ESRF	**Experiment title:** Temperature dependent phonon density of states of ^{119}Sn	**Experiment number:** HS-676
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Shifts: 18 (9+9)	**Local contact(s):** A. Barla	*Received at ESRF:*

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Report:

The experiment HS676 was mainly aimed at studying the lattice dynamics of β-Sn at low temperatures using Nuclear Inelastic Scattering (NIS). In a previous experiment (see, e.g., [1] and experimental report of HE282), we observed that, although the low energy acoustic phonons around 3.5 meV were clearly visible in our spectrum, thanks to the very good energy resolution (< 1 meV) of our monochromator (HRM), the dynamics of β-Sn at 300 K is dominated by multiphonon processes, because of the low Lamb-Mössbauer factor ($\sim 4\%$). As a consequence of this, the determination of the phonon density of states (DOS), which depends only on single phonon scattering, was not possible. It is therefore necessary to go to low temperatures, where the higher Lamb-Mössbauer factor and the reduced probability of phonon annihilation processes will reduce the occurrence of multiphonon events. In order to carry out low temperature measurements with a good countrate, new crystals for the HRM were prepared by the ESRF Optics Group, and then tested partly during the shifts in single bunch mode (~7 of the 9 shifts available). By using a Be compound refractive lens to reduce the divergence of the synchrotron beam, we obtained a flux of $1.1 \cdot 10^7$ photons/s in an energy bandwidth of ~ 1 meV, improving by a factor of ~ 5 with respect to HE282.

The NIS measurements on β-Sn at low temperature were carried out in 16-bunch mode (during ~ 4 shifts), with a maximum countrate of ~ 4 Hz. The sample (a $\sim 100\,\mu$m thick foil) was mounted in a closed cycle cryostat. Fig. 1 shows the probability density of NIS by β-Sn at $T = 100$ K, compared with the one at room temperature. One clearly sees the almost complete disappearance of multiphonon scattering and the shift to higher energies ($\sim 15\%$) of the low energy peaks. Fig. 2 shows the DOS extracted from the low temperature data, together with the one calculated in [2] for room temperature and shifted in energy by 15%. The agreement is good, even if the peaks in the experimental DOS are less pronounced due to the finite resolution.

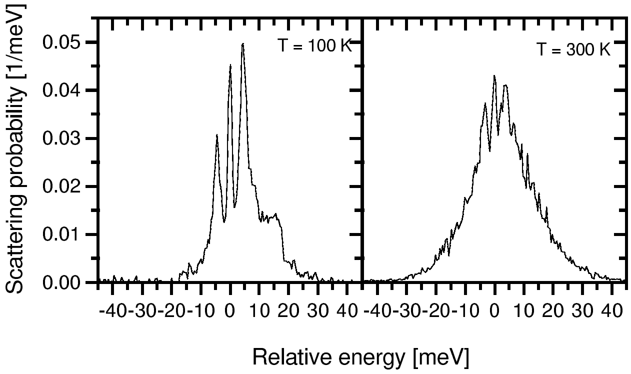

Fig. 1: NIS by β-Sn

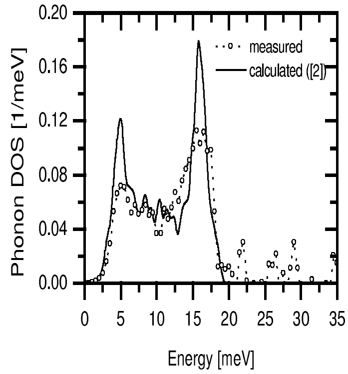

Fig. 2: DOS of β-Sn

The Lamb-Mössbauer factor obtained from the DOS is $f_{LM} = 0.41(3)$, in agreement with the one given in [3]. Using Lipkin's sum rules [4], we directly determined from the NIS spectrum the mean kinetic energy $< T >= 5.2(1.1)\,\mathrm{meV}$ and (in the harmonic approximation) the mean force constant $K = 95.6(9)\,\mathrm{N/m}$. From the DOS we obtain the specific heat at constant volume $c_V = 5.5(9)\,\mathrm{calK^{-1}mol^{-1}}$ and the molar vibrational entropy $S = -6.0(7)\,\mathrm{calK^{-1}mol^{-1}}$.
First test measurements on powder samples of SnO_2 and SnTe have been made at room temperature, but the lack of time didn't allow to get sufficient statistics to determine the DOS.
The precise determination of hyperfine splittings of the order of the natural linewidth or less is very difficult with conventional Mössbauer spectroscopy. Even if β-Sn and SnO_2 have been widely studied in the past with this technique, there especially the determination of the small quadrupole splittings (less than the natural linewidth) could not be done accurately. Therefore during the shifts in single bunch mode not used for the testing of the HRM crystals (and not usable for the NIS experiments due to the 6 times lower flux in comparison to 16-bunch mode), Nuclear Forward Scattering was used to measure their Lamb-Mössbauer factor f_{LM}, quadrupole splitting ΔE_Q and isomer shift IS at room temperature. Fig. 3 shows the time spectra of the two compounds separately (second and third spectra in the figure), together with a fit giving $\Delta E_Q = 0.24(1)\,\mathrm{mm/s}$ for β-Sn and $\Delta E_Q = 0.55(6)\,\mathrm{mm/s}$ for SnO_2, and $f_{LM} = 0.039(1)$ (in agreement with [3]) for β-Sn. The first spectrum in Fig. 3 was measured with both absorbers in the beam at the same time, allowing to determine the relative isomer shift $IS = 2.56(1)\,\mathrm{mm/s}$ of β-Sn with respect to SnO_2, in agreement with Mössbauer spectroscopy.

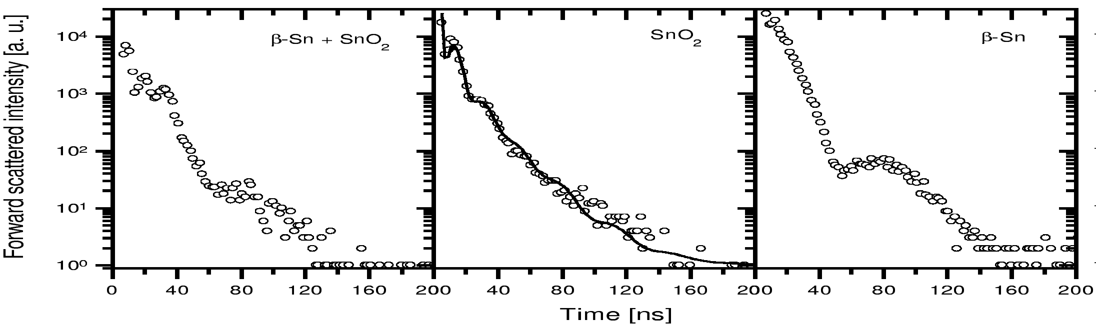

Fig. 3: NFS on β-Sn and SnO_2

References:

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