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A theory for a certain crossover in relaxation phenomena in glasses

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Abstract. It is shown that the simplified mode coupling theory for supercooled liquid dynamics allows for a scenario where a first component of the system is within an ideal glass state while the second component exhibits a crossover from an ideal glass state to a state of almost liquid behaviour. The dynamics near the crossover is governed by two relevant control parameters and it can be described by a scaling law. There appear fractal spectra. Under certain simplifying assumptions or symmetry conditions the crossover becomes an ergodic to non-ergodic transition, where the Edwards–Anderson parameter of the second component changes continuously from zero to a non-zero value.

1. Introduction

A glass is a state of condensed matter, where the particles are arrested in a spontaneously frozen disordered array. Conventionally one assumes that in an ideal glass the motion of all particles is non-ergodic. However, one can also imagine a system of two components so that one is in a non-ergodic glass state while the other exhibits liquid-like dynamics. Such a situation is anticipated for disordered crystals. In that case the disordered matrix, produced for example by impurities, can be viewed as a glassy state of one component. Other modes, such as for example the density fluctuations of electrons, may exhibit liquid flow. In this paper the scenario for a liquid-to-glass transition of one component of a many particle system occurring in a glassy matrix shall be analysed within mode coupling theory.

Let us assume that the dynamics of a classical many particle system is described by a set of M conventionally defined autocorrelation functions $\Phi_q(t)$, $q = 1, \dots, M$; $\Phi_q(0) = 1$, $\dot{\Phi}_q(0) = 0$. If the system is in an ergodic liquid state, the correlation functions decay to zero for large times. However, if it is in an ideal glass state, there is a spontaneous arrest of some fluctuations, i.e. $\Phi_q(t \rightarrow \infty) = f_q$ with $f_q > 0$ for some q (Edwards and Anderson 1975). Suppose one varies some control parameter x , such as the density, the composition or the inverse temperature. An ideal liquid-to-glass transition is said to occur at some critical point x_c if, for $x < x_c$, all long-time limits f_q vanish, while for $x > x_c$ an ideal glass state is found. Mode coupling theory (MCT) describes such liquid-to-glass transitions; for a review, see Götze and Sjögren (1992). The basic version of the MCT, to which the following discussion shall be restricted, deals with the set of equations of motion:

$$\ddot{\Phi}_q(t) + \nu_q \dot{\Phi}_q(t) + \Omega_q^2 \Phi_q(t) + \Omega_q^2 \int_0^t m_q(t-t') \dot{\Phi}_q(t') dt' = 0 \quad (1.1)$$

$$m_q(t) = \mathcal{F}_q(\Phi(t)). \quad (1.2)$$

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Here $\Omega_q > 0$, $v_q \geq 0$ are scales for the short-time transient dynamics. The mode coupling functional \mathcal{F} is an absolutely monotonic function; for simple liquids it is a second-order polynomial (Bengtzelius *et al* 1984):

$$\mathcal{F}_q(f) = \frac{1}{2} \sum_{kp} V(q, kp) f_k f_p \quad V(q, kp) \geq 0. \quad (1.3)$$

The vertices $V(q, kp)$ are the coupling constants of the theory; they are combined to a vector V in the N -dimensional control parameter space \mathbb{R} . In applications the system moves on a smooth path $V(x)$, traversed upon changing x . The long-time limit f , called the Edwards–Anderson parameter, the non-ergodicity parameter or the Debye–Waller factor, obeys the set of M implicit equations (Bengtzelius *et al* 1984)

$$f_q/(1 - f_q) = \mathcal{F}_q(V, f) \quad 0 \leq f_q < 1. \quad (1.4a)$$

Usually these equations have many solutions for given V . The long-time limit is the maximum solution in the following sense: if f' obeys (1.4a) one gets (Götze 1991)

$$f'_q \leq f_q \quad q = 1, \dots, M. \quad (1.4b)$$

Thus, glass transitions are bifurcations of (1.4), occurring at some critical points for the control parameters $V^c = V(x_c)$. There the long-time limit f exhibits a singularity if considered as function of V . The V^c are therefore called glass transition singularities (GTS). For $V \neq V^c$, the long-time limits f_q are smooth functions of V . The MCT deals with the dynamics near the bifurcation points V^c .

If x crosses x_c the non-ergodicity parameter f can either change continuously (type-A transition) or discontinuously (type-B transition). The simple $M = 1$ model with $\mathcal{F}(f) = v_1 f + v_2 f^2$ exhibits both types (Götze 1984). The quadratic polynomial (1.3) brings out generically only type-B transitions and the results for that case have been compared in detail with experiments during the past years. From an analysis of light scattering for the glass former $\text{Ca}(\text{NO}_3)_2\text{K}(\text{NO}_3)$ (Li *et al* 1992, Cummins *et al* 1993) and other simple systems, one concludes that the MCT type-B scenario describes a number of experimental features in supercooled liquids adequately; for a summary see Cummins *et al* (1994). Studies of the glass transition in hard sphere colloids with photon correlation spectroscopy confirmed MCT predictions to a 15% accuracy level (Van Meegen and Underwood 1993). Type-A transitions can occur generically only if the mode coupling functional \mathcal{F}_q contains linear terms. Such terms arise if one considers interactions of the particles with a static random potential as it may be provided by an arrested array of defects or impurities. MCT results for such transitions have been tested against computer experiments done for the Lorenz model (Götze *et al* 1981).

A static array of impurities can be viewed as a glass state for the impurity particles. Thus there appears the question: under which conditions can (1.1)–(1.3) imply a glass state, within which a type-A transition occurs? It shall be shown that the MCT with quadratic functional (1.3) can yield a glass state, where V is close to a type-A GTS. The long-time dynamics for such states shall be worked out. In this paper the equations (1.1)–(1.3) shall be considered as a mathematical model; its justification within a microscopic theory shall not be considered. It is the aim to provide a scenario for a possible crossover for the dynamics of superionic glasses, amorphous semiconductors, mixed crystals and the like.

Sjögren (1986) considered an $M = 2$ model with $\mathcal{F}_1(f) = v f_1^2$, $\mathcal{F}_2 = v_s f_1 f_2$. Within this model there is no effect of the second variable on the first. There is a type-B transition

where f_1 jumps from zero to $1/2$ if v shifts through $v_c = 4$. The second variable is zero if $\sigma = v_1 f_1 - 1$ is negative, and then $f_2 = \sigma + \mathcal{O}(\sigma^2)$ for $\sigma \geq 0$. Quite the same type-A transition occurs for an $M = 2$ model with $\mathcal{F}_1(f) = v_1 f_1^2 + v_2 f_2^2$, $\mathcal{F}_2(f) = v_s f_1 f_2$ (Krieger and Bosse 1987, Götze and Hausmann 1988). For both models the special forms for the mode coupling functionals was motivated by physical reasoning. In the first case Φ_1 referred to coherent density fluctuations while Φ_2 described the density of a single tagged particle. The second case modelled schematically a symmetric molten salt with Φ_1 referring to density and Φ_2 to charge fluctuations. The assumed symmetry of the interactions implies then the peculiar form of \mathcal{F}_2 (Bosse and Munakata 1982). For applications, one would also like to understand the case where there is a finite density of the tagged particles or where the interaction symmetry is broken. Furthermore, one would like to discuss the two specified problems on the basis of less schematic models. Therefore we shall consider how the quoted results get modified if more general situations are considered.

