



A brief introduction of Generative Adversarial Network for Superconductivity

Fang Han Lim
3/6/2024



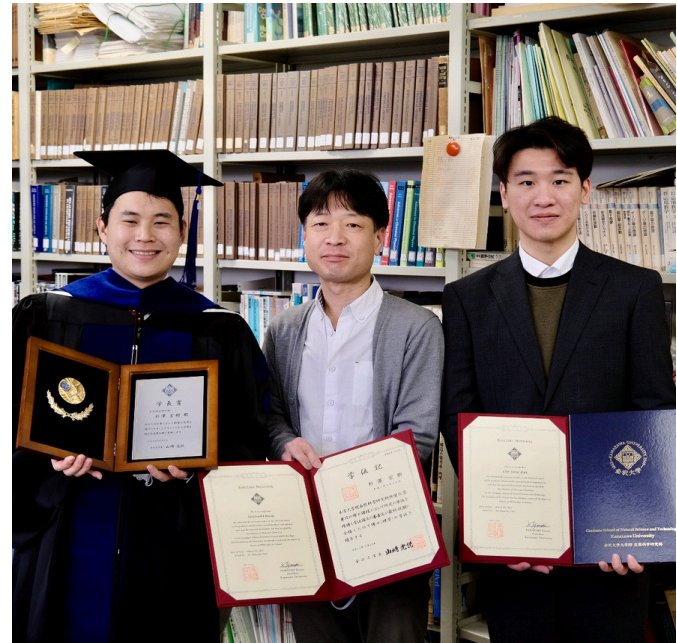
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About Me

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Fang Han, LIM:

- Nationality: Malaysia
- Education:
 1. Kanazawa University
 2. University of British Columbia
- Area of Research :
 1. Ab Initio method for Electronic Structure Theory
 2. Theory of Superconductivity
- Work Experience :
 1. Junior Financial Analyst
 2. Software Development Engineer
 3. Postdoc



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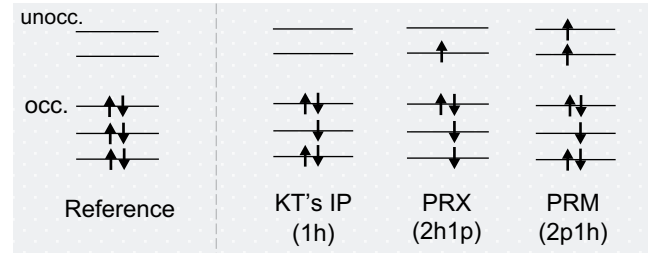
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Worked on...

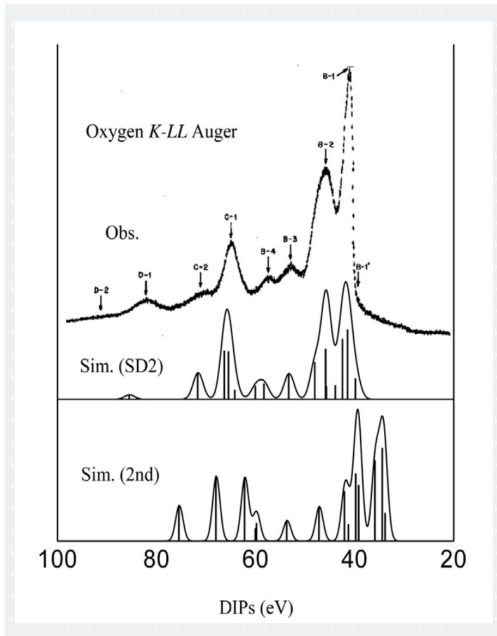
Dyson Equation, Theory of Superconductivity, & other

2-1 Dyson Propagator

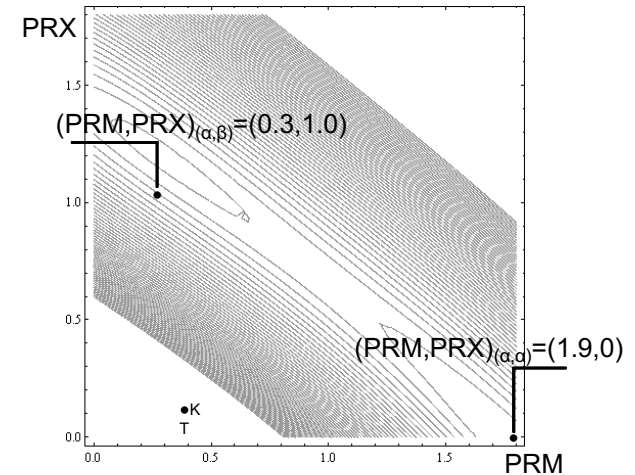
- ✓ The self-energy term for the 3hp configuration is included into 2nd order shifted denominator approximation (SD2)
- ✓ DIP results from SD2 are as accurate as ADC(2) with much lower computational cost



