Lattice Dynamics of Al-based Quasi-crystals

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Introduction

Quasi-crystals are aperiodic long-range ordered solids that are also expected to exhibit peculiar dynamical properties [1-4]. For the new intermetallic phases, previous theoretical work predicted the existence of phason dynamics and a highly structured vibrational density of states (VDOS). The most detailed information on phonon dynamics in quasi-crystals was previously obtained by inelastic neutron scattering measurements; however, specific heat, thermal conductivity, ultrasound, and light scattering experiments were also performed [5]. Very few experiments were done by using synchrotron radiation, however. The recently developed technique of inelastic nuclear resonant scattering [6, 7] was used to measure the Fe partial VDOS in an icosahedral Al-Cu-Fe powder sample fully enriched in ^{57}Fe [8]. This experiment yielded the first experimental observation of sharp features of the VDOS in quasi-crystals and outlined the potential of inelastic x-ray scattering (IXS) methods for studying the dynamic properties of aperiodic solids. The energy-resolved measurement of phonon dispersion laws by using the high-resolution IXS method became feasible [7, 9] once high-intensity x-ray sources were made available at modern synchrotron radiation facilities and storage rings. During the last decade, the IXS technique was optimized, and additional methods were developed. Meanwhile, synchrotron radiation IXS experiments may access a wide range of energy and momentum transfers, with an energy resolution of about 2 meV or better.

We used the high-resolution IXS method to investigate the lattice dynamics of Al-Cu-Fe and Al-Pd-Mn quasi-crystals, near the (18,29) diffraction peak situated on the fivefold axis. Phonon dispersion relations were determined for both quasi-crystals. In addition to propagating acoustic modes, dispersionless ("optic") low-energy modes were also observed.

Methods and Materials

Single-phase Al-Pd-Mn and Al-Cu-Fe mono-domain quasi-crystals were prepared at the Materials Science Center at Ames Laboratory, Iowa State University, by using the Bridgman method and/or specific thermal treatments. Surfaces are typically oriented to within ± 0.25º of the desired orientation (i.e., normal to the fivefold axis). The IXS method was used to investigate the lattice dynamics of both Al-Cu-Fe and Al-Pd-Mn quasi-crystals. The strong (18,29) Bragg peak lying along the fivefold symmetry axis served as the starting point of our measurements. Inelastic spectra were collected for energy transfers ΔE of up to 40 meV. The phonon dispersion relations (longitudinal and transverse) corresponding to the fivefold symmetry axis could be determined from momentum-resolved energy scans for both Al-Pd-Mn and Al-Cu-Fe quasi-crystals. Our IXS experiment took advantage of the high energy resolution attained at the 3-ID beamline at APS [10]. The energy resolution for the present experiments was equal to 1.87 meV, as determined from energy scans made by using a plexiglass sample. Rectangular collimation slits were mounted in front of the analyzer crystal in order to improve the momentum resolution. In turn, this is expected to lead to an increase of the energy resolution for propagating modes. We used a slit system defining a "low-resolution" setup as well as a smaller entrance slit for the "high-resolution" mode. While the "low-resolution" setup offers more intensity (an advantage for low-intensity excitations), the "high-resolution" measurements allow us to resolve nearby phonon excitations.

Results

Selected IXS patterns of Al-Pd-Mn are presented in Fig. 1. The momentum transfer $\Delta Q \equiv q$ is conventionally expressed in reduced units (the quasi-lattice constant for the $Al_{71.5}Pd_{20.2}Mn_{8.3}$ quasi-crystal is $a = 6.451$ Å). The corresponding dispersion relation is illustrated in Fig. 2. Apart from a well-defined propagating longitudinal mode (open symbols in Fig. 2), several low-energy dispersionless modes were also evidenced (filled symbols in Fig. 2; see also Fig. 3). We observed low-intensity "optic" modes at energies close to 5, 7, 12.4, and 15.7 meV (Fig. 2). For energy transfers larger than 20 meV, excitations are typically very broad and more difficult to resolve individually.

For comparison, inelastic neutron scattering (INS) experiments on Al-Pd-Mn quasi-crystals [11] provided evidence for "optic" modes located at 7.4, 12.4, 15.7, and 22.75 meV (corresponding to frequencies of 1.8, 3.0, 4.0, and 5.5 Thz), in good agreement with the present results. In the long-wavelength (continuum) limit, the dispersion relation of the longitudinal acoustic mode is linear, with the sound velocity close to 6.3×10^3 m·s^{-1}, also in agreement with INS measurements [11,12]. The "optic" mode located close to 5 meV was not previously observed by other groups. We have therefore investigated this dispersionless mode in some detail. Figure 3 illustrates the room-temperature IXS pattern taken at $q = 0.5$ (reduced units): two phonon excitations at 5 and 6.9 meV may clearly be identified, together with the major longitudinal acoustic phonon at 8 meV. At large q values, the 5-meV optic mode may still be observed in the IXS patterns, provided the "high-resolution" setup is used. Low-temperature (10K) measurements were also performed in order to certify the physical nature of the low-energy excitation at 5 meV: All features of the IXS spectra exhibit the expected intensity variation with temperature (Fig. 4).

