Inhomogeneous Mode-Coupling Theory and Growing Dynamic Length in Supercooled Liquids

Giulio Biroli,[1] Jean-Philippe Bouchaud,[2,3] Kunimasa Miyazaki,[4] and David R. Reichman[4]

[1]*Service de Physique Théorique, Orme des Merisiers-CEA Saclay, 91191 Gif sur Yvette Cedex, France*

[2]*Service de Physique de l'État Condensé, Orme des Merisiers-CEA Saclay, 91191 Gif sur Yvette Cedex, France*

[3]*Science and Finance, Capital Fund Management, 6 Bd Haussmann, 75009 Paris, France*

[4]*Department of Chemistry, Columbia University, 3000 Broadway, New York, New York 10027, USA*

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We extend mode-coupling theory (MCT) to inhomogeneous situations, relevant for supercooled liquid in an external field. We compute the response of the dynamical structure factor to a static inhomogeneous external potential and provide the first direct evidence that the standard formulation of MCT is associated with a diverging length scale. We find that the so-called cages are, in fact, extended objects. Although close to the transition the dynamic length grows as $|T - T_c|^{-1/4}$ in *both* the β and α regimes, our results suggest that the fractal dimension of correlated clusters is larger in the α regime. We derive inhomogeneous MCT equations valid to second order in gradients.

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It is becoming increasingly clear that the viscous slowing down of supercooled liquids, jammed colloids, and granular assemblies is accompanied by a growing dynamic length scale, whereas all static correlation functions remain short-ranged. This somewhat unusual scenario, suggested by the experimental discovery of strong dynamical heterogeneities in glass formers [1], has been substantiated by detailed numerical simulations [2–6], explicit solution of simplified models [7,8], and recent experiments [9,10] where 4-point spatiotemporal correlators are measured. From a theoretical point of view, our understanding of supercooled liquids owes much to the mode-coupling theory (MCT) of the glass transition. Although approximate in nature, MCT has achieved many qualitative and quantitative successes in explaining various experimental and numerical results [11,12]. Despite early insights [13], the freezing predicted by MCT is often argued to be a local caging phenomenon, without any diverging collective length scale. This, however, is rather surprising, since one expects on general grounds that diverging relaxation times involve an infinite number of particles in the absence of quenched disorder, defects, or fixed obstacles [14]. Building upon the important work of Franz and Parisi [15], two of us (BB) [16] suggested a way to reconcile MCT with physical intuition. Within a field theoretic formulation of MCT, BB showed that the 4-point dynamic density correlation function plays the role of 2-point static correlations in standard phase transitions and is characterized by a diverging dynamical correlation length and spatiotemporal scaling laws. BB also proposed a Ginzburg criterion that delineates the region of validity of MCT, which breaks down in low dimensions. Still, the field theory language used in Ref. [16] is not trivially related to the standard, liquid theory formulation of MCT [11]. Indeed, recent work has shown that the field theory is laden with subtleties [17–19], in particular, related to the fluctuation-dissipation relation. The aim of the present Letter is twofold. First, we show how the results of BB may be recovered, corrected, and extended to obtain testable, quantitative predictions on absolute dynamic length scales, entirely within the standard, projection-operator-based MCT [11]. Our analysis predicts a remarkable scaling behavior that has implications for the geometry of dynamic heterogeneities. Second, our formulation generalizes MCT to spatially inhomogeneous situations, of importance in a wide variety of physically interesting situations that include the study of confined fluids [20] and the influence of gravity on the dynamics of glassy colloidal suspensions [21].

In order to proceed, we consider a fluid subject to an arbitrary external potential $U(\mathbf{x})$, such that the equilibrium averages [e.g., the static density $\rho(\mathbf{x})$] vary in space. The relationship with the results of BB will be obtained using this inhomogeneous MCT formalism to compute the response of the dynamical structure factor to a localized external potential. In the limit where the wave vector $\mathbf{q}$ associated with the external field tends to zero, a connection to the 4-point correlator of BB emerges. The dynamical quantities of interest are the density fluctuations $\delta\rho$ and the currents J, defined in Fourier space as $\delta\rho_{\mathbf{k}} = \sum_{i=1}^N e^{i\mathbf{k}\cdot\mathbf{r}_i} - \langle\rho_{\mathbf{k}}\rangle$ and $J_{\mathbf{k}} = \sum_{i=1}^N \hat{\mathbf{k}}\cdot\mathbf{p}_i e^{i\mathbf{k}\cdot\mathbf{r}_i}/m$. Following, step by step, standard procedures based on the Mori-Zwanzig formalism [11,22], one can establish the following *exact* equation of motion for the dynamic structure factor $F(\mathbf{k}_1, \mathbf{k}_2; t) = 1/N\langle\delta\rho_{\mathbf{k}_1}(t)\delta\rho^*_{\mathbf{k}_2}(0)\rangle$:

$$\frac{\partial^2}{\partial t^2}F(\mathbf{k}_1, \mathbf{k}_2; t) + \int d\mathbf{k}_1' \Omega^2(\mathbf{k}_1, \mathbf{k}_1')F(\mathbf{k}_1', \mathbf{k}_2; t) + \int d\mathbf{k}_1' \int_0^t dt' M(\mathbf{k}_1, \mathbf{k}_1'; t-t')\frac{\partial}{\partial t'}F(\mathbf{k}_1', \mathbf{k}_2; t') = 0, \quad (1)$$

where $\Omega^2(\mathbf{k}_1, \mathbf{k}_1') \equiv (k_BT/m)(\mathbf{k}_1\cdot\mathbf{k}_1')\langle\rho_{\mathbf{k}_1-\mathbf{k}_1'}\rangle S^{-1}(\mathbf{k}_1, \mathbf{k}_1')$ [with $S^{-1}(\mathbf{k}_1, \mathbf{k}_2)$ the inverse of $F(\mathbf{k}_1, \mathbf{k}_2, t=0)$], and $M(\mathbf{k}_1, \mathbf{k}_2; t)$ is a memory kernel which can be expressed in terms of the fluctuating force. Performing the same factorization approximations used to derive standard

MCT [11,22], but without assuming translation invariance, we reduce this memory kernel to two-body correlation functions and obtain an inhomogeneous mode-coupling theory (IMCT). The corresponding IMCT equations are rather cumbersome (although straightforward to derive) and will be presented elsewhere [23]. By construction, in the limit $U(\mathbf{x}) \to 0$, they reduce to standard MCT.

In order to obtain somewhat tractable expressions, one can consider *weakly* inhomogeneous situations $U(\mathbf{x}) \ll k_B T$, such that one can expand all quantities to first order in $U/k_B T$. The aim is to compute the sensitivity of the dynamical structure factor to a small perturbation of arbitrary spatial structure, in particular, localized perturbations, which we can always decompose in Fourier modes: $[\delta F(\mathbf{x}, \mathbf{y}, t)/\delta U(\mathbf{z})]|_{U=0} = \int d\mathbf{k} d\mathbf{q} e^{-i\mathbf{k}\cdot(\mathbf{x}-\mathbf{y})+i\mathbf{q}\cdot(\mathbf{y}-\mathbf{z})} \times \chi_{\mathbf{q}}(\mathbf{k}, t)$, where $\chi_{\mathbf{q}}(\mathbf{k}, t) \propto [\delta F(\mathbf{k}, \mathbf{q} + \mathbf{k}, t)/\delta U(\mathbf{q})]|_{U=0}$ is the response of the dynamical structure factor to a static external perturbation in Fourier space. For a localized perturbation $U(\mathbf{x}) = U_0 \delta(\mathbf{x})$, one finds $\delta F(\mathbf{k}, \mathbf{y}, t) = U_0 \int d\mathbf{q} e^{i\mathbf{q}\cdot\mathbf{y}} \chi_{\mathbf{q}}(\mathbf{k}, t)$ [24]. This susceptibility is related to a 3-point density correlation function in the absence of the perturbation. Although different from the 4-point functions considered previously in the literature, $\chi_{\mathbf{q}}(\mathbf{k}, t)$ is expected to reveal the existence of a dynamical correlation length of the homogeneous liquid (see [10,25] for the particular case $\mathbf{q} = 0$) and to have a similar critical behavior. The physical reason is that *spontaneous* dynamical fluctuations measured by the 4-point function and *induced* dynamical fluctuations measured by $\chi_{\mathbf{q}}(\mathbf{k}, t)$ are intimately related. Intuitively, speeding up or slowing down the dynamics at one given point (i.e., by changing the local density) should perturb the dynamics on a length scale ξ over which spontaneous dynamical fluctuations themselves are correlated. As shown below, this is indeed the case within MCT. The formal reason is that, precisely as for standard phase transitions, a certain linear operator (the susceptibility) that governs both the correlation and the response of the system becomes critical at the transition (see [25] for a diagrammatic interpretation). Differentiating Eq. (1) with respect to $U(\mathbf{q})$ and then setting $U(\mathbf{q}) = 0$, the final equation for the susceptibility $\chi_{\mathbf{q}}(\mathbf{k}, t)$ reads:

