# Time-resolved optical conductivity and Higgs oscillations in two-band dirty superconductors

Rafael Haenel,[1,2] Paul Froese,[1,2] Dirk Manske,[1] and Lukas Schwarz[1]

[1]*Max Planck Institute for Solid State Research, 70569 Stuttgart, Germany*

[2]*Quantum Matter Institute, University of British Columbia, Vancouver V6T 1Z4, Canada*

(Dated: October 27, 2020)

...

## I. INTRODUCTION

- Ultrafast spectroscopy
- Collective modes in superconductors: Higgs, Goldstone (shifted to plasma energy due to Anderson-Higgs)
- In two-band superconductors: Additional out-of-phase Leggett mode, can couple to Higgs in nonequilibrium
- Difficulties excitation Higgs mode, in clean-limit only weak coupling
- In dirty superconductors: Coupling is enhanced. Activation of $\mathbf{p}\cdot\mathbf{A}$ term by momentum-converation braking impurity scattering.
- Discussion of Matthis Bardeen theory.
- This work: 1) Higgs oscillations in two-band sc. with bands in different limits, 2) Nonequilibrium optical conductivity, 3) Leggett mode in dirty-limit, 4) Prediction for $MgB_2$

This article is organized as follows. In Sec. II we briefly review key aspects of the model as introduced by Murotani and Shimano []. We then discuss the case of a single-band superconductor in a pump-probe scenario and additionally with an assymetric supercurrent-inducing probe pulse in Sec. III. We then extend these results to the two-band case in Sec. IV where we give explicit experimental predicitons for pump-probe characterization of collective modes in $MgB_2$.

## II. MODEL

- In this section show only the final formula, all derivation into the appendix A as the equations are similar to the Murotani paper.
- Show Hamiltonian, gap equation, Mattis-Bardeen replacement, general approach for calculating time evolution...
- I suggest putting the (final) equations for current, optical conductivity, $\delta\Delta(t)$ into the respective section, but in principle we could also put all equations in this section and show only the results in the following sections
- Used parameters (take general parameters for section III and IV where no Leggett mode occurs, Leggett mode is discussed later separately and $MgB_2$ will be also discussed later. Show the used parameters in these section)
- Implementation details? Are there any subtle points?

$$H_{\mathrm{BCS}} = \sum_{i\mathbf{k}\sigma} \varepsilon_{i\mathbf{k}} c^\dagger_{i\mathbf{k}\sigma} c_{i\mathbf{k}\sigma} + \sum_{i\mathbf{k}} \left( \Delta_i c^\dagger_{i-\mathbf{k}\uparrow} c^\dagger_{i\mathbf{k}\downarrow} \right), \quad (1)$$

where $\varepsilon_{i\mathbf{k}} = s_i\left(\mathbf{k}^2/2m_i - \varepsilon_{F_i}\right)$ and the superconducting order parameter is self-consistently determined by $\Delta_i = \sum_{j\mathbf{k}} U_{ij} \langle c_{j-\mathbf{k}\downarrow} c_{j\mathbf{k}\uparrow}\rangle$.

$$H_{\mathrm{p\text{-}p}} = -\sum_{i\mathbf{k}\mathbf{k}'\sigma} \mathbf{J}_{i\mathbf{k}\mathbf{k}'}\cdot\mathbf{A}\, c^\dagger_{i\mathbf{k}\sigma} c_{i\mathbf{k}'\sigma} + \sum_{i\mathbf{k}\sigma} \frac{s_i e^2}{2m_i} \mathbf{A}^2\, c^\dagger_{i\mathbf{k}\sigma} c_{i\mathbf{k}\sigma}$$

$$\begin{aligned}\langle |\mathbf{e}\cdot\mathbf{J}_{i\mathbf{k}\mathbf{k}'}|^2 \rangle_{\mathrm{Av}} &= \int \frac{d\Omega_{\mathbf{k}}}{4\pi}\frac{d\Omega'_{\mathbf{k}}}{4\pi} |\mathbf{e}\cdot\mathbf{J}_{i\mathbf{k}\mathbf{k}'}|^2 \\ &\approx \frac{(ev_{F_i})^2}{3\pi N_i(0)} \frac{\gamma_i}{(\varepsilon-\varepsilon')^2+\gamma_i^2}\end{aligned}$$

Discussion of $A$, $A^2$.

The full Hamiltonian is given by $H = H_{BCS} + H_{p-p}$.

