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Technical notes

On the line-shape analysis of Compton profiles and its application to neutron scattering

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ABSTRACT

Analytical properties of Compton profiles are used in order to simplify the analysis of neutron Compton scattering experiments. In particular, the possibility to fit the difference of Compton profiles is discussed as a way to greatly decrease the level of complexity of the data treatment, making the analysis easier, faster and more robust. In the context of the novel method proposed, two mathematical models describing the shapes of differenced Compton profiles are discussed: the simple Gaussian approximation for harmonic and isotropic local potential, and an analytical Gauss–Hermite expansion for an anharmonic or anisotropic potential. The method is applied to data collected by VESUVIO spectrometer at ISIS neutron and muon pulsed source (UK) on Copper and Aluminium samples at ambient and low temperatures.

1. Introduction

Deep inelastic neutron scattering (DINS), also acronymed as neutron Compton scattering (NCS), is a rare yet powerful technique for the investigation of nuclear quantum effects in condensed matter systems and molecules [1,2], with important ramifications for research in condensed matter physics [3,4], chemistry [5] and materials science [6]. NCS experiments are routinely performed using VESUVIO spectrometer [7] at ISIS neutron and muon pulsed source (UK). In this technique, very high energy, ΔE (tens-to-hundreds of electron-volts), and momentum, $\vec{q}$ (tens of inverse Angstroms), transfers are obtained, owing to the use of epithermal neutrons. A unique feature of the NCS is that experimental spectra are mass-resolved, i.e., composed of independent peaks for each element and isotope in the sample. Each peak is broadened because of the motion of the nucleus in the surrounding local potential due to nuclear zero-point and thermal energies. This broadening is the ultimate observable that makes NCS a unique technique and VESUVIO a unique instrument [8]. Despite clear advantage of the NCS technique over other neutron spectroscopic techniques with respect to mass-selective quantitation and qualitative description of nuclear dynamics, much of the work over recent years has focused on NCS data treatment schemes [1]. Towards this end, the following observation is in order. Usually, a raw NCS spectrum needs to be corrected for a series of environmental and sample-induced signals, thus adding a great deal of complexity to the data treatment [9]. However, a substantial part of the aforementioned corrections is independent or quite robust against changes of the Compton profile broadening (e.g., due to temperature and pressure changes, and phase transitions). As such, those corrections can be eliminated using data treatment schemes involving signal differencing. This simple idea has motivated the work presented here.

This work is organized as follows. In Section 2 a few properties of the Compton profiles are summarised and the suggested procedure for the data analysis is described. In Section 3 experimental data treatment employing the standard procedure is compared with the novel scheme, and finally, in Sections 4 and 5 the implementation of the proposed data treatment scheme is discussed in case of the NCS data analysis from experiments on flat Aluminium and Copper samples.

2. Gaussian Compton profiles

In the case of a nucleus in a harmonic and isotropic potential, its momentum distribution can be expressed as a Gaussian function

$$n(p_x, p_y, p_z) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left(-\frac{p^2}{2\sigma^2}\right) \tag{1}$$

with an explicit dependence on the modulus of the momentum vector only, $p = |(p_x, p_y, p_z)|$. The value of σ is related to the atom-projected vibrational density of states, $g_M(E)$, of the system

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through the equation

$$\sigma^2 = M \int_0^\infty \frac{E}{2} g_M(E) \coth\left(\frac{E}{2k_B T}\right) dE \tag{2}$$

with M the mass of the nucleus and k_B the Boltzmann constant. It is worth noting that, via Eq. (2), σ is a temperature-dependent observable.

In a NCS experiment, the projection $J(y)$ of $n(p_x, p_y, p_z)$ on the direction of the momentum transfer q is measured, where $y = \vec{p} \cdot \hat{q}$ and

$$J(y) = \int n(y, \vec{p_\perp})\, d\vec{p_\perp} = \frac{1}{\sqrt{2\pi}\sigma} \exp\left(-\frac{y^2}{2\sigma^2}\right). \tag{3}$$

As a consequence of energy and momentum conservation during the collision of an epithermal neutron with a nucleus of mass M, the y variable can also be expressed as [10,11]:

$$y_M = \frac{M}{q}\left(\Delta E - \frac{q^2}{2\,M}\right). \tag{4}$$

Thus, y_M combines the energy and momentum transfers in a scaling variable [12] that follows the recoil line of a nucleus.

A great deal of NCS experiments is concerned with tracing changes in Compton profiles induced by changes in the local environment (e.g., phase transitions) or external stimuli (e.g., temperature). Whenever the difference δ between the final and initial values of σ, say σ_2 and σ_1, is small compared to their average, the difference of NCPs can be expressed as a Taylor expansion in powers of δ

$$\mathcal{D}(y, \sigma, \delta) := J(y, \sigma+\delta) - J(y, \sigma) = \sum_{k=1}^{\infty} \frac{\delta^k}{k!}\left(\frac{\partial^k J(y, \sigma+\delta)}{\partial \sigma^k}\right)_{\delta=0}$$

$$= \sum_{k=1}^{\infty} \frac{\delta^k}{k!}\left(\frac{\partial^k J(y, \sigma)}{\partial \sigma^k}\right) \simeq \sum_{k=1}^{l} \frac{\delta^k}{k!}\left(\frac{\partial^k J(y, \sigma)}{\partial \sigma^k}\right) = \mathcal{D}_l(y, \sigma, \delta). \tag{5}$$

