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# Light-scattering spectrum of supercooled liquids

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Two versions of extended mode-coupling theory are compared in the long-wavelength limit. The results are relevant for the light-scattering spectrum of supercooled liquids. It is shown that both versions predict the existence of a central Mountain peak whose width is independent of the wave vector at small $k$.

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## I. INTRODUCTION

The light-scattering spectrum [1] of simple liquids consists of three Lorentzian lines: a Rayleigh line at frequency $\omega = 0$ and a pair of Brillouin lines located at $\omega = \pm ck$. Here $\mathbf{k}$ is the wave vector, $k = |\mathbf{k}|$, and $c$ is the speed of sound. This spectrum is determined by the long-wavelength hydrodynamic modes [2]: the Rayleigh line is due to the heat mode, and the Brillouin lines are caused by the sound modes. It is well known that the line-widths are proportional to $k^2$, the proportionality coefficients being the thermal diffusivity $D_T$ and the sound damping coefficient $\Gamma_s$, respectively.

This simple picture no longer applies for supercooled liquids. The reason is that nonhydrodynamic modes associated with structural relaxation processes on short (molecular) length scales become so slow that they fall into the GHz window probed by light scattering. As the glass transition is approached, the slowing down of structural relaxation becomes, in fact, dramatic with an increase of characteristic times by more than 10 orders of magnitude in a temperature range of only about 100 K (for a review on the dynamic aspects of the glass transition, see Ref. [3]). Hence there must be a temperature range where structural relaxation is slow enough to stand out against the background of the spectrum, but not yet so slow as to vanish within experimental resolution. Here the Rayleigh-Brillouin spectrum is expected to differ from that of a normal liquid.

Nonhydrodynamic modes also play a role in the case of molecular liquids. Mountain [4] has calculated the scattering spectrum for a fluid with a frequency dependent bulk viscosity, which serves as a model to describe the internal degrees of freedom. He considered, in particular, a Maxwell model with a single relaxation time $\tau_M$ and found [4,5] that the positions and widths of the Brillouin lines are modified, and that a new component to the Rayleigh line appears. Moreover, the width of this new "Mountain peak" (given by $1/\tau_M$) stays finite in the limit $k \to 0$ [6], which is in contrast to the behavior of the hydrodynamic peaks. As a consequence, for wave vectors $k < (D_T\, \tau_M)^{-1/2}$, the width of the Rayleigh line becomes independent of $k$. For undercooled simple liquids, where the slow nonhydrodynamic modes are due to structural relaxation, the $k$ independence of the width of the Rayleigh line has been established in recent experiments [7]. This very general property of the scattering spectrum can thus be used as a qualitative test for any theory of structural relaxation.

The most important theory of structural relaxation is the mode-coupling theory (MCT, for reviews, see Refs. [8–10]), which determines a set of nonlinear equations for the density-density correlation function $\Phi(k,t)$, the quantity measured in light-scattering experiments. A self-consistent treatment of these equations at finite wave numbers leads to the so-called $\alpha$ peak, which describes the slowest mode of structural relaxation. Due to the interplay between the structural and the hydrodynamic modes [11,12], the calculation of the light-scattering spectrum is rather subtle [13,14]. But it turns out that the width of the Mountain line is essentially given by the width of this $\alpha$ peak.

In its original form [11], the MCT predicts a sharp transition from ergodic behavior $[\lim_{t\to\infty} \Phi(k,t) = 0]$ at temperatures $T > T_c$ to nonergodic behavior $[\lim_{t\to\infty} \Phi(k,t) > 0]$ at temperatures $T < T_c$. Here $T_c$ is a well-defined critical temperature associated with a glass-transition singularity. In this *idealized* MCT the $\alpha$ peak exists only above $T_c$ (since, below $T_c$, there is no ultimate relaxation). For simple systems, the equations of $\alpha$ relaxation can be solved numerically for all wave vectors [15], and the width of the $\alpha$ peak is found to be only weakly wave vector dependent with a finite value at $k = 0$.

There is experimental evidence [16], however, indicating that there are very slow relaxation processes besides those treated in the idealized MCT, which prevent the system from complete structural arrest. To include such ergodicity-restoring processes, several generalizations of the original MCT have been proposed. They are referred to as *extended* MCTs. Common to these theories is that not only density-density couplings are taken into account, as in the idealized theory, but also density-current and current-current couplings. However, there is no agreement on the detailed form of the equations and the coupling vertices. One class of such theories is based on nonlinear hydrodynamics [17] and its generalization to finite wave numbers [18,19], while the other [20,21] is based on a renormalized kinetic approach [22].

