Viscous relaxation in metallic glasses

Subir Sachdev

AT&T Bell Laboratories, Murray Hill, New Jersey 07974

(Received 11 December 1985)

Recent theories of icosahedral ordering in supercooled liquids are used to define a model dynamics to describe the relaxation of momentum fluctuations in these systems. Glassy behavior of the system at microscopic distances is enforced by introducing a phenomenological relaxation parameter Γ^B which is anomalously small and strongly dependent upon temperature. Macroscopic consequences of this microscopic slowing down are examined. It is found that there is an upper bound on the magnitude of the translational correlation length, below which microscopic sluggishness is ineffective in producing a large macroscopic viscosity. If the correlation length is larger than this upper bound, then the macroscopic viscosity acquires a strong temperature dependence increasing like $1/\Gamma^B$ as the temperature falls. Binary metallic glasses have a correlation length which is just above this bound.

I. INTRODUCTION

A few recent studies of structural ordering in metallic glasses and undercooled liquids[1-5] have focussed on the importance of short-range icosahedral order in these systems. As a simple metal or metal-metalloid alloy is cooled below its melting temperature at a rate fast enough to avoid crystallization, many local icosahedral clusters of atoms are expected to form.[6] It is topologically impossible for the atoms to extend this icosahedral ordering to large distances. This "topological frustration" was identified as a key property of these systems. Building upon these ideas, a calculation of the structure factor of metallic glasses has been performed.[5] The effect of the icosahedral ordering upon the electronic density of states has been studied by calculations upon a finite cluster of atoms of polytope {3,3,5}.[7] This polytope exists upon the surface of a sphere in four dimensions with every atom sitting at the center of a perfect icosahedron.

While these approaches have, with some success, addressed the question of the structural ordering of the atoms, the precise role the icosahedral ordering plays in the dramatic increase of viscosity above the glass-transition temperature is, for the most part, not understood. It is clear that just above the glass-transition temperature, constraints on the rearrangement of atoms become very much more effective and essentially freeze the topological configuration of the atoms. An important unknown in this process is the length scale at which the rearrangement times of the atoms become comparable to the timescale of the experiment. A proper identification of this length scale requires that the correct order parameter which is driving the transition be identified. In the case of spin glasses, while the proper definition of the order parameter is still a matter of some controversy,[8] it is generally accepted that this order parameter acquires an infinite correlation length at the spin-glass-transition temperature. This implies that all length scales contribute to the diverging timescale. In conventional glasses and metallic glasses, the answer to this question is less clear. No diverging susceptibility has yet been discovered, implying

that the correlation length may be finite. In this paper we take the viewpoint that the important physics is contained in the short-range icosahedral ordering. With this identification of the relevant ordering, the question of the length scale at which the timescale for rearrangement becomes large will now be addressed.

An order parameter which measures the degree of short-range icosahedral ordering was introduced by Nelsen and Widom:[4] $Q^n_{m_1 m_2}(\mathbf{r})$. The value of $Q^n_{m_1 m_2}(\mathbf{r})$ at the point $\mathbf{r}$ depends upon the configuration of particles within a translational correlation length of $\mathbf{r}$. The order parameter transforms under the nth representation of SO(4), the m_1, m_2 are the basis indices. The $n = 12, 20, 24$ components of this order parameter are of particular importance in measuring the strength of the short-range icosahedral order. For the purposes of this paper it is sufficient to think of this order parameter as a complicated multiparticle correlation function which becomes large when the atoms near a given point are sitting at the vertices of tetrahedra, fivefold bipyramids, and icosahedra. A Landau expansion of the free energy in terms of this order parameter may now be performed. Up to quadratic order in the $\mathbf{Q}^n$ we have

$$F = \frac{1}{2} \sum_n \int d^3r \{ [K_n \,|\, (\partial_\mu - i\kappa L_{0\mu}) \mathbf{Q}^n \,|^2 + r_n \,|\, \mathbf{Q}^n \,|^2] \}$$

$$+ O((\mathbf{Q}^n)^3) \times \cdots . \tag{1.1}$$

Here, $L_{0\mu}$ is a generator of "translations" of SO(4); K_n and r_n are Landau-expansion parameters. The constant κ is a measure of the frustration in the system and is related to the interparticle spacing d by

$$\kappa d = \pi/5 . \tag{1.2}$$

This peculiar form of the gradient term was proposed by Sethna[3] and is necessary to account for the most favorable relative orientations and positions of neighboring icosahedra. Sachdev and Nelson[5] argued that a quadratic truncation of this free energy was a reasonable approximation, and were able to calculate density correlation functions. The results were in good agreement with the structure fac-

tors of amorphous vapor-deposited films and computer experiments.[9-12]

It was found that short-range icosahedral order gave rise to well-defined peaks in the structure factor. The structure factor of vapor-deposited films of amorphous cobalt is shown in Fig. 1.[11] The peak positions were determined by a combination of the icosahedral ordering and the frustration associated with the packing of icosahedra. The frustration was also responsible for the small but finite peak widths.[5] In a system where the icosahedra can pack together perfectly, e.g., the surface of a four-dimensional sphere, the peaks would have been Bragg spots.[4] From the widths of the peaks in the structure factor of a metallic glass, one can deduce a translational correlation length ξ of 2—3 atomic spacings. The length ξ is also the correlation length of the icosahedral order parameter. As ξ is finite, the assumption that the slowest process in the structural rearrangements of the atoms is the relaxation of the icosahedral order parameter implies that there is no diverging length scale in the system.

A possible mechanism for the slowing down of the dynamics was proposed by Nelson.[2] He argued that the defect lines in the icosahedral parameter field should suffer severe kinetic constraints in their movements. The non-Abelian nature of the symmetry group under which the order parameter transforms forces in an additional defect line whenever two defect lines try to pass through each other.[13] These kinetic constraints give rise to an entangled mass of defect lines, which freeze in at the glass-transition temperature. One would expect the entanglement constraints to become effective at the microscopic scale of a translational correlation length.

The question of the dynamic slowing down of the relaxation of the atoms is obviously an enormously complicated problem. In this paper we therefore choose to sidestep the question of the precise mechanism of the slowing down, entanglement or otherwise. Instead we assume that a microscopic slowing down exists on the scale of clusters
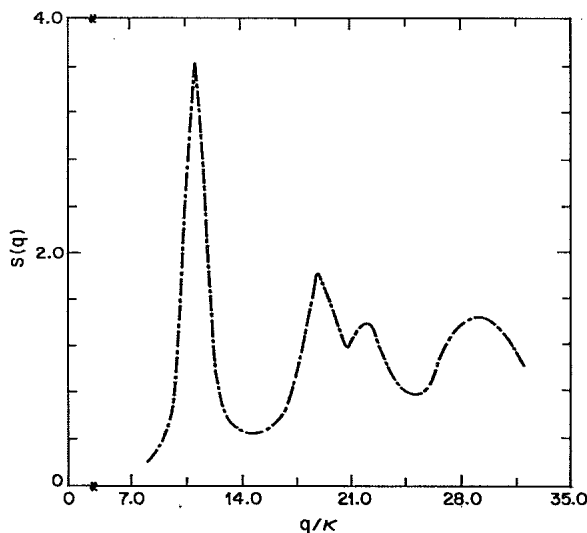

FIG. 1. Structure factor of a vapor-deposited film of amorphous cobalt from Ref. 11. Only the largest first peak is included in the dynamic calculations of this paper.

of 20—30 atoms and ask for possible macroscopic consequences as manifested in measurements of the shear viscosity. The relevant order parameter will be assumed to be the icosahedral order parameter. However, many features of the analysis would still hold even if some other type of ordering was found to be responsible for the dynamic slowing down.

