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► To cite this version:

J. Souletie, D. Bertrand. Glasses and spin glasses: a parallel. Journal de Physique I, 1991, 1 (11), pp.1627-1637. 10.1051/jp1:1991230 . jpa-00246440

HAL Id: jpa-00246440

<https://hal.science/jpa-00246440>

Submitted on 4 Feb 2008

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Classification

Physics Abstracts

64.60F— 64.60H — 64.70P — 75.40G — 75.10N

Glasses and spin glasses : a parallel

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(Received 21 May 1991, accepted in final form 18 July 1991)

Résumé. — Nous montrons que la classification entre verres « forts » et verres « fragiles » qui est populaire pour les liquides trempés peut être aussi appliquée aux verres de spins. Les caractères fragile, intermédiaire et fort distinguent des classes d'universalité différentes dans l'approche habituelle en lois d'échelles. Alors que la classe d'universalité d'un verre de spins reflète la plus ou moins grande anisotropie des interactions, on observe dans les liquides « fragiles » lorsqu'on baisse la température des cross-overs vers des régimes plus « forts » qui pourraient être interprétés comme une restriction de la dimensionnalité d'espace.

Abstract. — We show that the classification of glasses from « strong » to « fragile » can also be applied to spin glasses. Fragile, intermediate and strong behaviours would mean different universality classes and the strong limit would correspond to a lower critical dimension in the framework of the current scaling approach. While the class of a spin glass seems to be attached to the frozen in anisotropy of the interactions, there are experimental signs in glass-forming systems that a strong regime is the result of a cross-over from a more fragile high-temperature situation. The same fact can also be interpreted as a cross-over towards a more anisotropic state or as a restriction of space dimensionality.

1. Introduction.

In figure 1a, the logarithm of the viscosity η of a number of glass-forming systems [1-7] is represented vs. $\frac{T_g}{T}$ where $\eta \sim 10^{13}$ poises at T_g . The viscosity is related to the shear relaxation time τ by the equation $\eta = G_\infty \tau$. The high frequency shear modulus G_∞ is of the order of 10^9 . Figure 1 therefore is also a plot of $\log \tau$ vs. T_g/T where T_g corresponds to $\tau \sim 10^4$ s. As 10^4 s is a long time in laboratory scale, T_g represents a natural limit to the range where experiments have been performed at equilibrium. In some cases $\eta(T_g)$ is obtained by an extrapolation of the data at temperatures $T > T_g$ with one of the expressions which we will discuss below. This plot, which is due to Angell [1], contrasts different behaviours. The viscosity data of the « strong » systems align on a straight line in Angell's diagram and therefore obey an Arrhenius law

