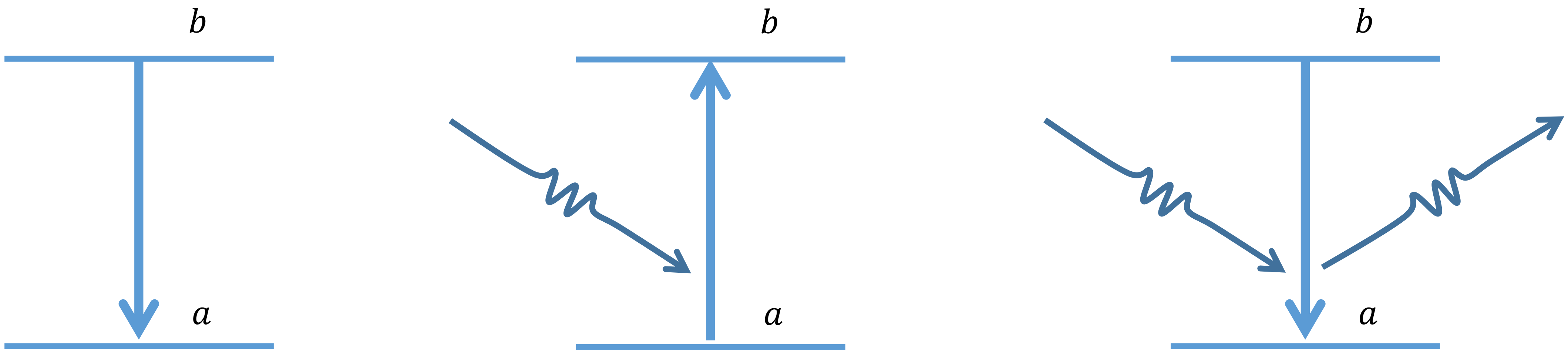
Interazione Radiazione materia (intro)
Sistema a due livelli e coefficienti di Einstein

Emissione Spontanea **Assorbimento** **<u>Emissione Stimolata</u>**



-Energia del fotone (<u>Emesso/Assorbito</u>) uguale alla differenza di energia tra i due livelli

-'Equazioni di bilanciamento' (Rate equation) delle popolazioni

-Condizioni di stazionarietà

-In condizioni di stazionarietà Einstein propone l'esistenza di un processo di <u>**emissione stimolata**</u> dal campo e.m.

-In cond. di equilibrio <u>**ottiene la legge di Plank**</u> (Nota: solo introducendo il processo di emissione stimolata!)

-LASER: <u>**L**</u>ight <u>**A**</u>mplification by <u>**S**</u>timulated <u>**E**</u>mission of <u>**R**</u>adiation

Interazione Radiazione materia

Approssimazione semiclassica: campi classici e atomi quantistici (no feedback dell'atomo sul campo)

L'Hamiltoniana di una particella in un campo e.m. è:

$$H = \frac{1}{2m}(\mathbf{p} - q\mathbf{A})^2 + q\phi$$

E l'eq. di Schroedinger diventa

$$i\hbar \frac{\partial}{\partial t}\Psi(\mathbf{r}, t) = \left[\frac{1}{2m}(-i\hbar\nabla + e\mathbf{A})^2 - \frac{Ze^2}{(4\pi\varepsilon_0)r}\right]\Psi(\mathbf{r}, t)$$

Nota: 'Eq. Di Schroedinger è invariante per la scelta di Gauge (la funzione d'onda acquisisce una fase).

Interazione di atomi ad 1e- con campi e.m.

La probabilità di transizione è data da:

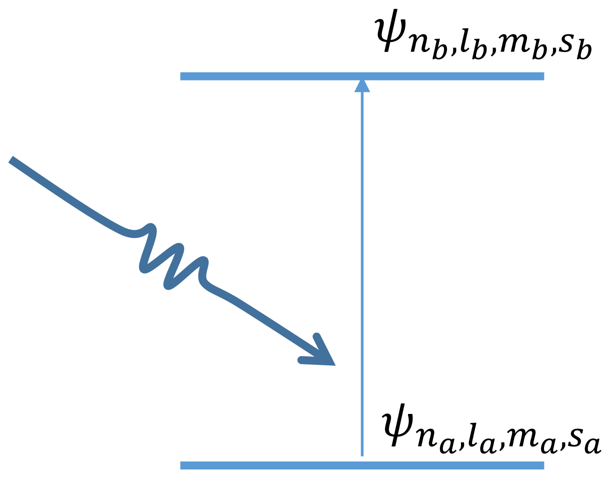
$$c_b^{(1)}(t) = -\frac{e}{m}\int_{\Delta\omega} \mathrm{d}\omega A_0(\omega)\left[\underbrace{e^{i\delta_\omega}\langle\psi_b|e^{i\mathbf{k}\cdot\mathbf{r}}\,\hat{\boldsymbol{\varepsilon}}\cdot\boldsymbol{\nabla}|\psi_a\rangle\int_0^t \mathrm{d}t' e^{i(\omega_{ba}-\omega)t'}} + \underbrace{e^{-i\delta_\omega}\langle\psi_b|e^{-i\mathbf{k}\cdot\mathbf{r}}\,\hat{\boldsymbol{\varepsilon}}\cdot\boldsymbol{\nabla}|\psi_a\rangle\int_0^t \mathrm{d}t' e^{i(\omega_{ba}+\omega)t'}}\right]$$