Auger Electron Spectra Simulation via SD2



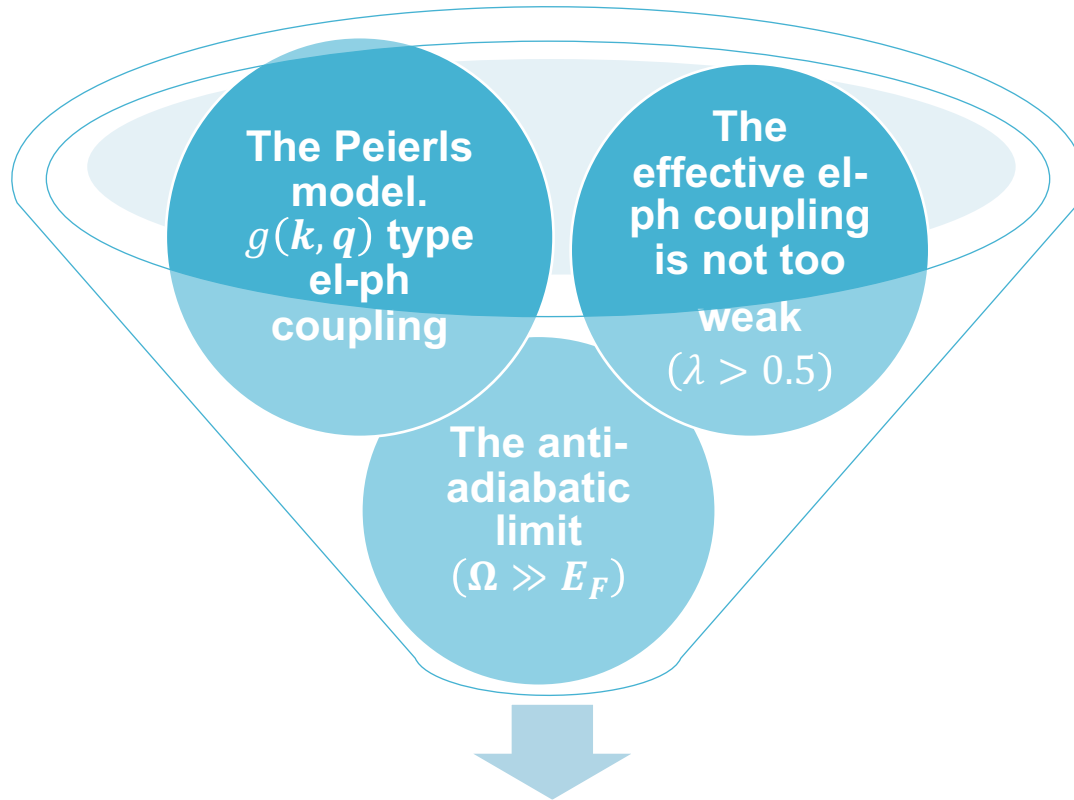
$$\mathbb{G}(E)^{-1} = \mathbb{G}_0(E)^{-1} - \Sigma(E)$$



Spin-Component-Scaled Electron Propagator

- ✓ Corrects fundamental errors from 2nd order perturbation
- ✓ Similar performance compared with 3rd order methods despite lower computational cost

2-2 Theory of Superconductivity

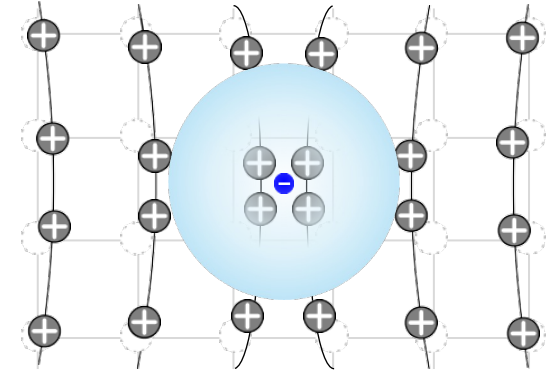


This study aims to prove that phonon-mediated superconductivity can be unconventional and with very high-TC

2-2 Theory of Superconductivity

Polaron:

The original electron surrounded by a cloud of phonons that propagates coherently with it (the particle keeps emitting and absorbing phonons).



- Holstein Model

The simplest most studied lattice model for electron-phonon (el-ph) coupling

- Fröhlich Model

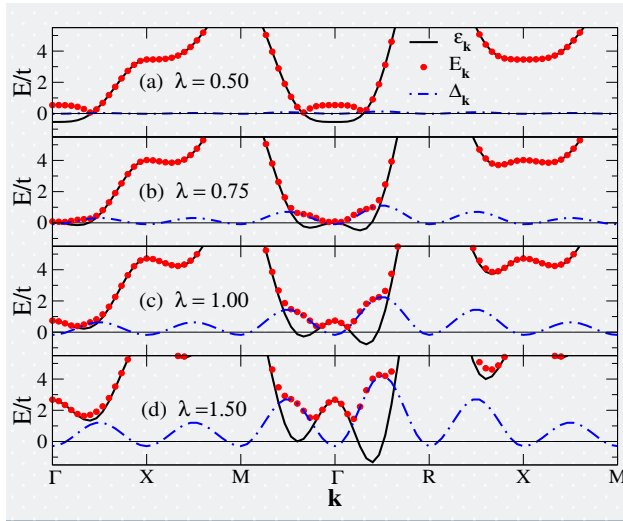
This describes free electrons coupled to the longitudinal acoustic phonons through unscreened Coulomb interactions, with a \mathbf{q} dependent $g(\mathbf{q})$ vertex.

- Peierls / SSH Model

Oscillation of lattice will modulate the hopping of electrons, describing one form of $g(\mathbf{k}, \mathbf{q})$ el-ph coupling. Research also shows that polaron can be light at strong λ . ^[1]

[1] J. Sous, M. Chakraborty, Roman V. Krems, M. Berciu, *Phys. Rev. Lett.* **121**, 247001 (2018).

2-2 Theory of Superconductivity



Results in a nutshell:

	T_c/K	$\Delta(0)/meV$
$\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$	96	30
This Study ($\lambda = 1.5,$ $n = 0.1$)	198	~ 30

Standard BCS

Adiabatic, $\omega_D \ll E_F$

Small el-ph coupling $\lambda \ll 1$

Simple Coulomb Interaction

$$V_{k,k'} = \begin{cases} -V_0, & \text{if } E_F \leq \epsilon_k, \epsilon_{k'} \leq E_F + \hbar\omega_D \\ 0, & \text{otherwise} \end{cases}$$

Simple Gap Equation

$$\frac{1}{2V} \sum_{k'} V_{k,k'} \frac{\Delta_{k'}}{\sqrt{\xi_k^2 + \Delta_{k'}^2}} \tanh\left(\frac{\beta E_{k'}}{2}\right)$$

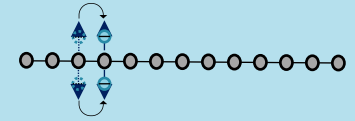
Present Study

Antiadiabatic, $\Omega \gg E_F$

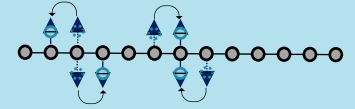
Strong el-ph coupling $\lambda > 0.5$

Polaron-induced Interaction

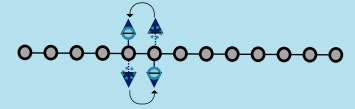
on-site NN pair hopping



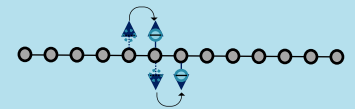
on-site and NNN singlet pair



NN AFM exchange



Pair hopping of a NN singlet pair



Gap Equation

$$\Delta_k = \Delta_s + \Delta_{s^*} \sum_{\gamma} \cos(2k_{\gamma}) + \Delta_p \sum_{\gamma} \sin(k_{\gamma})$$

