Out of the fire: Extracting the WAXS signal of soot

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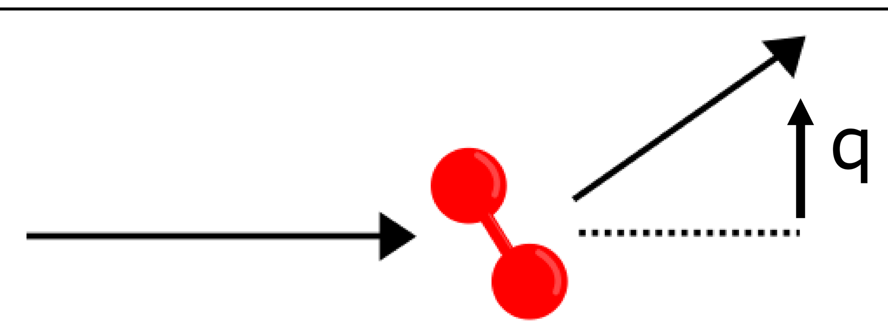
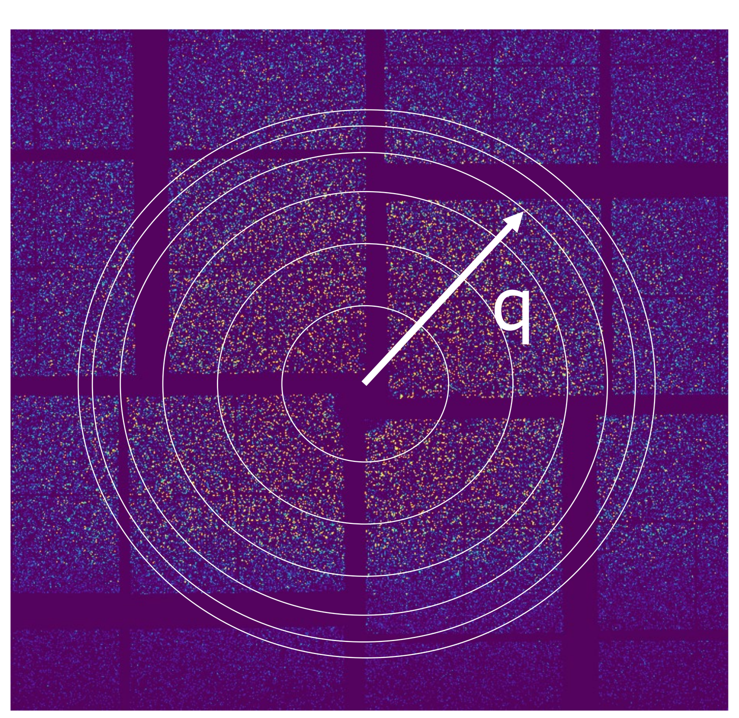
Introduction

Soot is a hazardous product of incomplete combustion in a flame. Previous synchrotron experiments have attempted to use Wide-Angle X-ray Scattering (WAXS) to study the in-situ composition of soot but have largely been unsuccessful. With the increased brightness provided by the free-electron laser at LCLS, we sought to determine the evolution of the chemical composition of soot using WAXS.

