Transient Dynamics and Gap Oscillations of Non-equilibrium Superconductors

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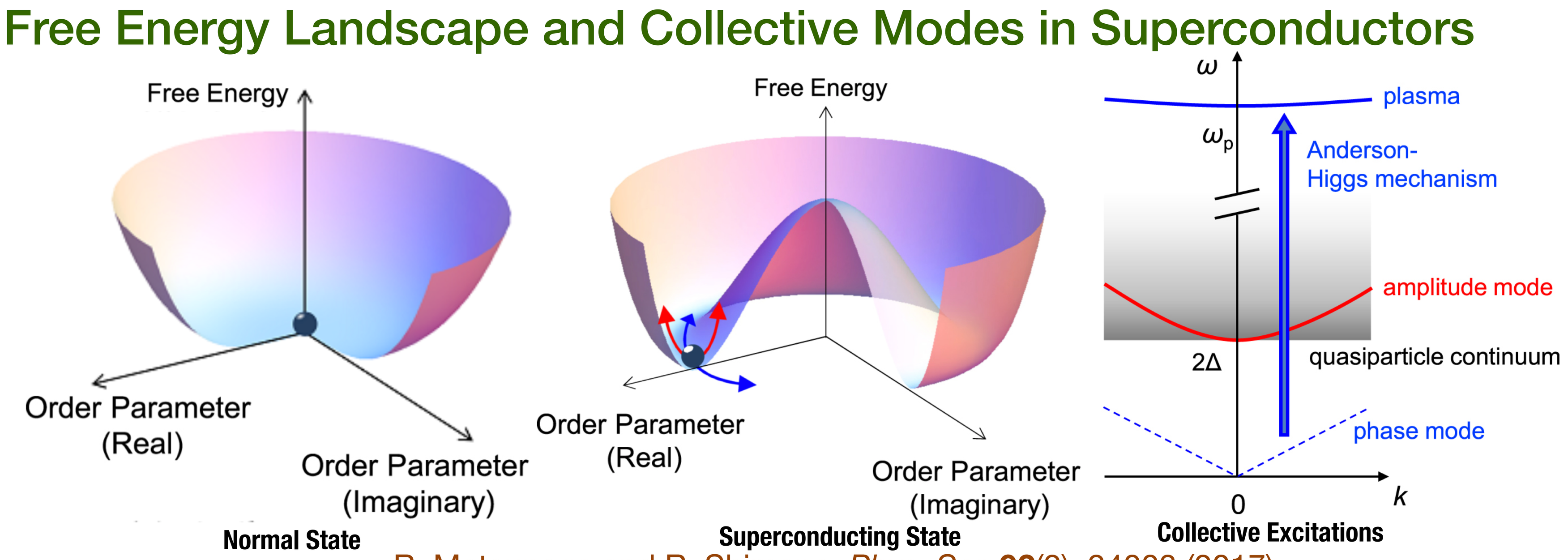
U.S. DEPARTMENT OF ENERGY

