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COMMENTS

Comments are short papers which criticize or correct papers of other authors previously published in the **Physical Review**. *Each Comment should state clearly to which paper it refers and must be accompanied by a brief abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Reexamination of the depolarized-light-scattering spectra of glass-forming liquids

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We reexamine the depolarized-light-scattering data of Cummins and co-workers [Phys. Rev. A **45**, 3867 (1992); **46**, 3343 (1992); Phys. Rev. E **47**, 4223 (1993)] and interpret these data rather differently than they. These authors make use of the mode-coupling theory of glasses which makes a number of significant predictions that have been associated with the observed behavior of supercooled liquids as they approach the glassy state. To date the strongest experimental support for the mode-coupling theory of glasses comes from the analysis by Cummins and co-workers of their depolarized-light-scattering data. For this reason it is particularly important to subject this analysis to careful scrutiny. Our conclusion is that these data do not show strong evidence for the critical behavior predicted by the mode-coupling theory of glasses.

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INTRODUCTION

Recently Cummins and co-workers [1–3] have reported extensive depolarized-light-scattering studies of liquid and supercooled CKN [2] and Salol [3], and they have interpreted these data in terms of the mode-coupling theory (MCT) of glasses. The standard MCT of glasses, as developed by Götze and others [4], describes relaxation processes in supercooled liquids in terms of collective modes. It predicts a dynamical critical temperature T_c, well above the standard glass temperature T_g, in the vicinity of which the dynamical behavior is divergent, and, in particular, the theory predicts a plot of relaxation time versus temperature which is cusplike about T_c. Cummins and co-workers extract such cusplike behavior from their analysis of the depolarized-light-scattering spectra of glass formers CKN [2] and Salol [3], and it is this result that has been taken as the most compelling supporting evidence for the theory. Before detailing our analysis of their work, we point out that their *raw data*, the logarithm of light-scattering intensity versus logarithm of frequency at different temperatures, ranging from well above to well below their putative T_c, suggest continuous change with no evidence of critical cusps; both the intensities and the slopes of the intensity versus ω curves decrease continuously as T is lowered. This, in and of itself, makes us hesitant to accept an analysis with an intrinsic critical point at T_c above T_g.

We will not discuss the theoretical validity of the MCT of glasses, nor the many other experimental tests to which it has been subjected; the principal question addressed here is not whether a critical point is encountered as a supercooled liquid is cooled, but whether such a critical point has been properly identified as a temperature well above T_g.

THE DATA

Cummins and co-workers have plotted $\log[\chi''(\omega)]$ versus $\log[\omega]$ curves for depolarized light scattering at different temperatures, where the susceptibility $\chi''(\omega)$ is equal to $I(\omega)[1+n(\omega)]^{-1}\approx\omega I(\omega)$, $I(\omega)$ is the intensity at frequency ω of the light-scattering spectrum, and $n(\omega)$ is the Bose factor. To the extent that the spectra are the consequence of rotational motions, this light-scattering susceptibility is the second rank analog of the imaginary part of the first rank dielectric susceptibility; however, it is likely, as noted by Cummins *et al.*, that a major part of the light-scattering spectrum, in contrast to the dielectric spectrum, arises from fluctuations in the dipole–induced-dipole (DID) interactions. It is interesting to note that these DID contributions are not expected in the corresponding dielectric relaxation spectra [6(a)], nor in the neutron-scattering spectra [6(b)].

The $\log[\chi''(\omega)]$ versus $\log[\omega]$ curves at a given temperature rise to a maximum at a relatively low frequency, $\omega_{\rm max}$, pass through a minimum at a higher frequency, $\omega_{\rm min}$, and rise to a second maximum at a high frequency, $\omega_{\rm mol}$, characteristic of individual molecular processes. (For Salol the high-frequency regime is slightly more complicated.) The frequency $\omega_{\rm mol}$, as well as the susceptibility $\chi''(\omega_{\rm mol})$ at that frequency, appear to be quite independent of temperature. Both the frequencies $\omega_{\rm max}$ and $\omega_{\rm min}$ decrease with decreasing temperature; at temperatures below the putative T_c, Cummins and co-workers found that both $\omega_{\max}$ and $\omega_{\min}$ were so small that they fell out of the resolution range, but there is no indication that these features suddenly disappear at lower temperatures. At frequencies above $\omega_{\min}$ the susceptibility at any given ω decreases continuously as the temperature is lowered.