The analysis begins in section 2 by identifying the indicated type-A GTS within the glass for the general $M = 2$ model. It will be shown (section 3) that the long-time dynamics near the GTS can be described by a two-parameter scaling law, which can be reduced to the one studied earlier (Götze 1984) for type-A transitions. The general case for arbitrary M shall be analysed in section 4. It will be shown that the long-time dynamics is described by the mentioned scaling law, which thereby is demonstrated to characterize the general dynamics near the specified GTS.

2. The type-A glass transition singularity for the two-component model

Let us begin with the general $M = 2$ model. Equation (1.4a) shall be rewritten as $f_q = \mathcal{T}_q$ where $\mathcal{T}_q = \mathcal{F}_q / (1 + \mathcal{F}_q)$. Let us simplify the notation by writing $\mathcal{F}_1 = v f_1^2 + v_1 f_1 f_2 + v_2 f_2^2$, $\mathcal{F}_2 = w f_1^2 + w_1 f_1 f_2 + w_2 f_2^2$.

To obtain a non-vanishing long-time limit for Φ_1 while that for Φ_2 is zero, one has to solve the cubic equation $f_1(1 + \mathcal{F}_1) = \mathcal{F}_1$ with $\mathcal{F}_1 = v f_1^2$ and to observe (1.4b). Such a solution exists if and only if $v \geq 4$ with $f_1 = f$, where we introduce $f = [1 + \sqrt{1 - 4/v}]/2$. Since the type-B liquid-to-glass transition occurring near $v \sim 4$ shall not be considered, we restrict the discussion to $v > 4$. Let us eliminate v in favour of f by writing

$$v = 1/f(1 - f) \quad 1/2 < f < 1. \tag{2.1}$$

Thus one gets for $f_2 = 0$: $f_1 = f$. For sufficiently small f_2 the three solutions of the cubic equation do not bifurcate. For f_1 one checks the small f_2 expansion $f_1 = f + h f_2 + \mathcal{O}(f_2^2)$, where

$$h = (1 - f)^2 v_1 f / (2f - 1). \tag{2.2}$$

Substitution of this result into \mathcal{T}_2 yields

$$\mathcal{T}_2 - f_2 = \gamma + \sigma f_2 + (\lambda - 1) f_2^2 + \mathcal{O}(\sigma f_2^2, \gamma^2, \gamma f_2, f_2^2). \tag{2.3}$$

Here the following abbreviations have been used:

$$\sigma = w_1 f - 1 \quad \gamma = w f^2 \tag{2.4}$$

$$\lambda = w_2 + h/f. \tag{2.5}$$

For the existence of the solution $f_2 = 0$ it is necessary that $\gamma = 0$. If $\lambda \neq 1$, the equation $T_2 - f_2 = 0$ indeed has two small f_2 -solutions: $f_2 = 0$ and $f_2 = \sigma/(1-\lambda) + \mathcal{O}(\sigma^2)$. Under these restrictions the described bifurcation may occur for $\sigma = 0$. The possibility $\lambda = 1$ shall not be considered, since it leads to higher-order singularities (Götze and Sjögren 1992). If $\lambda > 1$ one proves the existence of a solution $f_2^0 > 0$ of $f_2 = T_2(f_2)$ for $\gamma = \sigma = 0$, and the found bifurcation (1.4a) would violate (1.4b). Therefore one has to require

$$0 \leq \lambda < 1. \quad (2.6)$$

It is easy to check that solution of the equations (1.4a) and (1.4b) for positive but small v_1, v_2, w, w_2 is given by

$$f_1 = f + hg + \mathcal{O}(g^2) \quad (2.7a)$$

$$f_2 = g + \mathcal{O}(g^2) \quad (2.7b)$$

$$g = g(\sigma, \gamma) = [\sigma + \sqrt{\sigma^2 + 4\gamma(1-\lambda)}]/2(1-\lambda). \quad (2.8)$$

The model exhibits a glass transition singularity on a smooth manifold in \mathbb{R} of co-dimensionality 2, specified by the two equations

$$\sigma = 0, \gamma = 0 \quad \text{GTS.} \quad (2.9)$$

The rapid variation of f_q near the GTS is described by two smooth functions of the control parameter vector $V: \sigma(V)$ and $\gamma(V)$. They serve as the relevant control parameters. The leading-order terms $f_1 - f, f_2$ are given by g , which is a homogeneous function of σ and γ . If one varies the control parameter according to

$$\sigma = \hat{\sigma}\Omega \quad \gamma = \hat{\gamma}\Omega^2 \quad \Omega > 0 \quad (2.10)$$

the function g merely changes by a scale factor

$$g = \hat{g}\Omega \quad (2.11)$$

where $\hat{g} = g(\hat{\sigma}, \hat{\gamma})$.

Equations (2.10) describe scaling lines in the half plane of the relevant control parameters (σ, γ) . These are half parabolas terminating for $\Omega \rightarrow 0$ in the GTS. The function g describes the solution of (1.4) correctly in leading linear order of Ω ; the corrections to the results (2.7) and (2.8) are proportional to Ω^2 .

Let us consider the path $V(x)$ of the system, induced by a change of the external control parameter x . Equations (2.4) map this path on a curve C in the relevant control parameter half-plane $x \rightarrow (\sigma(x), \gamma(x))$. Generically one can write $\sigma(x) \propto (x - x_c)/x_c$ for $x \rightarrow x_c$ and therefore σ is called the separation parameter. Generically $\gamma > 0$, and therefore C avoids the GTS and the f_q are smooth functions of x . However, if $\gamma(x)$ is small, curve C can come close to the GTS. There is then a rapid crossover from a regime $\sigma < -d_0$, $d_0 = \sqrt{4\gamma(1-\lambda)}$, where f_2 is very small to a regime $\sigma > d_0$, where f_2 becomes of order unity. So there is a rapid crossover for $x \sim x_c$ from a glass with one component exhibiting a very small non-ergodicity parameter f_2 to a normal glass state.

If some speciality of the kind mentioned in the introduction enforces $\gamma = 0$ the result of the theory changes. The control parameter space is then reduced in its dimensionality. Curve C then consists of two scaling lines. For $x < x_c$ one gets $f_2 = 0$, i.e. a glass where one component behaves ergodically. This solution bifurcates for $x = x_c$ to $f_2 = \sigma/(1-\lambda) + \mathcal{O}(\sigma^2)$ for $x > x_c$. The long-time limit f_2 increases linearly with $(x - x_c)/x_c$ on the second scaling line $\sigma > 0$. Only for $\gamma \equiv 0$ does there occur a type-A transition within the glass.

