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Designing a Reactor Chamber for Hot Electron Chemistry on Bimetallic Plasmonic Nanoparticles

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Designing a Reactor Chamber for Hot Electron Chemistry on Bimetallic Plasmonic Nanoparticles

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SURF Summer Undergraduate Research Fellowship
Chapman University Center for Undergraduate Excellence
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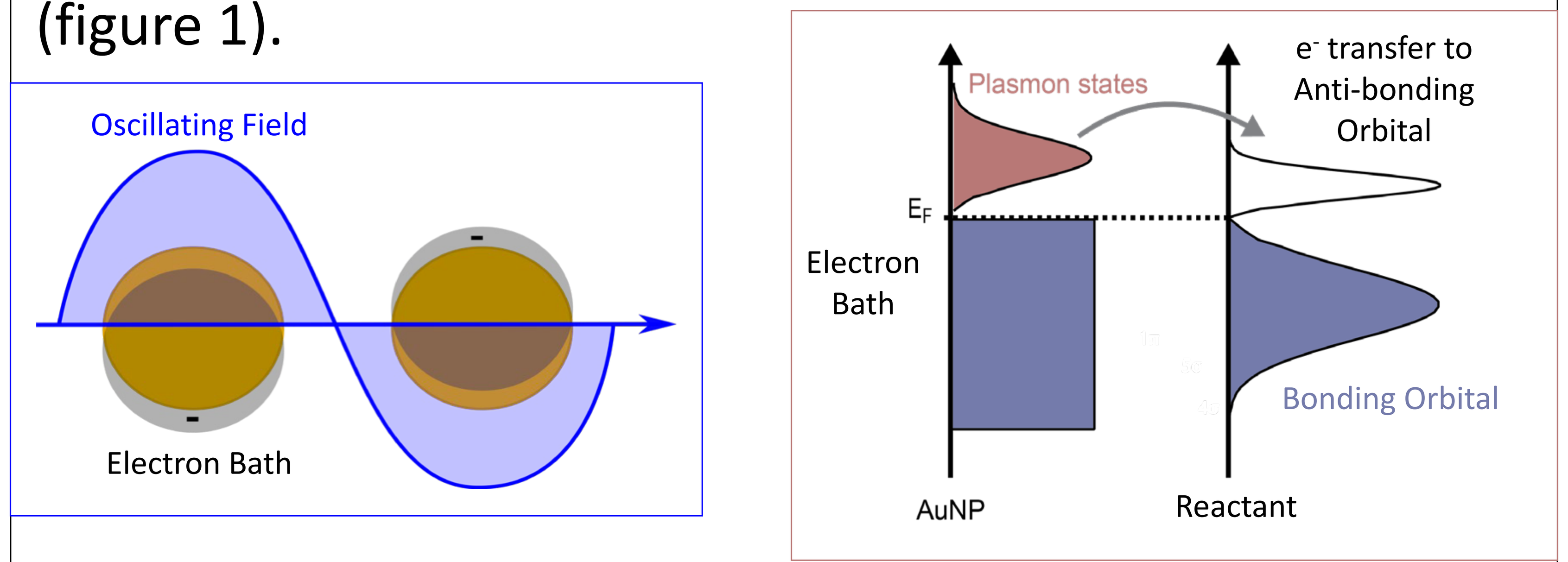
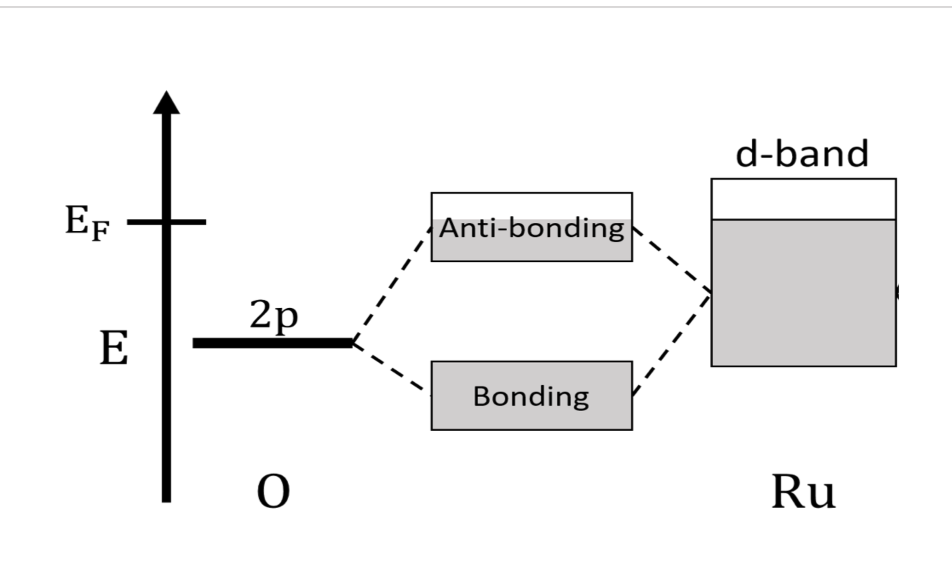
Background