2-3 Others

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Complete spin contamination-free MP2 method

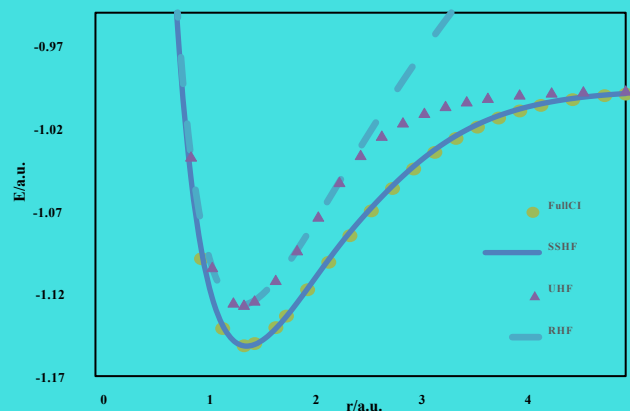
- ✓ Spin contamination from the reference space fails unrestricted MP2
- ✓ Applying spin-adapted formalism to spatial-orbital basis (LCAO-MO) to consider spin symmetry explicitly
- ✓ Removing triplet and quintet parts completely by performing unitary transform to diagonalize \hat{S}^2

Reaction	Expt.	UMP2	Present study	UCCSD(T)
$\text{H}_2 + \text{H} \rightarrow \text{H} + \text{H}_2$	9.7	13.2	9.1	10.4
$\text{H}_2\text{O} + \text{H} \rightarrow \text{OH} + \text{H}_2$	19.4	26.5	18.5	17.9
$\text{NH}_3 + \text{H} \rightarrow \text{NH}_2 + \text{H}_2$	11.4	20.1	14.6	13.6
$\text{CH}_4 + \text{H} \rightarrow \text{CH}_3 + \text{H}_2$	10.6	21.0	12.4	16.5
$\text{CH}_2\text{O} + \text{H} \rightarrow \text{HCO} + \text{H}_2$	7.6	9.8	8.9	7.4
$\text{CH}_3\text{F} + \text{H} \rightarrow \text{CH}_3 + \text{HF}$	30.4	39.0	35.9	35.4
$\text{H} + \text{CH}_2\text{O} \rightarrow \text{CH}_3\text{O}$	4.4	12.5	5.9	5.2
$\text{H} + \text{C}_2\text{H}_2 \rightarrow \text{CH}_2\text{CH}$	2.4	10.6	2.5	4.8
MAE		7.1	1.9	2.4
MAD		7.6	2.5	3.1

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Spin-Symmetrized Hartree-Fock

- ✓ Full-CI faces difficulties in non-orthogonal state calculation and SCF convergence
- ✓ Calculation of the dissociation potential curve of molecules using UHF method is difficult
- ✓ Spin-Symmetrized Hartree-Fock (SSHF) method uses time-reversal operator to generate new reference state

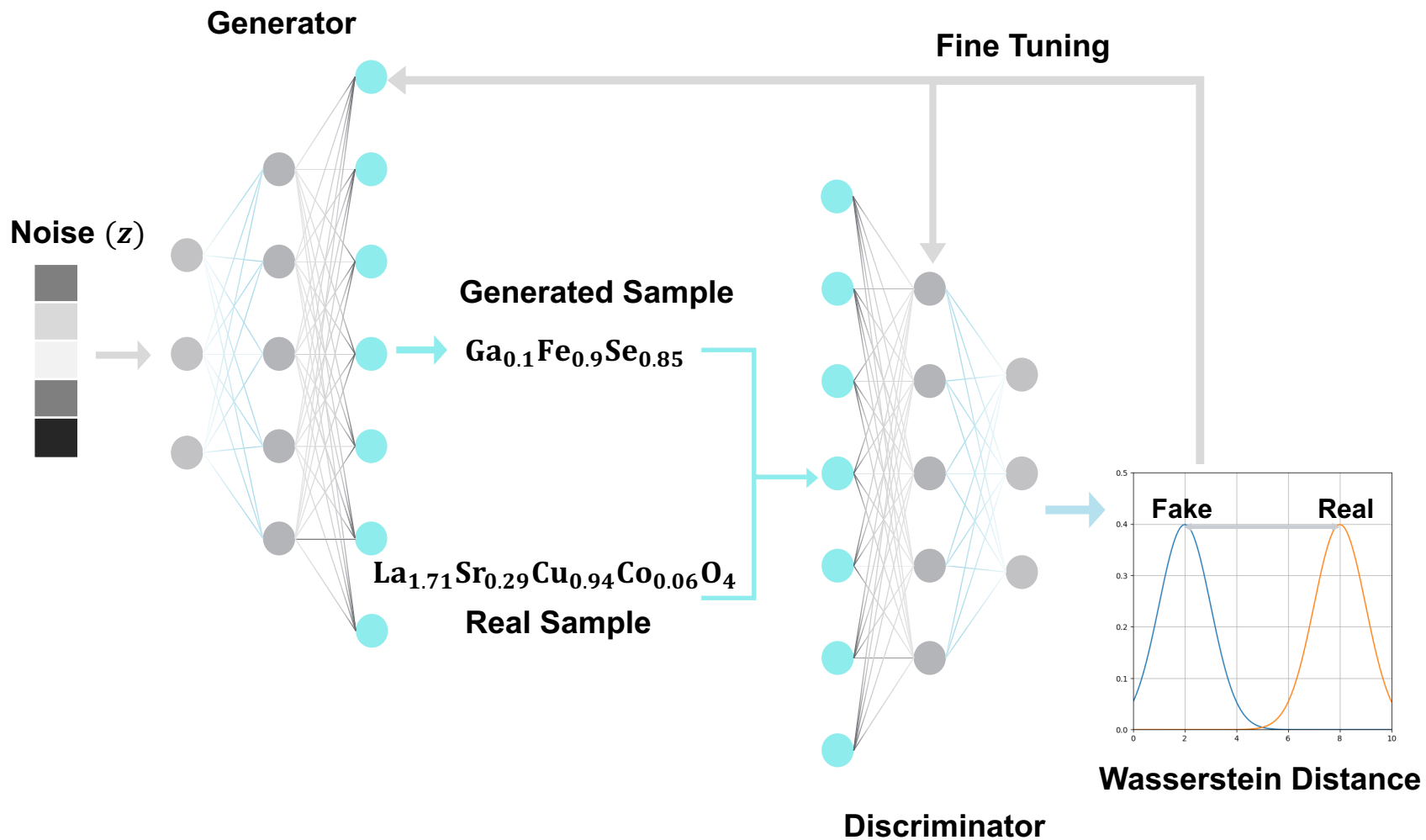




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Generative Adversarial Network

