Accelerating Inorganic Phosphor Development

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Acknowledgements

McElrath post-doctoral fellow
Dr. Martin Hermus (now at OSRAM)

Graduate Students
Dr. Ya Zhuo (Now at UCSB)
Shruti Hariyani
Dr. Anna Duke (Now at Sandia Nat. Lab)

NSF (CAREER 1847701) (DMR 1911311)

NIH (1R01AR072742-01)

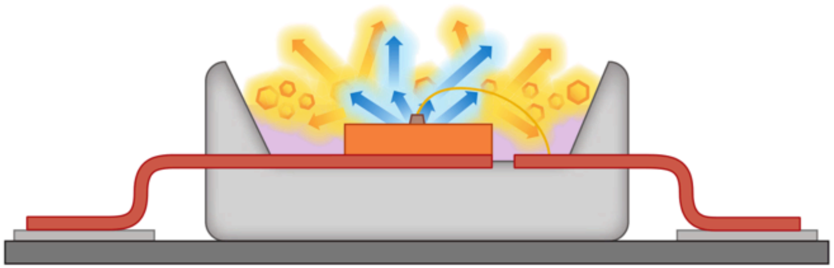
The Welch Foundation (E-1981)

UNIVERSITY of HOUSTON
TEXAS CENTER FOR SUPERCONDUCTIVITY

2020 Sloan Research Fellows

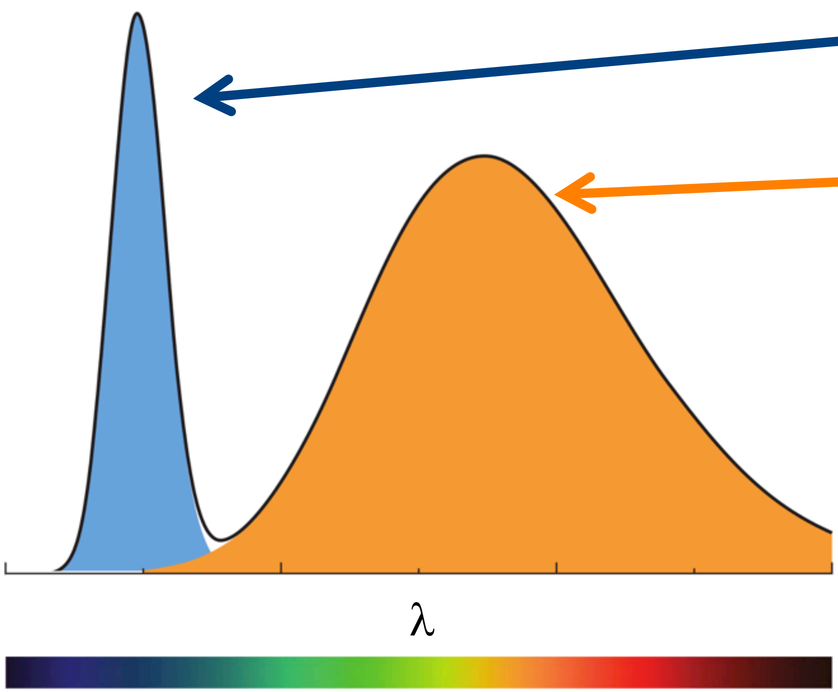
Phosphor converted solid-state (LED) lighting

LED + yellow phosphor



White LEDs use a blue (450 nm) or near-UV (405 nm) that is partially converted by a phosphor.
The combination of the LED emission and phosphor emission appear as white light.