Shortcoming of the model. Momentum-angle averaging removes information about polarization, momentum of photon. The gap oscillates with its equilibrium value and not $\Delta_\infty$. Also, the results are an expansion in $A$ up to third order. Specifically, the gap in only included up to second order (the only nonzero term) which makes it hard to say when this approximation becomes invalid. The dependence of the gap on fluence is linear up to this order.

## III. SINGLE-BAND SUPERCONDUCTIVITY

Motivated by the experiment of Matsunaga et al. [] we choose parameters $\Delta_{\mathrm{eq}} = 1.3\,\mathrm{meV}$, $\varepsilon_F = 1\,\mathrm{eV}$, $m = 0.78 m_e$, $s = 1$ that reflect measurements and ab-initio calculations on NbN [DFT-reference]. The Debye energy for NbN is order order of $\omega_D = 5\,\mathrm{meV}$, but for better numerical resolvability we choose $\omega_D = 2\,\mathrm{meV}$.

A characteristic property of a pump pulse is its pulse length $\tau$ in units of the natural timescale of the superconductor, $\hbar/\Delta$. For $\tau \ll \hbar/\Delta$ the superconductor is *quenched*, while it is adiabatically driven in the opposite limit of $\tau \gg \hbar/\Delta$. Here, we investigate a quench scenario with $\hbar\tau/\Delta = x$ by fitting the pulse form $A(t) = A_0 \exp\left(-(t-t')^2/2\tau^2\right) \cos \Omega t$ to the reported data of [Matsunaga]. The resulting electrical field waveform is shown in panel A of Fig. 4.

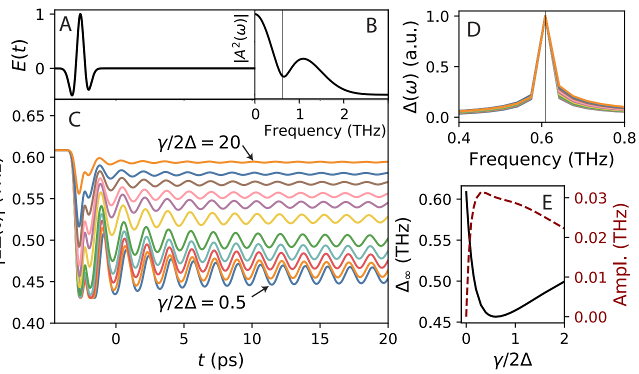


Figure 1. (A) Pulse field $E(t)$ realizing a qunech. (B) Spectral composition $|A(\omega)|$. The gray shaded area illustrates the quasi-particle continuum. (C) Spectral compositon $A^2(\omega)$ of the second order component $A^2(t)$ responsible for excitation of collective modes. The peak around zero frequency corresponds to a DFG process while the peak at finite 1.2 THz is a SFC process. (C) Evolution of the magnitude of the order parameter $|2\Delta(t)|$ for impuritiy strength varying from $\gamma/2\Delta = 0.5$ to 20 and Fourier spectrum of the gap oscillations (D). (E) Relaxation value $\Delta_\infty$ and amplitude of oscillation show a very similar dependence as a function of disorder strength which has maximum effect at around $\gamma \approx \Delta$.

$\Delta_\infty$ is given by the overlap of the pulse with the density of states of the quasiparticel continuum,

$$\Delta_\infty \propto \int d\omega N(\omega) A(\omega) \approx N(\varepsilon_F) \int_{\varepsilon_F}^{\infty} d\omega A(\omega)$$

.

## IV. MULTI-BAND SUPERCONDUCTIVITY

## V. HIGGS OSCILLATIONS

- Equation for Current
- Equation for optical conductivity
- Present and discuss equilibrium optical conductivity in Fig. 1 for the four different limit cases, used in the rest of the paper

