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14. ABSTRACT Density functional theory (DFT) and time-dependent DFT (TDDFT) have been employed to examine the optical absorption spectra of bare and passivated (thiolate-, phosphine-, or carboxylate-stabilized) gold and silver nanoparticles. The location of the ligands is responsible for causing splitting in the optical absorption spectra of thiolate-stabilized nanoparticles. The choice of metal and ligand affects the thermodynamic stability of the "staples" on the surface of the nanoparticle, and may affect the growth of these nanoparticles. The tail group on the ligand does not greatly affect the optical properties, although it does affect the redox potentials. For bare nanoparticles, sharp optical absorption spectra can be obtained depending on the number of electrons in the system; when the number of electrons is a "magic" number for a given shape, the spectrum is typically sharp. For small gold nanoparticles, interband transitions are also important and can be analyzed differently than intraband transitions. The width of the absorption peak is affected by temperature. For nanoparticle aggregates, the main effect on the spectrum arises from symmetry-lowering. However, orientation also plays a role.					
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FINAL PERFORMANCE REPORT

Structure and Optical Properties of Noble Metal Nanoparticles

Christine M. Aikens

Objectives

The statement of objectives for the work was:

- 1) Understand the structural and electronic origins and therefore the conditions under which gold and silver nanoparticles display broad/molecular or sharp surface plasmon resonance (SPR) optical absorption behavior.

The inconsistency between the sharp optical spectrum of Ag_{20} and related tetrahedra, the “molecular” optical spectrum of Au_{20} and $\text{Au}_{25}(\text{SCH}_2\text{CH}_2\text{Ph})_{18}^-$, the broadened optical spectrum and essentially nonexistent SPR peak of 1-2 nm gold particles, the sharp or molecular optical spectrum of 2 nm silver particles, and the narrow SPR peak of larger (> 5 nm) gold and silver nanoparticles is not understood at the current time. One of the aims of this proposal is to fill this gap in fundamental understanding so that we can control the optical properties of nanoparticles in the 1-5 nm regime.

- 2) Quantify the size-dependence of the absorption peak locations as a function of core shape, composition, and passivating ligand so that we can predict the optical properties of 1-20 nm clusters and choose synthetic targets for use in devices and sensors.

Because the goal of this research is to ascertain systematic structure-property relationships for use in designing nanostructured materials with prescribed optical properties, the association of the nanoparticle composition and structure to the optical absorption spectrum must be well understood. The **size**, **shape**, and **composition** of the metal core and the **head group**, **alkyl or aryl linker**, and **functionalized tail** are individual nanoparticle structural parameters that may be modified to change the **peak location**, **intensity**, and **full width at half maximum** (FWHM) of the predicted experimental optical absorption spectrum. **Efficient methods** of determining the FWHM will be assessed.

- 3) Predict distance and orientation effects on the SPR location and width of interacting nanoparticle dimers for use in photonic and plasmonic applications.

Aggregation of particles leads to changes in **peak location** and **intensity**, which depend on the **interacting metal cores** and the **interparticle distance** and **orientation**.

Technical Approach

Density functional theory (DFT) was the primary method employed to achieve the work described in this report. DFT is an electronic structure method based on quantum mechanics that is applicable to systems containing several hundred atoms such as the nanoparticles investigated in this work. DFT in its current form requires an approximation to the exchange-correlation functional; this research utilizes several different types of DFT including local density approximation (LDA), generalized gradient approximation (GGA), and asymptotically-corrected functionals. The Aikens group has found that LDA functionals are inexpensive and accurate for determining the geometric structure of noble metal nanoparticles. GGA functionals are useful when energetic quantities such as reaction energies and binding energies are needed, since LDA functionals are not accurate enough for this data. Asymptotically-corrected functionals have been found to be the most accurate for determining excited states and optical absorption spectra of gold and silver nanoparticles; excitation calculations are performed using linear-response time-dependent DFT (TDDFT).

