

Exploring Performance Degradation in Niobium Thin Film Radio-Frequency Cavities

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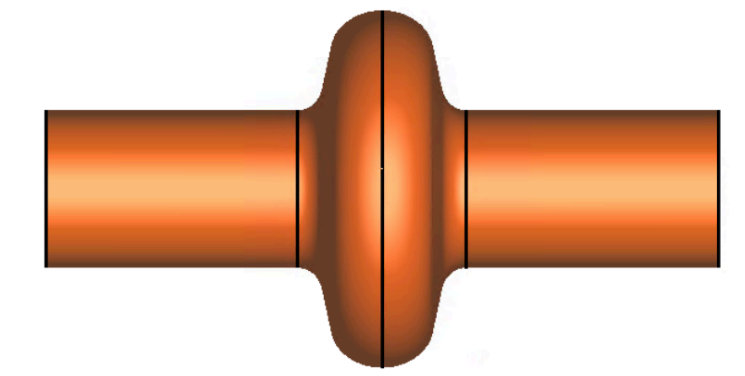
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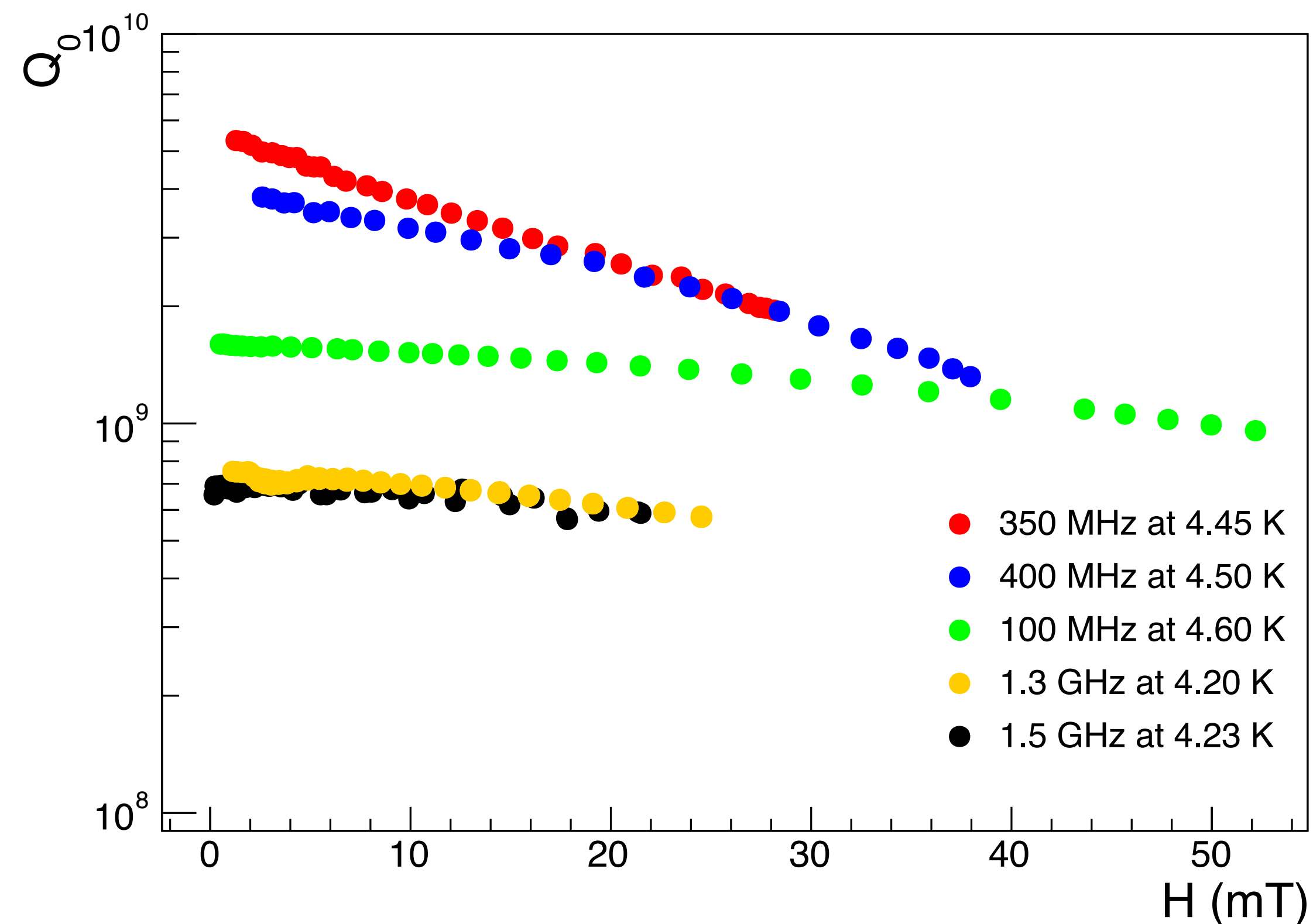
September, 16th 2024



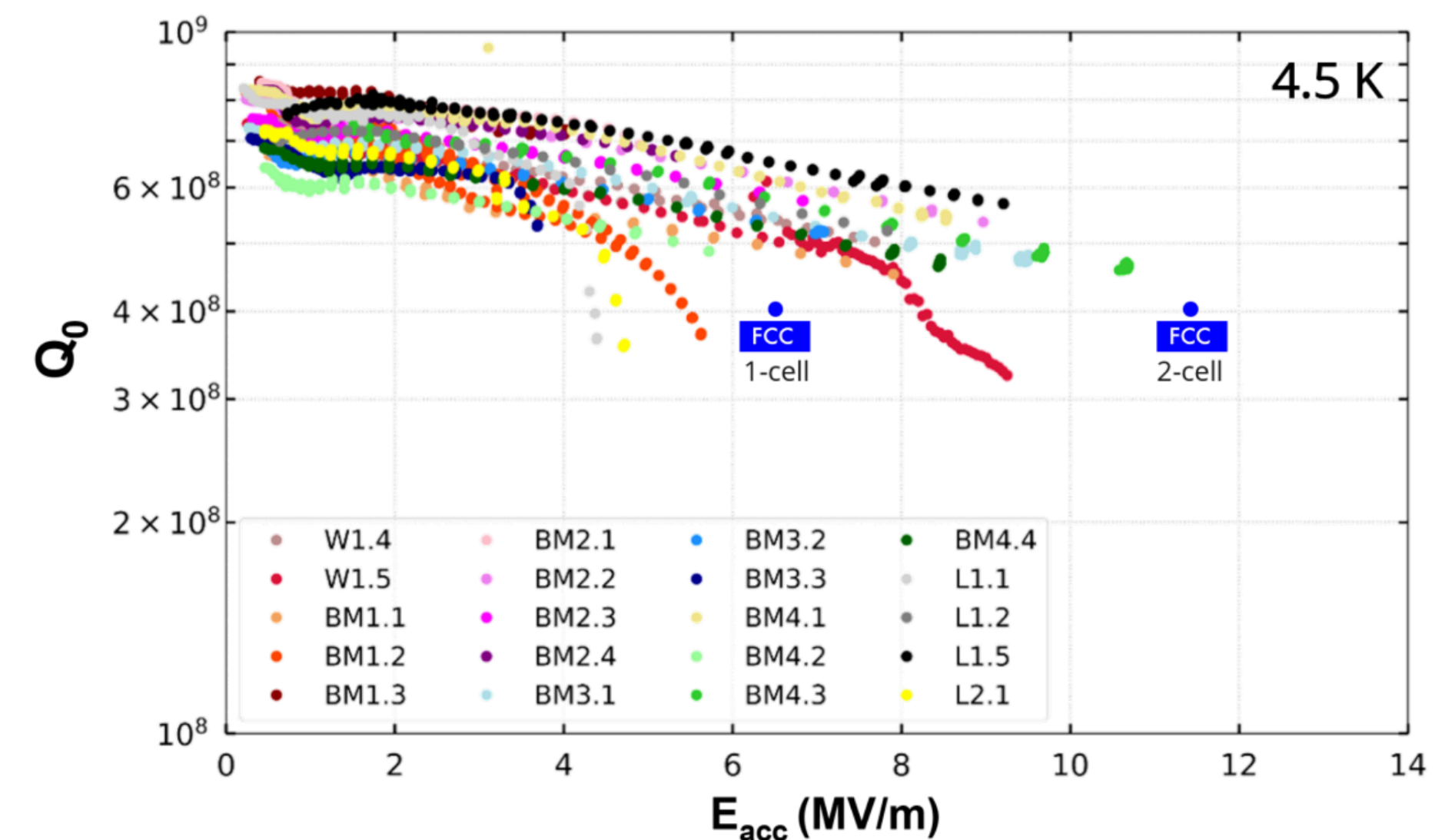
Medium-field Q-slope problem



Niobium thin film cavities generally exhibit **significant performance degradation** with increasing accelerating field (medium-field **Q-slope problem**). This issue is notably more pronounced in niobium thin film cavities compared to bulk niobium cavities.

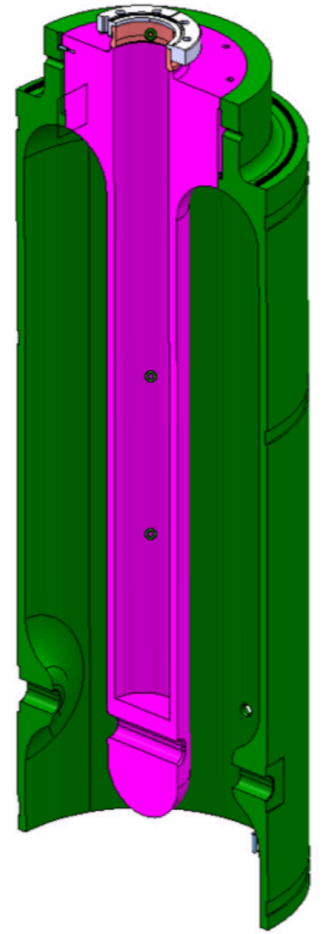


Many efforts are put in place in view of the potential implementation of **niobium thin film cavities** in future accelerators.

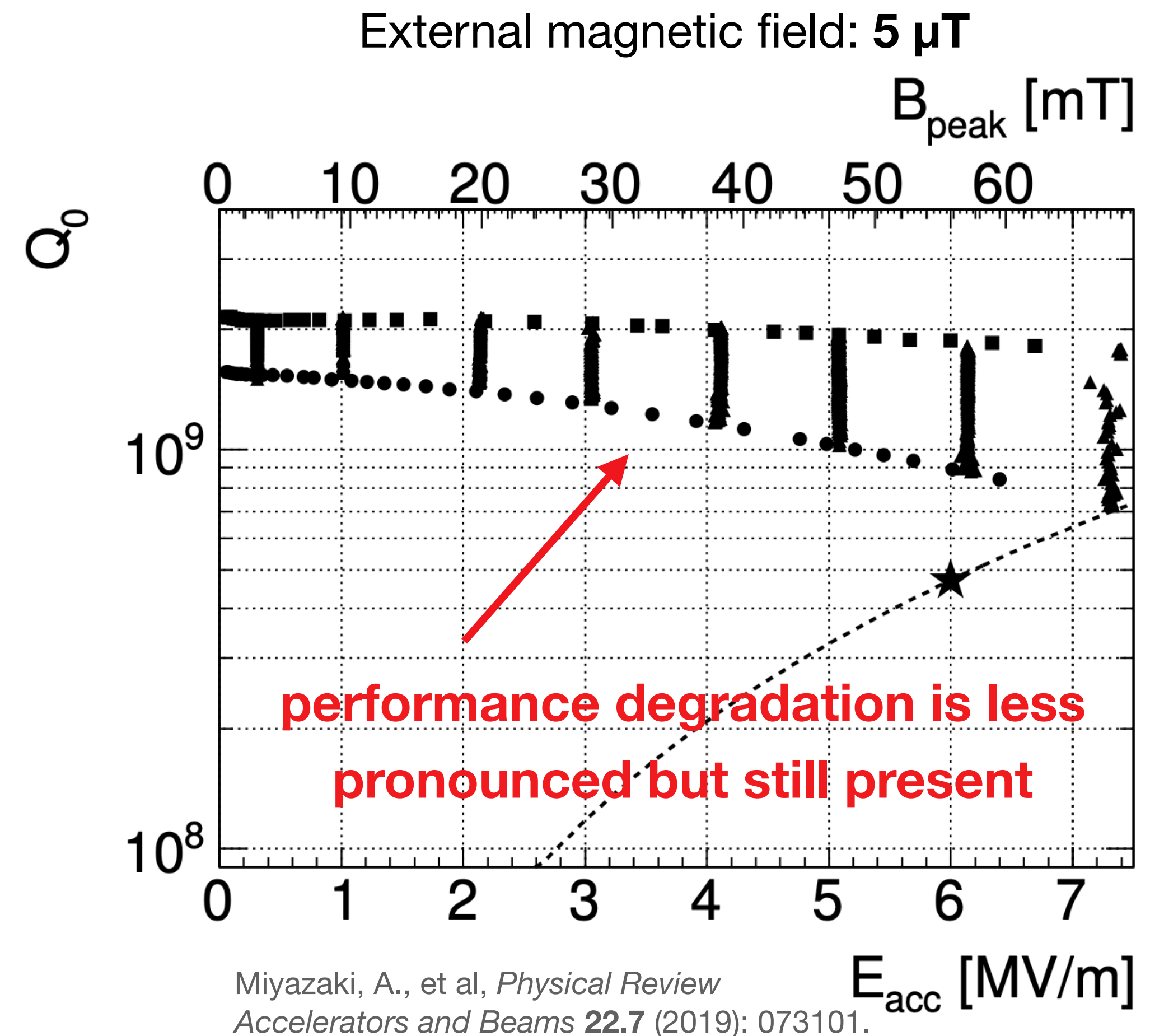
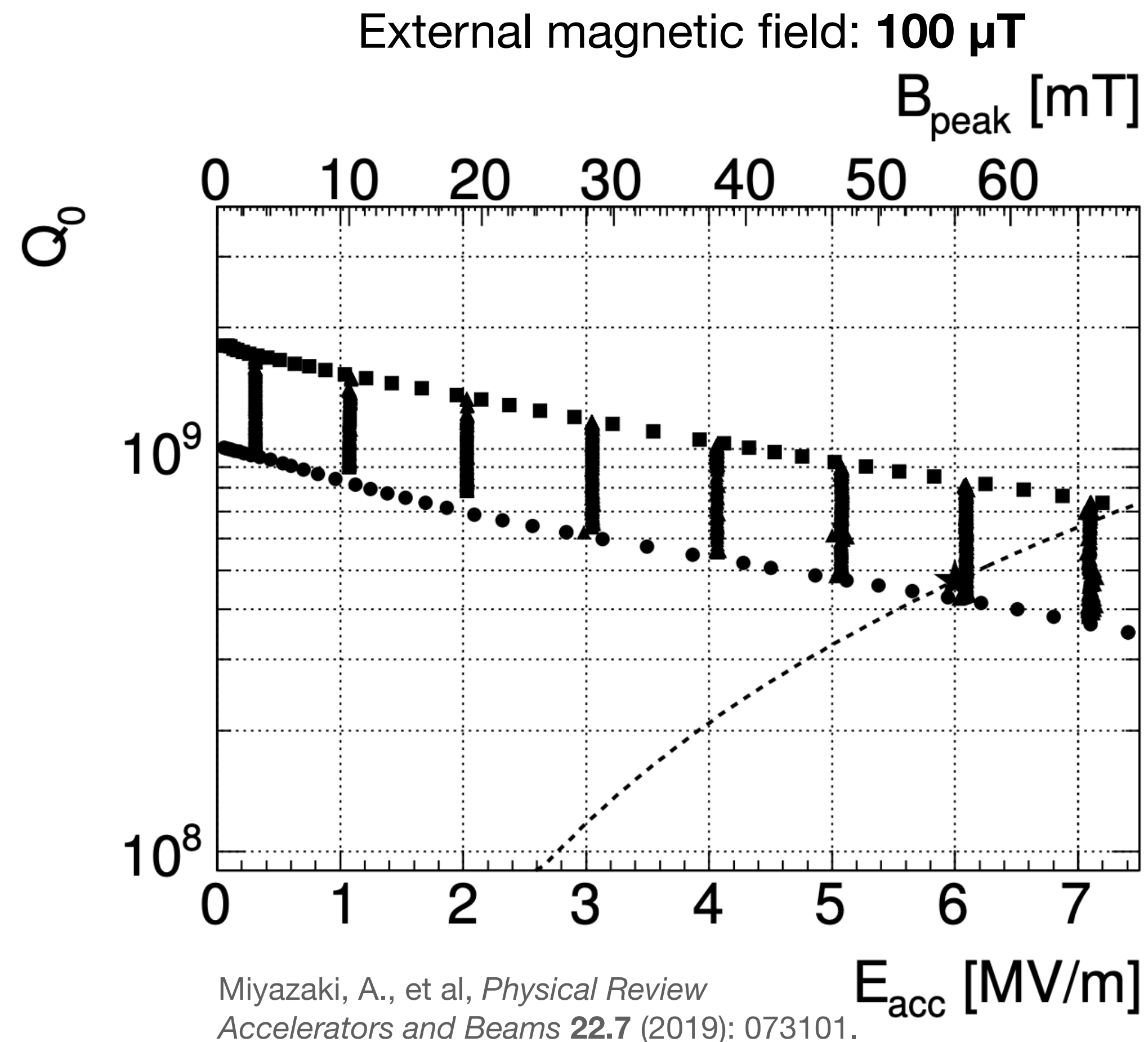


Vega Cid, L., et al, *JACoW SRF* (2023): 621-626.

