Supplementary Information

On-the-fly machine-learning for high-throughput experiments: search for rare-earth-free permanent magnets

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S1. Mean shift theory based clustering

The mean shift theory algorithm implementation used for this study is based on that of Comaniciu and Meer[1]. Each sample is represented by a feature vector concatenating the relative ternary composition values with the powder pattern intensities in order of corresponding q values. The vectors are normalized by subtracting the vector mean and dividing by the variable standard deviation. No noise smoothing is performed. Clusters with 4 or fewer members are integrated into the nearest cluster in composition space. All calculations were performed in 64bit MATLAB on a 3.1 GHz quad core machine with 12 GB of RAM running Windows 7.

The clustering resolution is set by the user through the selection of bandwidth parameters, one for the relative ternary composition and one for the diffraction pattern intensities. The user can select multiple values for each bandwidth and results are given for each pair of bandwidth values. Bandwidth values that are too small will result in an excessive number of clusters, the limit being that each sample is assigned to its own cluster. Bandwidth values that are too large will result in all samples being clustered together. For the FeGaPd system in Figure 2c and d, a composition bandwidth of 0.2 and a diffraction pattern bandwidth of 10^6 were chosen; for the FeCoMo system in Figure 3d and e, the composition and diffraction pattern bandwidths were 0.08 and 58,000 respectively. For both figures, an ICSD mass of 1 was used.

As mentioned in the Methods section, cluster assignment is determined by associating each converged sample vector with the nearest diffraction pattern. Those sample vectors that share the same associated diffraction pattern are then clustered together. This method assumes that the modes of each cluster fall near one of the original diffraction patterns. Other methods exist for cluster assignment. For example, the distance between each converged sample vector (in both composition and diffraction space) could be tabulated and all vectors separated by a length less than the bandwidths would be considered "connected" nodes in a graphical representation. Graph connectivity could then be used to identify cluster assignment, with all vectors connected through a path in the graph assigned to the same cluster. Such a method is not prone to uncertainty in cluster assignment.

A simple heuristic allows the user to narrow in on a reasonable choice of bandwidths within approximately three attempts. Both bandwidths are initially set to values that vary by order of magnitude and the clustering results are inspected, providing a general sense of the appropriate range for each bandwidth. The bandwidth ranges are then reduced, 'zooming in' to the bandwidth parameter values with an appropriate number of clusters. Other heuristics for bandwidth selection have been discussed in the literature[1,2]. A future implementation of CombiDM will include an option to automatically optimize bandwidths.

Fixed, user supplied bandwidths provide a flexibility in clustering that allow for expert input in clustering optimization. Alternatively, if reduced user interaction is desired, a variable bandwidth mean shift algorithm[2,3] can be used, removing the need for user selected bandwidths. These methods rely on a local estimation of sample density (e.g. through the use of k-nearest neighbors) in the feature space to adaptively set the bandwidth value.

There are various ensemble methods which can be used to combine the results of mean shift theory with that of other clustering methods. One method would be to run each clustering method on the data and then select the optimal clustering result based on a cluster evaluation metric. For example, the cluster with the greatest cluster stability measure could be selected.

S2. ICSD based simulated diffraction patterns

When using data from the ICSD, the materials structure data was exported in CIF format and transformed into simulated diffraction patterns using Bruker's Topas software. Typical x-ray parameters used and are listed in Table S2. The Lorentz-polarization correction factor was set to 0 for polarized radiation and 90 for unpolarized radiation. The texture parameter[4] was set to 0.7 for the [1 1 0] direction. The ICSD based diffraction patterns intensities were evaluated at the same 2θ (or q) values as the experimental diffraction patterns.

Primary radius [mm]	Secondary radius [mm]	Detector slit width [mm]	Tube filament length in axial plane [mm]	Sample length in axial direction [mm]	Receiving slit length in axial plane [mm]	Number of rays from point X-ray source in axial plane
217.5	217.5	0.2	12	15	12	30

