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Short Communication

Inelastic neutron scattering study of acoustic modes in a monodomain AlCuFe quasicrystal

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Abstract. — Inelastic neutron scattering results evidenced for the first time that acoustic phonon modes can be observed for a quasicrystal. The triple-axes experiments have been performed on a small monodomain $Al_{63}Cu_{25}Fe_{12}$ sample. The obtained TA and LA dispersion curves near to strong Bragg peaks do not show any variation with respect to different **Q** values. Their slopes are isotropic and do not depend on special symmetry directions; they are comparable to those of corresponding curves for pure aluminium. We have not been able to observe energy gaps in our measurements.

Introduction.

A stable icosahedral phase in the AlCuFe ternary alloys was obtained by slow solidification by Tsai *et al.* [1,2] . Ebalard and Spaepen [3] have identified the quasilattice with a face centered 6D-hypercubic lattice and the structure has been described by Devaud-Rzepski *et al.* [4] as a F superstructure with ordered domains and antiphase boundaries. Depending upon the preparation conditions and the thermal history of these AlCuFe alloys, it has been observed, even at room temperature, either a perfect quasicrystalline state without any indication of phason strain or a microcrystalline twin state with an overall pseudo-icosahedral symmetry [5-7].

In this paper we show for the first time phonon dispersion curves for a monodomain quasicrystal of $Al_{63}Cu_{25}Fe_{12}$. Dynamical properties of rapidly quenched as well as slowly cooled and annealed icosahedral $Al_{65}Cu_{20}Fe_{15}$ have been already studied by inelastic neutron time-of-flight experiments [8] . The obtained generalized vibrational densities of states show only very few structure and the low energy modes are related to atomic disorder introduced by rapid quench.

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In the large class of quasiperiodic structures, incommensurate modulated phases have been the most studied (both experimentally and theoretically) as regards to their dynamical properties. For quasiperiodic systems it is generally expected to observe acoustic phonon branches with the usual linear slopes. In an incommensurate modulated crystal where one can define a basic (or average) lattice and an associated Brillouin zone, the modulation and its harmonics lead to an infinite number of wave vectors within the Brillouin zone, and therefore an infinite number of energy gaps in the frequency dispersion relation is expected [9, 10]. In fact, only those which correspond to the **q** vectors of the modulation itself and its lowest order harmonics give experimentally detectable effects, similar to what is usually observed at the Brillouin zone boundary of a normal crystal.

In a quasicrystal no average lattice can be defined. Nevertheless acoustic modes should be observed around the Bragg reflections, and their intensity should fulfill the usual relationship which exists for crystals between the Bragg intensity and the acoustic mode intensity. Furthermore, because of the dense set of Bragg peaks, one expects energy gaps in the frequency dispersion of the acoustic phonons.

As an example of a 1D quasiperiodic system, a Fibonacci chain was treated in numerical model calculations by Benoit *et al.* [11] in order to show the existence of pseudoacoustic dispersion curves. They have pointed out that it is possible to define for stronger Bragg peaks pseudo Brillouin zones analogous to those of normal crystals.

Experimental conditions.

The alloy of composition $Al_{63}Cu_{25}Fe_{12}$ was prepared from the pure elements($Al_{99.99}Cu_{99.99}$ and $Fe_{99.95}$) by levitation melting in a helium atmosphere, followed by a quench from 1150° C by planar flow casting, as is described in [5]; part of the brittle flakes were annealed during many days in an alumina crucible under vacuum, at 860° C, just below the peritectic transformation and then slowly cooled. The quasicrystals grown from the melt are surrounded by small grains of slightly different composition.

