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Publisher: Taylor & Francis

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Philosophical Magazine Part B

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/tphb20>

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Version of record first published: 01 Dec 2006.

To cite this article: Y. Wolfus, Y. Yeshurun, I. Felner & J. Wolny (1987): Crystallization kinetics in amorphous ferromagnets effect of temperature and magnetic field, Philosophical Magazine Part B, 56:6, 963-968

To link to this article: <http://dx.doi.org/10.1080/13642818708215332>

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Crystallization kinetics in amorphous ferromagnets

Effect of temperature and magnetic field

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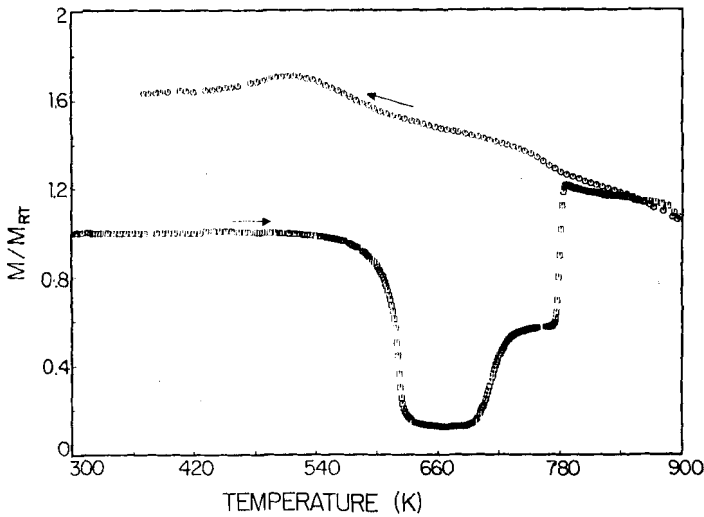
ABSTRACT

We study the time-evolution of the magnetization in the metallic glass $\text{Fe}_{83}\text{B}_{12}\text{Si}_5$ on different isotherms ($634\text{ K} \leq T \leq 666\text{ K}$) and at different magnetic fields ($30\text{ Oe} \leq H \leq 10\text{ kOe}$). The magnetization increases with time owing to the growth of a magnetic crystalline phase. The growth rate increases with increase of either temperature or magnetic field.

In the past few years there has been renewed interest in the process of nucleation and growth at first-order phase transformations. In particular, recent concepts of scaling have been applied to the description of the process in order to deduce the role of external parameters (temperature, pressure, etc.) on the growth rate (Wolny, Smardz, Zajac, Soltys and Dubois 1984), Wolny, Soltys, Smards, Dubois and Calka 1984, Yamada, Hamaya, Axe and Shapiro 1984, Hamaya, Yamada, Axe and Shapiro 1986, Roig, Munoz, Salamon and Rao 1987). Recently we demonstrated the success of scaling concepts in describing the crystallization kinetics of magnetic metallic glasses (Wolfus, Yeshurun, Felner and Wolny 1987). In the present work we extend our study of the crystallization kinetics to include the effect of *two* experimental variables, temperature and magnetic field, on the growth rate of the crystalline phase in metallic glasses. We demonstrate that the crystallization rate is accelerated by increasing either temperature or magnetic field.

Metallic glasses are non-equilibrium metallic solids, formed by very rapid solidification from the molten to the amorphous state (Moorjani and Coey 1984). Since the amorphous state is essentially metastable, it can transform into the more stable crystalline state. However, the most promising properties of metallic glasses, for example, the excellent magnetic behaviour, the high hardness and strength, ductility and high corrosion resistance, have been found to deteriorate drastically upon crystallization. Understanding the crystallization process is therefore not only of fundamental interest, but is very important in most applications. The amorphous to crystalline transition in metallic glasses has been studied by observing one or more of the physical properties of the material such as electrical resistivity, saturation magnetization and heat capacity, which change during the crystallization process.

Fig. 1

Temperature dependence of the magnetization for $\text{Fe}_{83}\text{B}_{12}\text{Si}_5$ at 250 Oe.

In the present work we focus attention on the magnetic metallic glass $\text{Fe}_{83}\text{B}_{12}\text{Si}_5$, for which we study the crystallization process by measuring the time-evolution of the magnetization on different isotherms and at different magnetic fields.

