

ULTRASONIC ATTENUATION IN QUASICRYSTALLINE AlPdMn STUDIED BY PICOSECOND ACOUSTICS

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Abstract

We report on the acoustic attenuation in an AlPdMn icosahedral quasicrystal between 15 K and 300 K, in the tens of GHz range. High frequency coherent longitudinal acoustic waves are excited and detected by an optical "pump and probe" technique, in an interferometric configuration. Our results reveal that, at high temperature ($T > 100$ K), the acoustic attenuation is rather small, close to the attenuation in crystals and much smaller than in disordered solids. We stress that Akhieser processes (phonon-phonon interactions) must be active in quasicrystals and give the correct behavior and order of magnitude of the acoustic attenuation, at high temperature. At low temperature, departure from the standard behavior arising from phonon-phonon interaction is not understood.

Introduction

The lattice dynamics of quasicrystals is a challenging problem in solid state physics. Quasicrystals exhibit a long range order which can be described using periodic structures in a higher dimensional space. Nevertheless, no powerful concept, equivalent to the Bloch theorem in crystals can be used to describe the vibrational excitations. Much theoretical and experimental work has been devoted to this problem [1] and the general agreement is that the lattices modes in 3D icosahedral solids are, so called, critical. However low frequency modes cannot practically be distinguished from extended modes and are similar to the acoustic modes of crystals. Inelastic neutron and X-ray scattering experiments set the limit between high and low frequencies around a few THz. Because of the unusual character of the lattice excitations, it is worthwhile investigating physical properties which are intimately related to them, such as thermal or acoustic properties. This paper reports on the acoustic attenuation in a single i-AlPdMn quasicrystal between 15 K and 300 K, in the tens of GHz range.

Experimental procedure

The sample is a $118 \mu\text{m}$ plate cut from a single icosahedral quasicrystalline domain $\text{Al}_{68.2}\text{Pd}_{22.8}\text{Mn}_{9.0}$ grown using the Czochralsky technique. The faces are normal to a twofold axis and are polished flat and parallel. The wedge angle and surface roughness were esti-

mated to be about 4×10^{-4} rd and 4 nm rms, respectively. For comparison, we also studied two Si[100] crystalline samples with low impurity content (phosphorus dopant). The samples are 110 and $198 \mu\text{m}$ thick with resistivity ρ : $165 < \rho < 240$ and $500 < \rho < 1000$ $\Omega\cdot\text{cm}$, respectively. The experiments are performed in a continuous-flow cryostat.

We performed the sound absorption measurements using a picosecond ultrasonics technique [2]. A Ti:sapphire mode-locked laser delivers linearly polarized pulses with a duration of 200 fs. The pump pulse is focused to a diameter of $18 \mu\text{m}$ on a thin (30 nm) aluminium film deposited on the sample. The film absorbs a fraction of the energy and generates a longitudinal acoustic pulse which propagates in the sample and gives rise to a series of echoes which are detected by a second, time delayed, probe pulse. The probe is focused to a diameter of $24 \mu\text{m}$. The pump and probe wavelength is 750 nm. The time delay T between two successive pulses of our laser source is typically around 12.2 ns which means that for a delay t between the probe and the pump we can detect any event occurring at times $t + nT$ (n is an integer), providing the laser has a good stability [3]. $n = 3$ or 6 in our experiments. Acoustic echoes induce a change of reflection coefficient r . The relative change writes $\frac{\Delta r}{r}(t) = \rho_e(t) + i\phi(t)$. The photoelastic effect contributes to both real and imaginary parts while the surface motion contributes only to the imaginary part. Interferometric detection can measure independently ρ_e and ϕ [4]. At low frequency the displacement contribution to the reflectivity change becomes larger than the deformation contribution and consequently ϕ is expected to be larger than ρ_e . Since high frequencies (~ 100 GHz) are highly damped at room temperature we use a Sagnac interferometer, sketched in Fig 1 [5] to measure the imaginary part. After the polarizing beamsplitter (PBS), the probe beam is split by a non polarizing beamsplitter (NPBS) in two equal parts which propagate in the same loop along opposite directions. The sample is located within the loop in a non symmetrical position in such a way that the two beams reach the sample at different times t and $(t + \theta)$ compared to the pump beam; the delay θ between the two beams is 800 ps. Another asymmetry is introduced by the half- and quarter-wave plate: the half-wave $\lambda/2$ plate rotates the beam polarization by

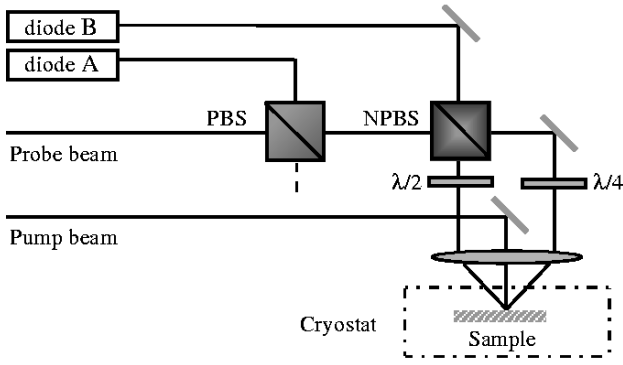


Figure 1: "Pump and probe" picosecond acoustics setup, in a Sagnac interferometric configuration.

