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Positron-annihilation studies on Al-Cu-Fe: Their significance for the structural models of icosahedral quasicrystals

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We have investigated the vacancy defects in samples of quasicrystalline Al-Cu-Fe using positron-lifetime measurements as a function of various heat treatments and alloy compositions. In general, a two-component positron-annihilation-lifetime spectrum seems characteristic of quasicrystalline Al-Cu-Fe and other icosahedral quasicrystals. This can be interpreted in terms of vacancy defects, whose concentrations are about three orders of magnitude more than what one usually observes in crystalline metals at room temperature. We discuss the significance of the results of our studies for the structure of icosahedral quasicrystals, for which several models have been postulated in the literature. The concentration of the vacancy defects observed by us seems incompatible with the icosahedral-twin model or, at first sight, with the space-filling Penrose tiling based models or random-tiling models.

I. INTRODUCTION

The Al-Cu-Fe quasicrystal is of special significance because it can also be grown from the melt[1] without employing a rapid solidification process, and has often been found to be nearly free of phason disorder.[2] We report here results of studies, using the positron-annihilation technique (PAT), on quasicrystalline Al-Cu-Fe alloys prepared from the melt and compare these results with those obtained from our earlier studies on rapidly solidified Al-Cu-Fe,[3] and other icosahedral quasicrystals such as Al-Mn,[4] Al-Mn-Si,[5] and Al-Cu-Li.[6] These studies have shown that the alloys, in the as-quenched state, contain vacancy defects, varying in size from monovacancies to six-vacancy size clusters in ppm concentrations. It was therefore important to examine by positron-annihilation technique (PAT) the vacancy defects in samples of quasicrystalline Al-Cu-Fe grown from melt with and without employing rapid solidification process. It is known that the quasicrystalline Al-Cu-Fe alloy exhibits resolution-limited diffraction peaks and as a result is believed to be a perfect quasicrystalline phase without phason disorder. We discuss in this paper the implications of our positron results on various proposed structural models of the quasicrystalline state.

Recently, different workers[7-13] have found very interesting effects of heat treatment on this quasicrystalline phase. Most of these changes occur due to heat treatment at or around 600 °C. The anomalous changes in x-ray-diffraction intensities and the resultant Debye-Waller factors between 400 and 700 °C have been interpreted by Bancel[9] as due to the softening of the phason modes. Janot *et al.*[11] studied *in situ* temperature patterns and high-resolution images and suggested that at 600 °C, the electron-diffraction spots have satellites along all fivefold directions, as observed with classical modulated structures. We have also studied, using PAT, quasicrystalline Al-Cu-Fe alloys after various kinds of heat treatments in the range 600 to 800 °C followed by either slow cooling or fast quenching.

II. EXPERIMENTAL PROCEDURES

It has been shown recently[10] that the phase diagram of Al-Cu-Fe is complicated and that it contains a number of phases related to the *i* phase. Although the composition of the first reported[1] quasicrystalline phase was $Al_{65}Cu_{20}Fe_{15}$, it is now known[10] that the samples with compositions around $Al_{63}Cu_{25}Fe_{12}$ do not generally decompose into other phases under heat treatment.

We present here results of four different samples. The first two samples (referred to as samples 1 and 2) are of compositions $Al_{63}Cu_{25}Fe_{12}$ and $Al_{62}Cu_{25.5}Fe_{12.5}$, respectively. These two samples were prepared by slow cooling from the melt in an inert atmosphere and were annealed for 63 and 60 h, respectively at 800 °C. We have also studied two other samples (referred to as samples 3 and 4), prepared by the usual melt-spinning technique, whose compositions are $Al_{61}Cu_{23}Fe_{16}$ and $Al_{65}Cu_{20}Fe_{15}$, respectively. The quasicrystalline character of all these samples was verified by electron- and powder x-ray-diffraction techniques. Positron-annihilation studies were made on powders obtained by gentle grinding of the ribbons in the case of samples 3 and 4. The thickness of the Al-Cu-Fe sample was at least 5 mm around the source to ensure that annihilation of positrons take place inside the sample only. The positron source was sandwiched between two tablets (5 mm×4 mm×1 mm) taken from the ingots in

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the case of samples 1 and 2. In all these experiments a weak (4 μCi) ^{22}Na source was evaporated on a thin (about 3 μm) nickel foil and was covered by an identical foil. The sample holder has been continuously evacuated during the measurements.