Introduction and Motivation

Free Energy Landscape and Collective Modes in Superconductors



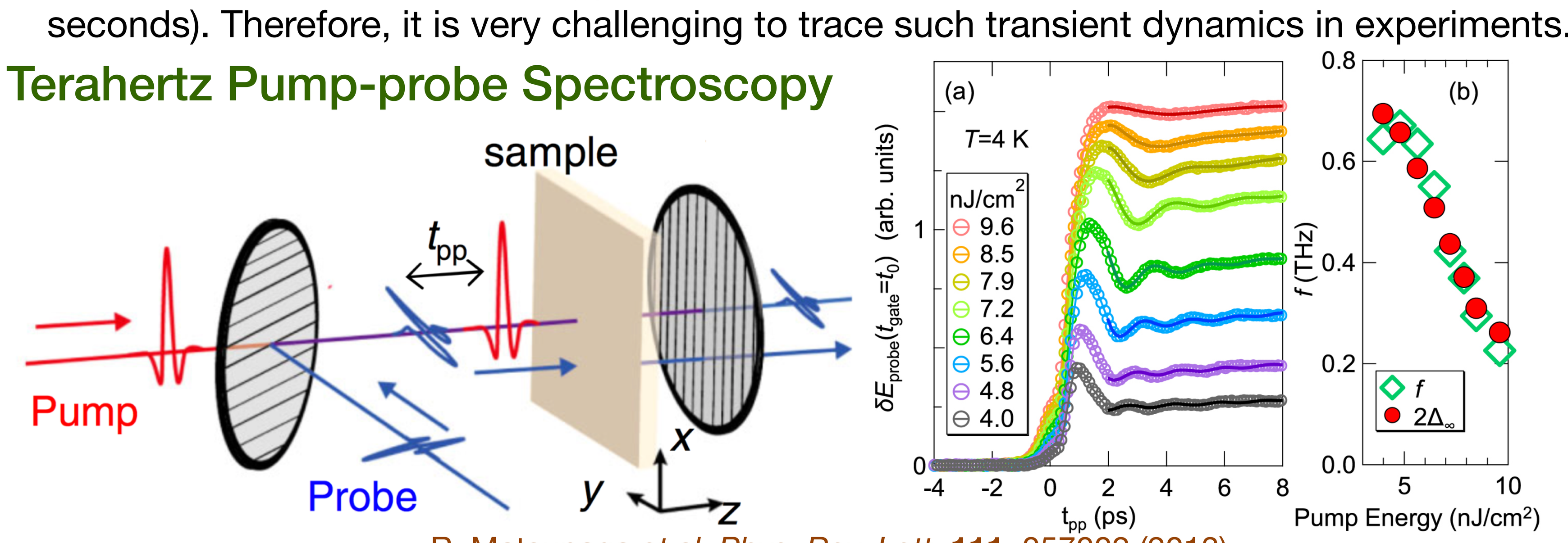
R. Matsunaga and R. Shimano, *Phys. Scr.* **92**(2), 24003 (2017).

- Due to the laws of thermodynamics, the free energy reaches its minimal in equilibrium.
- The free energy landscape changes at the superconducting phase transition.
- Various collective excitations exist in superconductors, including the Higgs amplitude mode.

Higgs Amplitude Mode in Superconductors

- Amplitude oscillation of the superconducting order parameter (shown as the red arrow in the figure of free energy landscape).
- The Higgs mode in superconductors has an analogy to the Higgs boson discovered at the Large Hadron Collider in 2012. But it can be observed in table-top experiments.
- The typical time scale of the Higgs amplitude mode is around 10 picoseconds (10^{-11} seconds). Therefore, it is very challenging to trace such transient dynamics in experiments.

Terahertz Pump-probe Spectroscopy



R. Matsunaga *et al*, *Phys. Rev. Lett.* **111**, 057002 (2013).

- Both the pump and probe beam are ultra-fast (around 2 ps) and in the terahertz regime.
- The change of the transmitted electric field traces the change of the superconducting order parameter, because the transmittance is inversely proportional to the order parameter.
- After the pump, the order parameter oscillates with frequency 2Δ, which coincides with previous prediction for Higgs mode in superconductors.

Motivation to Our Theoretical Study

- The ultra-fast pump-probe technique provides a new perspective to the study of superconductivity, and unveils the non-equilibrium properties on a picosecond time scale.
- The computational power of Minnesota Supercomputing Institute (MSI) allows us to simulate such complicated transient dynamics and explain experimental results.
- Furthermore, the question we want to address is: ***What is transient dynamics of unconventional superconductors where the order parameters have complex structures?***

