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Mössbauer effect study of the decagonal quasicrystal $Al_{65}Co_{15}Cu_{20}$

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Abstract The ^{57}Fe Mössbauer effect study at 5.0 K and in an external magnetic field of 9.0 T on a high-quality stable decagonal quasicrystal $Al_{65}Co_{15}Cu_{19.9}Fe_{0.1}$ is presented. It is shown that the iron atoms are located in two distinct classes of sites. The values of the principal component of the electric field gradient tensor and the asymmetry parameter at these sites are, respectively, $-1.90(10)\times10^{21}$ V/m^2, 0.97(15) and $-3.95(12)\times10^{21}$ V/m^2, 0.00(17).

Key words quasicrystal · decagonal alloy · electric field gradient

1 Introduction

One of the open questions in studies of quasicrystals (QCs) is to determine their atomic structure, which is a prerequisite to understand many unusual physical properties of these alloys [1]. In spite of significant progress in recent years, the complete determination of the structure of QCs has not yet been accomplished [2].

The first, and until now, the only quantitative X-ray diffraction (XRD) study of the structure of the decagonal Al–Co–Cu QC using the five-dimensional description of the structure of a decagonal QC was carried out by Steurer and Kuo [3, 4]. The difficulty with the five-dimensional approach is that only an average structure can be determined because disorder, which is clearly present in the decagonal Al–Co–Cu QC [5], cannot be treated properly. A relatively small number of Bragg reflections available [2] leads to some spurious atoms with unphysical inter-atomic separations in structural models. In addition, a problem of XRD analysis is that it is not possible to distinguish between the different transition metal (TM) atoms in the ternary Al–Co–Cu QC. It appears that the XRD

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investigations alone might not be able to solve the structure of the decagonal Al–Co–Cu QC.

Complimentary to the diffraction methods of structure determination are the local probes, such as extended X-ray absorption fine structure (EXFAS), nuclear quadrupole resonance (NQR), or Mössbauer spectroscopy (MS), which are element selective and sensitive to the local atomic structure. It has been recently demonstrated [6] how the combination of the complete set of the MS data and the calculations of the electric field gradients (EFGs) for several XRD-based structural models of the icosahedral Al-Cu-Fe QC led to the solution of the structure of this QC.

The main objective of the present study is to provide a complete set of the EFG parameters at the Cu sites in the decagonal Al-Cu-Co QC.

2 Experimental procedure

An ingot of nominal composition $Al_{65}Co_{15}Cu_{19.9}Fe_{0.1}$ was prepared by melting in an argon atmosphere of high-purity constituent elements using an arc furnace; the Fe metal used was enriched to 95.9% in the ^{57}Fe isotope. The ingot was annealed in vacuum at 1073 K for 48 h.

^{57}Fe MS measurements were performed using a standard Mössbauer spectrometer operating in a sine mode. Mössbauer spectra in zero external magnetic field and in an external magnetic field of 9.0 T parallel to the γ-ray propagation direction were measured with the $^{57}Co(Rh)$ source held at the same temperature as that of the sample. The spectrometer was calibrated with a 6.35-μm α-Fe foil (the surface density of 107×10^{-3} mg $^{57}Fe/cm^2$), and the spectra were folded. The full line width at half maximum of the inner pair of the α-Fe Zeeman pattern was 0.224(4) mm/s and this value can be regarded as the resolution of the Mössbauer spectrometer. The Mössbauer absorber was prepared by mixing the powdered alloy with powdered BN to ensure a uniform thickness of the absorber and the random orientation of sample particles. This mixture was then put into a plastic sample holder. The surface density of the Mössbauer absorber was 36×10^{-3} mg $^{57}Fe/cm^2$.

3 Results and discussion

The XRD analysis of the studied QC showed [7] that all the Bragg peaks could be indexed to the decagonal structure, with a quasilattice parameter a=7.185 Å perpendicular to the 10-fold axis and a quasilattice stacking periodicity c=4.129 Å along the 10-fold axis.