$$\tau/\tau_0 = \exp(\theta/T) \quad (1)$$

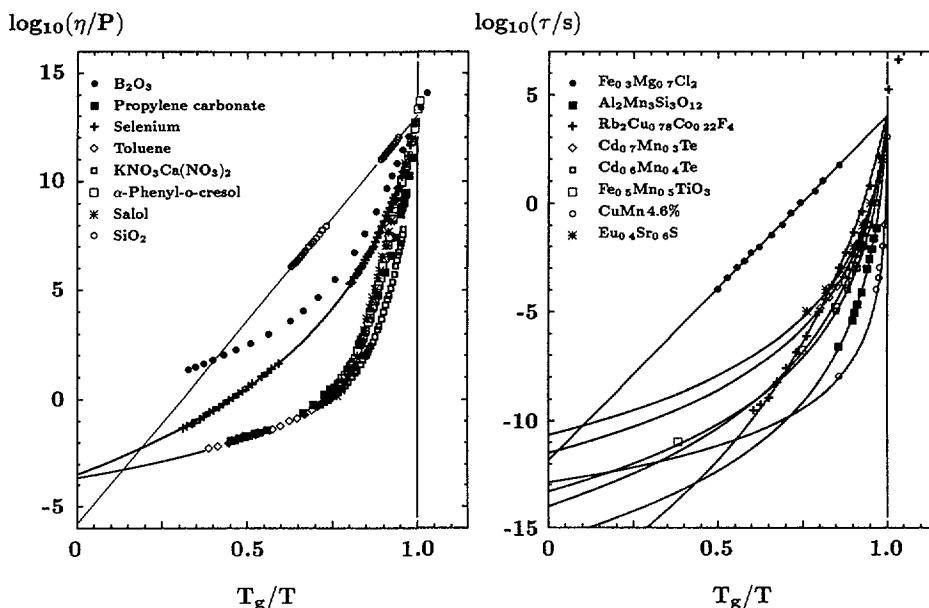


Fig. 1. — In figure 1a the logarithm of the viscosity of different glass-forming systems is represented vs. T_g/T such that $\eta = 10^{13}$ Poise at T_g . The strong systems show Arrhenius behaviour. The viscosity of several fragile systems is well described at high temperatures by a power law $\eta \sim \eta_0 \left(1 - \frac{T_c}{T}\right)^{-z\nu}$ with $z\nu \sim 6.2$ and $\eta_0 \sim 10^{-4}$ Poise (notice however that B_2O_3 extrapolates to a larger η_0). On approaching T_c deviations from this behaviour are observed (see Figs. 2 and 3). Amorphous selenium has a behaviour intermediate between strong and fragile. In figure 1b the logarithm of the relaxation time of different spin glasses is represented vs. T_g/T such that $\tau = 10^4$ s at T_g . Most standard spin glasses [CuMn ... EuSrS, ... Mn aluminosilicate ...] would appear as fragile, according to the current classification of glass forming liquids, when compared to anisotropic systems like $Fe_{0.3}Mg_{0.7}Cl_2$ and $Rb_2Cu_{0.78}Co_{0.22}F_4$.

where $\tau_0 \sim 10^{-13}$ s. The description of the «fragile» systems, by contrast, requires more parameters. They are often described with the law of activated dynamics

$$\tau/\tau_0 = \exp \left[\left(\frac{A}{T - T_0} \right)^\sigma \right]. \quad (2)$$

Even with 4 parameters, this equation (whose oldest and most popular form is the Fulcher law for which $\sigma = 1$) fails to describe the data over the entire ranges of up to 14 decades which are commonly covered by viscosity experiments: on approaching T_0 , before they reach T_g , many fragile systems are better described by an Arrhenius law with an unphysically small τ_0 .

In figure 1b, we have constructed the same plot with dynamic data of the spin-glass literature i.e. we plot the available data vs. T_g/T where T_g is such that $\tau(T_g) \sim 10^4$ s. This definition which stems from the history of glass manufacture differs from the definition which is in general used in spin-glasses and which refers to the temperature where the measurements become non ergodic at the frequency of the experiment. Both definitions would coincide for experiments performed in typically 10^4 s. This choice although arbitrary has the advantage to be independent of the instrument. Until recently most data would have been confined to the

fragile side, sitting between CuMn 4 at% [8] and Mn aluminosilicate [9]. However recent measurements in $\text{Fe}_{0.3}\text{Mg}_{0.7}\text{Cl}_2$ [10], $\text{Rb}_2\text{Cu}_{0.78}\text{Co}_{0.22}\text{F}_4$ [11], $\text{Cd}_{0.6}\text{Mn}_{0.4}\text{Te}$ [12], $\text{Fe}_{0.5}\text{Mn}_{0.5}\text{TiO}_3$ [13] have filled the range between the fragile and the strong limit. The dynamical data in spin glasses can be fitted with the same Fulcher law which was introduced for the structural glasses (Eq. (2) with $\sigma = 1$) [14]. However, when the existence of a phase transition was established on the basis of the divergence of the higher order susceptibilities, it became natural [8, 15] to use the slowing down equation

$$\tau/\tau_0 = (\xi/\xi_0)^z = (1 - T_c/T)^{-z\nu} \quad (3)$$

which is the dynamical counterpart of the same scaling theory which accounts for the static result. As Castaing and Souletie [17] have shown, the dynamic scaling follows in a model which assumes that a hierarchical procedure governs the evolution from step n to step $n+1$ of Kadanoff's renormalisation. More specifically, if we decide that a cluster of order $n+1$ is formed when μ clusters of order n are in a given position we have $\tau_{n+1} = 2^\mu \tau_n = 2^{\mu\nu} \tau_0$ when simultaneously $\xi_{n+1} = b\xi_n = b^n \xi_0$. On eliminating n between these two equations one justifies the dynamic scaling hypothesis

$$\tau/\tau_0 = (\xi/\xi_0)^z \quad \text{where} \quad z = \mu \ln 2 / \ln b. \quad (4)$$

The slowing down equation (3) follows from the static scaling hypothesis which states that there is a power law divergence at T_c in the temperature dependence of the correlation length ξ which is otherwise an analytic function of $1/T$ at all temperatures larger than T_c . We have therefore :

$$\begin{aligned} \xi(T)/\xi_0 &= \left(1 - \frac{T_c}{T}\right)^{-\nu} \left(1 + \frac{B}{T} + \frac{C}{T^2} + \dots\right) \\ &= \left(1 - \frac{T_c}{T}\right)^{-\nu} \times p(1/T). \end{aligned}$$