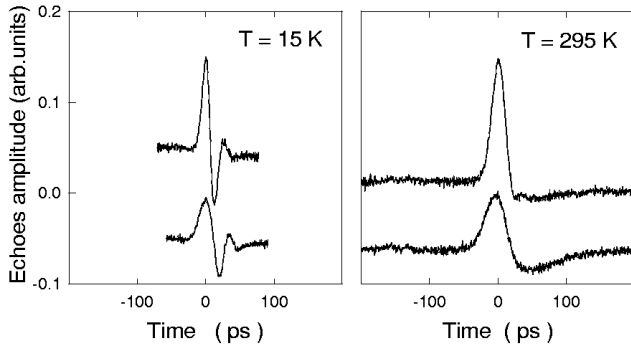


Figure 2: First (upper) and second (lower) acoustic echoes at 15 K and 295 K. For clarity, the echoes are arbitrarily translated along the time and amplitude axis.

90 degrees in such a way that the counter propagating beams in the loop pass through the quarter-wave plate $\lambda/4$ with a polarization either parallel to the ordinary or to the extraordinary optical axis of the plate. After the loop the two beams interfere and are detected by two photodiodes A and B. Due to the $\pi/2$ delay introduced by the quarter-wave plate between the beams propagating in opposite directions it can be shown, assuming the non polarizing beamsplitter is perfect, that:

$$S_A(t) - S_B(t) = \pm 2 [\phi(t + \theta) - \phi(t)] \quad (1)$$

where S_A and S_B are the photodiodes signals. The sign depends on the orientation of the quarter-wave plate. The pump is modulated at 1 MHz (duty factor: 20%) and the induced change $\Delta r/r$ is detected with a lock-in amplifier; the sensitivity in surface displacement detection is estimated to be 5×10^{-6} nm.

Due to the very low thermal conductivity of AlPdMn (0.1 to $1.5 \text{ W.m}^{-1}.\text{K}^{-1}$ from 10 to 300 K) and the small volume of the sample, special care must be payed to DC and AC heating problems.

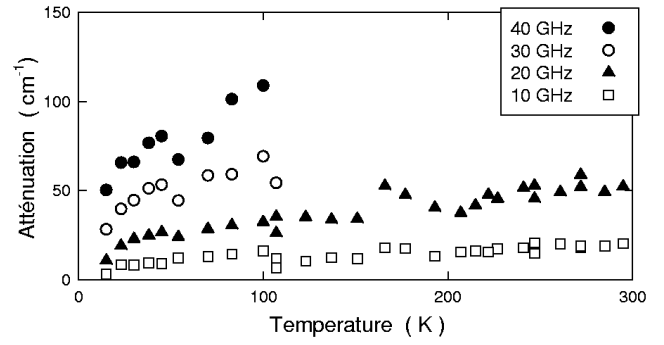


Figure 3: Attenuation of longitudinal waves versus temperature in $i\text{-Al}_{68.2}\text{Pd}_{22.8}\text{Mn}_{9.0}$.

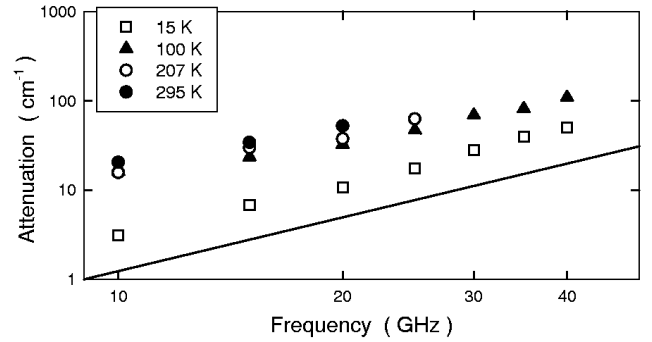


Figure 4: Attenuation of longitudinal waves versus frequency in $i\text{-Al}_{68.2}\text{Pd}_{22.8}\text{Mn}_{9.0}$. The full line displays a quadratic variation.