At the highest temperatures studied the minimum is at such high frequencies that there is almost no rise in $\chi''(\omega)$ between $\omega_{\min}$ and ω_{mol}. At lower temperatures where the rise is distinct, the presence of "leaked" Brillouin lines, at resonance frequencies which increase with decreasing temperature, tends to obscure spectral details.

A CRITIQUE OF THE ANALYSIS GIVEN IN REFS. [2, 3]

The simple form of the mode-coupling theory of glasses divides the spectrum into an α and a β component, the former giving rise to the spectrum in the vicinity of $\omega_{\max}$, the latter to the spectrum from somewhat below $\omega_{\min}$ on up, but terminating at frequencies somewhat below ω_{mol}. The simple MCT of glasses predicts nonergodic behavior for the α relaxation at temperatures below a critical temperature T_c, i.e., $\omega_{\max}\rightarrow 0$ as $T\rightarrow T_c$ from above, and at the same T_c it predicts that the minimum vanishes; it also predicts that the shape of the spectrum changes below T_c to one where $\chi''(\omega)$ is proportional to ω up to a "knee" frequency ω_e above which $\chi''(\omega)$ is proportional to ω^a, where a is a constant exponent.

Cummins and co-workers [2,3] fit the susceptibility in the region around its minimum (observed at temperatures above the putative T_c) to an interpolation formula

$$\chi''(\omega)=\chi''(\omega_{\min})\frac{1}{a+b}\left[a\left[\frac{\omega_{\min}}{\omega}\right]^{b}+b\left[\frac{\omega}{\omega_{\min}}\right]^{a}\right], \tag{1}$$

which is compatible with the mode-coupling interpolation formula

$$\chi''(\omega)=A|\varepsilon|^{(a+b)/2a}\omega^{-b}+B\omega^{a}\quad (T>T_c), \tag{2}$$

where

$$\varepsilon=\frac{T_c-T}{T_c}, \tag{3}$$

and a, b, A, B, and T_c are taken to be independent of temperature. In the same temperature range they fit the frequency $\omega_{\max}$ at which $\chi''(\omega)$ reaches its first maximum to

$$\omega_{\max}=C|\varepsilon|^{\gamma}, \tag{4}$$

where C and γ are constants. (One can equally well fit the viscosity since it is proportional to $\omega_{\max}^{-1}$.)

At low temperatures, $T<T_c$, they fit $\chi''(\omega)$ at frequencies well above the minimum to a formula which reduces to

$$\chi''(\omega)=B\omega^{a}\quad\text{for } \omega>\omega_0|\varepsilon|^{1/2a} \tag{5a}$$

and to

$$\chi''(\omega)=B\omega_0^{a-1}|\varepsilon|^{(a-1)/2a}\omega\quad\text{for } \omega<\omega_0|\varepsilon|^{1/2a}, \tag{5b}$$

where ω_0 is a constant. Note that there is no critical dependence in the second term of Eq. (2) nor in Eq. (5a), but in the MCT analysis these terms are expressed as the product of two factors, each separately dependent upon ε.

The simple MCT of glasses imposes the following interrelationships among the exponents a, b, γ:

$$\gamma=\frac{1}{2a}+\frac{1}{2b}, \tag{6}$$

$$\Gamma^2(1-a)/\Gamma(1-2a)=\Gamma^2(1+b)/\Gamma(1+2b)=\lambda . \tag{7}$$

Thus the experimental challenge to the MCT of glasses is to fit the data to Eqs. (1)–(5) and to see whether the parameters a, b, γ satisfy the interrelationships in Eqs. (6) and (7).