Per definition $p(1/T)$ remains finite for $T \geq T_c$ and it is assumed to be constant in the approximation which leads to equation (3). In the high temperature limit therefore

$$\xi(T)/\xi_0 = (1 + \nu T_c/T + \dots) p(1/T)$$

where $\nu T_c = \theta'$ should remain finite to insure that the expression remains analytic (that it can be expanded in terms of $1/T$). With this constraint equation (3) generates essential singularities [18] when T_c cancels as ν diverges like θ'/T_c and one obtains equation (1) which would be appropriate for « a lower critical dimensionality » for which $T_c = 0$. The exponents, it is expected, should depend only on features which survive when ξ diverges, like the space or the spin dimensions. Systems which share such features belong to the same universality class. In non frustrated systems the universality even resists the introduction of some disorder [19]. Very little however is known on the effect of frustration so that the principle of the universality of $z\nu$ in spin glasses is not evident.

One difficulty with equation (3) is that it has three parameters. We propose in this case, to take the logarithm and differentiate to obtain [18]

$$P_\tau(T) = - \frac{\partial \ln T}{\partial \ln \tau} = \frac{T - T_c}{z\nu T_c} = \frac{T - T_c}{\theta} \quad (5)$$

which has only two parameters. It is then possible, by differentiating the experimental data to

obtain an objective criterion which determines these two parameters in a unique step as the data of $P_\tau(T)$ vs. T should align on a straight line which intersects the T axis at T_c and the $T=0$ axis at $-1/z\nu$. The same line intersects the $P_\tau(T)=1$ line at $T_c + \theta$ where $\theta = z\theta' = z\nu T_c$.

2. Dynamic scaling and the glass transition.

Figure 2a shows the $P_\tau(T)$ plot for representative glass-forming systems between strong and fragile [20]. The following features are remarkable :

a) In many systems on the fragile side we have a high temperature regime where the viscosity is well approximated by a power law with exponent $z\nu \sim 6.2 \pm 1$.

b) On decreasing the temperature to T_R close to T_c , most of these systems escape towards a situation of lower T_c hence of higher $(z\nu)'$ and reach eventually Arrhenius (strong) behaviour which corresponds to the $T_c = 0$ limit. We have found $(z\nu)' \sim 15$ in $\text{KNO}_3\text{Ca}(\text{NO}_3)_2$ and LiCl , $5.75 \text{ H}_2\text{O}$ [21], $(z\nu)' \sim 23$ in propylene carbonate [4], $(z\nu)' \rightarrow \infty$ in B_2O_3 or α -Phenyl-o-Cresol. In these last two systems where the data cover a particularly wide range of decades