The sample used in our inelastic neutron study consisted of a 2 mm diameter monodomain quasicrystal of dodecahedral shape with pentagonal faces. It was characterised by a complete neutron diffraction study on a four-circle instrument. A set of symmetry related Bragg reflections was investigated with respect to their precise orientations and intensities in order to check the icosahedral symmetry, and no deviation was observed. The high perfection of the sample was ascertained by studying the reflection profiles which correspond to almost the instrumental resolution obtained by measurements on an ideal germanium single crystal. These results imply a perfect quasicrystal without any phason strain, especially when comparing them to those obtained on monodomain icosahedral AlLiCu samples where the best ones show a "mosaïcity" of about 0.8° and a strong and typical dependence of the reflection profiles on $Q_\perp$, which is completely absent for the AlCuFe sample. A data collection of the intensities of all Bragg peaks up to $Q = 11.6$ Å^{-1} has been achieved to allow a complete structure analysis of the AlCuFe icosahedral phase. First results have been published elsewhere [12]. Additional reflections, observed for the microcrystalline state of $Al_{63.5}Cu_{24}Fe_{12.5}$ by X-ray precession photographs [6] were checked, but no intensity could be detected. Finally, X-ray measurements on powdered single crystals of identical preparation show reflections of the F icosahedral phase, only. Even with the good resolution of this synchroton experiment no linewidth is observed [5].

Inelastic results . Discussion . Conclusion.

Inelastic neutron scattering measurements have been carried out on the three axis spectrometers 4Fl and IT, respectively on cold and thermal sources at the Orphée reactor in Saclay. In order to measure phonon dispersion curves for the main symmetry directions of the icosahedral phase, the sample was oriented with a scattering plane defined by two orthogonal two-fold axes. This plane also contains a three fold and a five fold axis, and its schematic diagram is shown in figure 1.