The extended MCTs replace the sharp glass transition by a soft one, in the sense that $\Phi(k,t)$ ultimately decays to zero for all temperatures, although, at sufficiently low $T$, there is a broad time window within which $\Phi(k,t)$ appears to be frozen. As a consequence, the $\alpha$ peak no longer ceases to exist below some $T_c$. Concerning the predictions of the extended MCTs on the Mountain peak, it was noted [13] that the kinetic version leads to a $k$ independent width, while for the hydrodynamic version a width proportional to $k^2$ was anticipated. It is the purpose of this paper to demonstrate that this is not the case: the hydrodynamic theory also leads to a $k$-independent width.

## II. IDEALIZED MODE-COUPLING THEORY

Structural relaxation is described in terms of the normalized density-density correlation function

$$\Phi(k,t) = \frac{\langle \delta n(\mathbf{k},t)\ \delta n(-\mathbf{k},0)\rangle}{\langle \delta n(\mathbf{k},0)\ \delta n(-\mathbf{k},0)\rangle}, \tag{1}$$

where $\delta n(\mathbf{k},t)$ denotes a density fluctuation about the mean value $n$ and the averages $\langle\cdot\rangle$ are taken over an equilibrium ensemble at temperature $T$. According to idealized MCT, the Laplace transform, $\tilde{\Phi}(k,z) = \int_0^\infty dt\ e^{-zt}\ \Phi(k,t)$, satisfies a Dyson-type equation

$$\tilde{\Phi}(k,z) = \cfrac{1}{z + \cfrac{\Omega^2(k)}{z + \nu_L(k)k^2 + \Omega^2(k)\ \tilde{m}(k,z)}} \tag{2}$$

where $\Omega(k)$ is the phonon frequency, $\nu_L(k)$ is a "bare" longitudinal viscosity (associated with dissipation on a short "Enskog" time scale), and $\tilde{m}(k,z)$ is a frequency dependent relaxation function, which dominates the long-time dynamics. This relaxation function is related back to $\Phi(k,t)$ via a nonlinear constitutive equation of the form

$$m(k,t) = \int d^3q\ \Lambda(\mathbf{k},\mathbf{q})\ \Phi(q,t)\ \Phi(|\mathbf{k}-\mathbf{q}|,t), \tag{3}$$

with coupling vertices $\Lambda(\mathbf{k},\mathbf{q})$ that are essentially determined by the static structure factor. It is important to note that $\Lambda(\mathbf{k},\mathbf{q})$ stays finite in the limit $k \to 0$. Equations (2) and (3) provide a self-consistent theory for the density-density correlation function $\Phi(k,t)$.

For small $k$, the density-density correlation function can be probed in light-scattering experiments. In this limit,

$$\Omega(k) \to c_L k, \qquad \nu_L(k) \to \nu_L, \qquad m(k,t) \to m(t), \tag{4}$$

where $c_L$ and $\nu_L$ are the bare sound velocity and the bare sound damping coefficient, respectively, and (2) is reduced to

$$\tilde{\Phi}(k,z) = \cfrac{1}{z + \cfrac{{c_L}^2k^2}{z + \nu_L k^2 + \tilde{m}(z)\ {c_L}^2k^2}}. \tag{5}$$

The contribution from the heat mode is neglected in this expression. The interplay of the glass singularity and the heat mode gives rise to frequency dependent thermodynamic derivatives [12], but does not qualitatively change the conclusion of this paper. We shall come back to this point in the Discussion.

The static structure factor, and hence the coupling vertices are controlled by the temperature. A major result of the idealized theory is the existence of a critical temperature $T_c$, below which the vertices become strong enough to cause a transition from ergodic to nonergodic behavior of $\Phi(k,t)$ [11]. This implies that, in the low-frequency limit,

$$\tilde{m}(z) = \frac{g}{z} \qquad (T < T_c), \tag{6}$$

where $g$ is a form factor that is determined by the self-consistent solution of (2) and (3) for all wave numbers. It turns out [11] that the dominant part of (3) is due to $q$ values close to the main peak of the static structure factor. The long-wavelength value $g$ is thus determined primarily by mode-coupling effects on molecular length scales.

