xanesnet

a deep neural network for x-ray spectroscopy

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the xanesnet project

~$ we think that computational simulations for X-ray absorption spectroscopy (XAS) should be fast, affordable, and accessible to everyone

~$ the popularity of XAS is on a steep upward trajectory globally, driven by advances at, and widening access to, high-brilliance light sources such as synchrotrons and X-ray free-electron lasers (XFELs), but the high resolution of modern X-ray spectra, coupled with ever-increasing data acquisition rates, brings into focus the challenge of accurately and cost-effectively analyzing these data

~$ decoding modern X-ray spectra demands detailed computational calculations that are capable of capturing the complexity of the underlying physics but that are - at the same time - fast, affordable, and accessible enough to appeal to researchers

~$ this is a tall order - but we're using deep neural networks (DNNs) to make it possible!

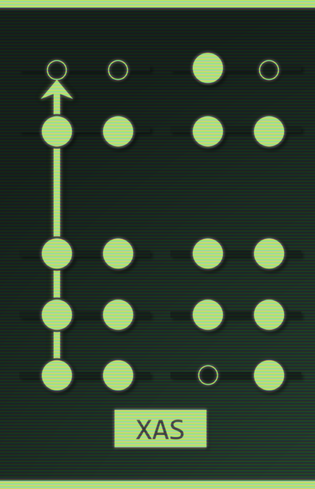
x-ray spectroscopy

~$ atoms are able to absorb and emit X-ray radiation

~$ absorption of X-ray radiation promotes electrons from core energy levels and creates high-energy, unstable core-hole states

~$ these states subsequently decay

~$ using X-ray spectroscopy, we can monitor these processes and, in some cases, what happens in between



deep neural networks

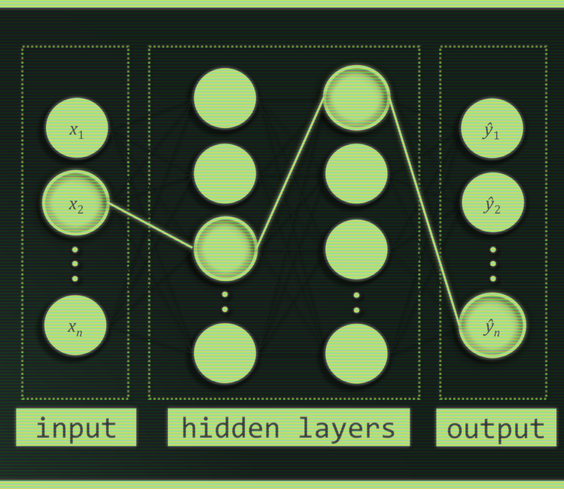
~$ DNNs are machine-learning models based on the architecture of the human brain

~$ neurons are containers for numbers; these numbers are their activations

~$ activations in one layer influence the activations in subsequent layers

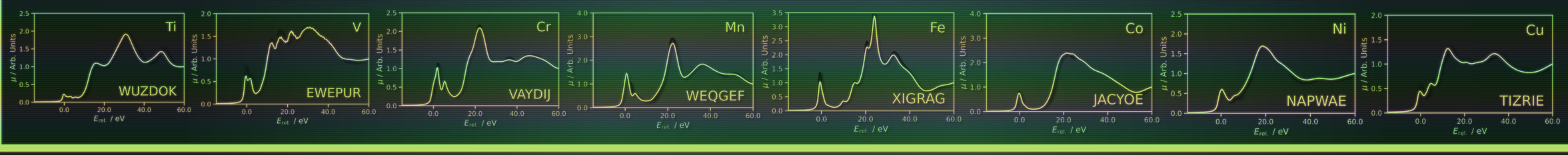
~$ activating the input layer by passing in data produces a particular response in the output layer that can be tuned iteratively

~$ DNNs can be used to produce predictions from, or to classify, input data



predictions

~$ xanesnet predicts the XANES spectra of arbitrary transition metal X-ray absorption sites using only the local geometric structure: the coordination environment

