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## A non-linear model of solid-state amorphization

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**Abstract.** A non-linear model of solid-state amorphization (SSA) is proposed which takes into account the interaction of the structural elements of the amorphous phase via a stress field. For the experimental parameters which correspond to semiconductors undergoing SSA by high-pressure phase retention, we obtain the solution in the form of a self-sustaining wave with a constant velocity  $v$ . This solution exists in the region  $v < v_c$ , where the critical velocity  $v_c$  is controlled by the thermal and concentration diffusivities  $D_T$  and  $D_N$ . A simple scaling relation is proposed for  $v_c$ :  $v_c(D_N, D_T) \sim D_T/(D_N)^{0.5}$ . It is found that the characteristic correlation length of the amorphous phase formed by SSA strongly depends on the interplay between the parameters  $v$ ,  $D_N$  and  $D_T$ .

### 1. Introduction

The field of non-equilibrium phase transitions in non-crystalline materials continues to attract much attention [1–4]. A metastable amorphous phase can, when heated, begin to crystallize and the latent heat thus released can accelerate the reaction, leading to the phenomenon of self-sustained explosive crystallization [3, 4]. This non-equilibrium problem has been described adequately only on a phenomenological level and a number of unsolved problems still remain. First of all, there is a serious discrepancy between the observed growth rates for such non-equilibrium processes and those obtained by extrapolating from the equilibrium growth rates to the relevant temperatures of the non-equilibrium process. The enhancement of the concentration diffusivity,  $D_N$ , can reach  $10^4$ – $10^5$  [5–7], suggesting that some extra physical mechanism may drive the non-equilibrium phase transition. Second, using the standard phonon thermal conductivity, it is easy to show that the heat disappears from the zone of reaction too rapidly to sustain a non-equilibrium regime [7, 8]. Third, in order to obtain agreement with the experimental data one must multiply the thermal diffusivity  $D_T$  by a factor of  $10^{-5}$ – $10^{-6}$  [7].

However, although explosive crystallization is a well known example of a non-equilibrium phase transition, it is not the only one. Another example is solid-state amorphization (SSA) and this is the subject of this paper. Generally, SSA occurs when a highly excited metastable solid is allowed to relax. Clear evidence for SSA is found in high-pressure phase transitions [9, 10]. In contrast with the case of explosive crystallization rather little theoretical analysis of SSA has been done thus far. The aim of the present work is therefore to construct a model of SSA which is based on a physical approach previously suggested [6, 7] for modelling explosive crystallization, and therefore to describe SSA

as a diffusion-controlled process, with the aim of explaining the observed thermal and concentration diffusivities. (An early version of the model was reported in [10].)

The paper is organized as follows: in section 2 we discuss the background physics to the model which is introduced in section 3. The results are presented in section 4.

## 2. Model background

We first review the current experimental situation concerning SSA. Methods which have been used to prepare amorphous alloys by SSA include solid-state reactions [11, 12], mechanical alloying by continuous milling of a two-component mixture [13, 14], ion implantation or radiation-induced disordering [15], hydrogen-induced vitrification [16] and high-pressure techniques [17]. A common feature of these SSA processes appears to be that relaxation of stress can aid the formation of the amorphous phase. In the case of ion implantation, Volkert [15] has shown that ion bombardment in silicon at first leads to a steadily increasing in-plane stress, and that at some critical dose of radiation the stress reaches a maximum. This point coincides with the formation of the amorphous phase and further growth of the amorphous layer is accompanied by a subsequent decrease in the amplitude of the stress field [15]. Similar stress relaxation has been proposed as a factor in milling, grinding and solid-state reactions [9, 10].

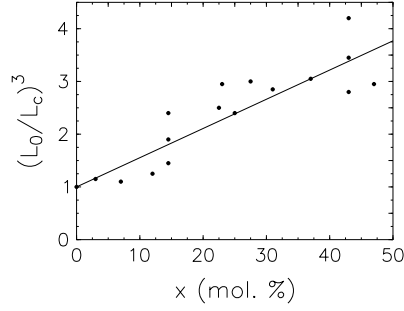
In high-pressure techniques, a high-pressure phase (HPP) of a solid is initially formed and then cooled to liquid nitrogen temperature and the pressure is released. At ambient pressure a metastable HPP is obtained. If this is heated, when some temperature  $T_a$  (the temperature of amorphization) is reached the HPP becomes unstable and the nearest-neighbour order changes to that appropriate for the low-pressure phase, but long-range order is simultaneously lost and an amorphous solid with a correlation length  $L_c \sim 10\text{--}20 \text{ \AA}$  is formed [9, 17–19].

