Adaptive Design of Experiment for X-Ray Magnetic Circular Dichroism Spectroscopy by Machine Learning

Machine learning (ML) is applied to X-ray magnetic circular dichroism (XMCD) spectroscopy to improve its efficiency. Adaptive design of an XMCD experiment by Gaussian process modeling successfully reduced the total energy points for obtaining an XMCD spectrum. An orbital magnetic moment is evaluated from an ML-predicted XMCD spectrum with the required accuracy. This methodology paves the way to improving the efficiency of various quantum beam measurements.

Machine learning (ML) is a field of artificial intelligence research. The primary objective of ML is to give computers the ability to learn like humans. Machine learning techniques can find patterns in data and are used for the classification, regression, and clustering of patterns. Recently, materials informatics, which is materials science using ML techniques, is attracting attention for the efficient discovery and development of novel materials. At the same time, metro-informatics, namely measurement techniques involving ML techniques, is necessary for accelerating materials informatics.

X-ray magnetic circular dichroism (XMCD) spectroscopy is an important experimental technique for investigating magnetic materials such as permanent magnet materials, magnetic recording, and spintronic materials. The spins and orbital magnetic moments of a specific element can be evaluated by XMCD spectroscopy using magneto-optical sum rules. In this study, we applied an ML technique to an XMCD spectroscopy experiment to improve its efficiency [1].

Gaussian process (GP) modeling has been used in meteorology and geology for kriging. An XMCD spectrum is represented as a nonlinear function of X-ray energy. A GP model is a generalized linear model that can approximate such nonlinear spectral shapes by linear regression in feature space. The GP model predicts a spectrum by learning experimental data points, namely photon energy versus intensity. Moreover, both the expectation and variance of the prediction can be evaluated.

We used an experimental XMCD spectrum to assess the applicability of GP modeling. Sm $M_{4,5}$ XMCD and XAS spectra of $SmCo_5$, a typical permanent-magnet material, were measured using a scanning transmission X-ray microscope (STXM) at BL-13A [2]. Details of the STXM experiment are described in the literature [3].

Figure 1 shows a flowchart of the adaptive design of the experiment for XMCD spectroscopy. First, initial data points are sampled to obtain an experimental spectrum as the training data. Next, a spectrum is predicted by GP modeling fitted to the training data. Then, magnetic moments are evaluated from the predicted spectra. The experiment is stopped if the values of the magnetic moments satisfy the convergence criterion. Otherwise, new data points are sampled, and the spectrum is predicted again. We examined three methods of selecting new sampling data points: (1) sample the data point with maximum variance (max. var.) of the predicted spectrum, (2) random sampling, and (3) random sampling weighted with variance (weighted sampling).

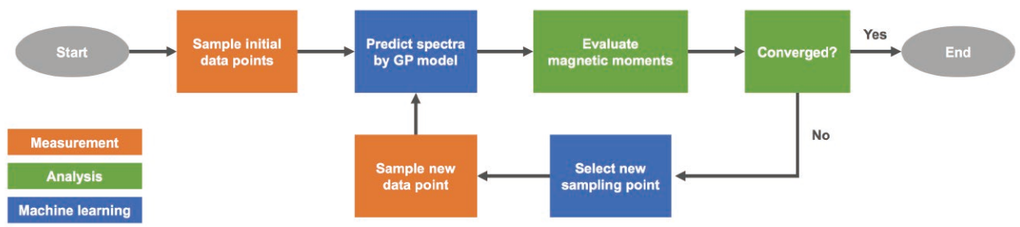

Figure 1: Flowchart of an adaptive design of an experiment for XMCD spectroscopy.

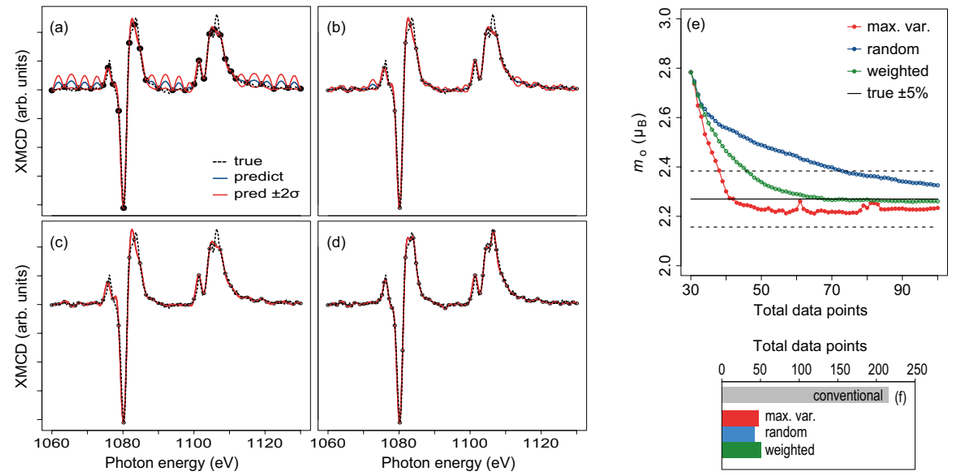

Figure 2: (a)–(d) XMCD spectra predicted by the GP model. Sm $M_{4,5}$ XMCD spectra for **(a)** 30, **(b)** 40, **(c)** 50, and **(d)** 70 data points in total. The black dashed and blue solid curves represent the true (experimental) spectrum and the spectrum predicted by the GP model, respectively. The open circles represent observed data points. The red solid curves indicate the variance with a 95% confidence interval (±2σ) of the predicted spectrum. **(e)** Orbital magnetic moment m_o versus total data points. The red, blue, and green markers represent the methods for data-point sampling: maximum variance (max. var.), random, and random sampling weighted by variance, respectively. The black solid and dashed lines represent the reference value and 5% deviations, respectively. **(f)** The total number of data points needed for convergence of m_o values.

Figure 2 shows the results of the adaptive design of the experiment for XMCD spectroscopy. Typical XMCD spectra predicted by the GP model are shown in **Figs. 2(a)–(d)**. The initial 30 data points were extracted from the experimental XMCD spectrum. Variances in the predicted spectra (red solid curves) became significant among the observed data points. By increasing the number of observed data points, the total variance of the predicted spectra decreased, and the spectral shape of the predicted spectrum became similar to that of the experimental (true) spectrum. **Figure 2(e)** shows the orbital magnetic moment m_o evaluated from the predicted spectrum plotted as a function of the total number of data points with different sampling methods. True values for the magnetic moments and the ±5% errors are indicated by black solid and dashed lines, respectively. The orbital magnetic moment converged to the true value at around 40 total data points with maximum variance sampling. Random sampling showed poor convergence to the true value even with 100 data points. Weighted sampling behaved halfway between maximum variance sampling and random sampling, and showed good convergence to the true value. **Figure 2(f)** shows the total number of data points for the various sampling methods to satisfy the convergence criterion. All sampling methods satisfied the convergence criterion at around 50 points, but the random sampling showed convergence with a large error from the true value. Thus the maximum variance sampling with fast convergence and small error is the best way to select the energy point to be measured.

In conclusion, we demonstrated the adaptive design of an experiment for XMCD spectroscopy with GP modeling. GP was found to predict the nonlinear spectral shapes of the XMCD spectrum successfully. Magnetic moments can be evaluated from the predicted spectra with the required level of accuracy. The present method reduces the total number of data points for an XMCD spectroscopy experiment. This method has potential applicability to various quantum beam measurements.

REFERENCES

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