Assorbimento
≠0 se
$w_{ba}=w$;
i.e. $E_b=E_a+\hbar\omega$

Emissione
≠0 se
$w_{ba}=w$;
i.e. $E_b=E_a-\hbar\omega$

In un processo di **assorbimento** la probabilità che un atomo si trovi in uno stato viene calcolata in teoria delle perturbazioni dipendente dal tempo e prende la forma:

$$|c_b^{(1)}(t)|^2 \sim \int_{\Delta\omega} \mathrm{d}\omega \left[\frac{eA_0(\omega)}{m}\right]^2 |M_{ba}(\omega)|^2$$



Dove il ruolo cruciale è quello dell'**Elemento di Matrice**:

$$M_{ba} = \langle\psi_b|e^{i\mathbf{k}\cdot\mathbf{r}}\hat{\boldsymbol{\varepsilon}}\cdot\boldsymbol{\nabla}|\psi_a\rangle = \int \psi_b^\star(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}}\hat{\boldsymbol{\varepsilon}}\cdot\boldsymbol{\nabla}\psi_a(\mathbf{r})\,\mathrm{d}\mathbf{r}$$

Che ci permette di definire la **probabilità di transizione di assorbimento** $$W_{ba} = \frac{4\pi^2}{m^2 c}\left(\frac{e^2}{4\pi\varepsilon_0}\right)\frac{I(\omega_{ba})}{\omega_{ba}^2}|M_{ba}(\omega_{ba})|^2$$

Legata alla sua **sezione d'urto** $$\sigma_{ba} = \frac{4\pi^2\alpha\hbar^2}{m^2\omega_{ba}}|M_{ba}(\omega_{ba})|^2$$

Oltre al processo di **Assorbimento** dal campo e.m.

Il processo in cui un fotone viene emesso da un atomo in un processo di diseccitazione è chiamato
<u>Emissione</u>

<u>Emissione Stimolata</u> è un processo per cui **la presenza di un campo e.m.** (risonante all'energia di transizione) stimola la diseccitazione di un atomo e di conseguenza l'emissione di un fotone

Al contrario il processo di **<u>Emissione Spontanea</u>** è un processo di decadimento spontaneo dello stato eccitato di un atomo che avviene in assenza di fotoni nel campo di stimolo;

$$\mathbf{A}_2 = \hat{\boldsymbol{\varepsilon}}\left[\frac{(N(\omega)+1)\hbar}{2V\varepsilon_0\omega}\right]^{1/2} e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega t+\delta_\omega)}$$

Nota: emissione spontanea è un processo di **emissione «stimolato» dal campo elettromagnetico di vuoto**. In formalismo quantistico esistono fluttuazioni di campo e.m. anche in assenza di eccitazioni (fotoni) in tale campo.
Effetto Purcell: probabilità di emissione spontanea degli atomi cambiano quando gli atomi sono incorporati in una cavità risonante

Quantizzazione del campo elettromagnetico

a) Troviamo l'Hamiltoniana H(A(r,t))

$$A_{\mathbf{k}\lambda}(\mathbf{r},t) = A_{\mathbf{k}\lambda}\exp(-\mathrm{i}\omega_k t + \mathrm{i}\mathbf{k}.\mathbf{r}) + A^*_{\mathbf{k}\lambda}\exp(\mathrm{i}\omega_k t - \mathrm{i}\mathbf{k}.\mathbf{r}) \quad \Rightarrow \quad \mathcal{E}_{\mathbf{k}\lambda} = \varepsilon_0 V\omega_k^2\left(A_{\mathbf{k}\lambda}A^*_{\mathbf{k}\lambda} + A^*_{\mathbf{k}\lambda}A_{\mathbf{k}\lambda}\right)$$

b) Troviamo le variabili canoniche Q, P

$$\hat{a} = (2m\hbar\omega)^{-1/2}(m\omega\hat{q} + \mathrm{i}\hat{p})$$

$$\hat{a}^\dagger = (2m\hbar\omega)^{-1/2}(m\omega\hat{q} - \mathrm{i}\hat{p})$$

c) Promuoviamo ad operatori le variabili canoniche e imoniamo le regole di commutazione

$$A_{\mathbf{k}\lambda} \to \left(\hbar/2\varepsilon_0 V\omega_k\right)^{1/2}\hat{a}_{\mathbf{k}\lambda} \qquad A^*_{\mathbf{k}\lambda} \to \left(\hbar/2\varepsilon_0 V\omega_k\right)^{1/2}\hat{a}^\dagger_{\mathbf{k}\lambda}$$

$$[\hat{q},\hat{p}] = \mathrm{i}\hbar \qquad \Leftrightarrow \qquad \left[\hat{a},\hat{a}^\dagger\right] = 1$$

Hamiltoniana del campo elettromagnetico quantizzato

$$\mathcal{E}_{\mathbf{k}\lambda} = \varepsilon_0 V\omega_k^2\left(A_{\mathbf{k}\lambda}A^*_{\mathbf{k}\lambda} + A^*_{\mathbf{k}\lambda}A_{\mathbf{k}\lambda}\right) \quad \text{-->} \quad \hat{\mathcal{H}} = \hbar\omega\left(\hat{a}^\dagger\hat{a} + \tfrac{1}{2}\right)$$