WGAN

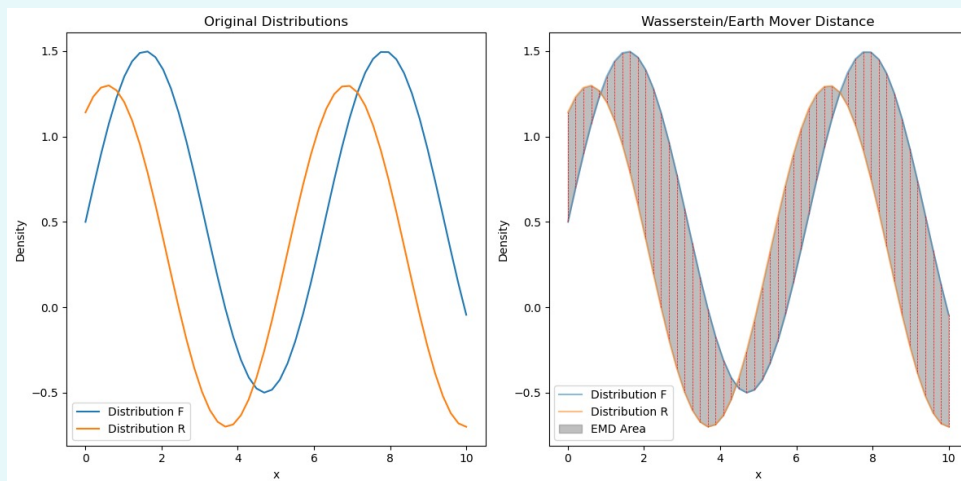


WGAN Loss Function

- Wasserstein Loss:

Earth Mover's Distance (EMD) measures the minimum cost of transporting mass to transform one distribution into another. In GAN it represents the distance between the generator's distribution and the real data distribution.

$$W_p(F, R) = \left(\inf_{\gamma \in \Gamma(F, R)} \int_{\mathcal{X} \times \mathcal{X}} d(x, y)^p d\gamma(x, y) \right)^{1/p}$$



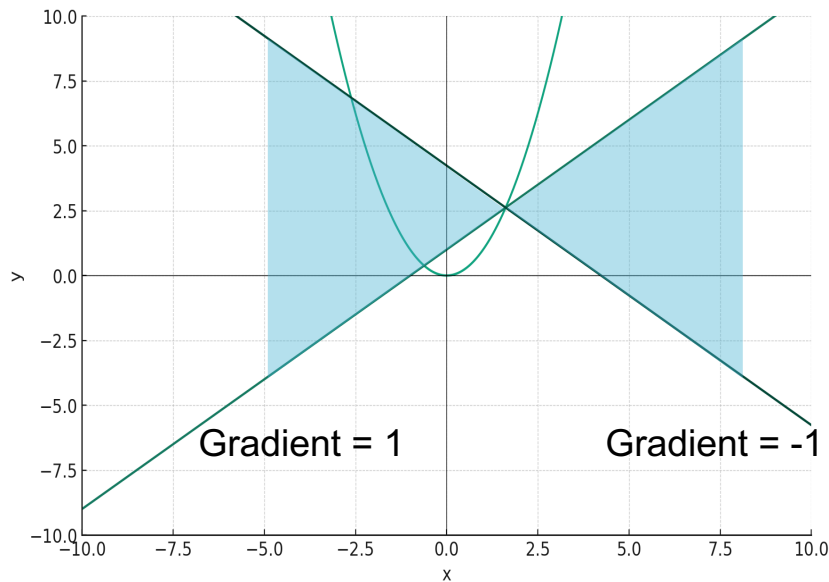
Usage in GAN:

Wasserstein distance is approximated with the interpolation method, and loss function is penalized with a regularization term

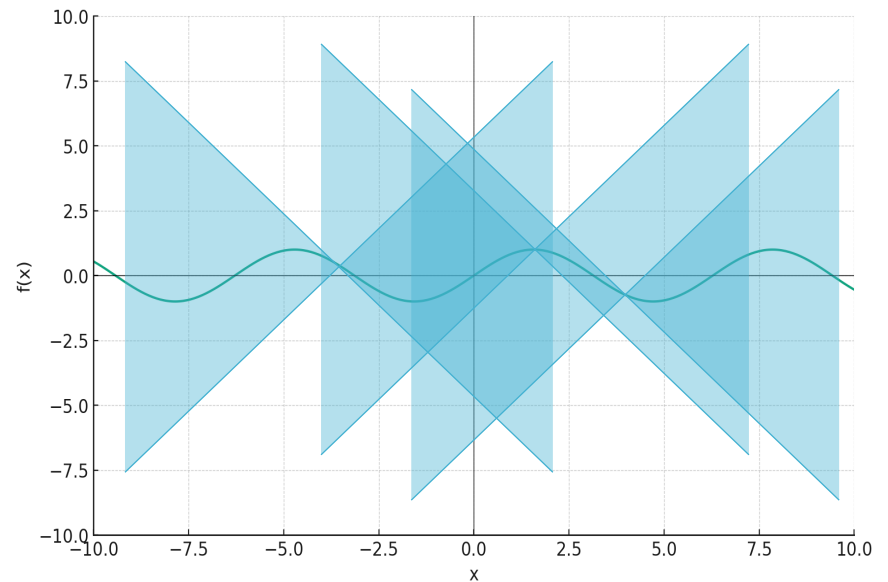
1-Lipschitz Continuity

- Wasserstein Loss:

$$Loss_W = \min_g \max_d E(d(x)) - E(d(g(z)))$$



Non-1-L continuous



1-L continuous

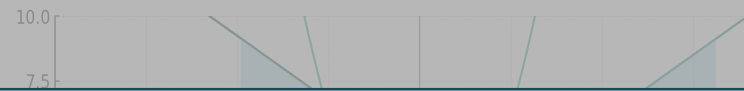
- Gradient Penalty (GP):

$$Loss_W = \min_g \max_d E(d(x)) - E(d(g(z))) + \lambda E(\|\nabla d(\hat{x})\| - 1)^2$$