The identified results of (1.4) are illustrated in figure 1 for $f = 0.8, v_1 = 1.0, w_2 = 0.3$. These parameters imply $v = 6.25, h = 0.32$ and $\lambda = 0.7$. The generic result $\gamma \neq 0$ appears as smearing of the ideal type-A transition.

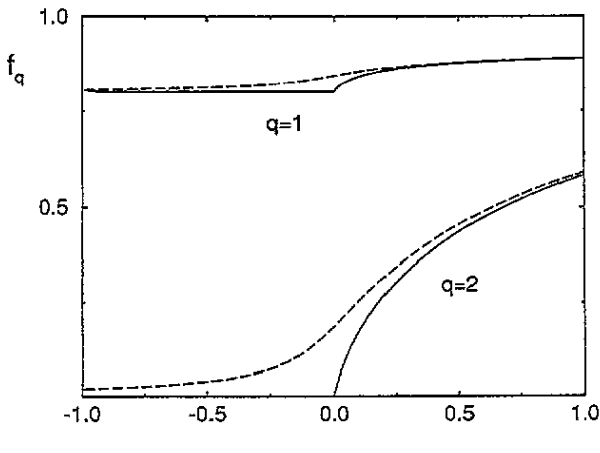


Figure 1. The non-ergodicity parameter f_q for the general $M = 2$ model. The full curves refer to $\gamma = 0$ and the dashed ones to $\gamma = 0.02$. The other parameters, specified in section 2, are: $v = 6.25, v_1 = 6.0, v_2 = 0, w_2 = 0.3$.

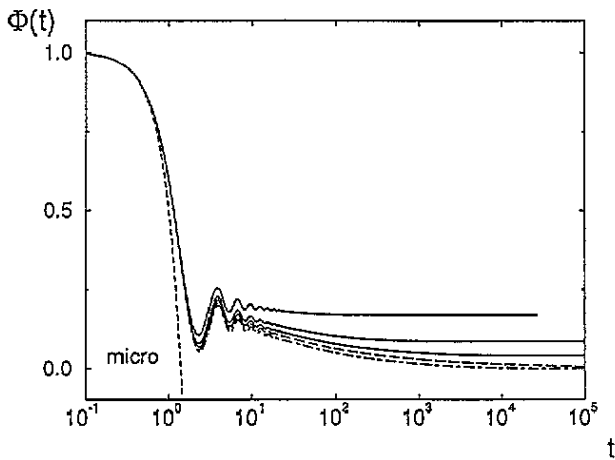


Figure 2. The second correlator $\Phi = \Phi_2$ for the $M = 2$ model for $\gamma = 0$ and constants specified in the caption of figure 1, and $\Omega_1 = \Omega_2 = 1, v_1 = v_2 = 0$. The full curves correspond, from the bottom to top, to $\sigma = 0.1, 0.04, 0.016$. The long-dash broken curve refers to $\sigma = 0$ and the chain curve to $\sigma = -0.016$. The short-dash broken curve is the short-time asymptote $\Phi = 1 - t^2/2$, denoted by 'micro'.

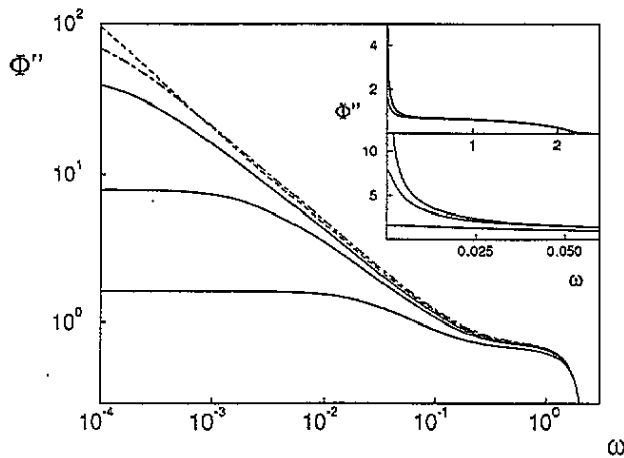


Figure 3. The correlation spectra Φ'' for the decay curves from figure 2. The inserts exhibit the spectra on linear scales for $\sigma = 0.1, 0.04, 0.016$ (from bottom to top).

3. The scaling law description of the long-time decay

Figure 2 exhibits the correlator $\Phi(t) = \Phi_2(t)$ for the previously defined two-component model for $\gamma = 0$ and with the same coupling constants as used in figure 1. The transient dynamics is specified by $v_1 = v_2 = 0$, $\Omega_1 = \Omega_2 = 1$. The full curves exhibit the decay curves for $\sigma = 0.1, 0.04$ and 0.016 and the chain one for $\sigma = -0.016$. The broken curve refers to the critical point $\sigma = 0$. Figure 3 exhibits the relaxation spectra $\Phi''(\omega)$, i.e. the Fourier-cosine transforms of $\Phi(t)$. Notice, that the area of the spectra decreases with increasing positive σ , since a part of the spectra is contained in the not explicitly shown elastic contribution $\pi f_2 \delta(\omega)$.

From (1.1) one derives the short-time asymptotics $\Phi(t) = 1 - t^2/2 + \mathcal{O}(t^4)$, shown as short-dash broken curve in figure 2. The transient dynamics is influenced by interaction effects only for the terms $\mathcal{O}(t^4)$ and it yields to pronounced oscillations extending up to $t = 10$. This transient motion for $t \lesssim 10$ produces a flat spectrum extending between $0.5 < \omega < 2$. For points off the GTS the decay curves reach their asymptote exponentially and this yields an ω -independent spectrum for very low frequencies (Götze and Sjögren 1993). However, this approach to the asymptote sets in only for times much larger than expected from the scale $1/\Omega = 1$ for the transient. The low-frequency spectrum $\Phi''(\omega = 0)$ is reached only for $\omega \ll \Omega$. The crossover to the low-frequency plateau occurs for smaller ω , and the larger the spectral enhancement $\Phi''(0)$, the smaller is $|\sigma|$. The insets of figure 3 demonstrate that it is difficult to appreciate the sensitive variation of the spectra near the singularity if one plots them on linear scales.