Our model rests on the assumption [10] that an amorphous network can be represented as a superstructure of certain structural elements which are absent in the crystalline state. These elements, which we will call ‘structure-forming defects’ (SFD), act as soft regions in the rigid covalent network which allow the bond angles and bond distances adjacent to the SFD to vary, and are thus responsible for the loss of long-range order in the amorphous state [20, 21]. The detailed microscopic structure of the SFD may be different for different amorphous solids, depending on their chemical composition. We assume only that they are characterized by energies of creation and annihilation and that they interact via a stress field, as discussed below. We neglect all orientation effects and average over nearest neighbours.

If the concentration of SFD is  $N$ , the correlation length  $L_c$  is

$$L_c \approx N^{-1/3}. \quad (1)$$

(The case of the perfect crystal corresponds to  $N = 0$ .) Additional arguments in favour of this model may be deduced from experimental data on the effect of doping of amorphous solids obtained by solid-state amorphization [18, 19]. For the case of amorphous gallium antimonide doped with copper, a-GaSb:Cu [18], and germanium a-GaSb:Ge [19], it was found that  $L_c$  decreases with the impurity concentration  $x$  up to several molecular per cent ( $L_c$  was calculated from x-ray scattering data). It may be supposed that the doping produces additional defect states leading to additional disorder, so in the doped case  $L_c^{-3} \sim N + \gamma x$ , where the coefficient  $\gamma$  takes into account the number of impurity atoms involved in the generation of these SFD. Experimental data from [18, 19] are replotted in figure 1 in terms of coordinates  $L_c^{-3} = f(x)$ , and it can be seen that they correlate well with the suggested linear relation.



**Figure 1.** The concentration dependence of  $L_c^{-3}$  with doping for a-GaSb samples obtained by solid-state amorphization under high pressure.  $L_0$  corresponds to  $L_c(x=0) \sim 20 \text{ \AA}$  [13, 14].

We assume that the SFD are characterized by an energy of creation  $U$ , an energy of annihilation  $\Delta$ , and a characteristic radius  $r_0$ . The key idea is that the SFD interact via a stress field, and that the presence of stress reduces the activation energy for SFD creation. (This assumption is typically applied to standard defects in solids [22].) If  $\varphi(r - r_i)$  is the potential energy associated with the stress field produced by the  $i$ th SFD, at position  $r_i$  then in the mean-field approximation, the activation energy can be written as

$$U = \langle U(r) \rangle = U_0 - \left\langle \sum_i \varphi(r - r_i) \right\rangle \approx U_0 - n_0 \varphi(\bar{r}) \quad (2)$$

where  $U_0$  is the self-energy of the SFD,  $n_0$  is the number of nearest neighbours, and  $\bar{r} = N^{-1/3}$  is the average distance between SFD (which we will henceforth identify with the correlation length  $L_c$ ). We now assume that the excited state has an energy given by the difference between the thermodynamic potential of the metastable HPP and that of the low-pressure crystalline phase. If no SFD are formed, the excited state will decay into a crystalline state and an excess of energy will be dissipated in phonon modes. If SFD are formed, they can serve as traps for energy from the excited solid, so for their internal energy we can write [10]

$$E = \langle UN \rangle \approx U(N)N \quad (3)$$

where  $U(N)$  is given by equation (2) when the substitution  $\bar{r} = N^{-1/3}$  is made. When a stationary concentration of SFD is reached, the value  $E(N)$  will describe the excess energy of the amorphous solid with respect to the crystalline solid, i.e. give an estimate of the heat of crystallization  $Q_{cr}$ . In the ideal case one might imagine all of the energy of the excited state ending up being stored in SFD, but since the formation of SFD at  $T_a$  is a dynamical process, some part may cause phonon excitation. Thus some heat, which is less than  $Q_{cr}$ , may be released near  $T_a$ . Note that the current model makes no assumptions about the diffusivities  $D_N$  and  $D_T$ , so we will keep them as free parameters and analyse the effect of the diffusion mechanism on the solid-state amorphization process.