Gold nanoparticles (AuNPs) are potentially effective photocatalysts due to their strong localized surface plasmon resonances (LSPRs). LSPRs are the oscillations of the electron bath excited by light at the surface of the nanoparticles (NPs). The LSPR decay into excited electrons that can occupy excited states when radiated with light (figure 1).



▲ ***Figure 1.*** *Excited electrons are created at the surface of the AuNP and transferred to the antibonding orbital of the reactant. Figures courtesy of Dr. LaRue.*

Excited states, such as antibonding orbitals, are destabilizing when occupied by electrons. Electron occupation of destabilizing antibonding orbitals can be facilitated by introducing a transition metal shell to the core AuNPs (figure 2).

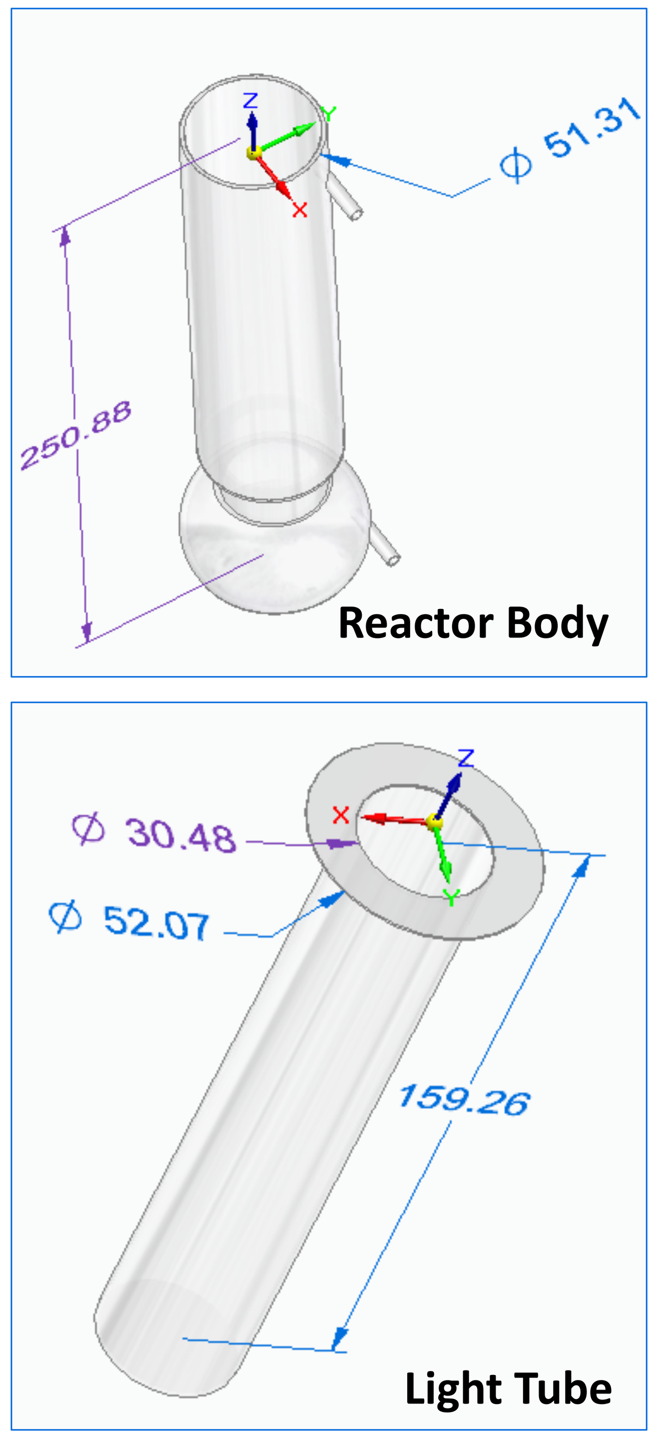


◄ ***Figure 2.*** *The d-band orbital of transition metal Ruthenium (Ru) facilitating the transfer of bonding orbital electrons to oxygen anti-bonding orbitals. Figure from LaRue et al.*[1]

Ruthenium was used in this experiment to create bimetallic ruthenium-gold nanoparticles (RuAuNPs). The reactivity of the RuAuNPs will be assessed by running well modeled reactions, such as CO Oxidation (Eq. 1) using a single-tangent reactor chamber (figures 3 and 4). Future reactions include CO hydrogenation and Nitrogen Oxide Reduction.

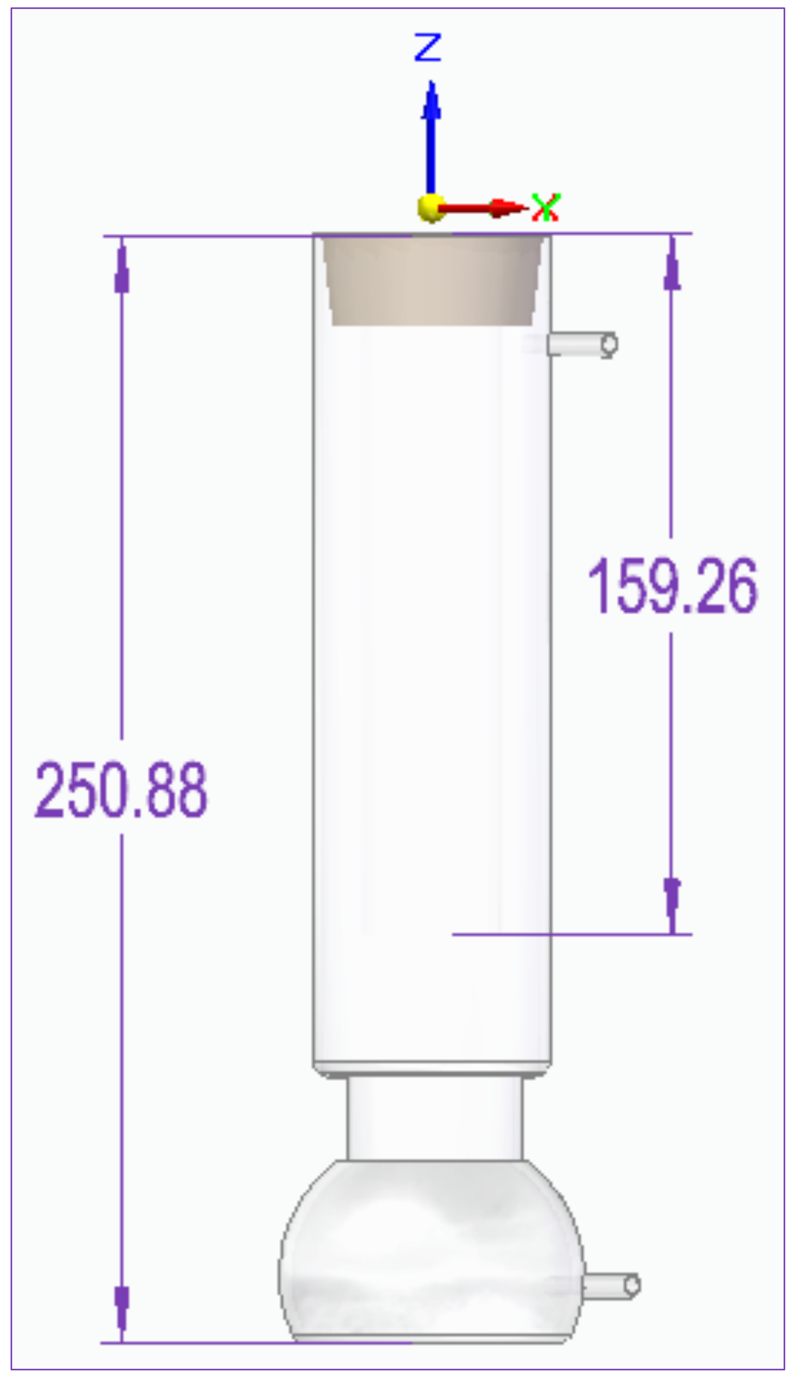
$$2CO + O_2 \longrightarrow 2CO_2 \qquad \text{Eq. 1}$$

