Positron lifetimes in a binary quasicrystal Cd-Yb and its crystalline approximant

K. Sato,* H. Uchiyama,† K. Arinuma, and I. Kanazawa
Department of Physics, Tokyo Gakugei University, 4-1-1 Koganei, Tokyo 184-8501, Japan

R. Tamura, T. Shibuya, and S. Takeuchi
Department of Materials Science and Technology, Science University of Tokyo, Noda, Chiba 278-8501, Japan
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The positron lifetime measurements for a recently discovered binary icosahedral quasicrystal ($Cd_{84.6}Yb_{15.4}$) and its cubic approximant (Cd_6Yb) have shown that the lifetime spectra in both alloys are composed of a single component with the lifetime of 230 ± 3 ps for the quasicrystal and 234 ± 3 ps for the cubic approximant; these values being close to the reported lifetime (250 ps) of positrons trapped in lattice vacancies in crystalline Cd. Based on the atomic structure of the cubic approximant, the positrons are most likely to be annihilated inside the dodecahedral Cd clusters in which half of the eight cubic Cd sites are unoccupied. Essentially the same lifetimes in the quasicrystal and the cubic approximant strongly suggest that the positrons are trapped in the same type of structural vacancies common to both alloys, implying that the quasicrystal is also composed of the same cluster units as the cubic approximant.

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Stable binary icosahedral quasicrystals (QC's) have been discovered recently in Cd-Yb and Cd-Ca alloys.[1,2] The discovery of the two-component QC's would make the theoretical and experimental investigations much simpler than previous ternary QC's due to much simpler chemical structure. Moreover, Takakura *et al.*[3] have shown that the structure of the cubic Cd_6Yb (Ref. 4) and Cd_6Ca crystals are closely related to those of icosahedral QC Cd-Yb and Cd-Ca (thus they are named the crystal approximant), and predicted that icosahedral QC's Cd-Yb and Cd-Ca belong to a new type of QC structure different from the previously established Mackay icosahedron (MI) type and rhombic triacontahedron (RT) type. Tamura *et al.*[5] first reported that electronic properties of the icosahedral QC Cd-Yb are remarkably different from those of ternary icosahedral QC's. Such reports mentioned above provide some insight into understanding the electronic and atomic structures of icosahedral QC Cd-Yb and Cd-Ca, but more experimental data should be urgently accumulated. In this paper, we present an important information of the structure of the binary icosahedral QC's by means of positron-annihilation measurements.

Up to now, positron-annihilation studies on the ternary icosahedral QC's have shown that extremely high concentration of the vacancy-type defect commonly exists in thermally stable icosahedral QC's.[6] Such a vacancy-type defect densely remains in the icosahedral QC's even after the long-time annealing of more than three months just below the melting point.[7] We believe that this kind of vacancy-type defect in the icosahedral QC's corresponds to the structural vacancy such as the vacant center site of the MI cluster in α-AlMnSi 1/1 approximant. In the conventional lifetime spectroscopy, all the incident positrons are naturally trapped into this type of structural vacancy, namely, the saturation trapping occurs due to the high structural vacancy density. Hence, the density cannot be determined. Recently, we have applied the slow positron beam technique to the icosahedral QC's in order to overcome this serious problem,[8] and successfully determined the structural vacancy density in the icosahedral QC's (on the order of 10^{20} cm^{-3}) under the saturation trapping condition.[9–11]

Alloys of the icosahedral QC $Cd_{84.6}Yb_{15.4}$ and the cubic crystal Cd_6Yb were prepared by the following procedure. Appropriate amounts of pure Cd grains (six-nine purity) and Yb flakes (three-nine purity) were charged in an alumina crucible with an inner diameter of 8 mm, which was sealed in a quartz tube in vacuum, heated to 973 K for 24 h to produce a homogeneous alloys melt and then furnace cooled. The obtained alloys were confirmed to be a single phase by powder x-ray spectroscopy, as shown in Fig. 1. Each alloy ingot was cut into 1 mm thick disks for the position annihilation measurements.

The positron-annihilation lifetime measurements were carried out at room temperature. The positron source (^{22}Na activity ~5 μCi), sealed in a thin foil of Kapton, was mounted in a specimen-source-specimen sandwich. The positron-annihilation lifetime spectra were recorded with a fast-fast coincidence system employing H2431 photomultiplier by HAMAMATSU with 1×1 in^2 BaF_2 scintillators. The time resolution of this system was 230 ps full width at

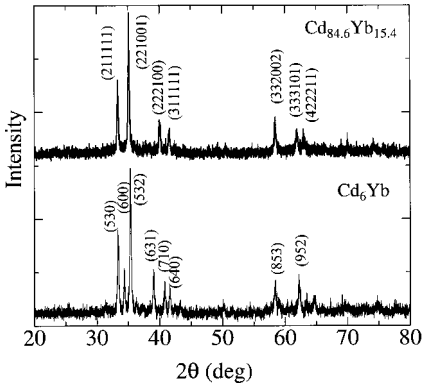

FIG. 1. Powder x-ray diffraction spectra for icosahedral QC $Cd_{84.6}Yb_{15.4}$ and cubic crystalline Cd_6Yb.

TABLE I. Positron lifetimes of the icosahedral QC $Cd_{84.6}Yb_{15.4}$ and the cubic crystal Cd_6Yb. Atomic density ρ_{at} presented in the table have been estimated from the composition and density. Experimental uncertainty is $\Delta\tau = \pm 3$ ps. The positron lifetime τ_f in the free state and the positron lifetime τ_v in lattice vacancies of the pure Cd and Yb are given for comparison.