It is straightforward to determine the poles and residues of Eq. (5), with $\tilde{m}(z)$ given by (6). As we focus on the small-$k$ regime, satisfying

$$\nu_L k \ll c_L \tag{7}$$

we can use first-order perturbation theory. Thus the spectrum is found to consist of two Brillouin lines and a Mountain line,

$$\tilde{\Phi}(k,z) = \tilde{\Phi}_B(k,z) + \tilde{\Phi}_M(k,z), \tag{8}$$

where

$$\begin{aligned} \tilde{\Phi}_B(k,z) &= \frac{1}{2(1+g)}\left[\frac{1}{z+z_{B+}(k)} + \frac{1}{z+z_{B-}(k)}\right], \\ \tilde{\Phi}_M(k,z) &= \frac{g}{1+g}\ \frac{1}{z+z_M(k)}, \end{aligned} \tag{9}$$

and the modes are given by

$$\begin{aligned} z_{B\pm}(k) &= \pm\ i\sqrt{1+g}\ c_L k + \frac{1}{2}\ \nu_L k^2 \\ z_M(k) &= 0. \end{aligned} \qquad (T < T_c) \tag{10}$$

The last result implies that the Mountain line is degenerate to an elastic line of zero width, which is a signature of an ideal glassy solid. Notice also that the Brillouin lines are shifted by a factor $\sqrt{1+g}$, as compared to a normal liquid. This is the well-known viscoelastic increase of the sound velocity (see, e.g., [23] and references therein).

At temperatures above $T_c$ (but still in the supercooled phase), the vicinity of the glass transition singularity is signaled by a broad time window within which $\Phi(k,t)$ appears to be frozen onto a plateau of finite height. Eventually, at still longer times, $\Phi(k,t)$ decays to zero by $\alpha$ relaxation [8-10]. The time scale of $\alpha$ relaxation provides $\tilde{m}(z)$ with a low-frequency cutoff. Hence we write in place of (6)

$$\tilde{m}(z) = \begin{cases} \frac{g}{z} & (|z| \gg g/\tau) \\ \tau & (|z| \ll g/\tau) \end{cases} \qquad (T > T_c). \quad (11)$$

Here we have spezified only the high- and low-frequency behavior of $\tilde{m}(z)$. In the crossover regime, it has a complicated structure that is not needed for the present purpose. The relaxation time $\tau$ is strongly temperature dependent and goes to infinity as $T$ approaches $T_c$. Therefore, at given $k$, there is a temperature range where

$$\tau c_L k \gg g. \quad (12)$$

Under the conditions given by (7) and (12), the sound modes fall into the high-frequency regime ($|z| \approx \sqrt{1+g}\ c_L k \gg g/\tau$), so that they have the same form as below $T_c$. On the other hand, the relaxation time $\tau$ becomes important for small frequencies ($|z| \ll g/\tau$) and leads to a finite lifetime of the elastic mode. Thus, from (5) with $\tilde{m}(z)$ given by (11), one finds that the spectrum is again of the form (8) and 9, but with

$$\begin{aligned} z_{B\pm}(k) &= \pm\, i\sqrt{1+g}\ c_L k + \frac{1}{2}\nu_L k^2 \\ z_M(k) &= \frac{1}{\tau} \end{aligned} \qquad (T > T_c). \quad (13)$$

The Brillouin lines are unchanged, while the Mountain line is broadened, its linewidth $\tau^{-1}$ being independent of $k$.

## III. EXTENDED MODE-COUPLING THEORY

In the extended MCTs, the Dyson equation (2) is replaced by a more complicated one, the form of which is controversial among different versions of the theory.

### A. Hydrodynamic versions

The hydrodynamic versions lead to a Dyson equation of the form

$$\tilde{\Phi}(k,z) = \cfrac{1}{z + \Omega^2(k)\ \tilde{m}_0(k,z) + \cfrac{\Omega^2(k)\ [1-\Omega(k)\ \tilde{m}_1(k,z)]^2}{z + \nu_L(k)k^2 + \Omega^2(k)\ \tilde{m}_2(k,z)}}, \quad (14)$$

with three relaxation functions $\tilde{m}_a(k,z)$ ($a = 0,1,2$). The function $\tilde{m}_2(k,z)$ plays the same role as $\tilde{m}(k,z)$ in the idealized theory, and the constitutive expression for $m_2(k,t)$ is again of the form ( 3). The other functions, $m_1(k,t)$ and $m_0(k,t)$ , are of a similar form, but involve couplings to the current (momentum density). It is observed from (14) that $\tilde{m}_2(k,z)$ describes a renormalization of the bare viscosity, while $\tilde{m}_1(k,z)$ give rise to a renormalization of the phonon frequency. Finally, $\tilde{m}_0(k,z)$ leads to a low-frequency cutoff that suppresses the $1/z$ singularity of $\tilde{\Phi}(k,z)$ below $T_c$.