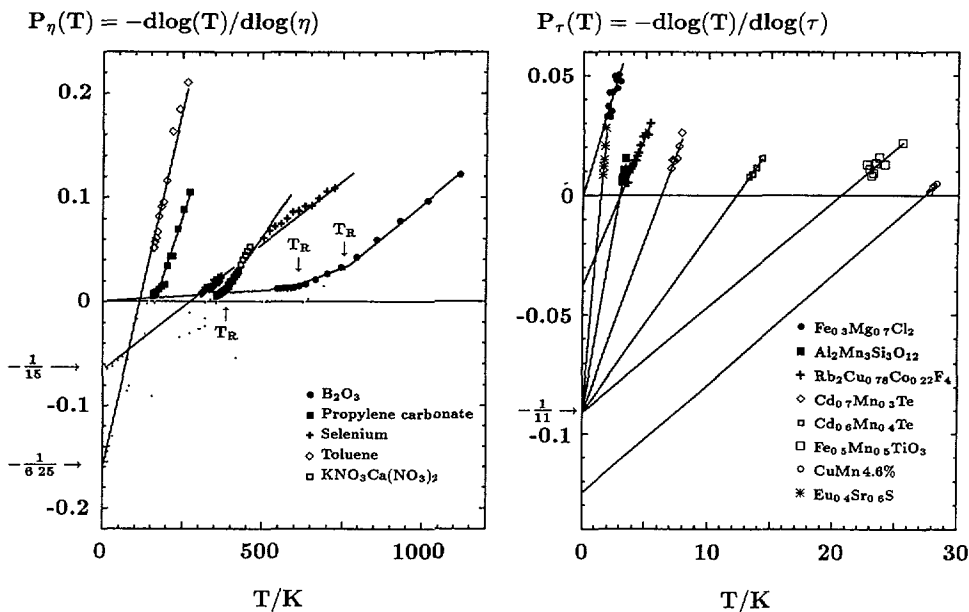


Fig. 2. — A plot of $-\frac{\partial \ln T}{\partial \ln \eta}$ vs. T shows that the data in the high temperature regime of several different fragile systems point towards an average $P(\eta)$ value which is of the order of -0.16 and therefore can be described with a universal dynamic exponent $z\nu$ of the order of 6.25 . The plot of figure 2a dramatizes the cross-over, at T_R , towards a stronger regime of lower T_c and larger $z\nu$. Strong systems, like silica, and intermediate systems like amorphous selenium can be entirely described with a unique regime where $z\nu \sim \infty$ and $z\nu \sim 15$ respectively. Notice that for selenium the same power law accounts for the data on both sides of a crystallisation window. In figure 2b CuMn, EuSrS or Mn aluminosilicate are on the fragile side as compared to stronger anisotropic spin glasses like $\text{Rb}_2\text{Cu}_{0.78}\text{Co}_{0.22}\text{F}_4$ or $\text{Fe}_{0.3}\text{Mg}_{0.7}\text{Cl}_2$ which exhibit a larger dynamical exponent. We do not know of any data which show a sign of a cross-over as observed in glass forming liquids. But this is perhaps because the data cover in general a much smaller range of decades.

one would easily be tempted to find three successive regimes with increasing exponents ($z\nu \sim 6$, then $z\nu \sim 15$, and then $z\nu \rightarrow \infty$). These changes of regime, although they are particularly apparent in the $P_\tau(T)$ diagram have also been noticed otherwise [22] and these systems are the same where Arrhenius deviations to the Fulcher law have been reported. In general no expression with 4 coefficients seems capable to describe both regimes with a unique set of parameters.

c) A number of systems like Se [23], glycerol [24], $\text{Pd}_{77.5}\text{Cu}_6\text{Si}_{16.5}$ [16] can be described by a unique power law with an intermediate $z\nu \sim 15$. Silica or GeO_2 on another hand can entirely be described by an Arrhenius law ($z\nu \sim \infty$). Perhaps these systems are the archetypes for two other (one intermediate, one strong) classes of universality. However it is not clear in these systems that the temperature has been raised high enough to reach the « universal » fragile regime. For example, in silica, we have not yet reached the viscosities below 1 poise which make the difference and exhibit fragile behaviour in the fragile systems.

d) Conversely one wonders if $T_c = 0$ ($z\nu \rightarrow \infty$) is not the ultimate limit for all systems and whether in these liquids where, like LiCl , $5.75 \text{ H}_2\text{O}$, an intermediate $(z\nu)' \sim 15$ to 25 is measured one would not reach still larger exponent values if the data for higher viscosities were available.

The discontinuities of the slope of the $P_\tau(T)$ dependence which are observed at T_R where ξ is finite can be interpreted as a cross-over from one to another regime which can both be described by a power law of the form of equation (4). But, as $P_\tau(T)$ is continuous, these two power laws should be tangent to one another at the cross-over point. The tangent itself intersects the $T_g/T = 0$ axis in the Arrhenius plot (Fig. 1) at τ'_0 such that [15]

$$\ln(\tau'_0/\tau_0) = -z\nu \left[\ln \left(1 - \frac{T_c}{T_R} \right) + \frac{T_R}{T_R - T_c} \right]. \quad (6)$$

The prefactor of any Arrhenius law fitted to the data in the regime below the cross-over would have this value and would appear unphysically small when compared with the generally accepted $\tau_0 \sim 10^{-13} \text{ s}$ (but τ'_0 is the result of an extrapolation to the high T limit of the analytical continuation of a law which is only valid below T_R). In any case τ'_0 will appear depressed if T'_c is decreased and the effect will be stronger when T_R is closer to T_c . The existence of a small τ_0 in some of the strongest systems like silica thus supports the conjecture that there exists a cross-over from a more fragile state at a higher temperature. Notice that the continuity of the $P_\tau(T)$ curve is a significant point in our description : equation (4) defines τ as a power of ξ ; the static scaling hypothesis on another hand defines the Gibbs potential as a power of ξ : we have $G/T \approx \xi^d$ in zero field. Discontinuities of $P_\tau(T)$ would mean a discontinuity of the entropy $S(T) = -\partial(G/T)/\partial(1/T)$ and a first order transition.