The annealing was carried out in an argon atmosphere, followed by either slow cooling, also in an argon atmosphere, or by fast quenching in liquid nitrogen. In the case of slow cooling, from the high temperature, the sample was brought down to room temperature in about 20 min. The process of fast quenching was achieved by transferring the tantalum foil containing the specimens from the furnace to liquid nitrogen in less than 10 s. The starting specimen for each of these annealing treatments is the originally prepared quasicrystalline alloy. Each time after the heat treatment, the source specimen geometry was reassembled. All the measurements were done at room temperature.

Positron-lifetime measurements were done using a conventional slow-fast coincidence spectrometer employing NE 111 plastic scintillators and RCA 8575 photomultiplier tubes. The prompt time resolution (full width at half maximum) of this spectrometer at the experimental window settings (with the upper 50% of the Compton spectra of 1.276 MeV and the 0.511-MeV γ rays accepted in the start and atop channels, respectively) was 260 ps for the coincidence γ rays from a ^{60}Co source. The total counts accumulated under the positron-lifetime spectra were about 1.5×10^6. Typical lifetime spectra are shown in Fig. 1. The lifetime spectra were analyzed with the help of standard computer codes RESOLUTION and POSITRONFIT.[14] All the spectra were corrected for background and source contribution. A single Gaussian was used in the resolution function throughout the analysis. Though the relevant fitting parameters such as the width and the centroid of the Gaussian were allowed to vary from spectrum to spectrum, no significant variation was observed. In all cases excellent variance of fit (VOF) were obtained.

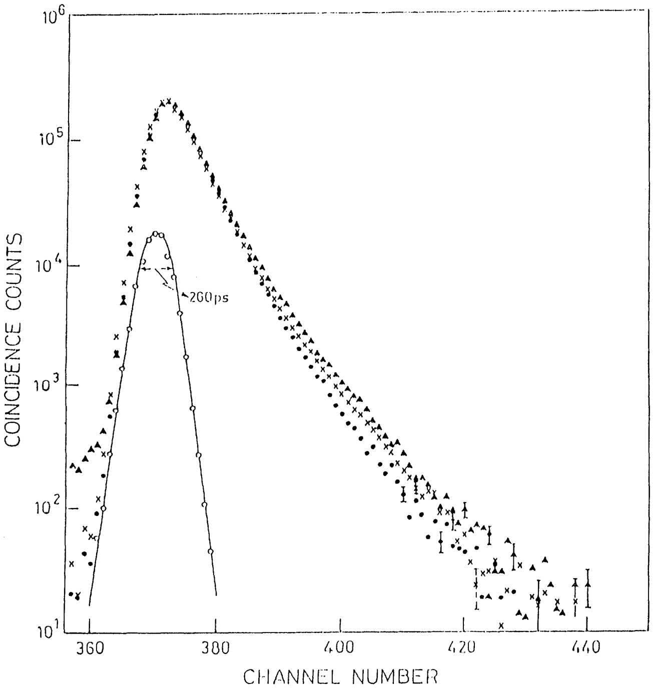

FIG. 1. Peak-normalized positron-lifetime spectra of $Al_{61}Cu_{23}Fe_{16}$ sample 3 in the as-quenched state (●) and after the respective heat treatments at 600 °C (▲) and 800 °C (×), along with the prompt time coincidence spectrum of ^{60}Co γ rays (○) 1 channel = 50 ps.

III. RESULTS AND DISCUSSION

A. Ingot-grown and melt-spun Al-Cu-Fe

We have tabulated in Table I the main results of our studies. All the lifetime spectra obtained in this work could be analyzed on the basis of two components. The second component indicates the presence of positron traps, which can be interpreted as vacancy clusters in the as-grown quasicrystalline Al-Cu-Fe phase. In the two-state trapping model (as discussed in our earlier publications[3,5]) used for the analysis, it is assumed that annihilations can only occur from either the bulk state or the positron trap state, giving rise to a two-component lifetime spectrum and the size and concentration of these vacancy clusters can be calculated. The sizes and concentrations of these vacancy clusters calculated using this method is tabulated in Table II.

In agreement with our measurements, Howell *et al.*[15] have also recently reported two-component lifetime spectra on quasicrystalline samples of $Al_{63}Cu_{25}Fe_{12}$ and suggested that there are "open volumes of fixed dimension in the structure." But Lawther and co-workers[16] claim to have shown that a quasicrystal (Al-Cu-Fe) can be prepared with a positron trapping site concentration below the sensitivity of detection by positrons, i.e., at or below 10^{-7}. The results shown in Table I are consistent with our earlier study[3] on rapidly solidified Al-Cu-Fe quasicrystalline alloy. In that study the positron-lifetime spectra could also be resolved into two distinct lifetime components. The present results from samples 1 and 2 clearly show that the presence of the second lifetime component is not due to the rapid solidification process.

