# Nuclear inelastic scattering of synchrotron radiation by $^{119}$Sn

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We have investigated inelastic scattering of synchrotron radiation by $^{119}$Sn nuclei using the radiative channel of nuclear deexcitation. The energy dependence of nuclear scattering of the 23.8795-keV x rays by a $\beta$-Sn foil was measured with sub-meV energy resolution. In the conditions of the experiment the scattering was incoherent over the various nuclei and averaged over the phonon momentum transfer. The experimental data are compared with the theory of nuclear inelastic absorption. The general agreement is reasonable, however, some deviation is noted. The possible origin of the discrepancy is discussed. [S0163-1829(98)02626-5]

The progress in nuclear scattering of synchrotron radiation (for a review see, e.g., Refs. 1,2) resulted in the recent observation of inelastic excitation of nuclei by x rays.[3] An incident photon with an energy close to the energy of the nuclear transition can excite the nucleus, even though it does not match the transition energy exactly. The energy conservation in this case is fulfilled by creation or annihilation of phonons in the sample. The intensity of nuclear inelastic excitation can be monitored through the yield of the products of nuclear deexcitation.

Nuclear deexcitation can proceed via two channels: radiative decay and internal conversion. Decaying via the radiative channel the nucleus emits $\gamma$-ray fluorescence radiation. While the nucleus decays via the channel of internal conversion, the energy of the nuclear excited state is transmitted to an electron of the atomic shell. This conversion electron leaves the atom, and the remaining hole results in a subsequent emission of atomic x-ray fluorescence radiation. Choosing in the experiment either nuclear $\gamma$-ray or atomic x-ray fluorescence radiation one may study nuclear inelastic *scattering* and nuclear inelastic *absorption*, respectively.

Most of the experiments on inelastic excitation of nuclei[3–7] were performed by monitoring x-ray fluorescence radiation. The reasons are the dominating probability of the internal conversion channel, the higher efficiency of the detectors for softer x-ray fluorescence radiation, and trapping of $\gamma$-ray fluorescence radiation in the sample.[8] Therefore, the up to now studied channel was nuclear inelastic absorption. This channel has been well investigated. The theory of nuclear inelastic absorption is available for both polycrystalline[9] and single crystal[10] samples, and shows good agreement with the experimental data.[3–5]

In contrast to nuclear inelastic absorption, the radiative channel of nuclear inelastic scattering has not yet been explored. There are no published experimental data on nuclear inelastic scattering, which would be suitable for the quantitative analysis, and the theoretical works have just started.[11,12] The aim of this work is to investigate the energy dependence of nuclear inelastic scattering through the radiative channel of nuclear deexcitation.

For this purpose we use the 23.8795-keV (Ref. 13) nuclear transition of $^{119}$Sn. For this isotope the energy of the $\gamma$-ray fluorescence radiation is much higher than the energy of the x-ray fluorescence radiation, therefore the channels of nuclear scattering and nuclear absorption may be easily distinguished. Synchrotron radiation experiments with $^{119}$Sn nuclei started several years ago.[14] It was this isotope that permitted the first observation of nuclear inelastic excitation.[13] However, the energy resolution in that early work ($\approx$35 meV) was not sufficient for the quantitative analysis of the data. Here we report on studies of nuclear inelastic scattering by $^{119}$Sn with sub-meV energy resolution.

The experiment was performed at the Nuclear Resonance Beamline[15] ID18 at the European Synchrotron Radiation Facility (ESRF). The storage ring was operated in 16-bunch mode with an averaged current of 70 mA. The details of the experimental technique can be found in Refs. 5,15.