Metallic glasses of $\text{Fe}_{83}\text{B}_{12}\text{Si}_5$, in the form of long ribbons, were produced by melt-spinning (Moorjani and Coey 1984). The measurement process is the following. One chip of the ribbon, typically $12 \times 1 \times 0.075 \text{ mm}^3$, was mounted in a vibrating sample magnetometer (VSM) at room temperature (RT), with the longest edge along the magnetic field \mathbf{H} . At the desired field strength, the magnetization M was measured against temperature T up to a certain, previously defined, temperature. The temperature was stabilized to better than 0.5 K for several hours. While the measurements were taken frequent isothermal measurements of the magnetization are taken. A new specimen was used for each run, and all magnetization data were normalized to their room-temperature values.

Figure 1 shows typical $M(T)$ data measured at $H = 250$ Oe. The key points to be noted in understanding the complex structure exhibited in this figure are the following: (1) Crystallization in $\text{Fe}_{83}\text{B}_{12}\text{Si}_5$ takes place in two stages (Ramanan and Fish 1982). At first α -(Fe, Si) phase is formed. Then, at higher temperatures, the crystallization is completed by formation of the unstable Fe_3B phase. (2) The ferromagnetic-paramagnetic (FN-PM) transition temperature T_c^a for glassy materials is usually lower than the FM-PM transition temperature T_c^c for the equivalent crystalline material. Similarly, the average magnetic moment for the ordered state is larger than that of the disordered phase. Bearing these points in mind, the interpretation of fig. 1 is straightforward. Upon increasing the temperature from RT, the behaviour of the magnetization is characteristic of a glass ferromagnet with $T_c^a \approx 610$ K. Any increase in temperature above T_c^a should yield a paramagnetic Curie-Weiss drop in the magnetization of the paramagnetic untransformed volume. On the other hand, the transformed α -(Fe, Si) crystalline volume contributes an increase in the magnetization. The competition between these two processes causes the

apparent deviations from a Curie–Weiss behaviour and the shallow minimum around 650 K. As temperature increases, the crystallization rate is accelerated and the α -(Fe, Si) growth dominates the features of the $M(T)$ curve. This is most apparent at $T_1 \simeq 700$ K, where M increases sharply and then saturates, signalling saturation in the growth of this phase. Similarly, the abrupt rise at $T_2 \simeq 785$ K signals the growth of the Fe_3B phase. On further warming, the crystallization process ends, and $M(T)$ reaches a maximum, then drops towards the crystalline FM–PM transition. The crystallization process is of course irreversible; hence the upper curve in fig. 1, which was taken during cooling.

In isothermal measurements of the magnetization in the amorphous phase, M increases with time owing to growth of the ordered phase. We assume that the changes in the bulk magnetization are linear with the volume of the transformed sample, and define the fraction of volume transformed at time t by

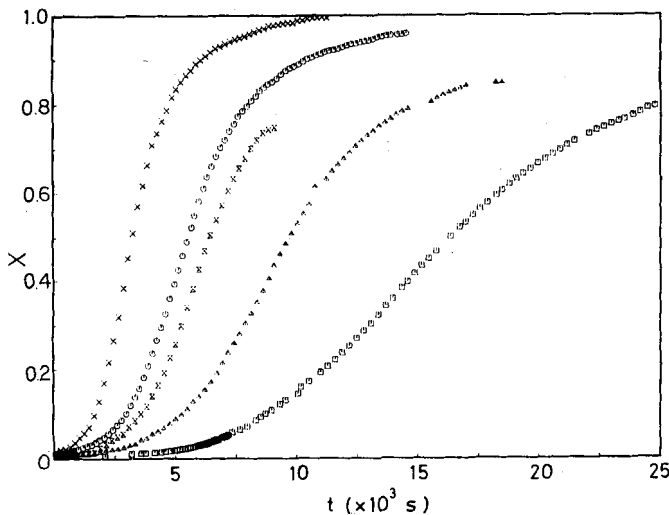
$$X(t) = \frac{M(t) - M(0)}{M(\infty) - M(0)}, \quad (1)$$

where $M(0)$ is the magnetization of the ‘pure’ glassy material (i.e. before any transformation has occurred) and $M(\infty)$ is the magnetization of the bulk after the crystallization process of α -(Fe, Si) has reached saturation.