Specimen	ρ_{at} [10^{28} m^{-3}]	τ_1 [ps]	τ_2 [ps]
QC $Cd_{84.6}Yb_{15.4}$	4.29		230
cubic Cd_6Yb	4.31		234
		τ_f [ps]	τ_v [ps]
pure Cd	4.63	190 [a]	250 [a]
pure Yb	2.42	260 [b]	

[a]Reference 13.
[b]Reference 14.

half maximum (FWHM). For each spectrum at least 1×10^6 annihilations were counted. The time resolution function was assumed to be composed of two Gaussian functions. Using this time resolution function, the lifetime in the bulk of well-annealed pure Al (six-nine purity) was measured as 165 ± 2 ps. After subtracting the background, positron-annihilation lifetime spectra were analyzed using POSITRONFIT.[12]

The observed positron-annihilation lifetime spectra for both specimens were composed of a single component, and the lifetime of 230 ± 3 and 234 ± 3 ps were evaluated for QC $Cd_{84.6}Yb_{15.4}$ and cubic crystal Cd_6Yb, respectively, from the observed lifetime spectra. Table I lists the positron lifetimes with the atomic densities of QC $Cd_{84.6}Yb_{15.4}$ and cubic approximant crystal Cd_6Yb together with those of the single elements. Considering that the atomic densities of QC $Cd_{84.6}Yb_{15.4}$ and cubic crystal Cd_6Yb, which are estimated from the composition and density,[4] are close to that of pure Cd, significantly high values were obtained (Table I). Here we have estimated the positron lifetimes in the free state for QC $Cd_{84.6}Yb_{15.4}$ and cubic crystal Cd_6Yb approximating those to be the compositionally weighted average of the constituent element values [$\tau_{Cd} = 190$ ps (Ref. 13) and $\tau_{Yb} = 260$ ps (Ref. 14)]. The estimated values are 201 and 200 ps for QC $Cd_{84.6}Yb_{15.4}$ and cubic approximant crystal Cd_6Yb, respectively, which are significantly shorter than the lifetimes τ_2 observed here. Therefore it is unlikely that the obtained positron lifetimes of 230 and 234 ps are due to annihilations in the defect-free region of the specimens.

In the case of the cubic approximant crystal Cd_6Yb, its atomic structure has been determined from a single crystal x-ray diffraction analysis:[4] it can be described as bcc packing of three-layered icosahedral atomic clusters. The first shell inside the dodecahedral second shell of 20 Cd atoms consists of four Cd atoms, which occupy four sites among equivalent eight sites, the other four sites remain vacant, as illustrated in Fig. 2. These vacant Cd sites are likely to serve as trapping sites for positrons and the observed long positron lifetime in the cubic crystal Cd_6Yb most probably corresponds to that at these vacancy sites.

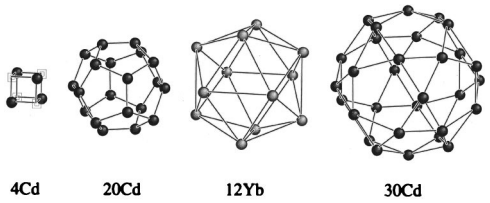

FIG. 2. Illustration of the successive icosahedral cluster shells in cubic crystalline Cd_6Yb.

Interestingly, we have observed essentially the same positron lifetime (230 ps) in the case of QC $Cd_{84.6}Yb_{15.4}$, which clearly indicates the existence of the same type of the structural vacancies in the QC. The almost identical lifetimes between the QC and the approximant are solely attributed to their common local environments. In addition to that, we should note that the observed lifetimes are close to the lifetime of positrons trapped at lattice vacancies in crystalline Cd (see Table I). It is worth noting that the nearest neighbors of these vacancy sites are Cd atoms in the cubic crystal Cd_6Yb. The slightly shorter lifetimes for the approximant and the QC may indicate smaller vacancies with respect to those of pure Cd.

As pointed out by Takakura *et al.*,[3] the local atomic structure of the icosahedral QC $Cd_{84.6}Yb_{15.4}$ is closely related to that of cubic Cd_6Yb. Also, the atomic densities of the QC and the approximant are nearly the same as described above. Therefore, the present results strongly support that the structure of QC $Cd_{84.6}Yb_{15.4}$ is composed of the same structural units (shown in Fig. 2) as the cubic crystal approximant of Cd_6Yb.

In conclusion, the positron lifetimes for a recently discovered binary icosahedral QC $Cd_{84.6}Yb_{15.4}$ and its approximant Cd6Yb are reported. The obtained lifetimes are found to be essentially the same between the two alloys and are close to the reported lifetime of positrons trapped in lattice vacancies in pure Cd. Furthermore, the long lifetime of the approximant is attributed to structural vacancies existing inside dodecahedral Cd clusters and the identical lifetime obtained for the QC strongly supports that the QC also has the same cluster units as the approximant.

*Present address: Institut für Theoretische und Angewandte Physik, Universitaet Stuttgart, Pfaffenwaldring 57, D-70550 Stuttgart, Germany.
†Present address: The Graduate University for Advanced Studies Department of Materials Structure Science, 1-1 Oho, Tsukuba, Ibaraki, 305-0801, Japan.

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