1-Lipschitz Continuity

- Wasserstein Loss:

$$Loss_W = \min_g \max_c E(c(x)) - E(c(g(z)))$$



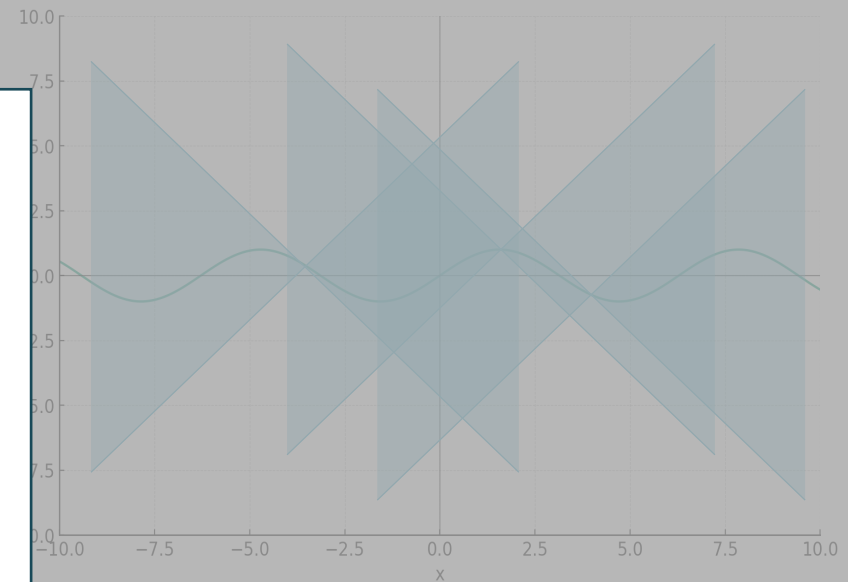
Regularizing Discriminator's Gradient:

For 1-Lipschitz Continuity to hold everywhere, it's sufficient to ensure it holds on the line segment between every pair of real and generated samples.

Gradient Penalty via interpolation:

$$GP = \lambda(\|\nabla d(\hat{x})\|_2 - 1)^2$$

$$\hat{x} = \epsilon x + (1 - \epsilon)g(z)$$



1-L
continuous

$$Loss_W = \min_g \max_c E(c(x)) - E(c(g(z))) + \lambda E(\|\nabla d(\hat{x})\|_2 - 1)^2$$

Material Descriptor



	H	He	...	O	...	Cu	...	Pu
0	1	1	0	0	0	0	0	1
1	0	.	.		1		1	0
2	.		1					.
3	.			.		1		.
4	.				.			.
...	.			1				.
9	.						.	.
10	0	0

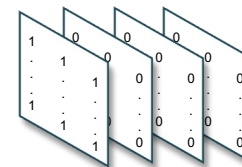
Composition Matrix^T
(adjusted one-hot)

	H	He	...	O	...	Cu	...	Pu
0	1	1	0	0	0	0	0	1
1	0	.	0		.4		.25	0
2	.		.625					.
3	.			.		1		.
4	.				.			.
...	.			.997				.
9	.						.	.
10	0	0

Normalized Weight Matrix^T

+

Labels : Cuprate, Pnictide, Both, Others



Neural Network Structure

Model	Layer	Input Shape	Kernal	Stride	Padding
Generator	Fc	[batch, 100 + No. of Classes]	-	-	-
	Reshape	[batch, 512×1×1]	-	-	-
	DeConvo1	[batch, 512, 1, 1]	(3,3)	(2,2)	0
	DeConvo2	[batch, 256, 3, 3]	(3,3)	(2,2)	1
	DeConvo3	[batch, 128, 5, 5]	(3,3)	(1,1)	1
	DeConvo4	[batch, 128, 5, 5]	(3,3)	(2,2)	0
	DeConvo5	[batch, 64, 11, 11]	(3,3)	(1,1)	1
	DeConvo6	[batch, 64, 11, 11]	(5,3)	(2,1)	1
	DeConvo7	[batch, 32, 23, 11]	(5,3)	(2,1)	1
	DeConvo8	[batch, 32, 47, 11]	(4,3)	(2,1)	1
	Output	[batch, 2, 94, 11]			
Critic	Convo1	[batch, 2+ No. of Classes, 94, 11]	(4,3)	(2,1)	1
	Convo2	[batch, 32, 47, 11]	(5,3)	(2,1)	1
	Convo3	[batch, 32, 23, 11]	(5,3)	(2,1)	1
	Convo4	[batch, 64, 11, 11]	(3,3)	(1,1)	1
	Convo5	[batch, 64, 11, 11]	(3,3)	(2,2)	0
	Convo6	[batch, 128, 5, 5]	(3,3)	(1,1)	1
	Convo7	[batch, 128, 5, 5]	(3,3)	(2,2)	1
	Convo8	[batch, 256, 3, 3]	(3,3)	(2,2)	0
	Flatten	[batch, 512, 1, 1]	-	-	-
	Fc	[batch, 512×1×1]	-	-	-