The time-evolution of the magnetization measured in ~ 25 Oe and normalized to its saturation value is exhibited in fig. 2 on five isotherms. The crystallization process is accelerated with increasing temperature, but a qualitative similarity between isotherms is apparent. Thus, for each isotherm we can find the time $\tau(T)$ at which dM/dt is maximal, and re-define the time-scale of each isotherm to be t/τ . Following this procedure, all isotherms collapse to a single curve, as illustrated in fig. 3. An Arrhenius law

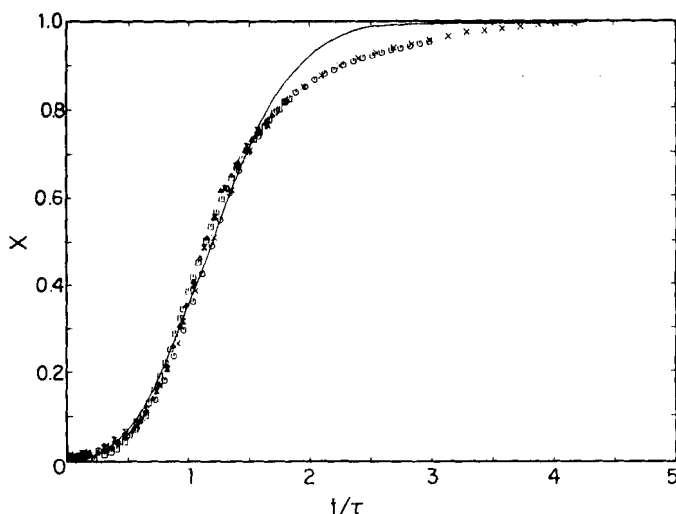
$$\tau^{-1} = \tau_0^{-1} \exp(-E_a/kT), \quad (2)$$

Fig. 2



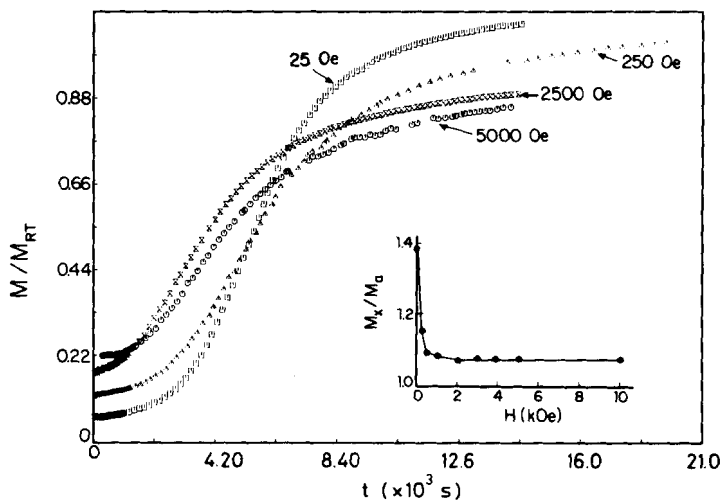
Time-evolution of isotherm-transformed volume: \times 666 K; \circ 660 K; γ 655 K; \triangle 647 K; \square 634 K.

Fig. 3



Isothermal curves of transformed volumes plotted against the scaled time t/τ . All curves fall onto a single universal line. The solid line is the curve calculated for the KJMA model (eqn. 4) with exponent $n = 2.5$. Symbols have the same meaning as in fig. 2. $H = 30$ Oe.

Fig. 4

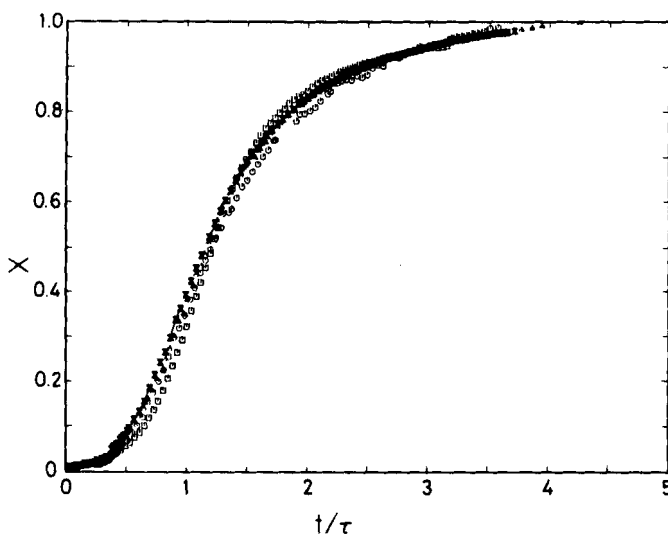


Time-evolution of the magnetization at 660 K for fields 25, 250, 2500 and 5000 Oe. The inset shows the room-temperature values for the ratio between the magnetization of the amorphous and crystalline phases as a function of the external field.

with $E_a = 1.85$ eV and $\tau_0^{-1} = 2.65 \times 10^{10} \text{ s}^{-1}$ fits the scaling parameter data satisfactorily. We stress, however, that the present data are limited to a relatively narrow temperature range and we are therefore not able to eliminate other possibilities (Wolfus *et al.* 1987).