We also rule out the possibility that the longer lifetime component τ_2 obtained in the alloys studied by us might arise from surface effects in the powder samples used. The mean positron diffusion lengths in these alloys are of the order of 0.2 μm and for the surface effects to dominate, the particle size would have to be much smaller than 1 μm. To see such surface effects, Morinaga[17] had started with ultrafine silver particles 70 nm in mean diameter and found that the critical diameter of silver particles below which almost all the positrons are annihilated from the trapped surface states may be estimated to be a few hundred nm. On the other hand, the particle size in our experiments was about 30 μm. Even in oxide powders where positroniumlike states weakly bound to the surfaces have been studied, particles of size about 10–50 nm have been generally used.[18]

TABLE I. Results of positron-lifetime measurements on quasicrystalline Al-Cu-Fe alloys. Typical errors in τ_1, τ_2, I_2, and τ_m are 4 ps, 4 ps, 3%, and 2 ps, respectively. VOF is the variance of fit in the data analysis.

	Heat treatment	VOF	τ_1 (ps)	τ_2 (ps)	I_2 (%)	τ_m (ps)
Ingot-grown Sample No. 1	As grown	1.047	170	375	28.9	229
	Heat treated at 600 °C for 24 h and fast quenched	0.897	170	375	32.3	236
	Heat treated at 700 °C for 24 h and fast quenched	1.217	169	377	32.4	236
Ingot-grown Sample No. 2	As grown	1.139	172	389	22.9	222
Melt-spun Sample No. 3	As quenched	1.010	150	238	50.2	194
	Heat treated at 600 °C for 6 h and slow cooled	1.441	158	308	36.6	212
	Heat treated at 800 °C for 6 h and slow cooled	1.001	158	292	35.1	205
Melt-spun Sample No. 4	As quenched	1.02	154	365	20	197

TABLE II. Defect size, concentration, and defect volume as deduced from positron-lifetime data in as-prepared icosahedral quasicrystals.

Quasicrystal	Size of defect N (in terms of monovacancies per cluster)	Concentration C (in ppm)	Defect volume $C \times N$ (in ppm)	Reference
1. Melt-spun Al-Mn	3–4	1	3–4	4
2. Melt-spun Al-Mn-Si	2	3	6	5
3. Ingot-grown Al-Cu-Li	2–3	1–2	3–4	6
4. Melt-spun Al_{61}-Cu_{23}-Fe_{16} (Sample 3)	1	8	8	3
5. Melt-spun Al_{65}-Cu_{20}-Fe_{15} (Sample 4)	6	1	6	Present work
6. Ingot-grown Al_{62}-$Cu_{25.5}$-$Fe_{12.5}$ (Sample 2)	6	1	6	Present work
7. Ingot-grown Al_{63}-Cu_{25}-Fe_{12} (Sample 1)	5	1	5	Present work

B. Heat-treated Al-Cu-Fe

We have carried out heat treatments of all the four samples at various temperatures. After each treatment, x-ray diffraction was performed to determine the quasicrystalline character of the sample. In the case of samples 2 and 4 extra diffraction peaks indicating the presence of crystalline phases was observed. The diffraction lines characteristic of pure quasicrystalline Al-Cu-Fe were only found in the case of samples 1 and 3 even after prolonged heat treatments, and we have included positron results of these heat-treated samples in Table I.

In the first set of experiments we studied effects of heat treatments on sample 1 annealed at 600 and 700 °C for 24 h each, followed by fast quenching to liquid-nitrogen temperature. As can be seen, all positron-lifetime spectra could be analyzed with two-component fittings giving rise to a good VOF (refer to Table I). The observed changes in the lifetime parameters are within error bars, and as a result the size (corresponding to pentavacancy) and concentration (corresponding to 1 ppm) of the vacancy clusters basically remains unchanged even after this prolonged heat treatment. In Figs. 2 and 3 we show the x-ray-diffraction patterns of sample 1 after heat treating it at various temperatures. These patterns are indexed according to the scheme described in Ref. 10. The x-ray-diffraction patterns of the as-grown sample is also shown for comparison. The peak widths for the 600 °C heat-treated specimen are found to be broader (refer to Fig. 3) compared to those for the as-grown and the 700 °C heat-treated sample in general agreement with the earlier observations.[7,9] The diffraction peak widths for the as-grown and 700 °C heat-treated samples were found to be resolution limited. The broadening of the diffraction peaks is generally explained in terms of an increase in "phason disorder" in the quasicrystalline alloys. It should be mentioned here that most of the other quasicrystalline phases apart from Al-Cu-Fe show much broader peak widths.