Optimization

- Model Details

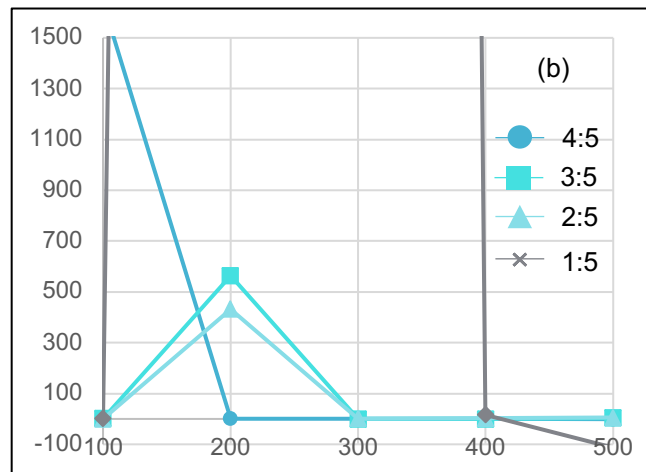
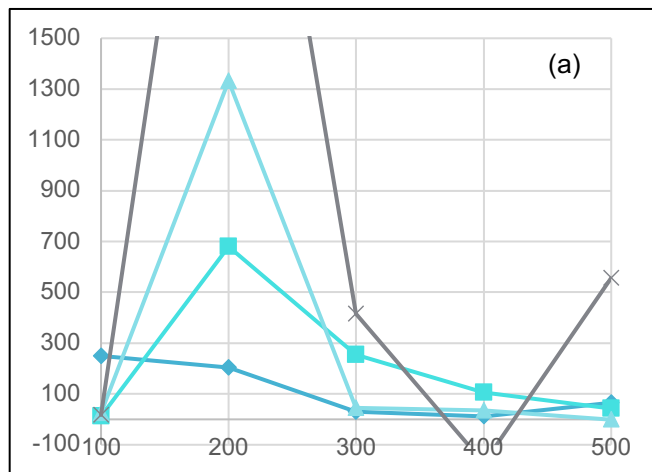
Hyperparameters			
Batch Size:	128	Learning Cycle: (Gen : Crit)	4 : 5
Epoch	500	Noise Dim.	128
Learning Rate:	0.001	Gradient Penalty :	10
No. of Labels:	4	Custom loss Penalty :	10
Optimizer :	Adam	θ_r :	0.1
Beta :	(0.5,0.9)	θ_b :	0.005

- Rate of Success Generation

Class	Rate of Recovery /100k generation	Stable Compound [1] /100k generation
Cuprate	25.6%	6.0%
Iron Pnictides/ Chalcogenides	51.1%	7.2%
Both	26.3%	6.1%
Others	55.3%	5.5%

[1] Davies, D. W. et al. *Chem.* 1, 617–627 (2016).

- Generator(a) and Discriminator(b) Loss at Different Learning Cycle (Gen : Crit)



T-SNE visualization

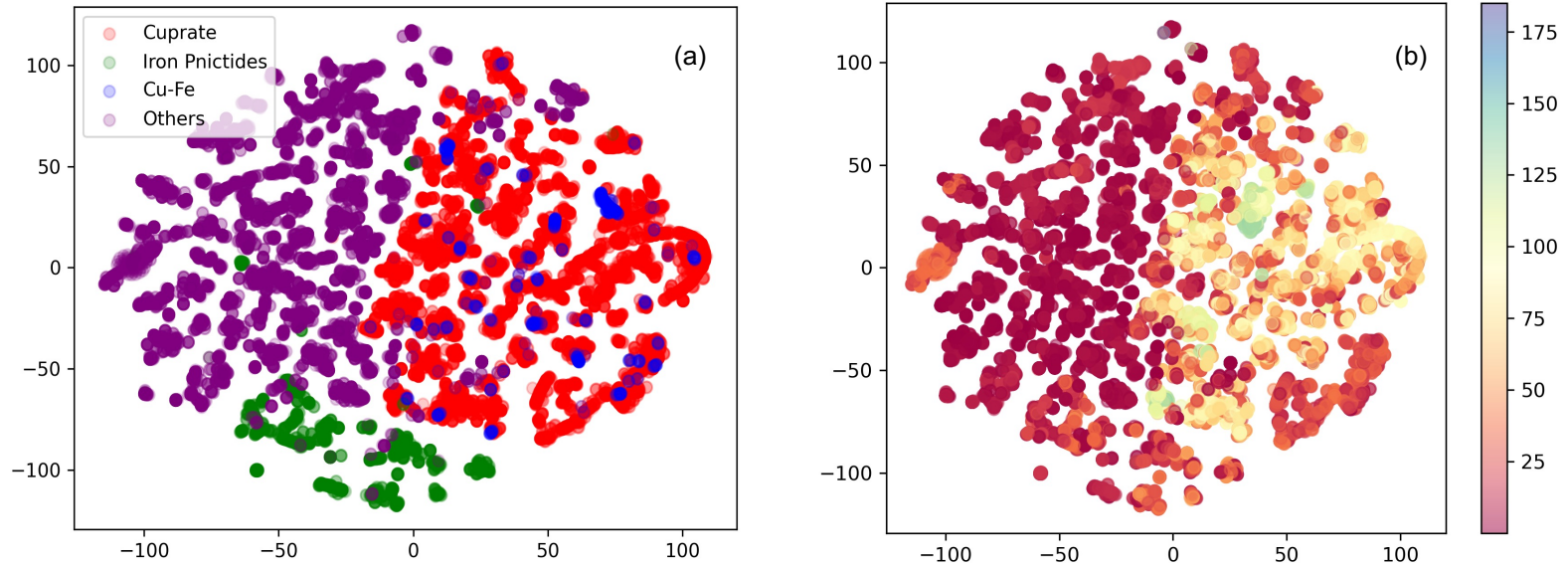
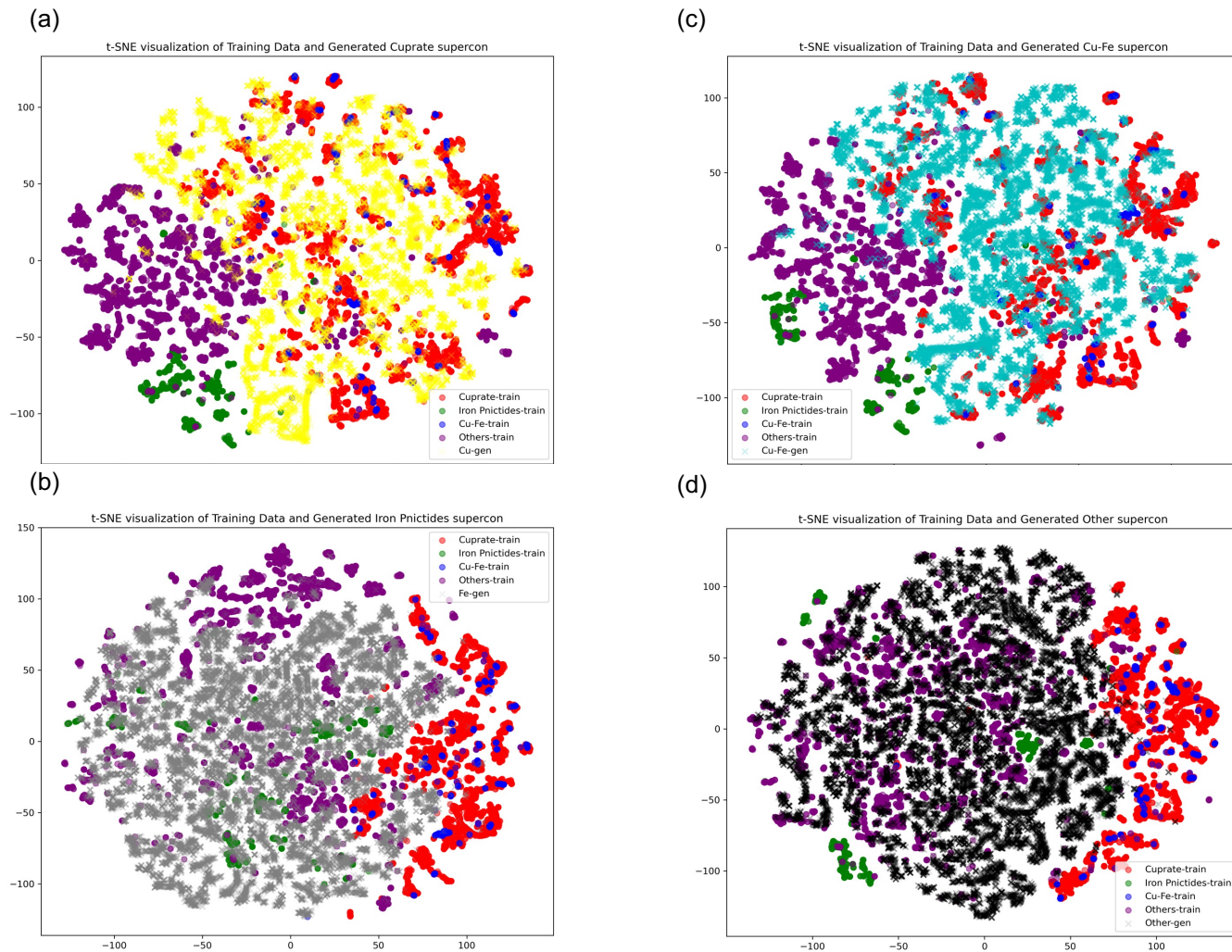


