Data Analysis for Inelastic Nuclear Resonant Absorption Experiments

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Inelastic nuclear resonant absorption (INRA) of synchrotron radiation is a unique technique to study phonon excitations [1, 2]. The inelastic excitation of nuclear resonance was studied theoretically in the 1960s [3, 4]. In contrast to the Mössbauer effect, for which the recoilless excitations are studied, this inelastic approach exploits the energy transfer between x-rays and lattice. Because only a particular type of isotope can be resonantly excited by x-rays with energy close to its resonant energy, this method provides a way to study the dynamics of selected atoms, i.e., nuclear resonant isotopes.

The resonant excitation, both elastic and inelastic, can be observed by detecting nuclear decay products. When the nuclear resonant lifetime is much longer than the synchrotron pulse length and the time scale of electronic scattering process, the nuclear resonant scattering process is distinguished by counting signals only after the disappearance of the prompt radiation and electronic scattering. This leads to an enormous background suppression. Thus noise level is basically determined by the detector and the associated electronics, which can be suppressed well below the signal level in most cases.

The experimental setup for INRA is shown in Fig. 1. The pre-monochromator reduces the x-ray energy bandwidth to a few eV and removes most of the heat load in the beam. The high-resolution monochromator further reduces the energy bandwidth to meV or less, which is the experimental resolution needed to study phonon excitations. The detector is placed very close to the sample to collect as many nuclear decay signals as possible. The intensity of delayed signal is proportional to nuclear resonant excitation cross section, which in turn is proportional to a Fourier transform, $S(\varepsilon, \mathbf{k})$, of the autocorrelation function of the scattering system [4]. The reason it is related to the auto- rather than the pair-correlation is that the inelastic resonant absorption is an incoherent process. In general, $S(\varepsilon, \mathbf{k})$ depends on the incident photon momentum $\mathbf{k}$. But in certain cases, e.g., for an isotropic lattice, this dependence can be dropped.

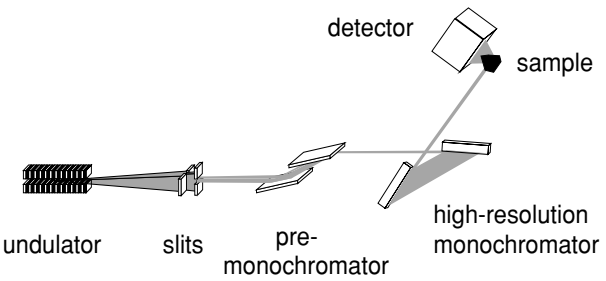

Figure 1: The experimental setup of inelastic nuclear resonant scattering experiments.

$$S(\varepsilon, \mathbf{k}) = \frac{1}{N} \int \frac{dt}{2\pi} e^{-i\varepsilon t} \sum_i g_i \langle i | \sum_l e^{-i\mathbf{k}\mathbf{r}_l(0)} e^{i\mathbf{k}\mathbf{r}_l(t)} | i \rangle \ , \tag{1}$$

where N is the number of resonant nuclei, σ_0 is the maximum resonant excitation cross section, and Γ is the natural linewidth of the resonance. The g_i is the statistical weight factor of initial lattice state $|i\rangle$. The $\mathbf{r}_l$ is the position of the l-th resonant nucleus.

In resonant absorption, there is a sudden momentum transfer and thus moment sum rules apply [5],

$$\int S(\varepsilon, \mathbf{k}) \, (\varepsilon - E_\mathrm{R}) \, d\varepsilon = 0 \tag{2}$$

$$\int S(\varepsilon, \mathbf{k}) \, (\varepsilon - E_\mathrm{R})^2 \, d\varepsilon = 4 \, E_\mathrm{R} \, \overline{T_\mathbf{k}} \tag{3}$$

$$\int S(\varepsilon, \mathbf{k}) \, (\varepsilon - E_\mathrm{R})^3 \, d\varepsilon = \frac{E_\mathrm{R}}{m} \, \hbar^2 \, \overline{F_\mathbf{k}} \tag{4}$$

where E_R is the recoil energy, m the nuclear mass, $\overline{T_\mathbf{k}}$ the mean kinetic energy of the resonant nuclei in the $\mathbf{k}$ direction, and $\overline{F_\mathbf{k}}$ the mean force constant experienced by the resonant nuclei in the $\mathbf{k}$ direction. While the higher moments give us information on vibrational dynamics of sample, the first moment is employed to normalize the spectrum and determine the recoilless fraction. The moments of $S(\varepsilon)$ can be calculated [6] from those of experimentally measured flux, $I_\mathrm{m}(\varepsilon)$, which is a convolution involving the instrument resolution function $R(\varepsilon)$.

