Journal of Alloys and Compounds, 211/212 (1994) 327–332
JALCOM 055

Theoretical basis and general applicability of the coupling model to relaxations in coupled systems*

K.L. Ngai[a], Y.-N. Wang[b] and L.B. Magalas[c]
[a]*Naval Research Laboratory, Washington, DC 20375-5320 (USA)*
[b]*National Laboratory of Solid State Microstructure, Nanjing University, Nanjing (China)*
[c]*Academy of Mining and Metallurgy, PL-30059 Cracow (Poland)*

Abstract

The coupling model proposed more than a decade ago for the description of relaxations in complex correlated systems by one of us (KLN) have been shown repeatedly to be applicable to amorphous polymers, viscous liquids, the glassy state, metallic glasses, glassy ionic conductors and *etc.* In these examples, the relaxing species are dense packed and mutually interacting. As a consequence, the constraints between them have a strong influence on the relaxation dynamics. Very recent quasielastic neutron scattering experimental data as well as molecular dynamics simulation results which provide dramatic proof of the validity of the coupling model will be discussed. The applicability of the coupling model to some relaxations in metals including precipitates and the Snoek–Köster relaxation is less obvious but has recently been proposed, justified and demonstrated to be relevant by one of us (YNW). The applicability of the coupling model to these and other problems of immediate interest to participants of ICIFUAS-10 will be summarized and discussed. For Snoek–Köster relaxation, additional experimental data in ultra-high purity iron containing low concentration of carbon obtained by one of us (LBM) show characteristics that cannot be explained by Seeger's model of thermally activated formation of kink pairs on screw dislocations in the presence of foreign interstitial atoms, but are consistent with the coupling model.

---

1. Recent direct experimental proofs of the coupling model

The coupling model since it was first introduced in 1979 [1, 2] has repeatedly been found to offer a faithful description of cooperative relaxation processes in dense packed correlated systems [3, 4]. It proposes the existence of a temperature-insensitive crossover time, t_c, separating two regimes in which the dynamics of relaxation are different. Although the existence of t_c is guaranteed from theoretical considerations [1, 2, 5, 6], its exact magnitude is not known *a priori*. Order of magnitude estimates of t_c obtained indirectly from comparing experimental data with the predictions of the coupling model (to be described immediately) have located it approximately within the range $10^{-12}<t_c<10^{-11}$ s for local segmental relaxation in polymers and small molecular van der Waal liquids [7–10]; and in the range of $10^{-13}<t_c<10^{-12}$ s for ionic motion in glassy ionics conductors [11]. At short times, for $t<t_c$, the units relax independently as if the mutual interactions between them have no effect with a rate W_0. However, for $t>t_c$ the effects of their mutual interactions take hold and slow down the independent relaxation rate W_0. The result is that the relaxation rate, after being averaged over all the units, has the time-dependent form $W(t) \propto W_0(t/t_c)^{-n}$, where n is the coupling parameter that can be considered as an indicator of the degree of correlation or cooperativity in the relaxation process originating from mutual interactions. The value of the coupling parameter falls within the range $0<n<1$. As far as the normalized correlation function, $C(t)$, of the relaxation process is concerned the immediate consequences are:

$$C(t)=\exp(-t/\tau_0) \quad \text{for } t<t_c \tag{1}$$

$$C(t)=\exp-(t/\tau^*)^{1-n} \quad \text{for } t>t_c \tag{2}$$

and the important relation between τ^* and τ_0 given by [12]

$$\tau^*=[t_c^{-n}\tau_0]^{1/(1-n)} \tag{3}$$

Relation (3) can be easily obtained from the condition of continuity of the correlation functions (1) and (2) that hold separately in the two time regimes at t_c, *i.e.*

$$\exp(-t_0/\tau_0)=\exp-(t_0/\tau^*)^{1-n} \tag{3a}$$

These three coupled relations first proposed more than 14 years ago [1, 2] have remained unmodified. They look deceptively simple but, in reality, when used together they have explained a host of important and

---

*Invited paper.