Table S1. Topas diffraction pattern simulation parameters

S3. Crystal structure of P4/m Fe$_8$CoMo

The P4/m Fe$_8$CoMo structure (Fig. S1) is a tetragonal structure containing 1 formula unit with lattice parameters Wyckoff positions shown in Table S2. In this structure, all nearest neighbors of the Co atom are 4k-site Fe atoms and those of the Mo atom are 4j-site Fe atoms. Even though the differences in the atomic radii of Fe, Co and Mo are small, they can lead to slight broadening of the (110) diffraction peak as observed in the experiment.
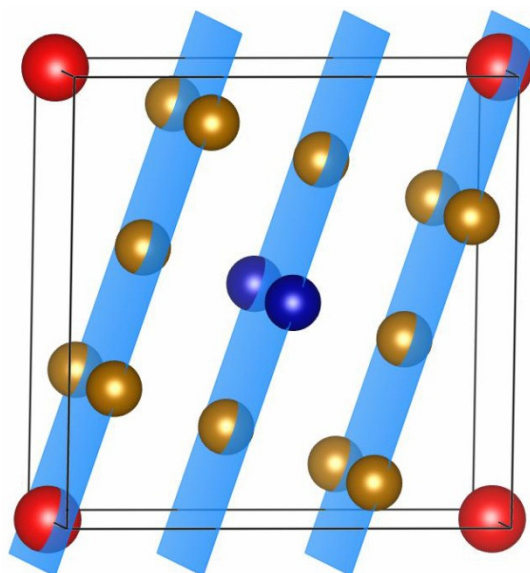

Figure S1. The crystal structure of P4/m Fe$_8$CoMo with (110) planes of the original bcc Fe indicated in light blue planes. The golden, blue and red balls are Fe, Co, and Mo atoms, respectively.

Parameters (Å)	Atom	x	y	z	$M(\mu_B)$
	Fe (4j)	0.68971	0.10091	0.00000	2.11
a = 6.4295	Fe (4k)	0.20417	0.60188	0.50000	2.55
c = 2.8840	Co (1c)	0.50000	0.50000	0.00000	1.67
	Mo (1b)	0.00000	0.00000	0.50000	-0.61

Table S2. The lattice parameters, atomic position and the magnetic moment of each atom of P4/m Fe_8CoMo structure

S4. Layered structure of Fe_8CoMo

By using the full potential (FP) LMTO method, we have also calculated the magnetocrystalline anisotropy of Fe_8CoMo with 5 different unit-cell structures using the experimental c/a ratio of 1.04. Figure S2 shows the schematic of five different structures where the Fe, Co and Mo atoms are represented by the golden, blue and red balls, respectively. The calculation results are shown in Table S3. The results indicate that the layered structure #5 has the lowest energy and highest anisotropy of 28.0 μeV/atom which is close to the value extracted from the experiment (27.0 μeV/atom). In this structure, the Co layers and Mo layers are sandwiched between Fe layers and the Mo layers tend to be separated from Co layers to lower the energy.
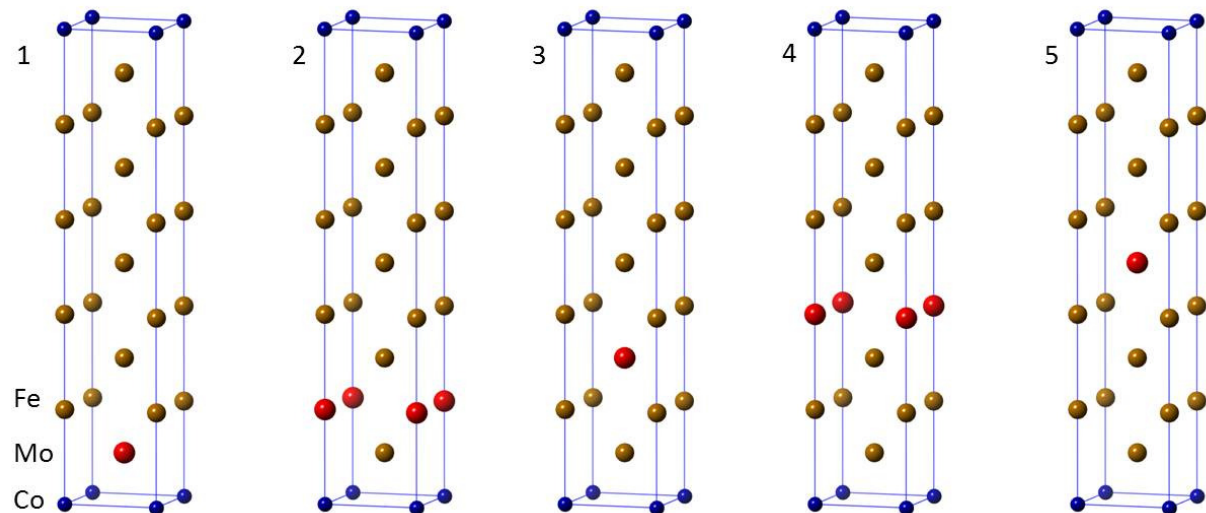

Figure S2. Five different unit cells of Fe_8CoMo with layered structures

Configuration	1	2	3	4	5
Energy(eV)	0.2474	0.1653	0.0312	0.0056	0
M(μ_B/cell)	19.79	20.12	20.22	20.41	20.38
K(μeV/atom)	-49.3	12.1	13.9	18.5	28

Table S3. The formation energy, magnetic moment and anisotropy constant of five different layered structures Fe_8CoMo

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