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LETTER TO THE EDITOR

Crystallisation and structural relaxation of quasicrystalline Al–Mn: a positron annihilation study

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Abstract. We report here for the first time the positron annihilation study of quasicrystalline $Al_{86}Mn_{14}$ alloy. Our study confirms calorimetric results reported earlier, which indicated that, before the crystallisation, the quasicrystalline phase in this alloy goes through a structural relaxation process. The underlying cause of the substantial drop in the line-shape parameter S during the crystallisation process is also considered.

Ever since the discovery [1] of the quasicrystalline phase in Al–Mn alloy by Shechtman and co-workers, there has been a continuous effort to understand the structure and growth mode of this new phase of matter [2]. Although the structure of the quasicrystalline icosahedral phase can be analysed using six-dimensional crystallography [3], three-dimensional Penrose tiling gives us, at present, more insight into the growth process of this phase. Recently it has been shown [4] that, if the growth process is restricted [5] within eight-vertex configurations, one can generate infinitely large perfect Penrose tiling in a simulation experiment by employing a set of local rules. In the actual growth of quasicrystals one can never achieve this perfect growth; as a result there will be trapped defects in as-grown samples, especially because this phase is generally formed by rapid solidification and other non-equilibrium techniques.

To understand the structure, growth and stability of this new phase, it is very important to know the nature of these trapped defects. Positron annihilation is known [6] to be a very sensitive technique with which to study the nature and concentration of defects in metallic alloys. Although calorimetric and other techniques have already been employed in the study of structural relaxation and crystallisation [7, 8] of the quasicrystalline phase, in this Letter we show for the first time—to the best of our knowledge—that the positron annihilation technique can be used to study these processes.

$Al_{86}Mn_{14}$ alloy was prepared by induction melting of high purity Al and Mn under argon atmosphere. Quasicrystalline ribbon samples of about 3 mm width and 30 μm thickness for the present study were obtained by the melt-spinning method. The samples were characterised by TEM and x-ray diffraction. The x-ray and positron annihilation study was performed after gently grinding the samples into powder form, as the as-grown ribbons were found to be very brittle and very thin for positron annihilation studies.

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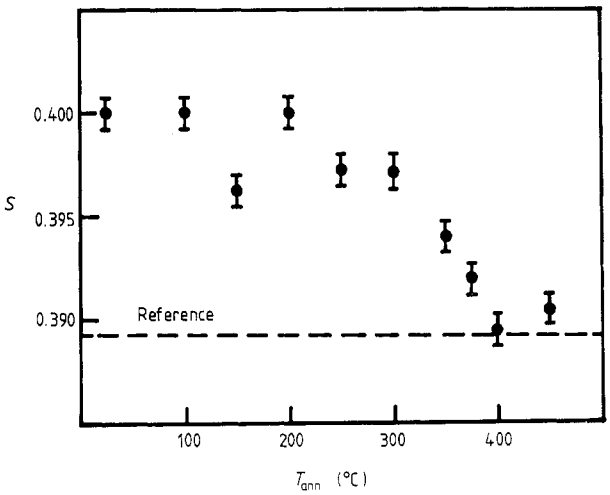

Figure 1. Change in Doppler-broadened annihilation line-shape parameter S as a function of the annealing temperature, T_{ann}.

A weak ^{22}Na positron source was evaporated on a thin Ni foil and an identical foil was used to cover the source. This source was placed inside a cylindrical glass sample holder and the quasicrystalline specimen, in powder form, was poured all around the source. A 40 cc HP Ge detector, with energy resolution (FWHM) of 1.10 keV at the 475 keV gamma line from a ^{102}Rh source, was used for this Doppler-broadening study. The structural relaxation and crystallisation of this quasicrystalline alloy were studied using isochronal annealing, which was on each occasion conducted in high vacuum. The sample was annealed at each set temperature for seven minutes, and then slowly cooled in vacuum for twenty minutes to avoid any quenching effects and surface contamination. The stability of any set temperature was better than ± 1 °C. All the positron measurements were performed at room temperature, keeping the specimens under vacuum throughout. The total counts accumulated under the Doppler spectra were about 0.8×10^6. Other experimental details have been given elsewhere [9].