Figure shows following details of Dataset used for GANs training:

(a) Distribution of classes of Superconductor, (b) Distribution of T_c

Material Generation



Distribution of training data and generated material of different classes : (a) Cuprate, (b) Iron Pnictides/Chalcogenides : (c) Mixture of Cuprate & Iron Pnictides/Chalcogenides, (d) Other else

Candidate Materials

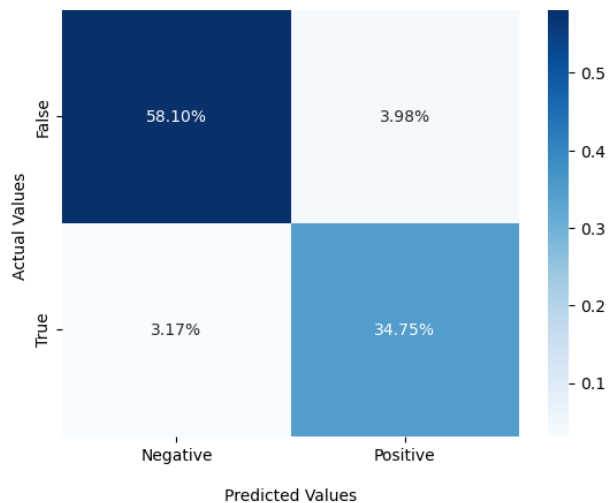
Chemical Composition	ΔE / eV	Stability / eV	Band gap / eV	Magmom/ pa	Chem Sys	Point Group	Experimentally Observed	Estimated T_c / K
B _{3.0} Si _{1.0} (MP)	0.827(-0.044)	0.929(0.000)	0.00(1.41)	0.0004	Orthorhombic	(mmm)	True	3.71
F _{6.0} S _{1.0} (MP)	-1.175(-1.846)	0.836(0.009)	0.00(5.81)	0.0005	Cubic	m $\bar{3}$ m	True	15.55
Eu _{1.0} S _{1.0} (MP)	-2.028(-2.462)	0.220(0.000)	0.00(0.00)	0.0010	Cubic	(m $\bar{3}$ m)	True	1.09
S _{1.0} Rb _{1.0} Au _{1.0} (MP)	-0.784(-0.927)	0.100(0.000)	0.00(1.99)	0.0021	Orthorhombic	(mmm)	True	14.89
B _{6.0} Si _{1.0} (MP)	-0.072(0.454)	0.071(0.479)	0.00(0.05)	0.0004	Cubic	(m $\bar{3}$ m)	True	7.21
S _{2.0} Pd _{1.0} (MP)	0.012(-0.338)	0.065(0.000)	0.00(0.42)	0.0004	Orthorhombic	mmm	True	1.03
P _{1.0} As _{1.0} (MP)	0.066(0.083)	0.063(0.083)	0.00(0.00)	0.0005	Trigonal	3m	False	1.47
Ce _{1.0} S _{2.0} (MP)	-1.917(-1.945)	0.002(0.104)	0.71(0.00)	0.0004	Tetragonal	4/mmm	True	0.05
S _{1.0} Pd _{1.0} (MP)	-0.450(-0.457)	0.001(0.000)	0.00(0.00)	0.0004	Tetragonal	(4/m)	True	1.02
Hf _{1.0} Ta _{1.0} Re _{2.0} (MP)	-0.298(1.274)	0.001(0.000)	0.00(0.00)	0.0004	Cubic	(m $\bar{3}$ m)	False	2.54
Cl _{0.1} Cu _{1.0} As _{1.9} (MP)	-0.050(0.085)	0.001(0.085)	0.00(0.00)	0.0004	Orthorhombic	(mmm)	False	1.59
B _{1.0} Mg _{8.0} Pt _{4.0} (MP)	-0.671(-0.645)	0.000(0.000)	0.00(0.00)	0.0007	Tetragonal	(m $\bar{3}$ m)	True	2.46
Y _{1.0} Pd _{3.0} (MP)	-0.712(-0.734)	0.000(0.000)	0.00(0.00)	0.0005	Cubic	(m $\bar{3}$ m)	True	0.78
Nb _{1.0} S _{3.0} (MP)	-0.747(-1.149)	0.000(0.000)	0.00(0.00)	0.0004	Monoclinic	2/m	True	1.22

[1] Gupta, V., Choudhary, K., Tavazza, F. et al. *Nat Commun* **12**, 6595 (2021).