0925-8388/94/$07.00
SSDI 0925-8388(93)00055-4

often critical experimental facts of relaxation phenomena in a number of fields [4]. Most effective is the relation (3) between τ^* and τ_0 through which many previously puzzling properties of viscoelastic properties of polymers, diffusion and conductivity relaxation in glassy as well as disordered crystalline ionic conductors, and other materials have been explained [4]. In all these applications, the experimental data usually considered were taken in the long time regime of $t \gg t_c$ which preclude direct observation of the crossover from exponential relaxation (1) to stretched exponential relaxation (2). Nevertheless, with the assumption of a temperature-insensitive t_c, the two coupled predictions (2) and (3) have been remarkably successful in explaining experimental data and molecular dynamics simulations.

Naturally, in view of the good agreements between the coupling model and experimental data found, it is desirable to directly verify the coupling model in its entirety (*i.e.* existence of a temperature-independent t_c and results (1)–(3)) either by a rigorous theoretical proof or by performing experiments in time windows that span across t_c. Solution of this problem from the theoretical physics side is not easy, principally because this is a most difficult problem involving both non-integrable many body interactions and irreversible statistical mechanics. Recent attempts based on non-linear classical Hamiltonian dynamics [6] have produced encouraging results (including the existence of a temperature-independent t_c and the time-dependent relaxation rate $W_0(t/t_c)^{-n}$), although there is still a long way to go before the problem can be considered rigorously solved. Non-linear classical Hamiltonian mechanics (the dynamics of which is popularly known to the general public as chaos) is the most fundamental starting point to describe most, if not all, of the systems we discuss in this paper. The basic relaxing units in all of our systems are interacting with each other. The interaction potentials are invariably anharmonic, which give rise to non-linear dynamics of evolution and lead to unusual (self-similar islands-around-island) structure in phase space (*i.e.* chaos). It is the formation of this self-similar structure in phase space which causes the slowing down in the relaxation rate. Before that, the relaxation of the basic units remain independent of each other with rate W_0. Therefore the crossover time t_c of the coupling model is given by the time-scale of the formation of the first "island". Since this t_c is determined by Hamiltonian mechanics and not by thermodynamics, its magnitude will be independent of temperature as the coupling model has postulated. It follows from non-linear Hamiltonian dynamics that the island formation time is shorter for stronger interaction. Hence we expect t_c to have the same dependence on the interaction strength. At times longer than t_c, the transport in phase space which governs the relaxation rate will be slowed down increasingly with time because of the corresponding increasing complexity in the structure of the phase space. Our theoretical developments in ref. 6 has led to the time-dependent relaxation rate $W_0(t/t_c)^{-n}$ where $0<n<1$ as stated above.

2. Polymers

On the other hand, direct experimental test of the coupling model can be performed by using time-of-flight (TOF) neutron scattering. This experimental technique can monitor the relaxation dynamics in the short time window of $10^{-13}<t<10^{-11}$ s where the expected crossover of relaxation dynamics at t_c can be observed directly. When complemented by other neutron scattering techniques (*e.g.* neutron back-scattering and neutron spin echo) [13], the time window can be extended to the range of $10^{-13}<t<10^{-8}$ s and the experimental data will provide stringent tests of the coupling model. Recent neutron TOF experimental work of Colmenero and coworkers in polyvinylchloride, an amorphous polymer, [14, 15] has proved explicitly the existence of a crossover time $t_c = 1.7$ ps. At times prior to t_c the experimental data verify that the relaxation is governed by a Debye process as indicated by eqn. (1) with the activation enthalpy of τ_0 the same as expected for the local segmental relaxation of polyvinylchloride in the absence of intermolecular interactions, *i.e.* 6 kcal mol^{-1} for the conformation energy barrier of an isolated polyvinylchloride chain. Across $t>t_c$, the TOF data clearly shows a switch from the $\exp(-t/\tau_0)$ for $t<t_c$ to the stretched exponential, $\exp-(t/\tau^*)^{1-n}$, as described by eqn. (2). The magnitude of t_c is independent of temperature. These experimental correlation functions in the two time regimes match their values at $t=t_c$ in exactly the same manner as eqn. (3a). From this it follows that eqn. (3) is automatically verified by neutron TOF data. Another direct experimental verification of the coupling model comes from molecular dynamics simulation [16–18] which can be performed now rather efficiently in the short time range of $10^{-15}<t<10^{-8}$ s. Recently molecular dynamics simulations have been performed in polymers [16]. The results of the simulation show the existence of a temperature-insensitive t_c. The magnitude of t_c found from molecular dynamics simulation for polymers is almost the same as that obtained from neutron scattering data, *i.e.* 2 ps. Thus we can conclude from this discussion that we now have direct microscopic experimental proof of the validity of the coupling model. These most recent exciting developments together with the unparalleled successes of the coupling model in explaining the low frequency (or $t \gg t_c$) relaxation data of polymers (we shall not list the successes made in the past here, but instead refer the reader to ref. 4) make it compelling to accept the coupling model.