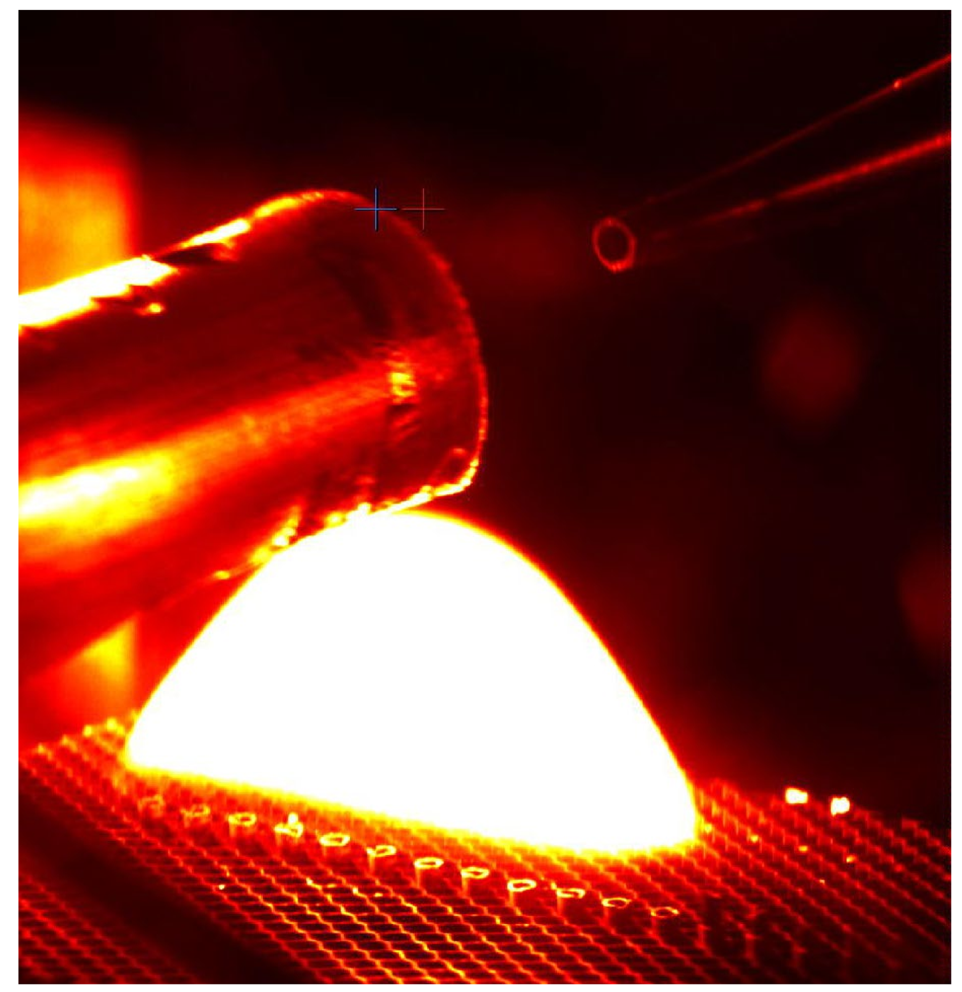
Wide Angle Scattering



When a photon elastically scatters off a molecule, it ricochets off with a change of momentum q, dependent of the distance between the atoms and the type of atom.



Since the sample is non-crystalline, the signal is diffuse and weak compared to Bragg diffraction. Thus, we needed the increased brightness of LCLS to get a signal.
By integrating over all angles, we can get a signal for a given value of q.

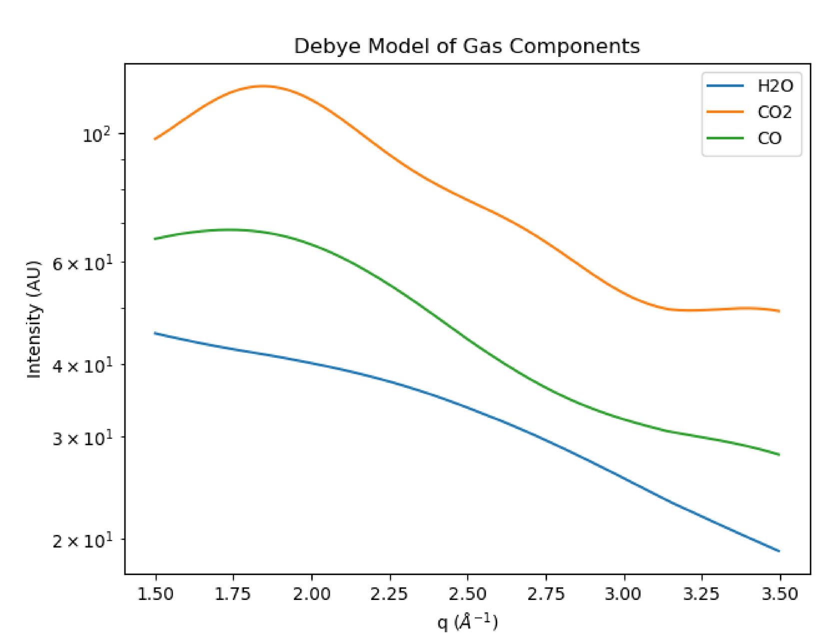


The experimental set up, with the X-ray beam passing through a variable height burner.

