**Abstract:** The proposed objective of this research is to use numerical simulation and quantum-mechanical modeling to guide the development of new paradigms for making artificial materials with an ever-larger nonlinear-optical (NLO) response that can apply to a broad range of technologies. The approach is to study how to use the effects of geometry on network of quantum loops to achieve optimized nonlinear properties.

**Introduction**

All light-matter interactions are governed by the nonlinear-optical susceptibility.

**Why the Gap?**

- Vibronic dilution effects not large enough to explain gap
- Oscillator strength is big enough in the best molecules
- Gap not due to any fundamental reason

**Scale Invariance[1]**

\[ H = \frac{1}{2} \sum_{k \in \text{bands}} \left[ v_k - \mu \left( n_1 \cdots n_p \right) \right] + V \left( n_1 \cdots n_p \cdot E_1 \cdots E_p \right) \]

1. Rescaling of energies and potentials leads to a wave function of the same shape.
2. Call this transformation Simple Scaling.
3. Intrinsic hyperpolarizability is invariant under simple scaling

**Universal Properties[1]**

Universal properties observed when the hyperpolarizability is near the limit of optimized I-D potentials, while very different, share similar universal properties.

**Motivation**

Is there a limit to the nonlinear susceptibility? YES!

\[ \beta_{\text{NLO}} = 3.0 \times 10^{-30} \text{ cm}^2 / \text{V}^2 \]

Similar results for higher-order nonlinearities

**Second-Order Susceptibility**

- Macroscopic
  - Electronic
  - Molecular
- Second Hyperpolarizability
  - Macroscopic
  - Electronic
  - Molecular

**The Model[3]**

- Schrödinger Equation is solved with following conditions and transition moments and energies are calculated.
- Electron moves freely along the quantum wire segments.
- To minimize confinement effects, a highly weighted Delta function potential confines electron in transverse direction!!

**Quantum-confined Systems (QCS)**

- Energy spectrum of most molecules impose a bond on the maximum attainable nonlinear response
- Artificial systems (Quantum wells, wires and dots) has demonstrated great potential for future optical devices
- Nonlinear response of QCSs depends on their degree of confinement

**Nanostructures and Quantum Graphs**

Is the NLO properties of quantum graphs sensitive to their geometry? YES!

- Quantum graphs can be built from nanowires
- Nanoparticles can be combined to optimize the system
- Closed triangle gives 3β\text{NLO} = 0.2

**Conclusion**

- The NLO response of quantum graph is sensitive to the geometry
- We have found a triangle quantum loop with 1.5 times larger NLO response compared to best chromophores ever measured!
- A combination of confinement and geometry effect can lead to extra-large response

**References**