A new high resolution monochromator for the 23.8795-keV radiation was built. It was elaborated according to the conventional ''nested'' design.[16,17] For the outer channel-cut crystal we have chosen a Si (6 4 2) reflection with asymmetry parameters $b_1 = -0.043$ and $b_2 = 1/b_1$. For the inner channel-cut crystal a symmetric Si (12 12 12) reflection was used. The angular acceptance of the monochromator was $\approx 7$ $\mu$rad. The bandpass of the monochromator of 0.97(5) meV [full width at half maximum (FWHM)] was determined from the fit of the instrumental function with a Gaussian distribution (Fig. 1). The throughput of the spectral intensity was smaller than expected, however, still sufficient to perform the experiment. The flux in the 0.97 meV bandwidth was about $2.4 \times 10^6$ photons/s.

The measurements were performed with the use of a polycrystalline foil of $\beta$-Sn. The abundance of the resonant $^{119}$Sn isotope in the sample was $\approx$90%. The thickness of the foil along the x-ray beam was $\approx 90$ $\mu$m. Incoherent scattering of radiation from the foil was measured with the large-area avalanche photo diode detector,[18] which covered the solid angle of $\approx 1\pi$ srad. The electronics was adjusted to count only the 23.8795-keV nuclear fluorescent radiation, the soft atomic $L$-fluorescent radiation ($\leqslant$4.1 keV) was rejected by the dis-

 

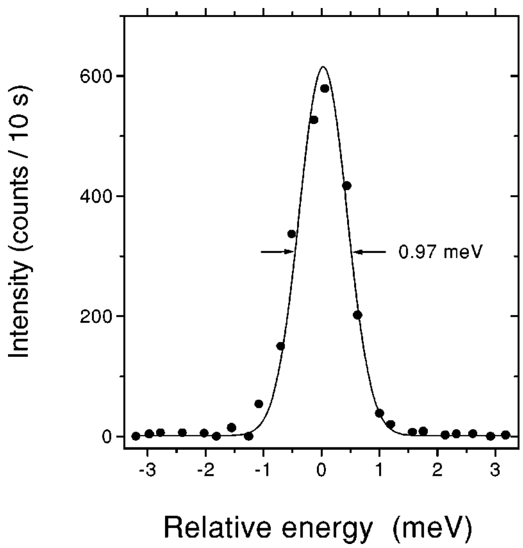


FIG. 1. Instrumental function of the high resolution Si (6 4 2) / Si (12 12 12) nested monochromator, which was measured with forward scattering of synchrotron radiation by the $^{119}Sn$ foil. The experimental data (dots) are fits by a Gaussian distribution (solid line). The best fit was reached with a width of 0.97(5) meV (FWHM).

criminator. During the measurements the instrumental function of the spectrometer was monitored via coherent nuclear forward scattering. This was measured with the stacked multidiode detector.[19] The typical count rate at resonance was $\approx 3.5$ counts/s for incoherent scattering and $\approx 6$ counts/s for forward scattering. The measurements were performed at room temperature.

The energy spectrum of incoherent nuclear scattering (Fig. 2) has a broad distribution with a width of $\approx 20$ meV, much wider than the instrumental function. This clearly demonstrates inelastic scattering. The spectrum has two peaks of inelastic scattering at about $\pm 4$ meV.

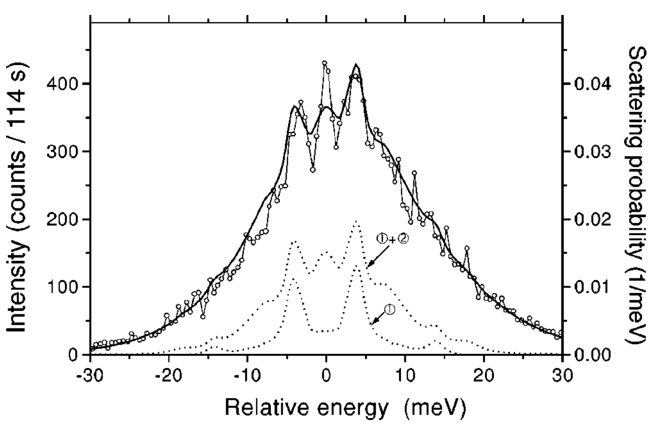


FIG. 2. Energy dependence of incoherent nuclear resonant scattering of 23.8795-keV quanta by the $^{119}Sn$ foil. The experimental data are shown by the open circles and the thin line (to guide the eye). The thick solid line shows the probability density of nuclear inelastic scattering, calculated using the density of phonon states from Ref. 20 according to Eq. (2) and convoluted with the instrumental function of the monochromator. The single-phonon contribution and the sum of single- and two-phonon contributions to the total probability of nuclear inelastic scattering are shown by the dotted lines 1 and 1+2, respectively.