In another series of heat treatment experiments we annealed two specimens of the same rapidly solidified alloy (sample 3) for 6 h each in an argon atmosphere, one at

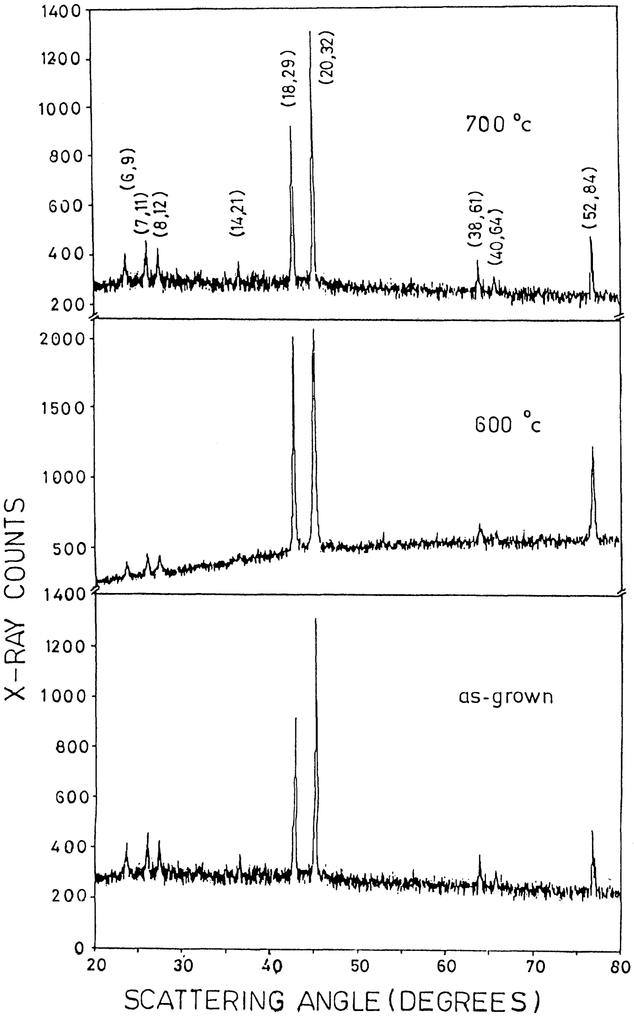

FIG. 2. X-ray-diffraction patterns of $Al_{63}Cu_{25}Fe_{12}$ sample 1 in the as-grown state (lower), heat treated at 600 °C (middle) and at 700 °C (top). The patterns have been offset vertically.

