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To cite this version:

M. Quilichini, B. Hennion, G. Heger, S. Lefebvre, A. Quivy. Inelastic neutron scattering study of icosahedral AlFeCu quasicrystal. Journal de Physique II, 1992, 2 (2), pp.125-130. 10.1051/jp2:1992118 . jpa-00247618

HAL Id: jpa-00247618

https://hal.science/jpa-00247618v1

Submitted on 4 Feb 2008

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Classification
Physics Abstracts
61.42 — 63.20

Short Communication

Inelastic neutron scattering study of icosahedral AlFeCu quasicrystal

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(Received 6 November 1991, accepted in final form 13 December 1991)

Résumé . — Les propriétés dynamiques des structures quasipériodiques sont complexes et pas encore complètement comprises. Pour les quasicristaux on ne possède que peu d'études dynamiques tant du point de vue théorique qu'expérimental. Dans cette lettre nous présentons des nouveaux résultats obtenus par diffusion inélastique de neutrons avec un quasicristal monodomaine de $Al_{63}Cu_{25}Fe_{12}$ que nous avions déjà étudié [1]. Dans la partie 1 nous rappelons quelques propriétés spécifiques des structures quasipériodiques et nous résumons brièvement les travaux théoriques qui nous permettent une interprétation qualitative des données expérimentales présentées dans la partie 2 et discutées dans la partie 3.

Abstract . — Dynamical properties of quasiperiodic structures are rather tricky and far from being understood. For quasicrystals only little information is available both theoretically and experimentally. In this paper we present new experimental results obtained by inelastic neutron scattering on a monodomain quasicrystal of $Al_{63}Cu_{25}Fe_{12}$ already investigated in a previous study [1]. In section 1 we recall the basic features of the quasiperiodic structures and briefly review theoretical works on the dynamics of quasicrystals which can be of some help to appreciate the experimental data presented in section 2 and discussed in section 3.

1. Introduction.

Since now three decades solid state physicists have observed that long range order may exist without translational symmetry. This has been first encountered in incommensurate (IC) crystals, including many systems such as alloys, ferroelectric insulators, 1D conductors. The diffraction patterns of these systems consist of sharp Bragg peaks. These cannot be indexed on the basis of a conventional three dimensional lattice. However, they can be indexed by a

set of n ($n > 3$) integer indices, and therefore the wave vectors of the Fourier transform read as

$$\mathbf{k} = \sum_{i=1}^{n} h_i \, \mathbf{a}_i^* \tag{1}$$

(h_i integer) and the structure is quasiperiodic [2].

By introducing the superspace formalism de Wolff, Janner and Janssen [3] gave a useful description of IC crystals and showed that periodicity is restored in an n dimensional space.

Furthermore they have shown that among IC crystals, modulated phases form an important class for which one can define a basic (average) structure with a 3D space group. It has been seen then [4] that excitations of IC modulated structures may be labelled by a wave vector $\mathbf{k}$ inside the Brillouin zone of the basic space group. However the frequency function is not continuous but shows many gaps of which only few have a substantial magnitude and occur at low multiples of $\frac{1}{2}\,\mathbf{q}$ (where $\mathbf{q}$ is the wave vector of the modulation).

The basic structure usually exists in a certain range of temperatures and pressures above the IC modulated phase. A phenomenological approach and group theory analysis are sufficient to give an interpretation of the transition from the basic structure to the IC phase. For the long wavelength limit they allow one to define the amplitudon and the phason which are specific excitations of the IC state. To go further requires attention to discreteness effects and to the microscopic origin of phenomenological parameters.

The so called quasicrystals have been found as a new class of alloys first in 1984 [5] and exhibit long range order too. Their diffraction pattern reveals a point group of icosahedral symmetry which is incompatible with the existence of a set of main reflections. Although significant progress has been made concerning their structural and static properties, so far the understanding of their dynamical properties has been rather limited. The lack of translational symmetry hampers the analysis of their vibrational state. Even though periodicity is restored in 6 dimensions this does not allow a finite number of Bloch type solutions, because in superspace atomic surfaces contain an infinite number of point atoms [6].