The analysis of the experimental data is based on several assumptions. First, we assume that nuclear inelastic scattering was incoherent over various nuclei, and the contribution of coherent inelastic scattering was negligible. Indeed, coherent inelastic scattering is likely to be significant only during a very short time after the excitation, when the special phase correlation of *inelastic* scattering over various nuclei is fulfilled. This phase correlation, which is specific for some modes of the lattice vibration, should remain only during the time, comparable with the phonon lifetime ($10^{-10}-10^{-12}$ s).[21] Therefore the decay of coherent nuclear inelastic scattering should be extremely fast. Since in our experiment the scattering events were collected starting $\approx 8$ ns after the excitation, we suppose that the contribution of coherent nuclear inelastic scattering was negligible. Secondly, we assume that due to the polycrystalline structure of the sample and due to the large angular acceptance of the detector, the collected signal of inelastic scattering was completely averaged over all possible momenta of the phonons in the lattice. Finally, we assume that Lipkin's sum rules,[22,23] which are derived for nuclear inelastic absorption or emission, are valid also for the case of incoherent nuclear inelastic scattering, since they result from the general conditions of the translation symmetry of the interatomic forces.

Based on these assumptions, we have normalized the experimental energy spectrum of nuclear scattering using the condition, that the first momentum of the spectrum equals the recoil energy $E_R=\hbar^2k^2/2M=2.572$ meV of a free $^{119}Sn$ nucleus (see Refs. 4,5 for details). Here $k$ is the wave vector of the resonant $\gamma$-ray quantum and $M$ is the mass of the nucleus. The vertical scale for the normalized data is given by the right vertical axes in Fig. 2.

It was shown[11] that in the harmonic approximation the energy dependence of incoherent nuclear inelastic scattering, which is accompanied by creation or annihilation of a single phonon and is averaged over all possible momenta of the phonon, is identical to the energy dependence of the single-phonon nuclear inelastic absorption:

$$S_1(E)=\frac{E_R g(|E|)}{E[1-\exp(-\beta E)]}, \tag{1}$$

where $S_1(E)$ is the normalized probability density of single-phonon scattering, $E$ is the energy of the incident x-ray photon relative to the energy of the nuclear transition, $\beta=(k_BT)^{-1}$, $k_B$ is the Boltzmann constant, $T$ is the temperature, and $g(|E|)$ is the density of phonon states (DOS). The data of the density of phonon states in $\beta$-Sn were taken from Ref. 20, where DOS was calculated within the framework of the Born-von Karman model with the account of the six coordination spheres. These calculations are in good agreement with the results of neutron scattering experiment.[24] We avoided using directly the experimental data from Ref. 24 because the energy resolution in those measurements varied with the energy transfer, and in the high-energy region it was two times larger than ours. The single-phonon part of inelastic scattering was calculated according to Eq. (1) and convoluted with the instrumental function. The results are shown by dotted line No. 1 in Fig. 2. Comparison of the single-phonon contribution of scattering to the experimental data shows that this term is relatively small, thus inelastic scattering is mainly dominated by the multiphonon processes. This agrees with the low Lamb-Mössbauer factor $f_{\rm LM}=0.042(2)$, which was derived from the time dependence of nuclear forward scattering.[25] The positions of the peaks in the energy spectrum of the single-phonon scattering ($\pm 4$ meV) correspond to the peaks of the experimental spectrum.