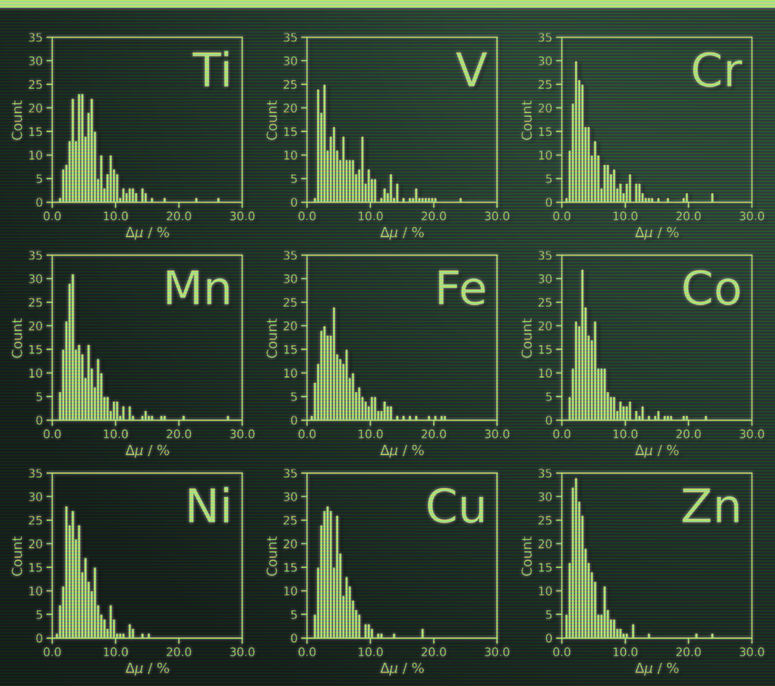


performance

~$ percentage differences, $\Delta\mu$, between predicted and target XANES spectra + skew metrics

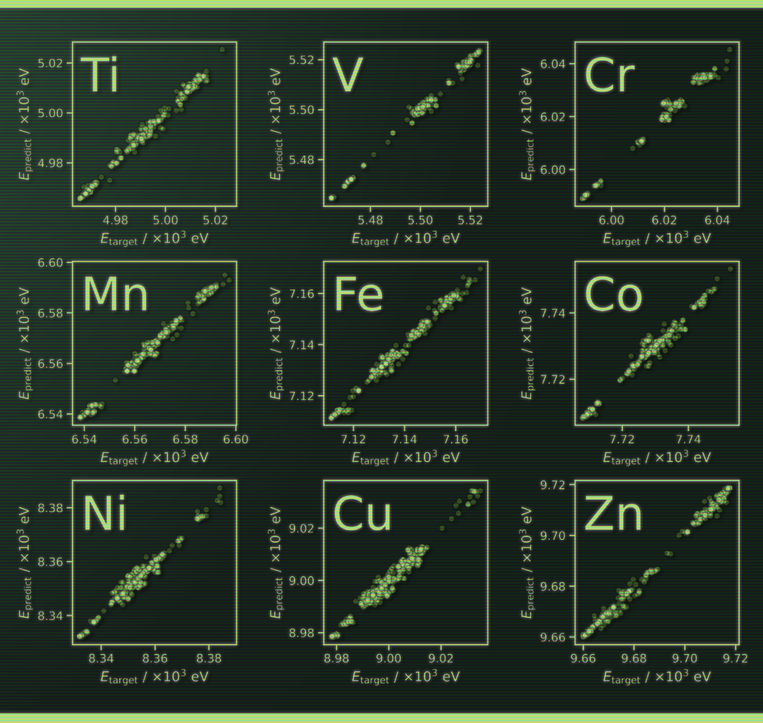
edge	$\Delta\mu_{avg.}$ / %	skew
Ti	5.5 (3.8)	1.898
V	5.2 (3.2)	1.625
Cr	3.8 (2.5)	1.926
Mn	4.3 (2.8)	2.242
Fe	4.7 (3.1)	1.607
Co	4.3 (2.8)	2.058
Ni	4.1 (2.6)	1.286
Cu	4.0 (2.7)	2.007
Zn	3.2 (2.2)	3.005

[values in parentheses are post-broadening]



~$ peak position errors, ΔE, between predicted and target XANES spectra + R^2 metrics