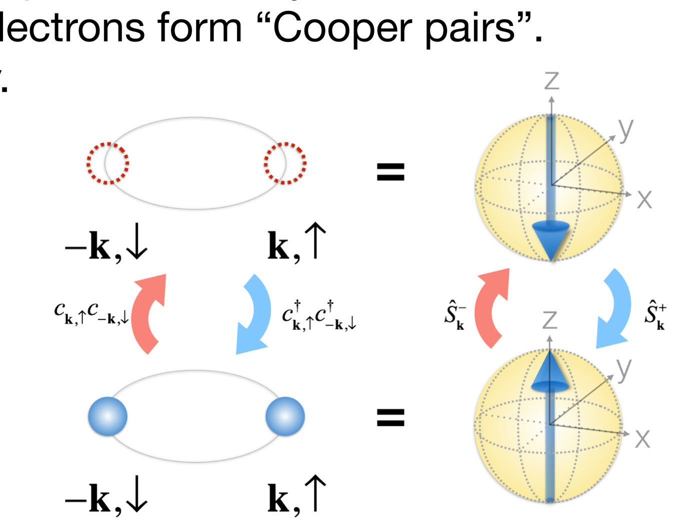
Model and Strategy

The Bardeen-Cooper-Schrieffer (BCS) Theory of Superconductivity

- In 1957, BCS proposed the first successful microscopic theory of superconductivity[1].
- Due to the effective attraction mediated by the ionic background, electrons form "Cooper pairs".
- Phase coherence between Cooper pairs leads to superconductivity.

➢ BCS Hamiltonian: $H_{\mathrm{BCS}}=\sum_{\mathbf{k},\sigma}\varepsilon_{\mathbf{k}}c^{\dagger}_{\mathbf{k},\sigma}c_{\mathbf{k},\sigma}+U\sum_{\mathbf{k},\mathbf{k}'}c^{\dagger}_{\mathbf{k},\uparrow}c^{\dagger}_{-\mathbf{k},\downarrow}c_{-\mathbf{k},\downarrow}c_{\mathbf{k},\uparrow}$

➢ Superconducting gap (order parameter): $\Delta=-U\sum_{\mathbf{k}}\langle c_{-\mathbf{k},\downarrow}c_{\mathbf{k},\uparrow}\rangle$



Pseudo-spin Formalism of the BCS Model

Mapping between the BCS model and the Spin model[2]:

❖ Pair empty state as spin-down and pair occupied state as spin-up.
❖ Pair creation as spin flip from down to up.
❖ Pair annihilation as spin flip from up to down.

In the mean-field limit, the BCS Hamiltonian is described by: $H_{\mathrm{BCS}}=-\sum_{\mathbf{k}}\mathbf{B}_{\mathbf{k}}\cdot\hat{\mathbf{S}}_{\mathbf{k}}$

➢ Pseudo-spins: $\hat{S}^{z}_{\mathbf{k}}=\frac{1}{2}\left(c^{\dagger}_{\mathbf{k},\uparrow}c_{\mathbf{k},\uparrow}+c^{\dagger}_{-\mathbf{k},\downarrow}c_{-\mathbf{k},\downarrow}-1\right)$ $\hat{S}^{+}_{\mathbf{k}}=\hat{S}^{x}_{\mathbf{k}}+i\hat{S}^{y}_{\mathbf{k}}=c^{\dagger}_{\mathbf{k},\uparrow}c^{\dagger}_{-\mathbf{k},\downarrow}$ $\hat{S}^{-}_{\mathbf{k}}=\hat{S}^{x}_{\mathbf{k}}-i\hat{S}^{y}_{\mathbf{k}}=c_{-\mathbf{k},\downarrow}c_{\mathbf{k},\uparrow}$

➢ Effective magnetic field: $\mathbf{B}_{\mathbf{k}}=\left(2\mathrm{Re}(\Delta),-2\mathrm{Im}(\Delta),-2\varepsilon_{\mathbf{k}}\right)$ where the superconducting gap is $\Delta=-U\sum_{\mathbf{k}}S^{-}_{\mathbf{k}}$

Equations of Motion for the Non-equilibrium Dynamics

Using the pseudo-spin formalism, it is straightforward to derive the equations of motion.

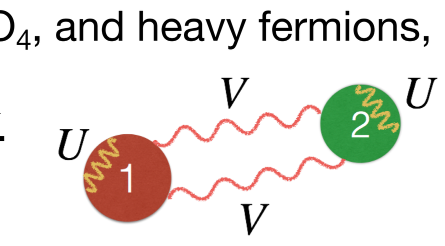
$\frac{d}{dt}\mathbf{S}_{\mathbf{k}}(t)=i\langle[H_{\mathrm{BCS}},\hat{\mathbf{S}}_{\mathbf{k}}]\rangle=-\mathbf{B}_{\mathbf{k}}(t)\times\mathbf{S}_{\mathbf{k}}(t)$ where the superconducting gap is time-dependent: $\Delta(t)=-U\sum_{\mathbf{k}}S^{-}_{\mathbf{k}}(t)$

