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Migration stage of the vacancylike defects in icosahedral $Al_{74}Mn_{20}Si_6$ by positron-annihilation techniques

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We have performed positron-annihilation lifetime and Doppler-broadening measurements of quasicrystalline-phase $Al_{74}Mn_{20}Si_6$. It is observed that the mean lifetime and line-shape parameter h decrease when going from room temperature to $\sim 250\,°C$. From fitting with two lifetime components, it is interpreted that the decreases of the mean lifetime and h correspond to the migration and annihilation of vacancylike defects. The implication of this is discussed.

Quasicrystalline phases, which Shechtman *et al.*[1] discovered, do not have long-range translational symmetry. However, the sharpness of the diffraction spots with 10-fold rotation indicates that quasicrystalline phases possess long-range orientational order belonging to the icosahedral point group. That is, these phases are midway between the usual crystalline and amorphous phases. Therefore, investigation of the structures of quasicrystalline phases is a very important subject. Computed diffraction patterns from two different models, the three-dimensional Penrose-tiling (3DPT) model,[2-6] and the dense packing of icosahedra model[7,8] agree well with diffraction patterns observed experimentally.

Subsequent experiments have shown that the diffraction-peak widths in quasicrystalline phases correlate strongly with $G_\perp$ (phason momentum).[9] It has been discussed that quenched strains in the phason or dislocations can lead to peak broadening in icosahedral-diffraction patterns.[10,11] Also, it is thought that the relaxation of the phason is related to the migration of vacancies.[12]

Recently, Mukhopadhyay, Ranganathan, and Chattopadhyay[13] have reported diffuse scattering developing upon low-temperature annealing of icosahedral i(Al-Mn). The new diffusion spots occur along the icosahedral axes at multiples τ [$\tau \equiv (1+\sqrt{5})/2$] and τ^{-1} of the positions of the sharp spots. More recently, Henley[14] suggested an interesting idea to explain the new diffusion spots. That is, if one is forced to generate an even-odd superlattice of occupied and vacant Al(δ) sites in the rhombic dodecahedron of the structure model of Elser and Henley,[15] the diffusion spots can be explained.

In this study, we have performed positron-annihilation lifetime and Doppler-broadening measurements of quasicrystalline-phase $Al_{74}Mn_{20}Si_6$. It is found that the migration stage of vacancylike defects exists in the temperature region from room temperature to $\sim 250\,°C$. The relations between the migration stage of vacancylike defects and the new diffusion spots discovered recently[13] or the relaxation of the phason will be discussed.

The $Al_{74}Mn_{20}Si_6$ alloy was prepared by arc melting of high-purity Al, Si, and Mn. Thin ribbons (~ 1 mm in width and ~ 30 μm in thickness) were fabricated by a single-roller spinning in a helium atmosphere. The quasicrystalline phase was confirmed by x-ray diffraction analysis and transmission electron microscopy. The positron source, ~ 5-μCi ^{22}NaCl sealed in an aluminum thin foil, was set at the center of the specimen. The specimen was sealed in a Pyrex glass tube in a vacuum of 10^{-5} Torr. The isochronal annealing was performed from 20 to 600 °C. The annealing time was 20 min. The measurements of Doppler-broadening profiles were carried out at room temperature with a pure germanium detector (Oak Ridge Technology). The energy resolution of the apparatus [full width at half maximum (FWHM)] was 1.19 keV using 512-keV γ rays from ^{106}Ru. The total counts in a spectrum were 1.2×10^6. The peak drift was less than 0.2 channel through this experiment (one channel corresponds to 0.0608 keV). The positron-lifetime spectra were obtained with a fast-fast coincidence system by using HAMAMATSU R2076 photomultipliers and $\frac{3}{4}$-in. BaF_2 scintillators. The time resolution of the system was 260 psec (FWHM) with the use of ^{60}Co. Total counts were 1.0×10^6 in one spectrum. The measurements were carried out at room temperature.