Figure 4 shows the time-evolution of the magnetization at fixed temperature

Fig. 5



Data of fig. 4 in terms of the scaled time t/τ . Symbols have the same meaning as in fig. 4.

($T = 660$ K) for four different magnetic fields normalized to its room-temperature value. The field apparently accelerates the crystallization process. The difference $M(\infty) - M(0)$ is field-dependent. This can be understood with the aid of the inset to fig. 4. The ratio between the magnetization of the amorphous and crystalline phases M_x/M_a decreases as the field increases, and as a result, the normalized saturation value M_∞/M_{RT} decreases with the field. The increase of the initial value $M(0)$ with field is due to the paramagnetic behaviour of the amorphous phase at 660 K.

After performing the *same* scaling procedure as in the temperature-dependence case and normalizing according to eqn. (1), the four curves collapse to a 'universal' growth curve in fig. 5. Since the run with $T = 660$ K and $H = 25$ Oe appears both in the temperature scaling and the field scaling, it is clear that the universal curve is unique and is not affected by the field, at least at this temperature. We stress, however, that the field data are still preliminary and we cannot clearly determine the field dependence of the scaling parameter $\tau(H)$. We believe, nevertheless, that the magnetic field accelerates the crystallization process by increasing the free-energy separation between the amorphous and crystalline phases by term of the form $\Delta\mu \cdot \mathbf{H}$ where $\Delta\mu$ is the difference of the magnetic moment between the phases.

This work presents a preliminary attempt to use scaling concepts for two experimental variables, field and temperature, in crystallization kinetics. Similar attempts have been applied by Hamaya *et al.* (1986) to their study of the structural transformation in RbI crystals. They used pressure and temperature as variables and were able to demonstrate scaling features for all isobars at a particular temperature. However, a change in temperature induces a change in the scaling function. Our preliminary data suggest that growth curves induced by either temperature or field have a universal form.

Phenomenological models for nucleation and growth were suggested decades ago (Kolmogorov 1937, Johnson and Mehl 1939, Avrami 1939, 1940, 1941) (KJMA). In these models, infinitesimal grains of crystalline phase which are randomly produced at

a constant rate $\Gamma(T)$ are the nuclei for a transformed volume. The transformed volume is given by

$$x(t) = 1 - \exp \left[-\Gamma \int_0^t V(t') dt' \right], \quad (3)$$

where $V(t')$ is the volume occupied by a grain at time t' after nucleation. Since $V(t') \propto (vt')^d$, where d is the growth dimensionality and v is the growth velocity, it is clear that

$$x(t) = 1 - \exp [-(kt)^n] \quad (4)$$

with $n = d + 1$. Thus, in the simplest approach, one would expect n to be an integer. This was indeed found to be the case in several recent experiments (Yamada *et al.* 1984, Roig *et al.* 1987), where n is 4 and 3, respectively. The solid line in fig. 3 is our least-square fit of the data to eqn. (4), which yields a non-integer value, $n = 2.5 \pm 0.2$. One interpretation of this result is to assume anisotropic growth velocity, i.e. d is effectively between 1 and 2. The second interpretation is to assume a diffusion-controlled process. The new α -(Fe, Si) phase grows at the expense of the old amorphous phase by the slow migration of the interphase boundary, and growth results when B atoms diffuse across this boundary. The migration, being dependent upon diffusion rates, implies that the radii of such spherical grains are proportional to the square root of the annealing time t . For three-dimensional processes we thus get $V \propto t^{3/2}$ and $n = \frac{5}{2}$ in accordance with our results. In this description of a diffusion-controlled process, the concentration of B atoms near the interphase is expected to increase with time and a concentration gradient of boron atoms is produced ahead of the crystallization front. Since long-range diffusion is involved here, the growth rate is expected to decrease with time. This description is consistent with the deviation (fig. 3) from the solid line at long times. It is also consistent with the activation energy of 1.85 eV, which is typical of atomic diffusion processes. However, in order to determine the correct description, one must not rely on bulk measurements but rather combine these with a study of the micromechanism of the crystallization process using microscopic methods.

ACKNOWLEDGMENTS

This work is supported in part by the National Council for Research and Development, the Israel Ministry of Science, and in part by Grant P9, Poland.

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