Gold is a heavy element, and scalar relativistic effects must be included in quantum chemical calculations on systems containing this element. In the work performed by the Aikens group, scalar relativistic effects are treated using the zeroth order regular approximation (ZORA). For consistency, ZORA is used for both gold and silver nanoparticles.

The Amsterdam Density Functional (ADF) program (a commercial software package) is used for the work described in this report.

Because DFT is still limited to systems with several hundred atoms, one additional approach examined by the Aikens group involved treating a nanoparticle as a particle in a spherical well surrounded by ligands that were modeled as point charges. The effects of the ligands on the orbital energy levels were determined by employing degenerate first order perturbation theory. This new method is called the charge-perturbed particle in a sphere (CP-PIS) model.

Accomplishments/New Findings

One of the hypotheses related to objective 1 was that the unique “staple” motif inherent in thiolated gold and some thiolated silver 1-2 nm nanoparticles is responsible for the “molecular” nature of the optical absorption spectrum of these systems. Several studies undertaken by the Aikens group have examined the origin of the optical absorption spectra of gold-thiolate and silver-thiolate nanoparticles and have shown that delocalized molecular orbitals in the nanoparticle core formed from the 6s(gold) or 5s(silver) atomic orbitals lie near the HOMO-LUMO gap. These delocalized orbitals are similar to orbitals expected for particles in a spherical well (for nanoparticles with approximately spherical cores) or particles in a cylindrical well (for nanorods and nanowires or other elongated nanoparticles). Since the core of a nanoparticle is well-described by a particle in a sphere (PIS) model, the Aikens group developed a new charge-perturbed PIS (CP-PIS) model to represent ligand-passivated nanoparticles. This relatively simple model provides a correct qualitative description of the orbital splittings in thiolated gold and silver nanoparticles. This splitting is responsible for the molecular-like optical absorption spectrum as well, so the CP-PIS model shows that the ligand arrangement is indeed responsible for the structured optical absorption spectra observed.

A second hypothesis related to objective 1 was that control over the thermodynamic stability of ligand-core interactions affects the final nanoparticle geometric structure (and hence its optical absorption spectrum). A study on the fragmentation energies of $\text{RS}(\text{AuSR})_n^-$ and $\text{RS}(\text{AgSR})_n^-$ ($\text{R} = \text{H}, \text{CH}_3, \text{Ph}$; $n = 1-4$) was proposed and carried out. The Aikens group determined that binding energies for silver-thiolate systems are smaller than those for gold-thiolate systems; in addition, binding energies for phenyl thiolate are smaller than those for hydrogen and methyl thiolates. However, it is still not known exactly how the differences in binding energies affect the final structure of the nanoparticle. Additional studies in the Aikens group have elucidated the role of the reducing agent in the formation of gold-gold bonds in gold-phosphine and gold-thiolate systems, which appears to depend on the type of ligand used in the system, but more work needs to be done to fully understand how the differing thermodynamic stability of gold and silver systems affects the resulting nanoparticle structure.

One of the hypotheses related to objective 2 was that multiple nanoparticle shapes can exhibit sharp surface plasmon resonance (SPR) absorption behavior that can be extrapolated to predict the peak locations for large nanoparticles. As part of this grant, the Aikens group has shown that silver nanowires, octahedra, truncated octahedra, and icosahedra each display optical absorption peaks whose broadness depends on the number of electrons in the system; a “magic” number of electrons typically correlates with a sharp absorption spectrum. This is due to the delocalized nature of the electrons and the resulting orbitals. The peak positions can be extrapolated with increasing size of the system. A second hypothesis related to this objective was that the interband (d to sp) transitions for gold decahedral systems would not increase in intensity at the same rate as the intraband (sp to sp) transitions, and thus these could be

separated out. This work is still ongoing, but it currently appears that the size of the system at which this would be possible may be larger than what is now practical with DFT.