Operatori di creazione e distruzione

$$\left[\hat{a},\hat{a}^{\dagger}\right]= \quad 1$$

a manda un autostato |n> in un autostato |n-1> con autovalore accresciuto o diminuito di $\hbar\omega$

$$\hat{a}|n\rangle = n^{1/2}|n-1\rangle \qquad \hat{a}^{\dagger}|n\rangle = (n+1)^{1/2}|n+1\rangle$$

Energia del Fotone: Variazione quantizzata dell'autovalore di energia

Stati numero della radiazione

Autostati di n
Risultato dell'azione ripetuta di operatori di creazione e distruzione

Stati Coerenti della radiazione

$$|\alpha\rangle = \exp\left(-\tfrac{1}{2}|\alpha|^2\right)\sum_{n=0}^{\infty}\frac{\alpha^n}{(n!)^{1/2}}|n\rangle$$

Numero di fotone indefinito
Campo elettrico classico
Fluttuazioni di vuoto

L’approssimazione di dipolo

Il calcolo dell’elemento di matrice $M_{ba} = \langle \psi_b | e^{i\mathbf{k}\cdot\mathbf{r}} \hat{\boldsymbol{\varepsilon}} \cdot \boldsymbol{\nabla} | \psi_a \rangle$ può essere semplificato

$$e^{i\mathbf{k}\cdot\mathbf{r}} = 1 + (i\mathbf{k} \cdot \mathbf{r}) + \frac{1}{2!}(i\mathbf{k} \cdot \mathbf{r})^2 + \cdots = 1$$

L’approx. corrisponde ad assumere che il campo sia uniforme sulle dimensioni atomiche; i.e. Campi ottici (10^{-6}m) e dimensioni atomiche 10^{-10}m

L’espressione per una probabilità di transizione si può quindi esprimere in funzione di $\mathbf{r}_{ba} = \langle \psi_b | \mathbf{r} | \psi_a \rangle$ (dove abbiamo espresso il nabla attraverso la sua equazione del moto al primo ordine):

$$W_{ba} = \frac{4\pi^2}{c\hbar^2}\left(\frac{e^2}{4\pi\varepsilon_0}\right) I(\omega_{ba}) |\hat{\boldsymbol{\varepsilon}} \cdot \mathbf{r}_{ba}|^2$$

Che possiamo esprimere introducendo l’operatore di momento di dipolo elettrico:

$$\mathbf{D}_{ba} = -e\mathbf{r}_{ba}$$

Il calcolo delle probabilità di transizione si riduce a calcolare la precedente o più in generale (q indice delle componenti sferiche).

$$I^{q}_{n'l'm';nlm} = \left(\frac{4\pi}{3}\right)^{1/2} \int_0^\infty \mathrm{d}r\, r^3 R_{n'l'}(r) R_{nl}(r) \times \int \mathrm{d}\Omega\, Y^{*}_{l'm'}(\theta, \phi) Y_{1,q}(\theta, \phi) Y_{lm}(\theta, \phi)$$

La componente radiale della funzione d’onda è non nulla, mentre la componente angolare ci da delle **<u>Regole di selezione di dipolo</u>**:

$$\Delta l = \pm 1$$

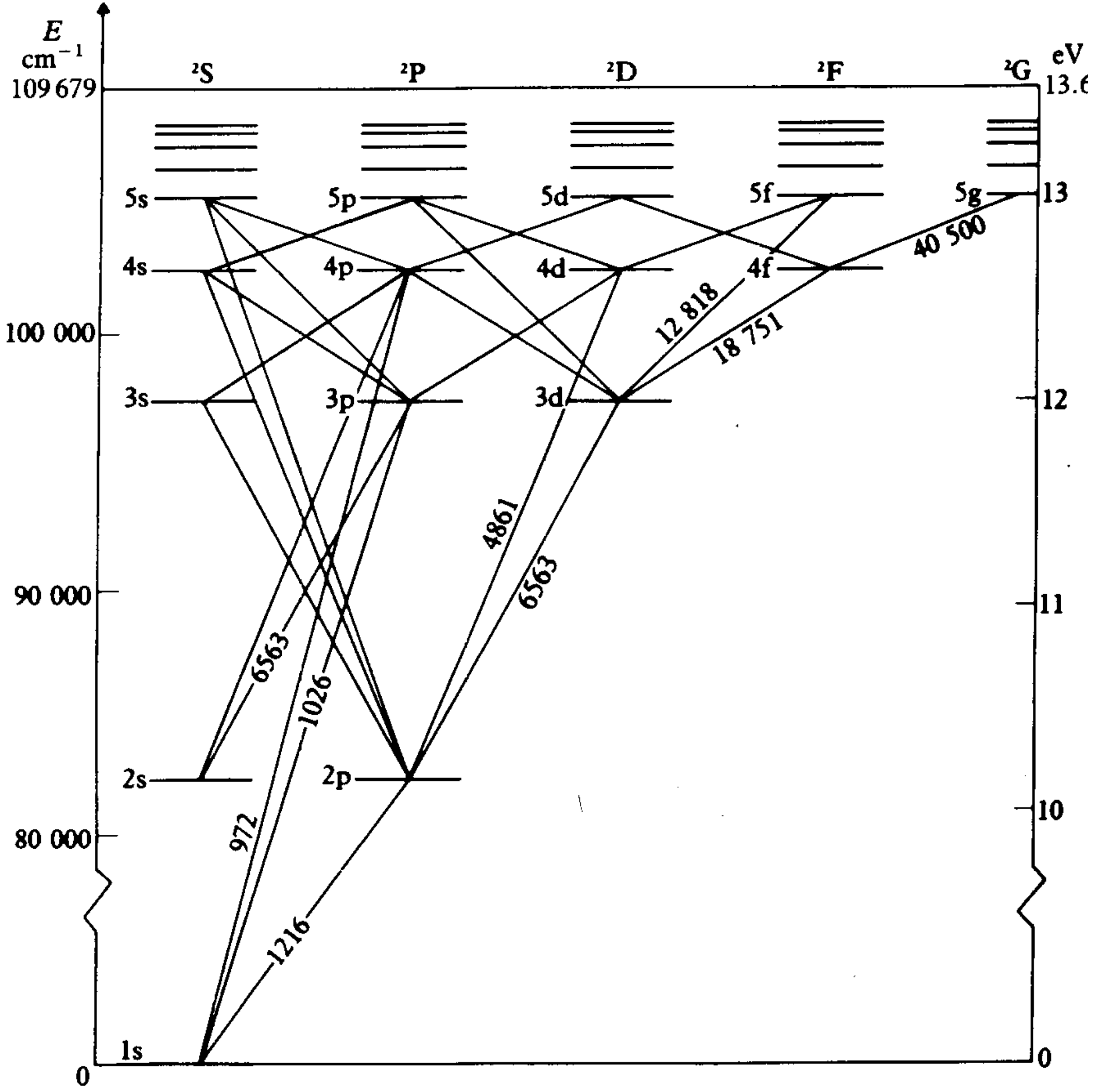
$$\Delta m = 0, \pm 1$$

Regola generale: vale per transizione tra stati elettronici

-**Stati vibrazionali** (IR and Raman activities)

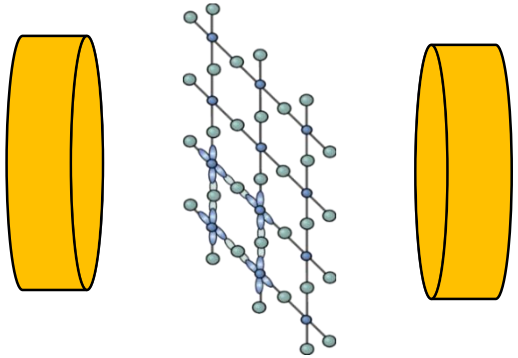
O—C—O	O—C—O
Simmetrico No dipolo	Dipolo (IR)

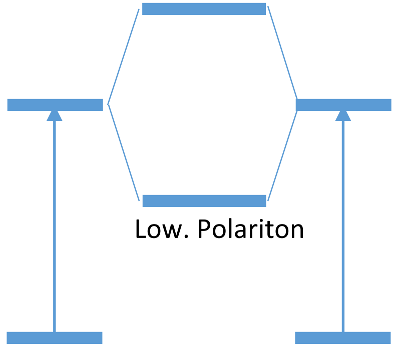
-**Magnetici** (Magnon absorption)



Detou «ExCursus»:
Can we use cavity electrodynamics to control complex materials?

Solid state samples in tunable cryogenic Fabry-Perot





-Materials embedded in tunable cryogenic optical cavities

Rev. Sci. Instr. 93, 033102 (2022)

* With Sample Limited by sample thickness (5-100μm)

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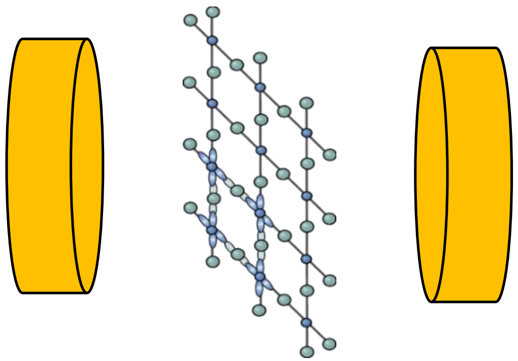
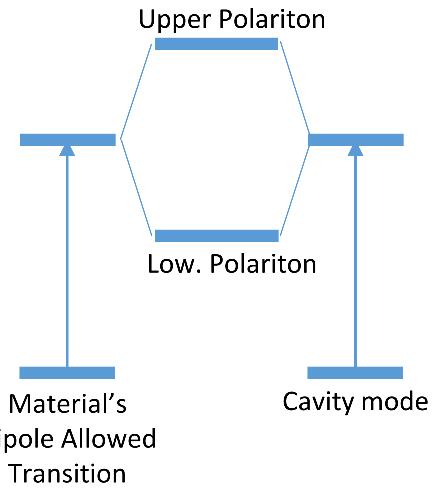
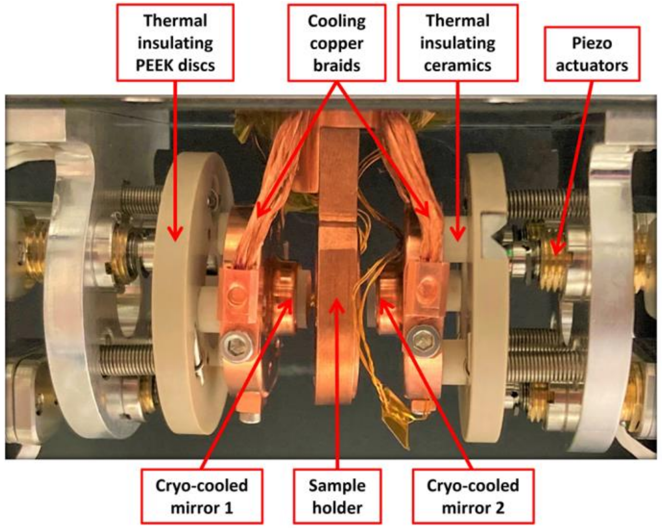
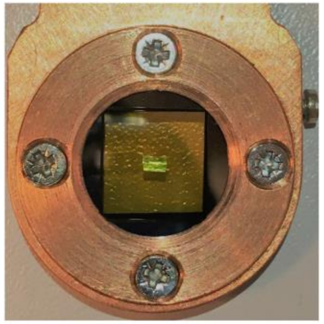