In the analysis of the statics of metallic glasses it was found that the strongest fluctuations were those contributing to the principal peak of the structure factor.[5] These fluctuations belonged to the $n=12$ component of the order parameter. Additionally, it was found that only one particular linear combination of the Q^{12}'s contributed appreciably to the structure factor at wave vectors near the peak. This linear combination of the Q^{12}'s was the eigenvector of the quadratic part of the free energy having the lowest eigenvalue at the wave vector of the first peak. We therefore make the approximation that the most important component of the order parameter is a one-component scalar ψ, corresponding to this particular linear combination of the Q^{12}'s. For wave vectors near the peak in the structure factor we can therefore write the free energy as a functional of ψ.

The simplest equation which can describe the relaxation of ψ consistent with the free energy $F\{\psi\}$ is[14]

$$\frac{\partial \psi}{\partial t} = -\Gamma^B(-i\partial_\mu)\frac{\delta F}{\delta \psi} + \zeta , \quad (1.3)$$

where Γ^B is a relaxation coefficient and ζ is a source of thermal white noise with correlations chosen to satisfy the fluctuation-dissipation theorem. We assume that all the microscopic constraints that we referred to are incorporated in the function $\Gamma^B(k)$. In particular, we assume that Γ^B is strongly temperature dependent near T_g and becomes anomalously small at temperatures just above T_g and at wave vectors near the first peak in the structure factor.

A possible source for the decrease in Γ^B could be the nonlinearities in the free energy F. To lowest order, a cubic nonlinearity will lead to a decrease in Γ^B.[15] In this paper we are able to sidestep these issues because the static analysis showed that at the scale of the translational correlation length a quadratic truncation of the free energy will yield acceptable results. We assume that the nonlinearities are important at shorter length scales and lead to the strongly-temperature-dependent decrease in Γ^B. If the relaxation of the order parameter is dominated by activated processes with a typical barrier height E_B, Γ^B will have an Arrhenius temperature dependence $\exp(-E_B/kT)$. To a good first approximation, the peak in the structure factor is a Lorentzian. So the free-energy functional can be taken to be

$$F\{\psi\} = \frac{1}{2}\sum_k |\psi(k)|^2 \chi_0^{-1}(1+(k-k_0)^2\xi^2) , \quad (1.4)$$

where χ_0 is the maximum susceptibility, k_0 is the position of the peak in the structure factor, and ξ is the translational correlation length.

We now turn to an analysis of the effects of this microscopic slowing down upon the long-wavelength viscosity. To do this, we introduce a fluctuating shear momentum

density $\mathbf{j}^T$. These momentum fluctuations will obviously cause topological rearrangements and lead to a change in the relaxation of ψ. The simplest possible coupling between the momentum and ψ is a convective coupling:

$$\frac{\partial \psi}{\partial t} = -\Gamma^B(-i\partial_\mu)\frac{\delta F}{\delta \psi} - g_0 \mathbf{j}^T \cdot \nabla \psi + \zeta ,$$

$$\frac{\partial \mathbf{j}^T}{\partial t} = \overset{\leftrightarrow}{\mathbf{T}}_k \cdot \left[\eta^B \nabla^2 \mathbf{j}^T + g_0 \nabla \psi \frac{\delta F}{\delta \psi} + \theta \right] . \tag{1.5}$$

The momentum density $\mathbf{j}^T$ is assumed to be transverse. The matrix $\overset{\leftrightarrow}{\mathbf{T}}_k$ is a transverse projection operator and g_0 is a coupling constant. We have scaled the momentum density so that kinetic energy associated with the momentum fluctuations is $\frac{1}{2}\mathbf{j}^2$. Longitudinal fluctuations are omitted for simplicity. They will be considered later and will not affect the result appreciably. η^B is a bare viscosity. Because momentum fluctuations are not directly associated with fluctuations in the order parameter, there is no reason to assume that η^B is large. We will assume it has a value typical of liquids well above T_g. We shall be interested in studying the corrections to η^B from the slowing down of fluctuations in ψ.

The equations above are identical in form to the equations of the model H Hohenberg and Halperin[16,17] for the liquid-gas critical point. There are, however, several crucial differences. The divergence of the viscosity near the liquid-gas critical point depends crucially upon the fact that the order parameter ψ is conserved. In the system of interest in this paper, the question of whether ψ is conserved or not is irrelevant, because the strongest fluctuations of ψ occur at a finite wave vector. The value of Γ^B at k_0 will be most effective in the renormalization of η. In addition, the form of the free energy of ψ is dictated by the underlying frustration in the system and is quite different from what would happen near a critical point. The correlation length ξ is finite and only *weakly* temperature dependent near T_g. The *strong* temperature dependence is in $\Gamma^B(k_0)$. Nevertheless, some of the techniques that have been developed for the analysis of the liquid-gas critical point can be applied to the problem of interest. In particular, some of the earliest attempts at understanding the liquid-gas critical point were made by Kawasaki and Lo[18–20] and Oxtoby and Gelbart.[21] They used a simple self-consistent one-loop analysis, ignoring retardation effects, to calculate the viscosity and the order-parameter relaxation rate. Their results were in remarkable agreement with the experiments and were confirmed later by a more rigorous renormalization-group analysis.[17] In this paper we shall obtain all our results in the Kawasaki approximation. The first corrections due to higher-order loops and retardation effects are discussed in Appendix A and B, respectively.

The results of the simple Kawasaki analysis are as follows. The behavior of the macroscopic viscosity depends crucially on the magnitude of the correlation length. The physics of the system changes dramatically depending on whether $k_0\xi$ is greater or smaller than a positive constant c. Neglecting longitudinal fluctuations this constant c is determined to be 11.25. We now discuss the behavior of the system in the two regimes.

A. $k_0\xi < c$

The coupling between the order parameter and the momentum fluctuation leads to an appreciable correction to Γ. The renormalized value of Γ is not anomalously small and this leads to a renormalized viscosity which is not anomalously large. In other words, for this range of parameters, momentum fluctuations are strong enough to stir up the order parameter and eliminate its sluggish behavior. As a consequence, the renormalized viscosity remains of order the bare viscosity. We expect such a regime to exist, even beyond the Kawasaki approximation.