A third hypothesis related to objective 2 was that the wavelength of the SPR band can be controlled by modifying the coordinating ligand head group and tail group. Different head groups dramatically affect the optical spectra. The effects of the tail groups are less noticeable. As proposed, the optical absorption spectra of *p*-substituted $\text{Au}_{25}(\text{SPhX})_{18}^-$ ($\text{X} = \text{H}, \text{F}, \text{Cl}, \text{Br}, \text{CH}_3, \text{OCH}_3$) [note: the $\text{Au}_{38}(\text{SPhX})_{24}$ stoichiometry originally reported by the experimental group was later corrected to $\text{Au}_{25}(\text{SPhX})_{18}^-$] were calculated and the HOMO-LUMO gaps were computed. The HOMO-LUMO gaps and the low-energy peaks in the optical absorption spectrum (due to HOMO-LUMO and related transitions) were not significantly affected by the change in the tail group as expected. The HOMO and LUMO energies and thus the redox potentials do change with the tail group, however. In addition, the higher energy peaks that arise from transitions out of orbitals with contributions from ligand-based orbitals show a slight (0.1 eV) variation with tail group. It was also determined by the Aikens group that the preferred orientation of the phenylthiol groups is slightly different than the established orientation of phenylethylthiol and other R groups, which has an impact on the optical absorption spectrum.

In order to determine the effect of vibrational motion on the width of the optical absorption peaks (objective 2), the Aikens group has undertaken combined molecular dynamics and TDDFT calculations on two systems: Ag_{20} and $\text{Au}_{25}(\text{SH})_{18}^-$. Work on these projects is still underway, but it appears that temperature effects leading to vibrational broadening of the absorption spectra could be sufficient to explain the origin of the peak width.

Finally, to address the effect of distance and orientation on the optical absorption spectra of nanoparticle aggregates (objective 3), eight different orientations of dimers of Ag_{20} tetrahedra were considered. The Aikens group determined that tip-to-tip, tip-to-face, face-to-face, and edge-to-edge dimers result in somewhat different excitation spectra, although all orientations lead to a splitting of the monomer peak due to symmetry-lowering. The Aikens group also demonstrated that TDDFT calculations can lead to low energy peaks that are thought to be charge-transfer artifacts, whereas configuration interaction singles (CIS) calculations do not exhibit these peaks. In consequence, this work shows that future researchers should take care when using TDDFT to interpret the excitation spectra of nanoparticle dimers.

Kansas State University Personnel Supported by the Project (8)

Prof. Christine M. Aikens (Faculty)

Dr. Gyun-Tack Bae (Postdoctoral Research Assistant)

Ms. Emilie B. Guidez (Graduate Research Assistant)

Mr. Brian M. Barngrover (Graduate Research Assistant)

Ms. Makenzie R. Provorse (Undergraduate Research Assistant)

Mr. Jacob M. Hull (Undergraduate Research Assistant)

Ms. Kathryn E. Brewer (Undergraduate Research Assistant)

Ms. Allison Hadley (Undergraduate Research Assistant)

Collaborators (4)

Sergei Ivanov (Los Alamos National Laboratory)

Hannu Häkkinen (University of Jyväskylä, Finland)

Olga Lopez-Acevedo (University of Jyväskylä, Finland)

Tetsuya Tsukuda (University of Tokyo, Japan)

Published Articles (15)

TDDFT and CIS Studies of Optical Properties of Dimers of Silver Tetrahedra. G.-T. Bae, C. M. Aikens, *J. Phys. Chem. A*, **2012**, *116*, 8260-8269.

Theoretical Analysis of the Optical Excitation Spectra of Silver and Gold Nanowires. E. B. Guidez, C. M. Aikens, *Nanoscale*, **2012**, 4, 4190-4198.

Modeling Small Gold and Silver Nanoparticles with Electronic Structure Methods. C. M. Aikens, *Molecular Simulation*, **2012**, 38, 607-614. (Invited article for special issue on New Developments in Molecular Simulation.)