Since 1986 several theoretical studies have investigated (numerically and analytically) vibrational modes properties of 1D and 2D quasiperiodic lattices. These works mainly concentrate on the density of states. Only few among them deal with properties of propagating modes and how well defined they are.

The simplest model of a 1D quasilattice is the Fibonacci chain (F.C.) which is generated by a deterministic process based on two basic parameters L and S with $L/S = \tau$ (τ golden ratio). This infinite sequence has a perfect long range order. Its Fourier transform consists of Bragg peaks which form a discrete dense set where intense peaks are separated. It has been demonstrated that [7] the weak peaks correspond to wave vectors with a large $q_\perp$ component (if $\hat{\mathbf{q}}_{/\!/}$ and $\hat{\mathbf{q}}_\perp$ are unit vectors in the Fourier space such that $\hat{\mathbf{q}}_{/\!/}$ is along the direction dual to the direction of the F.C. and $\hat{\mathbf{q}}_\perp$ is perpendicular to it).

Lu *et al.* [8] have studied analytically and numerically the dynamical behavior of this F.C.. They have shown that long wavelength modes propagate with sound velocity and that the wave functions are extended, whereas in higher frequency region they observed that the spectrum is self similar and "Cantor like" and therefore possesses many gaps. This gap structure (obtained by a technique which combines the continued-fraction expansion method and the decimation method) is shown [9] to be intimately related to the diffraction pattern. Also the gap width is directly proportional to the diffraction amplitude at the corresponding wave vector. These results agree with those of a renormalization group analysis by Kohmoto *et al.* [10] which have shown that the gaps are distributed densely and that a normal mode with a frequency ω is self similar and therefore neither a localized nor an extended state. Odagaki and Nguyen [11]

have considered the 2D case of a Penrose tiling ; their calculations have shown that gaps are present in the eigenfrequency functions, and that one observes Van Hove singularities.

More specifically related with our experimental work are the papers by Patel *et al.* [12], Benoit *et al.* [13], and Ashraff *et al.* [14], who have studied the dynamical response $S(\mathbf{Q},\omega)$ of a special model. $S(\mathbf{Q},\omega)$ is a quantity which can be obtained from neutron inelastic scattering measurements and for a one phonon process in crystals it reads as :

$$S(\mathbf{Q},\omega) = \sum_{j}^{\text{all modes}} S_j\ (\mathbf{Q},\omega) \tag{2}$$

and for one mode.

$$S_j(\mathbf{Q},\omega) = |G_j(\mathbf{q},\ \mathbf{Q})|^2\ F_j(\omega,\omega_j(\mathbf{q}),\ T) \tag{3}$$

where $\hbar\mathbf{Q}$ and $\hbar\omega$ are the momentum and energy transferred by a neutron to the sample. G_j is the structure factor and contains the eigenvector which describes the pattern of atomic displacements in one unit cell for the mode j, while F_j describes the inelasticity of the process (dispersion and damping).

In reference [12] the model consists of ferromagnetically coupled spins situated on the vertices of a 2D Penrose tiling. $S(\mathbf{Q},\omega)$ is computed for $\mathbf{Q}$ close to the strongest Bragg peaks of the static structure factor. The authors demonstrated that in the long wavelength limit propagating modes (spin waves, but potentially acoustic phonons) do exist with the same dispersion relation around every Bragg peak studied.

In reference [14] computations (by spectral moment methods) for a linear Fibonacci chain yielded pseudo dispersion curves near strong Bragg peaks and it has been shown that pseudo Brillouin zones can be defined for these stronger peaks.

The calculated and analytical results summarized above are quite encouraging when compared to the few measurements obtained up to now on the two quasicrystals $Al_{63}Cu_{25}Fe_{12}$ [1] and $Al_{60.3}Li_{29.2}Cu_{10\ 5}$ [15].

2. Experimental.

We have performed extended measurements on an almost perfect monodomain quasicrystal of $Al_{63}Cu_{25}Fe_{12}$ (7 mm^3) already described in a first paper [1]. The previous measurements have been limited by the weakness of scattered intensity, therefore we heated the sample to 400°C to increase the thermal population while staying in a temperature range where no damage was risked. For this purpose the sample was mounted in a furnace and oriented with a scattering plane defined by two orthogonal two fold axes and containing 3-fold and 5-fold axes (see Fig. 1 in Ref. [1]).