B. $k_0\xi > c$

In this regime, the renormalization of the viscosity η wins out and the system remains sluggish. There is no appreciable renormalization of Γ and the macroscopic viscosity is controlled by the freezing in of the microscopic clusters. The full frequency-dependent viscosity has the following behavior: At zero frequency, the viscosity increases with a decrease in Γ^B with the dependence

$$\eta \sim \frac{1}{\Gamma^B} . \tag{1.6}$$

For frequencies much greater than $\Gamma^B \chi_0^{-1}$, however, the viscosity acquires the form

$$\eta \sim -1/i\omega . \tag{1.7}$$

This indicates that at these frequencies the system has a reactive response and shear waves can propagate. The coefficient of $-1/i\omega$ can be identified as the infinite-frequency shear modulus G_∞. We also find that the zero-frequency viscosity η and the infinite-frequency shear modulus satisfy the simple relation

$$\eta\Gamma^B\chi_0^{-1} = \alpha G_\infty . \tag{1.8}$$

The constant α has a value of approximately 0.04. These results have, of course, been obtained in the simple Kawasaki approximation. As discussed in Appendix B, retardation corrections may reduce the value of the viscosity in this regime and lead to a dependence of η upon Γ^B which is weaker than that implied by Eq. (1.6).

Assuming that the bulk modulus of the material is of the same order of magnitude as its infinite-frequency shear modulus, we find that the phase space for the renormalization of Γ^B by longitudinal momentum fluctuations is very small. Thus there is a negligible correction in the value of the constant c when longitudinal fluctuations are included. The longitudinal viscosity D_l scales with the shear viscosity in the high-viscosity regime:

$$D_l = \frac{3}{2}\eta . \tag{1.9}$$

From the experimentally determined structure factor of amorphous cobalt shown in Fig. 1, we deduce a value of $k_0\xi$ of around 13.3. Most binary metallic glasses will have a value of $k_0\xi$ which is 10–20 % smaller than this value. It is noteworthy how close this value of $k_0\xi$ is to the value of the constant c in the Kawasaki approximation.

Nowhere in the analysis is the interpretation of ψ as an

icosahedral order parameter crucial. The scalar ψ may well be some other microscopic order parameter which is being frozen in. Only the fact that fluctuations of ψ are controlled by the free energy (1.4) is important in the subsequent analysis.

The outline of the remainder of the paper is as follows: In Sec. II we recapitulate the theory of the static structure factor of metallic glasses of Ref. 5. In Sec. III we define a model mode-coupling dynamics for the icosahedral order parameter and the momentum density. A graphical perturbation analysis of the model dynamics is presented in Sec. IV. The results of analyzing the self-consistent one-loop equations in the Kawasaki approximation are discussed in Sec. V. In Sec. VI we examine the effect of longitudinal momentum fluctuations which have been neglected so far. In Sec. VII we restate the main conclusions of the analysis of this paper. Corrections to the Kawasaki approximation are discussed briefly in the two appendixes.

II. REVIEW OF STATICS

We begin with a summary of the calculation of the static structure factor of Ref. 5. The density at a point $\mathbf{r}$ is expanded in terms of the order parameter $Q^n_{m_1 m_2}(\mathbf{r})$ as follows:[4]

$$\rho(\mathbf{r},\hat{\mathbf{u}}) = \sum_{n,m_1,m_2} Q^n_{m_1 m_2}(\mathbf{r})[Y^n_{m_1 m_2}(\hat{\mathbf{u}})]^* . \qquad (2.1)$$

Here, $\hat{\mathbf{u}}$ is a coordinate on a tangent four-dimensional sphere which is associated with every point in the physical space $\mathbf{r}$. The physical density is the density at the south pole of this tangent sphere. The $Y^n_{m_1 m_2}$ are the hyperspherical harmonics defined on the surface of a four-dimensional sphere and transform under the nth representation of SO(4). Using the value of the hyperspherical harmonics at the south pole, we obtain for the physical density

$$\rho(\mathbf{r}) = \sum_{n,m} (-1)^m \left[\frac{n+1}{2\pi^2} \right]^{1/2} Q^n_{mm}(\mathbf{r}) . \qquad (2.2)$$

The free energy which controls fluctuations in the order parameter $Q^n_{m_1 m_2}(\mathbf{r})$ was shown in Eq. (1.1). As argued in Ref. 5, a quadratic truncation of the free-energy expansion is adequate, facilitating the calculation of density correlations. We perform a Fourier transform in space and then diagonalize the quadratic form in the SO(4) representation basis. The eigenvalues of the quadratic form, $X^n_i(\mathbf{k})$, are all positive and we label them in ascending order by index i. If we label the ith eigenvector by $T^n_{m_1 m_2;i}(\mathbf{k})$, we may expand the order parameter Q^n in terms of the eigenvectors:

$$Q^n_{m_1 m_2}(\mathbf{k}) = \sum_i T^n_{m_1 m_2;i}(\mathbf{k})\alpha^n_i(\mathbf{k}) . \qquad (2.3)$$

The density-density correlations may now easily be calculated from the quadratic free energy (1.1) and the expansion (2.3). We obtain, for the structure factor of the liquid,

$$S(k) = \langle |\rho(k)|^2 \rangle$$

$$= \sum_{n,i} \frac{n+1}{2\pi^2} \frac{k_B T}{K_0 X^n_i(k) + r_n} \left| \sum_m T^n_{mm;i} \right|^2 . \qquad (2.4)$$

It is a good approximation to assume that for each representation n the contribution to the structure factor is dominated by the lowest eigenvalue $X^n_1(k)$. Therefore the structure factor goes through a maximum for each representation n, when $X^n_1(k)$ goes through a minimum as a function of k. Moreover, fluctuations in the density which do not have an icosahedral symmetry should be strongly suppressed from energetic considerations outlined earlier. This can be enforced by demanding that the masses r_n are very large for all representations n except those that allow fluctuations with an icosahedral symmetry, namely $n = 12,20,24,\ldots$. Thus we expect peaks in the structure factor associated only with these representations. Comparing the results of the calculation with experiments on vapor-deposited metal films, we find that the positions of the peaks (which are independent of the parameters K_n and r_n) are in good agreement, allowing one to determine r_{12}, r_{20}, r_{24} and K_{12}, K_{20}, K_{24}.

In the following analysis we shall make the approximation of including fluctuations only in the $n=12$ representation. We expect this to be a good approximation because the $n=12$ peak in the structure factor is much larger than the $n=20$ and 24 peaks. Besides, the factor of $(n+1)/2\pi^2$ which occurs in Eq. (2.4) for the expansion of the density also helps in making the order-parameter fluctuations in Q^n for $n=12$ larger. Therefore the relevant order parameter is $\alpha^{12}_1(\mathbf{k})$ and may be identified with the $\psi(\mathbf{k})$ discussed in the Introduction.