- The equations of motion describe the procession motions of pseudo-spins. The effective magnetic field, however, is also oscillating, because it depends on all the pseudo-spins in momentum space.
- For quantum quenches (a sudden change of the pairing interaction U), the equations of motion can be solved exactly due to the integrability of BCS model[3,4].
- Solution for weak quantum quenches: $t^{-1/2}$-damped $2\Delta_{\infty}$-oscillation[3-5] $\Delta(t)=\Delta_{\infty}+a\frac{\cos(2\Delta_{\infty}t+\phi)}{(\Delta_{\infty}t)^{1/2}}$

Quench Dynamics of Two-band Superconductors

In several unconventional superconductors, such as $SrTiO_3$, iron pnictides, Sr_2RuO_4, and heavy fermions, multiple bands cross the Fermi level, giving rise to multiband superconductivity.

Two-band BCS model with intra-band interaction, U, and inter-band interaction, V.



- Need two sets of pseudo-spins to describe electrons in two different bands.
- It is not exactly solvable, once the two bands have different density of states.

❖ **Strategy:** Numerically solve the equations of motion using the 4th-order Runge-Kutta method. Obtain the analytic form of long-time asymptotic using perturbation theory and Laplace analysis.

Damping Effects of Gap Oscillations in Terahertz Pump-probe Experiments

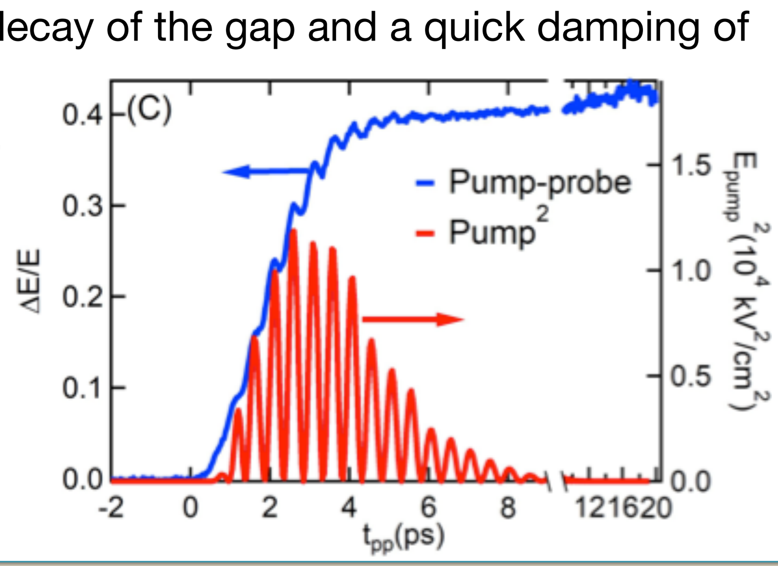
- Previous theoretical calculations only show coherent gap oscillations, due to the integrability of BCS.
- Recent terahertz pump-probe experiment observed a overall decay of the gap and a quick damping of the gap oscillation.

We consider two damping processes to the equations of motion.

$$\frac{d}{dt}\mathbf{S}_{\mathbf{k}}(t)=-\mathbf{B}_{\mathbf{k}}(t)\times\mathbf{S}_{\mathbf{k}}(t)-\frac{\mathbf{S}_{\mathbf{k}}(t)\cdot\hat{s}^{\parallel}_{\mathbf{k},\mathrm{eq}}-|\mathbf{S}_{\mathbf{k},\mathrm{eq}}|}{T_1}\hat{s}^{\parallel}_{\mathbf{k},\mathrm{eq}}-\sum_{i=1}^{2}\frac{\mathbf{S}_{\mathbf{k}}(t)\cdot\hat{s}^{\perp,i}_{\mathbf{k},\mathrm{eq}}}{T_2}\hat{s}^{\perp,i}_{\mathbf{k},\mathrm{eq}}$$

where $\mathbf{S}_{\mathbf{k},\mathrm{eq}}=\frac{\hat{s}_{\mathbf{k},\mathrm{eq}}}{2}\tanh\left(\frac{\sqrt{\varepsilon_{\mathbf{k}}^2+\Delta_*^2}}{2T_*}\right)$ is determined by the internal energy.

