Supplementary Materials of the paper titled

"A dynamic Bayesian optimized active recommender system for curiosity-driven partially "Human-in-the-loop" automated experiments"

Arpan Biswas,[1,*] Yongtao Liu[1], Nicole Creange[1], Yu-Chen Liu[2], Stephen Jesse,[1] Jan-Chi Yang[2], Sergei Kalinin[3], Maxim Ziatdinov,[1,4] Rama Vasudevan[1,**]

[1] Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, TN 37831

[2]Department of Physics, National Cheng Kung University, Tainan 70101, Taiwan

[3]Department of Materials Science and Engineering, University of Tennessee, Knoxville

[4] Computational Sciences and Engineering Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831

Appendix A. Gaussian Process Model (GPM)

The general form of the GPM is as follows:

$$y(x) = x^T\beta + z(x) \tag{A.1}$$

where $x^T\beta$ is the Polynomial Regression model. The polynomial regression model captures the global trend of the data. $z(x)$ is a realization of a correlated Gaussian Process with mean $E[z(x)]$ and covariance $cov(x^i, x^j)$ functions defined as follows:

$$z(x) \sim GP\left(E[z(x)], cov(x^i, x^j)\right); \tag{A.2}$$

$$E[z(x)] = 0, cov(x^i, x^j) = \sigma^2 R(x^i, x^j) \tag{A.3}$$

$$R(x^i, x^j) = \exp\left(-0.5 * \sum_{m=1}^{d} \frac{\left(x_m^i - x_m^j\right)^2}{\theta_m^2}\right); \tag{A.4}$$

$$\theta_m = (\theta_1, \theta_2, \ldots., \theta_d)$$

where σ^2 is the overall variance parameter and $\theta_m$is the correlation length scale parameter in dimension m of d dimension of x. These are termed as the hyper-parameters of GP model. $R(x^i, x^j)$ is the spatial correlation function. In this paper, we have considered a Radial Basis function which is given by eqn. A.4. The objective is to estimate (by MLE) the hyper-parameters $\sigma,\ \theta_m$ which creates the surrogate model that best explains the training data $\boldsymbol{D_k}$ at iteration k.

After the GP model is fitted, the next task of the GP model is to predict at an arbitrary (unexplored) location drawn from the parameter space. Assume $\boldsymbol{D_k} = \{\boldsymbol{X_k}, \boldsymbol{Y}(\boldsymbol{X_k})\}$ is the prior information from previous evaluations or experiments from high fidelity models, and $\bar{\bar{x}}_{k+1} \in \bar{\bar{\boldsymbol{X}}}$ is a new design within the unexplored locations in the parameter space, $\bar{\bar{\boldsymbol{X}}}$. The predictive output distribution of x_{k+1}, given the posterior GP model, is given by eqn A.5.

$$P(\bar{\bar{y}}_{k+1}|\boldsymbol{D_k}, \bar{\bar{x}}_{k+1}, \sigma_k^2, \boldsymbol{\theta_k}) = \boldsymbol{N}(\mu(\bar{\bar{y}}_{k+1}(\bar{\bar{x}}_{k+1})), \sigma^2(\bar{\bar{y}}_{k+1}(\bar{\bar{x}}_{k+1}))) \quad \text{(A.5)}$$

where:

$$\mu\left(\bar{\bar{y}}_{k+1}(\bar{\bar{x}}_{k+1})\right) = \boldsymbol{cov_{k+1}^T} \boldsymbol{COV_k^{-1}} \boldsymbol{Y_k}; \quad \text{(A.6)}$$

$$\sigma^2\left(\bar{\bar{y}}_{k+1}(\bar{\bar{x}}_{k+1})\right) = cov(\bar{\bar{x}}_{k+1}, \bar{\bar{x}}_{k+1}) - \boldsymbol{cov_{k+1}^T} \boldsymbol{COV_k^{-1}} \boldsymbol{cov_{k+1}} \quad \text{(A.7)}$$