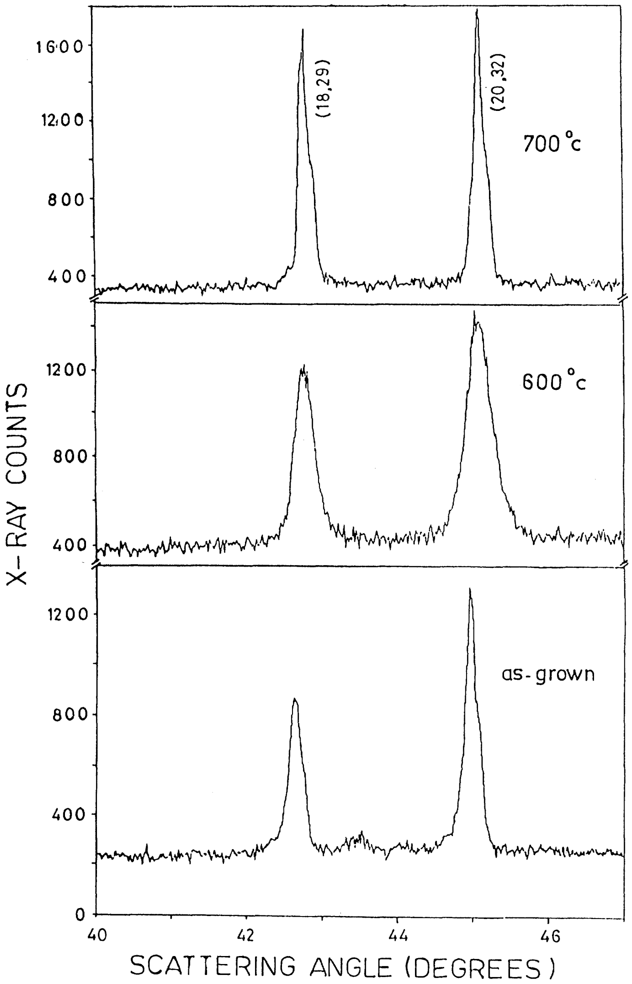

FIG. 3. Same as Fig. 2 but in an expanded scale between scattering angles of 40° and 47°.

600 °C and the other at 800 °C, and then slow cooled. Figure 1 shows the peak-normalized positron-lifetime spectra for this alloy in the as-quenched state and after the heat treatments. The positron-lifetime parameters are given in Table I. In this sample again we find that the x-ray-diffraction peak width of the sample increases when the sample is annealed at 600 °C. Like sample 1 here also we do not find the presence of crystalline phase in the x-ray-diffraction patterns. From Table I and Fig. 1, one can see that the mean positron-lifetime for the 600 °C heat-treated specimen has also been extended compared to that for the as-grown or the 800 °C annealed one. An analysis of these spectra indicates that the heat treatments at 600 and 800 °C have increased the vacancy size to trivacancy from the monovacancy size in the as-grown state, while the concentrations have been correspondingly reduced. One very significant observation from these studies is that the positron-lifetime spectra of the samples always showed two lifetime components, irrespective of the heat treatments. There is a high degree of stability of the vacancy defects in the alloy which do not anneal out even to a small extent on aging up to several days.

In case of samples 2 and 4, with identical heat treatments described above, a significant fraction of the crystalline phase was observed in the x-ray patterns of the heat-treated samples. However, the presence of the crystalline phase does not produce a change in the lifetime parameters. The extra lines present in the x-ray-diffraction pattern of the fast-quenched specimen as observed correspond to the monoclinic crystalline phase based on the stoichiometry Al_3Fe with some Cu substituted for Fe in the lattice. It has been shown[19] by one of us in an independent experiment that in the rapidly solidified Al-Cu-Fe quasicrystal, two distinct crystalline phases may coexist, albeit in very small amounts. One of the phases is AlCu (orthorhombic) and the other is Al_3Fe (monoclinic). The stability of vacancy defects in the quasicrystalline phase as a function of heat treatment up to even 800 °C implies a low mobility of these defects. When the crystalline phase grows in a region surrounded by the quasicrystalline matrix, the defects in the crystalline region would only be able to migrate and condense at the interface between the two phases. Hence it is reasonable to expect that the overall vacancy defect structure may not change significantly when small crystalline regions grow within the quasicrystalline matrix so that there is no significant change in the positron-lifetime spectrum, as observed in our experiments.

IV. IMPLICATIONS OF OUR POSITRON-ANNIHILATION RESULTS FOR THE MODEL OF ICOSAHEDRAL QUASICRYSTALS

In summary, a two-component positron-annihilation-lifetime spectrum seems characteristic of all icosahedral quasicrystals we have studied so far; melt-spun Al-Mn, Al-Mn-Si, and Al-Cu-Fe and ingot-grown Al-Cu-Li and Al-Cu-Fe. We give in Table II the vacancy size and concentration in various quasicrystalline alloys as deduced from the lifetime spectra. While the exact numbers in Table II are not significant—being sample dependent—it is clear that typical void volumes of a few ppm exist in these icosahedral quasicrystals. In the case of metastable quasicrystalline alloys we observe a single-component lifetime[5] spectra with a lifetime of 158±2 ps, for example, in the case of Al-Mn-Si after transformation to the crystalline phase.

There has been speculation[20] and somewhat widespread acceptance that the basic icosahedral cluster unit in the quasicrystalline phase is the so-called "Mackay" icosahedron with a hole in the center (in the original paper of Mackay,[21] it must be mentioned, the successive layers were built up around a central atom). If such a central hole existed, with a radius of about 90% of that of a monovacancy, it would act as a saturation trap for positrons and we would have obtained a single-component lifetime spectrum with a mean lifetime τ_m a little above 200 ps. Hence our positron-annihilation data rule out the possibility of a central hole in the icosahedral building block in the quasicrystalline phase. It has also been confirmed by us recently in more detailed band-structure calculations[22] that existence of central hole is not energetically favorable.