III. MODEL DYNAMICS

We will examine the relaxational dynamics of the order parameter Q^n in the presence of a convective coupling to a fluctuating momentum density $\mathbf{j}$. The equation of motion for Q^n has the form

$$\frac{\partial Q^n}{\partial t} + g_0 \mathbf{j} \cdot \nabla Q^n = -\Gamma^B \frac{\delta F}{\delta (Q^n)^*} + \zeta . \qquad (3.1)$$

This, in turn, implies that there exists a Poisson bracket between Q^n and $\mathbf{j}$ given by

$$\{Q^n_{m_1 m_2}(\mathbf{r}), j_\mu(\mathbf{r}')\} = -g_0 \partial_\mu Q^n_{m_1 m_2}(\mathbf{r})\delta(\mathbf{r}-\mathbf{r}') . \qquad (3.2)$$

The relaxation of the momentum density can be written in the form

$$\frac{\partial j_\mu}{\partial t} = \partial_\nu \sigma_{\mu\nu} , \qquad (3.3)$$

where $\sigma_{\mu\nu}$ is the stress tensor. Because of the Poisson-bracket coupling (3.2), there is a contribution to the stress tensor $\sigma_{\mu\nu}$ from the order-parameter fluctuations:

$$\sigma_{\mu\nu}=\sigma_{\mu\nu}^0-\frac{K_n}{2}[\partial_\mu Q^*\partial_\nu(Q^n)^*+\partial_\mu(Q^n)^*\partial_\nu Q^n-\delta_{\mu\nu}\partial_\sigma Q^*\partial_\sigma Q$$

$$+2i\kappa(Q^*L_{0\nu}\partial_\mu Q-\partial_\mu Q^*L_{0\nu}Q-\delta_{\mu\nu}Q^*L_{0\sigma}\partial_\sigma Q+\delta_{\mu\nu}\partial_\sigma Q^*L_{0\sigma}Q)-\kappa^2\delta_{\mu\nu}Q^*L_{0\sigma}L_{0\sigma}Q] . \tag{3.4}$$

For clarity we have suppressed the n, m_1, and m_2 indices from the expression above. $\sigma_{\mu\nu}^0$ is the stress arising from volume fluctuations and dissipation. It is nontrivial fact that the Poisson bracket (3.2) and the free energy (1.1) combine to give an expression for the relaxation of j which can be expressed in the form (3.3). It assures us that the formulation of the model dynamics is internally consistent even in the presence of frustration. These equations can be considerably simplified if we perform a Fourier transformation and express the equations in terms of the variables $\alpha_i^n(\mathbf{k})$, which are the coefficients of the eigenvectors of the quadratic form

$$\frac{\partial\alpha_i(\mathbf{k})}{\partial t}+g_0\frac{1}{\sqrt{V}}\sum_q \mathbf{j}^T(\mathbf{q})\cdot[-i(\mathbf{k}-\mathbf{q})]T_i^*(\mathbf{k})T_j(\mathbf{k}-\mathbf{q})\alpha_j(\mathbf{k}-\mathbf{q})=-\Gamma^B X_i(\mathbf{k})\alpha_i(\mathbf{k})+\zeta ,$$

$$\tag{3.5}$$

$$\frac{\partial\mathbf{j}^T(q)}{\partial t}+\eta^B\mathbf{j}^T(q)-g_0\frac{\overset{\leftrightarrow}{\mathbf{T}}_q}{\sqrt{V}}\sum_k X_i(-\mathbf{k})\alpha_i(-\mathbf{k})[-i(\mathbf{k}+\mathbf{q})]T_i^*(k)T_j(k+q)\otimes\alpha_j(\mathbf{k}+\mathbf{q})=\theta .$$

Again, redundant indices on α_i^n, X_i^n, and $T_{m_1m_2;j}^n$ have been suppressed, and a viscosity η^B has been introduced. The momentum fluctuations have been restricted to be purely transverse. This constraint will be removed later.

We will be interested in momentum fluctuations which occur at small wave vectors, while the order-parameter fluctuations will occur at the wave vectors near k_0. Under these conditions, we use the orthonormality of the eigenvectors T^n to approximate

$$\sum_{m_1,m_2} T_{m_1m_2;j}^*(k)T_{m_1m_2;j}(k+q)\approx\delta_{ij} . \tag{3.6}$$

If we now concentrate only upon the $\alpha_1^{12}(k)$ mode, the above equations reduce to the model equations (1.4) discussed in the Introduction. Focusing only on wave vectors for order-parameter fluctuations which are near the first peak in the structure factor, we can use the approximation (1.4) for the free energy. We will restrict the wave vector k of the order parameter to lie in the range

$$k_0-\frac{b}{\xi}\le k\le k_0+\frac{b}{\xi} . \tag{3.7}$$

b will be chosen to be 3, although the results are very insensitive to the value of b.

IV. PERTURBATION ANALYSIS

We use the standard Martin-Siggia-Rose[22] technique to perform the calculation. The techniques are very similar to calculations performed earlier on momentum relaxation near the liquid-gas critical point[17] and thermal conductivity near the λ transition[23,24] in liquid helium. We will therefore just define the notation and present the results of the graphical analysis. In this section and the next we will assume that the momentum fluctuations are purely transverse. This is equivalent to assuming a large bulk modulus. The effect of longitudinal fluctuations will be considered in Sec. VI.

We introduce the conjugate fields $\hat{\psi}$ and $\hat{j}$, which enables us to transform the equations into a Martin-Siggia-Rose functional. If we index the hatted fields by 1 and unhatted fields by 2, the matrix propagators for the ψ field, G, and the j field, D, are given by (we have set $k_BT=1$)

$$G^{-1}(\mathbf{k},\omega)=\begin{bmatrix} -2\Gamma^B(\mathbf{k})-\Sigma_{11}(\mathbf{k}) & i\omega-\Gamma^B(\mathbf{k})\chi^{-1}(\mathbf{k})-\Sigma_{12}(\mathbf{k},\omega) \\ -i\omega-\Gamma^B(\mathbf{k})\chi^{-1}(\mathbf{k})-\Sigma_{12}^*(\mathbf{k},\omega) & 0 \end{bmatrix},$$

$$D_*^{-1}(k,\omega)=\begin{bmatrix} -2\eta^B k^2-\Pi_{11}(\mathbf{k},\omega) & -i\omega-\eta^B k^2-\Pi_{12}^*(\mathbf{k},\omega) \\ i\omega-\eta^B k^2-\Pi_{12}(\mathbf{k},\omega) & 0 \end{bmatrix}.$$

$$\tag{4.1}$$

We have introduced the self-energies Π and Σ for the ψ and j fields. The renormalized viscosity and relaxation coefficients are related to these self-energies by

$$\eta(k,\omega)\equiv\eta^B+\frac{1}{k^2}\Pi_{12}(k,\omega)$$

$$\equiv\eta^B+\eta^C(k,\omega) ,$$

$$\Gamma(k,\omega)\equiv\Gamma^B+\chi(k)\Sigma_{12}(k,\omega)$$

$$\equiv\Gamma^B+\Gamma^C(k,\omega) . \tag{4.2}$$

Momentum autocorrelations are given by D_{22} and field autocorrelations are given by G_{22}. The physical response functions are not equal to G_{12} and D_{12} because of the presence of the convective coupling. In the absence of any nonlinearities in the statics, a simple relationship exists between the response functions R_ψ and R_j and the Green's functions:

$$R_\psi(k,\omega)=-[\Gamma^B+\Sigma_{12}(k,\omega)\chi(k)]G_{21}(k,\omega) ,$$

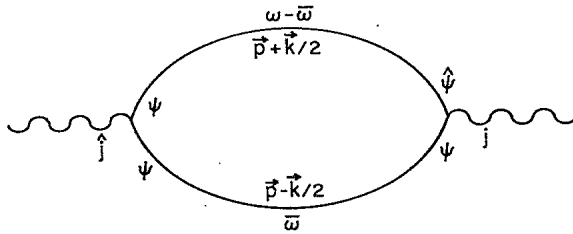$$R_j(k,\omega)=-[\eta^B k^2+\Pi_{21}(k,\omega)]D_{21}(k,\omega) . \tag{4.3}$$

FIG. 2. One-loop graph contributing to the correction to the viscosity. The internal lines are the exact Green's functions.