The suggestion therefore is that in supercooled liquids the situation is well described at high temperatures by a power law with a finite T_c where a correlation length would diverge, but that, on decreasing the temperature the system escapes by a (succession of) cross-over(s) towards a situation of lower T_c . The whole procedure is interrupted at the glass transition near T_g when the relaxation time becomes large compared to any reasonable experimental time and we are no longer able to observe the system at equilibrium. The case of B_2O_3 is particularly interesting because this system goes all the way through from a fragile to a strong situation. But in neither case even in the high-temperature fragile regime it is described with a « reasonable » τ_0 . This is certainly related to the fact that, in this system, undamped transverse sound waves are observed still much over the melting point and up to 1 300 K [25]. In our interpretation we certainly would expect something (another cross over to some

different situation) to occur at a still higher temperature. In other words it seems that this is a liquid which still has to melt.

In figure 2b we show that the $P_\tau(T)$ plot relative to the data of figure 1b. This plot calls the following comments :

a') Most spin glasses should point $z\nu \sim 8 \pm 2$ according to the admitted truth based mainly on early experiments on R.K.K.Y. systems like CuMn and on systems like EuSrS [8, 15]. The data in FeMnTiO₃, Cd_{0.6}Mn_{0.4}Te, Cd_{0.7}Mn_{0.4}Te or Mn aluminosilicate point a larger $z\nu \sim 11 \pm 2$. The criteria which permit to define the $\tau(T)$ dependence have been improved since the first experiments were performed [9-13]. Certainly it would be useful to reconsider all the former data with the new criteria. After this is done it is not completely clear if two classes or one will be needed to describe all these fragile systems. We had no difficulty to superimpose the data on a single curve shown in figure 3 with a $z\nu \sim 11$. The superposition of the CuMn data on the same curve is possible if we accept a $\tau_0 \sim 10^{-20}$ s. We should stress that the data for this sample are of 3 distinct origins for 3 very different ranges (neutrons, a.c., d.c...) and this implies further uncertainties.

b') Fe_{0.3}Mg_{0.7}Cl₂ or Rb₂Cu_{1-x}Co_xF₄ which point respectively $z\nu \sim \infty$ and $z\nu \sim 25$ show a different (stronger) behaviour. More examples would be needed in order to confirm that

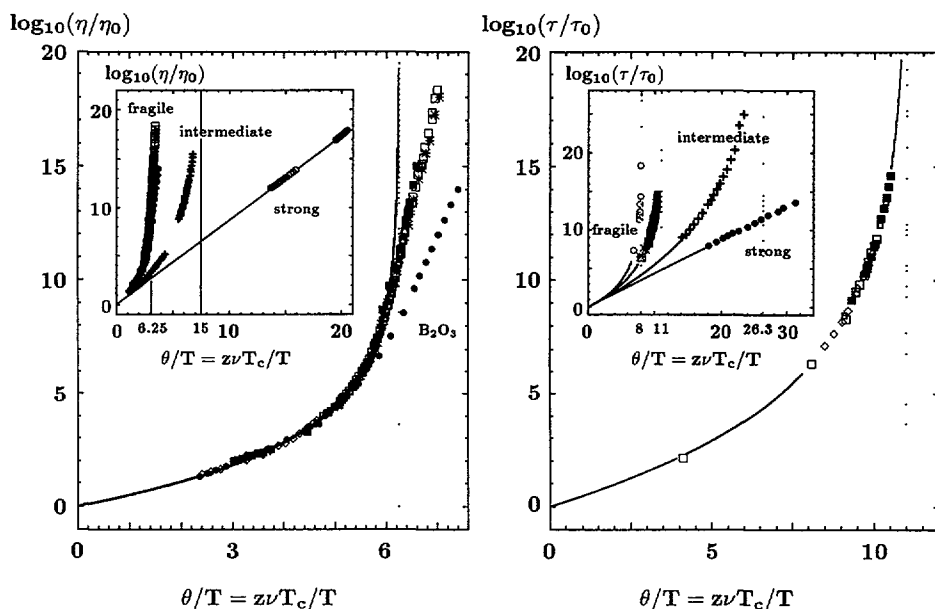


Fig. 3. — In the plot of $\log(\eta/\eta_0)$ vs. θ/T shown in the insert of figure 3a the data of figure 1a display three different universality behaviours : one fragile with $z\nu \sim 6.2$, one intermediate with $z\nu \sim 15$ and one strong with $z\nu \rightarrow \infty$. The curves representative of a pure case are tangent to each other at their common origin where $\eta = \eta_0$ (see Tab. I) and the positions of the asymptotes where η would diverge is at $\frac{\theta}{T} = z\nu$. For a mixed case which crosses over at T_R to a stronger universality class the contact point between two behaviours is at T_R so that τ_0 and the position of the new T_c depend on the T_R over T_c ratio. The main figure shows in more detail that B₂O₃ which belongs to the same fragile class as KNO₃-Ca(NO₃)₂, propylene carbonate, toluene, α -phenyl-o-cresol, salol crosses over towards stronger behaviour at a different T_R/T_c ratio. The same plot for spin glasses (Fig. 3b) shows similar features but no indication for a cross over from one to another regime.

