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

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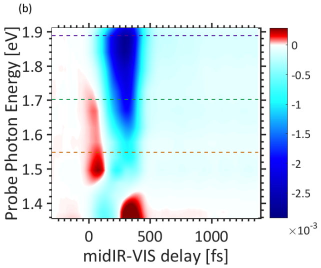
Outline

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 trasmitted 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 ($CuGeO_3$)
- 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

$$\varrho = D\varrho_\beta D^\dagger \otimes |0\rangle\langle 0|, \quad \varrho_\beta = \frac{\mathrm{e}^{-\beta\omega b^\dagger b}}{\mathrm{Tr}(\mathrm{e}^{-\beta\omega b^\dagger b})}, \quad D = \mathrm{e}^{B_t(b^\dagger - b)}$$

- Probe-target interaction modeled explicitly
  - Unperturbed Hamiltonian:

$$H_{ph-el} \equiv H = \omega\, b^\dagger b + \epsilon\, d^\dagger d + M d^\dagger d\,(b + b^\dagger), \tag{1}$$

  - Interaction Hamiltonian:

$$H_{int} = \mu_0 P \sum_k (a_k^\dagger + a_k), \quad P = (b + b^\dagger)(d + d^\dagger), \tag{2}$$

Results

- Computed observable: light intensity (mean photon number) at a certain frequency $I_j = \mathrm{Tr}\Big(\varrho\, U^\dagger(\tau) a_j^\dagger a_j U(\tau)\Big)$
- Approximation: second order perturbation theory
- First result: the temperature dependence of the total absorption

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

- 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

Alexandre Marciniak, Stefano Marcantoni, Francesca Giusti, Filippo Glerean, Giorgia Sparapassi, Tobia Nova, Andrea Cartella, Simone Latini, Francesco Valiera, Angel Rubio, Jeroen van den Brink, Fabio Benatti, and Daniele Fausti, **arXiv:2003.13447**

Thank you for your attention!