$\boldsymbol{COV_k}$ is the kernel matrix of already sampled designs $\boldsymbol{X_k}$ and $\boldsymbol{cov_{k+1}}$ is the covariance function of new design $\bar{\bar{x}}_{k+1}$ which is defined as follows:

$$\boldsymbol{COV_k} = \begin{bmatrix} cov(x_1, x_1) & \cdots & cov(x_1, x_k) \\ \vdots & \ddots & \vdots \\ cov(x_k, x_1) & \cdots & cov(x_k, x_k) \end{bmatrix}$$

$$\boldsymbol{cov_{k+1}} = [cov(\bar{\bar{x}}_{k+1}, x_1), cov(\bar{\bar{x}}_{k+1}, x_2), .., cov(\bar{\bar{x}}_{k+1}, x_k)]$$

Appendix B. Additional figures

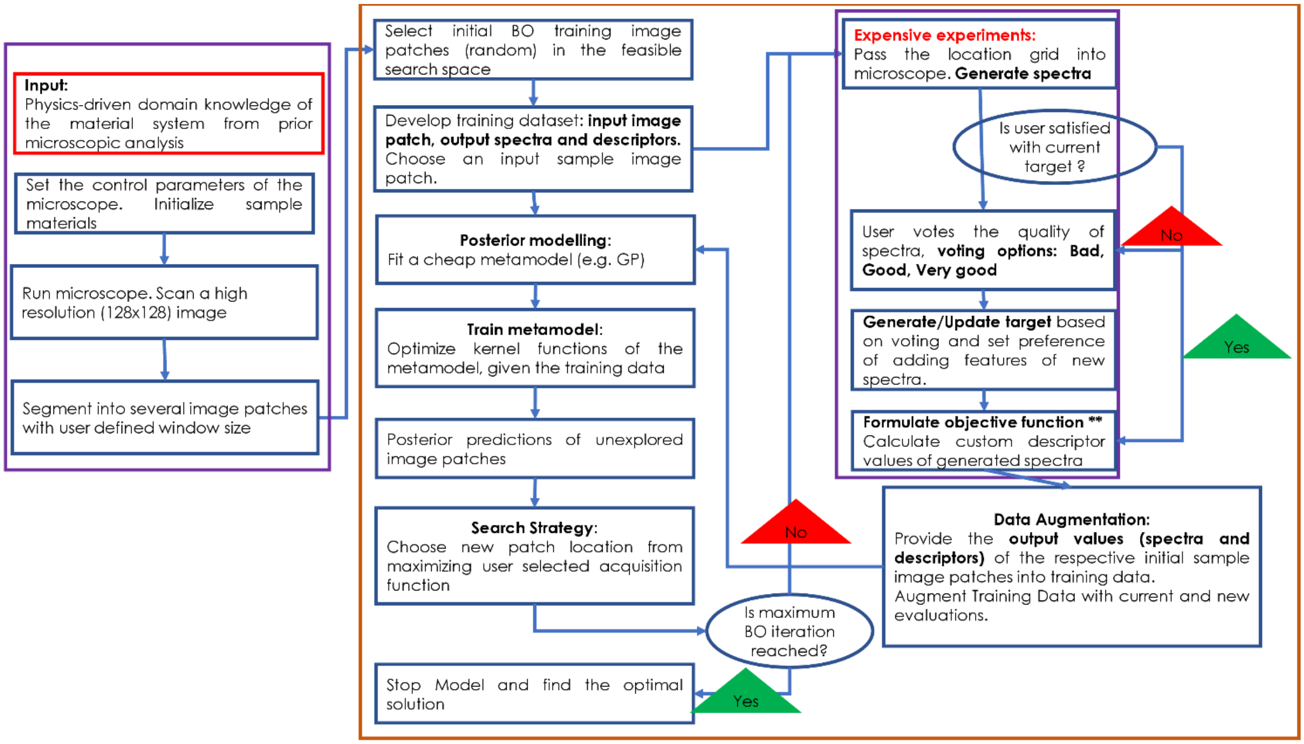

Figure B1. Flowchart of human augmented BOARS architecture

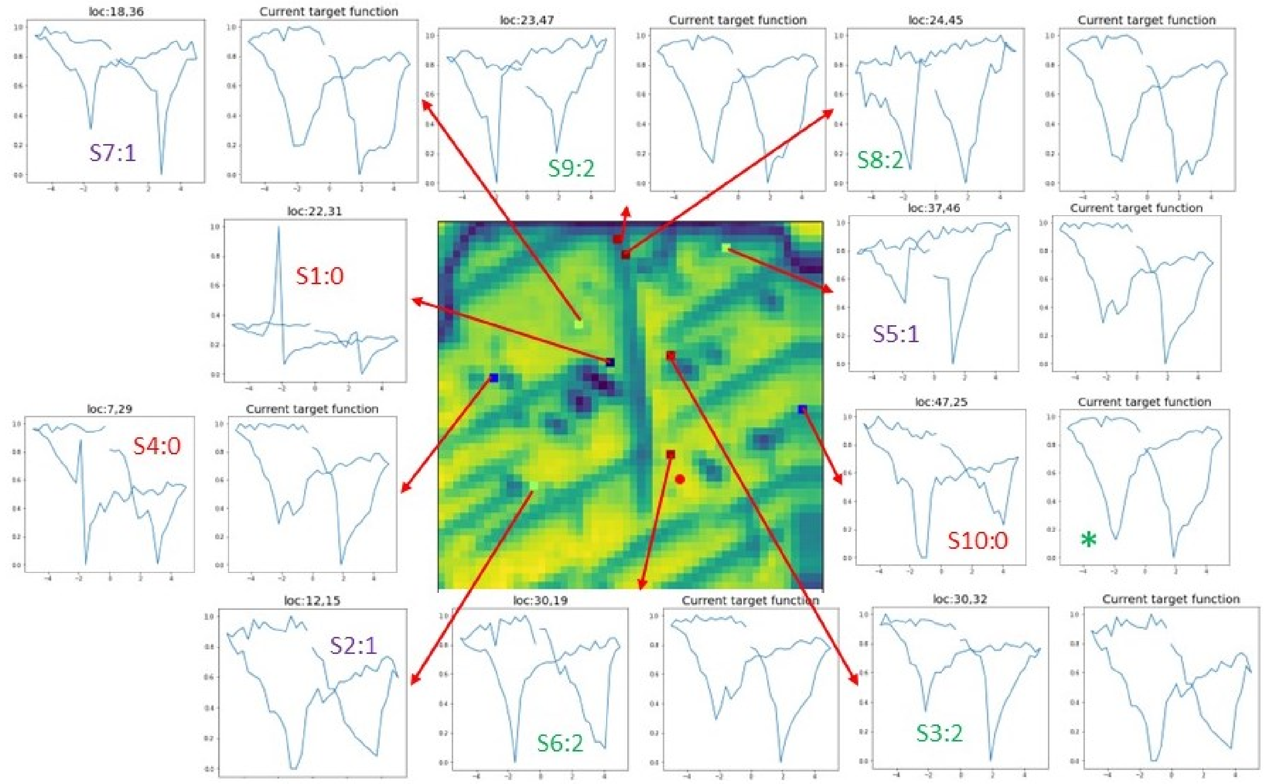

Figure B2. Analysis of Oxide sample 1. User-voting of the spectral at first 10 iterations. Rating for the votes is defined as: bad (0), good (1) and very good (2). We can see the dynamic changes of the target spectral in the current iteration, depending on the voting given in all the previous iterations. Finally, user satisfies with the target (represented with *) at 10th iteration. For the comparative study in this paper, this target is set for both BOARS system, with traditional periodic kernel and deep learning kernel. For all the spectral figures, the X axis ranges from -4V to 4V and the Y axis ranges from 0 A(a.u.) to 1 A(a.u).