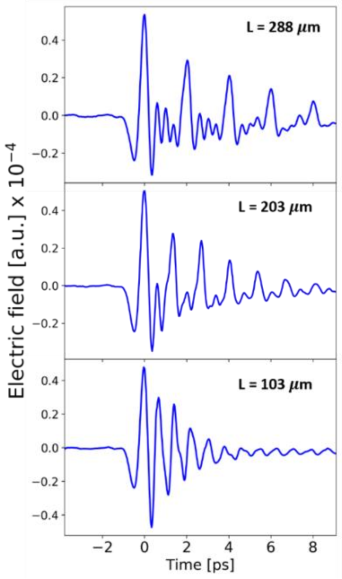


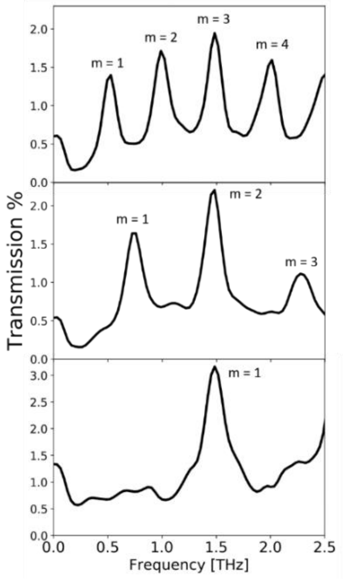
-Materials embedded in tunable cryogenic optical cavities

-Tuneable requency 0.01THz-100Thz*
-Quality factor 2<Q<10









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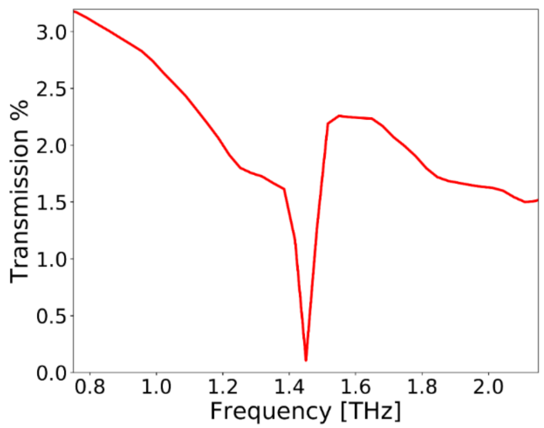
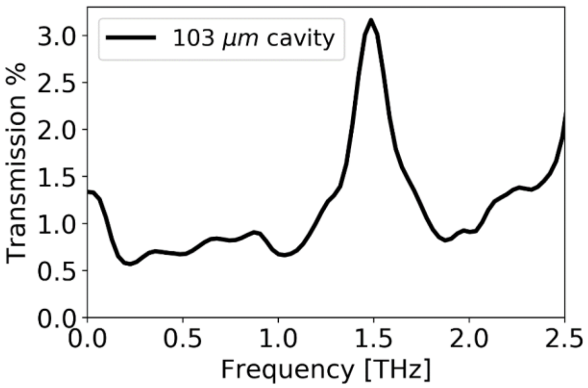
Solid state samples in tunable cryogenic Fabry-Perot

THz absorption of CuGeO3





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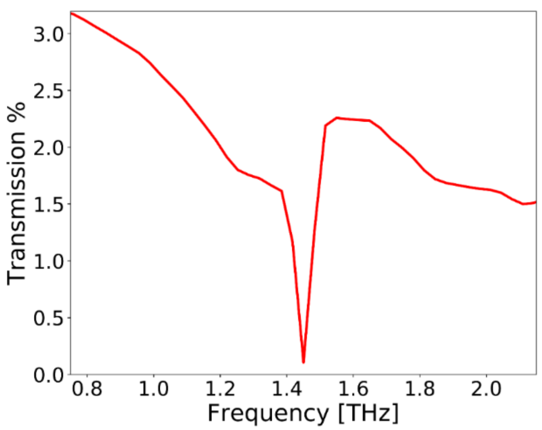