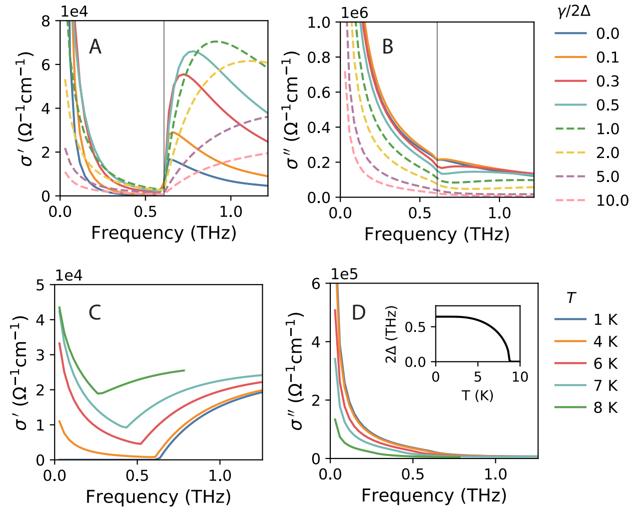


Figure 2. Real part $\sigma'$ (A,C) and imaginary part $\sigma''$ (B,D) of optical conductivities to first order in $A$ for various impurity scattering rates at $T = 4\,\mathrm{K}$ (A,B) and various temperatures at $\gamma/2\Delta = 10$. $\sigma'$ show a characteristic conductivity gap below $T_C$ and both $\sigma'$, $\sigma''$ diverge in the static limit.

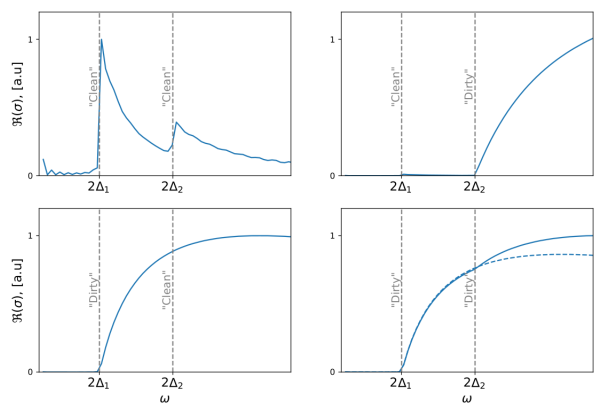


Figure 5. Equilibrium optical conductivity of a two-band superconductor with gaps in different impurity scattering limits (clean-clean case rescaled as response is much smaller?)

- Equation for $\delta\Delta(t)$
- Show and discuss Higgs oscillations in Fig. 2 of the four cases with a suitable pump pulse (refer to appendix B for details about the pump pulse)

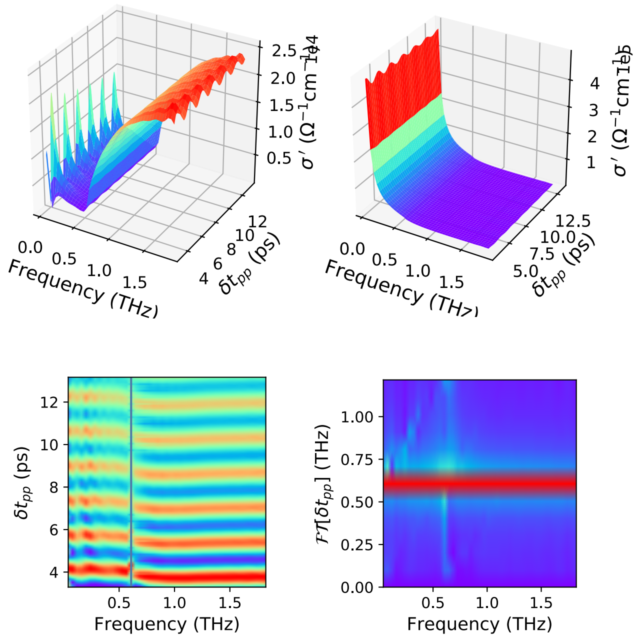


Figure 3. (A,B) Conductivity spectra for sweeped pump-probe delay $\delta t_{\text{p-p}}$ to third order in $A$. (C) False-color plot of $\sigma'$ which was average-subtracted and normalized to show the oscillations. A phase shift occurs across the resonance at $2\Delta_{\text{eq}}$ of the quench pulse frequency. (D) Fourier spectrum of panel (C) showing that frequency of conductivity oscillation is peaked at $2\Delta_{\text{eq}}$.