For $T>T_c$

A stringent test of the mode-coupling theory of glasses, at least in the temperature regime above the putative T_c, is whether the exponents a and b are temperature independent and related by Eq. (7). To carry out this test one can make use of Eqs. (1)–(4) and (6) to obtain the MCT relations

$$\omega_{\min}\propto|\varepsilon|^{1/2a}\propto\omega_{\max}^{b/(a+b)}, \tag{8}$$

$$\chi''(\omega_{\min})\propto|\varepsilon|^{1/2}\propto\omega_{\min}^{a}. \tag{9}$$

The most direct way to determine a and b from the data above the putative T_c is then to plot $\log[\chi''(\omega_{\min})]$ versus $\log[\omega_{\min}]$ and $\log[\omega_{\min}]$ versus $\log[\omega_{\max}]$; in all cases good linear plots are obtained which enable one to make a crisp determination of the parameters a and b; see Figs. 9 and 14 of Ref. [2] and Figs. 9 and 15 of Ref. [3]. The a and b so determined are indeed constant over the limited temperature range, but disagree appreciably with the requirement of Eq. (7). This can be cross-checked by looking at the consequences for the a determined as the slope of $\log[\chi''(\omega_{\min})]$ versus $\log[\omega_{\min}]$; using the slope $a=0.34$ for CKN (see Fig. 9 in Ref. [2]), one obtains, by means of Eq. (7), $\lambda=0.67$, which as seen in Fig. 6 of Ref. [2] does not lead to a good fit to the data. The same is true for Salol, studied in Ref. [3]. We believe this to be a significant contradiction of the simple mode-coupling theory of glasses.

Cummins and co-workers carried out the calculations outlined above, but they discarded the results in favor of those obtained by another (less direct) approach. At temperatures above the putative T_c they used the interpolation formula, Eq. (1), for all temperatures at which a minimum is identified. They let $\chi''(\omega_{\min})$ and $\omega_{\min}$ be freely adjustable parameters for each temperature, but constrained the fitting parameters a and b to be temperature independent. The agreement between Eq. (1) and the experimental data is far from convincing; see, for instance, the results for CKN in Fig. 6 of Ref. [2]: *at frequencies below the minimum* the agreement is poor for the lowest temperatures, not significant for the highest temperatures, and only good for one or two intermediate temperatures. Similarly, the agreement shown for Salol in Fig. 10(a) of Ref. [3] *at frequencies above the minimum* is good and significant only for the intermediate temperature $T=283$ K; on the other hand, it is good for all temperatures at frequencies below the minimum. The a and b determined in this way depend upon the frequency range over which the fit is made, and reported values are significantly different than those described above which do not obey Eq. (7).

In principle, one could test the scaling properties of the mode-coupling picture by directly examining the ω^{-b} dependence of $\chi''(\omega)$ at temperatures above T_c and at frequencies at and below $\omega_{\min}$, but well above $\omega_{\max}$, and by directly examining a within the same temperature range at frequencies at and above $\omega_{\min}$, but well below molecular frequencies. As can be seen from Figs. 5 and 6 of Ref. [2], this is not possible: b can be well determined over two decades of frequencies for Salol and indeed appears constant at temperatures above T_c; but from the same data, a cannot be determined because the frequency of the minimum and the molecular frequency are not well separated. For CKN, the reverse situation occurs: a seems to be reasonably well observed, but a direct determination of b is not possible.

For $T < T_c$

A most interesting aspect of the results reported by Cummins and co-workers is the characterization of the crossover in the $\log[\chi''(\omega)]$ curves at temperatures below the putative T_c: this predicted "knee" occurs where $\log[\chi''(\omega)]$ versus $\log[\omega]$ curves are thought to exhibit a change of slope from 1 to a. Not only does the simple MCT of glasses predict the existence of such a "knee," but it gives the "knee" frequency the critical temperature dependence $\omega_0|\varepsilon|^{1/2a}$ specified in Eqs. (5a) and (5b). The predicted and reported behavior of the "knee" frequency is startling simply because it decreases as T increases. Relaxation frequencies do not generally decrease as T is increased unless the system is approaching a critical point from below. Consequently, the behavior of the knee frequencies reported by Cummins and co-workers has been taken as strong support for the MCT of glasses. However, it appears to us, as explained below, that the data do not unambiguously substantiate the existence of such a "knee."

