf{q},\omega)}. \quad (5.1)$$

If the self-energies $\Sigma_{\hat{V}n}$ and $\Sigma_{\hat{n}\rho}$, as well as $\Sigma_{\hat{V}_\rho}$ are set to zero, $G_{\rho\hat{\rho}}(t)$ reaches a finite value as time $t\to\infty$. The presence of these self-energies makes $G_{\rho\hat{\rho}}(t)$ decay slowly and thus provides the cutoff effect. The evaluation of these self-energies in principle can be done to yield complicated expressions in terms of $\phi(t)$ and $\psi(t)$. The analysis, including this effect, can thus be performed, but it will be a very difficult task, even numerically.

Throughout this paper, we have concentrated on the time relaxation behavior of the system near the glass transition. At this stage, it is important to note the implication of our model on the temperature dependence of the viscosity: $\eta(T)$. Traditionally, many different expressions have been used to fit the experimentally observed $\eta(T)$. These include the Arrhenius form $\sim\exp(A/T)$, the Vogel-Fulcher form $\sim\exp[B/(T-T_{VF})]$, and the power law $\sim|T-T_0|^{-\gamma}$. These forms are able to fit the experimental data only over limited temperature ranges

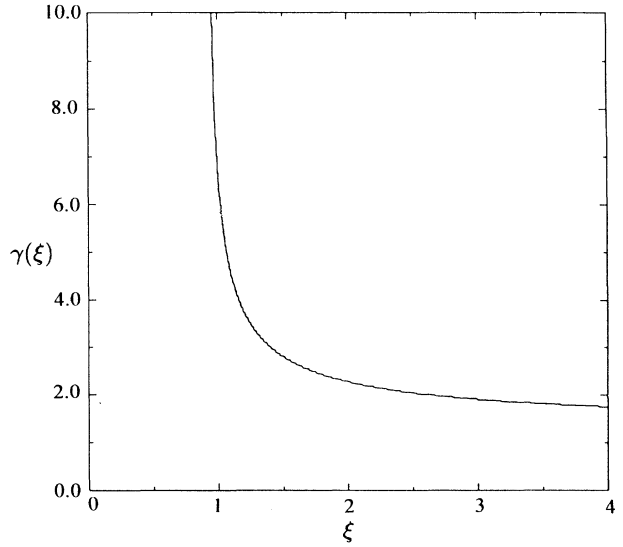

FIG. 6. Parameter $\gamma=1/(2a)+1/(2b)$ as a function of the temperature represented by ξ.

In the MCT, the temperature dependence of the viscosity is given by [1].

$$\eta(T)\sim\tau_\alpha(T)\sim|\sigma_0|^{-\gamma}\ , \tag{5.2}$$

where $\gamma=1/2a+1/2b$. According to the conventional MCT, σ_0 is assumed to be proportional to T_0-T and γ is temperature independent. Thus, the conventional MCT predicts that the viscosity shows a power-law divergence as the temperature approaches T_0. In our model, the situation is quite different. The temperature dependence of σ_0 is given by Eq. (3.13) without any special temperature. More importantly, $\gamma=1/(2a)+1/(2b)$ in this model is a function of temperature that is represented by the parameter ξ. Thus we have

$$\eta(\xi)=|f(\xi)\sigma(\xi)|^{-\gamma(\xi)}\ , \tag{5.3}$$

where $f(\xi)=\frac{2}{3}[3y+2C]\xi\lambda(1-\lambda)^2$. Therefore, the temperature dependence of the viscosity is governed mainly by the behavior of the exponent $\gamma(\xi)$ in Eq. (5.3) as a function of ξ, i.e., $\ln\eta(\xi)\sim\gamma(\xi)$. As shown in Fig. 6, $\gamma(\xi)$ increases as the temperature decreases. We do not expect, however, that Eq. (5.3) can be used directly to fit the experimental data, since a and b as functions of ξ are model dependent. Especially the rapid increase of $\ln\eta(\xi)$ near the lower bound of ξ should not be understood as a physical result, as noted in Sec. III. But this analysis clearly indicates that our model is consistent with the generic feature of the observed $\eta(T)$, which increases with decreasing temperature.

ACKNOWLEDGMENT

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