these systems are evidence for two other (one strong, one intermediate) classes of universality. Also in $\text{Rb}_2\text{Cu}_{0.78}\text{Co}_{0.22}\text{F}_4$ variations of $T_g(\omega)$ over 16 decades in frequency have been obtained by extrapolating with the Cole-Cole law measurements which actually cover a more conventional range of about 5 decades. This method assumes that the phenomenological Cole-Cole law is actually correct from the high frequency to the low frequency limit and that the data in these 2 regimes are consistent with the same values of the parameters. Right or wrong, this is still a hypothesis which needs further confirmations and can have an incidence on the « experimental data » which are derived for $\tau(T)$.

c') None of the spin glasses exhibits a clear cross over from one to another regime as observed with the glass forming liquids and a unique exponent characterizes the whole regime down to the « glass transition » where ergodicity is lost. But the relative range of $1/T$ where we have data is much smaller than for structural glasses (Fig. 2). In $\text{Rb}_2\text{Cu}_{1-x}\text{Co}_x\text{F}_4$

Table I. — We show for the different glass-forming systems « experimental » values of $z\nu$, T_c and η_0 which come from a best fit to the $P_\tau(T)$ plot of figure 2a. The corresponding « universal » values which correspond to the fits shown figures 1 and 3 have been obtained by imposing an average value of $z\nu$. For most fragile systems where $z\nu$ is of the order of 6.25 a cross-over at T_R towards a stronger regime is observed when T_c is approached. The data corresponding to this second regime when it exists are shown on the lower line for each system. T_g is the temperature where the viscosity becomes of the order of 10^{13} P and ergodicity is lost. The validity range of this regime is between T_R and T_g so that η_0 which is the $T \rightarrow \infty$ limit of the analytical continuation of the law appropriate to this range is not expected to have any simple, direct physical meaning (see Eq. (6)).

Glasses	$z\nu$	T_c/K	θ/K	η_0/P	T_R/K T_g/K	With average $z\nu$ imposed				Ref. of data
						$z\nu$	T_c/K	θ/K	η_0/P	
B_2O_3	6.65 ∞	618 0	4 110 46 350	1.06 2×10^{-24}	610 550	6.25	632	3 950	1.19	[3]
Propylene carbonate	5.31 20.5	173 143	921 2 935	1.9×10^{-4} 4.5×10^{-11}	187 154	6.25 15	166 148	1 040 2 220	1.26×10^{-4} 6.4×10^{-8}	[4]
Selenium	13.26	285	3 785	8.8×10^{-4}	304	15	284	3 830	8.6×10^{-4}	[23]
Toluene	4.76	130	620	3.4×10^{-4}		6.25	117	731	2.2×10^{-4}	[5]
KNO_3 $\text{Ca}(\text{NO}_3)_2$	4.96 15.83	368 327	1 825 5 170	3.4×10^{-4} 1.55×10^{-10}	383 340	6.25 15	356 329	2 232 4 930	8.5×10^{-5} 4.5×10^{-10}	[5]
α -Phenyl O Cresol	7.23 ∞	230 0	1 660 33 200	7.66×10^{-6} 2×10^{-56}	241 210	6.25	233	1 455	2.3×10^{-5}	[2]
Salol	5.34 ∞	243 0	1 300 31 200	1.9×10^{-5} 1.3×10^{-51}	254 212					[2]
SiO_2	∞	0	62 353	1.4×10^{-6}	1 440					[6]

moreover, a cross-over from fragile to intermediate state at a high temperature would permit us to account for the unphysical $\tau_0 \sim 10^{-19}$ s which is found with our interpretation. Perhaps such a cross-over is suggested by the experimental data (Fig. 1).

d') In spin glasses the stronger regime is clearly associated with a lower dimensionality. Thus $\text{Fe}_{0.3}\text{Mg}_{0.7}\text{Cl}_2$ is reputed Ising and two-dimensional.