Emission characteristics



LED + Phosphor

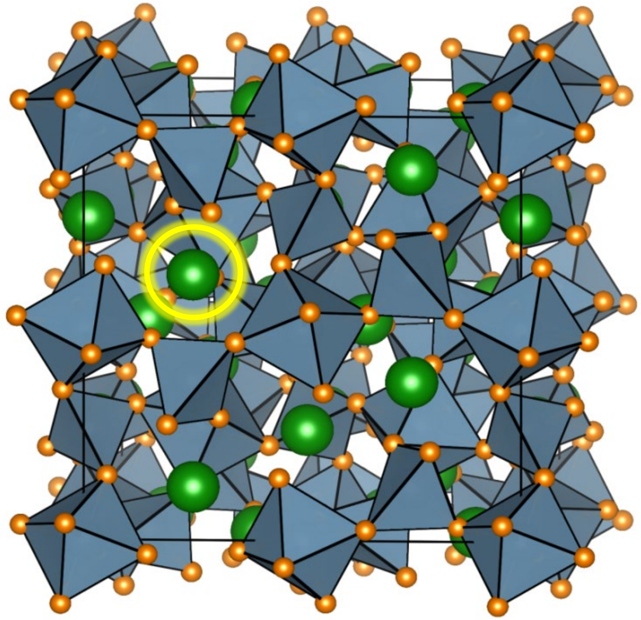
Radiation from LED
(absorbed by phosphor)

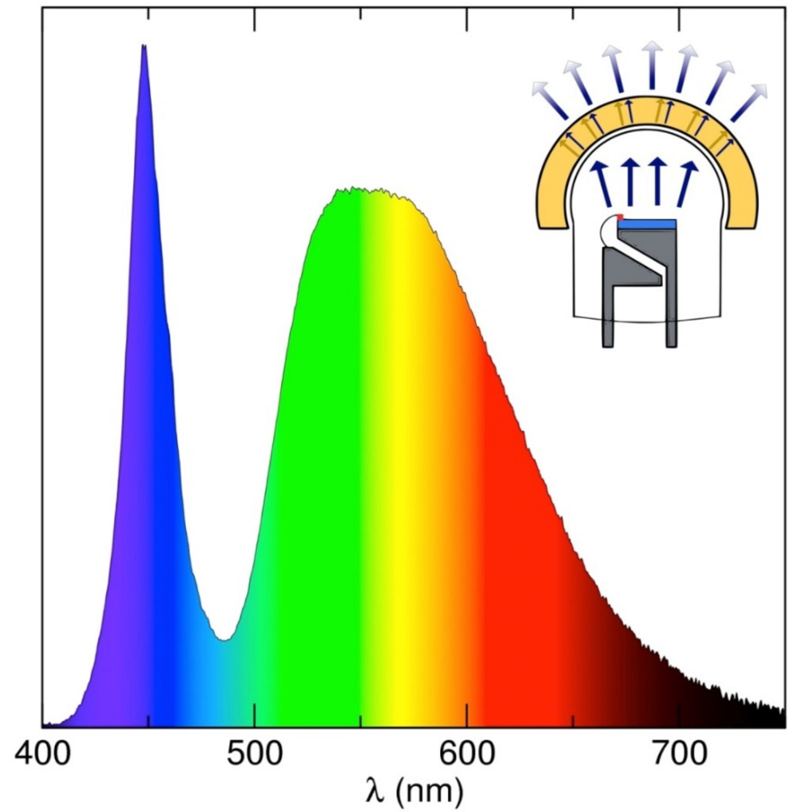
Radiation from phosphor down-conversion
(emission from phosphor)

George, Denault, and Seshadri *Annu. Rev. Mater. Sci*, **2013**, *43,* 2.1:2.21

Cerium-substituted yttrium aluminum garnet: The prototypical phosphor

$(Y_{2.97}Ce_{0.03})Al_5O_{12}$





YAG:Ce^{3+} is a widely used phosphor because of its high photoluminescent quantum yield (Φ = 80% to 95%)