3. Ionic conductors

Neutron TOF experiments in fast ionic conductors [19] have also been performed with the purpose of measuring the short time diffusion dynamics, which according to the coupling model should be governed by the independent relaxation rate

$$W_0 \equiv (\tau_0)^{-1} = (\tau_\infty)^{-1} \exp(-E_a/kT) \tag{4}$$

where E_a is the real microscopic energy barrier to ion jump from site to site and $(\tau_\infty)^{-1}$ the attempt frequency of the jump. Interestingly, the neutron TOF data in several fast ionic conductors [19] have found the activation energy E_a in this short time regime to be considerably smaller than the activation energy E_a^* of ion diffusion found in the long time regime by more conventional methods (the correlation function was not available in these experimental works). As can be verified by substituting expression (4) for τ_0 into eqn. (3), the coupling model indicates that ion diffusion correlation time τ^* is given by

$$\tau^* = \tau_\infty^* \exp(E_a^*/kT) \tag{5}$$

where the apparent preexponential is related to τ_∞ as

$$\tau_\infty^* = (t_c^{-n}\tau_\infty)^{1/(1-n)} \tag{6}$$

and

$$E_a^* = E_a/(1-n) \tag{7}$$

The activation energy of ion diffusion E_a^* has been measured using conventional techniques (dielectric and mechanical) at much lower frequencies compared with that of neutron scattering. The coupling parameter n has also been determined by comparing the experimental frequency dispersion with that predicted by eqn. (2). With all three quantities appearing in eqn. (7) independently determined, eqn. (7) is a stringent test of the coupling model. Remarkably it was verified [19].

Molecular dynamics simulation of ion motion has been performed for sodium ions in sodium silicate glasses [17]. Again, in this case a temperature-independent crossover time t_c has been found which is about an order of magnitude shorter than that for polymers. The ionic interaction in sodium silicate glasses is stronger than the van der Waal forces in polymers. Thus the shorter t_c found in the former is consistent with what we expect from the theoretical consideration given above [6], and from previous indirect deductions from low frequency relaxation data [7, 8, 11].

Ample experimental data have shown that the dielectric, nuclear spin lattice relaxation or the mechanical loss peak of the diffusing ions is broadened considerably compared with the Debye form for non-interacting ions. The relaxation time, $\tau^* = \tau_\infty^* \exp(E_a^*/kT)$, that corresponds to the experimentally observed relaxation loss maximum has an activation energy E_a^* much larger than the energy barrier E_a experienced by an individual ion [4]. The preexponential τ_∞^* differs from the reciprocal of the attempted angular frequency of vibration of an ion [4] and it even has an anomalous isotope mass dependence [4, 20]. Many dramatic effects, including those mentioned here, that interactions have on diffusion can be accounted for by a theory of relaxation that has built in the physics of many body interactions and correlations such as the coupling model. If one considers together the neutron TOF data, the most recent molecular dynamics simulation data [17] and the low frequency relaxation data [4] of glassy ionic conductors as well as disordered crystalline ionic conductor such as Naβ-Al$_2$O$_3$ [21–25], then the inevitable conclusion has to be made that the coupling model is in excellent agreement with all the experimental data.