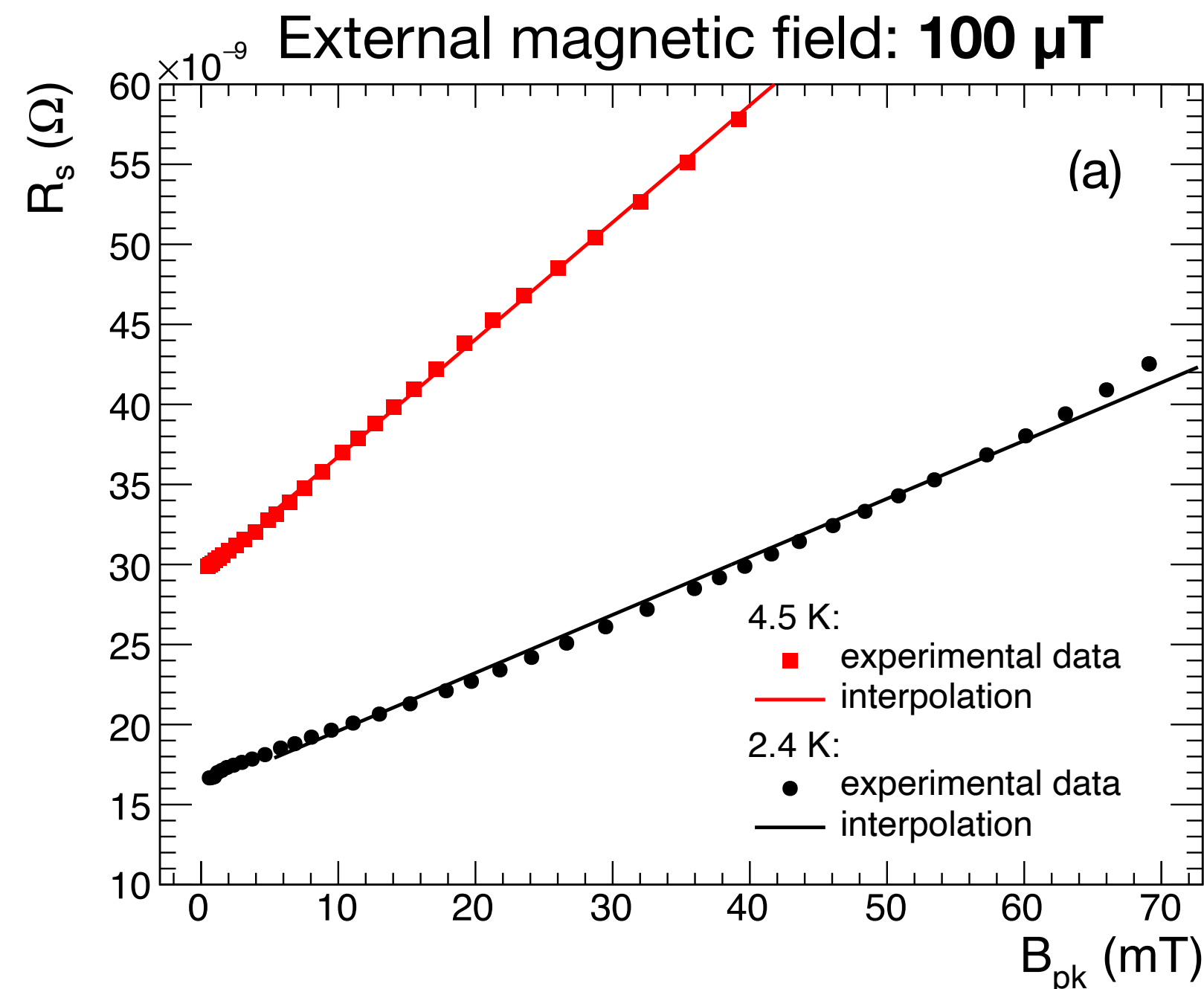
Effects of trapped magnetic field



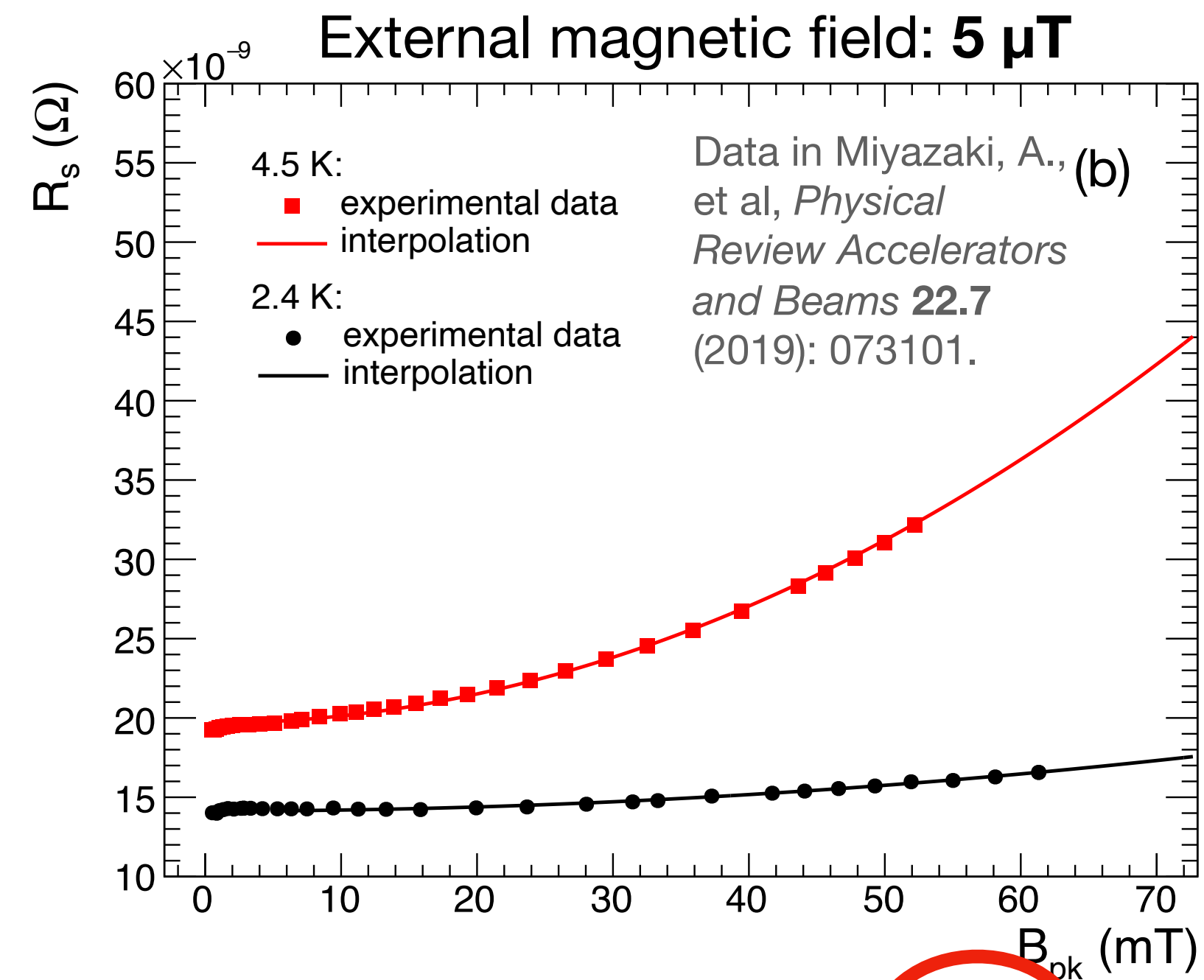
In a dedicated test on a 100 MHz thin film cavity for HIE-ISOLDE:



Effects of trapped magnetic field



$$R_s = R_0 + R_{\text{slope}} \times B_{\text{pk}}$$



$$R_s = R_0 + R_{\text{slope}} \times (B_{\text{pk}})^2$$

The different trends suggest that **two distinct mechanisms may contribute to the Q-slope problem** in niobium thin film cavities.

The present study focuses on cavities in which **R_s increases proportionally to $(B_{\text{pk}})^2$** with the intention to potentially disregarding the effects of trapped magnetic field in the analysis of performance degradation.

Existing works in the literature

- A. Gurevich, “[Multiscale mechanisms of SRF breakdown](#)”, *Physica C*, **441** (2006): 38–43 → The first work where the dependence of R_s on B^2 is introduced in clean bulk niobium by considering pair-breaking in a current-carrying state.
- P. Bauer, *et al.*, “[Evidence for non-linear BCS resistance in SRF cavities](#)”, *Physica C*, **441** (2006): 51–56 → Dependence of R_s on B^2 is introduced taking into account the impurity scattering

→ These models were developed specifically for **bulk niobium cavities** and **did NOT explain the strong B^2 dependence in niobium-copper cavities**

- Palmieri, V., and Vaglio, R., “[Thermal contact resistance at the Nb/Cu interface as a limiting factor for sputtered thin film RF superconducting cavities](#)”, *Superconductor Science and Technology* **29.1** (2015): 015004:
This is a film-specific model considering micro-quenches caused by limited thermal contacts at the niobium-copper interfaces in thermal equilibrium state

→ This was falsified by two major counter-evidences:

1. **micro-quenches cannot be thermodynamically stable** even in the Nb/Cu cavities;
2. **temperature dependence of Q-slope (2 K vs 4 K) was not explained.**

[Miyazaki, A., and Venturini Delsolaro, W., *Physical Review Accelerators and Beams* **22.7** (2019): 073101]

We need to develop a new model specifically to niobium-coated copper cavities and re-formulate the problem of interface in a NON-thermal way!

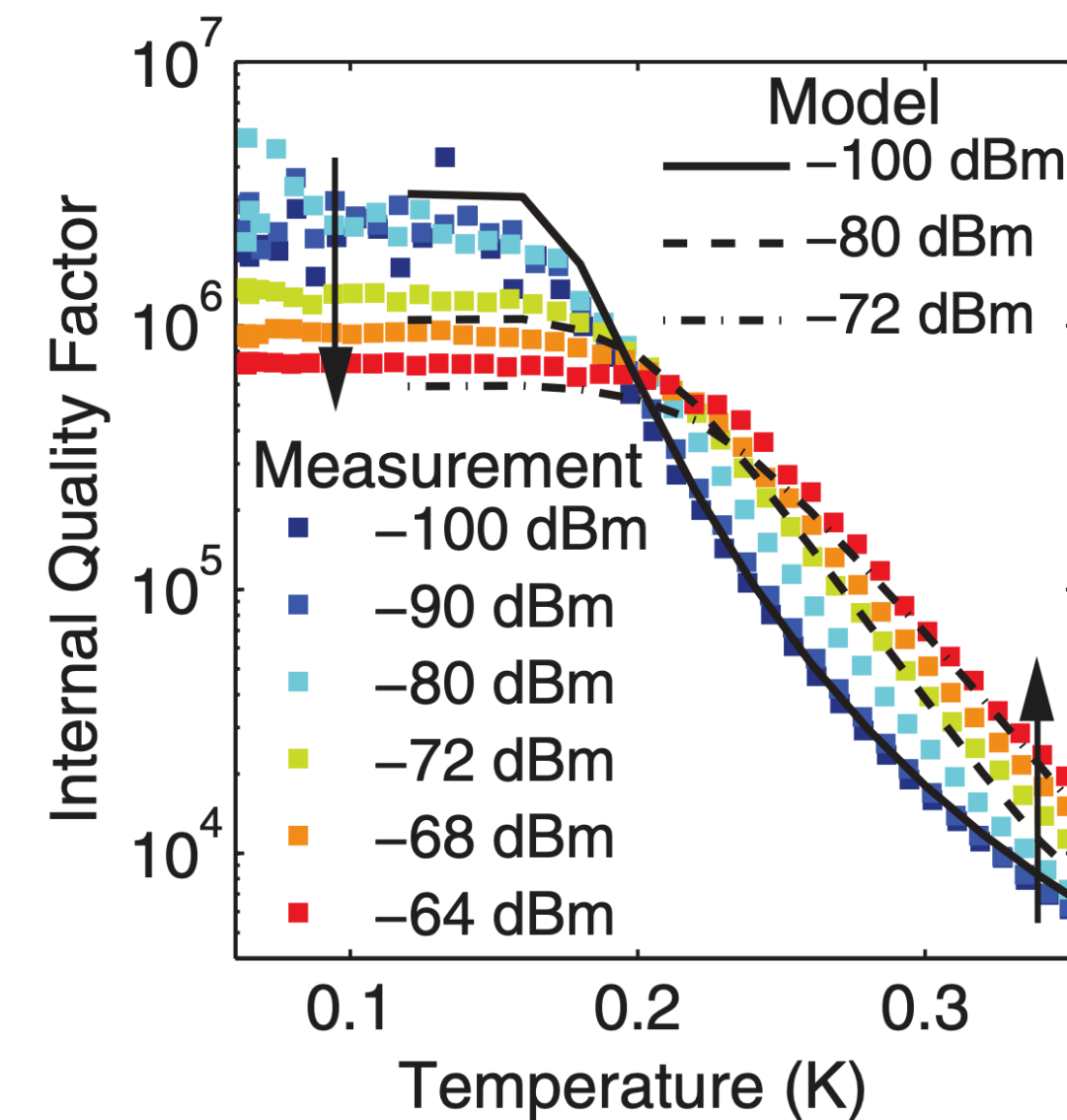
Eliashberg's theory

In recent years, a theory initially developed by Eliashberg has been suggested as a potential explanation for the **observed enhancement in performance** with increasing accelerating field in non-doped niobium cavities and nitrogen-doped cavities. **In this theory, both B_{pk}^2 and frequency play a crucial role.**

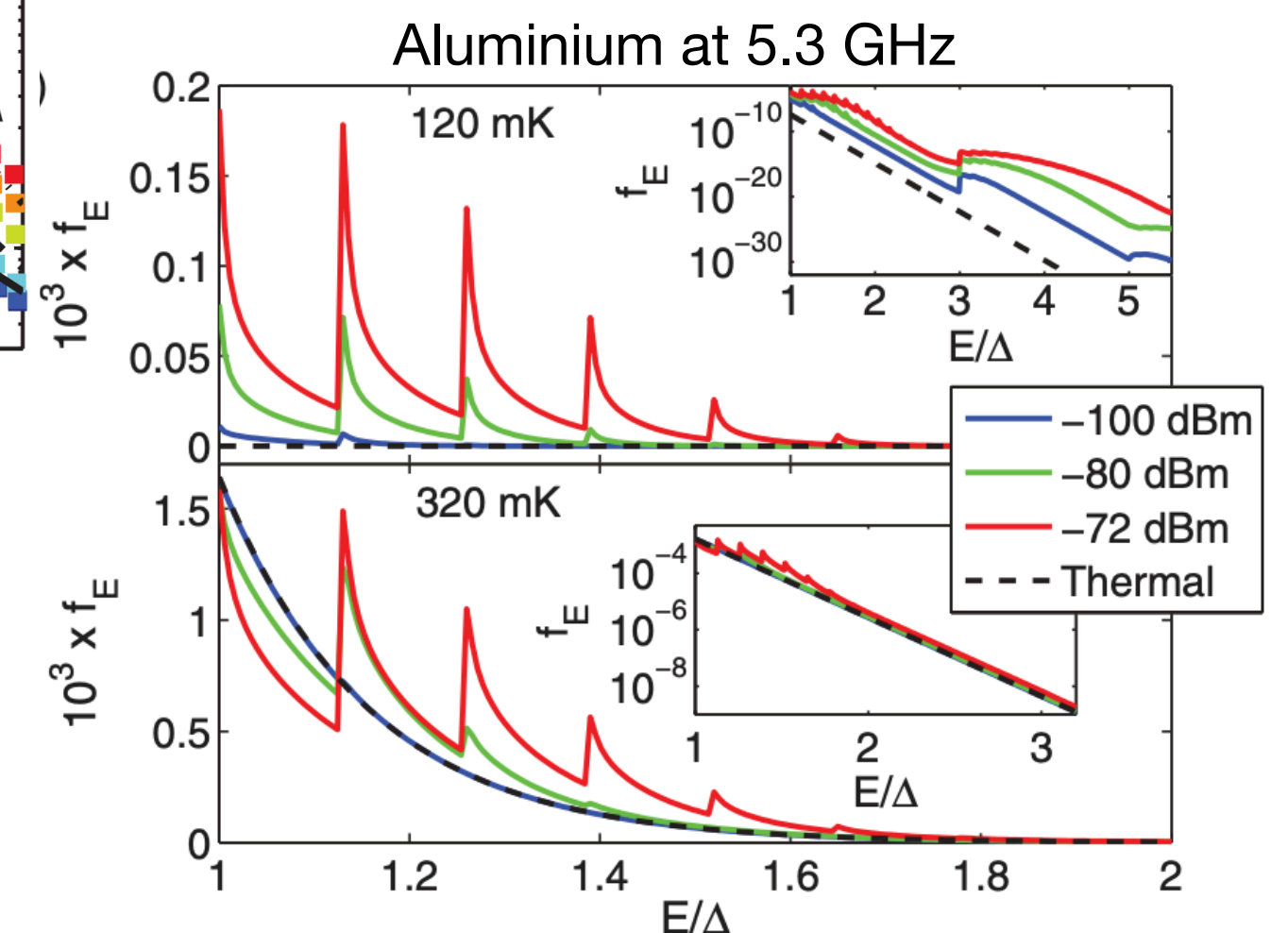
The hypothesis that the anti- Q slope of N-doped cavities may originate from a deviation of the quasiparticle energy distribution from thermal equilibrium was first proposed by Romanenko [32] and further corroborated in this Letter.