Absorbs blue and then down-converts to a yellow emission

George, Denault, and Seshadri *Annu. Rev. Mater. Sci*, **2013**, *43,* 2.1:2.21

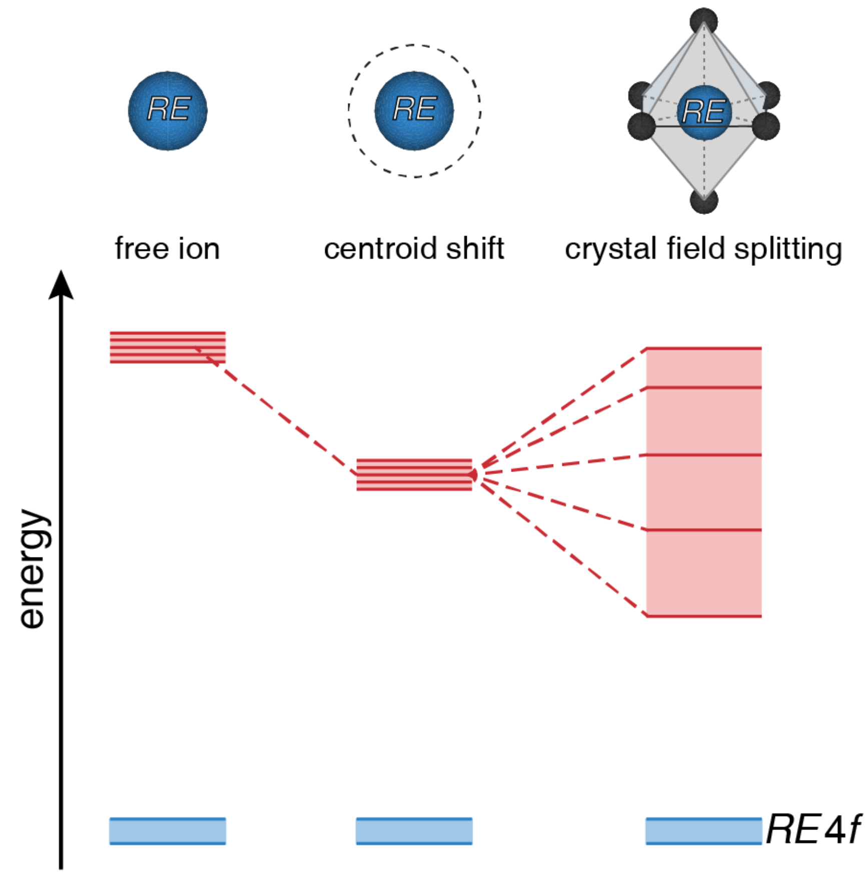
Mechanism of phosphor emission for white LEDs

The difference in energy between the 5*d* and 4*f* orbitals of free ion is in the UV region

5*d* orbitals stabilize when placing in a host

The different degree of covalency between the activator ion and surrounding anions causes the 5*d* orbitals to split