In treating this "knee," Cummins and co-workers rewrite Eqs. (5a) and (5b) in the scaled form

$$\frac{\chi''(\omega)}{\chi''_e}=\begin{cases}(\omega^*)^a & \omega^*>1 & \text{(10a)}\\ (\omega^*) & \omega^*<1 & \text{(10b)}\end{cases}$$

where

$$\omega^*=\frac{\omega}{\omega_0|\varepsilon|^{1/2a}}\ , \tag{11a}$$

$$\chi''_e=B\omega_0^a|\varepsilon|^{1/2}\ . \tag{11b}$$

Therefore, according to the simple MCT of glasses, below T_c the $\log[\chi''(\omega^*)/\chi''_e]$ versus $\log[\omega^*]$ curves should be universal, and by scaling all their data at $T<T_c$ Cummins and co-workers attempted to determine the "knee" frequency, $\omega_e=\omega_0|\varepsilon|^{1/2a}$, and its interesting predicted temperature dependence. But this is not what is observed in the experimental data. For Salol [Fig. 10(b) of Ref. [3]] some data points corresponding to the two lowest temperatures do lie quite well on the master curve at frequencies below $\omega^*=1$, but there are no data supporting the contention that there is a "knee" at $\omega^*=1$ or a universal straight line with constant slope a at frequencies above $\omega^*=1$. The situation seems more favorable for CKN since a small downward curvature in $\chi''(\omega)$ is indeed detectable below the molecular peak for several temperatures [cf. Fig. 2(b) of Ref. [2]]. However, the uncertainty in the determination of the "knee" frequency is very large and clouds any check of critical behavior. This ambiguity is confirmed by a different analysis of the data by Cummins *et al.* [5] (see below).

ALTERNATIVE DATA ANALYSIS

The critique given above of the analysis in terms of the simple mode-coupling theory of glasses *does not depend* upon the introduction of an alternative analysis, but some insight may be obtained by means of the following closely related analysis which is simple and rather general, but phenomenological rather than theoretical. In this analysis the α process is that which dominates between 0 and $\omega_{\min}$, the β process that which dominate between $\omega_{\min}$ and ω_{mol}. (This β process is likely to be associated with DID interactions, which is different than that detected in dielectric experiments [6(a)].) A difference between this analysis and the MCT model is that here the minimum is treated as a *crossover* between α and β, and not as part of the β process. No criticality need be assumed.

We seek the simplest non-Lorentzian functions that describe the data qualitatively at frequencies well below ω_{mol}. We describe the α process by a Cole-Davidson function [7]. The $\omega_{\max}$ is rewritten as

$$\omega_{\max}=K\tau_\alpha^{-1}\ , \tag{12a}$$

and the high-frequency wing of the α relaxation is given by

$$\chi''(\omega)=A(\omega\tau_\alpha)^{-b}\ , \tag{12b}$$

where only τ_α has a strong temperature dependence. We describe the β susceptibility at all temperatures and at frequencies well below ω_{mol} by

$$\chi''(\omega)=\begin{cases}B\omega^a \ \text{ for } \omega_{\text{mol}}\gg\omega>\tau_\beta^{-1} & \text{(13a)}\\ B\tau_\beta^{1-a}\omega \ \text{ for } \omega<\tau_\beta^{-1}\ll\omega_{\text{mol}}\ , & \text{(13b)}\end{cases}$$

where none of the parameters has a strong temperature dependence comparable to that of τ_α, but the T dependence of both B and a may, nevertheless, be important. Equations (13a) and (13b) are comparable with a more complete description which separates the non-α processes into DID and molecular components, and is valid for frequencies up to the second (molecular) maximum in $\chi''(\omega)$ [9].