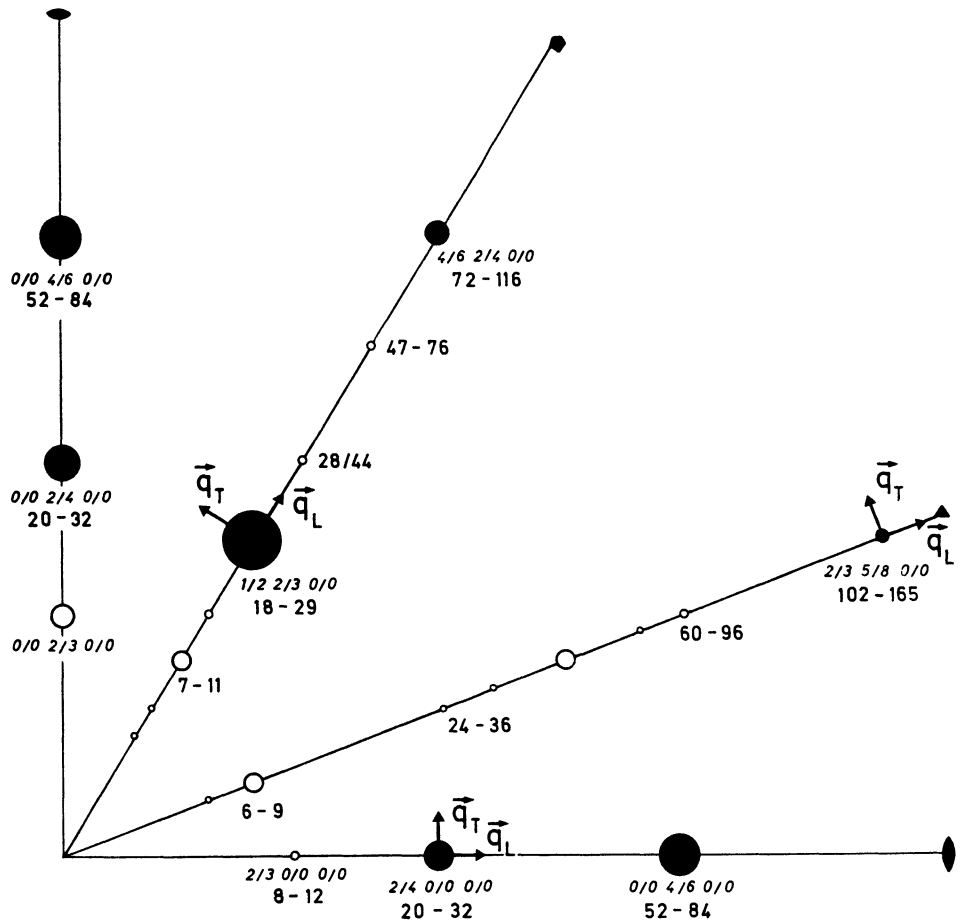


Fig. 1. — Scattering phase: circles locate Bragg reflections. Dark large circles indicate high intensity peaks around which acoustic modes were looked for. In calculating irrational pseudo indices H,K,L with $H = h+h'\tau$, $K = k+k'\tau$, $L = \ell+\ell'\tau$ (τ: golden mean), we took a pseudocubic lattice parameter $a = 16.944$ Å. Here Bragg peaks are labelled following the indexing method of icosahedral quasiperiodic crystals by Cahn *et al.* [13]. Arrows indicate typical phonon scans.

Intense Bragg reflections defined by a vector Q_B (Å^{-1}) have been selected on the different symmetry axes, and inelastic measurements have been performed with a propagation **q** vector parallel

or normal to symmetry axes around Bragg positions which have different Q_B values.

On both spectrometers pyrolytic graphite (PG[002]) has been used as monochromator (vertically bent) and analyser. On 4F1 spectrometer constant $k_I = 2.662$ Å^{-1} scans allowed us to measure phonons in neutron energy gain, with a horizontally bent analyser (energy resolution: $\simeq$0.23 THz, FWHM). On IT spectrometer constant $k_F = 2.662$, 3.85 or 4.25 Å^{-1} scans were used to measure phonons in neutron energy loss with a flat analyser.

Phonon modes have been detected in both, transverse and longitudinal, configurations and found to follow the usual linear dispersion. Systematic data analysis, accounting for the convolution of the instrumental resolution with the observed linear dispersion indicates that these phonon modes have no intrinsic linewidths within experimental accuracy. An example of typical neutron groups is shown in figure 2.

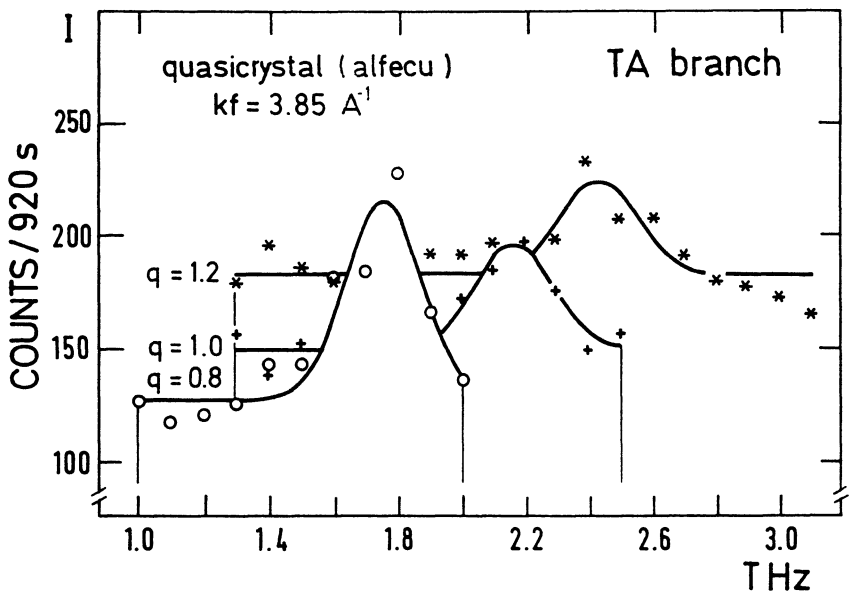
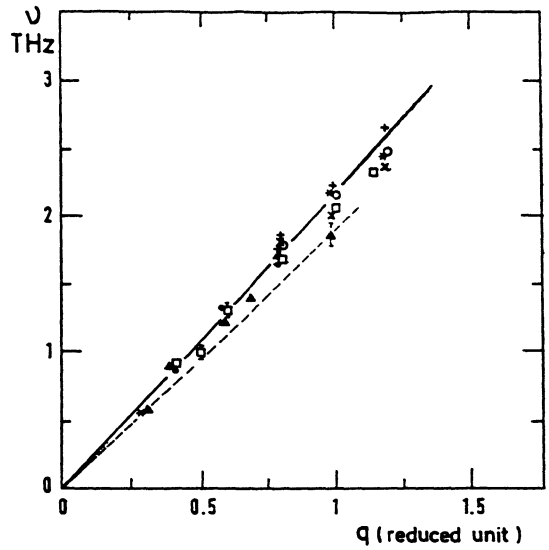


Fig. 2. — Typical neutron groups for the transverse acoustic mode emanating from (0/0 4/6 0/0). Experimental points are shown along with the fitted responses (full lines).