The well-shielded 4*f* orbitals are not strongly affected



George, Denault, and Seshadri *Annu. Rev. Mater. Sci*, **2013**, *43,* 2.1:2.21

Information on centroid shift and crystal field splitting

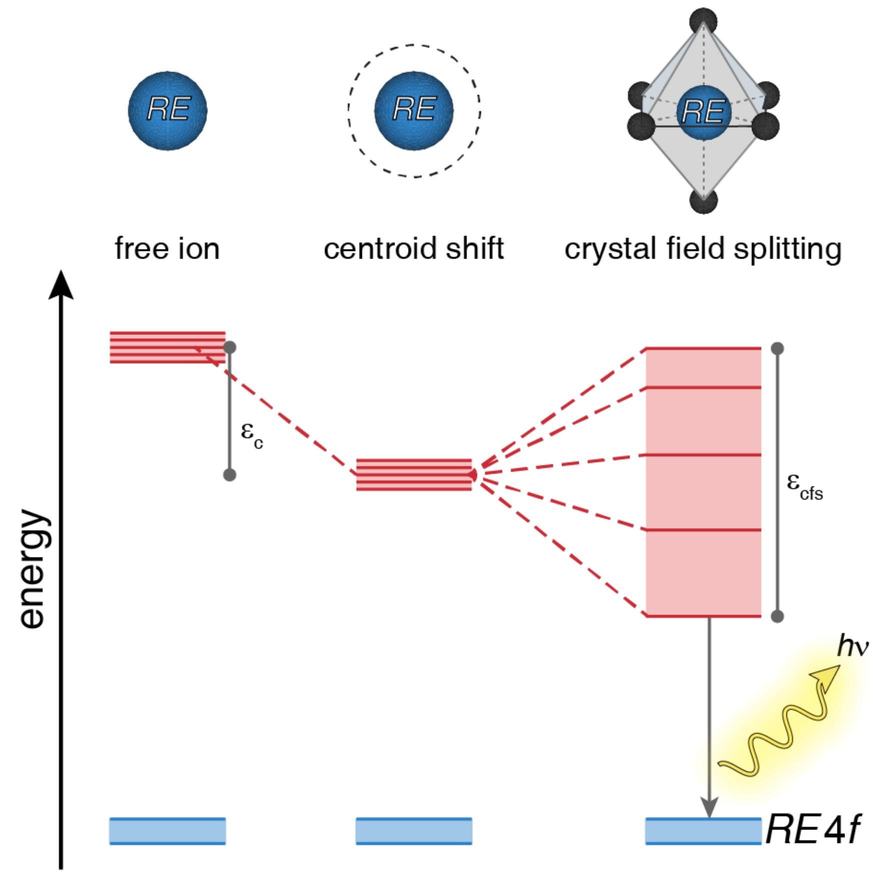
D(A) is determined by centroid shift (ε_c) and crystal field splitting (ε_{cfs})

ε_c - centroid shift

ε_{cfs} - size and shape of rare-earth polyhedron

ε_{cfs} and ε_c behave independently.

Access of ε_{cfs} and ε_c gives insight into the optical properties



P. Dorenbos, *Phy. Rev. B,* **2000**, *62*, 15640-15649.

Host band gap also plays an important role

D(A) is determined by centroid shift (ε_c) and crystal field splitting (ε_{cfs})

ε_c - centroid shift

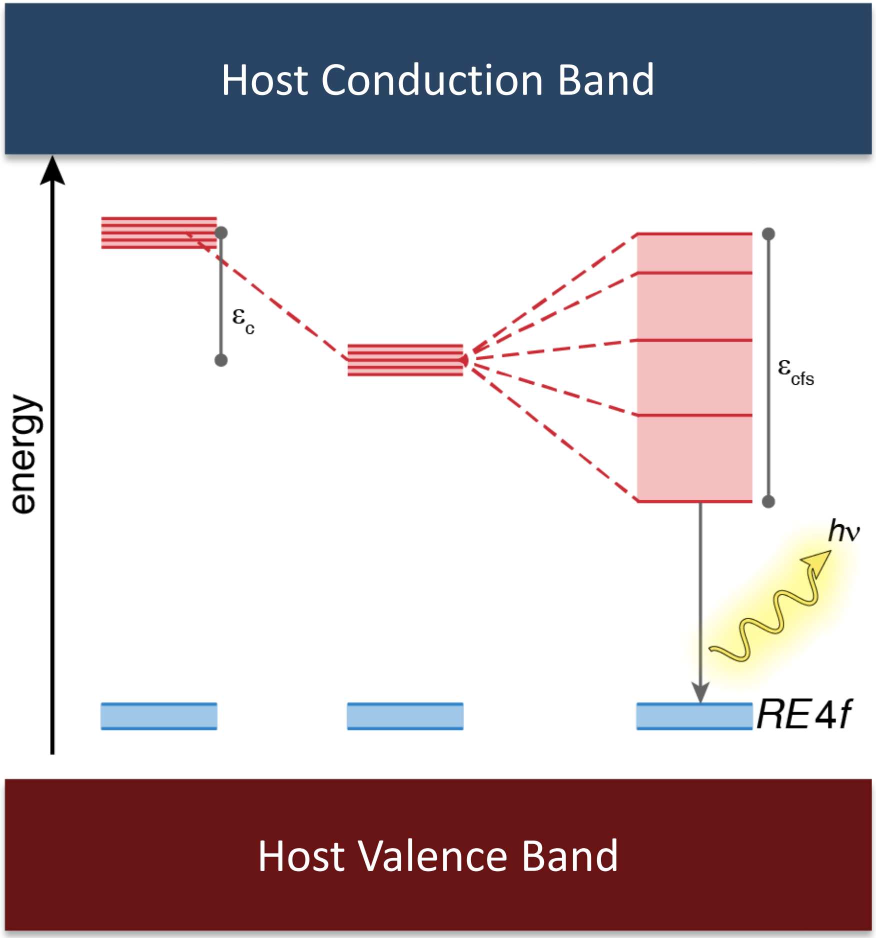
ε_{cfs} - size and shape of rare-earth polyhedron