$$\frac{\partial^2 \chi_{\mathbf{q}}(\mathbf{k}, t)}{\partial t^2} + \frac{k_B T k^2}{m S(k)} \chi_{\mathbf{q}}(\mathbf{k}, t) + \int_0^t dt' M_0(k, t-t') \frac{\partial \chi_{\mathbf{q}}(\mathbf{k}, t')}{\partial t'} + \int_0^t dt' \frac{k_B T \rho k}{m|\mathbf{k}+\mathbf{q}|} \times \int \frac{d\mathbf{k}'}{(2\pi)^3} v_{\mathbf{k}}(\mathbf{k}', \mathbf{k}-\mathbf{k}') v_{\mathbf{k}+\mathbf{q}}(\mathbf{k}-\mathbf{k}', \mathbf{q}+\mathbf{k}') \chi_{\mathbf{q}}(\mathbf{k}', t-t') F_0(|\mathbf{k}-\mathbf{k}'|, t-t') \frac{\partial F_0(|\mathbf{k}+\mathbf{q}|, t')}{\partial t'} = \mathcal{S}_{\mathbf{q}}(\mathbf{k}, t), \quad (2)$$

where $F_0(k, t)$ is the usual MCT solution for $U = 0$, $v_{\mathbf{k}}(\mathbf{k}_1, \mathbf{k}_2) \equiv \hat{\mathbf{k}} \cdot \mathbf{k}_1 c(k_1) + \hat{\mathbf{k}} \cdot \mathbf{k}_2 c(k_2)$ and $M_0(k, t) = k_B T \rho / 2m \int d\mathbf{k}'/(2\pi)^3 v_{\mathbf{k}}^2(\mathbf{k}', \mathbf{k}-\mathbf{k}') F_0(k', t) F_0(|\mathbf{k}' - \mathbf{k}|, t)$ are the standard MCT vertex and memory kernel, respectively [22]. The source term $\mathcal{S}_{\mathbf{q}}(\mathbf{k}, t)$, whose precise form will be presented elsewhere [23], depends on $F_0(k, t' \le t)$ and static correlation functions; the value of the dynamic length scale and the critical properties of $\chi_{\mathbf{q}}$ are, however, independent of the precise form of this source term. The above equation is of the type $\mathcal{L}_{\mathbf{q}} \chi_{\mathbf{q}} = \mathcal{S}_{\mathbf{q}}$, where $\mathcal{L}_{\mathbf{q}}$ is a linear operator, the structure of which, in particular, its smallest eigenvalue, contains the information we want to extract. One should first note that, in the limit $\mathbf{q} = 0$, the operator $\mathcal{L}_0$ simply encodes the change of the MCT dynamic structure factor when the coupling constant (i.e., the density or the temperature) is shifted uniformly in space. This remark allows one to compute $\chi_0(\mathbf{k}, t)$ from standard MCT results in the β and α regimes:

$$\chi_0(\mathbf{k}, t) = \begin{cases} \frac{S(k)h(k)}{\sqrt{\varepsilon}} g_\beta(q^2 = 0, \frac{t}{\tau_\beta}) & \tau_\beta = \varepsilon^{-1/2a}, \\ \frac{1}{\varepsilon} g_{\alpha,k}(\frac{t}{\tau_\alpha}) & \tau_\alpha = \varepsilon^{-1/2a-1/2b}. \end{cases} \quad (3)$$