The decay curve at the GTS is shown in figure 2 as a broken curve. The corresponding spectrum approaches a straight line on the double-logarithmic representation of figure 3 for small frequencies. Hence $\Phi''(\omega \rightarrow 0) \propto 1/\omega^{1-a}$ for $\sigma = 0$ and this is equivalent to a power law decay $\Phi(t \rightarrow \infty) = (t_0/t)^a$. From figure 3 one reads off $a = 0.33$ and from figure 2 $t_0 = 0.025$. For every finite time interval $0 \leq t \leq T < \infty$, the correlators depend smoothly on the control parameters like σ (Götze and Sjögren 1993). Thus there is a time interval $t_0 < t < t_d$ where $\Phi(t)$ is close to the mentioned power law decay. The time t_0 is σ -insensitive and marks the end of the short-time transient. The time t_d specifies the crossover to the long-time limit and it tends to infinity as σ tends to zero. The evolution of this interval $t_0 \leq t \leq t_d$ is the origin of the relaxation stretching exhibited in figure 2. Via Fourier transformation it leads to the spectral increase shown in figure 3. In order to understand the essence behind the described anomalous dynamics the equations of motion shall be solved for $t \gg 1$ or $\omega \ll 1$ and parameters approaching the GTS.

Using the notations of section 2 and writing $\Phi_1(t) = f_1 + \Psi(t)$, $\Phi_2(t) = \Phi(t)$ one can cast the equations of motion in the following form:

$$\Phi(t)[v_1 f(1-f)] - \Psi(t)(2f-1)/(1-f) = I_1(t) \quad (3.1)$$

$$\gamma + \sigma \Phi(t) + \lambda \Phi(t)^2 - \frac{d}{dt} \int_0^t \Phi(t-t')\Phi(t')dt' = I_2(t). \quad (3.2)$$

Here the following abbreviations have been used:

$$I_1(t) = [\ddot{\Psi}(t) + v_1 \dot{\Psi}(t)]/\Omega_1^2 - (1-f)\delta m_1(t) + \frac{d}{dt} \int_0^t \Psi(t-t')\{[2\Psi(t')/(1-f)] + f v_1 \Phi(t') + \delta m_1(t')\} dt' \quad (3.3a)$$

$$\delta m_1(t) = v\Psi(t)^2 + v_1 \Psi(t)\Phi(t) + v_2 \Phi(t)^2 \quad (3.3b)$$

$$\begin{aligned}
 I_2(t) = & [\ddot{\Phi}(t) + v_2\dot{\Phi}(t)]/\Omega_2^2 + \sigma + \frac{d}{dt} \int_0^t \Phi(t-t')\Phi(t') dt' - \sigma(h/f)\Phi(t)^2 \\
 & - w_1[\Psi(t) - h\Phi(t)]\Phi(t) - \gamma[2f + \Psi(t)]\Psi(t)/f^2 \\
 & \times \frac{d}{dt} \int_0^t \Phi(t-t')[w\Phi_1(t')^2 + w_1\Psi(t')\Phi(t') + w_2\Phi(t')^2] dt'. \quad (3.3c)
 \end{aligned}$$

The solution shall be worked out by asymptotic expansion in the limit $\sigma \rightarrow 0$, $\gamma \rightarrow 0$, $t \rightarrow \infty$. In this limit both Ψ and Φ tend to zero. Anticipating $I_{1,2}(t)$ to be of higher order than the left-hand sides of (3.1) or (3.2) respectively, one gets $\Psi = h\Phi$ in leading order. Thus one can generalize (2.7) to

$$\Phi_1(t) = f + hg(t) + \mathcal{O}_1 \quad (3.4a)$$

$$\Phi_2(t) = g(t) + \mathcal{O}_2. \quad (3.4b)$$

Here the correlator $g(t)$ is to be determined from

$$\gamma + \sigma g(t) + \lambda g(t)^2 - \frac{d}{dt} \int_0^t g(t-t')g(t') dt' = 0. \quad (3.5)$$

One has to show that the \mathcal{O}_q terms are asymptotically arbitrary small compared to $g(t)$.

First let us consider the relaxation at the bifurcation point $\sigma = 0$, $\gamma = 0$. Then (3.5) has the following solution, to be called critical decay:

$$g(t) = (t_0/t)^a \quad \text{GTS.} \quad (3.6)$$

The critical exponent a follows from $\lambda = \Gamma(1-a)^2/\Gamma(1-2a)$, $0 < a \leq 1/2$. For $\lambda = 0.7$ one obtains $a = 0.327\dots$, and the solution in figure 2, shown as a broken curve, follows this law for $t > 20$. The timescale t_0 has been determined by fitting to the curve $\sigma = 0$. The corresponding critical spectrum $\Phi''(\omega) = \sin(\pi a/2)\Gamma(1-a)(\omega t_0)^a/\omega$ is also shown as a broken asymptote in figure 3. Substitution of (3.4), (3.6) into (3.3) yields $I_1 \propto (t_0/t)^{2a}$, $I_2 \propto (t_0/t)^{3a}$ for large t . This in turn, with (3.1) and (3.2), implies

$$\mathcal{O}_q = A_q(t_0/t)^{2a} + \mathcal{O}((t_0/t)^{2a}) \quad \text{GTS.} \quad (3.7)$$

It is straightforward but cumbersome to evaluate the coefficients A_q .

The equation (3.5) for the leading contribution g to the long-time decay can be simplified by introducing another function G , related to g by

$$g(t) = \frac{\sigma + \sqrt{1-\lambda}G(t)}{2(1-\lambda)}. \quad (3.8)$$

This new correlator obeys

$$d^2 + \lambda G(t)^2 = \frac{d}{dt} \int_0^t G(t-t')G(t') dt'. \quad (3.9)$$

While g depends on the two parameters σ and γ , function G depends only on the combination

$$d = \sqrt{\sigma^2 + 4\gamma(1-\lambda)}. \quad (3.10)$$

If the two relevant control parameters shift along a scaling line (2.10), the function d rescales proportionally to Ω

$$d = \Omega \hat{d} \quad (3.11)$$

where \hat{d} is the value of d for the point $(\hat{\sigma}, \hat{\gamma})$. For vanishing γ , $d = |\sigma| \propto |x - x_c|/x_c$ is the distance from the GTS. The function d can be viewed as a renormalized distance. The parameter $\gamma \neq 0$ hinders the distance to vanish: $d \geq d_0 = \sqrt{4\gamma(1-\lambda)}$. Equation (3.9) is the scaling equation derived originally for the discussion of the ideal type-A transition and also for the description of the relaxation within the glass state near type-B transitions (Götze 1984). Obviously, one can write

$$G(t) = d \hat{G}(t/t_d) \quad (3.12)$$

where the control parameter independent master function \hat{G} obeys the equation

$$1 + \lambda \hat{G}(\hat{t})^2 = \frac{d}{d\hat{t}} \int_0^{\hat{t}} \hat{G}(\hat{t} - \hat{t}') \hat{G}(\hat{t}') d\hat{t}' \quad \hat{t} = t/t_d. \quad (3.13)$$

Equation (3.5) is scale invariant: with $g(t)$ also $g^y(t) = g(ty)$ is a solution for any $y > 0$. The same holds for (3.9) and (3.13). To fix the solution uniquely initial conditions shall be imposed for $t/t_0 \rightarrow 0$, or $\hat{t} \rightarrow 0$ respectively:

$$g(t)(t/t_0)^a \rightarrow 1 \quad \hat{G}(\hat{t})\hat{t}^a \rightarrow 1. \quad (3.14)$$