ε_{cfs} and ε_c behave independently.

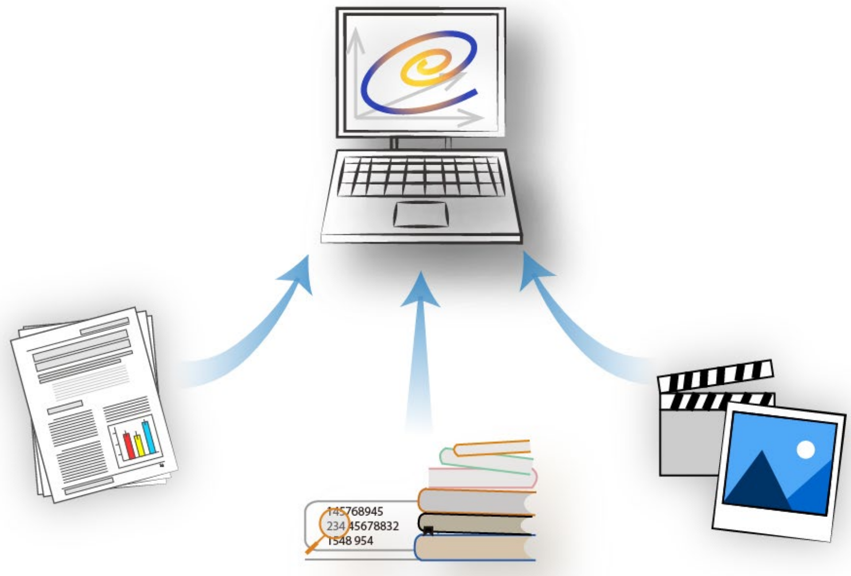
Access of ε_{cfs} and ε_c gives insight into the optical properties

The luminescence occurs in the band gap

- Need wide band gap materials that can accommodate Ce^{3+}



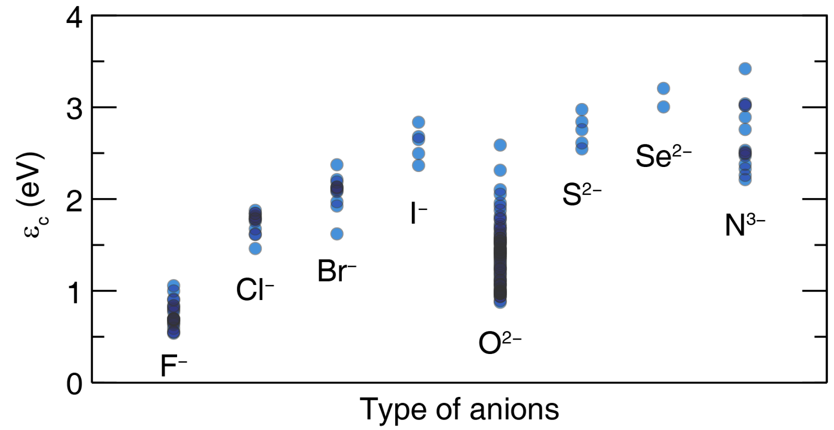
Using data science to predict the optical properties of phosphors