The model introduced above can be used to evaluate $\chi''(\omega)$ at its minimum, i.e., at $\omega=\omega_{\min}$. Provided the minimum occurs at $\omega_{\min}\tau_\beta>1$, one finds

$$\begin{aligned}\omega_{\min}&=(Ab/Ba)^{1/(a+b)}\tau_\alpha^{-b/(a+b)}\\ &=(Ab/Ba)^{1/(a+b)}K^{-b/(a+b)}\omega_{\max}^{b/(a+b)}\ ,\end{aligned} \tag{14}$$

$$\begin{aligned}\chi''(\omega_{\min})&=(a+b)[(B/b)^b(A/a)^a]^{1/(a+b)}\tau_\alpha^{-ab/(a+b)}\\ &=(a+b)(B/b)\omega_{\min}^a\end{aligned} \tag{15}$$

while if $\omega_{\min}\tau_\beta<1$, there are slight differences.

Equations (11a)–(15) correspond exactly to the simple MCT expressions in Eqs. (1)–(5), (8), and (9) if we assume

$$\tau_\alpha^{-1}=(C/K)|\varepsilon|^{(a+b)/2ab} \tag{16}$$

and

$$\tau_\beta^{-1}=\omega_0|\varepsilon|^{1/2a} . \tag{17}$$

But, of course, we do not wish to make these assumptions.

We note that in all the relevant expressions, i.e., Eqs. (12a)–(13a), (14), and (15), there is no dependence upon τ_β, and so we expect the temperature dependence of all quantities to be dominated by the strong temperature dependence of τ_α, whether or not Eq. (17) is valid. Therefore, if above a given T_c the relaxation time τ_α is assumed to be of the form in Eq. (6), the simple MCT expressions (2)–(4), (6), (8), and (9) follow necessarily provided $\omega_{\min}\tau_\beta > 1$. More specifically if τ_α is defined by Eq. (16), *all the* ε *dependences predicted by simple MCT for* $T > T_c$ *follow necessarily and do not offer independent checks of "criticality."* Furthermore, since τ_α is given quite well by a Vogel-Fulcher function over a much wider range of temperature than that described by Eq. (16) [8], a fit to Eq. (16) over a limited temperature range by means of adjustable (C/K), T_c, and $(a+b)/2ab$ is always possible without the need for a meaningful critical T_c. Therefore *no evidence for the validity of a critical theory can be found from the* ε *dependence of the data above* T_c *alone.* The only significant check of the MCT that can be found from the data above T_c alone is the interrelationship of a and b given by Eq. (7); this interrelationship is not required by our alternate analysis, and, as discussed above, does not seem to be consonant with the data.

Below the putative T_c, the simple MCT of glasses predicts the behavior given by Eqs. (5a) and (5b); in our analysis, for frequencies well above $\omega_{\min}$, which implies that $\omega_{\min}\tau_\beta < 1$, the behavior is described by Eqs. (13a) and (13b) which can be connected to the MCT results if Eq. (17) is assumed. But, as explained above, τ_β^{-1}, or equivalently the "knee" frequency, has not been adequately determined. In fact, qualitatively it seems to us that one can also get suitable fits by taking

$$\tau_\beta \approx \text{const} , \tag{18}$$

by which we mean that the temperature dependence is weak, much weaker than the critical dependence reported [2,3] and predicted by Eq. (17). Equation (18) is somewhat unexpected because temperature-independent relaxation times that are larger than the picosecond times associated with "molecular" relaxations are not common; however, the DID relaxation of molecular liquids has not been well studied and one does not yet know what to expect. Such constancy of scaling times has also been reported by Fayer and co-workers [10].

In contrasting the simple MCT of glasses with our alternate phenomenological analysis one should note that whereas the former predicts the disappearance of both the minimum and first maximum in $\chi''(\omega)$ at temperatures below T_c, the latter retains these features at all temperatures. And indeed, as recognized in a later work by Cummins and co-workers [5], there is no reason to believe that these features disappear at temperatures above T_g.