In agreement with the Eliashberg theory [33], superconductors start showing nonequilibrium effects above a certain frequency threshold at which quasiparticles populate high-energy states far from the gap edge. This redistribution of quasiparticles decreases the probability of photon absorption, lowering the dissipation and hence the surface resistance. As previously reported for aluminum cavities [34], such a regime of stimulated superconductivity

Martinello, M., et al, *Physical Review Letters* **121.22** (2018): 224801.

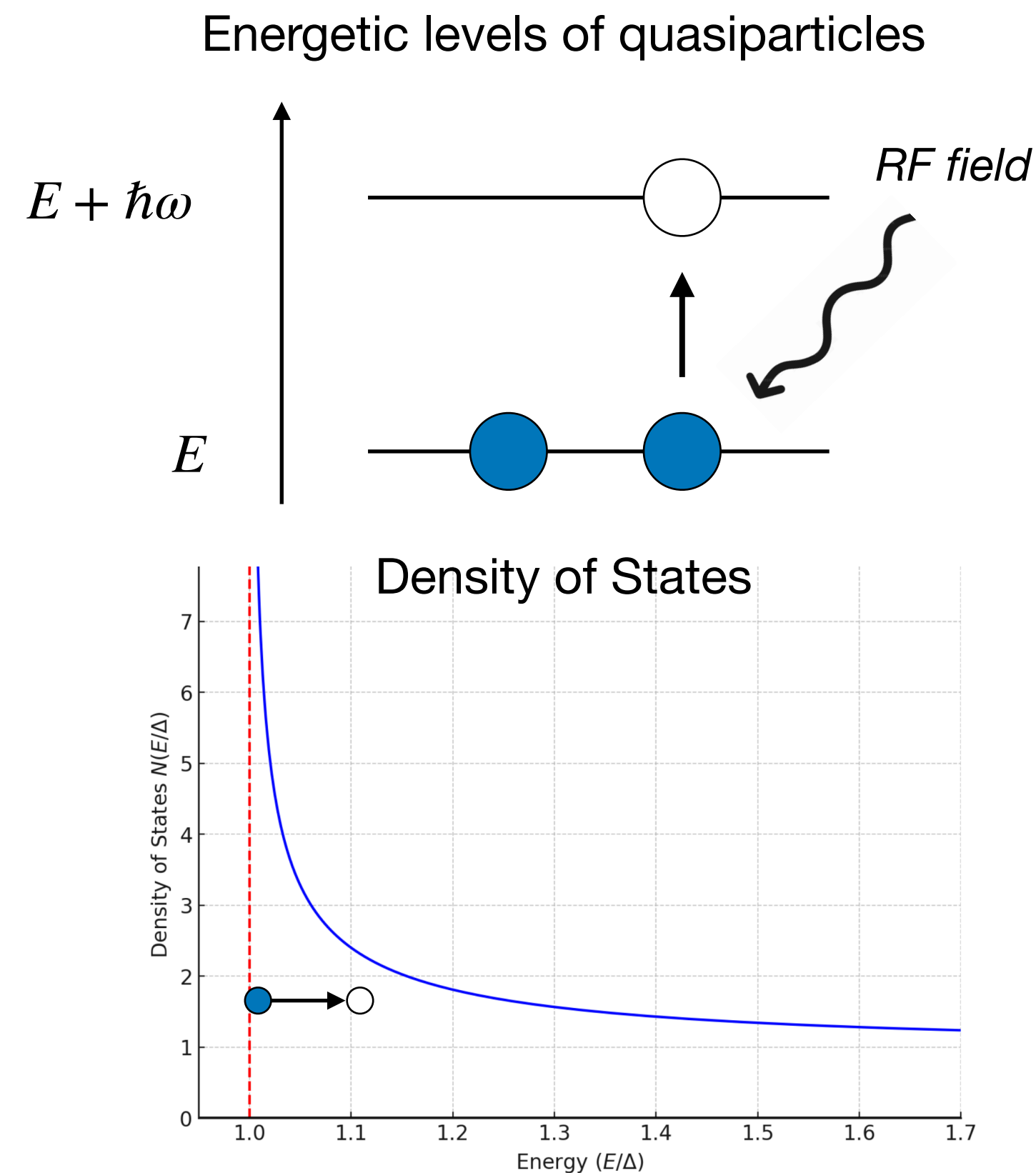


De Visser, P. J., et al, *Physical Review Letters* **112.4** (2014): 047004.



Enhancement or weakening of superconductivity

The quasiparticle population is driven out of thermal equilibrium by microwaves into either a steady-state dynamic equilibrium or a more general time-dependent regime. [Tinkham, M. *Introduction to superconductivity*, 2004]



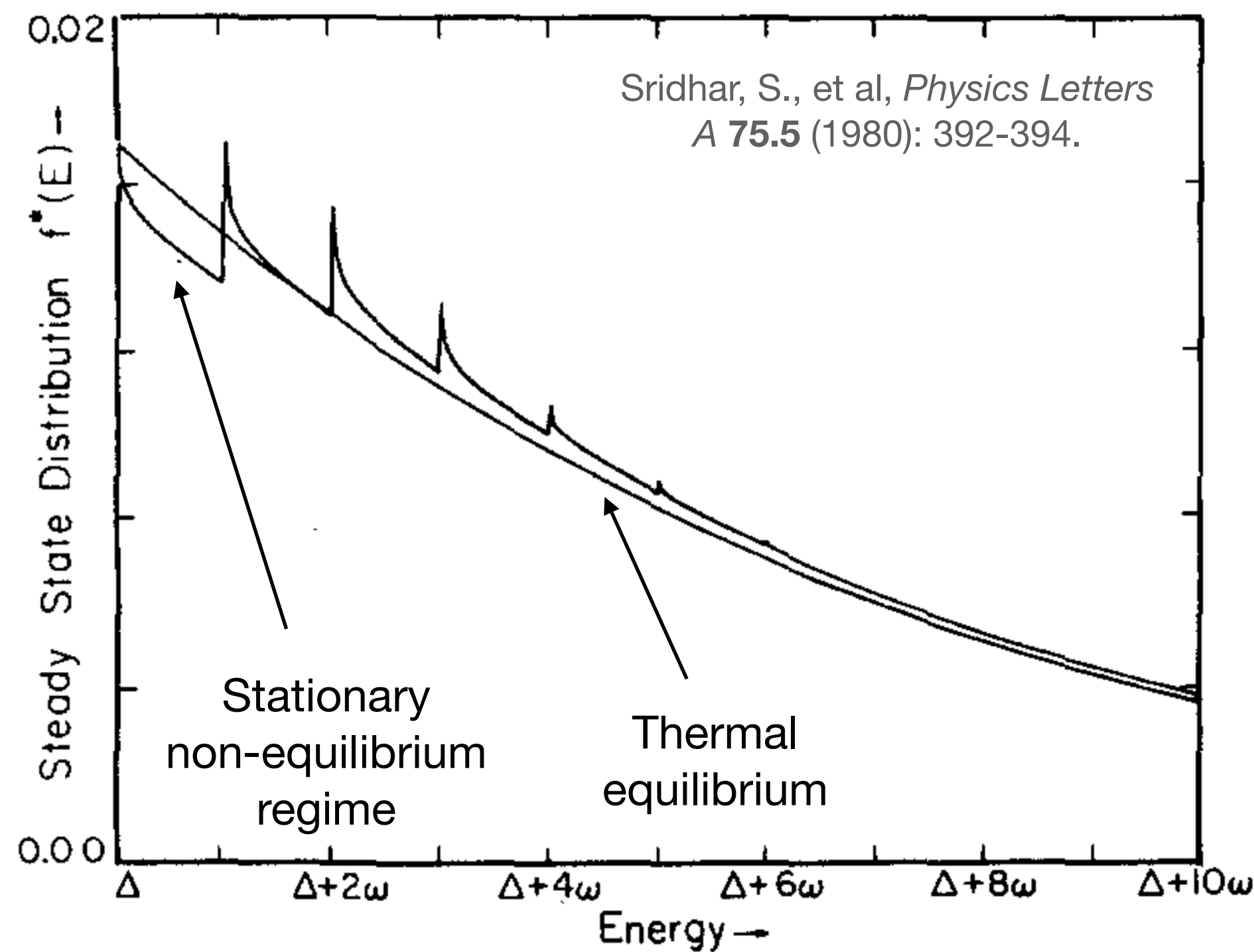
The consequence is an **effective enhancement or weakening of superconductivity due to non-equilibrium phenomena.**

Depending on the microwave frequency:

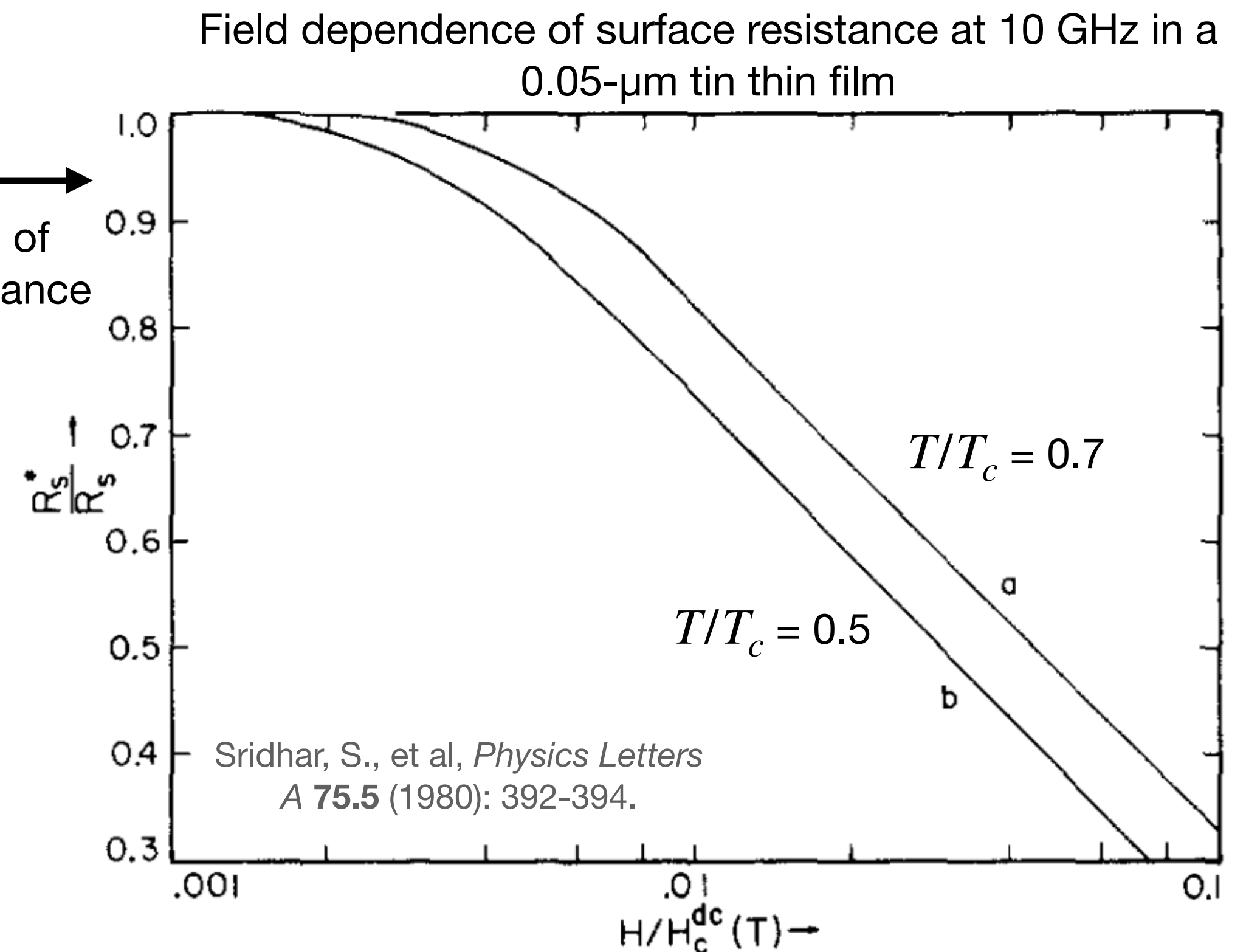
- A dynamic equilibrium is achieved when the perturbing source (microwaves) is balanced by relaxation, leading to a stationary non-equilibrium state that differs from thermal equilibrium. In this scenario, an enhancement of the superconducting energy gap can occur.
 - The commonly believed frequency threshold for the onset of this phenomenon is **higher than 10 GHz in niobium for $T \rightarrow T_c$.**
- Non-equilibrium phenomena at lower frequencies lead to a weakening of superconductivity. [Tinkham, M. *Introduction to superconductivity*, 2004]
 - **This may be the key to understand the Q-slope problem, considering the electron-phonon scattering and their respective dynamics simultaneously.**

Redistribution of quasiparticles

Quasiparticles pushed to higher energy states have fewer available states to occupy, while simultaneously inhibiting other quasiparticles from being promoted from lower energy states. This implies the reduction of surface resistance.



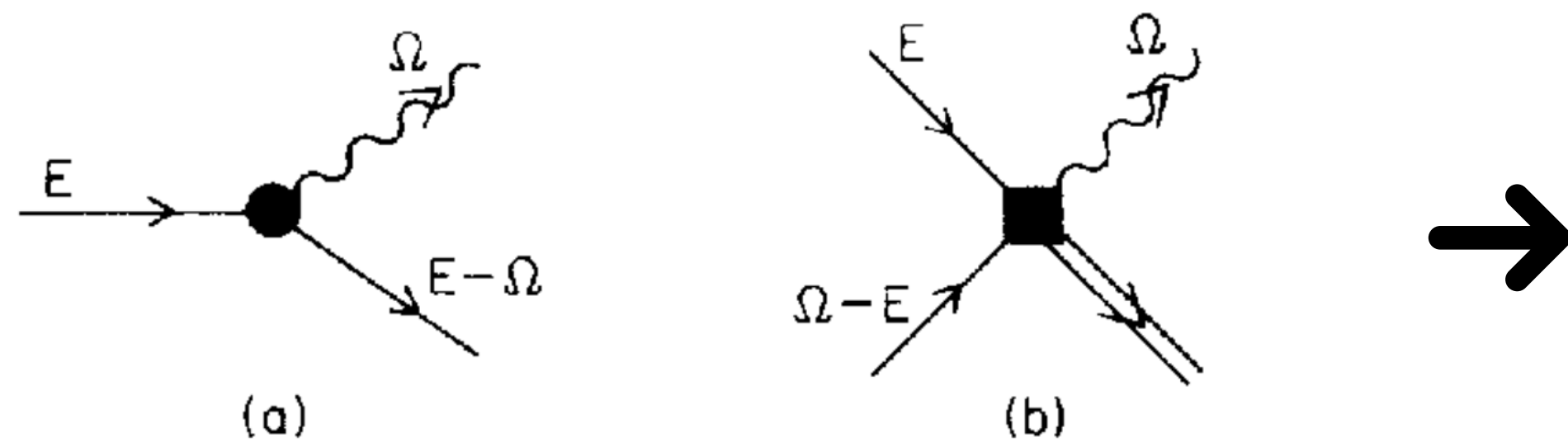
Calculation of surface resistance



In the calculation, phonons are considered in thermal equilibrium

The role of phonons

When perturbed out of equilibrium, a quasiparticle system tends to return to equilibrium through inelastic **scattering** of quasiparticles with all-energy phonons and quasiparticle **recombination** with phonons of energy $\Omega \geq 2\Delta$ (pair-breaking phonons).