Centroid shift from 160 Ce^{3+} phosphors collected and curated in a database

- In collaboration with Pieter Dorenbos – TU Delft

Centroid shift (ϵ_c) is closely related to host anion



Centroid shift from 160 Ce^{3+} phosphors collected and curated in a database

- In collaboration with Pieter Dorenbos – TU Delft

Fluorides have smallest centroid shift because of its ionic nature

Oxides and halides tend to have a wide range of centroid shift

Nitrides, sulfides, and selenides possess the largest centroid shift due to their extensive covalent bonding

Y. Zhuo, S. Hariyani, S. You, P. Dorenbos, and J. Brgoch *J. Appl. Phys.* **2020**, *128*, 013104.

Developing a physics-based feature set for machine learning

An ensemble learning method was constructed to predict ϵ_c using features based on numerical equations

$$\chi_{av} = \frac{\sum n_i z_i \chi_i}{\sum n_i z_i} \qquad \alpha_{av} = \frac{\sum m_e \alpha_e}{\sum m_e}$$

Variable	**Notation**
Relative permittivity	ε_r
Avg. cation EN	χ_{av}
Avg. anion polarizability	α_{av}
Ionic radius	R_M
Difference in radius	ΔR
Average bond lenghth	R_{av}
Coordination number	n_i
Condensation	cond.

Y. Zhuo, S. Hariyani, S. You, P. Dorenbos, and J. Brgoch *J. Appl. Phys.* **2020**, *128*, 013104.

Predicting centroid shift with ensemble machine learning

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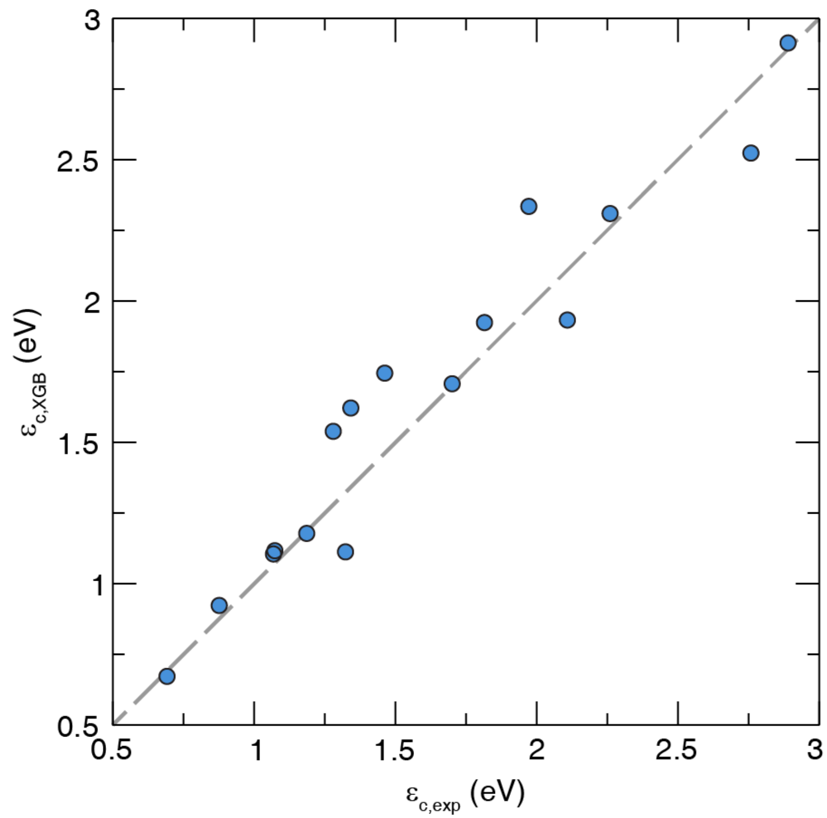