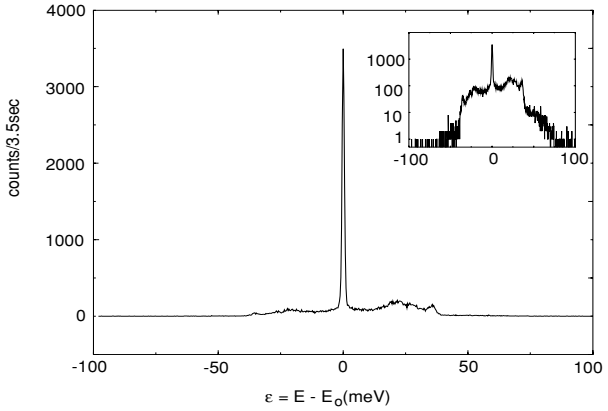

Figure 2: The data taken on an α-Fe foil, with energy resolution of 0.9 meV. The insert shows the same data on a log scale.

We measured an enriched α-Fe foil by INRA with an energy resolution of 0.9 *meV* [7]. A spectrum is shown in Fig. 2. The center spike is the elastic peak resulting from

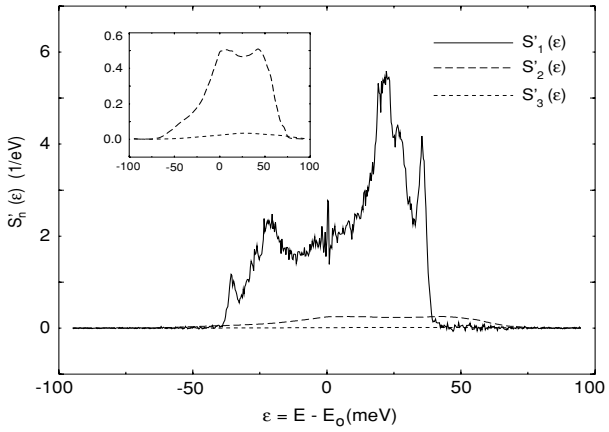

Figure 3: The single and multiple phonon contributions to $S'(\varepsilon)$. The inset shows $S'_2(\varepsilon)$ and $S'_3(\varepsilon)$ on an enlarged scale.

the recoilless nuclear resonant excitation. The inelastic excitation is clearly seen in the spectrum and extends to about 80 meV away from the peak. The low-energy side of the spectrum corresponds to phonon annihilation, the high-energy side, phonon creation.

The elastic part of the function $S(\varepsilon)$ is appreciably larger than the inelastic part. In the case of enriched iron metal, the former is six orders of magnitude larger. However, the elastic peak is suppressed in the measured spectrum, due to exactly the large cross section and the strong absorption of x-rays with the exact resonant energy that results in small penetration depth and less material contributing to the elastic peak. So, the first step in data evaluation is to fit the elastic peak with the instrument resolution function and remove the peak from the spectrum. Then one must to normalize the spectrum and at the same time determine the recoilless fraction. Because the elastic peak is suppressed by an unknown factor, it cannot be normalized by simply integrating the spectrum. Instead, we replace the elastic peak by the theoretically expected,

$$I(\varepsilon) = a\,S(\varepsilon) = a\,[S'(\varepsilon) + f\,\delta(\varepsilon)]\ , \qquad (5)$$

where a and f can be solved as follows,

$$a = \frac{1}{E_R}\left[\int I'_m(\varepsilon)\,\varepsilon\,d\varepsilon - \int I'_m(\varepsilon)\,d\varepsilon \int R(\varepsilon)\,\varepsilon\,d\varepsilon\right] \qquad (6)$$

