HERMES – a GUI-based software tool for pre-processing of X-ray absorption spectroscopy data from laboratory Rowland circle spectrometers

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HERMES, a graphical user interface software tool, is presented, for pre-processing X-ray absorption spectroscopy (XAS) data from laboratory Rowland circle spectrometers, to meet the data handling needs of a growing community of practice. HERMES enables laboratory XAS data to be displayed for quality assessment, merging of data sets, polynomial fitting of smoothly varying data, and correction of data to the true energy scale and for dead-time and leakage effects. The software is written in Java 15 programming language, and runs on major computer operating systems, with graphics implementation using the JFreeChart toolkit. HERMES is freely available and distributed under an open source licence.

1. Introduction

The renaissance of laboratory X-ray absorption spectroscopy (XAS) instrumentation is revolutionizing access to, and uptake of, this technique across the physical sciences and engineering, enabling application of this technique without the need for access to a synchrotron light source (Blachucki et al., 2019; Honkanen et al., 2019; Jahrman et al., 2019a; Schlesiger et al., 2015; Malzer et al., 2018; Mortensen et al., 2016; Németh et al., 2016; Seidler et al., 2014, 2016; Zeeshan et al., 2019). In particular, commercial and user-built instrumentation based on a Rowland circle spectrometer with spherically bent crystal analyzers (SBCAs) used in the Johann configuration, and utilizing an energy-dispersive X-ray (EDX) detector, are gaining adoption, both as laboratory and regional facilities with a role complementary to, and symbiotic with, use of synchrotron radiation sources (Ditter et al., 2019). Already, this spectrometer design has been exploited to address a wide range of problems in nuclear, functional, catalysis and geological materials, including operando studies (Bés et al., 2018; Bi et al., 2019a,b; Jahrman et al., 2019b; Kuai et al., 2018; Lutz & Fittschen, 2020; Mottram et al., 2020a,b,c; Moya-Cancino et al., 2019; Nolis et al., 2020; Sun et al., 2021; Wittkowski et al., 2021; Zimmermann et al., 2021).

The rapid uptake of the Rowland circle XAS spectrometer is driving an expansion of the user base for the technique, who require tools to integrate and pre-process data for further analysis. This need arises because, typically, several scans are acquired with \( I_t \) and without \( I_0 \) the sample, to compute the absorption; whereas, at a synchrotron source, \( I_0 \) and \( I_t \) would be acquired simultaneously. Recently, Bés et al. (2018, 2021) have demonstrated an elegant procedure for acquisition of \( I_0 \) and \( I_t \) simultaneously, by exploiting SBCA harmonics,
although this is not always applicable. Raw data also need
to be quality assessed, dead-time corrected, appropriately
merged, and corrected to the true energy scale and for leakage
effects. Although data processing codes have been developed
within Jupyter notebook and Mathematica environments, they
require some familiarity with coding to use efficiently and
troubleshoot problems. However, this may not necessarily be
within the grasp of a broad user base, for whom XAS is a
supplementary or infrequent analytical tool. We therefore
developed HERMES as software based on an intuitive
graphical user interface (GUI), to enable rapid and robust
pre-processing of laboratory XAS data from Rowland circle
spectrometers, for import into software such as ATHENA for
further analysis (Ravel & Newville, 2005). Subsequently, the
HERMES backronym was later coined – Handy Energy
Recalibration and Mu Evaluation Software.

HERMES is free to download and distributed under an
Open Source Initiative approved MIT Licence (Open Source
Initiative, 2021), and documentation is distributed under a
Creative Commons CC BY 4.0 licence (Creative Commons,
2021), enabling users to adapt and modify the source code to
better meet their needs, as may be desirable. HERMES is
written in Java 15 and compiled and tested to work on the
common laboratory Microsoft Windows and Macintosh
OSX platforms. Plotting graphics are implemented using
JFreeChart (JFreechart, 2021). Java was chosen for imple-
m entation due to its strong object orientation and type safety.

2. Features of HERMES

HERMES is a program for pre-processing of transmission
mode laboratory X-ray absorption spectroscopy data, from
Rowland circle spectrometers, to produce input files suitable
for further analysis using software such as ATHENA. It
provides the following functionality:

(i) Dead-time correction of raw data.
(ii) Plotting and comparison of multiple \( I_0 \) and \( I_t \) data.
(iii) Fitting of a polynomial to suitable \( I_0 \) and \( I_t \) data.
(iv) Merging of several \( I_0 \) and \( I_t \) data sets.
(v) Evaluation of absorption \( \mu(E) \) from \( I_0 \) and \( I_t \) data.
(vi) Correction of data for leakage effects.
(vii) Recalibration of data energy scale.