Y. Zhuo, S. Hariyani, S. You, P. Dorenbos, and J. Brgoch *J. Appl. Phys.* **2020**, *128*, 013104.

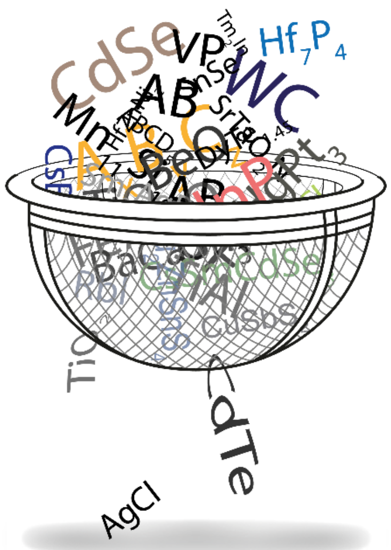
Machine learning also predicts the host bandgap (E_g)

A second machine-learning model was constructed to predict E_g using 4916 experimentally measured values

A classification algorithm was first used to separate metals from nonmetals

- 97% accuracy achieved

	Total
PCD*	102,532
Metals	66,819
Nonmetals	35,371



classification

Y. Zhuo, A. Mansouri Tehrani, and J. Brgoch *J. Phys. Chem. Lett.* **2018**, *9*, 1668-1673.

Using machine learning to predict the bandgap (E_g)

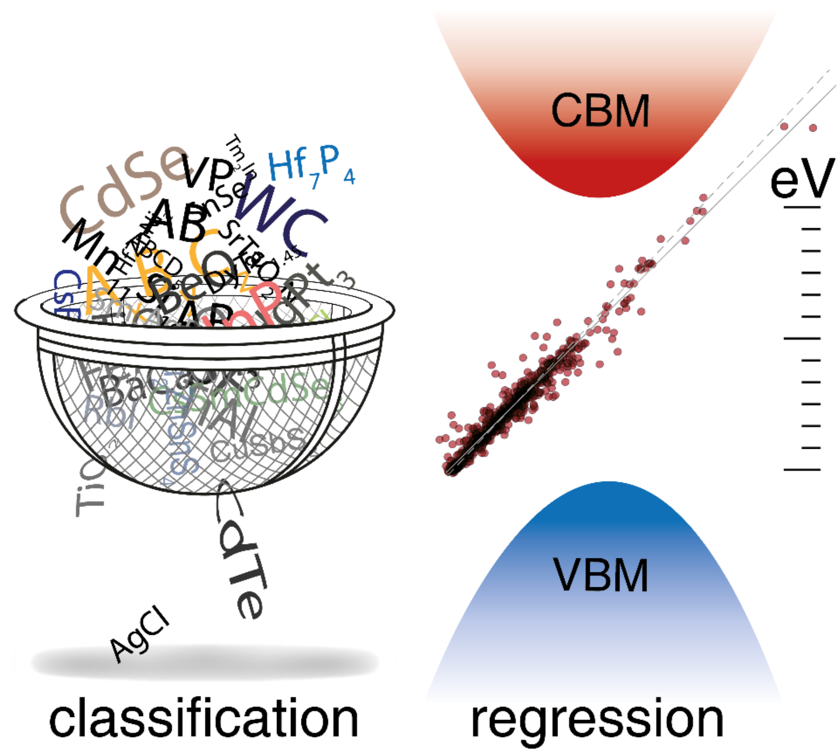
A second machine-learning model was constructed to predict E_g using 4916 experimentally measured values

