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APPLICATION OF MAXIMUM ENTROPY TO NEUTRON TUNNELING SPECTROSCOPY

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abstract

We demonstrate the maximum entropy method for the deconvolution of high resolution tunneling data acquired with a quasielastic spectrometer. Given a precise characterization of the instrument resolution function, a maximum entropy analysis of lutidine data obtained with the IRIS spectrometer at ISIS results in an effective factor of three improvement in resolution.

Keywords: Maximum Entropy, Quasielastic Neutron Scattering, Inelastic Neutron Scattering

INTRODUCTION:

The maximum entropy method (MaxEnt) [1,2] is an optimal technique for reconstructing positive images from noisy, incomplete and broadened data. It represents an important advance in data treatment and statistical inference, which has been widely used in fields such as radio astronomy, medical tomography, forensic deconvolution, etc. However, its applications in neutron scattering are only beginning [3]. The data in a neutron scattering experiment represent a convolution of the scattering law of the sample with the resolution function of the instrument, which in general varies with instrumental parameters such as energy transfer, to which are added a background and a statistical uncertainty. This results in the blurring of the data and raises problems in interpretation. In most data analysis approaches the scattering law is represented by a model, and the parameters of the model are extracted by least squares fitting to the data of the model convoluted with the measured resolution function. MaxEnt offers a method of inferring the scattering law without recourse to any specific model, and the reconstruction shows finer detail with minimal noise artefacts.

In this paper, we consider the application of the maximum entropy method to neutron tunneling spectroscopy data. We demonstrate that significant improvements in resolution up to a factor of three can be reliably achieved, and that statistical errors on inferred features in the spectrum can be provided.

THEORY:

The data in a neutron scattering experiment can be represented mathematically by

$$D = R \otimes f + b \pm \sigma \quad , \quad (1)$$

where D are the data, R is the instrument response function, f is the scattering law, b is the background, σ is the noise, and $\otimes$ represents a convolution. Given the data D and the resolution function R , the goal of any neutron scattering experiment is to infer the scattering law f . In general, this is an ill-posed problem because an infinity of f 's will fit the data. Our goal is to determine the most probable f as well as to estimate its reliability in the form of error bars.

An important mathematical property of f is that it is a positive additive distribution function.

Then, a variety of probability theory and logical statistical inference arguments lead to a variational principle for the most probable f . One maximizes

$$Q = \alpha S - \chi^2/2 \quad . \quad (2)$$

The variational principle, Eq. (2), represents a balance between the data embodied in χ^2 , which is the usual statistic for the quality of a fit, and one's prior knowledge of the scattering law before the experiment began embodied in the Shannon–Jaynes information theory “entropy”

$$S = \sum_i [f_i - m_i - f_i \ln(f_i/m_i)] \quad . \quad (3)$$

The parameter α is a Lagrange multiplier to be determined self-consistently from the data. Here m_i is termed the default model for the scattering law, as it is the result in the absence of data. If before the experiment began we are ignorant about the scattering law, the default model is usually chosen to be flat. Further arguments lead to estimates of statistical errors on integrated features in f . This procedure is implemented using a commercial MaxEnt image processing code, MEMSYS3 [4].

APPLICATION:

We have applied this technique to data obtained on the IRIS spectrometer at the ISIS pulsed neutron source using a sample of 2, 6 dimethyl pyridine (lutidine). This system is an appropriate subject of study since it exhibits a complex overlapping tunneling spectrum. Two methyl groups are linked to an aromatic pyridine ring, have relatively low potential barrier for rotation, and exhibit tunneling excitations in the 10 to 200 μ eV energy transfer range.

Our measurements used pyrolytic graphite 002 crystal analyzers with a full-width-half-maximum (FWHM) of 15 μ eV and mica 004 crystal analyzers providing a FWHM of 4.2 μ eV. The instrumental resolution function was determined for zero energy transfer from the measured vanadium spectrum by a numerical Wiener filter to reduce statistical noise. The measured variation of the resolution with energy transfer was then accounted for by varying the FWHM of this distribution over the entire energy transfer range of the spectrometer.