A theory of this type was first advocated by Das and Mazenko [17] on the basis of nonlinear fluctuating hydrodynamics. The short-wavelength structure, which is a main ingredient of the idealized mode-coupling theory, was initially not taken into account. But this was later corrected by Das [18]. In the Das-Mazenko theory (for a review, see [9]) it is argued that the Galilean invariance of fluctuating hydrodynamics is responsible for the existence of the cutoff. Another theory by Schmitz, Dufty, and De [19], also based on the short-wavelength generalized hydrodynamics, pursues the detailed-balance symmetry as the source of the cutoff. This also leads to a Dyson equation of the form (14). It should be emphasized, however, that the two theories are not equivalent, since the constitutive equations, relating the relaxation functions back to $\Phi(k,t)$, differ from each other. But for the present purpose this is of no concern.

For small $k$, the formulas (4) apply now with three relaxation functions that all stay finite. Moreover, since $\tilde{m}_0(z)$ and $\tilde{m}_1(z)$ involve couplings to the currents, one can argue that they are less frequency dependent than $\tilde{m}_2(z)$. For simplicity we therefore assume that

$$\tilde{m}_0(z) \to \frac{\delta}{{c_L}^2 {k_m}^2} \quad (15)$$

independently of $z$. Here, $k_m$ is the wave vector at the peak of the structure factor. The frequency scale $\delta$ is the natural scale for hopping processes as discussed in the kinetic MCT [8,20]. There it was understood that the most important contributions to $\tilde{m}_0(k,z)$ come from the peak of the structure factor. Then the Dyson equation (14) is reduced to

$$\tilde{\Phi}(k,z) = \cfrac{1}{z + \delta\,\cfrac{k^2}{{k_m}^2} + \cfrac{{c_L}^2 k^2}{z + \nu_L k^2 + \tilde{m}_2(z)\ {c_L}^2 k^2}}. \quad (16)$$

Notice that there is no contribution from $\tilde{m}_1(z)$, since we have only kept terms up to second order in $k$.

Although the sharp glass singularity is removed in extended MCT, it is still reminiscent at high frequencies. Thus we write

$$\tilde{m}_2(z) = \begin{cases} \frac{g}{z} & (|z| \gg g/\tau) \\ \tau & (|z| \ll g/\tau), \end{cases} \tag{17}$$

but now, in contrast to (11), for all temperatures (in the supercooled regime). Again, the values $g$ and $\tau$ are dominated by mode-coupling effects at wave numbers $q$ close to the main peak of the static structure factor. For low temperatues (below the $T_c$ of idealized MCT), it is the cutoff term $\tilde{m}_0(k,z)$ in (14) that causes $\alpha$ relaxation of $\Phi(q,t)$. This behavior is then transmitted via Eq. (3) to $\tilde{m}_2(z)$, giving rise to a finite value of $\tau$, which in turn influences via self-consistency the value of $\tilde{m}_0(k,z)$. For the mode-coupling scenario to be consistent, the frequency $\delta$ must be well separated from the microscopic frequencies [24]. In the hydrodynamic context we have to supplement the two conditions (7) and (12) by a third one, namely,

$$\frac{\delta}{c_L k_m} \frac{k}{k_m} \ll 1. \tag{18}$$

The above assumptions on $g$, $\tau$, and $\delta$ still need, of course, support from a fully self-consistent solution of the extended mode-coupling equations.

Using (7), (12), and (18), the poles and residues of (16) and (17) can be evaluated as before by a small $k$ expansion. The resulting spectrum is again of the form (8) and (9), while the modes are given by

$$\begin{aligned} z_{B\pm}(k) &= \pm\, i\sqrt{1+g}\; c_L k + \frac{1}{2}\left(\nu_L + \frac{\delta}{k_m^2}\,\frac{1}{1+g}\right) k^2, \\ z_M(k) &= \frac{1}{\tau} + \frac{\delta}{k_m^2} k^2. \end{aligned} \tag{19}$$

Observe that the $\delta$ terms only contribute in order $k^2$ to the linewidths. It is also important to note that the amplitude of the Mountain line (which is determined by the $\alpha$ process) is again $g/(1+g)$ as in the idealized theory. This is in contrast to the amplitude 1 found in the literature [9,17].