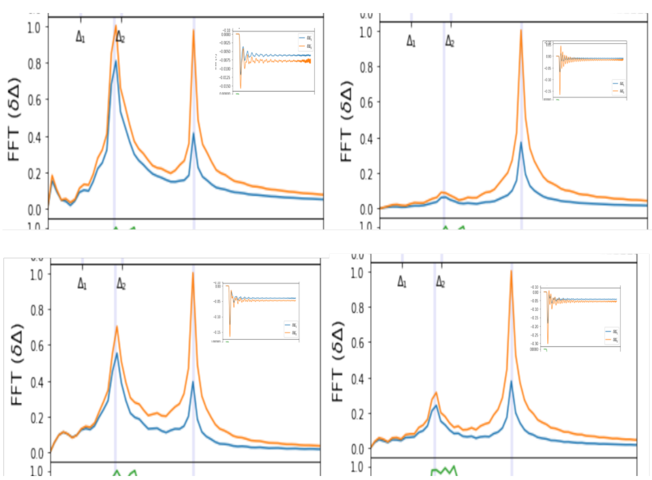


Figure 6. Higgs oscillations excited by a pump pulse with enough bandwidth to cover both gaps for the different impurity scattering limits shown in Fig. 5

## VI. NONEQUILIBRIUM OPTICAL CONDUCTIVITY

- Equation for nonequilibrium optical conductivity with two pulses. Maybe here more details (instead

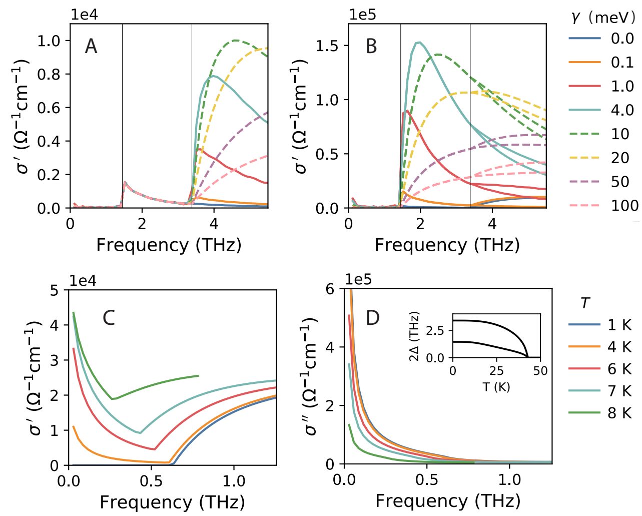


Figure 4. Real part $\sigma'$ (A,C) and imaginary part $\sigma''$ (B,D) of optical conductivities to first order in $A$ for various impurity scattering rates at $T = 4\,\text{K}$ (A,B) and various temperatures at $\gamma/2\Delta = 10$. $\sigma'$ show a characteristic conductivity gap below $T_C$ and both $\sigma'$, $\sigma''$ diverge in the static limit.

$\Delta_\infty$ is given by the overlap of the pulse with the density of states of the quasiparticel continuum,

$$\Delta_\infty \propto \int d\omega N(\omega) A(\omega) \approx N(\varepsilon_F) \int_{\varepsilon_F}^{\infty} d\omega A(\omega)$$

.

of appendix), as this is not covered by the Murotani paper

- Show and discuss nonequlibrium conductivity in Fig. 3.

- More figures of nonequilibrium conductivity? Suggestions: Imaginary part, maybe appendix. Nice 3d plot. Oscillations along cuts.

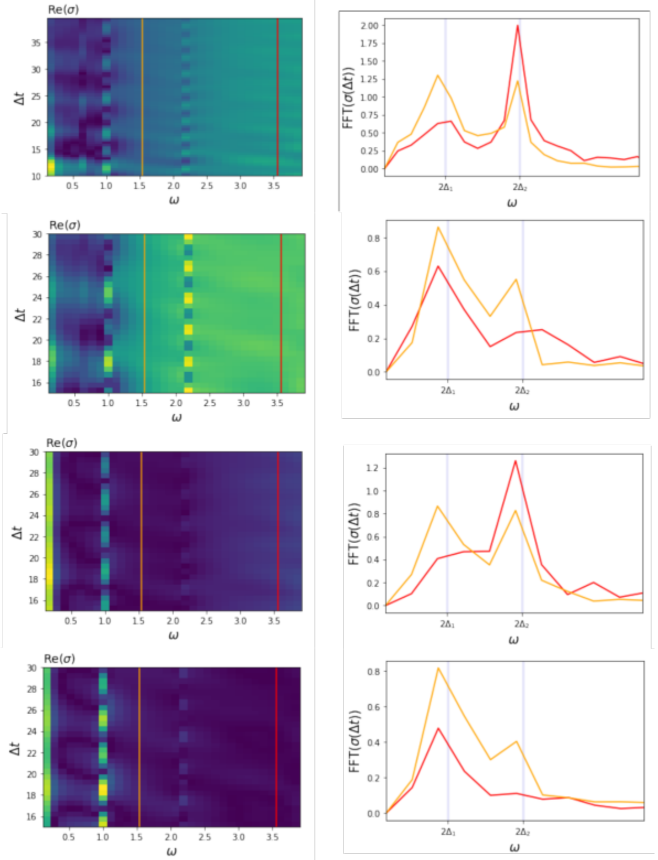

Figure 7. Nonequilibrium optical conductivity of a pump-probe experiment for the different impurity scattering limits shown in Fig. 5

## VII. LEGGETT MODE

- Definition, equation of Leggett mode
- Parameters for Fig.4 where Leggett mode occurs
- Discuss Fig. 4
- Maybe rethink what exactly to show in Fig. 4
- New plot of time-resolved conductivity showing Leggett mode?