The low-temperature Mössbauer spectrum of the decagonal $Al_{65}Co_{15}Cu_{19.9}Fe_{0.1}$ QC (Figure 1), in contrast to Mössbauer spectra of icosahedral QCs which are in a form of a somewhat broadened single quadrupole doublet [8], exhibits a clear structure. It can be fitted unequivocally with two symmetric quadrupole doublets (Figure 1). Each doublet is characterized by a full line width at half maximum Γ, a relative area A, a centre shift δ (relative to α-Fe at 298 K), and a quadrupole splitting $\Delta = \frac{1}{2}eQ|V_{zz}|\left(1+\frac{1}{3}\eta^2\right)^{1/2}$ where e is the proton charge and Q is the electric quadrupole moment of the nucleus. The asymmetry parameter is $\eta = \left|\left(V_{xx}-V_{yy}\right)/V_{zz}\right|, (0 \le \eta \le 1)$, where V_{xx}, V_{yy}, and V_{zz} are the eigenvalues of the electric field gradient (EFG) tensor in order of increasing magnitude. The values of Γ, A, δ, Δ determined from the fit (χ^2=1.09) for each quadrupole doublet are, respectively, 0.313(9) mm/s, 63.0(2.6)%, 0.299(2) mm/s, 0.389(7) mm/s and 0.258 (10) mm/s, 37.0(2.5)%, 0.235(3) mm/s, 0.706(7) mm/s. The fact that the Mössbauer

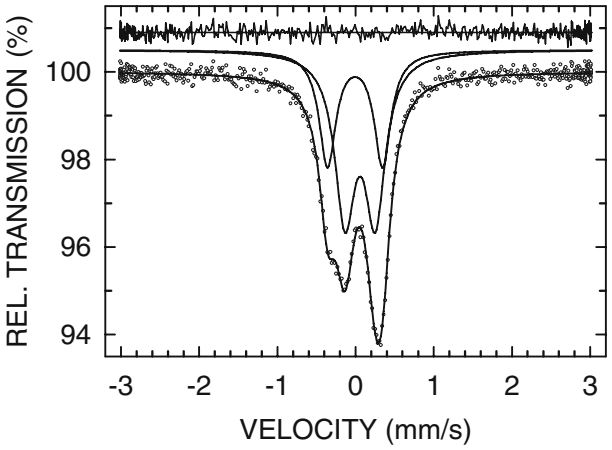

Figure 1 The ^{57}Fe Mössbauer spectrum of the decagonal $Al_{65}Co_{15}Cu_{19.9}Fe_{0.1}$ quasicrystal at 5.0 K fitted (*solid curve*) with two symmetric quadrupole doublets which are shown above the spectrum. The zero of the velocity scale is relative to the ^{57}Co (Rh) source at 5.0 K. The residuals are shown above the spectrum.

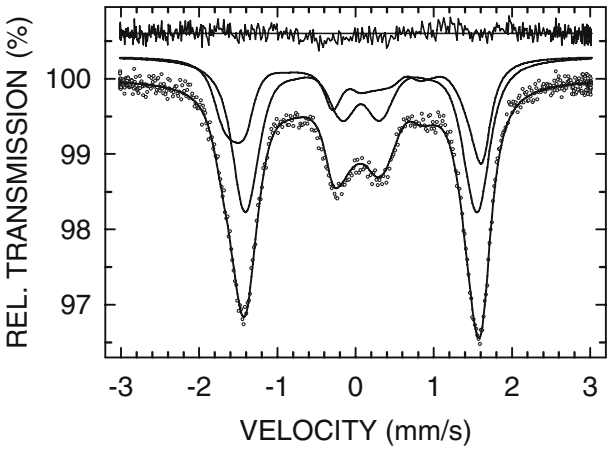

Figure 2 The ^{57}Fe Mössbauer spectrum of the decagonal $Al_{65}Co_{15}Cu_{19.9}Fe_{0.1}$ quasicrystal at 5.0 K in an external magnetic field of 9.0 T fitted (*solid line*) with two components which are shown above the spectrum. The zero velocity scale is relative to the ^{57}Co(Rh) source at 5.0 K. The residuals are shown above the spectrum.

spectrum of the decagonal $Al_{65}Co_{15}Cu_{19.9}Fe_{0.1}$ QC is indisputably composed of two quadrupole doublets (Figure 1) proves the existence of two distinct iron sites in this QC.