Why should one turn to an alternative, strictly phenomenological model in lieu of the MCT of glasses for analyzing data? Because major predictions of the simple MCT, such as the interrelationship in Eq. (7) and the critical behavior of τ_β, do not seem to be valid. Because the alternate model provides a simple, two-process picture for describing the behavior both below and above the spectral minimum, and does not require the introduction of additional "hopping" mechanisms to avoid a nonphysical predicted catastrophe at T_c. Because the alternate model does not incorporate critical behavior which is, in fact, not observed in the unmassaged data; critical behavior should be evident without preprogramming into the analysis. Because it can be used to exhibit those interrelationships that have been interpreted as reinforcing the case for MCT but actually occur quite independently of the MCT. Because it arguably requires fewer adjustable parameters (or at least no more) than does the MCT. Because it suggests to researchers that the search for an adequate theory of supercooled liquids and glasses is far from over. The simple MCT of glasses *does* predict, as observed, non-Lorentzian spectra, i.e., ω^a and ω^{-b} dependences, which in our alternate model is incorporated phenomenologically by Cole-Davidson related functions, but with no particular requirement that the exponents be temperature independent.

DISCUSSION OF RECENT WORK

In a recent work, Cummins *et al.* [5] recognize that constant exponents a and b and constant amplitudes A and B do not describe the observed power-law behavior of $\chi''(\omega)$ over a wide range of frequencies, and that the "knee" frequency is hard to determine. They have reanalyzed their light-scattering data in Refs. [2,3] in terms of an extended or modified MCT of glasses. In this modified version of the MCT [5,11], the critical features are attenuated due to the introduction of hopping as a relaxation mechanism. Although, in the simple theory, the α relaxation becomes nonergodic below T_c, in the modified MCT the hopping modes are then said to take over. At temperatures below T_c the hopping modes preserve both the maximum and minimum in $\chi''(\omega)$. The hopping parameter $\delta(T)$ is introduced as a temperature-dependent adjustable parameter. According to the modified MCT, the simple critical relationships are masked by the hopping processes; the only critical temperature dependence (i.e., a dependence of the form $|T-T_c|^\nu$) that may still be clearly exhibited by the data is that of Eq. (4) at temperatures above T_c. The simple scaling features of the theory, i.e., the ω^a, ω^1, and ω^{-b} dependences of $\chi''(\omega)$ are compromised by the hopping mechanisms, as are the critical temperature dependences of the "knee" frequency.

The introduction of additional temperature-dependent parameters allows for better fits to the data, but at the cost of the introduction of two strongly, but noncritical, temperature-dependent adjustable parameters, $\delta(T)$ and an amplitude $h(T)$. This poses a problem in the use of the data to validate the MCT of glasses because the central feature of such a theory is the critical temperature dependence around T_c, and this feature can be discerned only if it dominates all regular temperature dependences, which it does not. Although the *modified* MCT of glasses can provide better fits than the simple theory, the fits yield hopping contributions that are so large that they greatly weaken the significance of the fits as a check of the critical nature of the theory; in fact, it greatly reduces the significance of the *simple* MCT as a zeroth-order picture of supercooled liquid-glass behavior.

Turning to the β relaxation at temperatures below the putative T_c, we note a major change in the more recent analysis [5] from that in previous articles on the subject [1–3]. In Refs. [2,3] the striking cusplike behavior around T_c of the relaxation frequency ω_e, the same T_c used to fit the $\omega_{\max}$ and $\omega_{\min}$ at temperatures above T_c, gave strong testimony to the applicability of the simple MCT of glasses. In particular, the marked increase of ω_e as $T \rightarrow T_c$ from below seemed to give strong support to the critical nature of the theory. These values of ω_e at temperatures below T_c were associated with the "knee" frequency. In their more recent article Cummins *et al.* [5] cannot identify a "knee" for Salol; for CKN, the "knee" is not clearly discernible directly from the susceptibility data, and they determine it by identifying a maximum in $\chi''(\omega)/\omega^{1/2}$; this maximum has been roughly identified only over a small temperature range. The fact that in Ref. [2] the "knee" frequency of CKN, which is proportional to ω_e, shifts by a factor of 13 between 353 and 296 K (and a factor of 70 over the entire temperature range studied), whereas in Ref. [5] it shifts by a mere factor of 3 over the same 353–296 K range (over only part of which a "knee" can truly be discerned), suggests that the "knee" frequency is too elusive to play a major role in the validation of the theory.