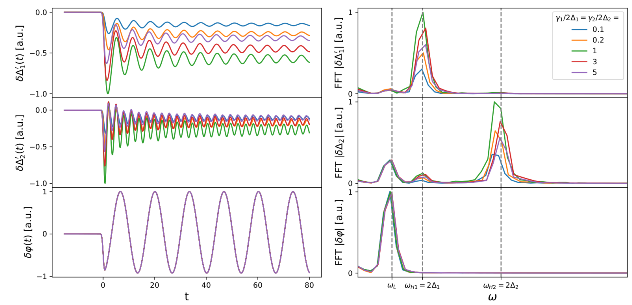

Figure 8. Induced amplitude (Higgs) and out-of-phase (Leggett) oscillations of a two-band superconductor for different impurity scattering rates

- Discuss Fig. 5 for varying coupling strength and compare with clean limit result

Leggett mode has been potentially measured in between the two gaps. Discuss this and also refer to Schnyder Nature Comm. Figure out the parameter regime that inverted the gaps. Reproduce (or see what was wrong) with Murotani who also choose parameters such that the Leggett mode is inside the gap.

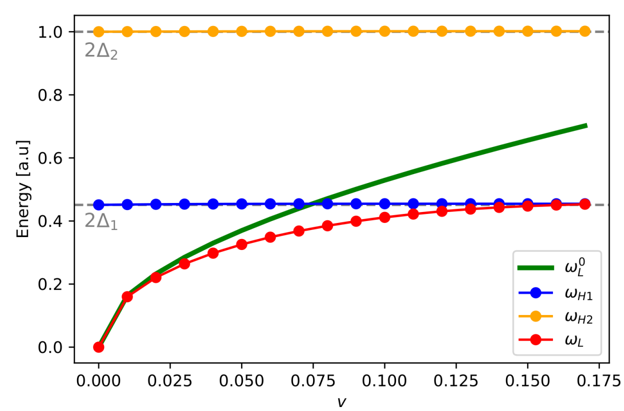


Figure 9. Frequency of Leggett mode as function of interband coupling strength in the dirty limit

## VIII. MGB$_2$

- Parameters which match MgB$_s$
- Show prediction in Fig. 6 of equilibrium conductivity, Higgs oscillations, nonequilibrium conductivity

# TODO

Figure 10. Result with parameters for $MgB_2$ a) Equilibrium optical conductivity, b) Higgs oscillations, c) pump-probe conductivity and spectra

## IX. CONCLUSION

...


## ACKNOWLEDGMENTS

...


## Appendix A: Derivation of nonequilibrium optical conductivity

- Put here all equations and derivations of the main results

## Appendix B: Influence of pump pulse frequency

- Discuss influence of pump pulse frequency and band-with to excite only one or both Higgs mode

- Show result in Fig. 7

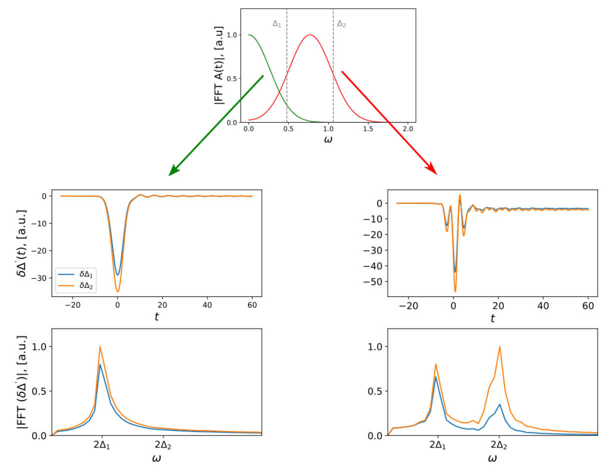


Figure 11. Influence of pump pulse frequency