ANDiE the Autonomous Neutron Diffraction Explorer

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We developed the Autonomous Neutron Diffraction Explorer (ANDiE) to autonomously perform neutron diffraction measurements to discover the magnetic ordering behavior in a material. Neutron diffraction is one of the few techniques that can directly probe the magnetic ordering of the atoms in a material. As such beamtime at neutron diffraction facilities is in high demand. ANDiE is able to increase the measurement efficiency for discovering the magnetic order parameter and ordering behavior by a factor of about 5, thus making efficient use of the beamtime.

The experiments to discover the magnetic order parameter and ordering behavior involve measuring the neutron diffraction patterns at many temperatures. Conventionally these measurements are scheduled *ad hoc* and take a significant amount of beamtime to perform. ANDiE, by contrast, uses Bayesian inference to determine the most informative temperatures at which to acquire diffraction patterns. ANDiE analyzes the powder neutron diffraction patterns, infers the temperature dependence, chooses the next temperature, and repeats until the magnetic transition behavior and magnetic order parameter is well known. The workflow is shown in Figure 1.

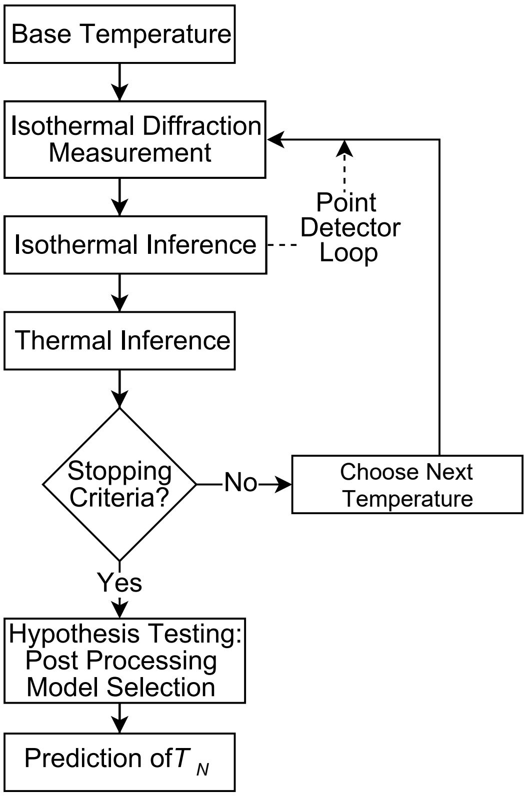

*Figure 1: A diagram of the algorithm used in the Autonomous Neutron Diffraction Explorer (ANDiE). ANDiE autonomously drives live neutron diffraction experiments to discover the Néel temperature (*TN*) and to perform subsequent hypothesis testing for the temperature dependence of magnetic structure. The solid lines show how ANDiE was implemented on the Wide-Angle Neutron Diffractometer (WAND2) at the HB-2C beamline at the Oak Ridge National Laboratory (ORNL) High Flux Isotope Reactor (HFIR), while the dashed line shows the additional active learning loop implemented on the BT-4 beamline at the National Institute of Standards and Technology (NIST) Center of Neutron Research (NCNR).*

To accomplish this task ANDiE uses several physics-informed constraints and models. For example, the magnetic transitions in materials can be hysteretic, so the experiments are performed by cooling to a base temperature then collecting data as the sample is warmed. Under this constraint, even popular active learning decision making schemes used to drive autonomous experiments would fail. Therefore, ANDiE uses a physics-based model to extrapolate – with quantified uncertainty – the temperature dependence of the magnetic contribution to the diffracted intensity to higher temperatures.

ANDiE then considers the predicted measurement uncertainty at those higher temperatures based on the predicted intensities and the Poissonian-like instrument uncertainty. The lowest temperature where the extrapolated uncertainty exceeds the measurement uncertainty by a threshold is the next temperature at which the neutron diffraction data is acquired. This threshold we call the Bravery Factor, as it controls how ambitiously ANDiE explores the search space. This data acquisition policy is illustrated in Figure 2.

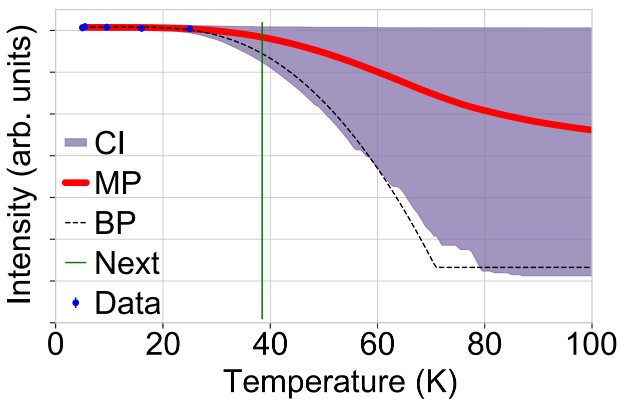

Figure 2: The temperature dependence of the magnetic component of the neutron diffraction intensity, for $Fe_{1.09}Te$ measured during an autonomous run using the $WAND^2$ instrument. The blue dots show the measured values with error bars. Additionally plotted: the confidence interval (CI) of the model, mean of the posterior (MP) distribution of the model, the model using the best (i.e., most likely) parameters (BP). The green vertical line shows the next temperature to measure at, where the model uncertainty relative to the estimated measurement uncertainty exceeds the Bravery Factor.

ANDiE was implemented both at the National Institute of Standards and Technology (NIST) Center of Neutron Research (NCNR) BT-4 Triple-Axis Spectrometer and on the Wide-Angle Neutron Diffractometer ($WAND^2$) at the HB-2C beamline at the Oak Ridge National Laboratory (ORNL) High Flux Isotope Reactor (HFIR) to drive autonomous experiments. As a case study, ANDiE was used at the $WAND^2$ instrument to discover the magnetic ordering behavior and transition temperature of MgO. In only 16 measurements, ANDiE successfully discovered the Ising-type behavior with a transition temperature at 120.81(56) K. In a further study, ANDiE was used to investigate the much less well known $Fe_{1.09}Te$ material. ANDiE autonomously discovered the much more difficult to study, abrupt, first-order transition behavior at 69.436(55) K. In both of these autonomous experiments ANDiE improved the measurement efficiency by a factor of about 5.

The full details of the work are available at: https://doi.org/10.1063/5.0082956. The notebooks and data acquired during the autonomous experiments with ANDiE are available at https://github.com/usnistgov/ANDiE-v1_0, and https://data.nist.gov/od/id/mds2-2449.

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