Single-tangent Reactor Chamber



▲ ***Figure 3.*** *The Reactor body (above) houses the light tube (below) with space between for the RuAuNP sample on a quartz fiber felt substrate.*

▼ ***Figure 4.*** *The assembled single-tangent reactor chamber with an inert rubber stopper for an airtight reaction environment.*

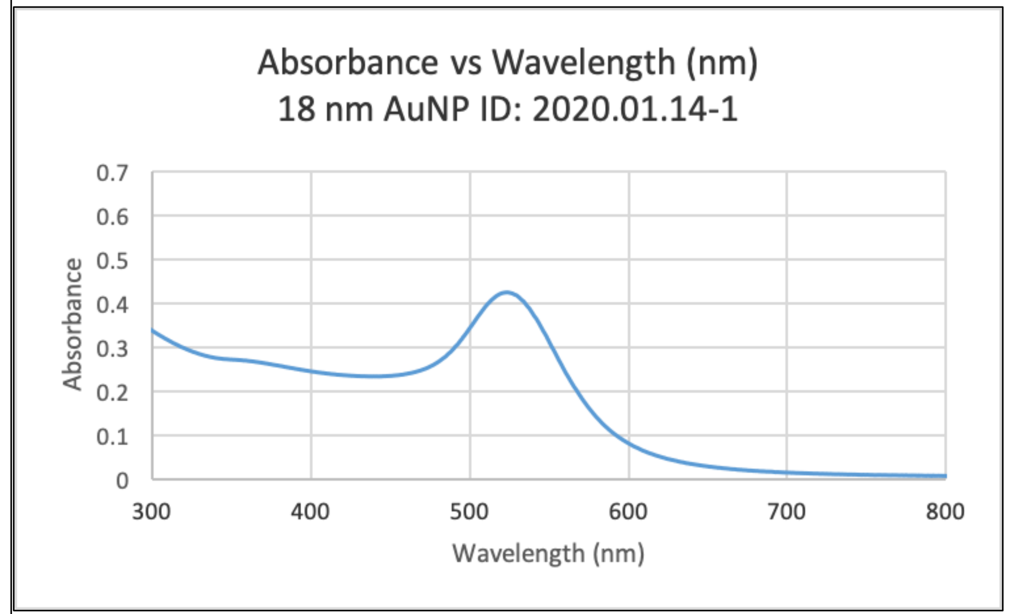


A light source (similar to figure 5) will be inserted into the light tube via a fiber optic cable. A mass spectrometer (similar to figure 6) will identify the compounds produced.