The fluctuation-dissipation theorem now implies the following relationship between the self-energies:

$$\Sigma_{11}(k,\omega)=2\mathcal{X}(k)\mathrm{Re}[\Sigma_{12}(k,\omega)] \ ,$$

$$\Pi_{11}(k,\omega)=2\,\mathrm{Re}[\Pi_{12}(k,\omega)] \ . \tag{4.4}$$

From the self-consistent one-loop equations for the Green's functions we may deduce relationships between the renormalized viscosity and Γ. The equations for the viscosity follow from the graph in Fig. 2. The wavy lines represent the momentum j and the smooth lines represent fluctuations of the order parameter ψ. We have
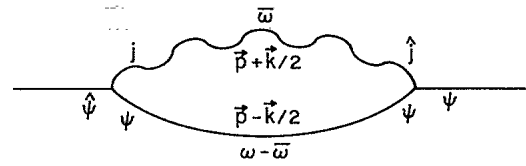

FIG. 3. Two one-loop graphs contributing to the relaxation coefficient of the order parameter. Again the internal lines are exact Green's functions.

$$\eta^C(k,\omega)=\frac{g_0^2}{k^2}\int\frac{d\bar{\omega}}{2\pi}\frac{d^3p}{(2\pi)^3}(\mathbf{p}\cdot\overset{\leftrightarrow}{\mathbf{T}}_k\cdot\mathbf{p})[\mathcal{X}^{-1}(p_+)-\mathcal{X}^{-1}(p_-)]\mathcal{X}(p_-)G_{21}(p_+,\omega-\bar{\omega})\mathrm{Re}[G_{21}(p_-,\omega)] \ , \tag{4.5}$$

where $\mathbf{p}_{\pm}=\mathbf{p}\pm\mathbf{k}/2$. Two graphs shown in Fig. 3 contribute to the renormalized relaxation coefficient Γ. Their sum yields the following equation for $\Gamma(k,\omega)$:

$$\Gamma^C(k,\omega)=2g_0^2\int\frac{d\bar{\omega}}{2\pi}\int\frac{d^3p}{(2\pi)^3}(\mathbf{k}\cdot\overset{\leftrightarrow}{\mathbf{T}}_{p_-}\cdot\mathbf{k})\mathcal{X}(p_+)D_{21}(p_-,\omega-\bar{\omega})\mathrm{Re}[G_{21}(p_+,\bar{\omega})] \ . \tag{4.6}$$

Equations (4.5) and (4.6) will form the basis of our further analysis.

V. RESULTS IN THE KAWASAKI APPROXIMATION

The Kawasaki approximation[18] involves neglecting the frequency dependence of η and Γ on the right-hand side of Eqs. (4.5) and (4.6). This will tend to *overestimate* the value of the zero-frequency viscosity. Thus, if we find that a microscopic slowing down does not yield an increasing macroscopic viscosity, then this result will hold for the full Eqs. (4.5) and (4.6). However, in the parameter regime where the viscosity increases it is possible that retardation effects may destroy this increase in viscosity.

Equations (4.5) and (4.6) yield, in the Kawasaki approximation for the zero-frequency viscosity η and Γ,

$$\eta^C(k)=\frac{g_0^2}{4k^2}\int\frac{d^3p}{(2\pi)^3}\mathbf{p}\cdot\overset{\leftrightarrow}{\mathbf{T}}_k\cdot\mathbf{p}\frac{[\mathcal{X}^{-1}(\mathbf{p}+\mathbf{k})-\mathcal{X}^{-1}(\mathbf{p})]^2}{[\Gamma(\mathbf{p}+\mathbf{k})\mathcal{X}^{-1}(\mathbf{p}+\mathbf{k})+\Gamma(\mathbf{p})\mathcal{X}^{-1}(\mathbf{p})]\mathcal{X}^{-1}(\mathbf{p}+\mathbf{k})\mathcal{X}^{-1}(\mathbf{p})} \ ,$$

$$\Gamma^C(k)=g_0^2\int\frac{d^3p}{(2\pi)^3}\frac{\mathbf{k}\cdot\overset{\leftrightarrow}{\mathbf{T}}_{p-k}\cdot\mathbf{k}\mathcal{X}(p)}{\eta(\mathbf{p}-\mathbf{k})\cdot(\mathbf{p}-\mathbf{k})^2+\Gamma(\mathbf{p})\mathcal{X}^{-1}(\mathbf{p})} \ . \tag{5.1}$$

In the limit that the order-parameter susceptibility is a Lorentzian with a small width, it is a good approximation to replace $\eta(k)$ by its value at $k=0$, and $\Gamma(k)$ by its value at $k=k_0$. Then the equations above become a pair of simultaneous equations for $\eta(k=0)$ and $\Gamma(k=k_0)$. In the following we shall drop the wave vector arguments of η and Γ,

$$\eta^C=\frac{g_0^2}{8}\lim_{k\to 0}\frac{1}{k^2}\int\frac{d^3p}{(2\pi)^3}\frac{\mathbf{p}\cdot\overset{\leftrightarrow}{\mathbf{T}}_k\cdot\mathbf{p}}{\Gamma\mathcal{X}^{-3}(\mathbf{p})}[\mathcal{X}^{-1}(\mathbf{p}+\mathbf{k})-\mathcal{X}^{-1}(\mathbf{p})]^2, \quad \Gamma^C=g_0^2\int\frac{d^3p}{(2\pi)^3}\frac{\mathbf{k}\cdot\overset{\leftrightarrow}{\mathbf{T}}_{p-k}\cdot\mathbf{k}\mathcal{X}(\mathbf{p})}{\eta(\mathbf{p}-\mathbf{k})^2}\Bigg| \ . \tag{5.2}$$

Using the expression for the susceptibility in (1.4), restricting the wave vector integrals to lie in the range in (3.7), and reinserting the missing factors of density and temperature, these equations may be reduced to a simple one-dimensional quadrature,

$$\eta^C = \frac{P}{\Gamma^B + \Gamma^C} \, ,$$

$$\Gamma^C = \frac{Q}{\eta^B + \eta^C} \, ,$$

(5.3)

where the variables P and Q are given by

$$P = \frac{k_B T \chi_0 k_0^2}{\pi^2 \xi \rho_0} \frac{l^2}{30} \int_{-b}^{b} \frac{x^2 (1 + x/l)^4}{(1 + x^2)^3} dx \, ,$$

(5.4a)

$$Q = \frac{k_B T \chi_0 k_0^2}{\pi^2 \xi \rho_0} \frac{1}{8} \int_{-b}^{b} dx \frac{(1 + x/l)^2}{1 + x^2} \left[\frac{(1 + x/l)^2 + 1}{1 + x/l} \ln \left[\frac{|2l + x|}{|x|} \right] - 2 \right] ,$$

(5.4b)

with $l = k_0 \xi$. Equations (5.3) can be solved and we obtain, for the correction to the viscosity,

$$\eta^C = \frac{P - Q - \Gamma^B \eta^B + [(\Gamma^B \eta^B + Q - P)^2 + 4 P \Gamma^B \eta^B]^{1/2}}{2 \Gamma^B} \, .$$

(5.5)

In the limit as Γ^B tends to 0, this equation yields

$$\eta^C = \begin{cases} \dfrac{P - Q}{\Gamma^B} & \text{for } P > Q \, , \\[3mm] \dfrac{P}{Q - P} \eta^B & \text{for } Q > P \, . \end{cases}$$

(5.6)

The condition of the relative magnitude of P and Q reduces to a comparison of the two integrals in Eqs. (5.4a) and (5.4b) as a function of $l = k_0 \xi$. We find that $P > Q$ for $k_0 \xi > 11.25$ and $P < Q$ otherwise.