### 3. Formalism

Following the discussion in section 2 we now write down a coupled pair of partial differential equations which will describe the model via the concentration and temperature fields,  $N(r, t)$  and  $T(r, t)$ , and which include terms corresponding to diffusion, and also creation and

annihilation of SFD. Thus we will need to solve

$$\frac{\partial N}{\partial t} = \omega_0 e^{-U/k_B T} - N \nu_1 e^{-\Delta/k_B T} + D_N \frac{\partial^2 N}{\partial r^2} \quad (4)$$

$$\frac{\partial T}{\partial t} = D_T \frac{\partial^2 T}{\partial r^2} - \frac{1}{c\rho} \frac{\partial E}{\partial t} \quad (5)$$

where the parameter  $\omega_0 \approx a^{-3}/\tau_0$ ,  $\nu_1$  is the characteristic phonon frequency ( $\tau_0 \approx 1/\nu_1 \approx 10^{-13}$  s),  $a$  is the lattice parameter,  $c$  is the specific heat capacity and  $\rho$  is the density. The first term on the R.H.S. of equation (4) describes the creation of SFD. The second term on the R.H.S. of equation (4) describes the annihilation of SFD, and we will take for simplicity  $\Delta \approx E_c \approx \text{constant}$ , where  $E_c$  is the activation energy of crystallization.

In order to evaluate equations (4) and (5), it is necessary to propose a function  $\varphi(r)$  in equation (2). Existing theories give an asymptotic form  $\varphi(r) \sim 1/r^\alpha$  for  $r \rightarrow \infty$ , where  $\alpha = 3$  or  $6$  [23]. There is no information about the asymptote in the other limit  $r \rightarrow 0$ . The effect of different possible asymptotic forms at  $r \rightarrow 0$  was considered in [10] and it was found that SSA may occur for almost all functional forms of  $\varphi(r)$  at small distances. In the present work we will consider a model form for  $\varphi(r)$ :

$$\begin{aligned} \varphi(r) &= \varphi_0 \chi(r/r_0) \\ \chi(x) &= 2x^3/(1+x^6) \end{aligned} \quad (6)$$

where  $\varphi_0$  is the amplitude of interaction between the SFD and  $r_0$  is their radius. Formulae (6) give  $\varphi(r) \approx r^{-3}$  at  $r \rightarrow \infty$  and  $\varphi(r) \approx r^3$  for  $r \rightarrow 0$ , so the stresses relax when  $N$  increases, i.e. when a ‘more amorphous’ phase is reached. This model behaviour is in agreement with the experimental results of Volkert [15], who showed that the relaxation of a stress field in the sample corresponds to the formation of an amorphous phase.

Equations (4) and (5) have either one ( $N_1$ ) or three stationary solutions ( $N_1 < N_2 < N_3$ ) and in both cases  $N_1$  is a stable solution, and in the latter case  $N_2$  is unstable while  $N_3$  is stable. The maximum stable solution is associated with the amorphous phase with correlation length  $L_c = N_1^{-1/3}$  or  $L_c = N_3^{-1/3}$ . In the case of one solution, the excited state always transforms into the amorphous state; for three solutions, in order to reach the amorphous state the initial concentration of SFD  $N(t=0)$  should exceed a critical value  $N_2$ , otherwise a crystalline state will be formed. We note that in non-linear dynamical processes, it is also possible that a transition wave between the stable points  $N_1$  and  $N_3$  may occur, so the amorphous state can be reached by a dynamical mechanism [24]. One of the aims of the present work is to find such a solution (in contrast to [10] where only static cases were analysed in detail).

Equations (4) and (5), together with (2) and (3), are strongly non-linear and it is difficult to solve them by analytical or numerical methods. We make the simplifying assumptions that the diffusivities  $D_N$  and  $D_T$  are constant. We expect that the qualitative results of the computation will not depend much on this assumption. Further calculations (see the next section) will show that the concentration profile is much sharper than the thermal one, so there is probably no sense in taking into account the exact temperature dependence of  $D_N$ . As for  $D_T$ , any temperature dependence will be no more than a power series in  $T$ , which is therefore weaker than the exponential terms in equations (4) and (5), so setting  $D_T$  constant is also reasonable.