❖ **Strategy:** Numerically solve the equations of motion using the 8th-order Runge-Kutta method with adaptive stepsize control. Study the impacts on the gap dynamics due to T_1 and T_2 relaxation processes.

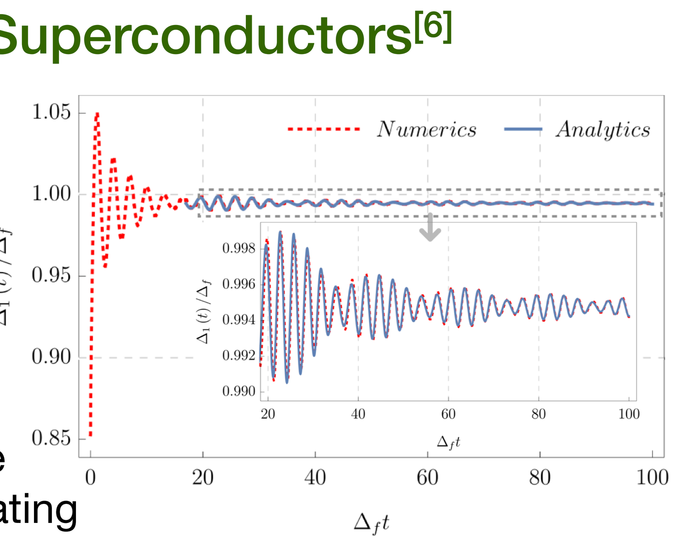


Results and Conclusions

Gap Oscillations in Two-band Superconductors[6]

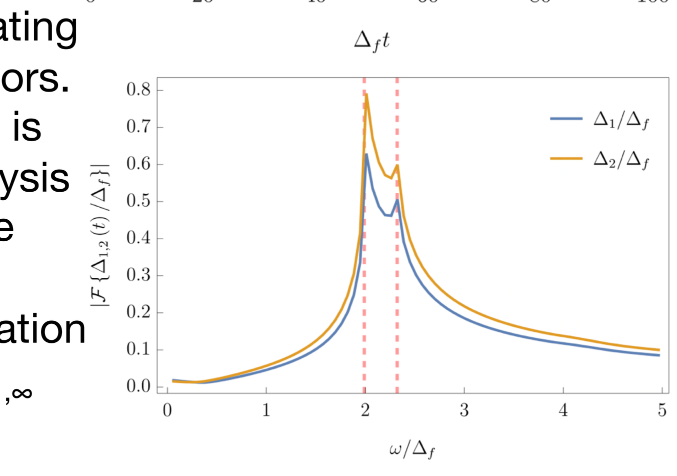
The quench dynamics of two-band superconductor is distinct from the single-band case once the two bands have different density of states.

Here we choose the ratio between the two density of states to be $\eta=\mathcal{N}_2/\mathcal{N}_1=0.7$



➢ In contrast to the $2\Delta_{\infty}$-oscillation in the single-band case, the gap exhibits beating oscillations in two-band superconductors.

➢ The damping of the beating oscillation is more rapid than $t^{-1/2}$. The Laplace analysis on the long-time asymptotic shows the damping is $t^{-3/2}$ instead.



➢ The Fourier spectrum of the gap oscillation shows two oscillation frequencies, $2\Delta_{1,\infty}$ and $2\Delta_{2,\infty}$, which promote the beating oscillation.

The long-time asymptotic of the gap oscillation:

$$\Delta_1(t)=\Delta_1^{\infty}+\mathcal{A}_1\frac{\sin\left(2\Delta_1^{\infty}t+\frac{\pi}{4}\right)}{(\Delta_1^{\infty}t)^{3/2}}+\mathcal{B}_1\frac{\sin\left(2|\Delta_2^{\infty}|t-\frac{\pi}{4}\right)}{(|\Delta_2^{\infty}|t)^{3/2}}+\mathcal{C}_1\frac{\sin\left(2|\Delta_2^{\infty}|t+\frac{\pi}{4}\right)}{(|\Delta_2^{\infty}|t)^{3/2}}$$

$$\Delta_2(t)=\Delta_2^{\infty}+\mathcal{A}_2\frac{\sin\left(2\Delta_2^{\infty}t+\frac{\pi}{4}\right)}{(\Delta_2^{\infty}t)^{3/2}}+\mathcal{B}_2\frac{\sin\left(2|\Delta_1^{\infty}|t-\frac{\pi}{4}\right)}{(|\Delta_1^{\infty}|t)^{3/2}}+\mathcal{C}_2\frac{\sin\left(2|\Delta_1^{\infty}|t+\frac{\pi}{4}\right)}{(|\Delta_1^{\infty}|t)^{3/2}}$$