The main result of the analysis above is that if $k_0 \xi$ is less than 11.25, then the renormalized viscosity will remain of the order of the bare viscosity and not increase dramatically even in the presence of a microscopic slowing down. The existence of such a regime transcends the Kawasaki approximation and should hold for the fully-self-consistent equations (4.5) and (4.6). The value of the upper bound 11.25 should increase slightly with a more complete analysis. If $k_0 \xi$ is greater than this upper bound, then the viscosity diverges like $1/\Gamma^B$ as Γ^B becomes small. The temperature dependence of the viscosity is then intimately related to the unknown temperature dependence to Γ^B. It is possible that retardation corrections could bring about a weaker dependence of η upon Γ^B.

Another useful result can be obtained from Eqs. (4.5) and (4.6) that is more general than the Kawasaki approximation. We look at the frequency-dependent viscosity in the regime $\omega \gg \Gamma \chi_0^{-1}$. Then Eq. (4.5) simplifies and we obtain

$$\eta^C(k = 0) = -\frac{1}{i\omega} \frac{g_0^2}{4} \lim_{k \to 0} \frac{1}{k^2} \int \frac{d^3 p}{(2\pi)^3} \mathbf{p} \cdot \overset{\leftrightarrow}{\mathbf{T}}_k \cdot \mathbf{p} \frac{[\chi^{-1}(p_+) - \chi^{-1}(p_-)]^2}{\chi^{-1}(p_-) \chi^{-1}(p_+)} \, .$$

(5.7)

Performing the angular integral and reverting to experimental units, we obtain

$$\eta^C = \frac{\chi_0^{-1}}{-i\omega} R \, ,$$

(5.8a)

where

$$R = \frac{k_B T \chi_0 k_0^2}{\pi^2 \xi \rho_0} \frac{l^2}{15} \int_{-b}^{b} dx \frac{(1 + x/l)^4 x^2}{(1 + x^2)^2} \, .$$

(5.8b)

This implies that the substance has a finite shear modulus at frequencies which are much greater than the order-parameter relaxation rate. The infinite-frequency shear modulus G_∞ is given by

$$G_\infty = \chi_0^{-1} R \, .$$

(5.9)

In the regime where the viscosity is diverging, this implies a simple relationship between Γ^B, η, and G_∞:

$$(\Gamma^B \chi_0^{-1}) \eta = \alpha G_\infty \, ,$$

(5.10a)

with

$$\alpha = (P - Q)/R \, .$$

(5.10b)

The numerical factor α can be evaluated. In the large-viscosity regime (for $l \sim 13$)

$$\alpha = (P - Q)/R \approx 0.04 \, .$$

(5.11)

VI. EFFECT OF LONGITUDINAL FLUCTUATIONS

We now check to see if the longitudinal fluctuations are effective in appreciably changing the results discussed above. We shall find, essentially from phase-space considerations, that the longitudinal fluctuations are quite ineffective in renormalizing the value of Γ^B and, hence, affecting the results discussed.

The equations of motion in the presence of longitudinal momentum fluctuations are as follows:

$$\frac{\partial \psi}{\partial t} = -\Gamma^B(-i\partial)\frac{\delta F}{\delta \psi} - g_0 \nabla \psi \cdot (j^T + j^2) + \zeta, \quad \frac{\partial j^T}{\partial t} = \overset{\leftrightarrow}{T} \cdot \left[\eta \nabla^2 j^T + g_0 \nabla \psi \frac{\delta F}{\delta \psi} + \theta_1 \right],$$

$$\frac{\partial j^L}{\partial t} = \overset{\leftrightarrow}{L} \cdot \left[D_l \nabla^2 j^L + g_0 \nabla \psi \frac{\delta F}{\delta \psi} - \frac{g_0 \nabla \delta \rho}{B} + \theta_2 \right], \quad \frac{\partial \delta \rho}{\partial t} = -g_0 \nabla \cdot j^L .$$

(6.1)

$\delta \rho$ represents the long-wavelength density fluctuations, B is the bulk modulus, D_l is the longitudinal viscosity, and $\overset{\leftrightarrow}{L}$ is a longitudinal projection operator. The perturbation analysis for this equation proceeds in much the same way as discussed in the preceding sections. Here we shall be primarily interested in the high-viscosity regime, and we wish to investigate whether there are appreciable corrections to the order-parameter relaxation rate.

As a first step we evaluate the corrections to the longitudinal viscosity from the order-parameter fluctuations. The one-loop corrections to D_l are found to be

$$D_l^C(k) = \frac{g_0^2}{2k^2} \int \frac{d^3 p}{(2\pi)^3} \frac{\mathbf{p}_- \overset{\leftrightarrow}{L}_k \cdot [\mathbf{p}_- \chi^{-1}(p_+) - \mathbf{p}_+ \chi^{-1}(p_-)]}{\chi^{-1}(\mathbf{p}_-)[\Gamma(p_+)\chi^{-1}(p_+) + \Gamma(p_-)\chi^{-1}(p_-)]} .$$

(6.2)

After evaluating this integral as $k \to 0$, we find

$$D_l^C = \tfrac{3}{2}\eta^C .$$

(6.3)

After a similar calculation in the high-frequency limit, we can find the corrections to the bulk modulus:

$$D_l^C(\omega \to \infty) = \frac{1}{-i\omega} \frac{g_0^2}{2k^2} \int \frac{d^3 p}{(2\pi)^3} \frac{\mathbf{p}_- \cdot \overset{\leftrightarrow}{L}_k \cdot [\mathbf{p}_- \cdot \chi^{-1}(p_+) - \mathbf{p}_+ \chi^{-1}(\mathbf{p}_-)]}{\chi^{-1}(p_-)} ,$$

(6.4)

which yields

$$B_x - B = \tfrac{3}{2} G_x .$$

(6.5)

We can now proceed to the calculation of corrections to Γ from longitudinal momentum fluctuations. The sum of the graphs of Fig. 3, where the momentum fluctuations are now longitudinal, is given by

$$\Gamma^C(k) = 2g_0^2 \int \frac{d^3 p}{(2\pi)^3} \frac{d\bar{\omega}}{2\pi} \frac{1}{-i\omega + \Gamma(p_+)X^{-1}(p_+)}$$

$$\times \text{Re}\left[\frac{i(\omega - \bar{\omega})}{(\omega - \bar{\omega})^2 + i(\omega - \bar{\omega})D_l(p_-)p_-^2 - Bp_-^2} \right] \otimes (\mathbf{p}_+ \cdot \overset{\leftrightarrow}{L}_{p_-} \cdot \mathbf{k}) X(p_+) .$$