This fixes the timescale t_d to

$$(t_d/t_0) = d^{-1/a} (2\sqrt{1-\lambda})^{1/a}. \quad (3.15)$$

A detailed discussion of \hat{G} can be found elsewhere (Götze 1990). The monotonically decreasing function \hat{G} describes the crossover from the short-time asymptote

$$\hat{G}(\hat{t}) = (1/\hat{t}^a) + A_1 \hat{t}^a + \dots \quad (3.16)$$

to the long-time limit $\hat{G}(\hat{t} \rightarrow \infty) = 1/\sqrt{1-\lambda}$. The coefficients A_i in the expansion (3.16) can be expressed in terms of a . The expansion up to \hat{t}^{5a} is sufficient in most cases to determine the complete \hat{G} . Equation (3.13) can be solved numerically. From (3.12) one obtains for the correlation spectrum

$$\Phi''(\omega) = C_d \hat{\Phi}''(\omega/\omega_d). \quad (3.17)$$

Here the spectral scale reads $C_d = dt_d$, the frequency scale is $\omega_d = 1/t_d$ and the master spectrum is given by the Fourier cosine transform $\hat{G}''(\hat{\omega})$ of $\hat{G}(\hat{t})$ by $\hat{\Phi}'' = \hat{G}''/(2\sqrt{1-\lambda})$.

Equations (3.12) and (3.17) are scaling laws expressing that the dependence of the solutions on the control parameter d is given by two scales only. The shape of the decay curves \hat{G} or spectra $\hat{\Phi}''$ is independent of d ; they are determined by the parameter λ . The timescale t_d marks the crossover from the regime $t_0 \ll t \ll t_d$ to $t_d \ll t$. In the former the correlator follows closely the critical law (3.6); it depends only weakly and regularly on the control parameters as follows from the substitution of (3.12) and (3.16) into (3.8):

$$g(t_0 \ll t \ll t_d) = \frac{\sigma}{2(1-\lambda)} + (t_0/t)^a + A_1 \frac{d^2}{4(1-\lambda)} (t/t_0)^a + \mathcal{O}(d^4 t^{3a}). \quad (3.18)$$

The correlator g depends on three variables: $g = g(t/t_0, \sigma, \gamma)$. It is a homogeneous function. If the control parameters vary along a scaling line (2.10) and if the time is rescaled according to

$$t = \hat{t}/\Omega^{1/a} \tag{3.19}$$

the correlator is altered merely by a scale factor:

$$g(t/t_0, \sigma, \gamma) = \Omega \hat{g}(\hat{t}). \tag{3.20}$$

Here $\hat{g}(\hat{t}) = [\hat{\sigma} + \sqrt{1 - \lambda \hat{d} \hat{G}(\hat{t})}]/[2(1 - \lambda)]$. Approaching the GTS on a scaling line with $\Omega \rightarrow 0$, the leading-order correlator g vanishes proportional to Ω in a self-similar manner. One checks that in (3.3) $I_1 \propto \Omega^2$, $I_2 \propto \Omega^3$ for $\Omega \rightarrow 0$. Therefore (3.1) and (3.2) imply in (3.4) $\mathcal{O}_q \propto \Omega^2$. The scaling law describes the dynamics near the GTS in leading order Ω while the leading correction terms vanish proportional to Ω^2 . To illustrate a possible procedure for an experimental test of the found scaling law, let us consider $\hat{\Phi}'' = \Phi''/\Phi''_{\max}$ as function of $\hat{\omega} = \omega/\omega_k$. Here Φ''_{\max} is the maximum of the spectrum, i.e. the plateau value for small frequencies $\Phi''_{\max} = \Phi''(\omega \rightarrow 0)$. The frequency ω_k is a scaling frequency, specifying the spectral knee. If scaling were valid, results for different σ would give the same $\hat{\Phi}''$ against $\hat{\omega}$ plot. Scaling is only an asymptotic result valid in the limit $\Omega \rightarrow 0$:

$$\hat{\Phi}'' = G''(\hat{\omega} t_d \omega_k) + \mathcal{O}(\Omega). \tag{3.21}$$

Therefore the $\hat{\omega}$ interval, where the various $\hat{\Phi}''$ against $\hat{\omega}$ curves coincide, expands for $\Omega \rightarrow 0$. The rescaling from Φ'' to $\hat{\Phi}''$ and from ω to $\hat{\omega}$ is done most conveniently by using double logarithmic diagrams. Rescaling then amounts to parallel shifts of the diagrams: $\log \Phi''_{\max}$ parallel to the vertical axis and $\log \omega_k$ parallel to the horizontal one. Such a scaling plot for the results from figure 3 is shown in figure 4. After having verified the scaling scenario one should compare the found master spectrum with the theoretical one, which can be obtained from Götze (1990). Finally, one should demonstrate that the found scales vary according to the predictions $\Phi''_{\max} \propto dt_d$ and $\omega_k \propto 1/t_d$ for $d \rightarrow 0$.

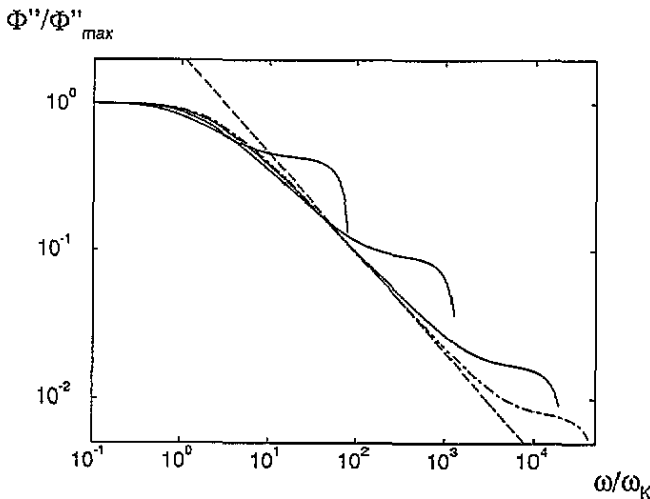


Figure 4. Rescaling of the spectra from figure 3, see text. The broken line exhibits the critical spectrum $\Phi'' \propto 1/\omega^{1-a}$ with $a = 0.327$. For $\sigma = 0.1, 0.04, 0.016, -0.016$ the following scales have been used respectively: $\Phi''_{\max} = 1.61, 7.84, 44.2, 94.6$ and $\omega_k = 39.4, 651, 10\,700, 26\,800$.

