Toward Enhanced Single-Molecule Cross-Sections in 3D Photonic Crystals

Brandon G. Hacha, Sam J. Evans, Anders J. Bergsten, Michael S. Mattei, Zongfu Yu†, Randall H. Goldsmith*

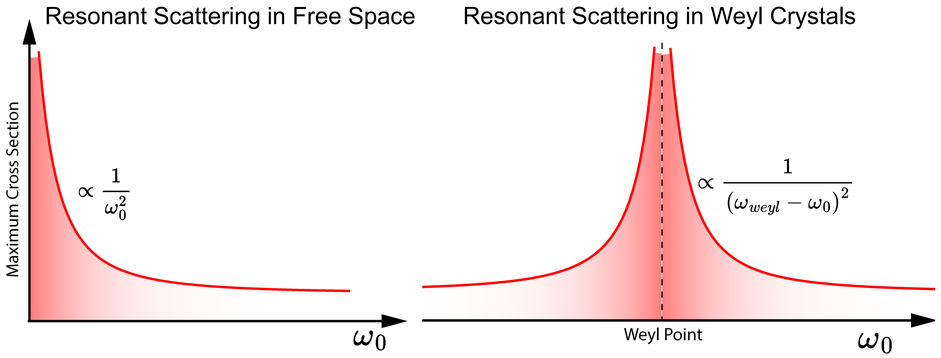
Department of Chemistry, University of Wisconsin-Madison, Madison, WI

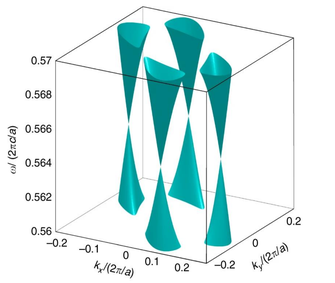
†Department of Electrical and Computer Engineering, University of Wisconsin-Madison, Madison, WI

Background and Motivation

- Photonic crystals provide unique optical environments, where the dispersion relation can be substantially different from that of free space
- Certain point degeneracies in momentum space, Weyl points, can enhance resonant cross sections at that frequency.

Can we use Weyl points for single-molecule spectroscopy?



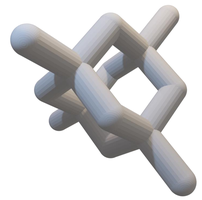


Weyl points in momentum space[1]

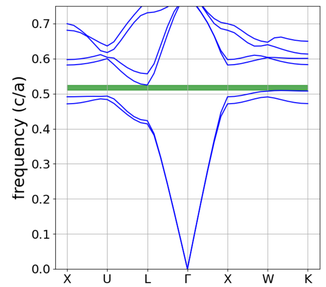
We have developed a fabrication process for making 3D photonic crystals with high refractive index contrast, the first step towards Weyl points at visible wavelengths

Simulation and Design

- Our fabrication method will result in a core-shell design, where a low-index core is coated in a high-index material.
- We use a custom Python package to create rod-connected diamond crystals that have a relatively large band gap.
- After simulating, we export the crystals for fabrication[2]



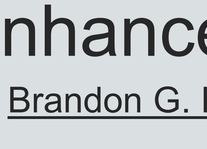
The conventional unit cell of a rod-connected diamond crystal



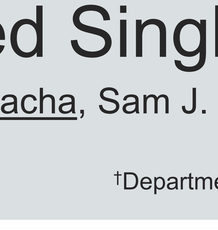
A simulated band structure diagram for a core-shell diamond crystal

Focused Electron Beam Induced Deposition (FEBID)

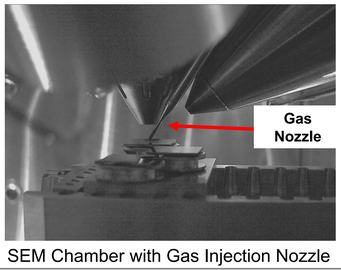
- A scanning electron microscope's electron beam is used to reduce a precursor gas
- Controlling beam position and exposure time allows for growing these deposits into nanoscale structures



(Trimethyl)methyl-cyclopentadienylplatinum(IV)



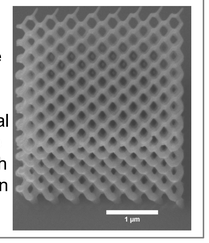
An early photonic crystal



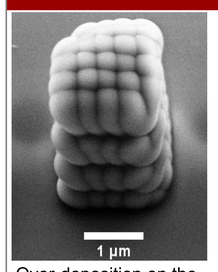
SEM Chamber with Gas Injection Nozzle

As-fabricated diamond-lattice crystal

The same crystal is shown in the next section with ~20 nm of silicon coating

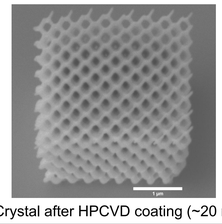


High-Pressure Chemical Vapor Deposition (HPCVD)



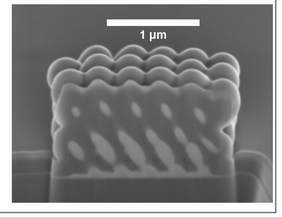
Over-deposition on the FEBID crystal shown to the left

- The FEBID material has too low of a refractive index, so we coat it in amorphous silicon
- Conventional low-pressure CVD is a ballistic process (mean free path around 1 micron)
- At high pressure, the process becomes diffusive and the deposition becomes conformal (mean free path around 1 nanometer)



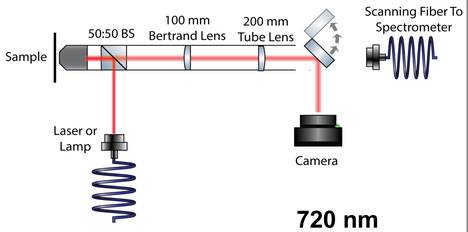
Crystal after HPCVD coating (~20 nm)

A FIB milled crystal showing conformal deposition. The light gray regions are the FEBID deposits and the dark gray is silicon.



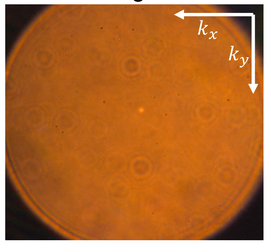
Optical Characterization

- Back focal plane reflectometry allows measuring angle-resolved reflection without rotating the sample

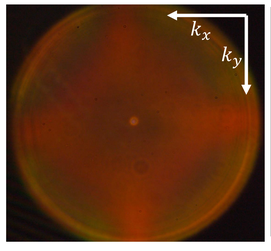


White Light

Background

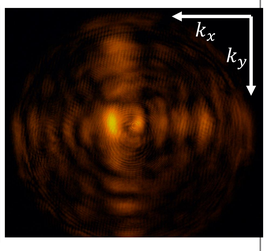


Diamond



720 nm

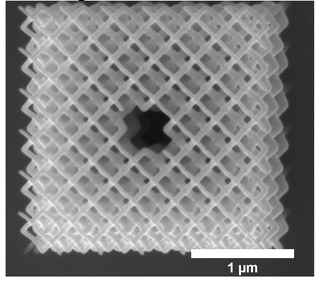
Diamond



Conclusions and Next Steps

- We can design, fabricate, and coat photonic crystals with band gaps in the visible and NIR.
- Comparing measurements to simulation will give information about our materials' optical properties
- Before Weyl points, we will characterize crystals with defects that emulate resonant features

Crystal Defects



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References

1. Zhou, M. et al. *Nat Commun* **2017**, *8* (1), 1388.
2. Skoric, L et al. *Nano Lett.* **2020**, *20* (1), 184–191.

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