One can suggest that for the momentum-averaged incoherent inelastic scattering not only the single-phonon contribution, but also the total scattering has the same energy dependence as the inelastic absorption.[12] Then the normalized probability density of inelastic scattering $S(E)$ is described by the sum over the $n$-phonon contributions $S_n(E)$:[9]

$$S(E)=f_{\rm LM}\sum_{n=1}^{\infty} S_n(E). \tag{2}$$

In harmonic approximation they can be found from the single-phonon term $S_1(E)$ through the recursive relation[9]

$$S_n(E)=\frac{1}{n}\int_{-\infty}^{\infty} dE'\ S_1(E')\ S_{n-1}(E-E'). \tag{3}$$

The probability density of inelastic scattering was calculated according to Eqs. (1)–(3) and convoluted with the instrumental function. The multiphonon contributions until $n=50$ were taken into account. Figure 2 shows the comparison of the normalized experimental energy spectrum (open circles) with the calculations (solid line). These two data sets are presented in an absolute scale without any additional fit. In general, the calculated scattering probability shows good agreement with the experimental data. However, one may note that the peaks at $\pm 4$ meV are sharper in the experimental spectrum than in the calculated curve. On the basis of the available data we cannot judge whether this discrepancy results from the uncertainties in the DOS data, or indicates that expression (3) does not precisely describe the multiphonon contribution of scattering.

The calculated probability of inelastic scattering has a peak at zero relative energy. As seen from Eq. (2), the elastic contribution to the scattering probability ($n=0$) is not taken into account in the calculations. The energy spectrum of the single-phonon scattering also does not contain the central peak. Thus, the central peak of the calculated scattering probability results only from the multiphonon scattering. In order to illustrate this point, we show in Fig. 2 the added contributions of the single- and two-phonon scattering (dotted line 1 + 2). This curve has the same peaks at $\pm 4$ meV as the single-phonon contribution, but contains also the peak at the center. Apparently, it arises from the convolution of the energy spectrum of the single-phonon scattering with itself, and accounts for the particular case of the two-phonon scattering, which is accompanied by the simultaneous creation and annihilation of the phonons with approximately the same energy. This inelastic contribution to the central peak was not previously noted in the energy spectra of nuclear inelastic absorption,[3–7] but here it is pronounced due to the sharp peak in low-energy region of the density of phonon states of $\beta$-Sn.

The significant contribution of inelastic scattering to the central peak does not allow us to subtract the elastic contribution of scattering according to the conventional procedure[4–7] and to calculate the Lamb-Mössbauer factor from the area of the inelastic part of the energy spectrum. Instead of this, we have determined the lower and the upper margins of $f_{\rm LM}$, considering the central peak to be either completely inelastic or elastic, respectively. This gives the following range for the Lamb-Mössbauer factor: $0.031(3) \leqslant f_{\rm LM} \leqslant 0.053(5)$. Comparing this estimation with the value obtained from forward nuclear scattering $f_{\rm LM}=0.042(2)$ (Ref. 25) we conclude that the central peak in the experimental data consists of approximately equal contributions of elastic and inelastic scattering.

In summary, we have measured the energy dependence of nuclear inelastic scattering of the 23.8795-keV x rays by $\beta$-Sn through the radiative channel of nuclear deexcitation with sub-meV energy resolution. The scattering was incoherent over the various nuclei and averaged over phonon momentum transfer. We suppose that in this approximation the energy dependence of inelastic scattering should be close to that of inelastic absorption. Comparison of the experimental spectrum of inelastic scattering with the calculated spectrum of inelastic absorption shows in general good agreement, however, the peaks of the experimental spectrum are somewhat sharper. This can hardly be attributed to the experimental uncertainty because any systematic error of the experiment (mechanical instability, thermal drift, etc.) would work in the opposite direction. One possible reason of the discrepancy may be the uncertainty of the data on the density of phonon states. On the other hand, it also may arise from the inadequate theoretical description of the multiphonon contribution of inelastic scattering by Eq. (3). In this respect we note that there is an urgent need of a complete theoretical description of nuclear inelastic scattering via the radiative channel of nuclear decay.

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