### B. Kinetic version

The kinetic version of extended MCT leads to a Dyson equation of a somewhat different form. Employing the notation of the preceding paragraph, it reads

$$\tilde{\Phi}(k,z) = \cfrac{1}{z + \cfrac{\Omega^2(k)}{z + \cfrac{\nu_L(k)k^2 + \Omega^2(k)\;\tilde{m}_2(k,z)}{1 + \tilde{m}_0(k,z)\;\left[\nu_L(k)k^2 + \Omega^2(k)\;\tilde{m}_2(k,z)\right]}}}. \tag{20}$$

Again $\tilde{m}_2(k,z)$ plays the same role as $\tilde{m}(k,z)$ in the idealized theory, while $\tilde{m}_0(k,z)$ describes the cutoff. The expression (20) has been derived by Götze and Sjögren [20,21] in a renormalized kinetic appoach [22]. The theory in this form was successfully used to describe the so-called $\beta$-relaxation regime in depolarized light-scattering and neutron-scattering experiments [25,16].

Making the same assumptions as in the hydrodynamic version, one obtains a spectrum of the same form, but with somewhat different modes, namely,

$$\begin{aligned} z_{B\pm}(k) &= \pm\, i\sqrt{1+g}\; c_L k + \frac{1}{2}\left(\nu_L + \frac{\delta}{k_m^2}\,\frac{g^2}{1+g}\right) k^2, \\ z_M(k) &= \frac{1}{\tau} + \frac{\delta}{k_m^2}\; k^2, \end{aligned} \tag{21}$$

Again the $\delta$ terms provide only $k^2$ corrections to the linewidths. The expressions (19) and (21) differ only with respect to the broadening of the Brillouin lines.

## IV. DISCUSSION

We conclude with a number of discussion remarks.

(1) To leading order in the wave number, the width of the Mountain line is independent of $k$. This is predicted by all versions of extended MCT.

(2) As mentioned above, we have ignored the heat mode. Its inclusion leads to an additional diffusive peak that is superimposed to the Mountain line [4,5]. But for small enough wave numbers, $k < (D_T\ \tau)^{-1/2}$, the heat mode becomes so narrow that the Mountain line can be resolved alone. Notice, however, that the heat mode will also affect the bare sound velocity and the bare sound damping, as in the case of normal liquids. Additionally the slowing down of structural relaxations leads to frequency-dependent renormalizations of thermodynamic derivatives [12].

(3) Several authors [26] have considered vacancy diffusion below $T_c$ as an alternative mechanism for ergodicity restauration. This implies an additional diffusion line in the spectrum, which is superimposed to the Mountain line. Nevertheless the mechanism described in the present paper leads also in this case to a $k$-independent width of the Mountain line. The difference will only show up in order $k^2$ with a model-dependent coefficient. For small enough wave numbers, $k < (D_V\ \tau)^{-1/2}$, the vacancy diffusion mode is covered by the Mountain line.

(4) In this paper we have only discussed the conse-

quences of the distinct Dyson equations being employed in the different versions of extended MCT. To actually calculate the parameters $(g, \tau, \delta)$, a self-consistent solution of the full set of equations (Dyson equation plus constitutive expressions for the relaxation functions) has to be undertaken. In a numerical study of the kinetic MCT for a Lennard-Jones liquid [27] the values of $1/(\omega_0 \tau)$ and $\delta/\omega_0$ were found to be of the order $10^{-7}$ and $10^{-10}$, respectively, where $\omega_0$ is a microscopic frequency defined in Ref. [27]. This means that a clear separation of the microscopic scale, the $\alpha$ scale, and the scale of activated hopping processes has been obtained. Moreover, the condition $\delta/\omega_0 \ll 1$ makes the so-called $\beta$ relaxation scaling regime stand out against the microscopic background [24] in agreement with the experimental findings [16]. Up until now, the numerical solutions of the generalized nonlinear hydrodynamic equations in the vicinity of the glass transition [18,28] were not reliable enough to draw conclusions about the values of $(g, \tau, \delta)$.

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