The Debye Equation

$$I_M(q) = \sum_{a=1}^{N_{at}} \left(|f_\alpha(q)|^2 + S_\alpha\right) + \sum_{\alpha \neq \beta}^{N_{at}} f_\alpha(q) f_\beta(q) \frac{\sin(qr_{\alpha\beta})}{qr_{\alpha\beta}}$$ [1]

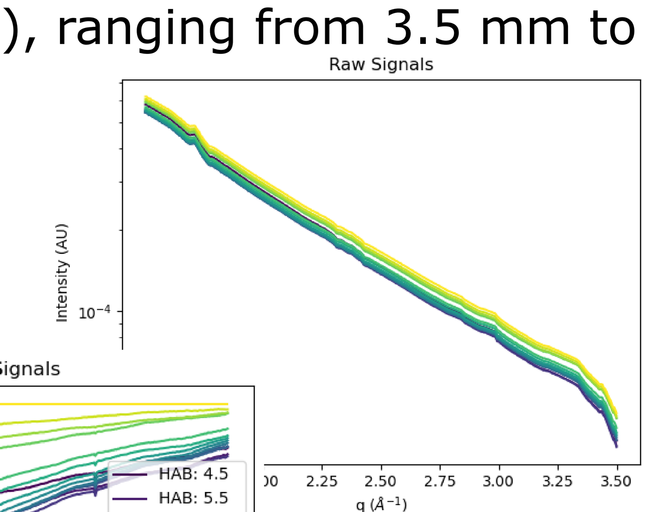
The Debye equation models how X-rays will scatter from molecules. The dominant component sums up a function of all distances between each pair of atoms. Thus, it contains information on the atomic structure of each molecule.

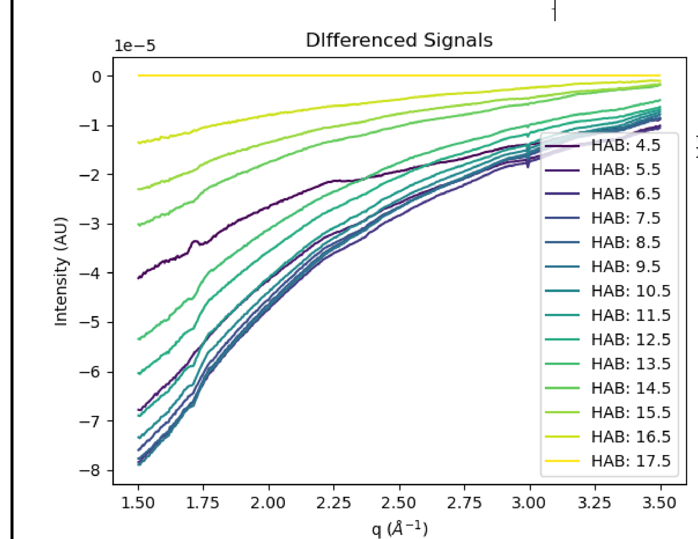


We used this equation to simulate the expected signal from the three largest gas phase components of the flame: water, carbon dioxide, and carbon monoxide.

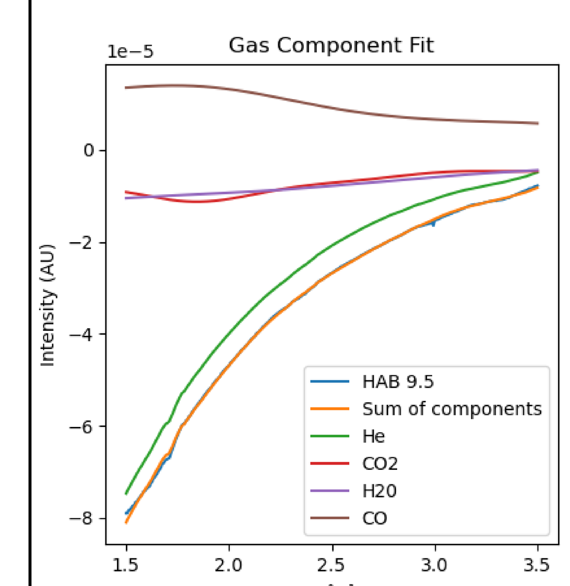
Analysis of Data

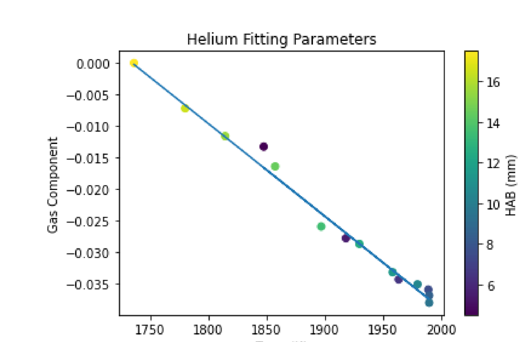
The composition and temperature of soot changes over its evolution in a flame. We measured WAXS at various heights above the burner (HAB), ranging from 3.5 mm to 17.5 mm.