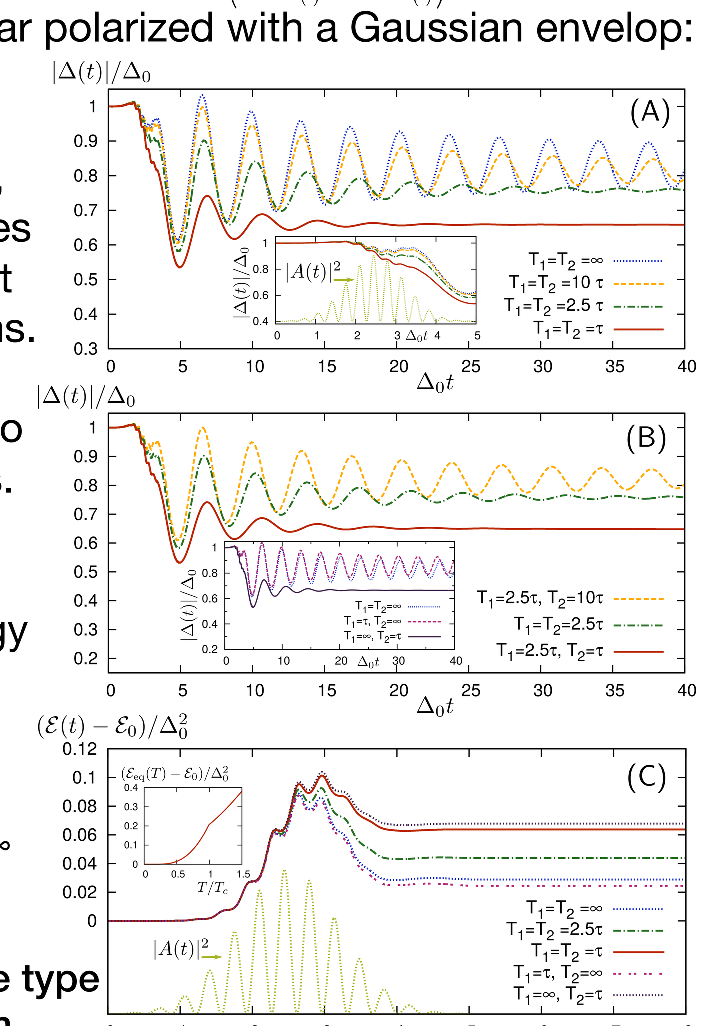
as shown in the figure, it agrees nicely with the numerical result.

Impact of Damping Induced by Terahertz pulse[7]

$$\frac{d}{dt}\mathbf{S}_{\mathbf{k}}(t)=-\mathbf{B}_{\mathbf{k}}(t)\times\mathbf{S}_{\mathbf{k}}(t)-\frac{\mathbf{S}_{\mathbf{k}}(t)\cdot\hat{s}^{\parallel}_{\mathbf{k},\mathrm{eq}}-|\mathbf{S}_{\mathbf{k},\mathrm{eq}}|}{T_1}\hat{s}^{\parallel}_{\mathbf{k},\mathrm{eq}}-\sum_{i=1}^{2}\frac{\mathbf{S}_{\mathbf{k}}(t)\cdot\hat{s}^{\perp,i}_{\mathbf{k},\mathrm{eq}}}{T_2}\hat{s}^{\perp,i}_{\mathbf{k},\mathrm{eq}}$$