Results

Figure 2 displays the first and second acoustic echoes, at 15 K and 300 K. We checked that no nonlinear effect is induced. To perform a quantitative analysis, we compute the Fourier transform after subtraction of a linear background from the raw data. The acoustic attenuation is then computed from the ratio of the Fourier amplitude of the first and second echoes [6]. Figure 3 displays the attenuation versus temperature, at various frequencies, between 15 K and 295 K. The attenuation decreases linearly with temperature between 300 K and 30 K. It seems that below 30 K (at 20 GHz) the attenuation decrease is becoming steeper. Unfortunately we could not perform experiments below 15 K to test this point. Figure 4 displays the attenuation versus frequency, at various temperatures, between 10 GHz and 40 GHz. The variation of attenuation is roughly quadratic.

Discussion

Two sources of acoustic attenuation must be considered: intrinsic microscopic processes giving a true acoustic attenuation and extrinsic sources originating from macroscopic defects, giving an apparent attenu-

Table 1: Longitudinal wave attenuation in different solids, at 300 K. UP, BS and BUP stand for picosecond ultrasonics, Brillouin scattering and Brillouin scattering by picosecond ultrasonics, respectively.

	a-TiNi	a-SiO ₂		a-SiO ₂		a-As ₂ Se ₃	c-SiO ₂		Si	i-AlPdMn
		(film)		(bulk)					[100]	
α (cm ⁻¹)	600	3200	5600	1500	1000	6900	230	380	80	50
F (GHz)	30	30	40	35	30	30	40	35	20	20
Method	UP	UP	UP	BS	BUP	UP	BS	BS	UP	UP
Ref.	[7]	[6]	[6]	[8]	[9]	[2]	[10]	[11]	This work	This work

ation (poor parallelism, surface roughness...). The non-parallelism effect was computed and found to be negligible. In principle, the surface roughness contribution can be estimated by making measurements on a series of samples with identical surface condition and different thickness. We did not performed such experiments but we notice that the rms surface roughness (4 nm) is much smaller than the shortest investigated acoustic wavelength (160 nm at 40 GHz). All together, the effect of the different extrinsic sources can be assumed as temperature independent and their contributions can be estimated, at every frequency, as a residual attenuation at 0 K. Unfortunately, our experiments do not extend to sufficiently low temperature to make it possible to measure this residual attenuation. Nevertheless, we observe that the attenuation at 15 K is smaller than the attenuation at high temperature. At 20 GHz and above 100 K, ($\alpha(T) - \alpha(15\text{ K})$) is more than 70 % of the total attenuation $\alpha(T)$. Therefore, above 100 K, the main contribution to the attenuation comes from intrinsic processes. It is interesting to compare the attenuation in i-AlPdMn with the attenuation in other solids, at room temperature and in our frequency range. Table 1 gives various data at room temperature. For comparison, results at different frequencies can be scaled to 20 GHz assuming a quadratic variation of the attenuation versus frequency. At 300 K, the attenuation in i-AlPdMn is much smaller than in the amorphous metal or in the covalent glasses. On the contrary, it is quite close to the attenuation in crystalline Si or crystalline SiO₂. Moreover, we have previously noted that extrinsic processes could give a small contribution to the total attenuation. Then the actual intrinsic attenuation in i-AlPdMn could still be slightly smaller than measured. Therefore, from the point of view of the acoustic attenuation, i-AlPdMn compares to dielectric crystalline solids, not to disordered solids, at room temperature. This suggest a common origin of the sound absorption process.

In crystalline solids, it is well known that the acoustic waves decay because of their interaction with the thermal phonons [12], [13]. At high temperature, the absorption can be calculated thanks to the Boltzmann

equation, in the collision time approximation [13]. Two contributions are found: the Akhieser and the thermoelastic contributions. The thermoelastic contribution is usually negligible and will not be considered here. In the Akhieser mechanism, the acoustic wave acts as an external field which modulates the energies of the thermal lattice excitations through the Grüneisen parameters. These excitations readjust their energy distribution via anharmonic interactions on the time scale of the collision time and this delay gives rise to sound absorption. In the derivation of the Akhieser contribution, the character of the thermal lattice excitations does not play any part. Thus we stress that this mechanism must be active, at room temperature, whatever the true nature of the lattice thermal excitations is (extended, localized, critical...) and that the expression derived for crystals is also valid in quasicrystals. Then, the Akhieser contribution to the absorption α_A writes, if $\omega\tau \ll 1$:

$$\alpha_A = \frac{CT\omega^2\Gamma^2\tau}{2\rho s^3} \quad (2)$$

where C is the specific heat per unit volume, T the temperature, ρ the mass density, τ the mean lifetime of the thermal excitations. $\omega/2\pi$ and s are the frequency and velocity of the sound wave. Γ is related to the Grüneisen parameters through thermal averages.