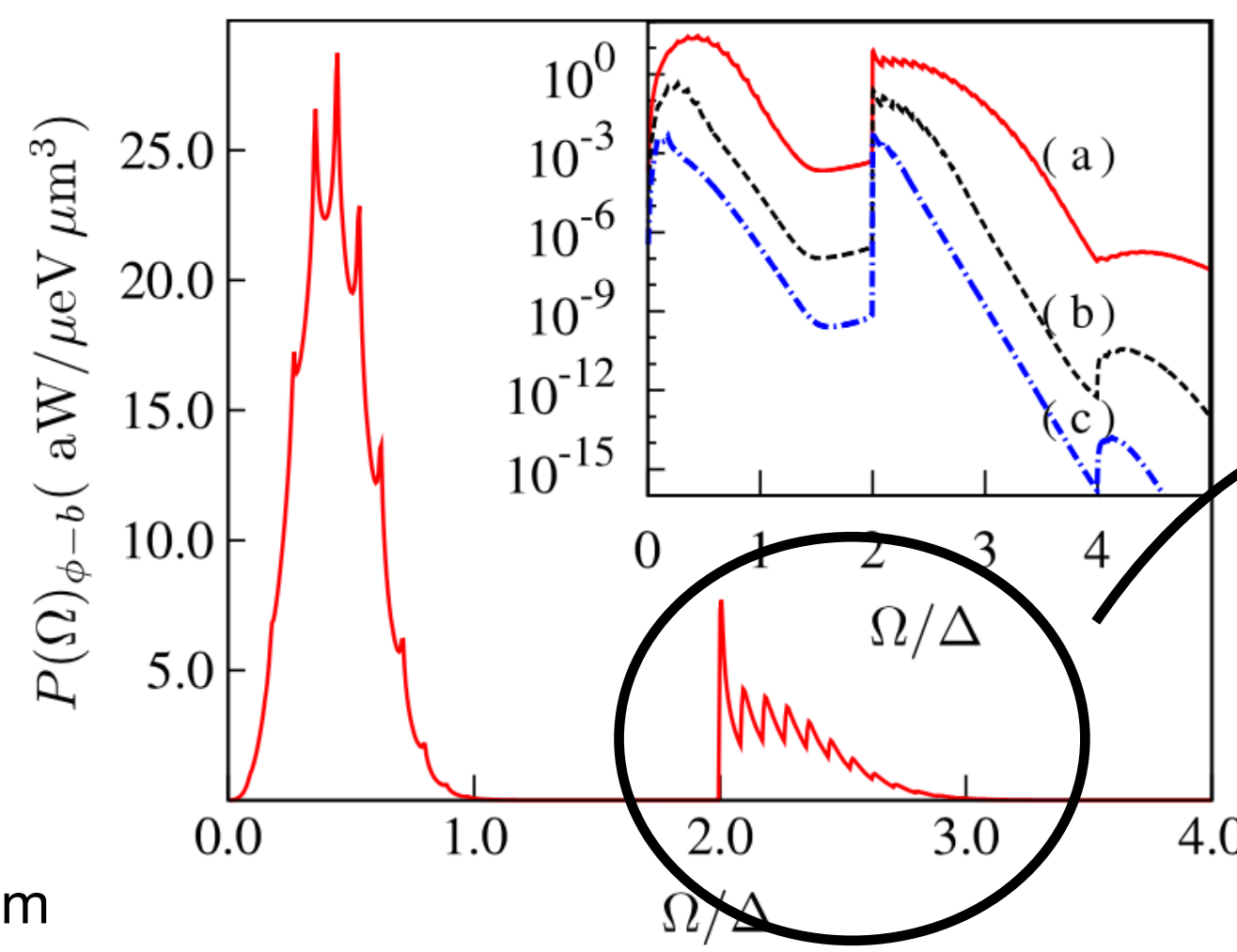
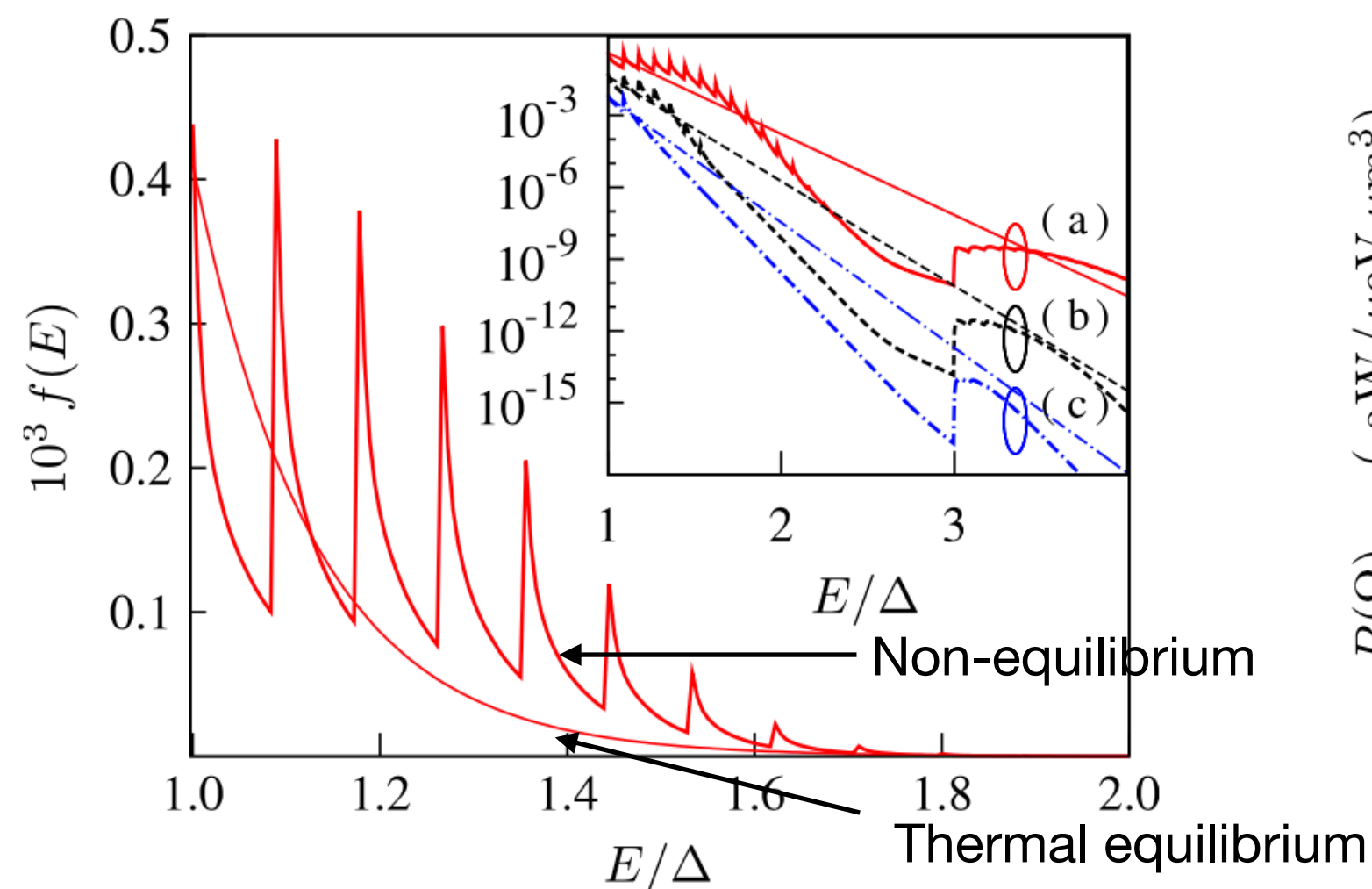


The total relaxation rate is: $1/\tau = 1/\tau_s + 1/\tau_r$ where τ_s and τ_r are the scattering and recombination lifetimes, respectively.

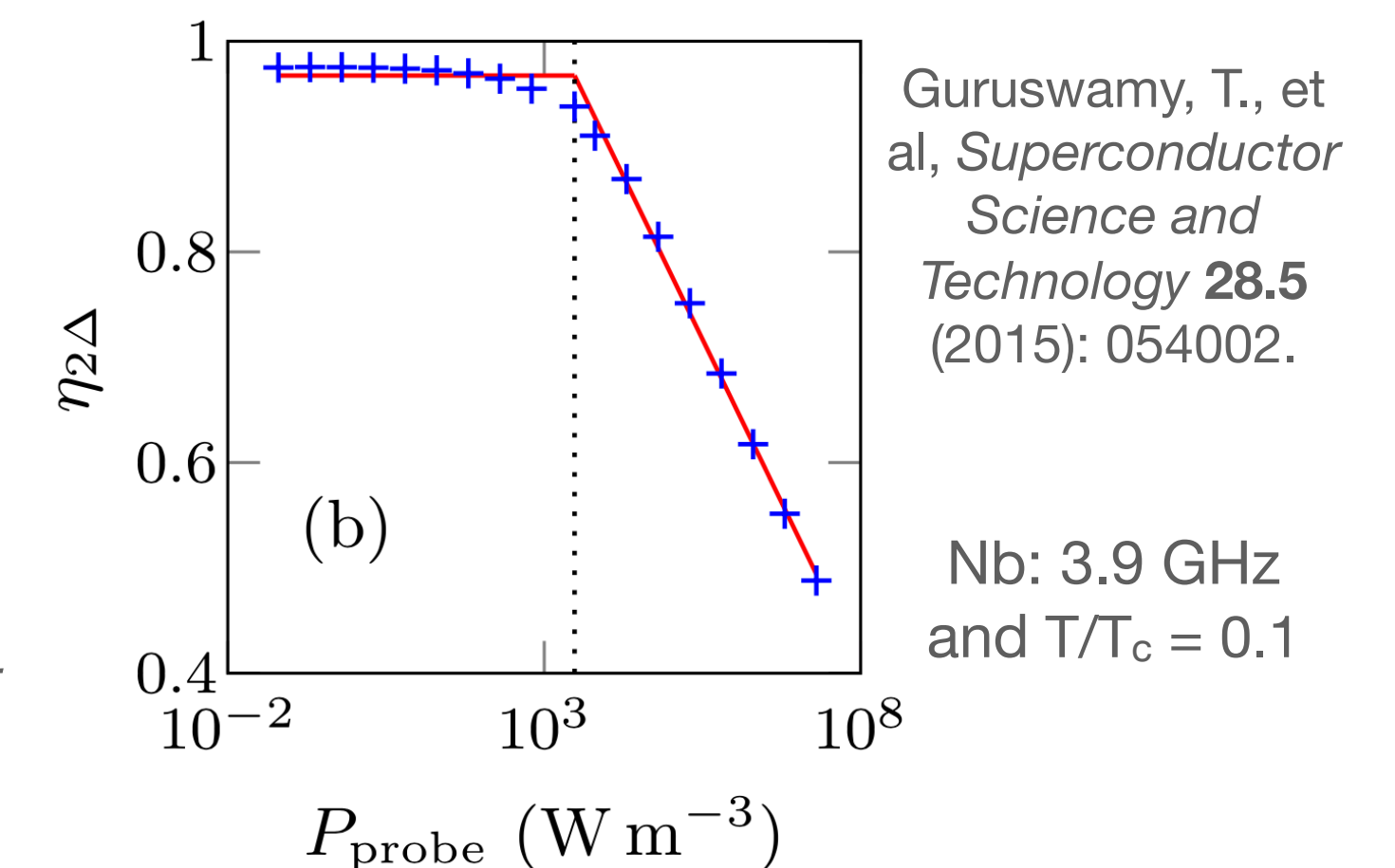
$$\tau_s(\Delta, T) = \tau_0 \frac{1}{\Gamma(\frac{7}{2})\zeta(\frac{7}{2})} \left[\frac{2\Delta(0)}{kT} \right]^{1/2} \left[\frac{T_c}{T} \right]^{7/2},$$

$$\tau_R(\Delta, T) = \tau_0 \frac{1}{\sqrt{\pi}} \left[\frac{kT_c}{2\Delta(0)} \right]^{5/2} \left[\frac{T_c}{T} \right]^{1/2} \exp(\Delta(0)/kT)$$

Kaplan, S. B., et al, *Physical Review B* **14.11** (1976): 4854



Goldie, D. J., et al, *Superconductor Science and Technology* **26.1** (2012): 015004.



Guruswamy, T., et al, *Superconductor Science and Technology* **28.5** (2015): 054002.

Nb: 3.9 GHz and $T/T_c = 0.1$

Chang–Scalapino mechanism

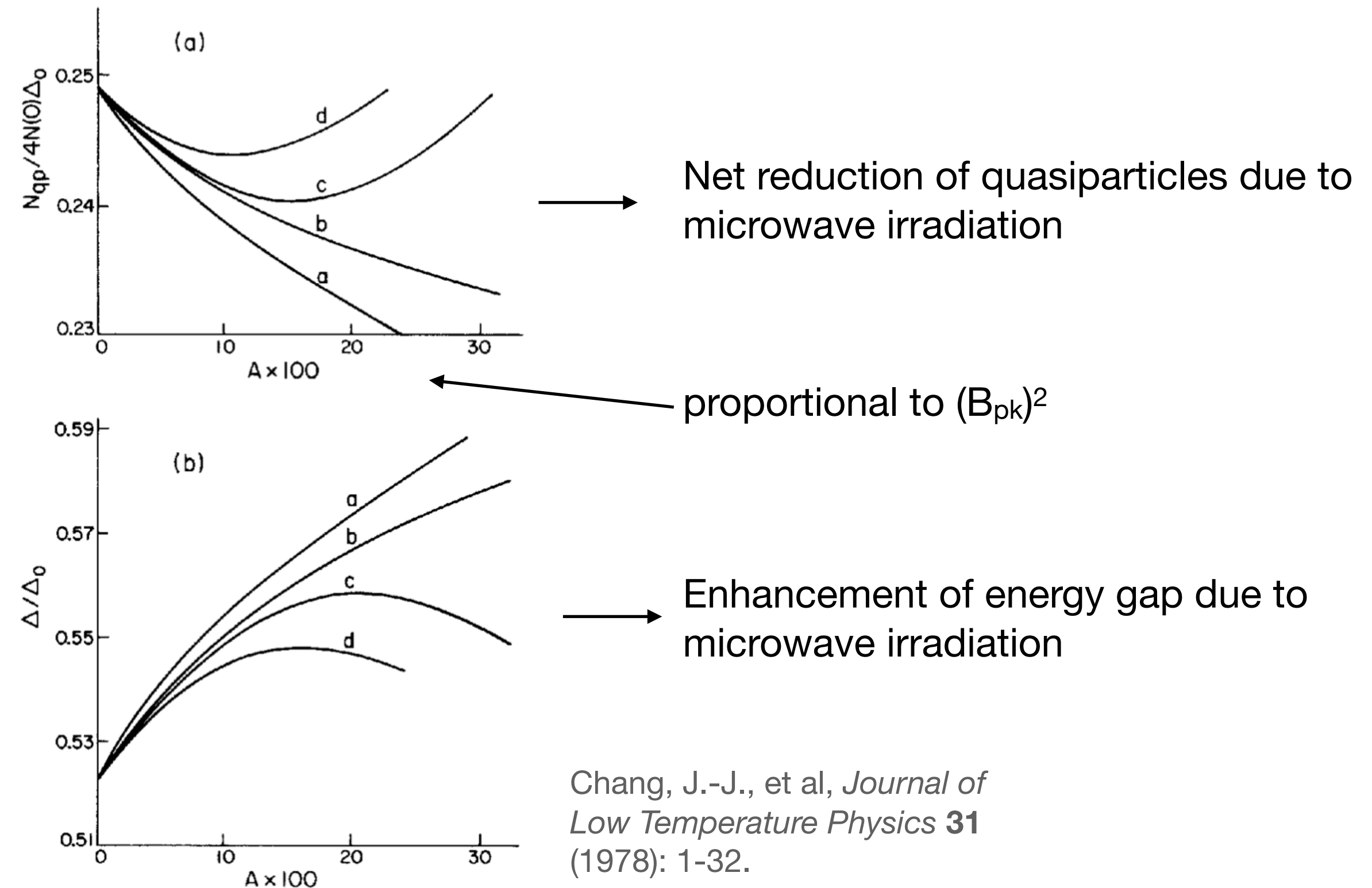
By solving the kinetic equations for quasiparticle and phonon distributions coupled with the BCS gap equation:

$$\begin{aligned} \frac{df(E)}{dt} = & I_{qp}(E) - \frac{2\pi}{\hbar} \int_0^\infty d\Omega \alpha^2(\Omega) F(\Omega) \rho(E+\Omega) \left(1 - \frac{\Delta^2}{E(E+\Omega)}\right) \\ & \times \{f(E)[1-f(E+\Omega)]n(\Omega) - f(E+\Omega)[1-f(E)][n(\Omega)+1]\} \\ & - \frac{2\pi}{\hbar} \int_0^{E-\Delta} d\Omega \alpha^2(\Omega) F(\Omega) \rho(E-\Omega) \left(1 - \frac{\Delta^2}{E(E-\Omega)}\right) \\ & \times \{f(E)[1-f(E-\Omega)][n(\Omega)+1] - [1-f(E)]f(E-\Omega)n(\Omega)\} \\ & - \frac{2\pi}{\hbar} \int_{E+\Delta}^\infty d\Omega \alpha^2(\Omega) F(\Omega) \rho(\Omega-E) \left(1 + \frac{\Delta^2}{E(\Omega-E)}\right) \\ & \times \{f(E)f(\Omega-E)[n(\Omega)+1] - [1-f(E)][1-f(\Omega-E)]n(\Omega)\} \end{aligned}$$

and

$$\begin{aligned} \frac{dn(\Omega)}{dt} = & I_{ph}(\Omega) - \frac{8\pi}{\hbar} \frac{N(0)}{N} \int_\Delta^\infty dE \int_\Delta^\infty dE' \alpha^2(\Omega) \rho(E) \rho(E') \\ & \times \left(\left(1 - \frac{\Delta^2}{EE'}\right) \{f(E)[1-f(E')]n(\Omega) \right. \\ & \left. - f(E')[1-f(E)][n(\Omega)+1]\} \right. \\ & \times \delta(E+\Omega-E') + \frac{1}{2} \left(1 + \frac{\Delta^2}{EE'}\right) \{[1-f(E)][1-f(E')]n(\Omega) \\ & \left. - f(E)f(E')[n(\Omega)+1]\} \delta(E+E'-\Omega) \right) - \frac{n(\Omega) - n(\Omega, T)}{\tau_{es}} \end{aligned}$$

The **recombination of higher-energy quasiparticles is more rapid than that of lower-energy quasiparticles**, due to the larger phonon density of states involved:



This second enhancement effect is generally comparable in importance to the effect caused by recombination of quasiparticles.