The fit of the zero-field Mössbauer spectrum in Figure 1 gives information on the magnitude of Δ, but not on the sign of the main component of the EFG, V_{zz}, or the value of η. The complete information of the sign of V_{zz} and the value of η can be obtained from the Mössbauer spectra measured in external magnetic fields such that the magnetic dipole interaction becomes of a similar magnitude as the electric quadrupole interaction. To determine the sign of V_{zz} and the value of η in the studied QC, a Mössbauer spectrum was measured in an external magnetic field of 9.0 T (Figure 2). The Mössbauer spectra exhibiting mixed hyperfine magnetic dipole and electric quadrupole interactions must be treated using the exact Hamiltonian. If texture effects are negligible one can assume, similarly to the case of powder samples, that the principal axes of the EFG tensor are randomly oriented with respect to the external magnetic field. The algorithm for calculating the spectra in such a case was given by Blaes et al. [9] and was used to fit the spectrum in Figure 2. As there are two classes of iron sites (Figure 1), it is clear that there are four possible combinations of signs for $q = \frac{1}{2}eQV_{zz}$: (+,+), (+,−), (−,−), and (−,+). The Mössbauer spectrum in Figure 2 was fitted with two components corresponding to these

four combinations of q signs; the value of Γ of two component subspectra was taken from the zero-field fit (Figure 1) and was fixed in the fit. The best fit (χ^2=1.24) (Figure 2) was obtained for the following values of A, δ, q, η corresponding, respectively, to two iron sites: 59.5(2.7)%, 0.312(10) mm/s, −0.316(13) mm/s, 0.97(15) and 39.5(2.6)%, 0.236(12) mm/s, −0.658(15) mm/s, 0.00(17). Thus, V_{zz} is negative at two iron sites and has the value of $-1.90(10)\times10^{21}$ and $-3.95(12)\times10^{21}$ V/m^2. In converting from the measured q to V_{zz} we have used the value Q=16 fm^2, which is based on a systematic comparison of experimentally determined quadrupole splittings and calculated EFGs [10] and which has been confirmed by a nuclear shell-model calculations [11].

The analysis presented above enabled us to determine precisely the values of the EFG at the two classes of iron sites in the decagonal $Al_{65}Co_{15}Cu_{19.9}Fe_{0.1}$ QC. What is now required are ab initio calculations of the EFGs for the Al-Cu-Co QC for several available structural models of this QC. Comparing these calculated EFGs with the experimentally determined EFG here, similarly as has been done for the icosahedral Al-Cu-Fe QC [6], could lead to the solution of the structure of the decagonal Al–Co–Cu QC.

4 Conclusions

The ^{57}Fe Mössbauer effect study at 5.0 K and in an external magnetic field of 9.0 T on a high-quality stable decagonal quasicrystal $Al_{65}Co_{15}Cu_{19.9}Fe_{0.1}$ has been presented. The iron atoms are shown to be located in two distinct classes of sites. The values of the principal component of the electric field gradient tensor and the asymmetry parameter at these sites are determined to be, respectively, $-1.90(10)\times10^{21}$ V/m^2, 0.97(15) and $-3.95(12)\times10^{21}$ V/m^2, 0.00(17).

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References

1. Stadnik, Z.M. (ed.): Physical properties of quasicrystals. Springer, Berlin Heidelberg New York (1999)
2. Steurer, W.: J. Non-Cryst. Solids **334–335**, 137 (2004)
3. Steurer, W., Kuo, K.H.: Acta Cryst. **B46**, 703 (1990)
4. Steurer, W., Kuo, K.H.: Philos. Mag. Lett. **62**, 175 (1990)
5. Frey, F., Steurer, W.: J. Non-Cryst. Solids **153−154**, 600 (1993)
6. Zijlstra, E.S., Kortus, J., Krajčí, M., Stadnik, Z.M., Bose S.K.: Phys. Rev., B **69**, 094206 (2004)
7. Stadnik, Z.M., Zhang, G.: J. Phys. Condens. Matter **17**, 6599 (2005)
8. Stadnik, Z.M. In Long, G.J., Grandjean, F. (eds.): Mössbauer Spectroscopy Applied to Magnetism and Materials Science, vol. 2, p. 125. Plenum, New York (1996)
9. Blaes, N., Fischer, H., Gonser, U.: Nucl. Instrum. Methods Phys. Res. B **9**, 201 (1985)
10. Dufek, P., Blaha, P., Schwarz, K.: Phys. Rev. Lett. **75**, 3545 (1995)
11. Martínez-Pinedo, G., Schwerdtfeger, P., Caurier, E., Langanke, K. Nazarewicz, W., Söhnel, T.: Phys. Rev. Lett. **87**, 062701 (2001)