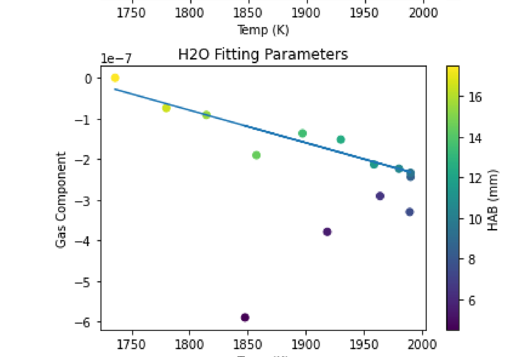




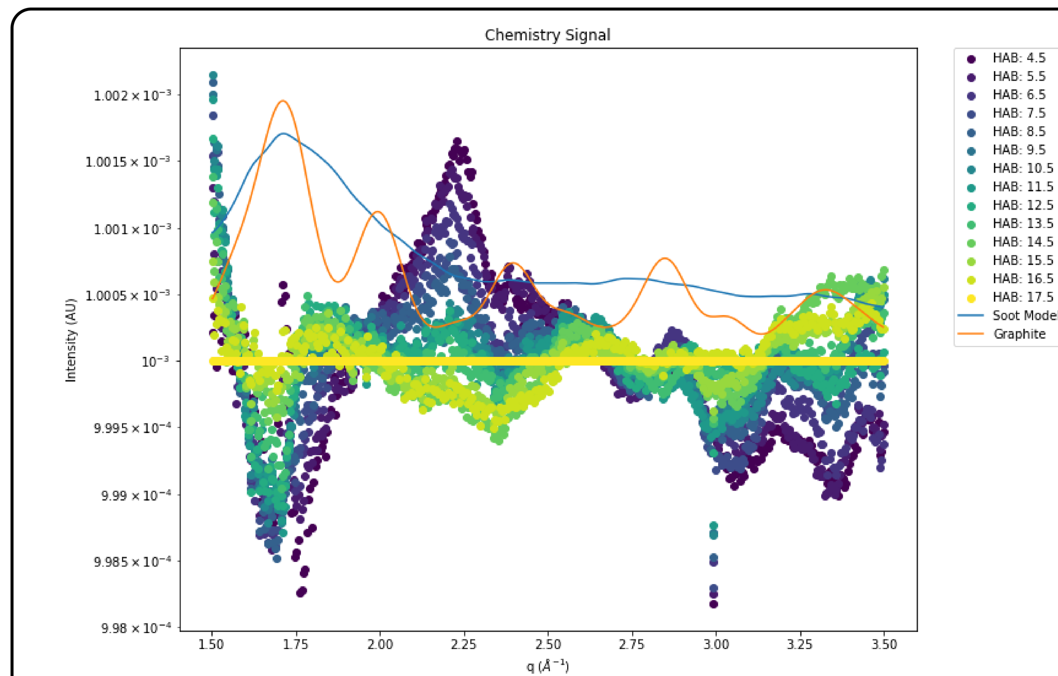
Each of the different HAB signals have similar static backgrounds. So, we expect the largest signal component to be a constant that is HAB-independent. Subtracting off the 17.5 mm signal removes this part.



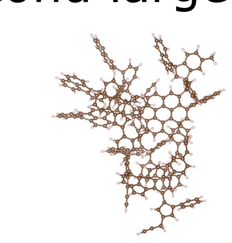




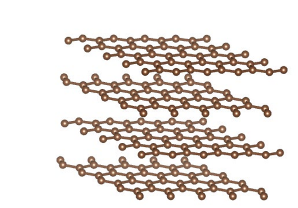
The second largest background component is the signal from the most prominent gasses: H_2O, CO_2, CO, and He. (The helium WAXS signal was taken from a flameless background run.) These components were fit to each signal using least squares linear regression. We would expect these fitting parameters to be linear with temperature except at HABs where the gas is produced by the reaction. We see this at all HABs for He and at HABs above 9mm for H_2O.



Subtracting off the gas phase components leaves only the signal attributable to the complex chemistry of combustion and soot formation. This signal has clear HAB-dependent peaks, some of which line up with predicted features in graphite and soot models, such as the large peak at 1.6 and smaller peak at 2.85. Other features are unexplained by the model, such as the second large peak at 2.2.



Soot Model



Graphite Model

Conclusions

By combining multiple methods of background subtraction and curve fitting, we were able to extract a smoothly varying HAB-dependent signal. Some of the features could indicate the presence of our predicted model of soot, but future modeling will be required to explain the entirety of the signal.

Acknowledgments

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[1] Brian Stankus et al 2020 J. Phys. B: At. Mol. Opt. Phys. 53 234004