In the above, $h(k)$, a, and b, are standard MCT notations describing the solution of MCT equations at $U = 0$ [11,22] and ε is the distance from the MCT critical point T_c. The behavior of the scaling functions at large and small arguments can be found directly by analyzing Eq. (2) or by scaling: In the early β regime $u = t/\tau_\beta \to 0$, the ε dependence should drop out; hence, $g_\beta(0, u) \propto u^a$. The matching between α and β regimes implies $g_\beta(0, u) \propto u^b$ at large u, whereas $g_{\alpha,k}(u) \propto S(k)h(k)u^b$ at small u. How are these results affected when $\mathbf{q} \neq 0$? The analysis is simple for $\chi_{\mathbf{q}}(\mathbf{k}, \infty)$ in the glass phase, where straightforward manipulations of Eq. (2) allow one to show that it satisfies the matrix equation $(I - M_{\mathbf{q}}) \cdot \chi_{\mathbf{q}}(\mathbf{k}, \infty) = \mathcal{S}_{\mathbf{q}}^\infty$ with a source term that is regular and of order one in the limit $q \to 0$ and:

$$M_{\mathbf{q}}(\mathbf{k}_1, \mathbf{k}_2) = \rho \frac{G(\mathbf{k}_1) G(\mathbf{q}+\mathbf{k}_1) S(\mathbf{k}_1 - \mathbf{k}_2)}{(2\pi)^3 k_1 |\mathbf{k}_1 + \mathbf{q}|} f_{\mathbf{k}_1 - \mathbf{k}_2} \times v_{\mathbf{k}_1}(\mathbf{k}_2, \mathbf{k}_1 - \mathbf{k}_2) v_{\mathbf{k}_1+\mathbf{q}}(\mathbf{k}_1 - \mathbf{k}_2, \mathbf{q} + \mathbf{k}_2), \quad (4)$$

where $G(\mathbf{k}_1) = S(\mathbf{k}_1)(1 - f_{\mathbf{k}_1})$ and $f_{\mathbf{k}}$ is the nonergodic parameter. Interestingly, the matrix $M_{\mathbf{q}}$ is *exactly* the same as the one obtained from the resummation of the ladder diagrams in the field theoretical framework of BB. Note also that the source $\mathcal{S}_{\mathbf{q}}^\infty$ is irrelevant provided it is not orthogonal to the lowest eigenvector of $M_{\mathbf{q}}$. For $q = 0$ and $\varepsilon < 0$, the largest eigenvalue of M_0 was shown by Götze to be $\lambda = 1 - O(\sqrt{-\varepsilon})$ [26] and its right eigenvector is $S(k)h(k)$. The correction $\delta\lambda$ to this eigenvalue at $q \to 0$ can be computed by perturbation theory. By symmetry, one expects that, in general, $\delta\lambda = -\Gamma q^2$, where Γ is a certain coefficient, leading to $\chi_{\mathbf{q}}(\mathbf{k}, \infty) \sim S(k)h(k)/(\sqrt{|\varepsilon|} + \Gamma q^2)$ [16]. In the schematic limit where $S(k)$ is sharply peaked around $k = k_0$, with a small width ΔK, one can compute Γ exactly; one finds that Γ is positive and $\propto$

ΔK^{-2}. We have performed a microscopic calculation that includes the full $S(k)$ and have determined numerically that for the hard-sphere structure factor computed within the Percus-Yevick approximation at the MCT critical density $\phi_c = 0.515$, $\Gamma = 0.072\sigma^2$. For more realistic hard-sphere structure factors, Γ may be as large as $\Gamma = 0.3\sigma^2$. We have not been able to show in full generality that Γ should always be positive. A negative Γ would predict a remarkable "modulated" glass transition, with the nonergodic factor displaying periodic oscillations in space [27].

The analysis of the full temporal behavior is more involved. The operator that becomes critical at the transition again turns out to be the same as the one considered in BB. We find that $(I - M_c)\chi_{\mathbf{q}}(t\varepsilon^{1/2a}) = s_{\mathbf{q}}(t\varepsilon^{1/2a})$, where M_c is the matrix M at the transition and the new source term is of order one in the limit $q \to 0$. As a consequence, one finds [23]:

$$\chi_{\mathbf{q}}(\mathbf{k}, t) = \frac{1}{\sqrt{\varepsilon} + \Gamma q^2} S(k)h(k)g_\beta\left(\frac{\Gamma q^2}{\sqrt{\varepsilon}}, t\varepsilon^{1/2a}\right), \tag{5}$$