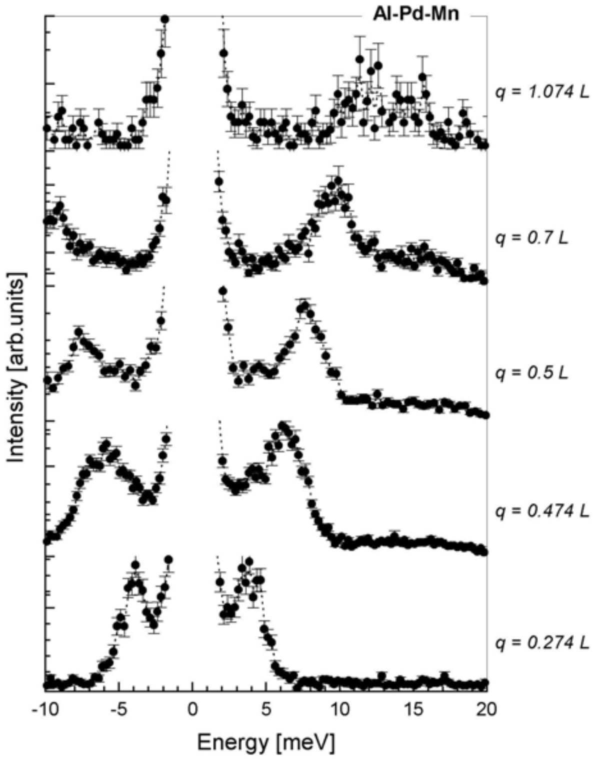

FIG. 1. IXS spectra of Al-Pd-Mn quasi-crystal.

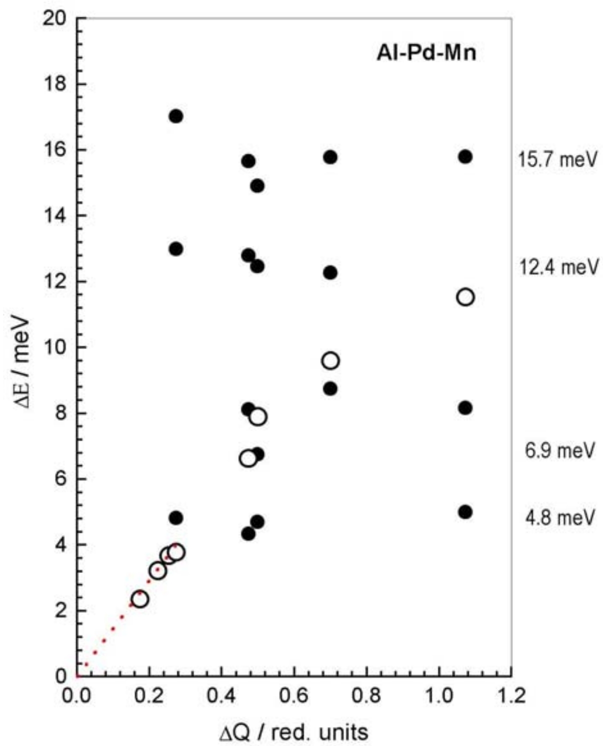

FIG. 2. Phonon dispersion relation for the Al-Pd-Mn quasi-crystal (longitudinal).

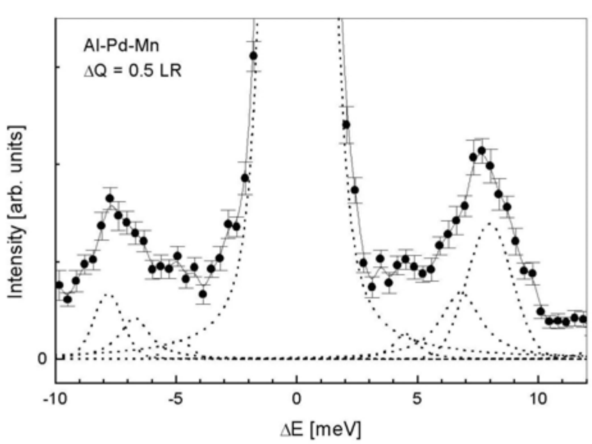

FIG. 3. IXS pattern at $\Delta Q = 0.5$ (red. units).

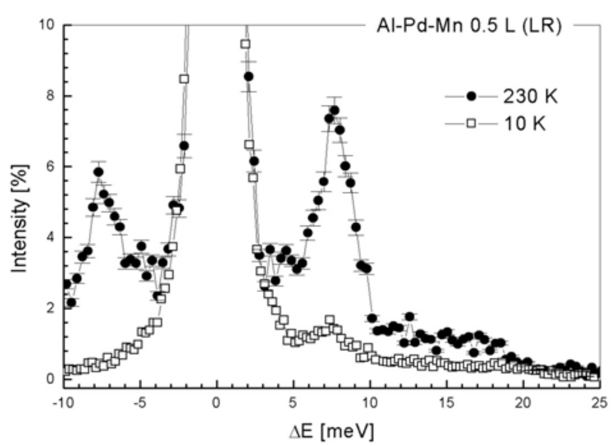

FIG. 4. Comparison between IXS patterns at room temperature and 10K (Al-Pd-Mn).

Discussion

In quasi-crystals, dispersionless modes are tentatively associated to the existence of a dense set of gaps in the VDOS and to the "critical" nature of the wave functions [5, 12]. We have determined the phonon dispersion relations for Al-Pd-Mn and Al-Cu-Fe quasi-crystals by using the IXS method with an energy resolution better than 2 meV. Apart from well-defined acoustic modes, dispersionless modes were also found. A low-energy "optic" band located at about 5 meV could be clearly quantified for the first time. It is expected that further high-resolution inelastic scattering experiments in the low-energy low-momentum transfer range as well as theoretical modelling of the available experimental data will contribute to a more accurate understanding of the physical origin of "optic" vibrational modes in quasi-crystals.

Acknowledgments

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