The result that follows from eqn. (7) when the relaxation is highly coupled to have a large coupling parameter n and the activation energy E_a^* obtained by conventional relaxation methods can be much larger than the actual energy barrier E_a, has repeated occurrence in other fields of research [4]. In the field of internal friction, this has been invoked by other workers in connection with high temperature relaxation in aluminium [26, 27]. While this applications appears to be interesting, yet more works, both experimental and theoretical, are needed before one is justified in applying the coupling model to these areas. In the next section we shall focus on a relaxation process in metals, the Snoek–Köster relaxation, that we have confidence in applying the coupling model to. An explanation of this relaxation by a theory based on the coupling model [3, 28] will be discussed.

4. The Snoek–Köster relaxation

The summary of the coupling model given above has paved the way for the discussion of its applications to a problem of internal friction in metals. It is by now generally agreed that the Snoek–Köster (SK) relaxation [29] is caused by the bowing out of a dislocation segment under applied stress, dragging along with it the surrounding atmosphere of rather high density of interstitial solute atoms (ISAs). This mechanism for SK relaxation first suggested by Schoeck [30] accounts for many established experimental facts and has been widely accepted to be basically correct. Seeger proposed a theory that involves the dragging of mobile ISAs by a dislocation but in addition, the thermally activated formation of kink pairs in a $a_0/2\langle 111\rangle$ screw dislocations. In this theory the activation enthalpy H^{SK} is essentially the sum of the formation enthalpy $2H_{\mathrm{k}}$ of kink pairs in $a_0/2\langle 111\rangle$ screw dislocations and the migration energy H^{M} of foreign interstitials, *i.e.* $H^{\mathrm{SK}}=2H_{\mathrm{k}}+H^{\mathrm{M}}$. The quantity $2H_{\mathrm{k}}$ can be identified with the activation enthalpy of the γ-relaxation, known also as the Bordoni peak due to kink pair formation on screw dislocations. This theory retains all the desirable features of Schoeck's model and provides a viable explanation of H^{SK}.

In spite of the fact that Seeger's theory is overall quite successful, there are still issues that have remained unresolved. The major problem we see in both Schoeck's and Seeger's theories is the neglect of the interactions between the ISAs and the effects these interactions have on the migration of ISAs. It is generally recognized that the local concentration of foreign interstitial atoms along or near the dislocation core can be quite high and certainly much higher than the concentration of interstitial atoms in the same metal before cold work. Even in the Snoek relaxation observed without cold work, Haneczok *et al.* [33] have shown the effect of long range interactions between the interstitial atoms through the shift of the Snoek maximum to higher temperatures, and the broadening of the Snoek peak with increasing interstitial atom content. It is thus clear that the interactions between the ISAs in the Cottrell atmosphere surrounding the dislocations will be even stronger than the long range interaction between the interstitial atoms seen in the Snoek relaxation. Therefore the effects of interactions should be even more prominent in the Snoek–Köster relaxation than in the Snoek relaxation, and cannot be neglected without justification as in Schoeck's and Seeger's treatment. This was first pointed out by Sun *et al.* [28] who also constructed a theory of SK relaxation based on Schoeck's original model, but on top of it used the coupling model to take into account the correlated diffusive motion of foreign atoms in the Cottrell cloud surrounding the dislocations when dragged by the dislocations. The jump relaxation time, τ_0, of the ISA is given by $\tau_0=\tau_\infty{}^{\mathrm{S}}\exp(H^{\mathrm{S}}/kT)$. According to the coupling model, interacting and correlations between the ISAs in the Cottrell cloud will slow down the Snoek relaxation rate for $t>t_{\mathrm{c}}$ and modifying the correlation function to the stretched exponential form given by eqn. (2). The effective correlation time for diffusion of the interaction ISAs is determined by eqn. (3). The size of n can be determined from the shape of the SK peak. Also the exact magnitude of t_{c} for this system is not known as yet. We expect it has an order of magnitude of about 10^{-13} s as found for alkali ions in sodium silicate glasses [11, 17]. For the purpose of this work, we need only know that it is temperature-independent as assured by evidence from neutron scattering experiments and molecular dynamics simulation in other materials. Written out explicitly, we have $\tau^*=[t_{\mathrm{c}}{}^{-n}\tau_\infty{}^{\mathrm{S}}\exp(H^{\mathrm{S}}/kT)]^{1/(1-n)}$. The effective diffusion coefficient D^* of the ISAs can be obtained in the form $D^*=6a^2/\tau^*$. On replacing the diffusion coefficient in Schoeck's expression for the SK relaxation time by D^*, we obtain the new expression:

$$\begin{aligned}\tau^{\mathrm{SK}}&\equiv\tau_\infty{}^*\exp(H^{\mathrm{SK}}/kT)\\&=\{6\alpha kTc_{\mathrm{d}}L^2a^2/(t_{\mathrm{c}}{}^{-n}\tau_\infty{}^{\mathrm{S}})^{1/(1-n)}\}\exp[H^{\mathrm{S}}/(1-n)kT]\end{aligned}\tag{8}$$

The non-exponential time dependence of the relaxation of the cloud of ISAs described by eqn. (2) is transferred to the SK relaxation making its correlation function, $C^{\mathrm{SK}}(t)$, also have the stretched exponential form of $C^{\mathrm{SK}}(t)=\exp[-(t/\tau^{\mathrm{SK}})^{1-n}]$. We can calculate the internal friction peak, Q^{-1}, via the Fourier transform of $C^{\mathrm{SK}}(t)$ which has a skew asymmetric shape and its peak width increases monotonically with n. As a function of temperature, Q^{-1} shows a maximum at the temperature for which the condition $\omega\tau^{\mathrm{SK}}\approx 1$ is satisfied. An Arrhenius plot of τ^{SK} s obtained by varying the measurement frequency will yield an activation enthalpy for the SK relaxation given by

$$H^{\mathrm{SK}}=H^{\mathrm{S}}/(1-n)\tag{9}$$

From our physical picture, we expect the coupling parameter n to be non-zero for the SK relaxation and thus the predicted H^{SK} can be substantially larger than H^{S}. It is clear that while H^{S} corresponds to a real energy barrier of a foreign interstitial atom jump from one site to another, H^{SK} is only an apparent activation enthalpy which reflects the many body correlations of the relaxation of the ISA cloud. After n has been determined and H^{S} known from the Snoek peak data, we have a quantitative prediction of H^{SK}.

The values of the coupling parameter determined by fitting the shape of the SK relaxation peaks of Fe, Ta and Nb (and their monocrystals) doped with several foreign interstitial impurities are shown in Table 1. In this table we have also entered the experimental values of the activation enthalpy H^{SK} and the calculated values of the product $(1-n)H^{\mathrm{SK}}$. According to this theory this product should be equal to or approximately equal to the activation enthalpy H^{S} of the Snoek peak for the same interstitial atom: $H^{\mathrm{S}}=(1-n)H^{\mathrm{SK}}$. This prediction can now be tested quantitatively. On comparing these two quantities in Table 1 we can conclude that they are nearly equal, thus verifying the theory. In most cases, we find the product $(1-n)H^{\mathrm{SK}}$ to be slightly smaller than H^{S}. This may be due to the fact that the values of n obtained from our fits of the SK peaks may be overestimated because a distribution of dislocation loop lengths, which is likely to exist, also

TABLE 1.

High purity specimen	Doped ISA	H^{SK} (eV)	n	H^{S} (eV)	$(1-n)H^{SK}$ (eV)	Ref.
α-Fe	N	1.40	0.38	0.796	0.868	32
α-Fe	C	1.84	0.60	0.87	0.736	33
Ta(A)	O	1.45	0.30	1.10	1.015	29
Ta(B)	O	1.68	0.48	1.10	0.874	29
Ta monocrystal	O	2.24	0.46	1.10	1.21	34
Nb	O	1.49	0.40	1.15	0.894	29
Nb monocrystal	O	1.68	0.50	1.15	0.840	29

contributes to the width of the SK peak. The results in Table 1 lend strong support to Wang's proposal that relaxation of the ISAs in the Cottrell cloud while dragged by a dislocation segment, is responsible for the activation energy and the breadth of the SK relaxation peak, although its large relaxation strength peak still comes from the area swept out by the segment.