In this paper, we will restrict ourselves to the case of a one-dimensional solution for a self-sustaining phase transition wave. For the new variable  $\xi = x - vt$ , where  $v$  is the

velocity of the phase transformation wave, we can write

$$-v \frac{dy}{d\xi} = D_N \frac{d^2y}{d\xi^2} - \frac{y}{\tau_0} e^{-\Delta/\theta} + \frac{b}{\tau_0} e^{-U(y)/\theta} \quad (7)$$

$$-v \frac{d\theta}{d\xi} = D_T \frac{d^2\theta}{d\xi^2} + \frac{vk_B}{c\rho a^3 b} \frac{d}{d\xi}(yU(y)) \quad (8)$$

$$U(y) = \Delta(1 + \delta - s\chi(y^{-1/3})) \quad (9)$$

where  $y = Nr_0^3$ ,  $b = r_0^3/a^3$ ,  $\theta = k_B T$ ,  $\delta = (U_0 - \Delta)/\Delta$ ,  $s = \varphi_0 n_o/\Delta$ , and  $k_B$  is the Boltzmann constant. In accordance with reference [10], the following experimental values were chosen:  $a = 3 \text{ \AA}$ ,  $b = 15.7$ ,  $T_0 = 365 \text{ K}$ ,  $Q_{cr}/c\rho = 94 \text{ K}$ ,  $\Delta = 0.5 \text{ eV}$  and  $k_B/c\rho a^3 b = 0.44$ , representing the case of a typical semiconductor which undergoes solid-state amorphization via high-pressure phase retention. The values of  $\delta$  and  $s$  are not known and are therefore kept as free parameters.

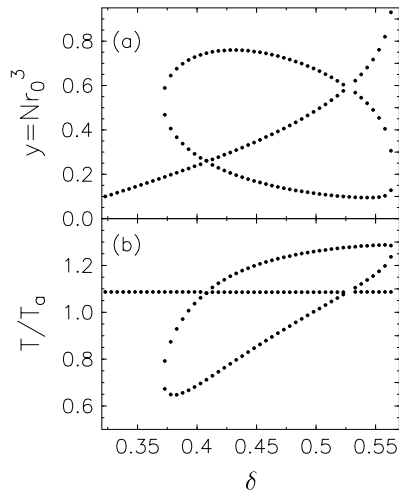
Equations (7) and (8) were solved numerically by means of an iterative method. Initially, the exact solution of equation (7) was replaced by some reasonable profile  $N(\xi)$ , and  $T(\xi)$  was calculated exactly using equation (8). The  $T(\xi)$  value thus obtained was put into equation (7) to calculate  $N(\xi)$ , etc. The iteration process was continued until the difference between subsequent profiles  $N(\xi)$  became less than  $10^{-4}$ – $10^{-5}$ , which typically happened after 40–50 iterations. Several methods of discretizing  $\xi$  were also examined and all gave similar results. To check the accuracy of the calculations, the step of the grid was decreased until the required accuracy ( $10^{-4}$ – $10^{-5}$ ) for  $N(\xi)$  and  $\theta(\xi)$  was achieved.

The boundary conditions for the initial distribution function as  $\xi \rightarrow -\infty$  should correspond to the case of the amorphous state, i.e.  $N = L_c^{-3} = \text{constant}$  and  $T = T_0 \approx \text{constant}$ . In the opposite case we have an excited solid, and  $N$  is small, whereas the temperature is higher:  $T = T_0 + Q_{cr}/c\rho$ . Here  $T_0$  is the equilibrium temperature of the sample after the phase transition and  $Q_{cr}$  is the crystallization heat captured by structure-forming defects:  $Q_{cr} \approx E \approx U(N)N$  [10]. We used zero-flux boundary conditions for  $N$  and  $T$  at  $\xi = \pm\infty$ .