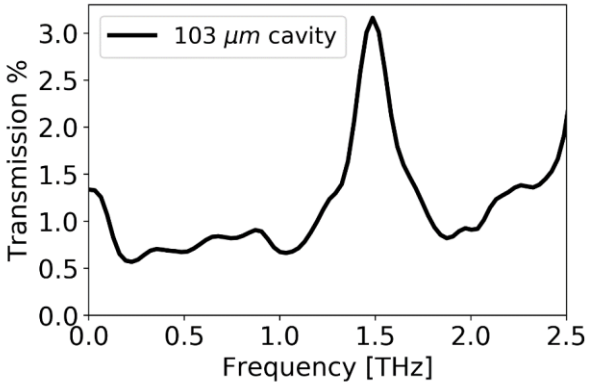

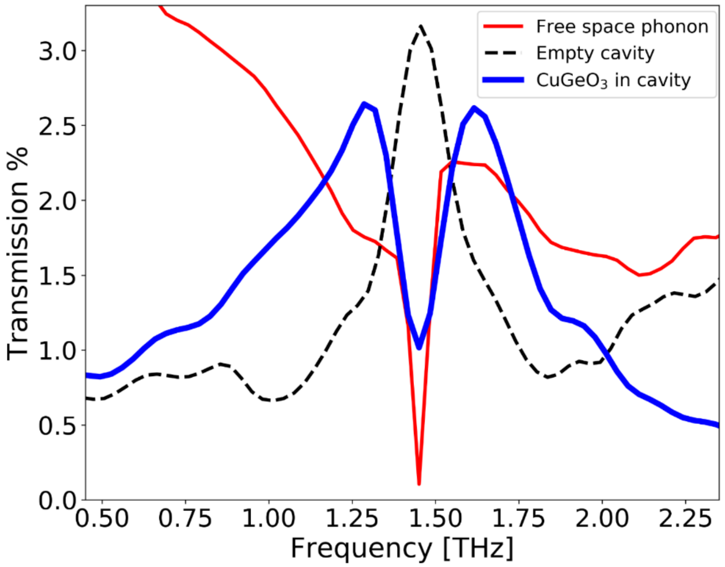

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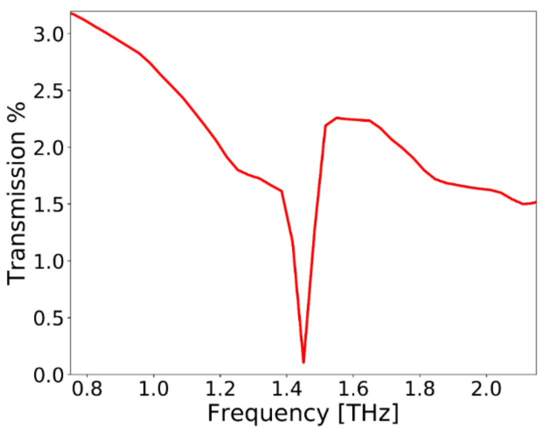

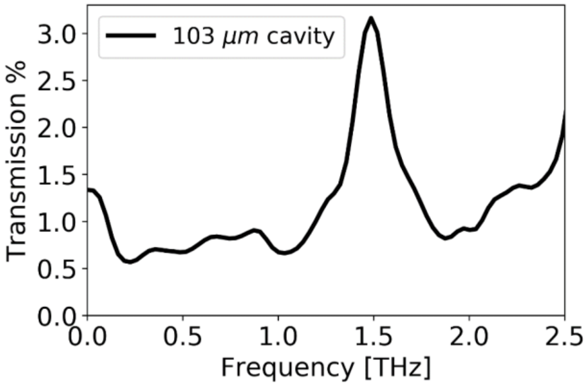

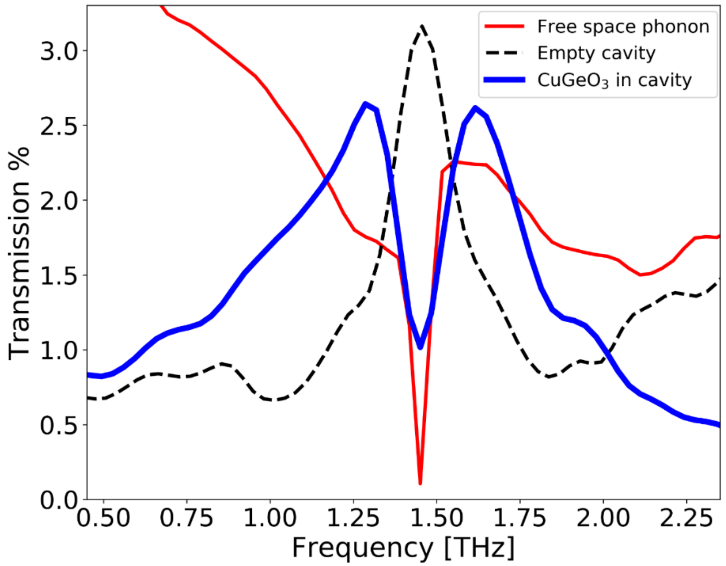

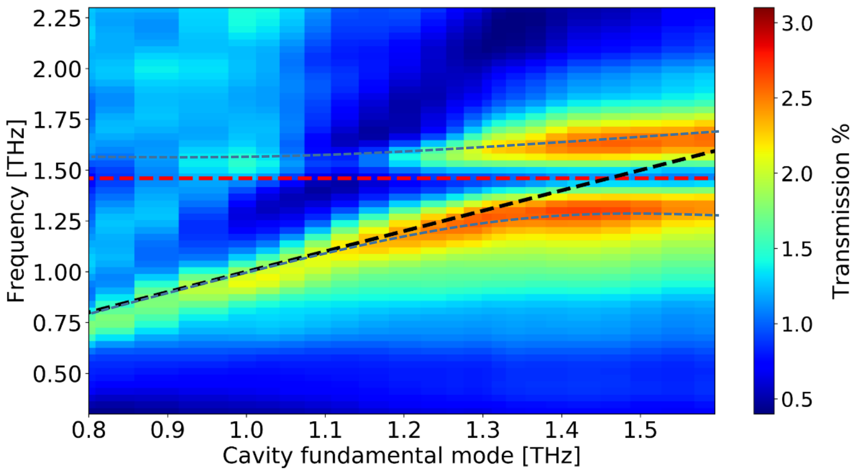

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Quantum spectroscopies for quantum materials