The Doppler-broadening spectra were deconvoluted by the fast-Fourier-transform–power-spectrum method.[16] After background subtraction the line-shape parameter h was determined by the ratio of the central area over 20 channels to the total area of the spectrum and was normalized by setting the value of the h parameter of well-annealed Al to 1. Positron-lifetime spectra were analyzed by POSITRONFIT (Ref. 17) after subtracting the background and were resolved into exponential-type multicomponent curves which were broadened by the time-resolution function. The time resolution used was assumed to be composed of two Gaussian functions. With

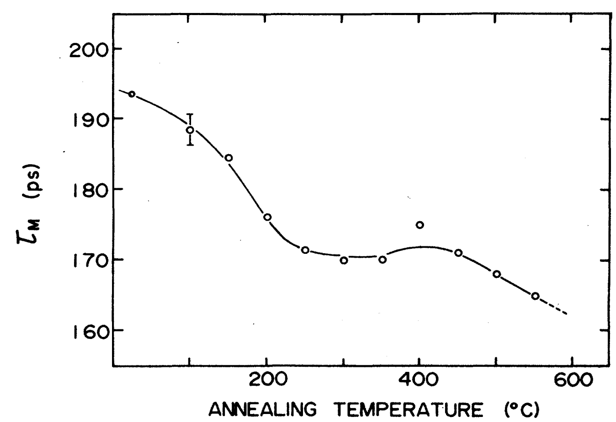

FIG. 1. The change in the mean lifetime during isochronal aging for 20-min intervals at temperatures 50 °C apart in the quasicrystalline-phase $Al_{74}Mn_{20}Si_6$.

use of this time-resolution function, the lifetime in the bulk of well-annealed pure Al (99.9999 wt.%) was estimated to be 165 ± 2 psec. Each χ^2/q was below 1.2.

Figure 1 shows the change in the mean lifetime during the isochronal aging for 20-min intervals at temperatures 50 °C apart in the quasicrystalline-phase $Al_{74}Mn_{20}Si_6$. The mean lifetimes decrease upon going from room temperature to ~250 °C. The mean lifetimes from ~250 to ~450 °C are ~170 psec. It looks as though the mean lifetimes decrease again from ~450 to ~600 °C. This change from ~450 to ~600 °C corresponds to the crystallization. Figure 2 shows the change in h during the isochronal aging for 20-min intervals at temperatures 50 °C apart in the quasicrystalline phase of $Al_{74}Mn_{20}Si_6$. The values of h decrease upon going from room temperature to ~300 °C. Then they decrease gradually between ~450 and ~600 °C.

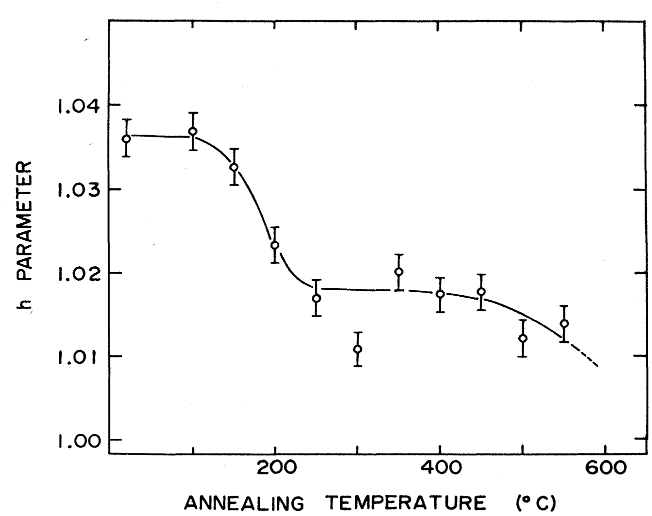

FIG. 2. The change in h during isochronal aging for 20-min intervals at temperatures 50 °C apart in quasicrystalline-phase $Al_{74}Mn_{20}Si_6$.

Now, we turn our attention to the influence of crystalline interfaces in quasicrystalline phases on the positron-lifetime spectra. The crystalline diameter in quasicrystalline-phase $Al_{74}Mn_{20}Si_6$ is ~1 μm. So far, the positron-diffusion length in quasicrystalline phases is not known. However, taking account of the fact that the properties of the quasicrystalline phase are located between those of amorphous alloys and the usual crystalline metals, it can be deduced that the positron-diffusion length at room temperature in the quasicrystalline phase is much shorter than ~1 μm. This fact shows that thermalized positrons mainly annihilate in the quasicrystalline phase without crystalline interfaces.