Figure 4 shows that the scaling laws do indeed explain the qualitative features of the anomalous spectra shown in figure 3 for the dynamics near the GTS. But the figure also shows that rescaled results show appreciable deviations from the correct asymptotic results. These findings imply in particular the following hazard for data analysis. The $\log \Phi''$ against $\log \omega$ diagram exhibits an inflection point on the wing of the low-frequency spectral peak. So there is always a large frequency interval where a fit according to $\Phi'' \propto 1/\omega^{1-a'}$ is quite good. But if the separation from the critical point is not small enough, one gets $a' > a$. This is demonstrated in figure 4 for the $\sigma = 0.016$ result and even more for the spectrum for $\sigma = 0.04$. The true critical spectrum is shown as a broken line. In order to exclude the misinterpretation of a' as the critical exponent a , one ought to compare data with the full theoretical master spectrum.

A test of the scaling laws can also be done for the correlation functions $\Phi_q(t)$. One can, for example, discuss the rescaled deviation of the correlator from its long-time asymptote $\tilde{\Phi}_q = (\Phi_q - f_q)/H_q$. If the rescaling factor is chosen to be proportional to the critical amplitude and to the distance, $H_q = H h_q |d|$, all curves $\tilde{\Phi}_q$, if considered as a function of the rescaled time t_d , should coalesce on a master curve:

$$\tilde{\Phi} = H[\hat{G}(\hat{t}) - 1/\sqrt{1-\lambda}] \quad \hat{t} = t/t_d. \quad (3.22)$$

The master curve, i.e. the bracket in the preceding equation, is determined by λ and can be inferred from Götze (1990). Scaling holds only in the asymptotic limit $d \rightarrow 0$. Thus the prediction is that the interval of rescaled times \hat{t} , where (3.22) is valid, should expand upon approaching the GTS. To cover a sufficient time interval one uses conveniently a logarithmic abscissa. Figure 5 exhibits such a scaling plot for the data from figure 2.

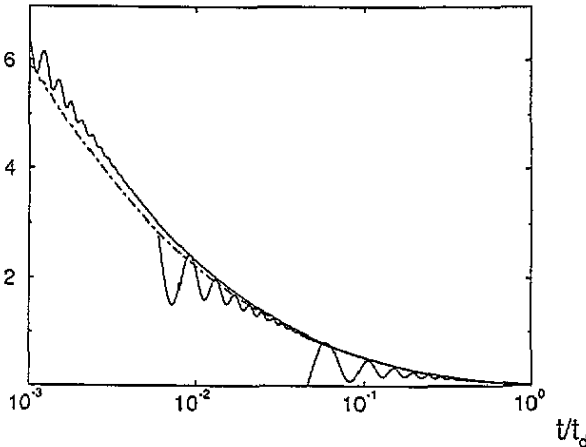
 $\tilde{\Phi}$


Figure 5. Rescaled decay curves $\tilde{\Phi} = (\Phi - f)/H$ from figure 2 as a function of rescaled times \hat{t} (see text). For $\sigma = 0.1, 0.04, 0.016, -0.016$ the following scales have been used respectively: $H = 0.1, 0.04, 0.016, 0.016$ and $t_d = 65.5, 738, 10250, 22540$.

4. General derivation of the scaling laws near type-A glass transition singularities

An ideal type-A transition can be obtained for a one-component model, provided the mode coupling functional contains a linear term: $\mathcal{F}(f_2) = \sigma f_2 + \lambda f_2^2$. This model can be extended

to describe the complete scenario, discussed in sections 2 and 3, if one generalizes the functional to $\mathcal{F}(f_2) = \gamma + \sigma f_2 + \lambda f_2^2$. It was shown that this scenario can be obtained from a quadratic functional (1.3) provided one considers a two-component model. The two parameters γ and σ are then caused by the coupling of one component to another background component. The latter provides an effective defect potential, which leads to the linear term in \mathcal{F} . In this section the general case $M \geq 2$ shall be studied. It will be shown that in this case too a GTS can occur, which is described by a single g , obeying the equations (3.5)–(3.18). Thus the results of section 3 demonstrate the general case. The full theory merely provides more general formulae for σ , γ and λ in terms of the mode coupling functional (1.3) and generalizes the formulae (2.7) and (3.4). The following derivation extends and modifies the corresponding reasoning, made originally for a type-B transition (Götze 1991).

We start by introducing some notation. If f^c is a solution of (1.4) for control parameters V^c , we write

$$f_q = f_q^c + (1 - f_q^c)^2 g_q \quad V = V^c + v. \tag{4.1}$$

The various Taylor coefficients of the mode coupling functional \mathcal{F}_q shall be denoted as

$$C_q(V) = \mathcal{F}_q(V, f^c) \tag{4.2a}$$

$$C_{qk}(V) = \frac{\partial \mathcal{F}_q(V, f^c)}{\partial f_k} (1 - f_k^c)^2 \tag{4.2b}$$

$$C_{qkp}(V) = \frac{1}{2} \frac{\partial^2 \mathcal{F}_q(V, f^c)}{\partial f_k \partial f_p} (1 - f_k^c)^2 (1 - f_p^c)^2. \tag{4.2c}$$

These are linear functions of V and one gets

$$\mathcal{F}_q(V, f) = f_q^c / (1 - f_q^c) + C_q(v) + \sum_k [C_{qk}^c + C_{qk}(v)] g_k + \sum_{kp} C_{qkp}(V) g_k g_p. \tag{4.3}$$

These formulae would also hold if a general polynomial \mathcal{F} were to be considered in (1.3); but (4.3) would then have to be completed by a term of higher order in g . The matrix $C^c = C_{qk}(V^c)$ is called the stability matrix; it is the non-trivial part of the Jacobian of the implicit equation (1.4a). If the eigenvalues of C^c are not unity, the solution f_q^c can be uniquely continued to a solution f_q of (1.4) for small v . This solution depends smoothly on v . A bifurcation singularity is connected with an eigenvalue of C^c tending to unity.

The point V^c shall now be specialized to the singularity of interest. Let us split the variables into two groups: the first one is specified by L variables $q = 1, \dots, L$ and the second one by $M - L$ variables $q = L + 1, \dots, M$. In an obvious generalization of section 2 we demand that

$$V_{qkp}^c = 0 \quad q > L \quad k, p \leq L. \tag{4.4}$$

We analyse f_q^c for $q > L$. The other components are non-negative and generically $0 < f_q^c < 1$, $q \leq L$. The stability matrix consists of four blocks. There are the two diagonal blocks, to be denoted by $C^{(\alpha)}$, $\alpha = 1, 2$. They are of degree L and $(M - L)$ respectively: $C_{qk}^{(\alpha)} = C_{qk}^c$ where $q, k \leq L$ for $\alpha = 1$ and $q, k > L$ for $\alpha = 2$. The L by $(M - L)$ matrix $C^{(1,2)}$ we shall abbreviate by $C_{qk}^{(1,2)} = C_{qk}^c$ for $q \leq L, k > L$. Equation