The measurements were performed on the 1T three-axis spectrometer with thermal neutrons at the Orphée reactor, (LLB-CEN-Saclay, France). All energy scans (in neutron energy loss mode) were obtained with a fixed outgoing neutron wavevector of $k_F = 3.85$ Å^{-1} or 4.25 Å^{-1} Pyrolytic graphite (PG [002]) was used as monochromator (vertically bent) as well as analyser. The instrumental resolution was in the range of 0.7 - 0.95 THz.

As expected at 400°C we obtained a better signal-to-background ratio. Figure 1 shows the response obtained from transverse acoustic modes with $q = 1.2$ (0.44 Å^{-1}) (q normal to 2-fold and 5-fold axis respectively) which can be compared to figure 2 of reference [1].

In the earlier experiment at room temperature we have demonstrated that there exists only one unique transverse acoustic branch and one unique longitudinal acoustic branch with

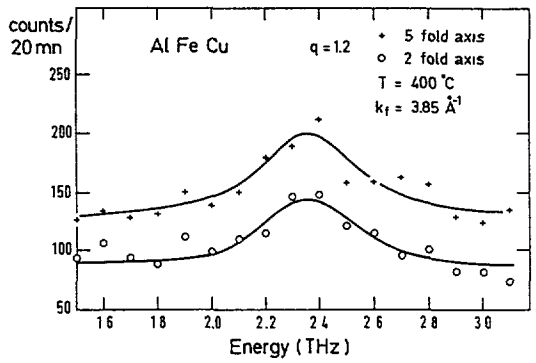

Fig. 1. — Energy runs for TA modes (solid lines are results of fit with a DHO response function). The symbols refer to the two directions of the wave vector **q** ((+) **q** ⊥ 5-fold-axis, (o) **q** ⊥ 2-fold-axis).

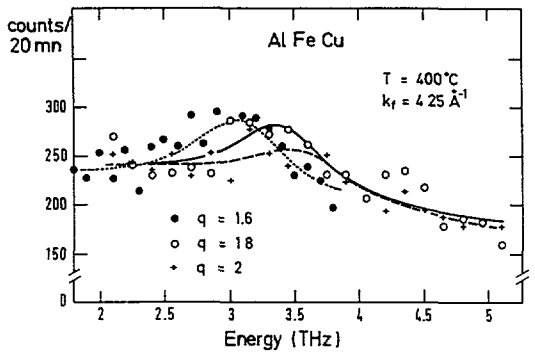

Fig. 2. — Energy runs for the TA mode with **q** normal to the 5-fold axis (solid lines are results of fits with a DHO response function).

isotropic slopes, but our measurements were limited to energies of 2.5 THz. In the present experiment the signal to background ratio allowed a reliable investigation up to 3.8 THz as shown in figure 2. Still no optic mode has been detected.

Systematic data analysis accounting for the convolution of the instrumental resolution with the observed linear dispersion were undertaken. In the fitting procedure, the phonon response function F_j in $S_j(Q,\omega)$ is described by the response function of a damped harmonic oscillator (DHO)

$$F_j(\omega,\omega_j(q),T) = \frac{\omega}{1-\exp(-\hbar\omega/kT)}\frac{\Gamma_j(q)}{\left(\omega^2-\omega_j^2(q)\right)^2+\omega^2\Gamma_j^2(q)} \tag{4}$$

where the first factor is the detailed balance factor, $\omega_j(q)$ the quasiharmonic frequency of the mode j and Γ_j its damping constant.

Main results are reported in figure 3. Data points have been obtained for **q** normal to the 2-fold axis near the (4/6 0/0 0/0) Bragg peak and for **q** normal to the 5-fold axis near the (2/4 4/6 0/0) Bragg peak.