TDDFT Studies of Optical Properties of Ag Nanoparticles: Octahedra, Cuboctahedra and Icosahedra. G.-T. Bae, C. M. Aikens, *J. Phys. Chem. C*, **2012**, 116, 10356-10367.

Formyloxyl Radical-Gold Nanoparticle Binding: A Theoretical Study. J. M. Hull, M. R. Provorse, C. M. Aikens, *J. Phys. Chem. A*, **2012**, 116, 5445-5452.

Development of a Charge-Perturbed Particle-in-a-Sphere Model for Nanoparticle Electronic Structure. E. B. Guidez, C. M. Aikens, *Phys. Chem. Chem. Phys.*, **2012**, 14, 4287-4295.

Binding of Carboxylates to Gold Nanoparticles: A Theoretical Study of the Adsorption of Formate on Au₂₀. M. R. Provorse, C. M. Aikens, *Comput. Theor. Chem.*, **2012**, 987, 16-21. (Invited article for special issue on Modeling in Materials Chemistry.)

Incremental Binding Energies of Gold(I) and Silver(I) Thiolate Clusters. B. M. Barngrover, C. M. Aikens, *J. Phys. Chem. A*, **2011**, 115, 11818-11823.

Electron and Hydride Addition to Gold(I) Thiolate Oligomers: Implications for Gold-Thiolate Nanoparticle Growth Mechanisms: B. M. Barngrover, C. M. Aikens, *J. Phys. Chem. Lett.*, **2011**, 2, 990-994.

Density Functional Analysis of Geometries and Electronic Structures of Gold-Phosphine Clusters: The Case of Au₄(PR₃)₄²⁺ and Au₄(μ₂-I)₂(PR₃)₄. S. A. Ivanov, I. Arachchige, C. M. Aikens, *J. Phys. Chem. A*, **2011**, 115, 8017-8031.

Initial Growth Mechanisms of Gold-Phosphine Clusters. E. B. Guidez, A. Hadley, C. M. Aikens, *J. Phys. Chem. C*, **2011**, 115, 6305-6316.

Perspective: Electronic Structure of Ligand-Passivated Gold and Silver Nanoclusters. C. M. Aikens, *J. Phys. Chem. Lett.*, **2011**, 2, 99-104. (Invited Perspective)

Geometric and Electronic Structure of Au₂₅(SPhX)₁₈⁻ (X = H, F, Cl, Br, CH₃, and OCH₃). C. M. Aikens, *J. Phys. Chem. Lett.*, **2010**, 1, 2594-2599.

TDDFT Investigation of Surface-Enhanced Raman Scattering of HCN and CN⁻ on Ag₂₀. K. E. Brewer, C. M. Aikens, *J. Phys. Chem. A*, **2010**, 114, 8858-8863. (Invited article for Klaus Ruedenberg Festschrift.)

Chirality and Electronic Structure of the Thiolate-Protected Au₃₈ Nanocluster. O. Lopez-Acevedo, H. Tsunoyama, T. Tsukuda, H. Häkkinen, C. M. Aikens, *J. Am. Chem. Soc.*, **2010**, 132, 8210-8218. (Featured in JACS Select)

Interactions/Transitions

Research supported by this grant was presented at the following meetings, conferences, and seminars:

TDDFT Optical Absorption Characteristics of Silver and Gold Nanoparticles: Effect of Size, Shape, and Composition. C. M. Aikens, poster presentation, Air Force Office of Scientific Research Molecular Dynamics Contractors' Meeting, Arlington, VA: May 22-24, 2012.

Complex Optical Absorption Spectra of Gold and Silver Nanoparticles. C. M. Aikens, poster presentation, 18th Annual German-American Frontiers of Science Symposium, Potsdam, Germany: May 10-13, 2012.