We notice that the experimental results are in accordance with the quadratic frequency variation displayed by Eq.2. Using $C=2.7 \times 10^6 \text{ J.m}^{-3}.\text{K}^{-1}$ [14], $\rho=5130 \text{ kg.m}^{-3}$, $s=6500 \text{ m.s}^{-1}$, $\alpha=50 \text{ cm}^{-1}$ at 20 GHz and 300 K, we find: $\Gamma^2\tau = 1.1 \times 10^{-12} \text{ s}$ at 300 K. This value compares to typical values in crystals where Γ and τ are of the order of 10^0 and 10^{-13} s , respectively (in c-Si, using $\alpha=80 \text{ cm}^{-1}$ at 20 GHz and published data, we find $\Gamma^2\tau = 2.4 \times 10^{-12} \text{ s}$ at 300 K). Moreover, the Grüneisen constant $\langle \gamma \rangle = 1.7$ which can be extracted from dilation is also close to typical values in crystals. We can check that our result is consistent with the assumption $\omega\tau \ll 1$. When the temperature is lowered, the same calculation can be performed consistently down to $T \sim 100 \text{ K}$. Figure 5 displays $\Gamma^2\omega\tau$ between 100 and 300 K, derived using the attenuation at 20 GHz and the experimental specific heat [14]. In that

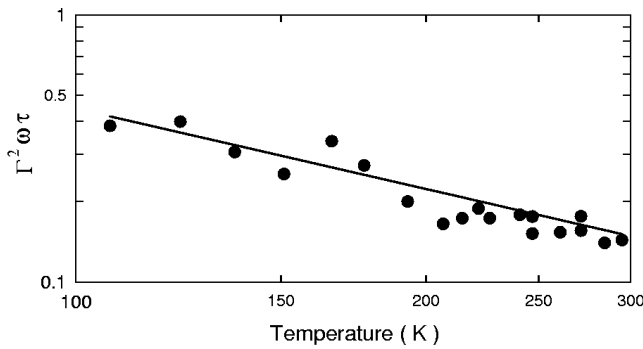


Figure 5: $\Gamma^2\omega\tau$ derived from Eq.2, using the attenuation measurement at 20 GHz (see text). The full line is a T^{-1} function.

range, we notice that $\Gamma^2\omega\tau$ varies roughly as T^{-1} . Thus, assuming that Γ is constant between 100 and 300 K, τ is found to vary as T^{-1} at high temperature. That kind of behavior is well known in crystals and arises from the density of phonons being proportional to the temperature, at high temperature. In quasicrystals, the density of lattice excitations must also be proportional to the temperature, at high temperature, so that the T^{-1} variation of τ is also expected.

Eq.2 is only valid if $\omega\tau \ll 1$. In crystals, if $\omega\tau \gg 1$ the Landau-Rumer regime is reached. In this regime, the acoustic wave is regarded as a collection of phonons decaying because of their anharmonic interaction with the thermal phonons. In quasicrystals, we still expect the attenuation to drop below some temperature because of the depletion of the thermal lattice modes. However, in the present state of knowledge, calculations in the transition regime cannot be performed. We notice that below 100 K, the attenuation is much larger than in dielectric crystalline solids [15], contrary to room temperature. Moreover, the attenuation still varies as ω^2 down to 15 K so that, surprisingly, no crossover to a new regime is observed. Then, clearly, below 100 K, the attenuation does not behave as in crystalline dielectric solids.

Conclusion

We have measured the acoustic attenuation in the icosahedral quasicrystal $\text{Al}_{68.2}\text{Pd}_{22.8}\text{Mn}_{9.0}$, between 15 K and 300 K, and in the tens of GHz range. We find that, above 100 K, the attenuation compares more to crystals than to amorphous solids. In disordered solids, the Akhieser process alone cannot explain the acoustic attenuation. Other relaxation processes are active (thermal activation of localized excitations, structural relaxation...). In a quasicrystal, we stress that, on the contrary, the attenuation behavior between 100 and 300 K is consistent with an Akhieser relaxation process in-

volving the relaxation of lattices modes perturbed by the acoustic wave. In this temperature range, the mean lifetime of the thermal excitations varies in the same way as in crystals, i.e. according to a T^{-1} law, and with the same order of magnitude ($\Gamma^2\tau \sim 10^{-12}$ s). Below 100 K, the frequency dependence of the attenuation is still quadratic so that we do not observe a crossover to a low temperature regime similar to the Landau-Rumer regime which exists in crystals. The behavior of the attenuation is not clearly understood in this temperature range. It would be interesting to perform experiments down to much lower temperature ($T \sim 1$ K).

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