Sub-meV Monochromator Development for 14.4 keV X-rays

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Momentum-resolved inelastic x-ray scattering has become a complementary technique to coherent neutron scattering for the determination of lattice dynamics. Both techniques involve partial measurement of a select group of phonon dispersion curves that is used as an empirical basis for a force-constant model, e.g., a Born-von Karmon harmonic oscillator model, from which one reconstructs the complete dispersion surface. Despite the enormous utility of these two techniques, the extraction of phonon density of states (DOS) with such methods produces model-dependent results and typically requires anharmonic terms that give rise to phonon lifetime effects to be small. In cases in which it is sufficient to have the phonon DOS in place of the complete dispersion surface, a more direct measurement without restrictions on anharmonicity is desirable. This is possible with incoherent neutron scattering, as well as with incoherent nuclear resonant γ-ray scattering. Specifically, the technique of incoherent inelastic resonant scattering of synchrotron radiation from the 14.4 keV nuclear resonance in ^{57}Fe can be used to determine the partial phonon DOS in any ^{57}Fe compound [1,2]. Furthermore, with this technique the nuclear resonance effectively performs the energy analysis, and the resolution of the measurement is determined solely by the energy bandwidth of the incident radiation. This allows the possibility of measuring ^{57}Fe partial phonon DOS with excellent resolution but requires one to achieve sub-meV monochromatization of hard x-rays. Performing inelastic x-ray scattering with sub-meV energy resolution requires both a high-brightness radiation source, as well as an efficient high-resolution x-ray monochromator.

Monochromatization at the nuclear transition energy precludes the use of crystal back-reflections as has been customary in the past to achieve the highest energy resolutions. In addition to the energy constraint, the x-ray monochromator must be tunable over a region sufficient to measure lattice excitations, viz. a few hundred meV. The use of two high-order symmetrically cut crystal reflections in a dispersive geometry offers energy resolutions of a few meV at this energy but suffers from low transmission [3]. In order to achieve sub-meV resolution with good efficiency, the use of extreme asymmetry angles on high-order crystal reflections was suggested [4]. A sub-meV energy bandwidth of 0.92 meV has been achieved with this method [5]. We have improved the resolution beyond this by employing even more extreme asymmetry angles on the diffracting crystals. This method of achieving sub-meV resolution may be applied at other energies in the range of 2-30 keV. In addition, the monochromator's tunability extends over a range of $\approx$ 50-100 eV. We demonstrate the performance of this monochromator by using it to measure the ^{57}Fe partial phonon DOS in α-$^{57}Fe_2O_3$ using the technique of inelastic nuclear resonant scattering.

The design of the high-resolution monochromator requires that one use crystal reflections with the largest Bragg angle at the x-ray energy of interest. In our case, we used two silicon (9 7 5) reflections, which have a room-temperature Bragg angle of $\Theta = 80.40°$ for 14.413 keV x-rays. The first crystal has an asymmetry angle of $-79.94°$ (asymmetry factor $b \approx 0.024$). Consequently, the angular acceptance for monochromatic radiation is approximately 11.9 μradian and the diffracted divergence is 0.28 μradian. The asymmetry angle for the second crystal may be relaxed slightly in comparison to the first crystal in return for greater reflectivity and hence greater overall throughput. The asymmetry angle chosen for the second crystal is $+79.52°$ (asymmetry factor $b \approx 22.5$). Due to the asymmetry angle on the second crystal, its angular acceptance is approximately 0.39 μradian, which is 0.11 μradian larger than the divergence of the monochromatic radiation incident upon it. A diagram of the crystal arrangement is shown in figure 1.

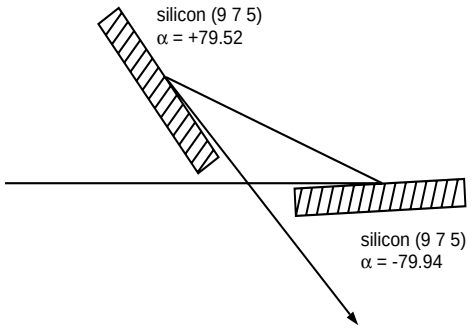

FIG. 1. Crystal arrangement of the high-resolution monochromator.

The asymmetry angle on the first crystal results in an incidence angle of 0.46° with respect to the surface, which is well above the critical angle for total external reflection of 0.13°. Note that extreme asymmetry angles that result in incidence angles approaching the critical angle for total external reflection effect a saturation of the Darwin width, as well as a reduction of the efficiency due to flux lost to the specular beam [6]. Consequently, even with the application of higher asymmetry angles and assuming perfect crystallinity, producing energy bandwidths with this design below approximately 300 μeV at this energy will be difficult.