- To simulate the terahertz-pump-probe experiment, we coupled the electrons with the laser field via the Peierls substitution, which changes the effective magnetic field in the pseudo-spin formalism: $\mathbf{B}_{\mathbf{k}}(t)=2\Delta'(t)\hat{x}-2\Delta''(t)\hat{y}-\left(\varepsilon_{\mathbf{k}-e\mathbf{A}(t)}+\varepsilon_{\mathbf{k}+e\mathbf{A}(t)}\right)\hat{z}$
- The laser field is chosen to be 0 degree linear polarized with a Gaussian envelop:

$$\boldsymbol{A}(t)=\hat{x}A_0\exp\left[\frac{(t-\tau/2)^2}{2\sigma^2}\right]\cos(\omega_{\mathrm{pump}}t)$$



➢ By taking relaxation processes into account, our numerical simulation perfectly reproduces the recent terahertz-pump-probe experiment on superconducting niobium-nitride thin films.

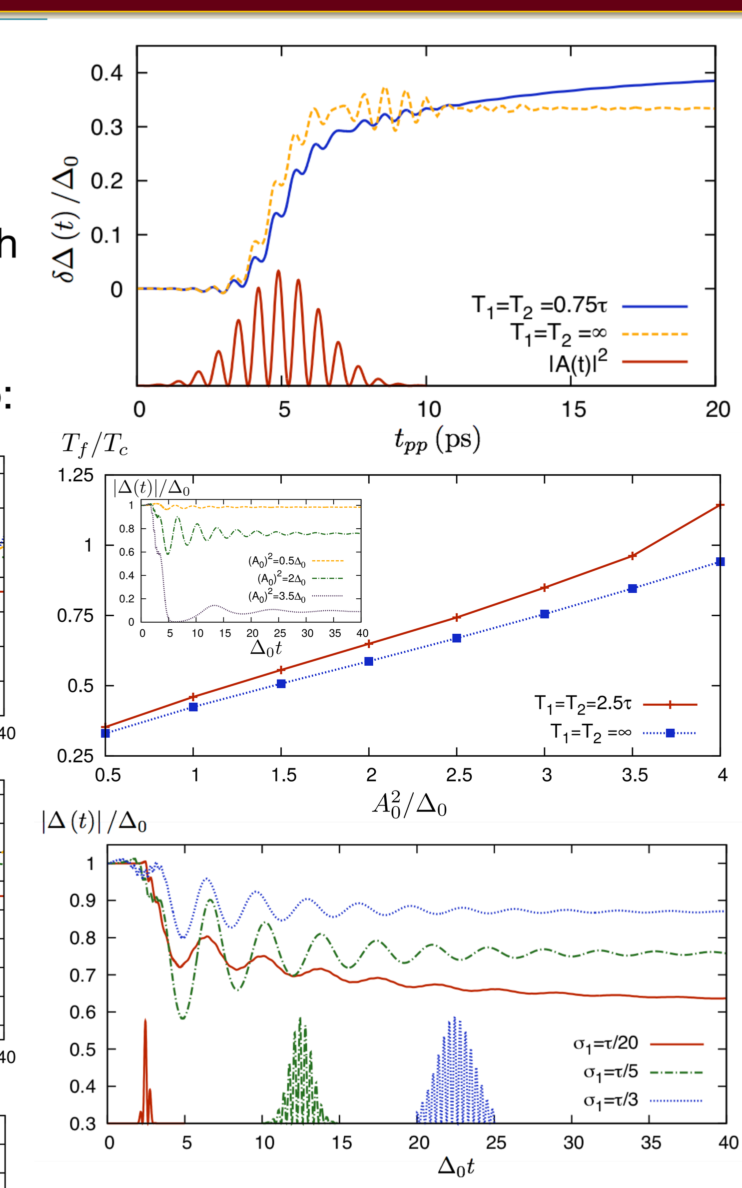
➢ The T_1 relaxation process corresponds to relaxation of the electronic distribution due to residue interactions between quasi-particles.

➢ The T_2 relaxation process describes the dephasing of the coherent oscillation.

➢ The efficiency of the system to absorb energy from the terahertz pump is more affected by the T_2 process than the T_1 process.

➢ Our simulation shows also that a shorter pump pulse is more efficient to drive the $2\Delta_{\infty}$ oscillation of the gap.

Further application of this approach to different superconductors will allow one to distinguish the type of relaxation processes dominant in each system.



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