The observed lifetime spectra are well fitted with two lifetime components. Of course, we have tried analyzing the observed lifetime spectra with three lifetime components. Especially, we checked to see whether the third longer lifetime component existed in the temperature region above ~250 °C. However, it was very difficult to separate the observed lifetime spectra into three lifetime components. Figure 3 shows the changes in the intensity, I_2, of component 2, the lifetime, τ_2, of component 2, and the lifetime, τ_1, of component 1. The lifetime, τ_1, of component 1 is 170 ± 5 psec from room temperature to ~350 °C and subsequently decreases from ~350 to ~550 °C. It is thought that the value, ~170 psec, of τ_1 corresponds to the lifetime of positrons in the bulk of the

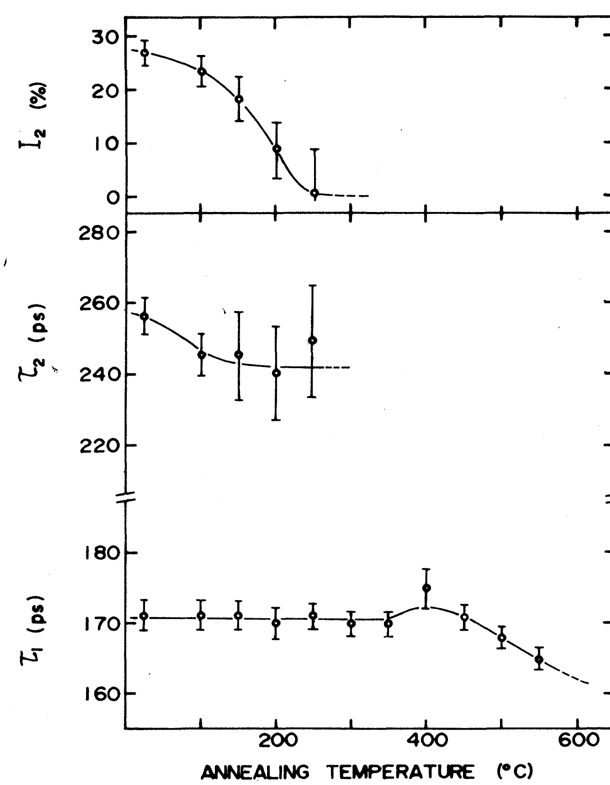

FIG. 3. The changes in the intensity, I_2, of component 2, the lifetime, τ_2, of component 2, and the lifetime, τ_1, of component 1 during isochronal aging for 20-min intervals at temperatures 50 °C apart in quasicrystalline-phase $Al_{74}Mn_{20}Si_6$.

quasicrystalline-phase $Al_{74}Mn_{20}Si_6$, which may include trapping sites such as Mn vacancy centers in Mackay icosahedra.[18] The decrease from ~350 to ~600 °C can be attributed to the crystallization. The value of the lifetime, τ_2, of component 2 is within 250 ± 10 psec as shown in Fig. 3. The intensity, I_2, of component 2 decreases upon going from room temperature to ~250 °C. Taking account of the value, ~250 psec, of τ_2,[19] it is thought that this component corresponds to that of vacancylike defects in the quasicrystalline phase. It should be noticed that the component-2 vacancylike defects do not indicate only one kind of trapping site but rather an average of distributional trapping sites. Therefore, we believe that the decreases of the mean lifetime and *h* from room temperature to ~250 °C reflect the migration and annihilation of vacancylike defects.

Recently, Elser[20] has shown that phason strains are contained in the quasicrystal during solidification. In fact, it has been found that the peak broadening and distortion in quasicrystal-diffraction patterns correlate strongly with $G_\perp$ (phason momentum). This effect can be explained by the presence of phason strain in quasicrystals. Levine *et al.*[21] have shown that dislocations in quasicrystals always have nonzero phonon fields, *u*, and phason fields, *w*. Therefore, to move a dislocation requires moving the associated phason field. This means that the motion of dislocations in a quasicrystal is expected to be comparable to the climb of dislocations. It is known that the climb of dislocations requires mostly migration of vacancies. In addition, Socolar, Lubensky, and Steinhardt[22] have discussed that the uniform shift in the phason field, *w*, corresponds to arrangements of the unit cells. This demonstrates that the relaxation of the phason requires the diffusion of atoms among the unit cells, which corresponds to the migration of vacancies. Taking the relaxation phenomena during the low-temperature aging[23,24] into consideration, it can be deduced that the migration of vacancylike defects from room temperature to ~250 °C in Figs. 1 and 2 causes the climb of dislocations or rearrangements of the unit cells in the quasicrystal. In addition, the present study suggests that the formation of the new diffraction spots upon low-temperature annealing[13] is closely related to the migration of vacancylike defects.

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