Figure 1 shows the experimental spectrum for lutidine obtained with the PG 002 analyzer

(solid line) along with the instrumental resolution function (dashed line). A linear background and an elastic scattering component of about 80%, determined by minimizing the remaining scattering at zero energy transfer, were subtracted from the data before processing with the MaxEnt code. The MaxEnt image shown in Fig. 2 has considerably improved resolution of about $4 \mu\text{eV}$ and reveals a multiplet structure. Note the rough symmetry between energy loss and energy gain. The MaxEnt images represent the peak of the probability distribution of possible scattering laws which fit the data, and MaxEnt also provides statistical errors on these curves. Also shown in this figure are the fractions of the total intensity provided by integrated features in the image, where the vertical lines represent statistical error estimates and the horizontal lines represent the energy transfer region integrated over.

Figure 3 presents the experimental spectrum and resolution function obtained with the Mica 004 analyzer. Comparison with Fig. 2 shows that the multiplet structure obtained by the MaxEnt analysis of the PG 002 data was reliable. These mica data were then corrected for background and elastic scattering component in the same way as above, and processed with the MaxEnt code. The MaxEnt image of the mica 004 confirms the integrated fractions calculated for PG 002. A portion of the mica 004 MaxEnt image in Figure 4 shows that each of the lines observed by MaxEnt analysis of the PG002 data is broken up into a further multiplet (probably triplet) structure. Again integrated intensities and associated statistical errors are also shown. The effective resolution obtained by MaxEnt is about $1.5 \mu\text{eV}$. To test our MaxEnt results for mica 004, we can compare directly to raw data (dashed line) on lutidine obtained at $1 \mu\text{eV}$ resolution over a limited energy transfer range using the back scattering spectrometers at ILL and KFA Julich with offset monochromators. This clearly shows the existence of corresponding three peaked structure and relative intensities in the scattering law in the energy range from $8\text{--}22 \mu\text{eV}$, although there does appear to be some shift in energy scale. A more careful resolution function characterization and data analysis for the backscattering spectrometer is required to determine if the small differences between the our MaxEnt images and the backscattering raw data are significant.

DISCUSSION AND CONCLUSIONS:

Although the maximum entropy method has been widely used in other fields of research, there have been only limited applications in neutron scattering [3]. Nevertheless, neutron scattering, being an intensity and resolution limited technique, is well suited to the application of MaxEnt. Our results provide confidence in the application of maximum entropy data analysis methods to neutron quasielastic and tunneling spectroscopy. Reliable improvements in resolution by a factor of three were achieved by software. This conclusion is established by comparison of our MaxEnt results directly with raw data from higher resolution experiments. Comparable resolution would be much more expensive to achieve by improvements to spectrometer and neutron source hardware. The full confidence of the user community will only be achieved after further experience has been gained, but the minimum benefit must at least be that such data analysis methods are a valid aid in pointing the need for higher resolution measurements on a given sample. Further, it has been previously demonstrated [5,6] that, when using maximum entropy methods, an asymmetric resolution function yields greater information about the scattering law than a symmetric resolution function of the same intensity and width. More generally, modern data analysis software such as MaxEnt may have a role [7] in optimizing the hardware of pulsed neutron source and instrument design.

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FIGURE CAPTIONS:

- Fig. 1 The solid line is the raw data for lutidine obtained with a pyrolytic graphite 002 crystal analyzer on the IRIS spectrometer at the ISIS pulsed neutron facility. The dashed line is the resolution function of the spectrometer for zero energy transfer deduced from a vanadium spectrum.
- Fig. 2 The solid line is the MaxEnt image of the scattering law for lutidine obtained from the data in Fig. 1. The crosses represent the integrated intensities of features in the scattering law, with vertical bars representing statistical errors and horizontal bars indicating the energy transfer region integrated over.
- Fig. 3 The solid line is the raw data for lutidine obtained with a mica 004 crystal analyzer. The dashed line is the corresponding instrument resolution function.
- Fig. 4 The solid line is a portion of the MaxEnt image of the scattering law for lutidine obtained from the data in Fig. 3. The crosses again represent integrated intensities of features in the scattering law, as in Fig. 2. The dashed line is raw data for lutidine obtained at approximately $1 \mu\text{eV}$ resolution with the backscattering spectrometer at KFA Julich.

PG 002 Lutidine

Lutidine PG002

Lutidine Mica004

Lutidine Mica004

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