We now discuss the possibility of accommodating such a large concentration of vacancy defects found in our studies in the proposed structural models of quasicrystalline state, such as the Penrose tiling model,[23] the random-tiling model[24] or the icosahedral-twinning model.[25] Our results are clearly incompatible with the icosahedral-twinning model, since this model is essentially based on the crystalline state itself. The basic motivation in the perfect Penrose tiling and the random-tiling models is space filling as in the crystalline state and hence one should expect to find here also vacancy concentrations similar to those in the crystalline state. The fact that one finds vacancy concentrations three orders of magnitude more in the icosahedral quasicrystals compared to metallic crystals makes tiling models seem at first sight incompatible with the positron-annihilation results. However, it is very interesting to note[26] that the growth process of Penrose tiles is favored by vacancy—such as defects called decapods,[27] and it might be possible to interpret our results in terms of such defects.

On the other hand, the vacancy concentrations are at least three orders of magnitude lower than what one usually observes in metallic glasses.[28] This indicates that the quasicrystalline state is in some sense intermediate between the crystalline state and the amorphous state. In fact, this conclusion seems to be supported by several nondiffraction experiments. For example, some recent interesting experiments by Eckert, Schultz, and Urban,[29] show that ball milling at various speeds of the $Al_{65}Cu_{20}Mn_{15}$ alloy can mutually transform the crystalline, the quasicrystalline and the amorphous states and that the quasicrystalline phase occurs at speeds intermediate to those for the crystalline and the amorphous phases!

All these studies, in particular our positron-annihilation results, would tend to favor the icosahedral glass model[30] (with suitable modifications to reduce the predicted excessive vacancy regions; these could be reduced by annealing as has been shown in computer simulation studies by Elser[31] or the more recent icosahedral cluster-based model[32]).

It is clear from, say, extended x-ray-absorption fine-structure experiments[33] that icosahedral clusters of atoms exist in all the phases—crystalline, quasicrystalline, and amorphous phases, which can be produced in some rapidly quenched alloys just by varying the rate of quenching. If given adequate time during the solidification process, they tend to organize themselves into a crystalline phase (except in some alloys such as Al-Cu-Fe or Al-Cu-Ru). At the highest rates of quenching, the orientations of the clusters would be expected to be random leading to the amorphous phase and quite likely most of the clusters would be imperfectly formed. At intermediate rates of quenching, the clusters are formed and link themselves up sharing symmetry axes or vertices but may not be able to get the full coordination of, e.g., eight around each cluster to get a bcc coordination. But in the process all the clusters are aligned parallel to each other so that one gets Bragg-like peaks during diffraction experiments.

There is some evidence from electron channeling experiments[34] in quasicrystalline Al-Cu-Fe that the atoms occupy more specific atomic positions in the icosahedral clusters of this alloy while, in quasicrystalline Al-Mn-Si there is more disorder among the Al(Si) and Mn atoms. One consequence of this is that the sizes of the icosahedral clusters would be more uniform in Al-Cu-Fe than in Al-Mn-Si and also the symmetry axes would be more nearly oriented towards the angles expected for a perfect icosahedron. It would be interesting to correlate the observation of lower phason disorder in Al-Cu-Fe to the experimental results presented here.

V. CONCLUSIONS

A two-component positron-annihilation-lifetime spectrum seems characteristic of the icosahedral quasicrystalline phase. As indicated in Table II, in the large number of quasicrystals that we have studied, their lifetime spectra correspond to vacancy defects of 1–8 ppm, the vacancy size varying from monovacancies to hexavacancies, leading to typical void volumes of a few ppm. The vacancy concentrations seen by us in icosahedral quasicrystals are about three orders of magnitude more than what one observed in crystalline alloys and about three orders of magnitude lower than what one usually observed in metallic glasses. Thus, in some sense, the quasicrystalline state is intermediate to the crystalline state and the amorphous state. This conclusion is supported by several other nondiffraction experiments. We feel that our positron-annihilation data are compatible with the icosahedral glass model (with suitable modifications to anneal the excessive vacancy regions predicted by this model in its simplistic form) or the more recent icosahedral cluster-based model. While there is some entropy inherent in these two models, they do not seem to be dependent for their stabilization on entropy as the random-tiling models do. In fact, some recent calculations[35] and photoemission studies[36] indicate the existence of a universal pseudogap at the Fermi energy of icosahedral quasicrystals and this could be the factor which enhances the cohesive energy of these structures and leads to their stabilization.

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