Energy gap equation in Eliashberg's theory

For low absorbed power values, the quasiparticle density can be expressed as:

$$n(\epsilon) = n_0(\epsilon) + n_1(\epsilon)$$

Fermi-Dirac distribution
small, field-induced quasiparticle density

For temperatures close to T_c , the gap equation is:

$$\frac{T_c - T}{T_c} - \frac{7\zeta(3)}{8\pi^2} \frac{\Delta^2}{(k_B T_c)^2} - 2 \int_{\Delta}^{\infty} \frac{n_1(\epsilon)}{\sqrt{\epsilon^2 - \Delta^2}} d\epsilon = 0$$

non-equilibrium term

As reported in the literature, the non-equilibrium contribution can be expressed as the sum of three terms:

$$\frac{T_c - T}{T_c} - \frac{7\zeta(3)}{8\pi^2} \left(\frac{\Delta}{k_B T_c}\right)^2 + \frac{\alpha}{\gamma} \left[-\frac{\pi\gamma}{2k_B T_c} - 0.172 \left(\frac{\hbar\omega}{k_B T_c}\right)^2 + \frac{\hbar\omega}{4k_B T_c} G\left(\frac{\Delta}{\hbar\omega}\right) \right] = 0$$

$\alpha = 2e^2 D A_{\omega}^2 / \hbar$
 $\gamma = \hbar\tau^{-1}$
 where $D \propto l$ and $A_{\omega}^2 \propto B_{pk}^2$

[This term] represents the pair breaking by the time-averaged field. For small ω , this term dominates and the gap is depressed by the field.

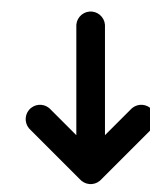
For $\hbar\omega \ll 2\Delta$: $G(\Delta/\hbar\omega) = 2(\hbar\omega/\Delta)[\ln(8\Delta/\hbar\omega) - 1]$

Van Son, P. C., et al, *Physical Review B* **29.3** (1984): 1503.

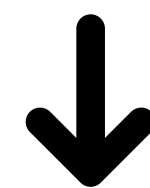
Model of Q-slope in thin film cavities:

The model is **specifically developed for describing Q-slope in niobium-coated copper cavities:**

- it is irrelevant to bulk niobium cavities
- it is a **non-thermal model**, unlike the model proposed by Palmieri and Vaglio

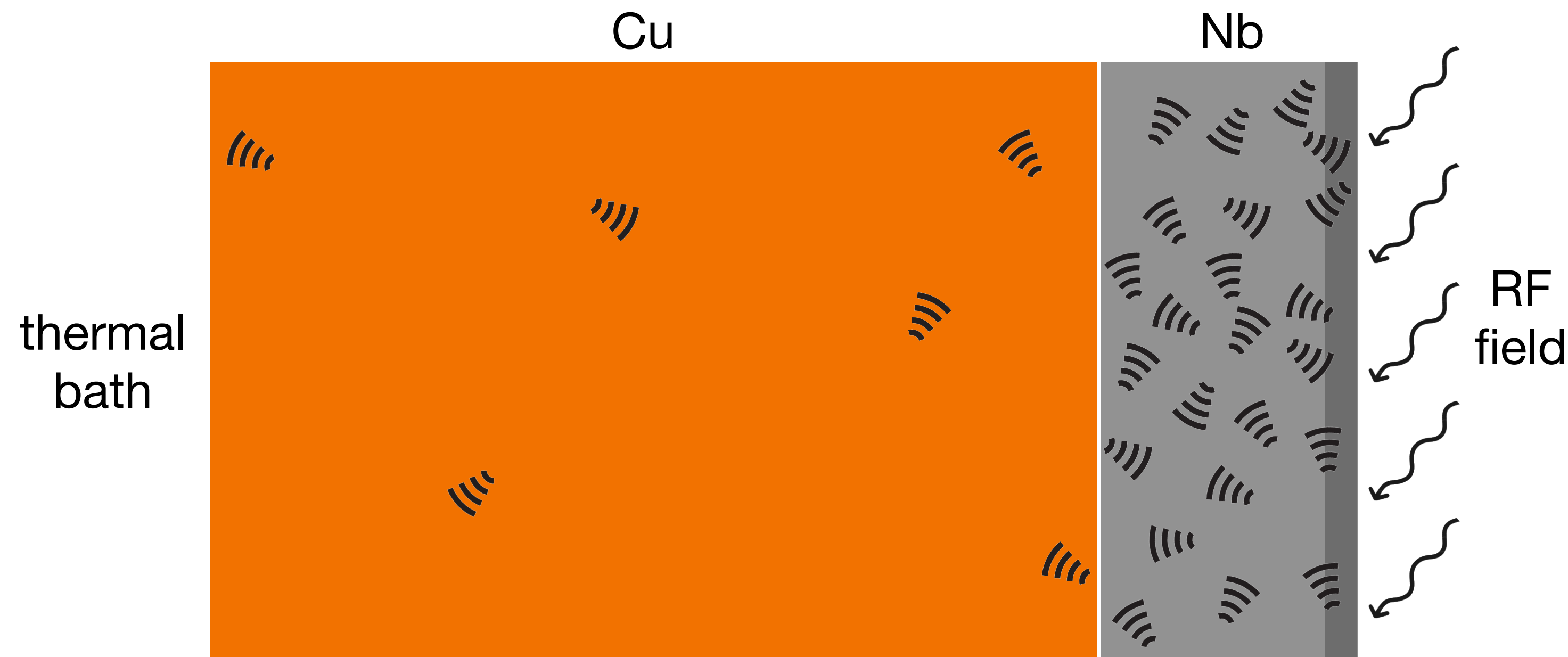


The core principle underlying the model is the **phonon escape mechanism**: this concept was originally developed in a different field and firstly introduced to the SRF community



Non-equilibrium phenomena at low frequencies are amplified by phonons confined within the superconducting film due to the **niobium-copper interface.**

Model of Q-slope in thin film cavities:



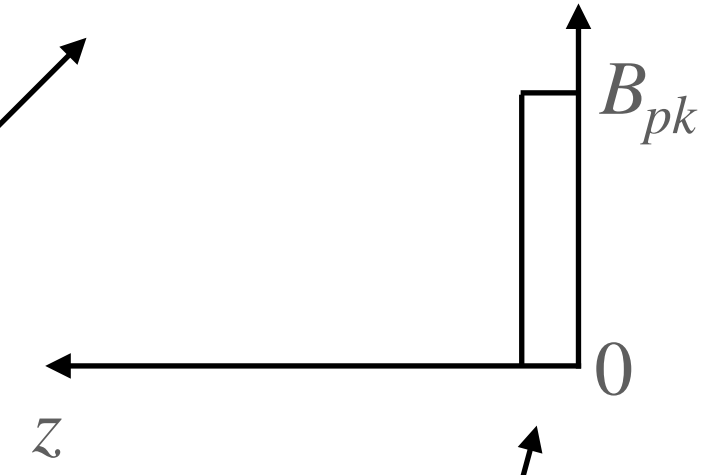
thermal bath

Cu

Nb

RF field

Phonons may remain largely confined in the superconducting film due to the **niobium-copper interface**



this is an assumption of the model

The magnitude of non-equilibrium effects may depend on the **phonon escape mechanism**



the longer it takes for the emitted phonons to leave the superconductor, the higher the probability of repeated interactions with quasiparticles

To describe the Q-slope of thin film cavities in the model, the role of phonons needs to be included in the energy gap equation of Eliashberg's theory.

Phonon escape mechanism

The more the phonon escape mechanism is limited, the greater the effective generation of excess quasiparticles as a result of repeated pair-breaking and recombination processes with phonons.

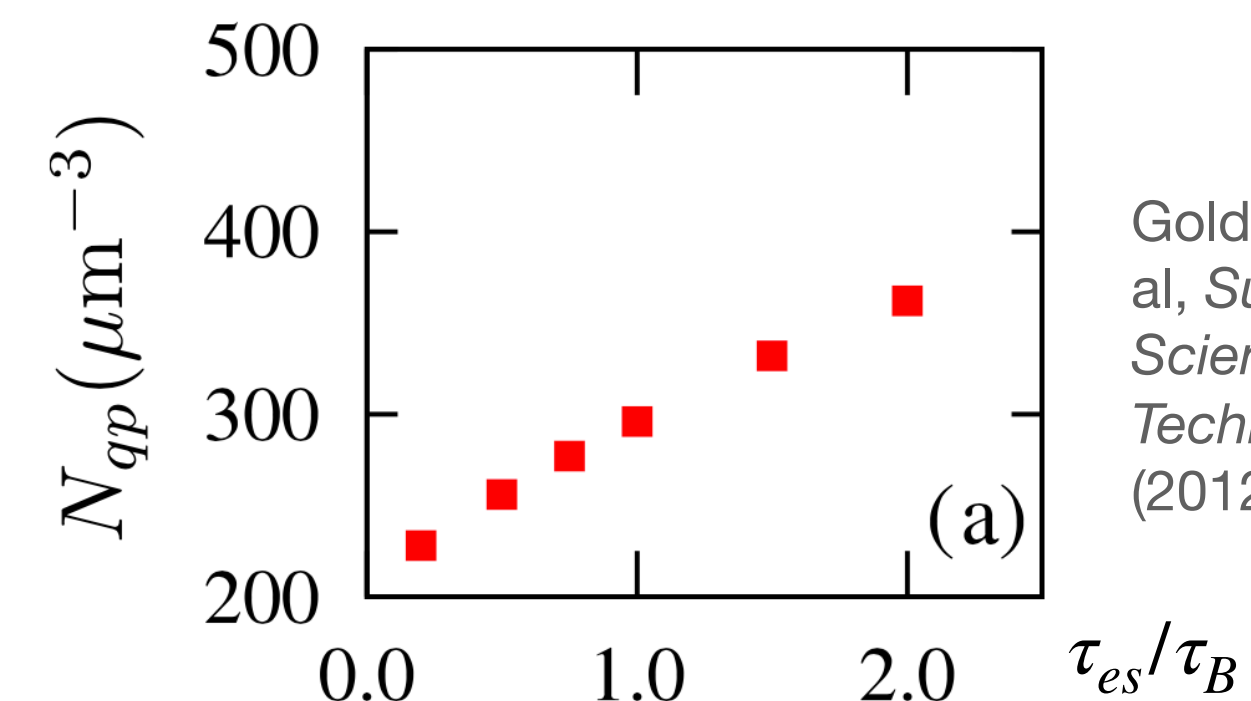
For the sake of simplicity, the phonon escape time τ_{es} is generally related to the phonon pair-breaking time τ_B :

- for $\tau_{es}/\tau_B \rightarrow 0$: pair-breaking phonons escape from the film without inducing further pair-breaking and recombination processes
→ the film is in good thermal contact with the substrate or thermal bath → the phonon distribution function in the film closely matches that in the substrate
 - for large values of τ_{es}/τ_B : weak phonon coupling between the film and the substrate
- **phonon trapping factor = $(1 + \tau_{es}/\tau_B)$: non-zero phonon trapping factor is expected due to the niobium-copper interface**

In the model, n_1 is assumed proportional to the phonon trapping factor. This is roughly in agreement with detailed calculations by Goldie and Withington.

“The phonon trapping effect can be minimized by optimizing the acoustic match between the superconducting film and the substrate”

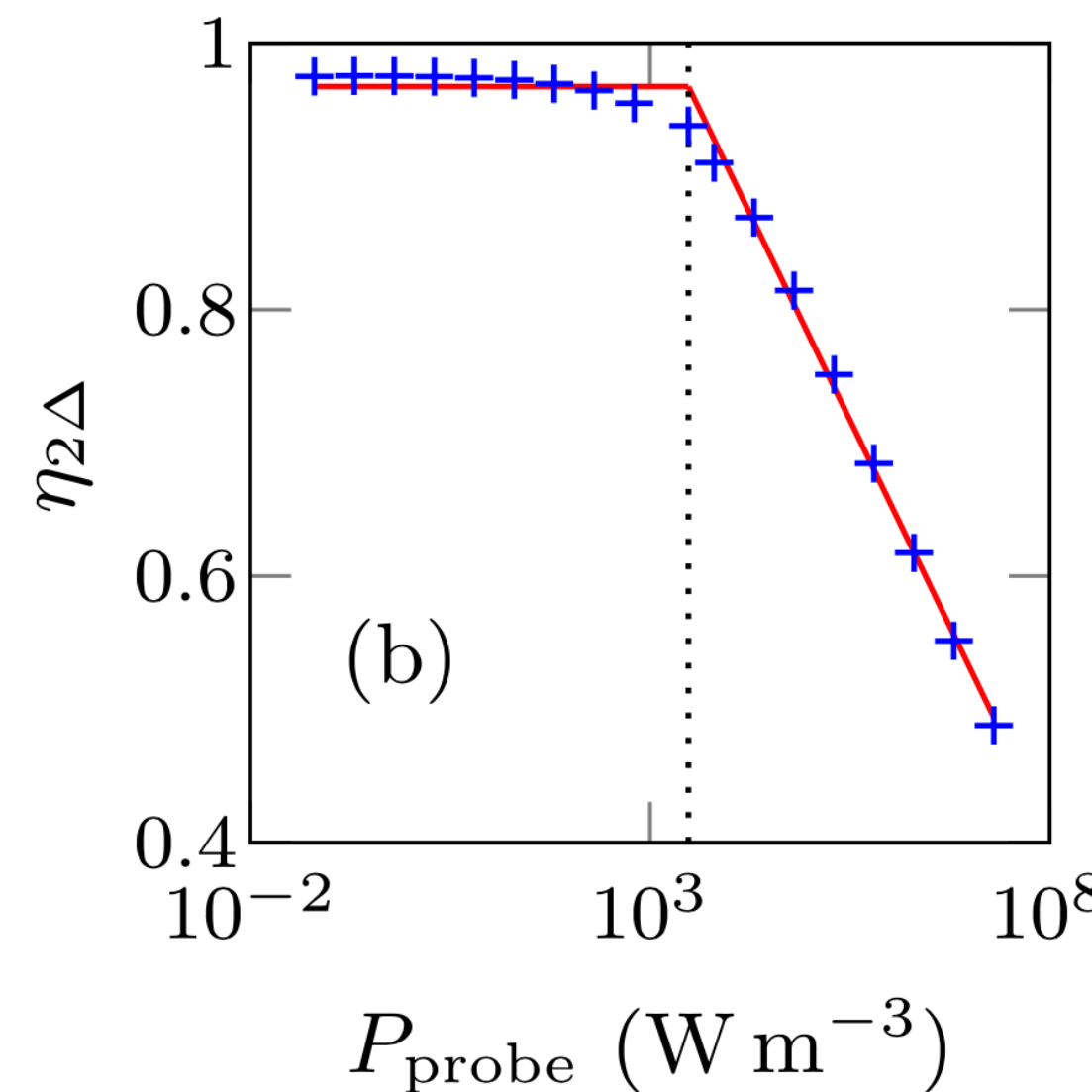
Kaplan, S. B, *Journal of Low Temperature Physics* **37.3** (1979): 343-365.



Goldie, D. J., et al, *Superconductor Science and Technology* **26.1** (2012): 015004.

Fraction of power carried by pair-breaking phonons

Guruswamy *et al.* calculated that the fraction of power transferred to the thermal bath by pair-breaking phonons remains approximately constant up to a certain threshold of absorbed power. Beyond this threshold, it decreases proportionally to $\log(P_{abs})$, as approximated in their cooling model for superconductors.



Based on these calculations, it is reasonable to expect that, in the model, **the fraction of power carried by pair-breaking phonons confined in the superconducting film contributes to the variation of the excess quasiparticle density n_1**

Guruswamy, T., et al, *Superconductor Science and Technology* **28.5** (2015): 054002.

Therefore, n_1 in the gap equation is scaled by the term $\eta_{2\Delta, B_{pk}} / \eta_{2\Delta, B_{pk} \rightarrow 0}$, which represents the fraction of power carried by pair-breaking phonons at a specific microwave field strength $\eta_{2\Delta, B_{pk}}$ normalized to its value at a field close to zero.