While $\mathcal{D}(y, \sigma, \delta)$ is a series defined by considering all the term in the Taylor expansion, $\mathcal{D}_l(y, \sigma, \delta)$ is a partial sum truncated at a certain term $k = l$, provided that any further term is negligible. In what follows, we used the truncated expansion with $l = 2$ that can be expressed in series of the dimensionless ratio δ/σ

$$\mathcal{D}_2(y, \sigma, \delta) = J(y, \sigma)\left[\left(\frac{y^2}{\sigma^2} - 1\right)\frac{\delta}{\sigma} + \left(\frac{y^4}{\sigma^4} - 5\frac{y^2}{\sigma^2} + 2\right)\frac{\delta^2}{\sigma^2}\right]. \tag{6}$$

Moreover, we note that Eq. (6) can be expressed as combination of Hermite polynomials $H_n(x)$ via the substitutions $x = \frac{y}{\sqrt{2}\sigma}$, $(y^2/\sigma^2 - 1) = H_2(x)/2$ and $(y^4/\sigma^4 - 5y^2/\sigma^2 + 2) = H_4(x)/4 + H_2(x)/2$.

2.1. Non-Gaussian Compton profiles

The generalization of Gaussian Compton profiles that takes into account the effects of anharmonicity and anisotropy on the local potential is obtained in a model-independent way by employing the Gauss–Hermite (GH) expansion [13]

$$J_A(y) = \frac{\exp\left(-\frac{y^2}{2\sigma^2}\right)}{\sqrt{2\pi}\sigma} \sum_n \frac{c_{2n}}{n!2^{2n}} H_{2n}\left(\frac{y}{\sqrt{2}\sigma}\right) \tag{7}$$

with $c_2 \equiv 0$ as the consequence of the enforcement of the first-order sum rule [1]. En passant, we note that in case of a disordered material and anisotropy of the local potential experienced by the nucleus, a spherically averaged multivariate Gaussian distribution, defined by the parameters σ_x, σ_y and σ_z (the standard deviations of the nuclear momentum distribution along the x, y, and z directions in space), is expressed as a GH expansion with $\sigma^2 = (\sigma_x^2 + \sigma_y^2 + \sigma_z^2)/3$ and

$$c_4 = \frac{2}{15}\left[\frac{\sigma_x^4 + \sigma_y^4 + \sigma_z^4}{\sigma^4} - 3\right]$$

$$c_6 = \frac{8}{315}\left[\frac{\sigma_x^6 + \sigma_y^6 + \sigma_z^6 + 6\sigma_x^2\sigma_y^2\sigma_z^2}{\sigma^6} - 9\right]. \tag{8}$$

With this in mind, one can argue that for both anharmonic and anisotropic systems, the main correction to Gaussian line-shapes will be of the order of c_4. When $\mathcal{D}_2(y, \sigma, \delta)$ is applied, the $H_4(x)$ term contains contributions from the difference of c_4 terms, $\Delta c_4/32$ (Eq. (7)), and the contribution $\delta^2/(4\sigma^2)$ (Eq. (6)). The former of these contributions is usually (including the case described in this work) an order of magnitude smaller than the latter and therefore can be neglected.

3. Experiment

The experiment was performed using VESUVIO spectrometer at ISIS. Flat Aluminium alloy and Copper samples with a scattering power of 2% and 10% respectively were used. The spectra were collected through the backscattering detectors in the standard double difference acquisition mode [14]. Each measurement corresponded to an integrated synchrotron proton current of ca. 1.5 mAh. The samples were placed in a Closed Circuit Refrigerator (CCR) that allowed the measurement at 30 K and 300 K in the case of Al, and at 80 K and 270 K in the case of Cu. Gold resonant foils were used to fix the neutron final energy at 4.9 eV. Experiments were carefully designed in order to avoid multiple scattering in the samples and to minimize the influence of the environmental background. However, in case of the Al sample the possibility of impurities present in the sample material cannot be excluded as the Al grade 7075 alloy was used. This, however, presented an opportunity to test the influence of impurities in sample composition on the robustness of the proposed method.

The entire NCS data analysis was performed in the Mantid data treatment environment [15,16]. For each backscattering detector, an empty instrument spectrum (dominated by the CCR neutronic response) was subtracted from the collected sample spectrum. The spectra were then transformed into the y_M scaling variable domain of the appropriate mass M = Al or Cu. Following this, spectra for all detectors expressed in the respective y_M domains were added to form the focused Compton profile for mass M. The final spectra have been then normalised to unity to fulfil the sum rule for a probability distribution. Similarly, resolution functions for each detector were simulated in the y_M space and averaged. Within the Mantid software, it is also possible to define a fictitious average detector that can be used to define the average resolution function. Both approaches have been tested and compared to results from routines used in the past [17], and their consistency was checked. The experimental profiles for Al at T = 30 K and 300 K as well as the resolution profile are shown in Fig. 1.

Two methods, referred to as (1) and (2), were adopted:

1. *Method* 1: Each Compton profile was fitted through the GH expansion expressed using Eq. (7) numerically convoluted with the resolution function. The resulting parameters σ_1 and σ_2 (see Table 1) were then used to define the value of δ.
2. *Method* 2: The difference of the two profiles was taken and fitted with the expansion given by Eq. (5). The parameters σ_1 and δ, obtained through the fit of the difference spectrum shown in Fig. 2, were combined to obtain σ_2.