- ✓ **Can we induce order from disorder?**

  Fluctuations of low energy modes is the key

- ✓ **How can we measure fluctuations?**

  We can use quantum and classical statistical properties of light

  Covariance Based Raman Spectroscopy

- ✓ **<u>How do we control fluctuations in matter?</u>**

  Cavity electrodynamics to control complex materials

  Controlling metal-insulator transition temperature in 1T-TaS2 in cavity

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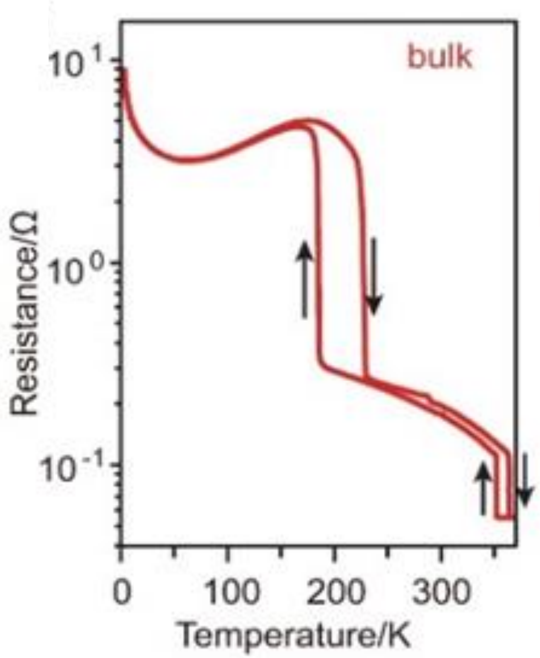
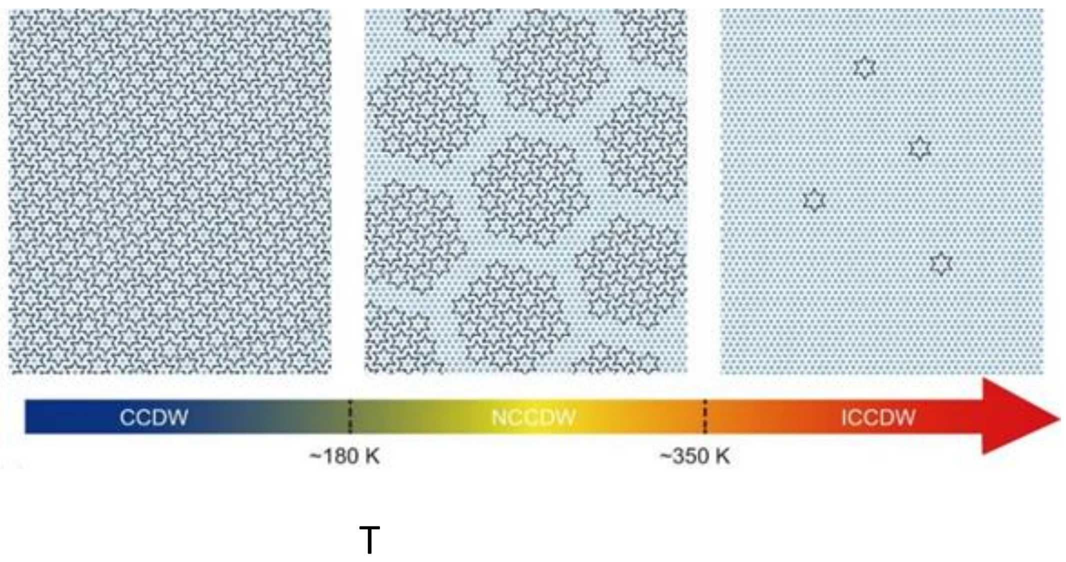
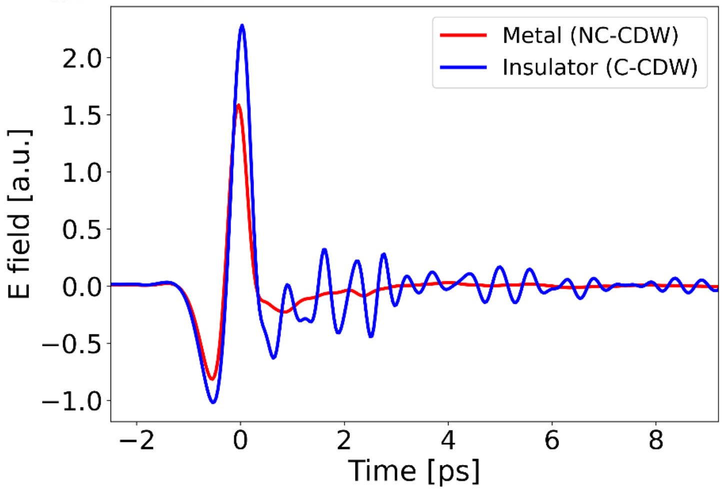
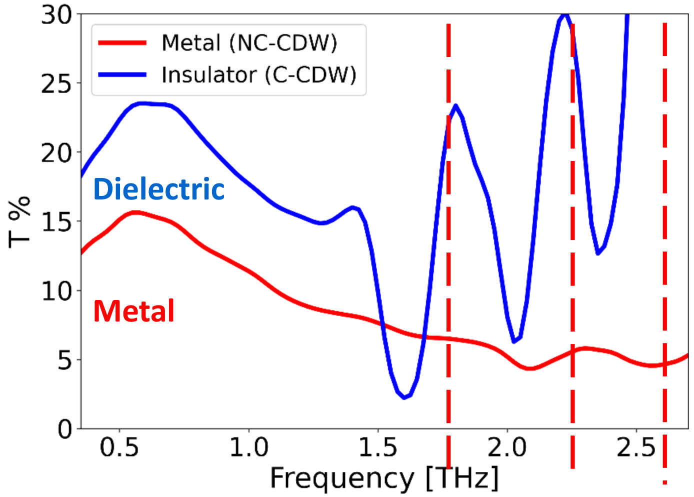
Cavity control of M-I transition in 1T-TaS2





