Vibrational modulation of electronic transitions in Copper Germanate: a theoretical model

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Introduction

- Motivation: understand the dynamics of complex materials
- Pump-probe scheme:
  1. First intense light pulse (pump): excites some degrees of freedom in the material
  2. The sample evolves for a certain time $t$
  3. Second light pulse (probe): interacts with the sample encoding useful information about the dynamics up to time $t$
  4. The transmitted probe light is measured
  5. Repeat 1 – 4 with different delay time $t$

- A theoretical model is then used to infer the dynamics of the material
The experiment

- Pump-Probe on Copper Germanate (CuGeO₃)
- Infrared pump (to excite vibrational degrees of freedom in the sample)
- Visible probe (to probe d-d electronic transitions)
- Transmissivity measure (shift spectral weight)
The model

- **Pump treated implicitly**: it sets the "initial state" of the phonon-electron system

\[ \rho = D \rho_\beta D^\dagger \otimes |0\rangle \langle 0|, \quad \rho_\beta = \frac{e^{-\beta \omega b\dagger b}}{\text{Tr}(e^{-\beta \omega b\dagger b})}, \quad D = e^{B_t(b\dagger - b)} \]

- **Probe-target interaction** modeled explicitly
  - Unperturbed Hamiltonian:
    \[ H_{ph-e} \equiv H = \omega b\dagger b + \epsilon d\dagger d + M d\dagger d (b + b\dagger), \quad (1) \]
  - Interaction Hamiltonian:
    \[ H_{int} = \mu_0 P \sum_k (a\dagger_k + a_k), \quad P = (b + b\dagger)(d + d\dagger), \quad (2) \]
Computed observable: light intensity (mean photon number) at a certain frequency \( I_j = \text{Tr}\left( \varrho \ U^\dagger(\tau) a_j^\dagger a_j U(\tau) \right) \)

Approximation: second order perturbation theory

First result: the temperature dependence of the total absorption

\[ \int \text{d}\nu (I(\nu) - I^0(\nu)) \propto \left( \text{coth} \left( \frac{\beta \omega}{2} \right) + 4B_t^2 \right). \]

Second result: shift of the d-d absorption peak depending on the pump-induced displacement \( B_t \)
Conclusion

- We provided a simple theoretical model that:
  - is consistent with known experimental evidence
  - can capture qualitative features of our pump-probe experiment and allows to interpret the result as a vibrational control of the electronic transition

- Outlook: An explicit treatment of the pump-sample interaction could allow a more quantitative estimate of the magnitude and time-evolution of the displacement

Thank you for your attention!