The source–specimen assembly inside the cylindrical glass container was made only once at the start of the measurements. This assembly was not disturbed throughout the annealing steps and line-shape measurements. Of necessity, the positron source was also put inside the furnace along with the sample during isochronal-annealing steps. Since the highest annealing temperature in the present study was only 450 °C, which is much below the start of sublimation of the ^{22}Na source [6], the above procedure could be adopted. This procedure removed any uncertainty in the measurements arising out of the source–specimen geometry, the reproducibility of the assembly etc.

In figure 1 the Doppler-broadened annihilation line-shape parameter S has been plotted against the annealing temperature. S has its usual definition, i.e. the ratio of the area under the central channels to the total area under the annihilation curve. The broken line in the figure shows the parameter S for the reference crystalline phase, which was obtained by annealing the quasicrystalline sample in high vacuum for one hour at 450 °C after taking the last point of the figure at same temperature.

From the calorimetric study [8] of quasicrystalline Al–Mn alloy we know that this phase undergoes structural relaxation. Our positron annihilation result supports the calorimetric observations. Before the crystallisation process starts above 300 °C, the parameter S of the quasicrystalline phase slowly decreases from the room temperature

value of 0.3999 to 0.3973 (with an error of ±0.0008) at 250 °C and stabilises there. This process indicates the annealing of defects, which can trap positrons, in the as-grown sample. The sudden drop of S at 150 °C and its recovery at 200 °C may be due to the formation of a cluster of the aluminium precipitates and generation of vacancies respectively.

After 300 °C, figure 1 clearly indicates the crystallisation process, which is completed at 400 °C. The slow fall in S confirms the earlier suggestions [2, 8] that quasicrystalline-to-crystalline phase transition involves diffusion. Nucleation and growth of the crystalline phase occur at the interface between quasicrystalline grains and Al precipitates and are controlled by the diffusion of the Mn atoms across this interface.

It is well known that change in S at the solid–solid phase transition point can be attributed to the change in electron momentum distribution of the new phase. It is also known that the electronic structure of the quasicrystalline phase has many exotic features [10]. The fall of S in the crystalline phase suggests that the average electron momentum in the crystalline phase has increased. This fall of S can also be explained on the basis of an icosahedral glass model [2], which inherently needs disorder and defects in the quasicrystalline phase. While the qualitative information presented here on the basis of the parameter S is interesting and new, we feel that a detailed quantitative analysis of the defect kinetics based on the positron life-time measurements, which is under way, will shed more light on the structure of this new phase of matter.

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References

[1] Shechtman D, Blech I, Gratias D and Cahn J W 1984 *Phys. Rev. Lett.* **53** 1951
[2] Janot Ch and Dubois J M 1988 *J. Phys. F: Met. Phys.* **18** 2303
[3] Gratias D, Cahn J W and Mozer B 1988 *Phys. Rev.* B **38** 1643
[4] Onada G Y, Steinhardt P J, DiVincenzo D P and Socolar J E S 1988 *Phys. Rev. Lett.* **60** 2653
[5] Sahni V C and Sanyal M K 1989 *Pramana* **32** L77
[6] Mackenzie I K 1983 *Proc. Int. Sch. Phys. 'Enrico Fermi'* ed. W Brandt and A Dupasquier, p 196
[7] Sanyal M K, Sahni V C and Dey G K 1987 *Nature* **328** 704
Sanyal M K, Sahni V C and Chidambaram R 1988 *Solid State Commun.* **66** 1043
[8] Kelton K F and Holzer J C 1988 *Phys. Rev.* B **37** 3940
Chen H S and Chen C H 1986 *Phys. Rev.* B **33** 668
[9] Naidu S V, Sengupta A, Sen P and Bhandari R K 1985 *Solid State Commun.* **55** 27
[10] Janssen T 1988 *Phys. Rep.* **168** 55
Chakrabarti A, Karmakar S N and Moitra R K 1989 *J. Phys.: Condens. Matter* **1** 1017