Revised gap equation of the model

Original energy gap equation in the Eliashberg's theory:

$$\frac{T_c - T}{T_c} - \frac{7\zeta(3)}{8\pi^2} \left(\frac{\Delta}{k_B T_c} \right)^2 + \underbrace{\frac{\alpha}{\gamma} \left[-\frac{\pi\gamma}{2k_B T_c} - 0.172 \left(\frac{\hbar\omega}{k_B T_c} \right)^2 + \frac{\hbar\omega}{4k_B T_c} G \left(\frac{\Delta}{\hbar\omega} \right) \right]}_{\text{non-equilibrium term } \propto n_1} = 0$$

Revised energy gap equation:

$$\frac{T_c - T}{T_c} - \frac{7\zeta(3)}{8\pi^2} \left(\frac{\Delta}{k_B T_c} \right)^2 + \left(1 + \frac{\tau_{es}}{\tau_B} \right) \times \frac{\eta_{2\Delta, B_{pk}}}{\eta_{2\Delta, B_{pk} \rightarrow 0}} \times \frac{\alpha}{\gamma} \left[-\frac{\pi\gamma}{2k_B T_c} - 0.172 \left(\frac{\hbar\omega}{k_B T_c} \right)^2 + \frac{\hbar\omega}{4k_B T_c} G \left(\frac{\Delta}{\hbar\omega} \right) \right] = 0$$

- If the phonon coupling between the film and its environment is optimum:

$$(1 + \tau_{es}/\tau_B) \rightarrow 1$$

- For low values of peak magnetic field:

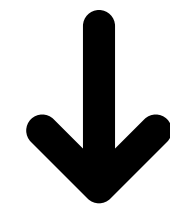
$$(\eta_{2\Delta, B_{pk}} / \eta_{2\Delta, B_{pk} \rightarrow 0}) \rightarrow 1$$

The gap equation has been modified based on **phenomenological considerations** to include the role of phonons. These considerations are inspired by the works of Chang-Scalapino and Guruswamy-Goldie-Withington

Revised gap equation of the model

Revised energy gap equation:

$$\frac{T_c - T}{T_c} - \frac{7\zeta(3)}{8\pi^2} \left(\frac{\Delta}{k_B T_c} \right)^2 + \left(1 + \frac{\tau_{es}}{\tau_B} \right) \times \frac{\eta_{2\Delta, B_{pk}}}{\eta_{2\Delta, B_{pk} \rightarrow 0}} \times \frac{\alpha}{\gamma} \left[-\frac{\pi\gamma}{2k_B T_c} - 0.172 \left(\frac{\hbar\omega}{k_B T_c} \right)^2 + \frac{\hbar\omega}{4k_B T_c} G \left(\frac{\Delta}{\hbar\omega} \right) \right] = 0$$



Solution of equation as a function of B_{pk} :

$$\Delta_{B_{pk}} / \Delta_{B_{pk} \rightarrow 0}$$

To (roughly) evaluate the **surface resistance** R_s :

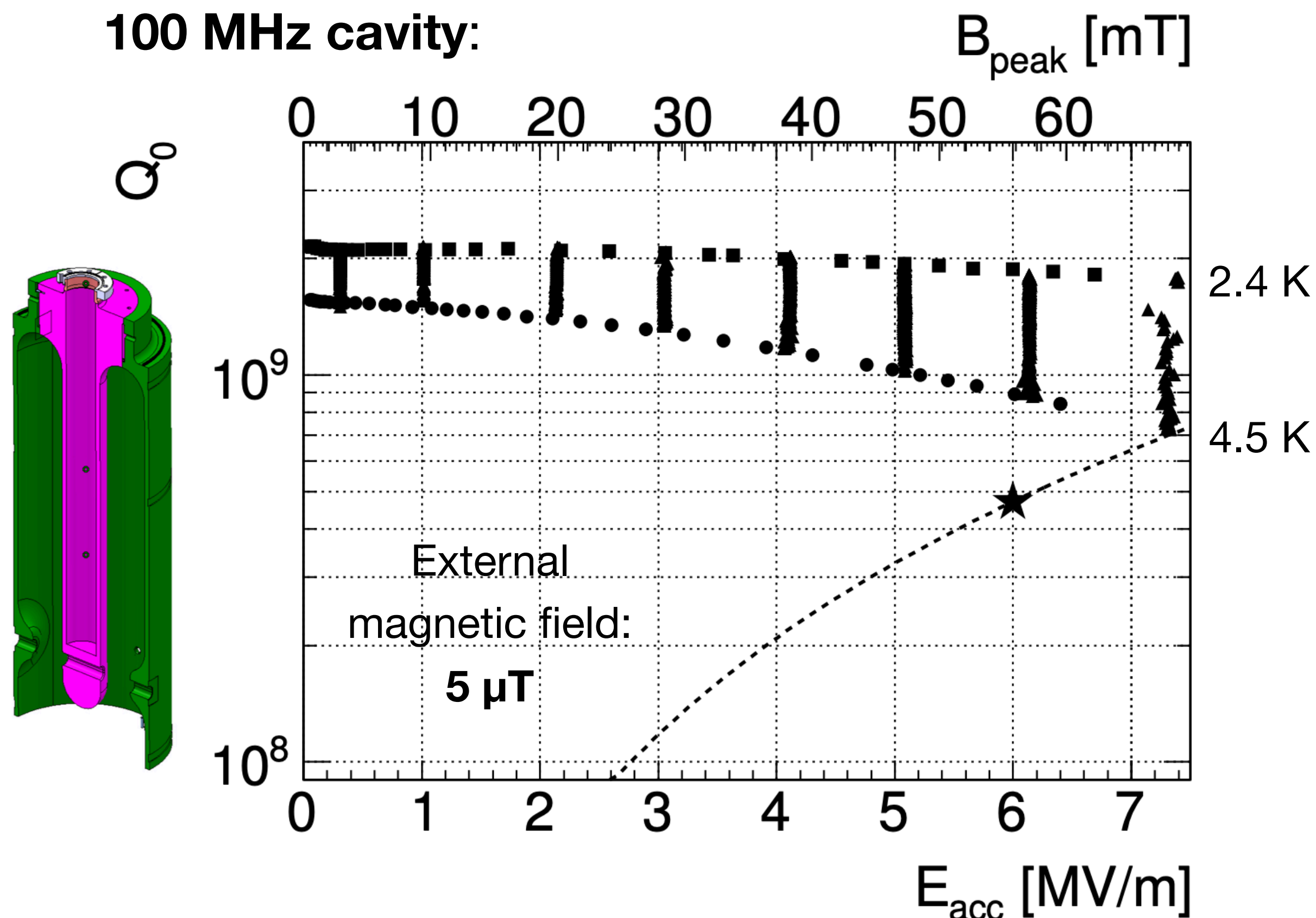
$$R_s = \frac{A_0}{T} \omega^\beta \exp \left(-\frac{\Delta_0}{k_B T} \right) + R_{res}$$

A_0 , Δ_0 and R_{res} are determined by interpolating the values of R_s as a function of T for $B_{pk} \rightarrow 0$

where Δ_0 is multiplied by $\Delta_{B_{pk}} / \Delta_{B_{pk} \rightarrow 0}$

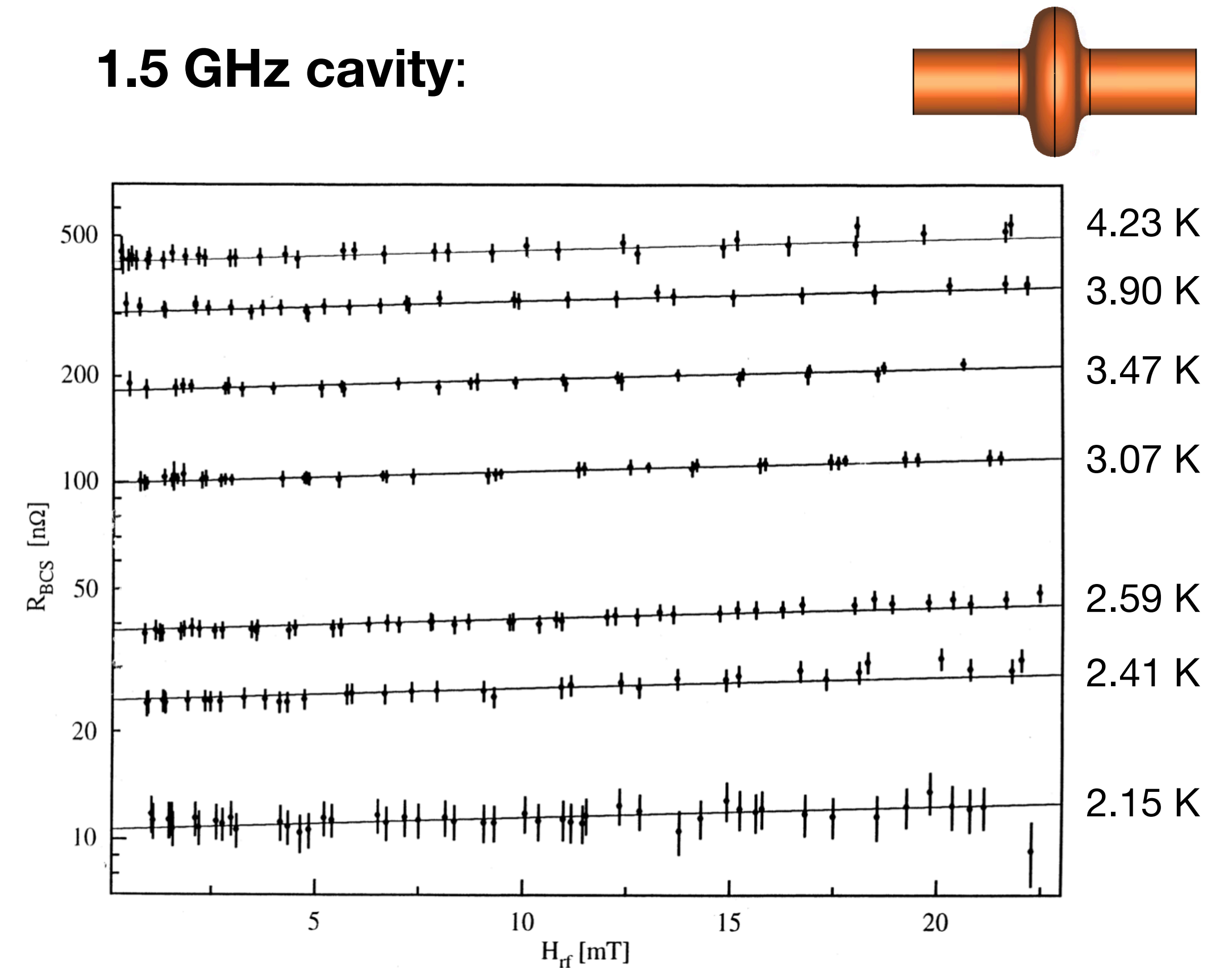
Dataset

The present study focuses on cavities where R_s increases proportionally to $(B_{pk})^2$. This is the only available dataset on cavities tested at various temperatures and for which the mean free path is known!



Miyazaki, A., et al, *Physical Review Accelerators and Beams* **22.7** (2019): 073101.

1.5 GHz cavity:

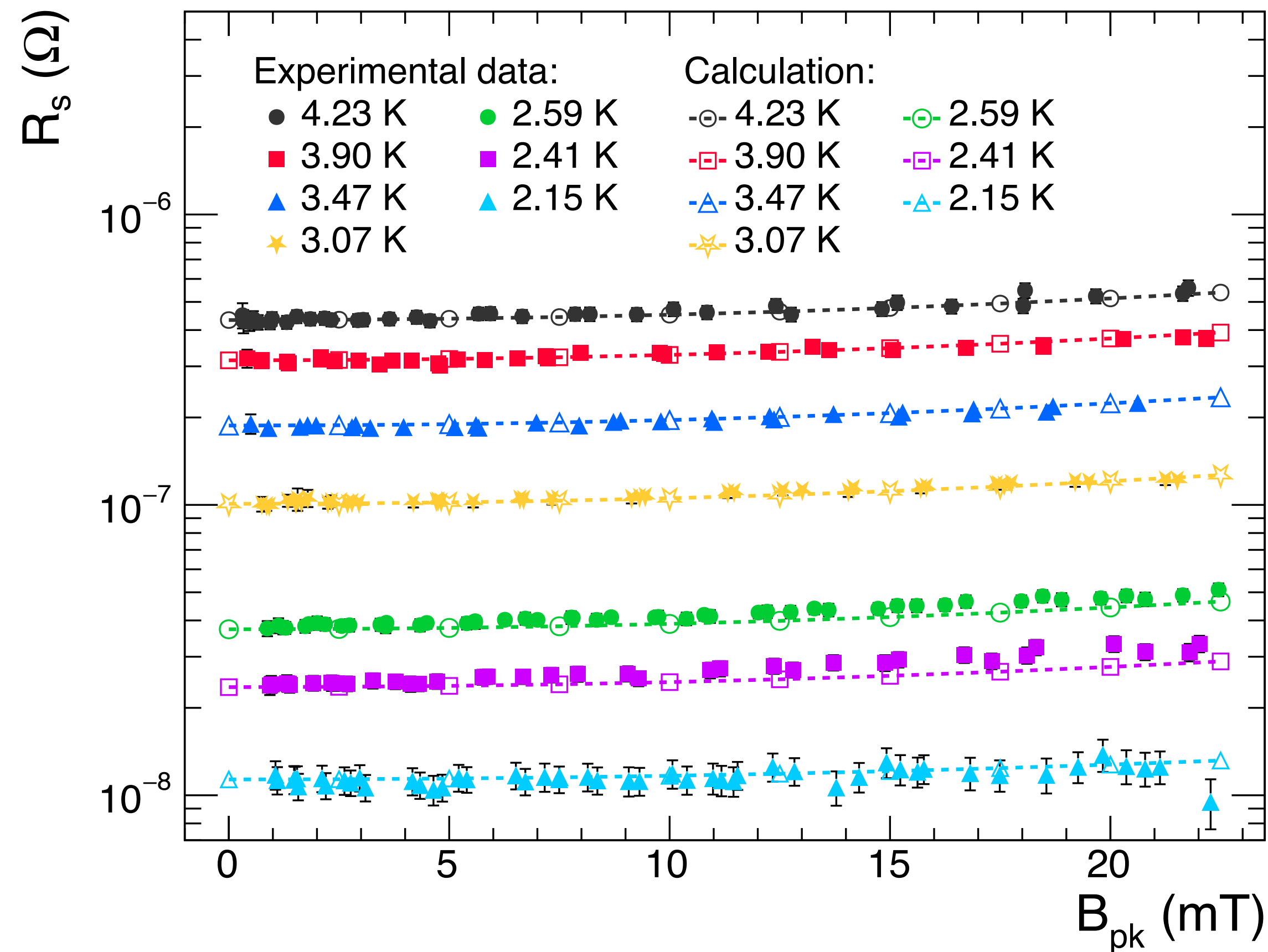


Benvenuti, C., et al. *Physica C: Superconductivity* **316.3-4** (1999): 153-188.

1.5 GHz Nb/Cu cavity

Mean free path:
27 nm

Value determined
experimentally



Data are corrected using
Longuevergne-Delavenne method

$$(1 + \tau_{es}/\tau_B) = 2.5$$

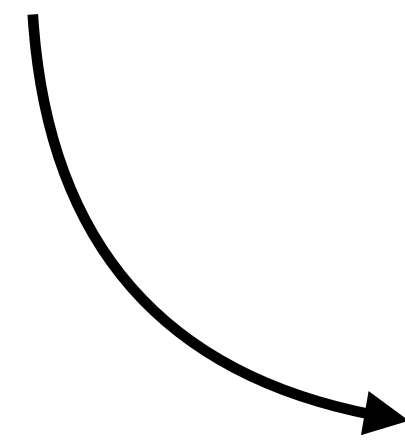
and

$$(\eta_{2\Delta, B_{pk}} / \eta_{2\Delta, B_{pk} \rightarrow 0}) = 1$$

Results of the model agree with the experimental data

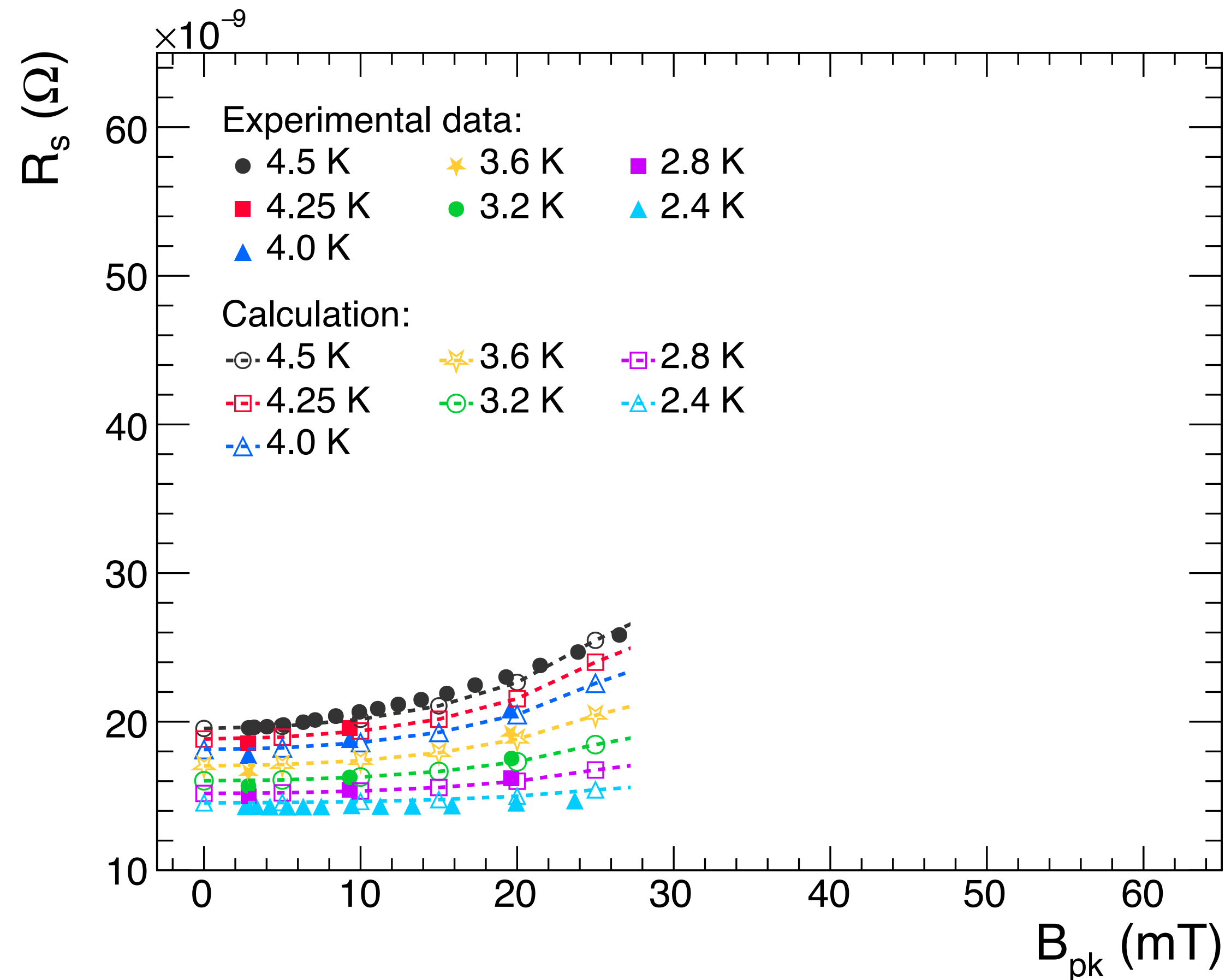
100 MHz Nb/Cu cavity

Mean free path:
100 nm

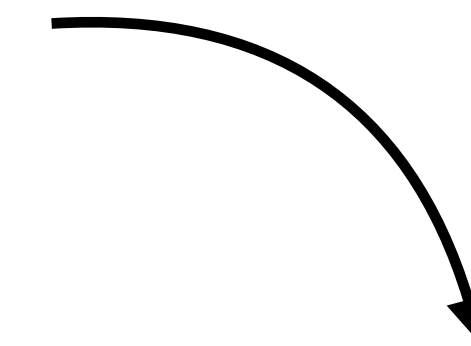


Value determined through a fitting procedure based on the BCS-Mattis-Bardeen theory to access material parameters and further confirmed by DC four-contact measurements

Miyazaki, A., et al, *Superconductor Science and Technology* **32.2** (2019): 025002.