where $q^2 g_\beta(\Gamma q^2/\sqrt{\varepsilon}, t\varepsilon^{1/2a}) = (\sqrt{\varepsilon} + \Gamma q^2)\langle l|s'_{\mathbf{q}}(t\varepsilon^{1/2a})\rangle$, where $\langle l|$ is the left eigenvector conjugated to $S(k)h(k)$. The analysis of $\chi_{\mathbf{q}}(\mathbf{k}, t)$ in the α-relaxation regime is subtle and will be detailed elsewhere [23]. The results turn out to be different from the naive guess presented in BB. We have established that, for small q and fixed ε,

$$\chi_{\mathbf{q}}(\mathbf{k}, t) = \frac{\Xi(\Gamma q^2/\sqrt{\varepsilon})}{\sqrt{\varepsilon}(\sqrt{\varepsilon} + \Gamma q^2)} g_{\alpha,k}\left(\frac{t}{\tau_\alpha}\right), \tag{6}$$

with Ξ a certain regular function with $\Xi(0) \neq 0$ and $\Xi(v \gg 1) \sim 1/v$ such that $\chi_{\mathbf{q}}$ behaves as q^{-4} for large $q\epsilon^{-1/4}$, independently of ε. Also, $g_{\alpha,k}(u \ll 1) = S(k)h(k)u^b$, as to match the β regime, and $g_\alpha(u \gg 1, k) \to 0$. In order to confirm the above analytical predictions, we performed a numerical integration of both the standard MCT equation and Eq. (2) in the schematic Leutheusser approximation [28]. We neglected all k dependence, while keeping q dependence to lowest order [24]; replacing $F_0(k, t) \to F(t)$, $\chi_{\mathbf{q}}(k, t) \to \chi_{\mathbf{q}}(t)$, $M_0(k, t) \to 4\lambda F^2(t)$, $\mathcal{S}_{\mathbf{q}}(\mathbf{k}, t) \to F(t)$, and, finally, the memory kernel in the fourth term of Eq. (2) by $8\lambda(1 - q^2)\chi_q(t)F(t)$. The result is shown in Fig. 1. Note that the scaling variable is still $q^2\varepsilon^{-1/2}$ in the α regime, rather than $q^2\varepsilon^{-1}$ as surmised in BB [16]. The physical consequence of the above analysis is the existence of a *unique* diverging dynamic correlation length $\xi \sim \sqrt{\Gamma}|\varepsilon|^{-1/4}$ that rules the response of the system to a space-dependent perturbation. The analysis of the early β regime where $t \ll |\varepsilon|^{-1/2a}$ shows that this length, in fact, first increases as $t^{a/2}$ and then saturates at ξ. Furthermore, Eq. (6) indicates that, although the integrated dynamic correlation $\chi_{\mathbf{q}=0}(\mathbf{k}, t)$ increases in the α regime as $\varepsilon^{(b-a)/2a}t^b$ (from $\varepsilon^{-1/2}$ for $t = \tau_\beta$ to ε^{-1} for $t = \tau_\alpha$), the dynamic length scale itself remains fixed at ξ. Interestingly, this suggests that, while keeping a fixed extension ξ, the (fractal) geometrical structures carrying the dynamic correlations significantly "fatten" [29] between τ_β (where the structures could correspond to the strings reported in recent simulations [5,30]) and τ_α, where more compact structures are expected, as indeed suggested by the results of Ref. [31]. For $\tau_\beta \ll t \ll \tau_\alpha$, we expect a crossover between dense and dilute structures at a new, time-dependent crossover length [23].

Starting from the general IMCT equation (1), one could have chosen to follow a slightly different path and only assume that the length scale ℓ of the imposed inhomogeneities is large. Performing a gradient expansion to order ℓ^{-2}, one obtains an equation on the space-dependent structure factor $F(\mathbf{k}, \mathbf{r}, t)$. This space-dependent Ginzburg-Landau-like MCT equation has one part identical to the standard MCT equation (with space-dependent coefficients) plus nonlinear contributions (see [23]) containing a $\nabla^2 F$ term and, interestingly, a Burgers nonlinear term $(\nabla F)^2$ [32]. When inhomogeneities are small, one recovers Eq. (2) above.