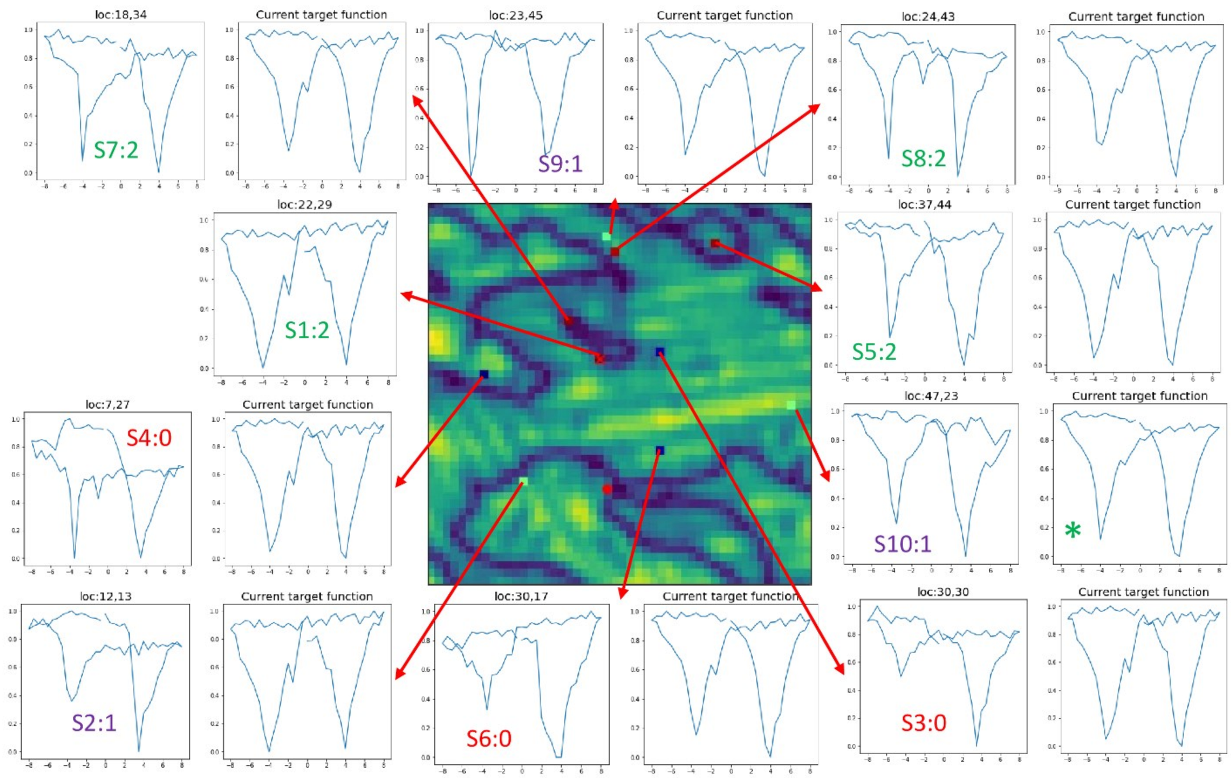

Figure B3. Analysis of Oxide sample 2. User-voting of the spectral at first 10 iterations. Rating for the votes is defined as: bad (0), good (1) and very good (2). We can see the dynamic changes of the target spectral in the current iteration, depending on the voting given in all the previous iterations. Finally, user satisfies with the target (represented with *) at 10^{th} iteration. For the comparative study in this paper, this target is set for both BOARS system, with traditional periodic kernel and deep learning kernel. For all the spectral figures, the X axis ranges from -8V to 8V and the Y axis ranges from 0 A(a.u.) to 1 A(a.u).

We can see the voting are also given based on the current target spectral structure, in other words, the votes are correlated. For example, in fig. B3, the user votes spectral 2 (S2) as good (1) whereas votes spectral 6 (S6) as bad (0), though both are spectral does not have symmetric features. This is because, with more iterations, the user is better informed about the general spectral features over the material sample and the target that can be achieved. Thus, with updated knowledge the user choice can be more specific in updating to a desired target, which is an expected human-thinking approach. In this research, we extend the application of BO to tackle this dynamic human-augmented voting functionalities in ARS system.