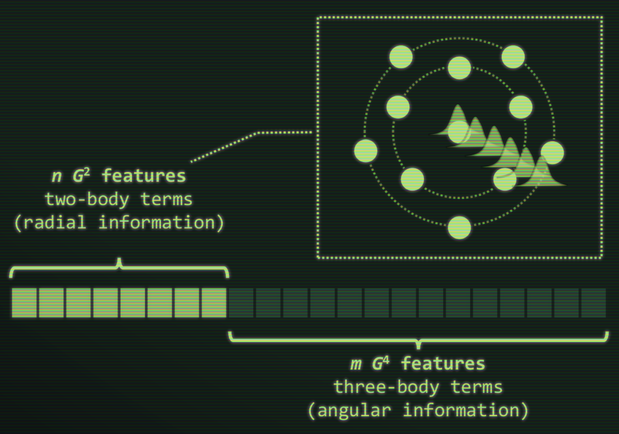
edge	$\Delta E_{avg.}$ / eV	R^2
Ti	0.86	0.996
V	0.54	0.999
Cr	0.65	0.997
Mn	0.76	0.997
Fe	0.83	0.996
Co	0.74	0.993
Ni	0.88	0.993
Cu	0.99	0.991
Zn	0.95	0.997



physical insight

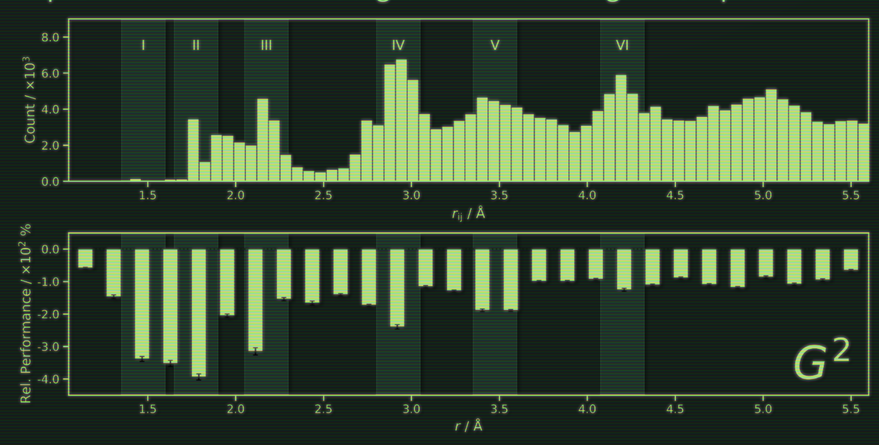
~$ local environments around X-ray absorption sites are encoded in XANESNET as feature vectors

~$ feature vectors comprise informative features that XANESNET uses to predict XANES spectra



~$ scrambling the feature vectors featurewise and assessing the performance penalty tells us about the importance of each individual feature

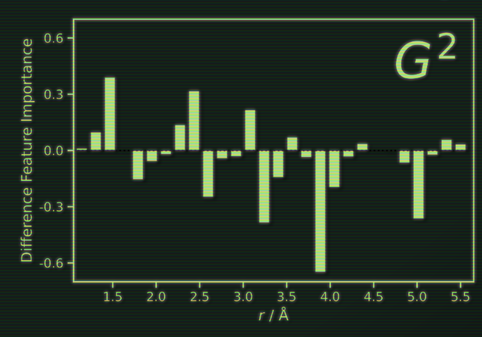
pair-distribution histogram for Fe K-edge absorption sites



feature importance for G^2 feature functions placed at a radial distance, r; Fe K-edge

shell	label(s)
1st coordination	**I** (H), **II** (C, N, O, …), **III** (Si, P, S, …)
2nd coordination	**IV** (C, N, O, …), **V** (Si, P, S, …)
3rd coordination	**VI** (...)

difference (high-energy window minus low-energy window) feature importance for G^2 feature functions placed at a radial distance, r; Fe K-edge



~$ feature importance in the high-energy window (+50.0 → +56.0 eV) is greater close to the absorption site: single scattering

~$ feature importance in the low-energy window (-3.0 → +3.0 eV) is greater further from the absorption site: multiple scattering