HERMES uses a logical workflow to guide the user through
the steps of data pre-processing. The dashboard has a simple
and intuitive interface, with four data workspaces and
processing tools, shown in Fig. 1. The user specifies the
Measurement Type to be loaded or processed using a drop-
down menu (\( I_0 \), \( I_t \), \( I_0 \) leakage, \( I_t \) leakage). The user is required
to select appropriate columns for energy, theta, detector raw
counts, detector input count rate (ICR), and detector output
count rate (OCR). A first-order dead-time correction is
applied to raw detector counts, valid for dead-time up to 50% (XIA LLX, 2009). The plotting function supports enlargement
of regions of interest and data may be displayed individually,
overlaid or offset (by a user-specified amount), as shown in
Fig. 2. An \( n \)th-order polynomial (where \( n \) is user specified)
may be fitted to any appropriate and smoothly varying data set
selected (i.e. \( I_0 \), \( I_{0,\text{lk}} \), \( I_{t,\text{lk}} \); where \( \text{lk} \) denotes a leakage
measurement, as discussed below).

After the user has completed plotting, assessment, merging
and polynomial fitting of raw data in the first tab of the
dashboard, the workflow progresses naturally to the second
tab where the user may evaluate and inspect the absorption
spectrum, \( \mu(E) \). If the specimen is sufficiently thick and/or
attenuating it may be desirable to correct the computed
absorption data for ‘leakage effects’ (Stern & Kim, 1981; Mottram
et al., 2020a) which may arise from contamination of
the transmission data by harmonics, stray scatter and the low
energy tail of the monochromator function; this correction is
effected by a tick box. In the Rowland circle geometry, it
may be necessary to measure transmission data with a large
detector offset (\( I_{0,\text{lk}} \) and \( I_{t,\text{lk}} \)), to correct the absorption
data for distortion arising from leakage effects according
to equation (1),

\[
\mu(E) = \int \frac{I_0 - I_t}{I_0} \, dE
\]

Figure 1
HERMES dashboard showing the Data Input tab, with four file
workspaces, measurement type dropdown menu, data processing and
plotting tools.

Figure 2
Example of the HERMES plotting window, showing a stack plot of \( I_t \) data
at the U \( L_3 \)-edge, separated by a user-specified offset.

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Where leakage effects are not important, then \( I_{0,\text{IR}} = I_{\text{T},\text{IR}} = 0 \). The fitted polynomials, without Poisson noise, may be used to evaluate the absorption, if desirable and appropriate.

Evaluation of absorption requires the user to specify one data set each of \( I_0 \) and \( I_t \) (plus \( I_{0,\text{IR}} \) and \( I_{\text{T},\text{IR}} \) if required), which may be raw, merged or polynomial fitted data. The absorption is displayed as a function of energy and theta. The computed absorption, merged, and polynomial fit data are written as text files, at the point of computation, together with a list file to enable data provenance and curation.

The third tab in the HERMES workflow enables calibration of the absolute energy scale of absorption spectra evaluated in the previous workspace. In general, the absolute energy scale of laboratory XAS data requires calibration using a suitable reference material, for which there are calibrated literature, open source or user-acquired XAS data. Rowland circle spectrometers function on an angle-dispersive principle to maintain the required focusing condition. Steps in energy or \( k \)-space, within user-specified ranges, determine the required steps in theta space according to the Bragg Law,

\[
\mu = \ln \left( \frac{I_0 - I_{0,\text{IR}}}{I_t - I_{\text{T},\text{IR}}} \right),
\]

where \( E_{\text{mono}} \) is the characteristic backscatter energy of the SBCA. HERMES determines \( E_{\text{mono}} \) from a user-specified absorption spectrum, within the workspace. Since the relationship between energy and theta is non-linear, it is necessary to apply the correction in theta space and then recalibrate the energy scale. HERMES requires the user to specify the observed and true energy (\( E_{\text{obs}} \) and \( E_{\text{true}} \)) of some feature in the absorption spectrum, such as the maximum in the first derivative of \( \mu (E) \). From equation (2), the corresponding \( \theta_{\text{obs}} \) and \( \theta_{\text{true}} \) are determined, the difference between these values being the \( \Delta \theta_{\text{shift}} \) required to align the absorption spectrum in theta space. The absorption spectrum is calibrated by applying the theta shift to the observed data and calculation of the true energy, from equation (2). The original and calibrated absorption spectrum are plotted in energy and theta space for inspection, post calibration. The calibrated absorption spectrum is written as a text file, at the point of computation, with both original and calibrated energy and theta scales, together with a list file detailing the key calibration parameters.

A comprehensive user guide and video tutorial are provided to support use of the software.

### 3. Conclusions

We have presented the HERMES software for pre-processing of laboratory X-ray absorption spectroscopy data from Rowland circle spectrometers. A simple GUI and intuitive workflow enable integration, correction and calibration of raw data to output data files suitable for further analysis in software such as ATHENA. This software contributes to meeting the need of a rapidly growing community of practitioners, who require freely available tools for rapid and robust pre-processing of laboratory XAS data.

### 4. Resources

A project page for HERMES exists at https://github.com/xasheffield/hermes. HERMES is available as an executable .jar file (Windows, MacOS, requiring an existing installation of Java) or as a .exe file with the necessary Java Runtime bundled (Windows only), and both are freely available at the link above, as well as a complete user manual; a tutorial video is available (see the supporting information).

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