$$f = 1 - \frac{1}{a}\int I'_m(\varepsilon)\,d\varepsilon\ . \qquad (7)$$

With the normalized spectrum, the mean kinetic energy and the mean force constant experienced by the nuclei are derived by the sum rules (Eqs. 3 and 4). In this experiment, the recoilless fraction f is found to be 0.796(2), the mean kinetic energy per nucleus 42.9(3) meV, and the mean force constant $1.74(6) \times 10^8$ N/m.

Finally, if a harmonic lattice model is assumed, the inelastic part of the function $S(\varepsilon)$ can be further separated into single- and multiphonon contributions, i.e.,

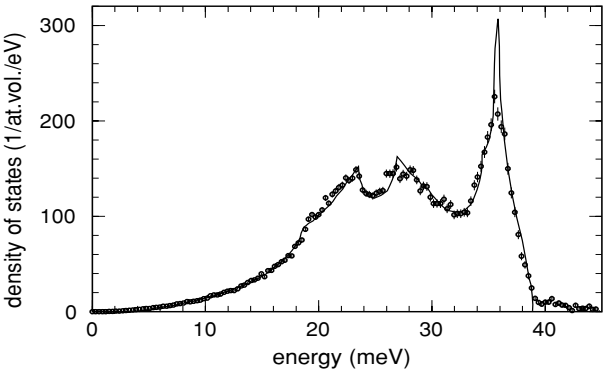

Figure 4: The phonon DOS of α-Fe(circles), compared with a neutron result(solid line).

$S'(\varepsilon) = S'_1(\varepsilon) + S'_2(\varepsilon) + S'_3(\varepsilon) + \cdots$. These terms follow certain recursive relations and can be calculated by the method of Fourier-Log decomposition (Fig. 3). The phonon density of states per unit energy per atomic volume, $\mathcal{D}(\varepsilon)$, is related to the single-phonon term [4, 2],

$$\mathcal{D}(\varepsilon) = 3\,\frac{\varepsilon}{E_{\rm R}}\,\frac{S'_1(\varepsilon)}{f}\,[1 - exp(-\frac{\varepsilon}{k_B T})]\ . \qquad (8)$$

The above-mentioned Fourier-Log decomposition is carried out to separate multiphonon contributions (Fig. 3) and to extract the phonon DOS (Fig. 4). The single phonon contribution can be seen to dominate and it can be attributed to the high recoilless fraction of 0.796 of α-iron. The phonon DOS derived from the experiment is compared with the calculated DOS from coherent inelastic neutron scattering data [8].

Use of the Advanced Photon Source is supported by the U. S. Department of Energy, Basic Energy Sciences, Office of Science, under contract No. W-31-109-ENG-38.

References

[1] M. Seto, Y. Yoda, S. kikuta, X. W. Zhang, M. Ando, *Phys. Rev. Lett.* **74**, 3828 (1995).

[2] W. Sturhahn, T. S. Toellner, E. E. Alp, X. Zhang, M. Ando, Y. Yoda, S. Kikuta, M. Seto, C. W. Kimball, B. Dabrowski, *Phys. Rev. Lett.* **74**, 3832 (1995).

[3] W. M. Visscher, *Annals of Phys.* **9**, 194 (1960).

[4] K. S. Singwi, A. Sjolander, *Phys. Rev.* **120**, 1093 (1960).

[5] H. J. Lipkin, *Phys. Rev.* **B 52**, 10073 (1995).

[6] M. Y. Hu, W. Sturhahn, T. S. Toellner, P. M. Hession, J. P. Sutter, E. E. Alp, *Nucl. Instrum. Methods A,* **428**, 551 (1999).

[7] T. S. Toellner, M. Y. Hu, W. Sturhahn, K. Quast, E. E. Alp, *Appl. Phys. Lett.* **71**, 2112 (1997).

[8] V. J. Minkiewicz, G. Shirane, R. Nathans, *Phys. Rev.* **162**, 528 (1967).