(6.6)

The velocity of sound in this liquid is $\sqrt{B}$. If we now assume that sound can travel across a correlation length in a time shorter than the order-parameter relaxation time [this amounts to ignoring the $(\omega - \bar{\omega})^2$ in the denominator], and make the Kawasaki approximation, we find, for Γ,

$$\Gamma^C(k) = g_0^2 \int \frac{d^3 p}{(2\pi)^3} \frac{\mathbf{p} \cdot \overset{\leftrightarrow}{L}_{p-k} \cdot \mathbf{p}}{(\mathbf{p} - \mathbf{k})^2} \frac{\Gamma(p)}{D_l(p-k)\Gamma(p)X^{-1}(p) + B} .$$

(6.7)

Evaluating Γ^C for $k = k_0$, we find

$$\Gamma^C = \frac{k_B T X_0 k_0^2}{\pi^2 \xi \rho_0} \frac{1}{D_l} \frac{1}{4} \int_{-b}^{b} dx \frac{(1+x/l)^2}{1+x^2+\beta} \left[1 + \frac{(1+x/l)^2 - 1}{2(1+x/l)} \ln \left[\frac{2l+x}{|x|} \right] \right] .$$

(6.8)

We have introduced a parameter β which is given by

$$\beta = \frac{B}{D_l X_0^{-1} \Gamma} = \frac{B}{\tfrac{3}{2} G_\infty \alpha} = \frac{B}{B_\infty - B} \frac{1}{\alpha} .$$

(6.9)

From the expression above we can compare the relative magnitudes of the corrections to Γ^C from longitudinal and transverse fluctuations. It is clear that for $\beta \gg 1$ the contributions from the transverse part will be quite small. Also, even if we assume that B is as small as G_∞, β will be quite large and suppress the longitudinal fluctuations. For $l = 13$ and $\beta = 20$, we find that the longitudinal contri-

bution is 2.5% of the transverse contribution. Thus we do not expect a significant change in the value $k_0 \xi$ at which the system crosses over from small to large viscosities.

VII. CONCLUSION

We have described a simple model of dynamic relaxation in metallic glasses. Microscopic slowing down is described by a parameter $\Gamma^B(\mathbf{k})$ which is *assumed* to become anomalously small just above T_g for $k \simeq k_0$. The macroscopic consequences of this short-distance sluggishness were then explored. It was found that if the correla-

tion length ξ was smaller than some upper bound, the microscopic slowing down was ineffective in slowing down momentum fluctuations. Instead, the momentum fluctuations destroy the microscopic freezing in, and lead to large-distance behavior which is liquidlike. In the Kawasaki approximation the upper bound on the correlation length was given by $k_0\xi < 11.25$. Remarkably, the correlation length of a typical metallic glass is just above this bound.

In the large-correlation-length regime, the decrease in Γ^B led to an increase in the zero-freezing viscosity η of the form $\eta \sim 1/\Gamma^B$. Retardation corrections, which were ignored, may lead to a weaker dependence of η upon Γ^B. At frequencies much greater than $\Gamma^B \chi_0^{-1}$, the system acquired a finite shear modulus G_∞. Thus if the time $\chi_0 \Gamma^{B-1}$ becomes larger than the experimental time, the system will behave like a glass with a reactive shear response. The shear modulus G_∞, the viscosity η, and the relaxation time $\chi_0 (\Gamma^B)^{-1}$ are related by

$$\eta = \alpha G_\infty \chi_0 (\Gamma^B)^{-1} , \qquad (7.1)$$

with $\alpha \approx 0.04$.

ACKNOWLEDGMENTS

APPENDIX A: EFFECT OF VERTEX CORRECTIONS

In this appendix we shall examine the effects of including some of the graphs that have so far been omitted. A typical example of such a graph is shown in Fig. 4. We shall, in particular, be interested in whether any of the graphs have a dependence on Γ^B which is stronger than $1/\Gamma^B$. The result that $\eta \sim 1/\Gamma^B$ depends crucially on their being no such terms. We shall find indeed that there are no such terms.

To begin the analysis, let us perform a simplified estimate of the value of the graph in Fig. 4. We shall neglect the effect of the wave-vector fluctuations and assume that on the average each momentum line carries a wave vector of $1/\xi$ and each order-parameter line carries a wave vector of k_0. Then in the time domain each order parameter behaves like $\exp[-\Gamma(k_0)\chi_0^{-1} t]$ and the momentum lines behave like $\exp(-\eta t/\xi^2)$. For brevity of notation, we shall denote these time dependences as $\exp(-\bar{\Gamma}t)$ and $\exp(-\bar{\eta}t)$, respectively. The contribution of Fig. 4 to the zero-frequency viscosity behaves like
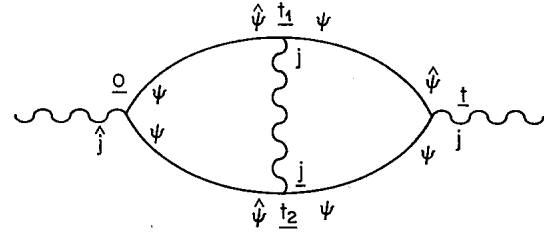

FIG. 4. Typical higher-order graph which has been neglected. The underlined symbols at the vertices denote the time at each vertex.

$$\delta\eta^C \sim \int_0^x dt \int_0^x dt_2 \int_0^t dt_1 \, e^{-\bar{\Gamma}t_2} e^{-\bar{\Gamma}|t-t_2|}$$
$$\times e^{-\bar{\eta}|t_1-t_2|} e^{-\bar{\Gamma}t}$$
$$\sim \frac{1}{2\bar{\Gamma}^2\bar{\eta}} + O(1/\bar{\eta}^2\bar{\Gamma}) . \qquad (A1)$$

The largest contribution to this integral comes from the time interval in which the two vertices which are connected by the momentum propagator are consecutive in time. This fact will be true for *any* graph.

The contribution of a general graph can be written as a sum of terms in which the vertices have various time orderings. As in the graph already considered, the largest contribution arises when the vertices at the ends of a momentum line define a time interval with no other vertex time. Pick any such graph. Each interval with a momentum fluctuation will contribute a factor $1/\bar{\eta}$ and the remaining intervals will give factor $1/\bar{\Gamma}$. Since there is at least one more order-parameter interval than a momentum interval, the contribution of a graph with I vertices will have to be less divergent than $1/\bar{\Gamma}^{n+1}\bar{\eta}^n$ with $I = 2n + 2$. From this we see that the ansatz $\eta \sim 1/\Gamma$ is self-consistent since no terms with a strong dependence on Γ can be obtained. In a similar manner we may show that Γ^C consists of a sum of terms none of which is more divergent than $1/\bar{\Gamma}^n\bar{\eta}^{n+1}$.