(4.4) implies $C_{qk}^c = 0$ for $q > L, k \leq L$. The elements of the matrices $C^c, C^{(1)}, C^{(2)}$ are non-negative. Frobenius theory (Gantmacher 1960) implies that the matrices have non-negative eigenvalues $E, E^{(1)}, E^{(2)}$ respectively, which agree with the corresponding spectral radius. The matrix C^c is reducible and $E = \text{Max}(E^{(1)}, E^{(2)})$. The results of the general MCT (Götze and Sjögren 1993) ensure $E \leq 1$. The condition $E^{(1)} = 1$ would imply generically a type-B transition for the first L components. This case shall not be considered in this paper. Therefore it will be demanded that $E^{(1)} < 1$. Thus the L by L matrix $(1 - C^{(1)})$ has an inverse, which can be evaluated as a Neumann series

$$R = (1 - C^{(1)})^{-1} = \sum_l C^{(1)l}. \quad (4.5)$$

Obviously, the L^2 elements of R are non-negative: $R_{qk} \geq 0, q, k \leq L$. To get a GTS one has to request $E^{(2)} = E = 1$. According to Frobenius theory matrix $C^{(2)}$ has right and left eigenvectors, to be denoted by e and \hat{e} respectively:

$$\sum_{k>L} C_{qk}^{(2)} e_k = e_q \quad \sum_{q>L} \hat{e}_q C_{qk}^{(2)} = \hat{e}_k \quad e_k \geq 0 \quad \hat{e}_k \geq 0. \quad (4.6)$$

It shall be required that the matrix $C^{(2)}$ is non-degenerate. Then the eigenvalue $E^{(2)}$ is non-degenerate and the eigenvectors are unique up to a positive scale factor. They are uniquely fixed if one imposes the conditions

$$\sum_{q>L} \hat{e}_q e_q = 1 \quad \sum_{q>L} \hat{e}_q (1 - f_q^c) e_q e_q = 1. \quad (4.7)$$

Generically none of the numbers e_q and \hat{e}_q is zero. A left eigenvector \hat{e}^c of C^c for eigenvalue $E = 1$ is given by

$$\begin{aligned} \hat{e}_q^c &= 0 & q &\leq L \\ \hat{e}_q^c &= \hat{e}_q^c & q &> L. \end{aligned} \quad (4.8)$$

A right eigenvector e^c can easily be found as

$$e_q^c = \sum_{k \leq L} \sum_{p > L} R_{qk} C_{kp}^{(1,2)} e_p \quad q \leq L \quad (4.9a)$$

$$e_q^c = e_q \quad q > L. \quad (4.9b)$$

These vectors obey normalizations analogous to (4.7).

The solution shall be written as $\Phi_q(t) = f_q^c + (1 - f_q^c)^2 g_q(t)$. With

$$\chi_q(t) = -[\dot{g}_q(t) + \nu_q \dot{g}_q(t)](1 - f_q^c) / \Omega_q^2 \quad (4.10)$$

and the help of

$$\int_0^t m_q(t-t') \dot{\Phi}_q(t') dt' = \frac{d}{dt} \int_0^t m_q(t-t') (1 - f_q^c)^2 g_q(t') dt' - (1 - f_q^c) m_q(0) \quad (4.11)$$

and substitution of (4.3) one derives for the equations of motion (1.1) the equivalent set of equations

$$\sum_k [\delta_{qk} - C_{qk}^c] g_k(t) = R_q(t). \tag{4.12}$$

Here the following abbreviations are used:

$$R_q(t) = C_q(v) + \sum_k C_{qk}(v) g_k(t) + \sum_{kp} C_{qkp}(V^c) g_k(t) g_p(t) - \frac{d}{dt} \int_0^t g_q(t-t')(1-f_q^c) \sum_k C_{qk}^c g_k(t') dt' + I_q(t) \tag{4.13}$$

$$I_q(t) = \chi_q(t) - (1-f_q^c) C_q(v) g_q(t) + \sum_{kp} C_{qkp}(v) g_k(t) g_p(t) - (1-f_q^c) \frac{d}{dt} \int_0^t g_q(t-t') \left(\sum_k C_{qk}(v) g_k(t') + \sum_{kp} C_{qkp}(V) g_k(t') g_p(t') \right) dt'. \tag{4.14}$$

Let us introduce $\Phi(t) = \sum_q \hat{e}_q^c g_q(t)$. Then one writes

$$g_k(t) = \Phi(t) e_k^c + \delta g_k(t) \quad \sum_k \hat{e}_k \delta g_k(t) = 0 \tag{4.15a}$$

and the equations of motion (4.12) has the form

$$\sum_k [\delta_{qk} - C_{qk}^c] \delta g_k(t) = C_q(v) + \sum_k C_{qk}(v) e_k^c \Phi(t) + \Phi(t)^2 \sum_{kp} C_{qkp}(V^c) e_k^c e_p^c - \frac{d}{dt} \int_0^t \Phi(t-t') \Phi(t') dt' (1-f_q^c) e_q^c e_q^c + J_q(t) + I_q(t) \tag{4.15b}$$

where

$$J_q(t) = \sum_k C_{qk}(v) \delta g_k(t) + \sum_{kp} [2C_{qkp}(V^c) e_k^c \delta g_p(t) \Phi(t) + C_{qkp}(V^c) \delta g_k(t) \delta g_p(t)] - (1-f_q^c) \frac{d}{dt} \int_0^t [g_q(t-t') \sum_k C_{qk}^c \delta g_k(t') + \delta g_q(t-t') e_q^c \Phi(t')] dt'. \tag{4.15c}$$

The condition for the solvability of (4.15b) is that the right-hand side is perpendicular to \hat{e}_q^c . With (4.7) this leads to

$$\gamma + \sigma \Phi(t) + \lambda \Phi(t)^2 - \frac{d}{dt} \int_0^t \Phi(t-t') \Phi(t') dt' = I(t). \tag{4.16}$$

Here the following abbreviations are introduced:

$$\sigma = \sum_{qk} \hat{e}_q C_{qk}(v) e_k^c \tag{4.17a}$$

$$\gamma = \sum_q \hat{e}_q C_q(V) \tag{4.17b}$$

$$\lambda = \sum_{qkp} \hat{e}_q C_{qkp}(V^c) e_k^c e_p^c \tag{4.17c}$$

$$I(t) = \sum_q \hat{e}_q [I_q(t) + J_q(t)]. \tag{4.18}$$

In noting (4.17b) we used $C_q(v) = C_q(V)$ for $q > L$ because of (4.4). It is then evident that $\gamma \geq 0, \lambda \geq 0$. Let us emphasize that the equations (4.15)–(4.18) for the $(M + 1)$ variables $\Phi(t)$ and $\delta g_q(t)$, obeying one subsidiary condition (4.15a), are an exact reformulation of the original equations of motion (1.1)–(1.3).