We show in the insert of figure 3a a representation of $\ln(\tau/\tau_0)$ vs. θ/T where $\theta = z\nu T_c$ has been defined in equation (5). We have found an average $z\nu \sim 6.2 \pm 2$ in the fragile regime of the fragile systems and determined T_c and τ_0 which best fit the data for this value (see Tabs. I and II). The fits are shown in figures 1 and 3. Selenium is fitted with a $z\nu \sim 13.5$ and is given here as an example of an intermediate class which would also include e.g. glycerol or PdCuSi. Silica is shown as an example of the strong systems for which $T_c = 0$ and $z\nu \rightarrow \infty$ and which would also include GeO_2 , etc. In this representation all systems have the same slope at their common high temperature origin where $\tau = \tau_0$ and they exhibit a divergence at $\frac{\theta}{T} = z\nu$ which defines T_c . The main figure emphasises the situation of several fragile systems which cross over to intermediate behaviour at a T_R/T_c ratio which is of the order of 1.1 in o-terphenyl, salol or propylene carbonate and of the order of 1.2 in B_2O_3 .

Figure 3b is the corresponding plot for spin glasses. We have imposed an average $z\nu \sim 11$ for the « fragile » systems shown and $\text{Rb}_2\text{Cu}_{0.78}\text{Co}_{0.22}\text{F}_4$ and $\text{Fe}_{0.3}\text{Mg}_{0.7}\text{Cl}_2$ are proposed as illustrations for intermediate and strong behaviours with $z\nu \sim 25$ and $z\nu \sim \infty$ respectively.

Table II. — We give for the different spin-glasses the values of $z\nu$, T_c and τ_0 which come out from a best fit to the data of figure 2b. The corresponding values obtained with an average $z\nu = 11$ imposed correspond to the fits of figures 1 and 3.

						With average $z\nu$ imposed				
Spin glasses	$z\nu$	T_c/K	θ/K	τ_0/s	T_g/K	$z\nu$	T_c/K	θ/K	τ_0/s	Ref. of data
$\text{Fe}_{0.3}\text{Mg}_{0.7}\text{Cl}_2$	∞	0	56.72	1.48×10	1.56					[10]
Mn alumi- no-silicate	11.06	2.95	32.74	2.56×10^{-16}	3.00	11	2.96	32.58	1.81×10^{-16}	[9]
$\text{Cd}_{0.7}\text{Mn}_{0.3}\text{Te}$	10.1	6.34	64.2	3.16×10^{-12}	6.53	11	6.29	69.17	9.62×10^{-13}	[12]
$\text{Rb}_2\text{Cu}_{0.78}\text{Co}_{0.22}\text{F}_4$	26.33	3.02	79.54	5.58×10^{-19}	3.52					
$\text{Cd}_{0.6}\text{Mn}_{0.4}\text{Te}$	10.84	12.26	133	4.89×10^{-14}	12.58	11	12.25	135	3.66×10^{-14}	[12]
$\text{Fe}_{0.5}\text{Mn}_{0.5}\text{TiO}_3$	12.87	20.21	260.1	10^{-14}	21.05	11	20.64	227	7.74×10^{-14}	[13]
CuMn 4.6 %	5.73	27.47	157.3	1.3×10^{-13}	27.50	8	27.3	218.41	5.3×10^{-16}	[8]
$\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$	9.48	1.48	14.07	2.20×10^{-11}	1.53	11	1.46	16.04	4.18×10^{-12}	[15]

3. Discussion.

We have shown that critical slowing down offers an alternative to the phenomenological Fulcher law to fit the viscosity of glasses or the relaxation times of spin glasses. Besides it introduces some elements of rationalisation such as the convergence (shown in Fig. 2) of the $P_\tau(T)$ lines towards a limited number of « universal » exponent values. Until now, however, no conclusive evidence has been found in favour of either law from the strict point of view of the quality of the fits. The reason is apparent in figure 2. With equation (2) we have

$$P_\tau(T) = - \frac{\partial \ln T}{\partial \ln \tau} = \frac{(T - T_0)^{\sigma+1}}{\sigma B^\sigma T} \quad (7)$$

so that we expect a power law rather than its tangent which would correspond to equation (5) but the slow dynamics forbid the region near T_c where the difference would be conclusive. The case however of selenium deserves special consideration. The measurements are interrupted on lowering the temperature when the nucleation of crystallites becomes too considerable but it is possible to obtain another set of data by melting the quenched amorphous state in a range near T_g where the growth of the crystallites can be maintained sufficiently slow. In figure 2, we see that a unique exponent is necessary when two distinct Fulcher laws were needed. This stresses the continuity of the physics which describes both sides of the crystallisation window and its relevance to critical slowing down rather than to activated dynamics.