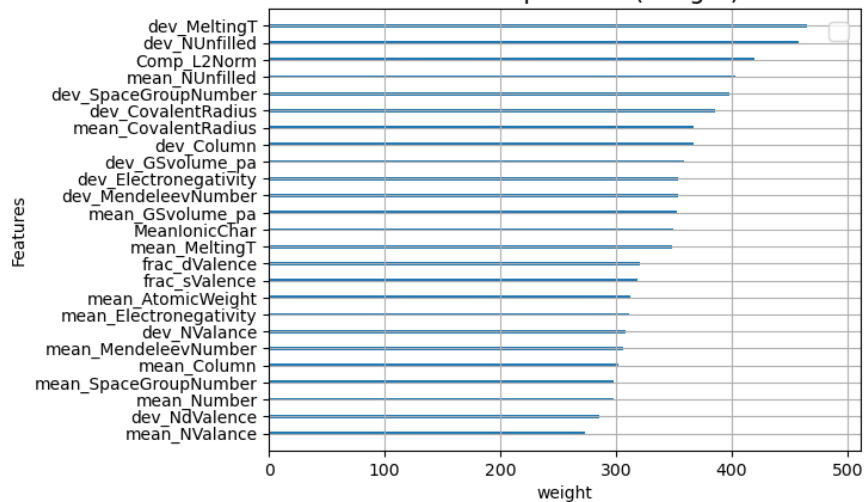
[2] Minoru Kusaba, Chang Liu, Ryo Yoshida, *Com Mat Sci* **211** 111496 (2022)

Classifier & Regressor with XGB

Tc > 10K classifier(N_estimator:300)



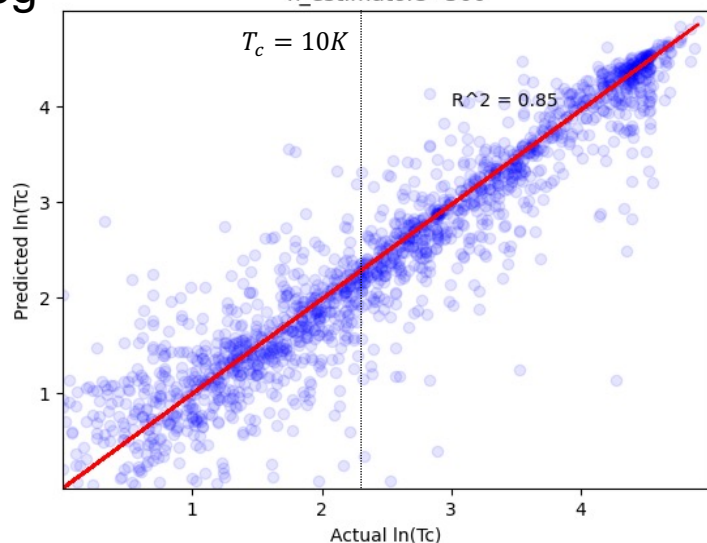
Feature Importance(Weight)



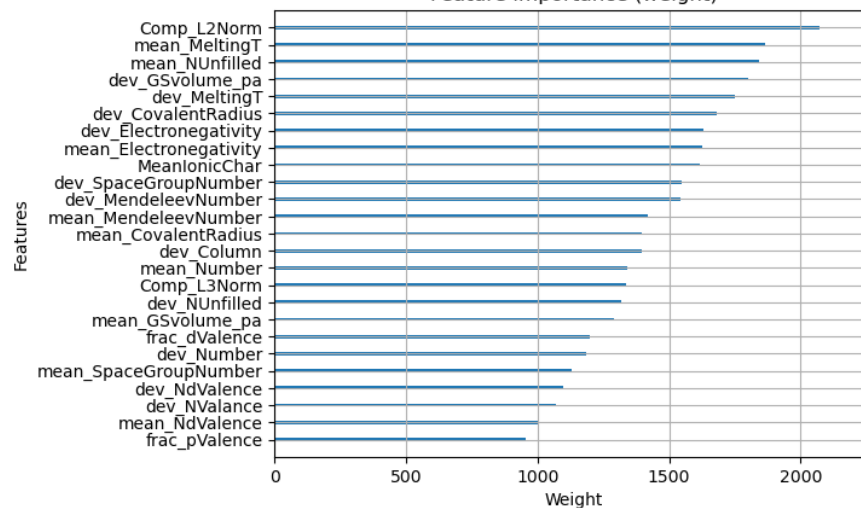
Clf

Reg

n_estimators=500



Feature importance (weight)



Improvement on WGAN

Group	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18										
Period																												
Nonmetals	1																	2										
	H																	He										
Metals	3	4											5	6	7	8	9	10										
	Li	Be											B	C	N	O	F	Ne										
	11	12											13	14	15	16	17	18										
	Na	Mg											Al	Si	P	S	Cl	Ar										
	19	20											21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36
	K	Ca											Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
	37	38											39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54
	Rb	Sr											Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
	55	56	La to Yb										71	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86
	Cs	Ba											Lu	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
	87	88	Ac to No										103	104	105	106	107	108	109	110	111	112	113	114	115	116	117	118
	Fr	Ra											Lr	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Nh	Fl	Mc	Lv	Ts	Og
	s-block (incl. He)		f-block		d-block								p-block (excl. He)															
Lanthanides			57		58	59	60	61	62	63	64	65	66	67	68	69	70											
			La		Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb											
Actinides			89		90	91	92	93	94	95	96	97	98	99	100	101	102											
			Ac		Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No											

From wikipedia