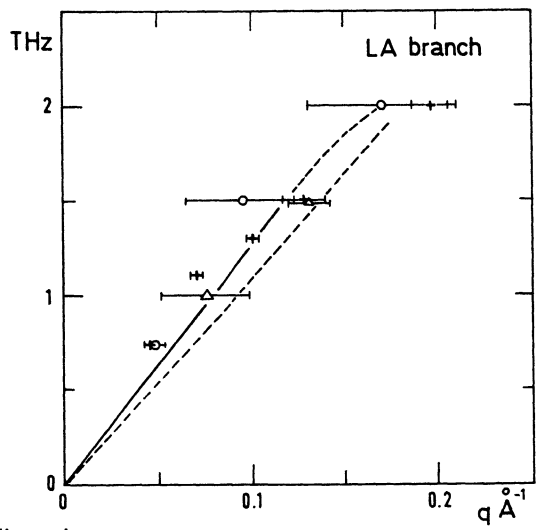
According to the signal to background ratio we were not able to ascertain any phonon peaks at energies higher than about 2.5 THz. The slopes of the linear dispersion curves of transverse acoustic (TA) and longitudinal acoustic (LA) branches are independent of the direction of propagation and of the Q_B values. This is clearly seen in figure 3 where the fitted measured values are reported (together with the slopes for pure aluminium).

We wish to emphasize that the independence of the results with respect to Q_B values supports the concept of the existence of a pseudo Brillouin zone introduced by Benoit *et al.* [11] for a Fibonacci chain, and that the isotropy of the slopes of phonon branches (LA and TA modes) for diffferent symmetry directions is not very surprising, due to icosahedral point group symmetry. But since this first experiment was performed on a very small sample ($\simeq$ 7 mm^3), a small anisotropy of the slopes may have not been detected.

We have found that we could follow all the studied TA modes up to 0.44 Å^{-1} with no significant linewidth. This corresponds to the propagation of plane waves with wavelength of about 14 Å



(a)



(b)

Fig. 3. — a) Unique TA dispersion curve.

- • 2/4 0/0 0/0 } $Q_B = 3.14$ Å^{-1}
- ▲0/0 2/4 0/0 } 4F1 Results
- ■ 1/2 2/3 0/0 $Q_B = 2.988$ Å^{-1}
- * 0/0 4/6 0/0 } $Q_B = 5.975$ Å^{-1}
- ∘ 4/6 0/0 0/0 }
- △0/0 2/4 0/0
- × 4/6 2/4 0/0 } 1T Results
- +2/3 5/8 0/0 $Q_B = 7.122$ Å^{-1}
- □ 1/2 2/3 0/90

q (reduced unit) $= q\left(\text{Å}^{-1}\right)\dfrac{a}{2\pi}$

b) Unique LA dispersion curves : ∘ 2/3 5/8 0/0; + 0/0 4.6 0/0; △ 4/6 2.4 0/0. Full lines are guides to the eye. Dash lines give the sound velocities of aluminium [14].

in real space without strong scattering. We have not found any evidence for energy gap in our measurements.

Furthermore, the comparison between results obtained in very different experimental conditions on the two spectrometers favors the assumption that the lack of observation in the higher energy range is due to experimental limitations rather than to physical reasons. as described above, this first experiment on a monodomain sample does show that at leat acoustic modes exist in a quasicrystal, in agreement with the quasiperiodicity of the lattice; but larger high quality samples are needed to obtain more complete data, especially when one wants to observe energy gaps and optic modes.

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