For q smaller than 0.45 Å^{-1} we have checked that at 400°C we always have a unique

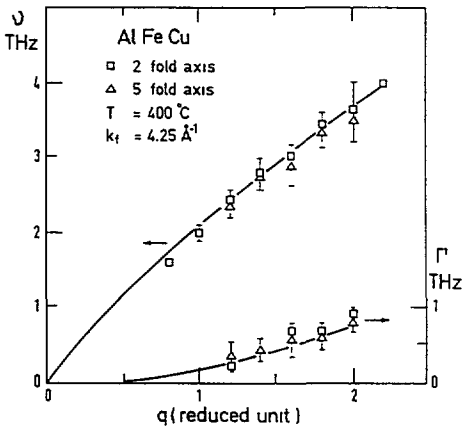

Fig. 3. — Dispersion curve $\nu(q)$ for the unique TA mode; dependence with q of the width Γ of this mode; q (reduced unit) $= q\left(\text{Å}^{-1}\right)\frac{a}{2\pi}$ ($a = 17.09$ Å at 400 °C).

transverse acoustic branch having the same slope of 5.5 THz Å^{-1} as at room temperature (earlier experimental points are not given on the curve) and that the modes have no intrinsic width. For q greater than 0.45 Å^{-1} a careful analysis indicates that the branch remains unique within experimental error bars but its slope decreases indicating a bending of the dispersion curve, while the width of the phonon (Γ) increases. Note that along **q** normal to the 2-fold axis we have, at $q = 0.457$ Å^{-1} and 0.74 Å^{-1} respectively two Bragg peaks having a very weak intensity. The TA modes which are associated to them are not observable which is in agreement with the calculations by Benoit *et al.* [13]. Finally for q values greater than 0.78 Å^{-1} the response signal is difficult to analyze because it is weak and it is spread over a relatively wide frequency range.

3. Discussion - conclusions.

The results presented earlier in [1] and here are reminiscent of the lattice dynamics of a crystal. In the long wavelength limit well defined acoustic modes are observed and we have shown that the slope of the TA and LA dispersion curves is isotropic. This is in good agreement with the high symmetry of the icosahedral point group. Furthermore, the long range ordering of the system evidenced by the existence of narrow Bragg peaks fits well with the fact that the observed vibrational modes have no intrinsic width.

For smaller wavelength values (larger q) the vibrations are more sensitive to the microscopic details of the structure. In this q range the inelastic signal loses its intensity while it is spread over an appreciable energy range. This has been accounted for in our analysis by introducing an intrinsic phonon linewidth Γ and a decrease of the slope of the dispersion curve. The physical origin of the linewidth is not well understood. It is certainly related to the fact that the eigenstates are neither collective nor localized modes. The decrease of the integrated intensity and the increase of the linewidth could indicate the possible existence of energy gaps in the dispersion curves. These gaps have to be small because there are no strong Bragg peaks in this Q region of the scattering plane and we know from the study by Lu *et al.* [9] that the gap width

is directly proportional to the diffraction amplitude at the corresponding wave-vector. They are therefore difficult to observed experimentally within the energy resolution of our setting. However they could manifest themselves *via* a broadening of the experimental response.

In conclusion, let us compare our results to those obtained by Goldman *et al.* [15] in a very similar experiment on $Al_{60.3}Li_{29.2}Cu_{10.5}$. The latter work and the neutron diffraction study of our sample [16] make clear that the distribution of strong Bragg peaks is quite different for AlFeCu and AlLiCu samples. In the AlLiCu compound it has been shown, as for the AlFeCu sample presented here, that the slope of the dispersion curves of the transverse and longitudinal acoustic modes are isotropic. As shown in figure 3 of reference [15] a nice Brillouin zone like behavior for a longitudinal acoustic mode which propagates along the 5-fold axis between two Bragg peaks of the scattering plane exists. This experimental result supports one of the main conclusions of the study by Benoit *et al.* [13] quoted in the introduction. Finally, one optical mode has been found along the 2-fold direction [15] at $\simeq$ 2.5 THz. This might indicate a principal difference between the two systems studied up to now.

Future measurements at higher energies have to await the synthesis of larger mono-domain samples.

Acknowledgements.

We wish to thank P. Boutrouille for his kind, constant and efficient technical assistance and C. Pomeau for her kind help in preparing this typescript.

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