Unraveling Nanoparticle Properties using Density Functional Theory. C. M. Aikens, Materials Research Science and Engineering Center seminar, Northwestern University, Evanston, IL: February 24, 2012.

Unraveling Nanoparticle Properties using Theoretical Methods. C. M. Aikens, Physical Chemistry and Integrative Nanoscience Institute seminar, Florida State University, Tallahassee, FL: November 14, 2011.

Unraveling Nanoparticle Properties using Density Functional Theory. C. M. Aikens, Student-Invited Chemistry Department seminar, University of Minnesota, Minneapolis, MN: November 3, 2011.

Achieving Theory-Experiment Agreement for the Optical Absorption, Electron Paramagnetic Resonance, and Chiroptical Properties of Gold and Silver Nanoparticles. Chemistry Department seminar, University of North Texas, Denton, TX: October 28, 2011.

DFT Optical Properties and Growth Mechanisms of Gold Nanoparticles. C. M. Aikens, B. M. Barngrover, E. B. Guidez, invited talk, Joint 46th Midwest and 36th Great Lakes Regional Meeting, St. Louis, MO: October 17-22, 2011.

Unraveling Nanoparticle Properties using Density Functional Theory. C. M. Aikens, invited talk, 16th International Workshop on Quantum Systems in Chemistry and Physics, Kanazawa, Japan: September 11-17, 2011.

Growth Mechanisms of Gold Nanoparticles. C. M. Aikens, B. M. Barngrover, E. B. Guidez, invited poster presentation – HP Junior Faculty Award, 242nd National American Chemical Society Meeting, Denver, CO: August 28-September 1, 2011.

Large-Scale TDDFT Calculations of Plasmon Resonances. C. M. Aikens, contributed talk, 242nd National American Chemical Society Meeting, Denver, CO: August 28-September 1, 2011.

Theoretical Studies of Noble Metal Nanoparticle Physical Properties. C. M. Aikens, invited talk – Marie Curie symposium, 242nd National American Chemical Society Meeting, Denver, CO: August 28-September 1, 2011.

Optical and Photophysical Properties of Gold Nanoparticles. C. M. Aikens, invited talk, Gordon Research Conference on Time-Dependent Density Functional Theory, University of New England, Biddeford, ME: August 14-19, 2011.

Growth Mechanisms of Gold Nanoparticles. C. M. Aikens, B. M. Barngrover, E. B. Guidez, poster presentation, American Conference on Theoretical Chemistry, Telluride, CO: July 17-22, 2011.

DFT Optical Properties and Growth Mechanisms of Gold-Phosphine and Gold-Thiolate Nanoparticles. C. M. Aikens, B. M. Barngrover, E. B. Guidez, invited talk, Midwest Theoretical Chemistry Conference, South Bend, IN: June 9-11, 2011.

DFT Optical Properties and Growth Mechanisms of Gold-Phosphine and Gold-Thiolate Nanoparticles. C. M. Aikens, B. M. Barngrover, E. B. Guidez, invited talk, International Symposium on Monolayer Protected Clusters (ISMPC11), Jyväskylä, Finland: June 6-9, 2011.

Structure and Optical Properties of Gold and Silver Monolayer-Protected Nanoparticles. C. M. Aikens, invited speaker, Air Force Office of Scientific Research Molecular Dynamics Contractors' Meeting, Pasadena, CA: May 15-17, 2011.

Optical Absorption, EPR, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, Physical Chemistry seminar, University of Washington, Seattle, WA: April 26, 2011.

Optical Absorption, EPR, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, Chemistry Department seminar, Washington University, St. Louis, MO: March 31, 2011.

Application of Chiroptical Properties with Nanoparticle Structural Prediction. C. M. Aikens, M. Provorse, O. Lopez-Acevedo, H. Häkkinen, poster presentation, Pacificchem 2010, Honolulu, HI: December 15-20, 2010.