Despite the uncertainties in evaluating the "knee" frequency, the fact that in all the analyses of the CKN data below the putative T_c this frequency appears to increase slightly with decreasing temperature suggests the rather unusual situation, often associated with a critical phenomenon, of a relaxation frequency increasing with decreasing temperature. However, the uncertainty and smallness of its temperature variation compared with the $|T-T_c|^{1/2a}$ prediction, the fact that the relatively intense "leaked" Brillouin lines (also with increasing frequency as T decreases) appear at nearby frequencies, and *the fact that such variations are found even if* τ_β *is constant, provided the exponent a increases slightly* (as expected and observed) *with decreasing temperature* [12], all tend to diminish the importance of the "knee" frequency analysis as evidence of critical behavior.

Taken merely as a fitting scheme, the MCT analysis requires many adjustable parameters. The simple MCT analysis requires at least eight such temperature-independent parameters to describe the spectrum: a, T_c, $\omega_{\min}/|\varepsilon|^{1/2a}$, $\chi''(\omega_{\min})/|\varepsilon|^{1/2}$, ω_0 in Eq. (11), $\chi''(\omega_e)/|\varepsilon|^{1/2}$, C in Eq. (4), and a parameter related to the amplitude A of the α peak; this count assumes a relationship between b and the exponent of the Williams-Watts function; it also assumes that Eq. (7) holds. Of course, at T's below T_c this description fails even qualitatively to describe $\chi''(\omega)$ at its minimum and below, and at all T's it fails to describe the molecular peak. In the modified MCT, at least four more temperature-independent adjustable parameters are needed: the amplitude of the separation parameter σ/ε, the two parameters needed to specify the Arrhenius expression for the hopping parameter $\delta(T)$, and a parameter indicating the dependence of the critical amplitude $h(T)$ upon temperature (although a temperature-independent h is incorporated into the parameters associated with the simple MCT theory, the significant temperature dependence of h has only been included in the analysis in Ref. [5] which makes use of the modified MCT of glasses). The modified MCT does not describe $\chi''(\omega)$ in the regime of the molecular maximum.

The modified MCT of glasses also predicts a sharp change in the temperature behavior of $\omega_{\max}$ and $\omega_{\min}$ below T_c because of a change from density-related modes to a hopping mechanism. The data presented [5] do not report $\omega_{\min}$ and $\omega_{\max}$ over a sufficiently wide temperature range below T_c to answer this last question, and after many years controversy still rages over the question of whether the viscosity (which is inversely proportional to $\omega_{\max}$) sharply changes its behavior at some temperature above T_g, or whether simple functions, such as the Vogel-Fulcher function, are merely inadequate to describe 14 orders of magnitude change in a quantity.

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[12] For example, if the β process well below ω_{mol} is described by $\chi''(\omega)=\mathrm{Re}[\omega\tau_\beta B(1-i\omega\tau_\beta)^{-1+a}]$, the maximum in $\chi''(\omega)/\omega^{1/2}$ shifts to higher ω as a increases, which is what one would expect as T is lowered. Alternatively, if $\chi''(\omega)=B(1-i\omega\tau_\beta)^{a-1}+C(1-i\omega\tau_{\mathrm{mol}})^{-1}$, where $C\geq 10B$, $\tau_{\mathrm{mol}}\ll\tau_\beta$, and only B varies with T, one finds that as B decreases (which is what is observed as T is lowered) the maximum in $\chi''(\omega)/\omega^{1/2}$ shifts to higher ω [9].