All measurements were carried out at the 3-ID undulator beamline of the Advanced Photon Source. The energy-resolution function is measured by energy scanning the monochromator through the 14.4 keV nuclear resonance of ^{57}Fe while monitoring the delayed x-ray flux

emitted into the forward direction. The result is shown in figure 2. This is an elastic scattering measurement and consequently represents the energy-resolution function of the instrument since the energy width of the nuclear hyperfine spectrum ($\approx 0.5\,\mu$eV) is three orders of magnitude smaller than the energy bandwidth transmitted by the monochromator. The result of a simulation gives an energy width of $660\pm40\,\mu$eV FWHM. This degree of monochromatization corresponds to $E/\Delta E \approx 2.2\times10^7$ and a longitudinal coherence length of 1.8 mm. Our estimate for the uncertainty in the energy width is based on the distribution of widths used to simulate a number of different measurements under a variety of environmental conditions including: positive and negative temperature drifts, as well as positive and negative crystal rotations. The transmitted flux is $\approx 6\times10^8$ ph/s at 100 mA storage ring current, which represents approximately 9 % of the incident spectral flux.

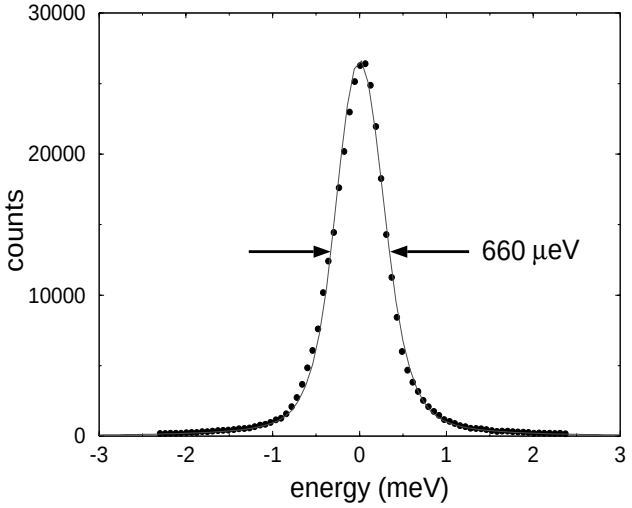

FIG. 2. Energy resolution function of the monochromator as measured by coherent elastic nuclear resonant scattering from ^{57}Fe. The solid curve is a simulation. The energy width is $660\pm40\,\mu$eV FWHM.

The high spectral intensity of undulator-based synchrotron radiation makes extreme energy resolutions (a few parts in 10^8) practical. To demonstrate this, we used the high-resolution monochromator to measure the ^{57}Fe partial phonon DOS in ^{57}Fe-enriched hematite, α-$^{57}Fe_2O_3$ (>90% enriched in ^{57}Fe), using the method of inelastic nuclear resonant scattering. This involves energy scanning the monochromator around the nuclear transition energy while detecting the yield of delayed K-fluorescence from the hematite using an avalanche photodiode. The ^{57}Fe-partial phonon DOS can be extracted directly from the measurement using a Fourier-log deconvolution procedure [2].

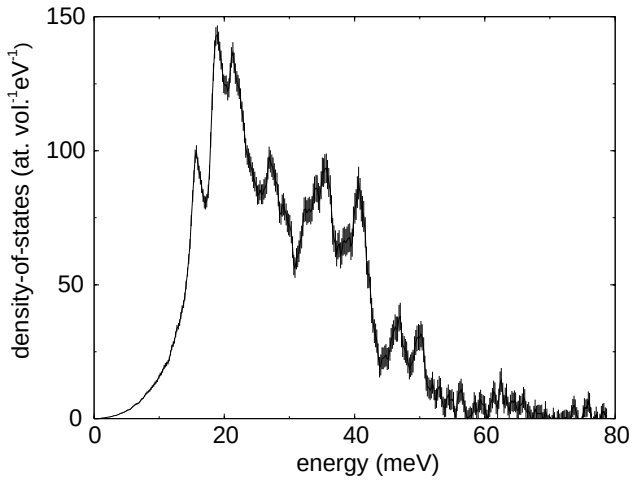

FIG. 3. ^{57}Fe-partial phonon density-of-states in hematite (α-$^{57}Fe_2O_3$) measured with an energy resolution of $660\,\mu$eV.

The result is shown in figure 3. Other useful thermodynamic quantities are also obtainable from the data. In the present case, for ^{57}Fe in hematite at room temperature, we calculate a room-temperature Lamb-Mossbauer factor of 0.788(2), an average kinetic energy per atom of 14.51(6) meV, a mean force constant of 225(3) N/m, a vibrational specific heat of 2.67(1) k_B/atom, and a vibrational entropy of 3.08(1) k_B/atom.

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