Structure and Optical Properties of Small (1-1.5 nm) Gold and Silver Nanoparticles. C. M. Aikens, Condensed Matter Physics seminar, Kansas State University, Manhattan, KS: December 3, 2010.

Optical Absorption, EPR, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, Physical, Theoretical, and Computational Chemistry seminar, Texas Tech University, Lubbock, TX: November 19, 2010.

Optical Absorption, EPR, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, Inorganic and Physical Chemistry seminar, Colorado State University, Fort Collins, CO: November 9, 2010.

Optical Absorption, EPR, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, Chemistry Department seminar, University of Nebraska at Kearney, Kearney, NE: October 8, 2010.

Optical Absorption, EPR, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, Chemistry Department seminar, University of Toledo, Toledo, OH: September 22, 2010.

Optical Absorption, EPR, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, Laboratory for Nanophotonics seminar, Rice University, Houston, TX: September 16, 2010.

Ligand Effects on the Geometric and Electronic Structure of the $\text{Au}_{25}(\text{SR})_{18}^-$ Nanoparticle. C. M. Aikens, poster presentation, 240th National American Chemical Society Meeting, Boston, MA: August 22-26, 2010.

Complex Structure and Optical Spectra of Small Gold Nanoparticles. C. M. Aikens, poster presentation, DARPA MEMS PI Meeting, San Francisco, CA: July 21-23, 2010.

Achieving Theory-Experiment Agreement for the Optical Absorption, Electron Paramagnetic Resonance, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, invited speaker, Gordon Research Conference on Noble Metal Nanoparticles, Mt. Holyoke College, South Hadley, MA: June 20-25, 2010.

Optical Absorption, Electron Paramagnetic Resonance, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, invited speaker, Molecular Quantum Mechanics 2010, Berkeley, CA: May 27-29, 2010.

Chiral Nanorod Structure of "Magic" $\text{Au}_{38}(\text{SR})_{24}$ Nanocluster. C. M. Aikens, poster presentation, Air Force Office of Scientific Research Molecular Dynamics Contractors' Meeting, Chantilly, VA: May 24-26, 2010.

Optical Absorption, EPR, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, Chemistry Department seminar, University of Missouri, Columbia, MO: March 12, 2010.

Achieving Theory-Experiment Agreement for the Optical Absorption, EPR, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, Nanoscience and Physical/Analytical Chemistry seminar, University of Utah, Salt Lake City, UT: February 22, 2010.

Theoretical Studies of Optical Absorption, EPR, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, AMO Physics Division seminar, Kansas State University, Manhattan, KS: October 28, 2009.

Optical Absorption, EPR, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, Chemistry Department seminar, Carnegie Mellon University, Pittsburgh, PA: October 8, 2009.

Optical Absorption, EPR, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, Chemistry Department seminar, Montana State University, Bozeman, MT: October 2, 2009.

Optical Absorption, EPR, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, Physical Chemistry seminar, University of Nebraska, Lincoln, NE: September 14, 2009.

Theoretical Examination of the Optical Absorption, Electron Paramagnetic Resonance, and Chiroptical Properties of Gold and Silver Nanoparticles. C. M. Aikens, invited speaker, 238th National American Chemical Society Meeting, Washington, DC: August 16-20, 2009.

No discoveries, inventions, nor patent disclosures have resulted from this work

Honors/Awards

Honors and awards for Prof. Aikens during the period of the AFOSR grant:
Kavli Fellow (2012)

Camille Dreyfus Teacher-Scholar (2011)

American Chemical Society COMP Division: Hewlett-Packard Outstanding Junior Faculty Award (2011)

Selected as one of the Top 150 Scientists in Kansas (Ad Astra Kansas Initiative for the Sesquicentennial, 2011)

Alfred P. Sloan Research Fellowship (2011-2013)

National Science Foundation CAREER awardee (2010)

Sigma Xi Junior Faculty Award (Kansas State University, 2010)