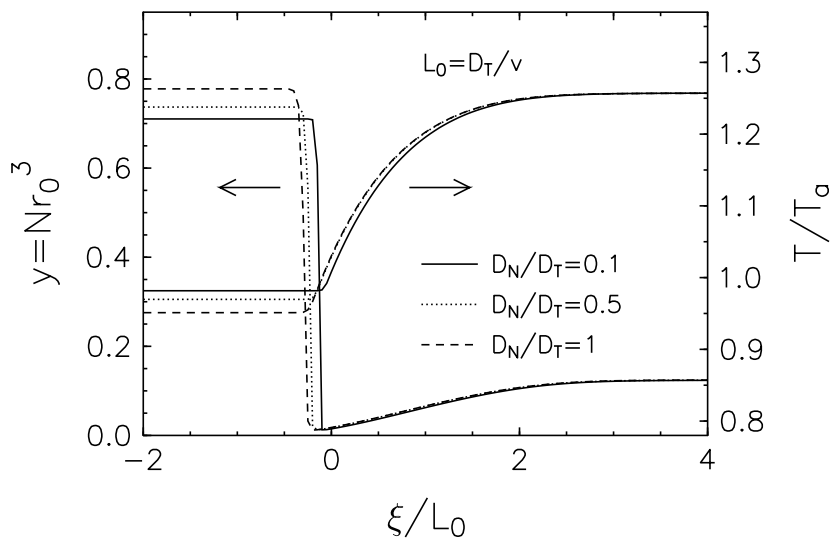
## 4. Results and discussion

### 4.1. Stable points

By analysing equations (7) and (8) [24] it is possible to show that a phase transition wave with constant velocity  $v$  corresponds to a process in which the system moves between the stable solutions of equations (7) and (8) or (4) and (5), which are given by the solutions of equations (7) and (8) with all  $d(\cdot\cdot)/d\xi = 0$ . In agreement with the results of [10] we find that for the experimental parameters corresponding to semiconductors like GaSb, CdSb and ZnSb which undergo solid-state amorphization by high-pressure phase retention [10, 11], equations (7) and (8) may have one or three stationary solutions in the  $(y, T)$  plane depending on the interplay between the parameters  $s$  and  $\delta$  described above. The particular case  $s = 0.3$  is illustrated by figure 2 for different choices of the parameter  $\delta$ . If there are three solutions  $y_1 < y_2 < y_3$  then  $(y_1, T_1)$  and  $(y_3, T_3)$  will represent stable points, and  $(y_2, T_2)$  will be unstable, and the case of a single solution is always stable [10]. Note that according to the basic assumptions of the model the biggest root  $y_{\max}$  should be associated with the amorphous state having a coherence length  $L_c = r_0 y_{\max}^{-1/3}$ ; see equation (1).



**Figure 2.** Stationary points for equations (7) and (8) for the case of  $s = 0.3$  as a function of  $\delta$ . If there are three solutions  $y_1 < y_2 < y_3$  for a particular choice of  $\delta$  then  $(y_1, T_1)$  and  $(y_3, T_3)$  will represent stable points, and  $(y_2, T_2)$  will be unstable. If there is a single solution it will be stable. The largest root is associated with the amorphous state with a coherence length  $L_c = r_0 y_{\max}^{-1/3}$ .

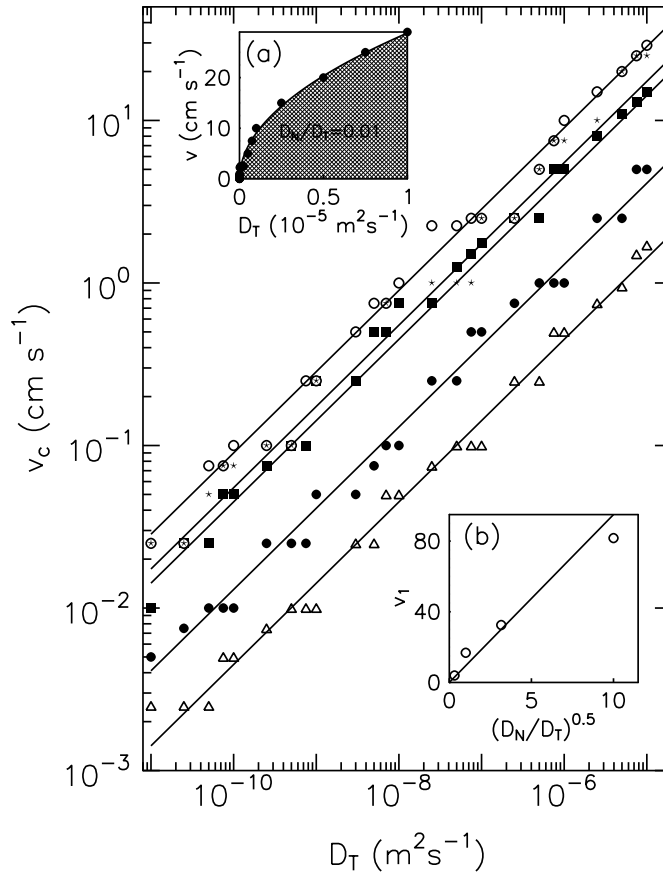


**Figure 3.** Concentration ( $y = Nr_0^3$ ) and temperature ( $T/T_a$ ) profiles for a self-sustaining wave with constant  $v$  for different choices of the ratio  $D_N/D_T$  for the case  $s = 0.3$  and  $\delta = 0.5$ .