In summary, we have extended the standard framework of MCT to inherently inhomogeneous physical situations. This allowed us to compute the response of the dynamical structure factor to spatial perturbations. The case of a localized perturbation shows directly that the dynamical structure factor is affected on a dynamic length scale ξ that diverges $(T - T_c)^{-\nu}$ as T_c is approached. Note that ξ, as in ordinary critical phenomena, diverges only at T_c, reflecting the critical fragility of the system right at the transition. It is therefore clearly distinct from the diverging viscous length $\sqrt{\eta\tau_\alpha}$ that sets the scale below which the liquid sustains shear waves [33], which is infinite in the whole glass phase.

Our most striking predictions are that (i) multipoint functions violate Ornstein-Zernicke scaling in the α regime and, instead, have a much stronger q dependence, encoded in Eq. (6), and (ii) the dynamical length scale ξ has a rather modest growth with thermodynamic control variables that is reflected in the small value of the exponent $\nu = 1/4$, different from $\nu = 1/2$ surmised in BB [34]. This slow growth is in harmony with our current experimental understanding of the magnitude of cooperative length scales at the glass transition. It should be also remarked that our work predicts that ξ governs both the β and α relaxation regimes, showing that the standard interpretation of the β regime as the vibrations of particles trapped in independent cages formed by nearest neighbors is somewhat misleading: As the MCT transition is approached, ξ grows and the "cages" become more and more collective. Between τ_β and τ_α, the dynamical length does not grow, but the geometrical structures responsible for dynamic fluctuations thicken with time [29], and this leads to the increase of $\chi_{\mathbf{q}}(\mathbf{k}, t)$. The detailed and novel predictions outlined above may be tested by molecular dynamics simulations, provided system sizes are large enough to explore the full scaling window. Experimentally, $\chi_{\mathbf{q}}(\mathbf{k}, t)$ could be accessible in colloids by use of an optical tweezer array, imposing a periodic dielectric force on the particles.

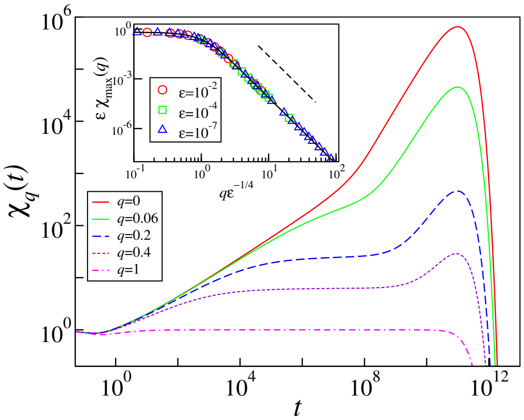

FIG. 1 (color online). Numerical solution of the schematic IMCT equations for $T > T_c$ (see [23]). Main plot: $\chi_{\mathbf{q}}(t)$ for different $\mathbf{q}$ as a function of time and for $\varepsilon = 10^{-6}$. From top to bottom: $q = 0$, 0.06, 0.2, 0.4, and 1. Note that the *shape* of $\chi_{\mathbf{q}}(t)$ in the α regime is independent of $\mathbf{q}$, as predicted by Eq. (6). We have, in fact, checked that the predicted scaling is very well obeyed in that region. Inset: $\varepsilon\chi_{\max}(\mathbf{q}) \equiv \varepsilon\chi_{\mathbf{q}}(t = \tau_\alpha)$ as a function of $q\varepsilon^{-1/4}$ in log-log, for different q's and ε's. Note the q^{-4} behavior for large $q\varepsilon^{-1/4}$, as indicated by the dashed line.

As argued in the introduction, a diverging length scale at the MCT transition is expected on general grounds; although not obvious at first sight, the MCT transition is akin to a standard phase transition, except that the order parameter is itself a 2-point correlation function, and, therefore, susceptibilities and correlation functions displaying critical behavior are 3- and 4-point objects. Physically, MCT can be interpreted as describing the appearance of marginally metastable states that slow down the dynamics. These states are characterized by soft modes that involve a diverging number of particles moving in correlated clusters. The physical idea put forward here is that the *intrinsic* size and characteristics of such clusters may be probed by an external potential that pins the particles over an *extrinsic* length scale, which may be freely and independently tuned.

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