The obtained equations can be solved by asymptotic expansion for large times and near the GTS, i.e. in the limit $v \rightarrow 0$. If $V \rightarrow V^c$ one finds $\sigma \rightarrow 0$ and $\gamma \rightarrow 0$. One starts with the ansatz $\Phi(t) = g(t) + \mathcal{O}$ and treats g and v as leading-order small quantities, while $I_q, J_q, \delta g_q$ are considered as small in higher order. Thus one arrives at

$$\Phi_q = f_q^c + h_q^{(1)} g(t) + \mathcal{O}_q^{(1)} \quad q \leq L \quad (4.19a)$$

$$\Phi_q = h_q^{(2)} g(t) + \mathcal{O}_q^{(2)} \quad q > L. \quad (4.19b)$$

Here the critical amplitudes $h_q \geq 0$ are given in terms of the eigenvector e_q^c of the stability matrix, as specified in (4.9):

$$h_q^{(1)} = (1 - f_q^c)^2 e_q^c \quad q \leq L \quad (4.20a)$$

$$h_q^{(2)} = e_q^c \quad q > L. \quad (4.20b)$$

The function $g(t)$ obeys (4.16) with $I(t)$ dropped. Hence $g(t)$ is identical with the quantity discussed in section 3. The proof is finished by verifying, that $\mathcal{O}_q^{(\alpha)} \propto g^2$. This task, which is left to the reader, can be done as demonstrated in section 3.

The results found formulate the reduction theorem of the MCT for the specified GTS. The task to determine the long-time dynamics near the singularity for M correlators $\Phi_q(t)$ is reduced to the evaluation of a single function $g(t)$. The task of studying the dependence of $\Phi_q(t)$ on the large number of control parameters V is reduced to studying the dependence on only the two relevant parameters σ and γ . The shape functions for the correlators or the spectra depend on the single parameter λ . All systems with the same λ exhibit the same dynamics, up to two scales C_d and t_d and up to the amplitude vectors $h_q^{(\alpha)}$.

5. Conclusions

The motion of electrons between impurities is usually modelled by assuming the latter as fixed in a frozen array. Depending on the density of the impurities one gets a percolation transition from extended to localized motion for the electrons. This transition can be viewed as the simplest example of a glass transition in the Edwards–Anderson sense. It can be treated approximately within the simplified mode coupling theory, which deals with the cage effect for the dynamics of interacting particles. In this case the cages are formed by the impurities. In this paper it was shown that the mentioned *ad hoc* model can be replaced within the MCT by analogous *ad hoc* assumptions. One can start with the standard equations dealing with quadratic mode coupling terms, equation (1.3), and impose certain conditions on the interaction constants $V(q, k, p)$. Then one can get a part of the variables frozen in an ideal glass state. These variables act as an impurity background for the second part of the modes. The latter get a mode coupling functional, where linear terms also appear, equation (2.3). These linear terms are the necessary conditions for the generalized percolation transition. We have developed the general theory for the long-time dynamics near such a transition. It was shown that a single control parameter σ governs the sensitive dependence of the correlators on the coupling coefficients. The separation parameter σ is

proportional to the difference of some external control parameter x from its critical value x_c : $\sigma \propto (x - x_c)/x_c$.

Application of the MCT to the description of supercooled liquids dealt with type-B transitions, where the Edwards–Anderson parameters exhibit discontinuities at the critical point x_c . This renders the long-time decay a two-step process, characterized by two diverging timescales and two time fractals. The dynamics for $x > x_c$ differs drastically from that for $x < x_c$ in this case (Götze and Sjögren 1992). The type-A transition, discussed in this paper, is quite different. The Edwards–Anderson parameter is continuous at x_c ; as a function of x it exhibits a kink only. The long-time decay is governed by a single timescale t_d and the critical decay (3.6) is the only time fractal appearing. There is symmetry of the dynamics with respect to the critical point x_c . The same master functions describe the relaxation for $x > x_c$ as for $x < x_c$ and the scales depend on the distance $d = |\sigma|$ but not on the sign of σ .

The described type-A transition can also occur for the model of a symmetric molten salt. In this case one considers as the first component the density and as the second the charge fluctuations. The assumed symmetry for the interactions restricts the parameter space \mathbb{R} so that type-A bifurcations for the charge fluctuations within the frozen density fluctuations occur on hypersurfaces in \mathbb{R} . Shifting the system on a path $V(x)$ through \mathbb{R} by a change of the single control parameter x can generically lead to crossings with the bifurcation hyper-surface and hence to the transitions under study. However, if the symmetry is broken such crossings do not occur generically. Within general models the mentioned singularities V^c occur on sets of co-dimensionality larger than unity. Generically the path $V(x)$ avoids the singularities and either all components exhibit glass behaviour or the system is in an ergodic liquid state. But it is a generic possibility that the system is in a close neighbourhood of the mentioned type-A bifurcation points V^c . In this case there are also long-time or low-frequency anomalies for the dynamics. It was shown that near the singularities these anomalies are described by only two relevant control parameters. In addition to the separation parameter σ only some smearing parameter $\gamma \geq 0$ plays a role. The Edwards–Anderson parameter exhibits a smooth but rapid crossover, equation (2.8). For large positive σ there is a normal glass state with all components frozen so that all Debye–Waller factors are of order unity. However, for large negative σ the Debye–Waller factors of the second components, though non-zero, are very small. For many purposes this state will appear as a liquid moving in the glass formed by the first component.

The identified crossover is connected with dynamical anomalies. Surprisingly, it is the same reduction theorem connected with the same scaling law and master functions as found for the ideal type-A transition, which describes the crossover. The smearing parameter γ merely alters the distance $|\sigma|$ to an effective distance d , equation (3.10). As far as the long-time motion or the low-frequency spectra are concerned, the smearing is accounted for by renormalizing $|\sigma|$ to a function d , which can become small but must not vanish. Figures 2–5, which were calculated for the ideal transition, automatically exhibit all the leading order dynamical effects for the general case $\gamma > 0$ as well.

Michel (1987) and Bostoen and Michel (1991) worked out a MCT for the deformations and orientations in mixed cyanid crystals. They predicted a type-A transition. Neutron scattering work (Wochner *et al* 1993) verified the predicted anomalies for the Debye–Waller factor and also found the expected anomalous low-frequency spectrum, thereby strongly supporting the proposed theoretical picture. The present paper extends Michel's theory in two respects. It is shown that there may occur a non-trivial exponent parameter λ . This $\lambda \neq 0$ alters, for example, the critical exponent $1/2$ to some number $a < 0.5$. Furthermore, there may occur a smearing parameter $\gamma > 0$, which prevents the system from

reaching the glass transition singularity. In order to test the theory further, the factorization property (4.19) has to be verified. The insensitivity of the dynamics on variations of the wavevector q is a crucial feature, which distinguishes the MCT scenario from conventional phase transitions. Then one has to verify that the spectral enhancement near the critical point follows the critical law (3.18) with $\alpha \leq 1/2$. Finally, one should test the scaling laws as indicated by figure 4 and test the predicted power law variations for the scales.

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