Another question is relative to frustration which is believed to be responsible for the slow dynamics in spin glasses. The question then is what would be the origin of frustration in glasses? A key to the answer could be the observation which is made of a highly universal regime at high temperatures in the fragile systems. The presence of the same $z\nu \sim 6$ in systems *a priori* as different as Salol and $\text{KNO}_3\text{Ca}(\text{NO}_3)_2$ reflects the fact that there is little energy difference between different crystalline structures which may eventually set-in. Hence the suggestion of a multivalleyed phase space with nearly equivalent minima: hence the presence of frustration as each structure has no compatibility with the others. In contrast with the spin-glass case where there is one response for one situation, fragile liquids seem capable to adapt their criticality through a succession of liquid-to-liquid transitions towards stronger situations. There are many general reasons by which one would justify that a phase space becomes stronger (or more anisotropic if we interpret the situation at the light of the spin glass data). This may be that one or several particular valleys are favoured (e.g. at temperatures close to the melting point a deeper valley ought to correspond to the crystalline structure of the system); on the contrary, valleys which have some probability at short range become hampered when ξ grows (e.g. the periodic structures which fill the 3d space are favoured with respect to 5-fold symmetries which do not). We would expect a similar situation in a spin glass layer where ξ would become of the order of the thickness and the correlations which initially developed in 3 dimensions would become restricted to the plane. Castaing and Souletie [17] observe that the model of activated dynamics describes critical dynamics in a phase space which shrinks as ξ grows. Indeed equation (7) can be interpreted as featuring a continuous evolution towards stronger situations as the slope of the $P_\tau(T)$ curves decreases and cancels when T_c is approached. The problem is that in actual liquids this evolution is realized by steps and a succession of cross-overs is more appropriate to describe the successive tiers of this hierarchy. This is particularly clear on the $P_\tau(T)$ plot which we are tempted to describe with a succession of segments rather than with the continuous curve or the hierarchy of tiers that the activated dynamic equation would imply (see Fig. 2). It would be of course very interesting to check whether each tier can be related to particular physics that a particular system would

exemplify (e.g. whether a $z\nu \sim 15$ would mean a polymeric situation as is observed in selenium²⁶).

It seems logical, in the framework of the above picture, to consider that the fluctuations of the ordered amorphous phase are complicated intermixtures of the different pure phases which constitute the different valleys. Among these fluctuations pure crystallites of these phases would occur with a certain probability and, among those crystallites, those which have the appropriate symmetry would act as the germs which would motivate the occurrence of the first order melting transition at T_M . The T_M/T_c ratio is an important parameter in this picture. If T_c is much smaller than T_M the liquid becomes a crystal at T_M but the model explains viscosity increases that a first order transition would not justify. If T_c is close to T_M , then high viscosities are reached in one or the other of the successive regimes which we have described above. Pure crystallites of only one phase become unlikely at large ξ and they would grow very slowly. High viscosity gradients which can reach orders of magnitudes in a few degrees affect the transport in the liquid towards the liquid to solid interface: the definition of what is liquid and what is solid is in question; it becomes frequency dependent! The ultimate consequences are known: the freezing becomes homogeneous and we have a glass. An intermediate case would be that of Se where a crystallisation window hampers viscosity measurements in an intermediate range of temperatures.

4. Conclusion.

We have found that spin glasses show similar features from fragile to strong as usual glass forming liquids. These features can be accounted for in the framework of usual scaling theory where the strong limit would correspond to a lower critical dimensionality. Data on real glasses of the fragile side suggest that the strong regime could result from a cross-over from a universal high temperature fragile situation occurring at T_R which depends on the microscopic properties of the system. It is the magnitude of the T_R/T_c ratio which decides whether the system will appear stronger or more fragile in the fixed experimental window. Another mark of the cross-over would be the presence of an unphysical τ_0 value in the regime below the cross-over. Before we finish we should mention that a power law behaviour for shear viscosity is also predicted by mode-coupling theory which has been rather successful in explaining some data on hard system glasses and polymeric ones as well [27]. It seems premature to decide whether the present analysis is or is not contradictory with these calculations which do not introduce any prerequisite that there should exist a diverging length ξ in the system. In any case the present description is phenomenological. The existence of a phenomenology, even successful, does not exempt us of seeking for a microscopical theory and we believe the reverse also should be true, specially with mode-coupling theory whose calculations are by no means transparent.

Acknowledgements.

We are grateful to P. Chieux for several illuminating discussions and to G. Faivre and Ch. Simon who pointed out to us the interesting situation of Se. Part of this work was motivated by the workshop on « Glasses, biomolecules and evolution » in May 1990 at the Santa Fe Institute that one of the authors (J.S.) wants to thank for its hospitality.

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