A well known feature of SK relaxation is that the experimentally determined activation enthalpy sometimes exhibits large fluctuations in its value for samples containing different amounts of ISAs and subjected to different levels of plastic deformation. For example, the commonly studied stable SK-2 peak of Ta/O has its experimental H^{SK} varying from 1.4 to 2.1 eV. This variation of H^{SK} may be difficult to explain by other models because the size of H^{SK} is usually fixed, but can easily be done in our model. This is because under different experimental conditions the concentration of ISAs in the Cottrell cloud can be different. There is, then, the possibility of varying degree of correlation among the ISAs and hence different coupling parameter values. The different values of the coupling parameter will give rise to correspondingly different experimentally observed values of $H^{SK}=H^{S}/(1-n)$, even though H^{S} remains constant. The result shown in Table 1 gives such an example. Table 1 shows the SK relaxation in monocrystalline and in polycrystalline tantalum has very different activation enthalpy and peak temperature. This large difference in H^{SK} values is correlated with a corresponding difference in n value. The product $(1-n)H^{SK}$ remains relatively constant and nearly equal to H^{S} of the Snoek peak.

This discussion suggests that a critical test of theoretical models of the SK relaxation is to do systematic measurements on samples with lower and lower concentrations of ISA. According to our theory, the SK peak temperature (at a fixed frequency), H^{SK} as well as n will decrease. In the limit of low concentrations, H^{SK} should approach H^{S} as n goes to zero. By inspection of Table 1 one finds that the largest coupling parameter occurs for α-Fe doped with carbon. From this we expect a large reduction of H^{SK} will take place when the concentration of carbon in α-Fe is reduced. Internal friction measurements were made by one of us (LBM) on such samples after deformation to test this prediction. A stable SK peak that is slightly asymmetric in shape was observed. The data and analysis will be published in detail elsewhere. We summarize only the results here. In samples with low C concentration, the peak temperature of the SK relaxation moves down to 461.3 K (for $f=1$ Hz), and H^{SK} assumes the low value of 0.95 eV (to be compared with $H^{S}=0.835$). This is to be contrasted with the peak temperature of 550 K and $H^{SK}=1.84$ eV determined for the SK relaxation in samples having higher C concentrations. The width of the SK peak is also reduced in the low C concentration samples but is still broader than a Debye peak. This residual width of the SK peak may reflect the contribution to peak broadening from the distribution of the dislocation loops. While the characteristics of the SK relaxation in these low C concentration samples are consistent with our theory of the SK relaxation based on the coupling model, they contradict the kink-pair-formation theory of Seeger because the latter would predict, even in ultra-pure iron, a much larger SK relaxation activation enthalpy over the Snoek relaxation activation enthalpy than we have observed experimentally.

Due to space limitation we cannot discuss another application of the coupling model to internal friction in metals: *i.e.* that associated with precipitation in alloys. The interested reader can find the results in ref. 35.

5. Conclusion

While the coupling model has widely been known in its applications to polymers, viscous liquids and ionic conductors, it is relatively unfamiliar to the community of researchers in internal friction. The applicability of the coupling model to problems of interest to this community is not as clear as in the other fields. However, as discussed in this work, we can now see the possibility that some relaxation problems of immediate interest to the participants of ICIFUAS-10 are falling into the realm of applicability of the coupling model. More work in the future is still needed to bear out this expectation.

Acknowledgment

K.L. Ngai is supported in part by ONR contract N0001494WX23010.