A classification algorithm was first used to separate metals from nonmetals

- 97% accuracy achieved

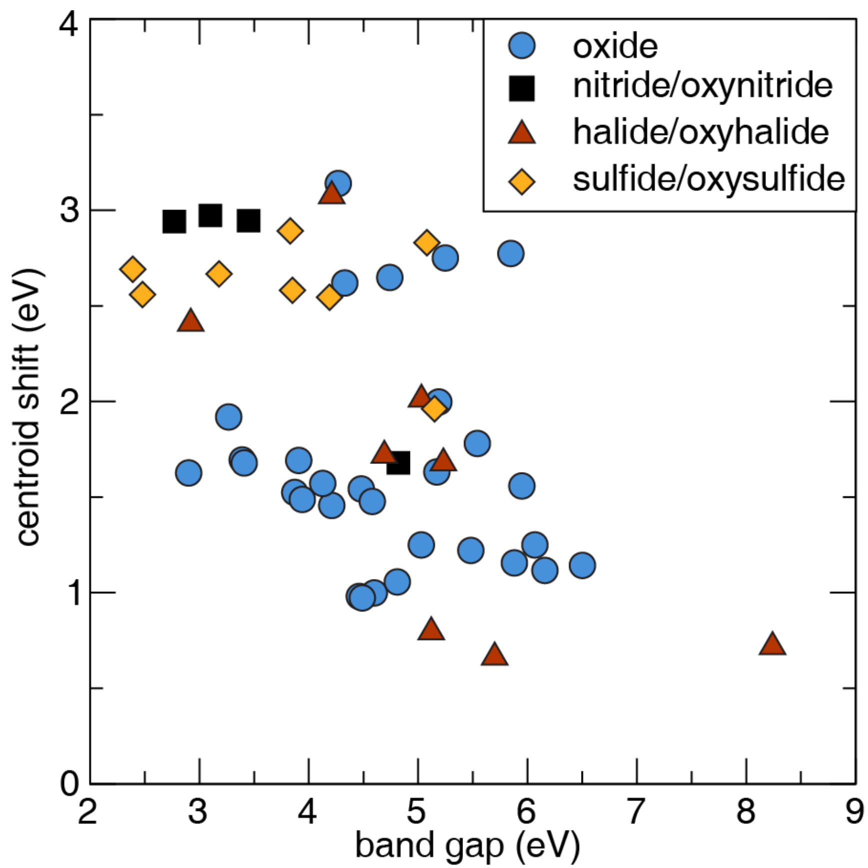
A regression model was then constructed to quantitatively estimate E_g of 35,371 nonmetals contained in PCD

- RMSE = 0.45 eV



Y. Zhuo, A. Mansouri Tehrani, and J. Brgoch *J. Phys. Chem. Lett.* **2018**, *9*, 1668-1673.

Maximizing centroid shift and band gap in inorganic phosphors

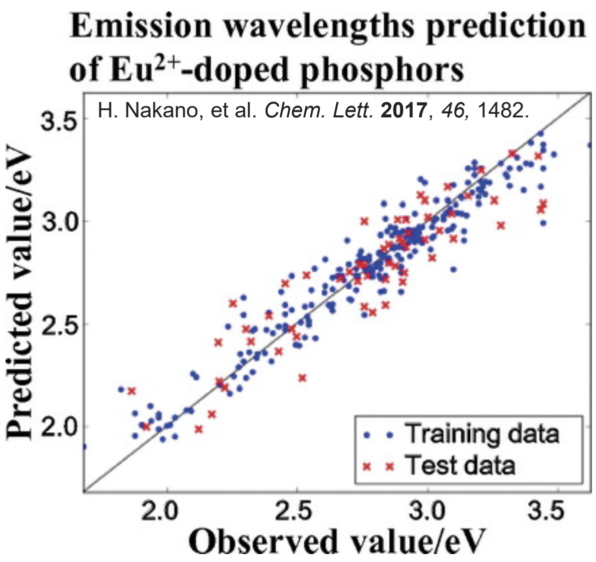


Plotting the machine-learning predicted centroid shift and band gap for unknown compounds provides high-level Ce^{3+} phosphor screening

Y. Zhuo and J. Brgoch, *unpublished.*

Research opportunities for developing inorganic phosphors

Predicting a material's emission spectrum using data science



Accelerated phosphor discovery using robotics

Y. Zhuo and J. Brgoch, *Opportunities for Next-Generation Luminescent Materials through Artificial Intelligence*, *J. Phys. Chem. Lett.* **2021**, *12*, 764.