◄ ***Figure 5.*** *Visible LED Light Source with a fiber optic coupling output from Newport*^{TM}*.* https://www.newport.com/p/66088-LED

► ***Figure 6.*** *Residual Gas Analyzer Mass Spectrometer from Newport*^{TM} *with an open ion source.* https://www.newport.com/p/EV2-210-000FT

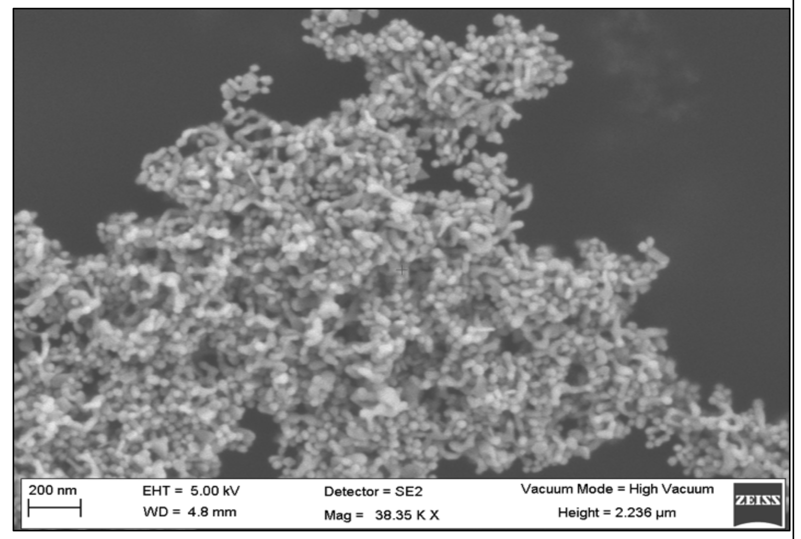
Nanoparticles

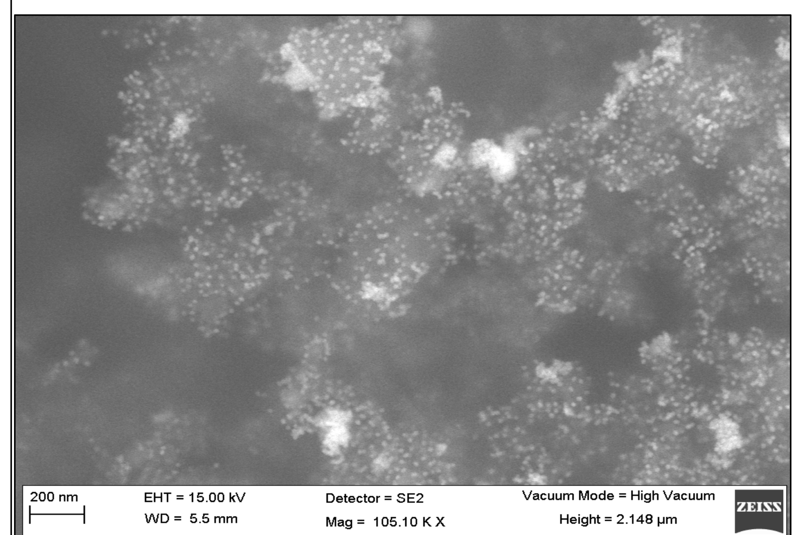


◄ ***Figure 7.*** *UV-Vis absorbance spectrum of 18 nm AuNP solution synthesized on 01/14/2020. This spectrum shows the light absorbance of AuNP LSPRs.*

Figure 7 shows the greatest absorbance of light by the AuNPs' LSPRs occur at about 530 nm, which means that the hot electrons are produced most effectively when irradiated with green light. The scanning electron microscope images in figures 8 and 9 show clusters of AuNPs and RuAuNPs respectively.

► ***Figure 8.*** *SEM image of 18 nm AuNP solution synthesized on 01/14/2020. 200 nm bar on bottom left for scale.*





◄ ***Figure 9.*** *SEM image of RuAuNP sample synthesized on 01/15/2020. 200 nm bar at bottom left shows scale.*

Conclusion

UV-Vis and SEM have shown that the synthesized nanoparticles have the ideal physical characteristics for photocatalysis. Testing the photocatalytic activity of the bimetallic nanoparticles will begin with CO Oxidation once the single-tangent reactor chamber is machined.

References

1) LaRue, J. L.; Katayama, T.; Lindenberg, A.; Fisher, A. S.; Öström, H.; Nilsson, A.; Ogasawara, H., THz-Pulse-Induced Selective Catalytic CO Oxidation on Ru. *Physical Review Letters* **2015,** *115* (3), 036103.