References

1 K.L. Ngai and C.T. White, *Phys. Rev. B, 20* (1979) 2475.
2 K.L. Ngai, *Comments Solid State Phys., 9* (1979) 129.
3 K.L. Ngai and L.B. Magalas, *Mater. Sci. Forum, 119–121* (1993) 49.
4 For a most recent review see K.L. Ngai, in R. Richtert and A. Blumen (eds.), *Disorder Effects on Relaxation Processes,* Springer-Verlag, Heidelberg, 1994.
5 A.K. Rajagopal, K.L. Ngai and S. Teitler, *J. Phys. C, 17* (1984) 6611.
6 K.L. Ngai, S.L. Peng and K.-Y. Tsang, *Physica A, 191* (1992) 523.
K.-Y. Tsang and K.L. Ngai, to be published.
7 K.L. Ngai, *J. Chem. Phys., 98* (1993) 7588.
8 K.L. Ngai, R.W. Rendell, A.F. Yee and D.J. Plazek, *Macromolecules, 24* (1991) 61.
9 K.L. Ngai, R.W. Rendell and D.J. Plazek, *J. Chem. Phys., 94* (1991) 3018.
10 K.L. Ngai, R.W. Rendell, A.K. Rajagopal and S. Teitler, *Annals New York Acad. Sci., 484* (1986) 150.
11 K.L. Ngai and H. Jain, *Solid State Ionics, 18/19* (1986) 362.
12 The familiar factor $(1-n)$ that appears in an earlier version of this relation is absent because here we impose the condition of continuity of the correlation functions instead of the relaxation rates at $t=t_c$.
13 See papers presented in the *Workshop on Dynamics of Disordered Materials II, March 1993, Grenoble, France, Physica A,* in press.
14 J. Colmenero, A. Arbe and A. Alegría, *Phys. Rev. Lett., 71* (1993) 2603.
J. Colmenero, in *Workshop on Dynamics of Disordered Materials II, Physica A,* in press.
15 J. Colmenero, A. Arbe, A. Allegria and K.L. Ngai, *Polym. Mater. Sci. Eng., ACS Spring Meeting, San Diego, CA, March 1994,* American Chemical Society, USA.
16 R.J. Roe, *J. Chem. Phys.,* in press.
17 W. Smith, G.N. Greaves and K.L. Ngai, to be published.
18 G.F. Signorini, J.-L. Barrat and M.L. Klein, *J. Chem. Phys., 92* (1990) 1294.
19 K.L. Ngai and O. Kanert, *Solid State Ionics, 53–56* (1992) 936.
20 K.L. Ngai, R.W. Rendell and H. Jain, *Phys. Rev. B, 30* (1984) 2133.
21 K.L. Ngai, *Solid State Ionics, 5* (1981) 27.
22 K.L. Ngai and U. Strom, *Phys. Rev. B, 27* (1983) 6031; *B, 38* (1988) 10 350.
23 K.L. Ngai, *Solid State Ionics, 61* (1993) 345.
24 D.J. Almond and A.R. West, *Phys. Lett., 69A* (1978) 130.
25 D.J. Almond and A.R. West, *Solid State Ionics, 3/4* (1981) 73.
26 For a review see B.J. Molinas and F. Povolo, *Mater. Sci. Forum, 119–121* (1993) 39.
27 A. Rivière and J. Woirgard, *Mater. Sci. Forum, 119–121* (1993) 125.
28 L.H. Sun, Y.N. Wang and M. Gu, in T.S. Kê (ed.), *Proc. ICIFUAS-9, Beijing, People's Republic of China, 1990,* p. 61.
29 For a review see (a) A. Seeger, M. Weller, J. Diehl, Z.L. Pan, J.X. Zhang and T.S. Kê, *Z. Metallkd., 73* (1982) 1; (b) M. Weller, *J. Phys. C, 9* (1983) 63.
30 G. Schoeck, *Acta Metall., 11* (1963) 617; G. Schoeck and M. Mondino, *J. Phys. Soc. Japan, 18* Suppl. 1 (1963) 149.
31 G. Haneczok, M. Weller and J. Diehl, *Phys. Stat. Solidi (b), 172* (1992) 557; G. Haneczok, M. Weller and J. Diehl, *Mater. Sci. Forum, 119–121* (1993) 101.
32 D.P. Petarra and D.N. Beshers, *Acta Metall., 15* (1967) 791.
33 L.B. Magalas, J.E. Defresne and P. Moser, *J. Phys. C, 5* (1981) 127.
34 U. Rodrian and H. Schultz, *Z. Metallkd., 73* (1982) 21.
35 Y.N. Wang, M. Gu and L.H. Sun, *J. Phys. C: Condensed Matter, 1* (1989) 10 039.