Data are corrected using Longuevergne-Delavenne method



$$(1 + \tau_{es}/\tau_B) = 2.3$$

and

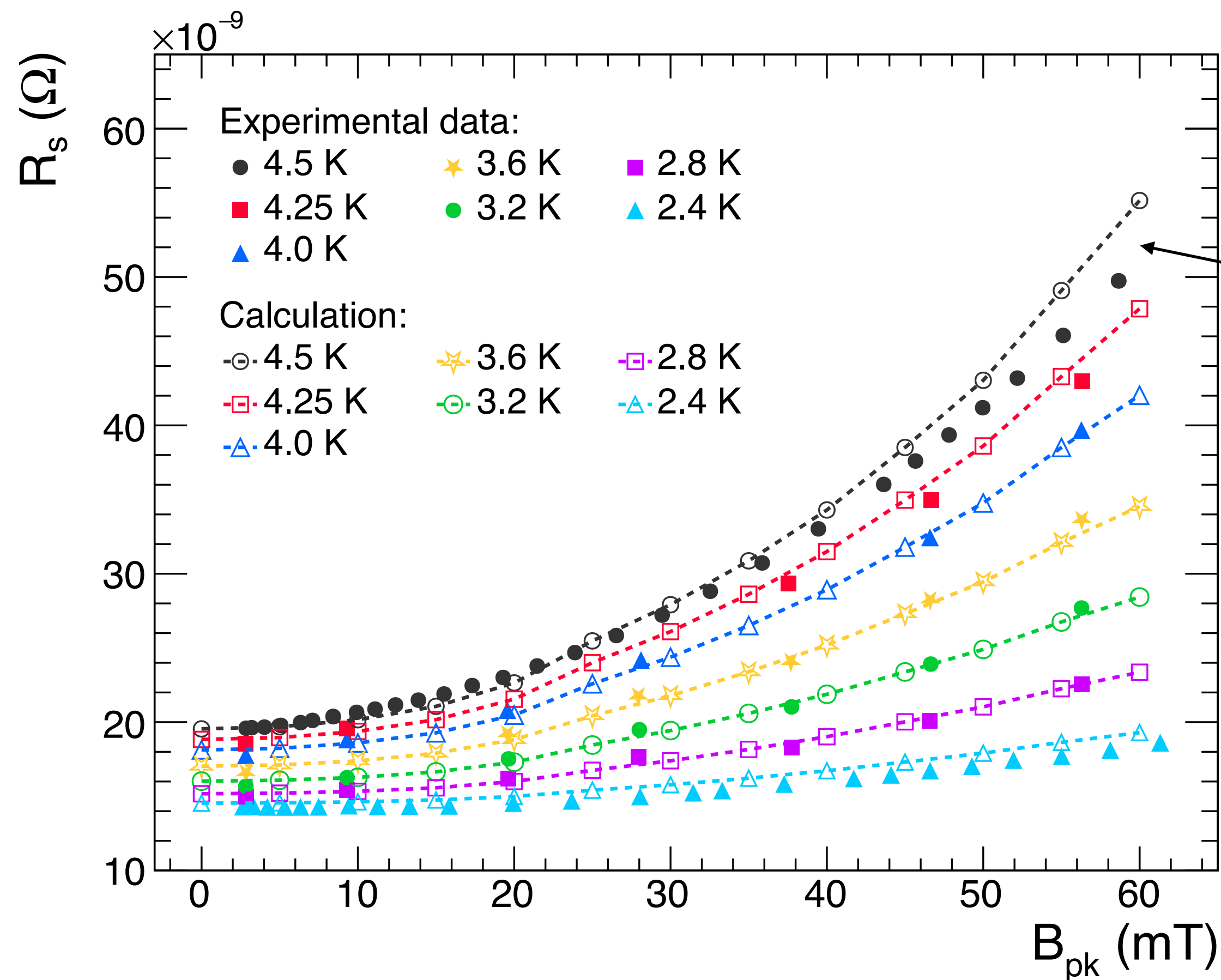
$$(\eta_{2\Delta, B_{pk}} / \eta_{2\Delta, B_{pk} \rightarrow 0}) = 1$$

100 MHz Nb/Cu cavity

Mean free path:
100 nm

Value determined through a fitting procedure based on the BCS-Mattis-Bardeen theory to access material parameters and further confirmed by DC four-contact measurements

Miyazaki, A., et al, *Superconductor Science and Technology* **32.2** (2019): 025002.



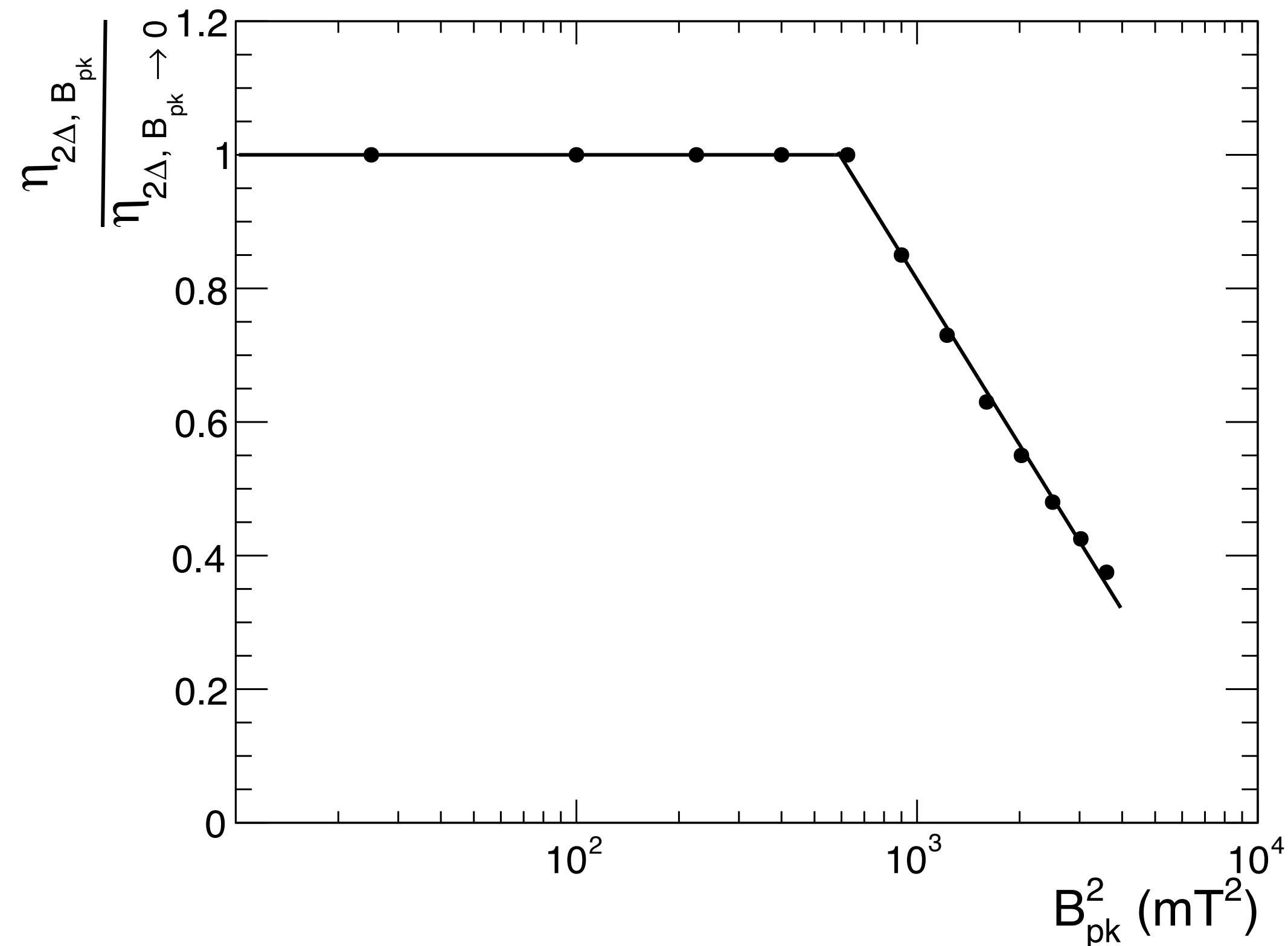
Data are corrected using Longuevergne-Delagen method

Maximum discrepancy: 10%

$$(1 + \tau_{es}/\tau_B) = 2.3$$

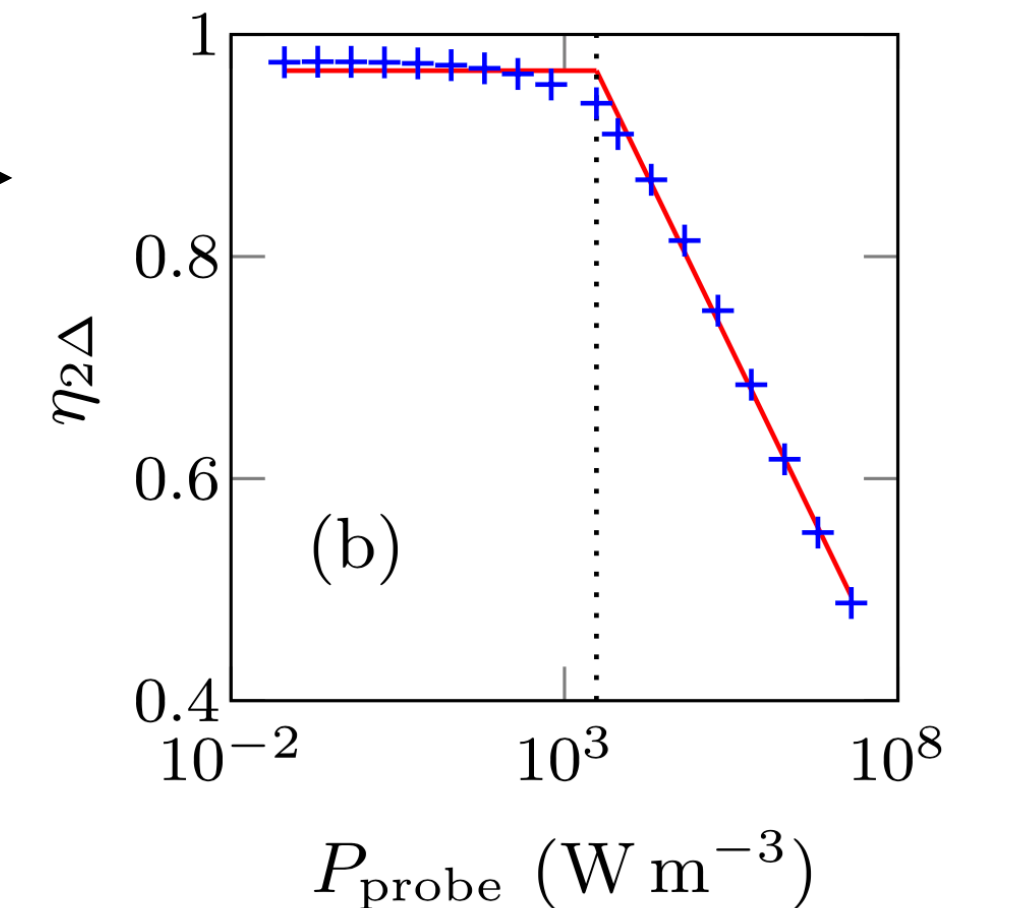
$(\eta_{2\Delta, B_{pk}} / \eta_{2\Delta, B_{pk} \rightarrow 0})$ remains constant at 1 until around 25-30 mT, after which a progressive decrease is required to achieve a satisfactory agreement with measurements

100 MHz Nb/Cu cavity



→

This trend closely resembles the fraction of phonon-bath power flow carried by pair-breaking phonons as a function of absorbed power, calculated by solving the kinetic equations for quasiparticle and phonon distributions

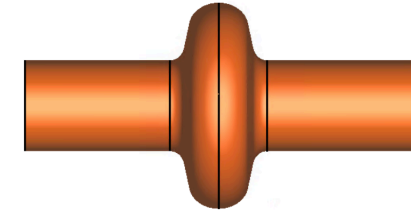


Guruswamy, T., et al,
Superconductor Science and Technology **28.5** (2015): 054002.

Since P_{abs} is proportional to $R_s B_{pk}^2$, the direct comparison of the two trends is justified because P_{abs} values in the 100 MHz cavity are mainly influenced by B_{pk}^2 , given the limited variation of R_s from 15 to 50 nΩ in the entire temperature range between 2.4 and 4.5 K.

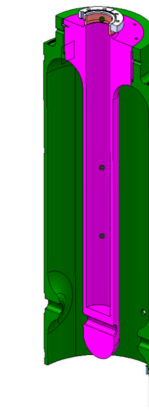
Comparison between 1.5 GHz and 100 MHz cavities

For the **1.5 GHz cavity**:

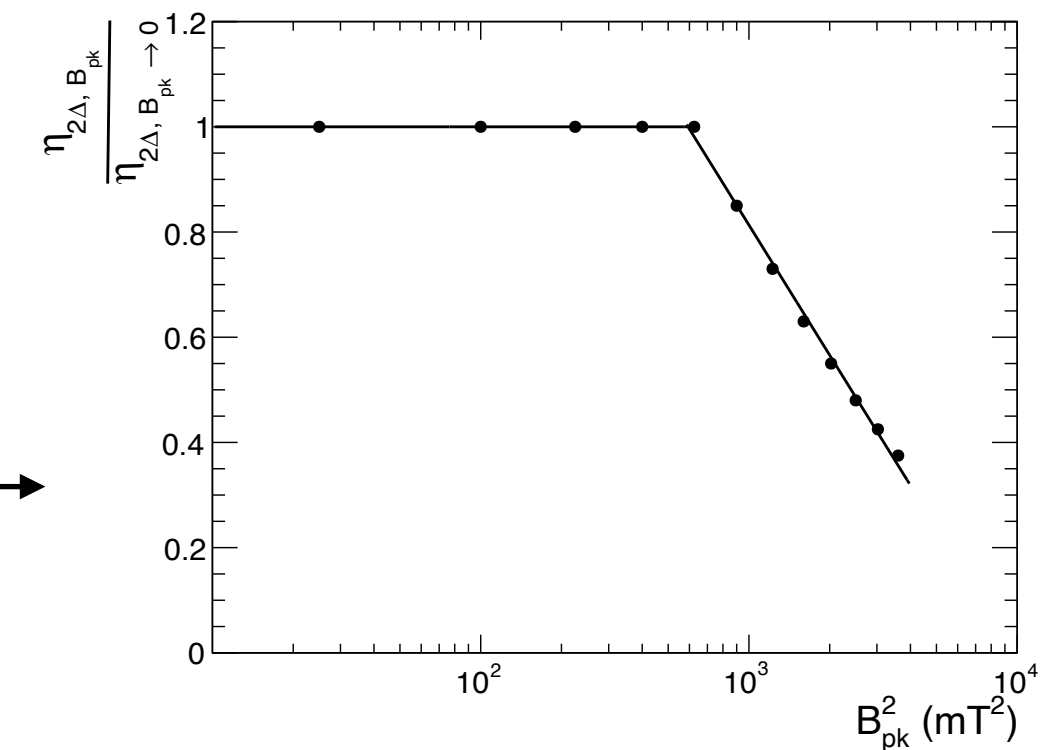


- $(1 + \tau_{es}/\tau_B) = 2.5$
- $(\eta_{2\Delta, B_{pk}} / \eta_{2\Delta, B_{pk} \rightarrow 0}) = 1$

For the **100 MHz cavity**:



- $(1 + \tau_{es}/\tau_B) = 2.3$
- $(\eta_{2\Delta, B_{pk}} / \eta_{2\Delta, B_{pk} \rightarrow 0})$: piecewise



→ The **phonon trapping factors for both cavities are in agreement**, as expected, since both have a **niobium-copper interface**.

