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DISCOVERY OF NEW PLASMONIC MATERIALS VIA HIGH-THROUGHPUT MACHINE LEARNING

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EXECUTIVE SUMMARY

Plasmonics aims to manipulate light through choice of materials and nanoscale structure. Finding materials that exhibit low-loss responses to applied optical fields while remaining feasible for widespread use is an outstanding challenge. Online databases have compiled computational data for numerous properties of tens of thousands of materials. Owing to the large number of materials and high computational cost, it is not viable to compute optical properties for all materials from first principles.

For this project, plasmonic quality factors for a training set of 1,000 metals and 2,000 semiconductors were computed using density functional theory (DFT) and the Drude model. The research team trained regressors to rapidly screen the Materials Project (MP) database to identify potential new plasmonic metals. Descriptors were limited to symmetry and quantities obtained using the chemical formula. The machine learning models filtered through 7,445 metals in the MP database. From this, the team predicted $AlCu_3$, ZnCu, and $ZnGa_3$ as candidates and verified their quality factors with DFT.

RESEARCH CHALLENGE

As mentioned before, the field of plasmonics seeks to manipulate light at the nanoscale. Precise control over plasmon response enables many applications including subwavelength waveguides [1], nanoantennas [2], superlenses [3], subwavelength imaging [4], nanocircuitry [5], and biosensors [6]. Currently used plasmonic materials consist of noble metals and electron-doped semiconductors with high electrical conductivity [7]. Unfortunately, noble metals suffer from large losses in the visible spectrum owing to absorption while semiconductors require high electron doping concentrations. Further advances of plasmon-based technology require finding new high-performance materials.

METHODS & CODES

To quantify the response of a material to an applied electrical field, the team computed its dielectric function. Two dominant contributions to the dielectric function were considered: interband transitions of electrons from valence states to conduction states and intraband oscillations of electrons near the Fermi energy. Interband transitions were captured through DFT calculations performed using the Vienna Ab initio Software Package (VASP) [8]. The contribution to the dielectric function from intraband oscillations was described by the Drude conduction model with a plasma frequency obtained through DFT. From the dielectric functions, plasmonic quality factors were computed based on analytic equations. DFT input files for 1,000 metals and 2,000 semiconductors to form the training sets were randomly chosen from the MP database with the pymatgen open source library [9,10].

Random forest regressors with adaptive boosting implemented in scikit-learn [11] related material descriptors to the DFT-calculated plasmonic quality factors. The descriptors consist of material properties readily obtainable from the chemical formulas and standard tables, *e.g.*, atomic masses and electronegativities. Constructed models were validated using an 80–10–10 validation scheme. With this approach, 10% of materials in the training set were randomly selected to form a testing set to be left fixed throughout the process. The remaining materials were randomly divided 80%–10% into fitting and validation sets. The fitting set was used to fit the model, with the model subsequently applied to the validation set. Model construction was then iterated over 100 random fitting–validation divisions. Each model was applied to all metals and semiconductors in the MP database to predict quality factors for all available materials. For the 200 materials with the largest predicted quality factors, the dielectric functions and quality factors were explicitly calculated with DFT.

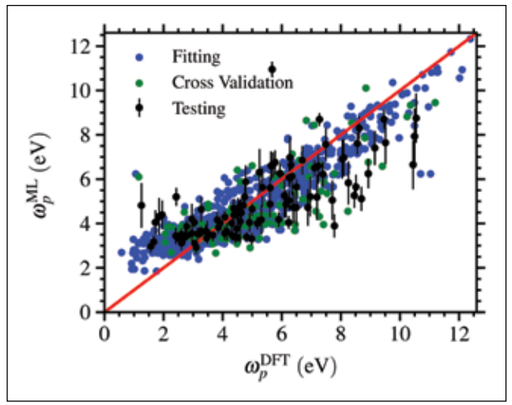

Figure 1: Comparison of metal plasma frequency predicted via machine-learned regressor vs. calculated with DFT. Plotted are the results of one fitting–validation iteration and average values and error bars for 100 iterations for testing. The red line is to guide the eye for machine learning predictions matching the DFT value.

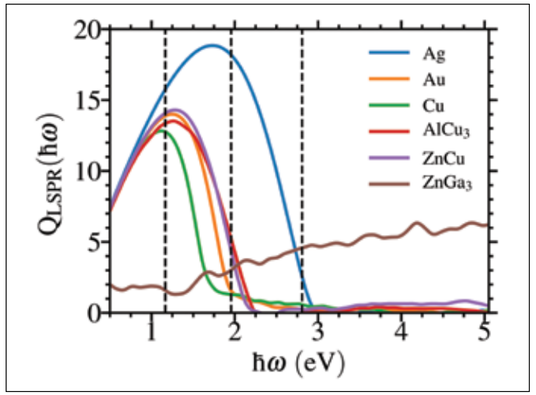

Figure 2: Computed plasmonic quality factor for currently used metals, Ag, Au, and Cu, and proposed new plasmonic metals, $AlCu_3$, ZnCu, and $ZnGa_3$. Vertical dashed lines correspond to operating energies of commonly used lasers. $AlCu_3$ and ZnCu are candidate plasmonic metals at low energy while $ZnGa_3$ shows large-quality factors in the UV spectrum.

RESULTS & IMPACT

We have used DFT calculations of materials' dielectric functions to train and validate machine learning models which predict plasma frequencies and plasmonic quality factors for metals and semiconductors in the Materials Project database. The constructed models allow for rapid calculation of the optical properties necessary to predict the suitability of a material for plasmonic applications. By applying the models to a large database, we have been able to search for predicted high-quality factor metals and semiconductors without requiring explicit DFT calculations for all materials in the database, significantly reducing the computational cost. From the database metals, we have identified three potential new metals, $AlCu_3$, ZnCu, and $ZnGa_3$. $AlCu_3$ and ZnCu are predicted to match or outperform the commonly used Au, and Cu for energies between 1 eV and 2 eV, the infrared and low energy end of the visible spectrum. $ZnGa_3$ shows high-quality factors for energies between 2 eV and 3 eV, outperforming currently used metals for higher-energy plasmonics.

WHY BLUE WATERS

The present research requires DFT simulations for thousands of materials with up to dozens of atoms. The computational cost for DFT calculations is further increased by the need for accurately mapping the electronic energies near the Fermi surface. Dense 31 x 31 x 31 point meshes to sample momentum space are required to obtain converged calculations of the electronic properties near the Fermi energy. This imposes challenging requirements for CPU hours that cannot be met by a system such as the Campus Cluster. The computational cost of DFT scales as the cube of the number of electrons; for large system sizes such as in this project, available computing power can quickly become a limiting factor. Efficiently carrying out electronic structure DFT calculations for this work requires nodes with fast communication. In addition, this research requires multiple of these runs and, hence, it needs a machine such as Blue Waters that allows the research team to routinely carry out this work.