APPENDIX B: EFFECT OF RETARDATION CORRECTIONS

In this appendix we will make a very simple estimate of the first correction due to dynamic effects. Because of the complexity of the integral equations (4.5) and (4.6), we will be forced to decouple the ω and k-space integrals: the wave-vector integrals will just give phase-space factors and the momentum and order-parameter fluctuations will be governed by a single frequency. As discussed by Siggia,[25] this procedure is hazardous because it will reproduce features characteristic of single particles. The results in this appendix are therefore only a first estimate. However, the conclusion that dynamic corrections will *reduce* the viscosity is expected to be true on more general grounds.

Performing the frequency integrals in Eqs. (4.5) and (4.6), we find

$$\Gamma^C(k,\omega) = g_0^2 \int \frac{d^3p}{(2\pi)^3} \frac{\mathbf{k} \cdot \overset{\leftrightarrow}{T}_{p_-} \cdot \mathbf{k} \chi(p_+) Z}{-i\omega + \eta(p_-, \omega - \omega^*) p_-^2 + \Gamma(p_+, \omega^*) \chi^{-1}(p_+)} \qquad (B1)$$

and

$$\eta^C(k,\omega) = \frac{g_0^2}{2k^2} \int \frac{d^3p}{(2\pi)^3} \frac{\mathbf{p} \cdot \overset{\leftrightarrow}{T}_k \cdot \mathbf{p} [\chi^{-1}(p_+) - \chi^{-1}(p_-)] \chi(p_-) Z}{-i\omega + \Gamma(p_-, \omega - \omega^*) \chi^{-1}(p_-) + \Gamma(p_+, \omega^*) \chi^{-1}(p_+)} , \qquad (B2)$$

with

$$\omega^* = -i\Gamma(p_+, \omega^*) \chi^{-1}(p_+) , \qquad (B3)$$

and Z given by

$$Z^{-1} = 1 + i\chi^{-1}(p_+) \left. \frac{\partial \Gamma(p_+, \omega)}{\partial \omega} \right|_{\omega = \omega^*} . \qquad (B4)$$

Ignoring the wave-vector dependence of ω^*, and using the approximations $k_0 \xi \gg 1$ and $\eta/\xi^2 \gg \omega^*, \Gamma_0^{-1}$, we find

$$\Gamma^C(\omega) = \frac{g_0^2 \chi_0 k_0^2}{4\pi\xi} Z \frac{\ln(2k_0\xi) - 1}{\eta(\omega - \omega^*)} \qquad (B5)$$

and

$$\eta^C(\omega) = \frac{g_0^2 \chi_0 k_0^2}{30\pi\xi} (k_0\xi)^2 \frac{Z}{\Gamma(\omega^*) + \Gamma(\omega - \omega^*)} \left[\frac{1 - \sqrt{x}}{1 - x} \right]^2 , \qquad (B6)$$

with

$$x = 1 - \frac{i\omega}{[\Gamma(\omega^*) + \Gamma(\omega - \omega^*)] \chi_0^{-1}} . \qquad (B7)$$

The wave-vector arguments of Γ and η are presumed to be k_0 and 0, respectively, and have been suppressed. From Eq. (B6) the percentage change in η^C in the high-viscosity regime is dependent on the fractional change in $\Gamma(\omega^*) + \Gamma(-\omega^*)$. $\Gamma(\omega^*)$ depends on $\eta(\omega = 0)$, so to this order is unchanged. $\Gamma(-\omega^*)$ depends on $\eta^C(-2\omega^*)$. Using $\omega^* = -i\Gamma(\omega = 0) \chi_0^{-1}$, we see—to lowest order from Eqs. (B6) and (B7)—that

$$\frac{\eta^C(-2\omega^*)}{\eta^C(0)} = \frac{[(1 - \sqrt{x})/(1 - x)]^2 |_{x = 1.5}}{[(1 - \sqrt{x})/(1 - x)]^2 |_{x = 1}} = 0.8 . \qquad (B8)$$

So there is a 20% decrease in $\Gamma(-\omega^*)$ and no change in $\Gamma(\omega^*)$. Since $\eta(\omega = 0)$ depends on $\Gamma(\omega^*) + \Gamma(-\omega^*)$, the dynamic corrections amount to a 10% decrease in the viscosity.

[1] P. J. Steinhardt, D. R. Nelson, and M. Ronchetti, Phys. Rev. Lett. 47, 1297 (1981); Phys. Rev. B 28, 784 (1983).

[2] D. R. Nelson, Phys. Rev. Lett. 50, 982 (1983); Phys. Rev. B 28, 5515 (1983).

[3] J. P. Sethna, Phys. Rev. Lett. 50, 2198 (1983); Phys. Rev. B 31, 6278 (1985).

[4] D. R. Nelson and M. Widom, Nucl. Phys. B240 [FS12], 113 (1984).

[5] S. Sachdev and D. R. Nelson, Phys. Rev. Lett. 53, 1947 (1984); Phys. Rev. B 32, 1480 (1985).

[6] F. C. Frank, Proc. R. Soc. London, Ser. A 215, 43 (1952).

[7] M. Widom, Phys. Rev. B 31, 6456 (1985).

[8] See, for example, M. Mezard, G. Parisi, N. Sourlas, G. Toulouse, and M. Virasoro, Phys. Rev. Lett. 52, 1156 (1984); D. J. Gross and M. Mezard, Nucl. Phys. B240 [FS12], 431 (1984).

[9] G. S. Cargill III, in Solid State Physics, edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic, New York, 1975), Vol. 30, p. 227.

[10] T. Ichikawa, Phys. Status Solidi A 19, 707 (1973).

[11] P. K. Leung and J. C. Wright, Philos. Mag. 30, 185 (1974).

[12] J.-P. Lauriat, J. Non-Cryst. Solids 55, 77 (1983).

[13] N. D. Mermin, Rev. Mod. Phys. 51, 591 (1979).

[14] B. I. Halperin, P. C. Hohenberg, and S.-k. Ma, Phys. Rev. Lett. 29, 1548 (1972); Phys. Rev. B 10, 139 (1974).

[15] S. P. Das, G. F. Mazenko, S. Ramaswamy, and J. J. Toner, Phys. Rev. Lett. 54, 118 (1985); S. P. Das and G. F. Mazenko (unpublished).

[16] P. C. Hohenberg and B. I. Halperin, Rev. Mod. Phys. 49, 435 (1977).

[17] E. D. Siggia, B. I. Halperin, and P. C. Hohenberg, Phys. Rev. B 13, 2110 (1976).

[18] K. Kawasaki and S. Lo, Phys. Rev. Lett. 29, 48 (1972).

[19] S. Lo and K. Kawasaki, Phys. Rev. A 5, 421 (1972).

[20] S. Lo and K. Kawasaki, Phys. Rev. A 8, 2176 (1973).

[21] D. Oxtoby and W. M. Gelbart, J. Chem. Phys. 61, 2957 (1974).

[22] P. C. Martin, E. D. Siggia, and H. A. Rose, Phys. Rev. A 8, 423 (1973).

[23] B. I. Halperin, P. C. Hohenberg, and E. D. Siggia, Phys. Rev. B 13, 1299 (1976).

[24] P. C. Hohenberg, E. D. Siggia, and B. I. Halperin, Phys. Rev. B 14, 2865 (1976).

[25] E. D. Siggia, Phys. Rev. A 32, 3135 (1985).