→ The reason why the ratio $\eta_{2\Delta, B_{pk}} / \eta_{2\Delta, B_{pk} \rightarrow 0}$ does not vary for the 1.5 GHz cavity at high values of absorbed power is still **under investigation**:

The same piecewise trend of $\eta_{2\Delta, B_{pk}} / \eta_{2\Delta, B_{pk} \rightarrow 0}$ was also observed in a

bulk niobium cavity, assuming the **phonon trapping factor equal to 1** due to the absence of the niobium-copper interface.

Is it a universal behavior?

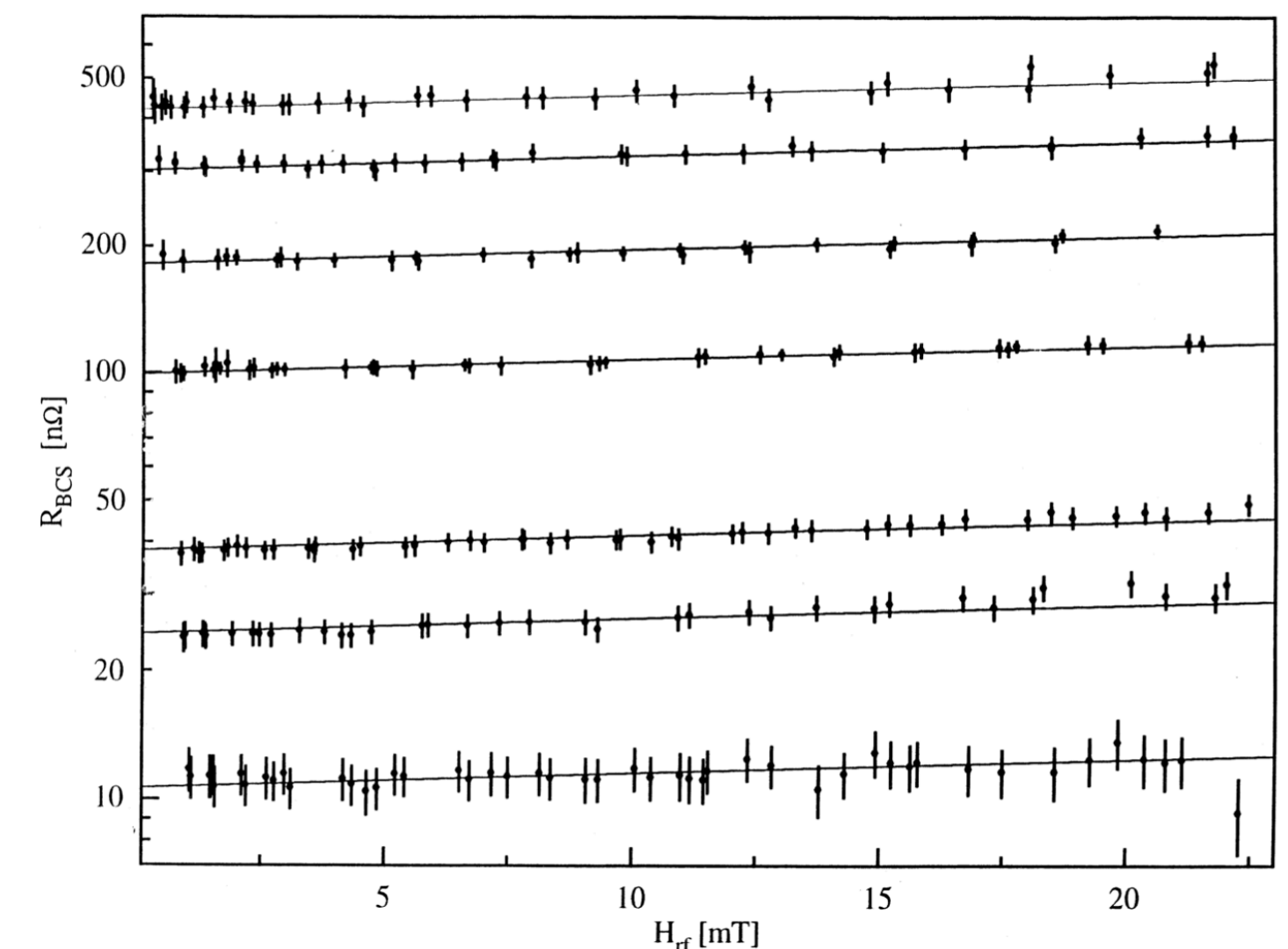
It does not seem to be film-specific.

Limitations of the model and future prospect

- Limitation 1: The gap equation in the Eliashberg's theory is only valid for temperatures close to T_c , so **it provides an approximate estimation for $T \ll T_c$**
 - More general calculation are needed by solving numerically the **kinetic equations** for quasiparticle and phonon distribution functions (**work in progress!**)
- Limitation 2: The validity of this argument at **high fields**
 - **Kinetic equations at high fields**; deviation from Guruswamy's calculations?
 - **Beyond the linear response to estimate surface resistance**
(outstanding challenge in fundamental condensed matter physics)

Suggestion to the community: conduct systematic measurements of **Q(B, T) and mean free path** similar to the datasets in this study

→ the measurements may be challenging, time-consuming, and expensive, but extremely valuable and useful!

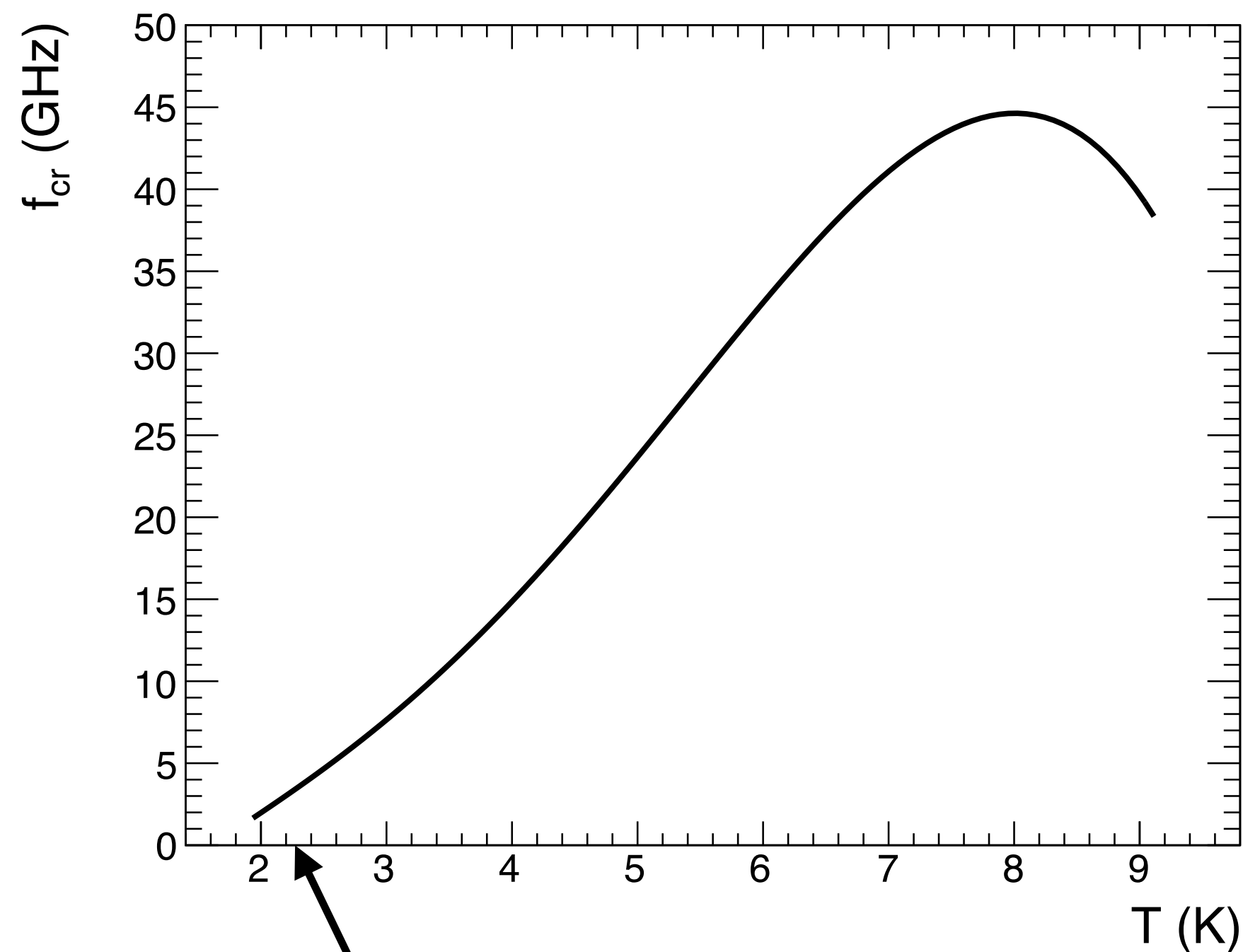


Benvenuti, C., et al. *Physica C: Superconductivity* **316.3-4** (1999): 153-188.

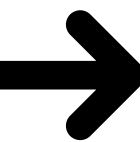
Crossover frequency

As pointed out by Eliashberg, there is a **minimum frequency threshold** beyond which the system enters a stationary non-equilibrium state. This frequency is usually called crossover frequency f_{cr}

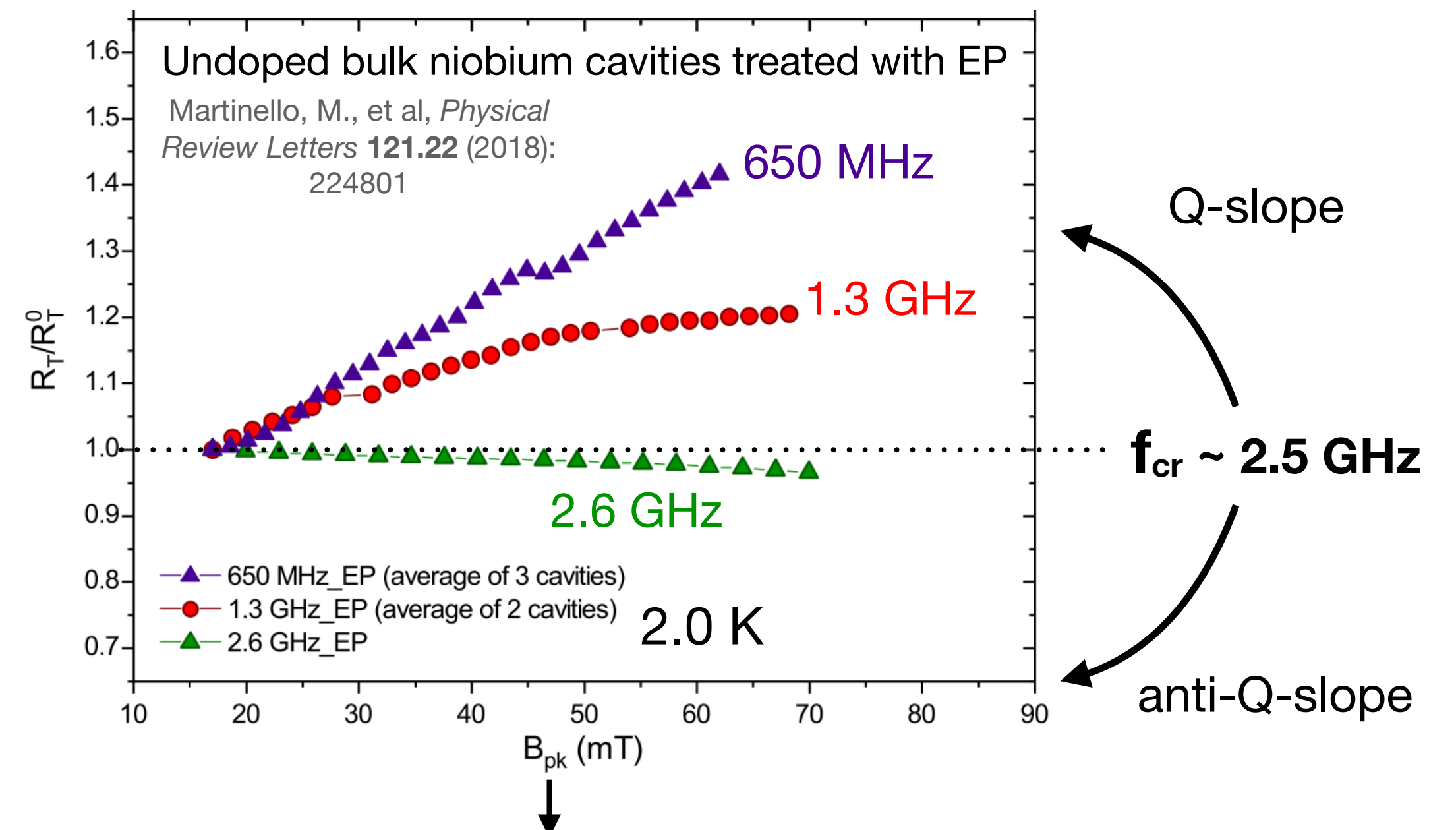
In niobium:



approximately **2.1 GHz at 2.0 K**

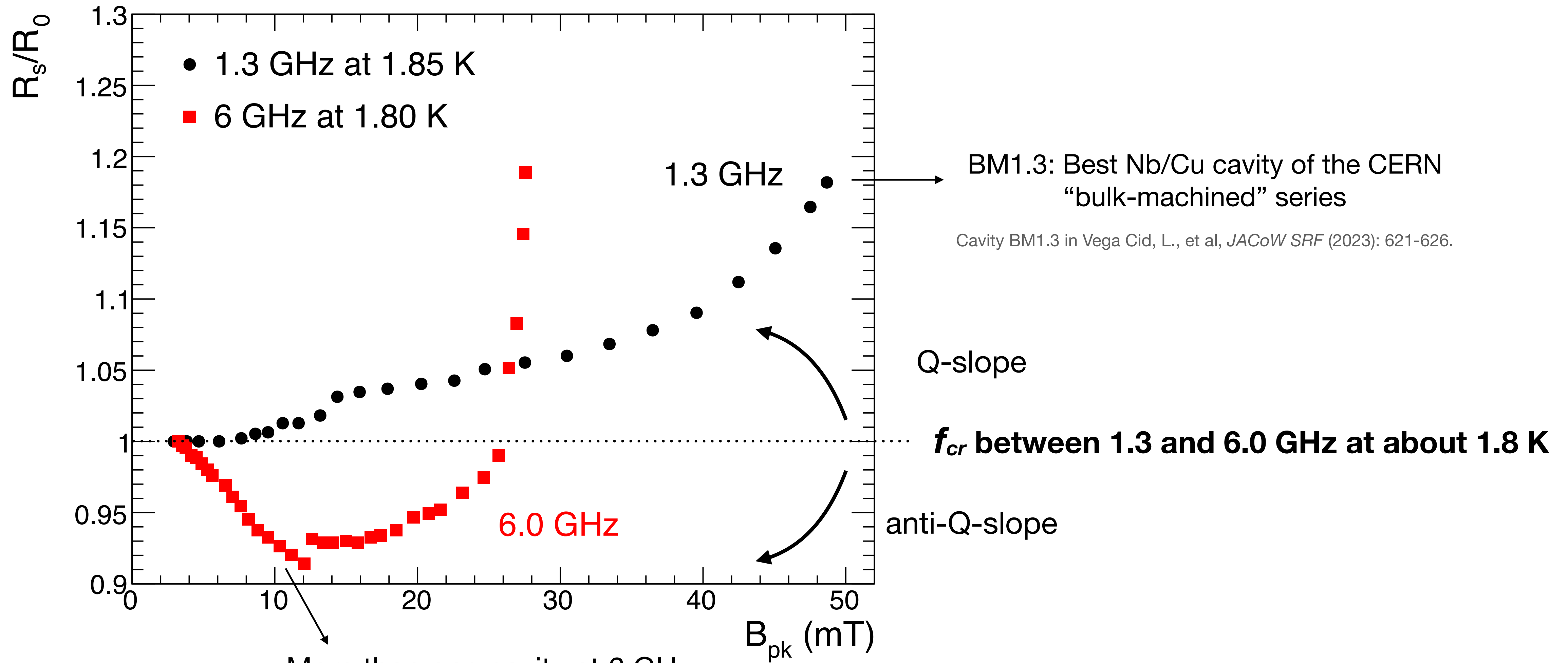


Even though the gap equation is derived for temperatures close to T_c , it seems possible to obtain fairly accurate predictions of crossover frequency at low temperatures



Differences in crossover frequencies may be related to variations in the material constant τ_0 , depending on the treatment or doping.

Crossover frequency in Nb/Cu cavities



More than one cavity at 6 GHz shows anti-Q-slope at 1.8 K!

Cavity Cav3,1 is shown in the graph: data in Pira, C. (2019), PhD thesis, University of Padova

Conclusions

- A new model that addresses the medium-field **Q-slope problem in niobium thin film cavities** is introduced:

- At low fields, the **phonon trapping factors for both cavities** are in agreement.
- At higher fields, a **progressive decrease of $\eta_{2\Delta, B_{pk}} / \eta_{2\Delta, B_{pk} \rightarrow 0}$** is required to achieve a satisfactory agreement with measurements
 → its trend is in substantial agreement with the fraction of phonon-bath power flow carried by pair-breaking phonons, **as expected.**

- Findings of this study are consistent with a scenario in which the medium-field **Q-slope problem in niobium thin film cavities arises from non-equilibrium superconductivity effects at low frequencies.** Nevertheless, **further theoretical and experimental studies are essential** to understand the Q-slope phenomenon in niobium thin film cavities.