#### 4.2. The phase transition wave

Here we will consider the case of a stable wave front with constant  $v$  for which the iteration procedure converges. The case of dissipative structures observed in some cases will be described elsewhere. In order to illustrate the basic stable behaviour we consider the case  $s = 0.3$  and  $\delta = 0.5$  (compare figure 2) for which there are three roots, two of them stable. The system may then exhibit a dynamical transition  $((y_1, T_1) \rightarrow (y_3, T_3))$ , skipping

the unstable root ( $y_2, T_2$ ). Typical results for the concentration and temperature profiles are given in figure 3. As can be expected, the temperature front is much wider than the concentration front, and scales with the diffusion broadening length  $L_0 = D_T/v$ , which gives the characteristic size of the thermal gradient (figure 3). The change of the  $D_N/D_T$  ratio alters the stationary asymptote for  $\xi \rightarrow \infty$ . This result supports the proposition that SSA is essentially a dynamical process, and the amorphous state formed will depend on the speed of the process and the diffusion coefficients.



**Figure 4.** Numerically calculated wave velocities for the case of  $D_N/D_T = 0.001$  (open circles), 0.01 (stars), 0.1 (filled squares), 1 (filled circles), and 10 (triangles). The slopes of the lines are found to be  $0.5 \pm 0.03$ . Insets: (a) the phase transformation wave (the case shown has  $D_N/D_T = 0.01$ ) can only exist if its velocity  $v$  is less than some critical value  $v_c = v_1 D_T^{0.5}$ ; the allowed region is shown shaded; and (b) the parameter  $v_1$  fits well to the relation  $v_1 \propto (D_N/D_T)^{-0.5}$ .

### 4.3. The phase diagram

Our calculations have shown that the phase transition wave, as described above (see figure 3), can exist only if its velocity is smaller than some critical value  $v_c(D_N, D_T)$  (see inset (a) in figure 4; the allowed region is shaded). For a fixed ratio  $D_N/D_T$  over a very large interval of thermal diffusivity,  $10^{-11} \text{ m}^2 \text{ s}^{-1} < D_T < 10^{-5} \text{ m}^2 \text{ s}^{-1}$  the following scaling relation



for a phase boundary holds:

$$v_c = v_1 D_T^{0.5} \quad (10)$$

(see the main diagram of figure 4 where the slope of the lines corresponds to the index  $0.5 \pm 0.03$ ). The change of the ratio  $D_N/D_T$  was found to alter the coefficient  $v_1$  in formula (10):  $v_1$  decreases when  $D_N/D_T$  decreases.

This type of  $v_c(D_N, D_T)$  dependence may have a natural explanation if one assumes that for the self-sustaining wave of the type considered above, the thermal gradient length  $L_0$  should be bigger than the length scale associated with the concentration diffusivity  $L_N$ , as one might expect for the temperature-driven process. Thus, we can write the condition for the existence of the wave which defines the critical velocity:

$$L_N = (D_N \tau_N)^{0.5} < L_T = D_T/v_c. \quad (11)$$

Here  $\tau_N$  is the relaxation time of the SFD. From equation (11) we immediately get equation (10) with  $v_1 = (D_N \tau_N/D_T)^{-0.5}$ . Inset (b) in figure 4 indicates that this estimate is consistent with the results of the computation.

## 5. Conclusions

A non-linear model of solid-state amorphization (SSA) taking into account the interaction of the structural elements of the amorphous phase via a stress field has been described. The proposed model represents SSA as a thermal diffusion-controlled process in which the concentration of SFD ‘follows’ the temperature. However, it is impossible to neglect the concentration diffusivity as, in accordance with the data displayed in figure 3, the characteristics of the amorphous state formed should depend on the  $D_N$ -parameter. Looking at the scale in figure 4 we see that SSA may be very fast, and hence appear in an experiment as a phase jump. The velocity of SSA remains high even in the localized regime ( $D_T \approx 10^{-11} \text{ m}^2 \text{ s}^{-1}$ ), being about  $0.01 \text{ cm s}^{-1}$ . Further experiments should check this theoretical prediction.

## Acknowledgments

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