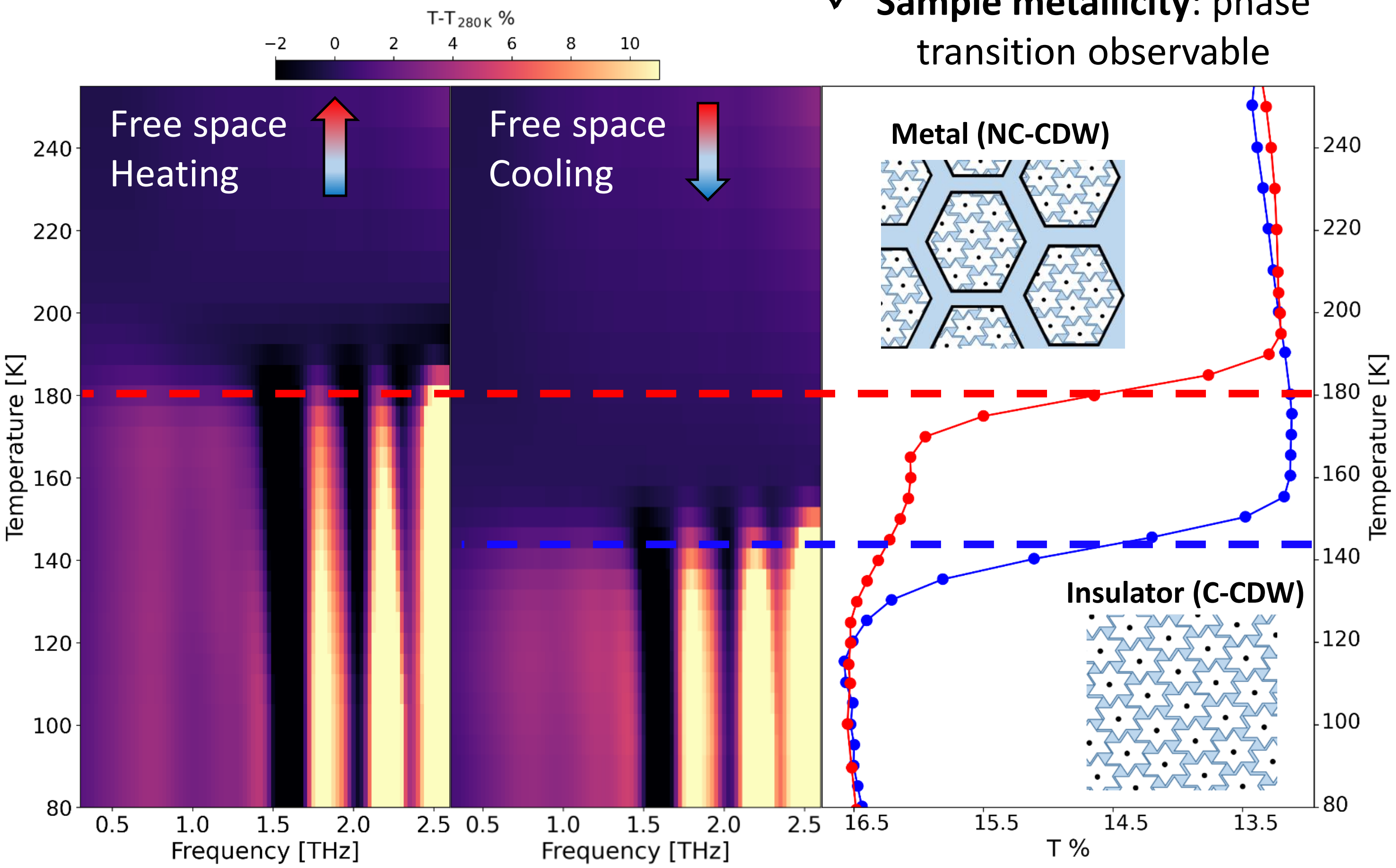




Cavity control of M-I transition in 1T-TaS2

✓ THz tracking of **1st order** metal-to-insulator phase transition

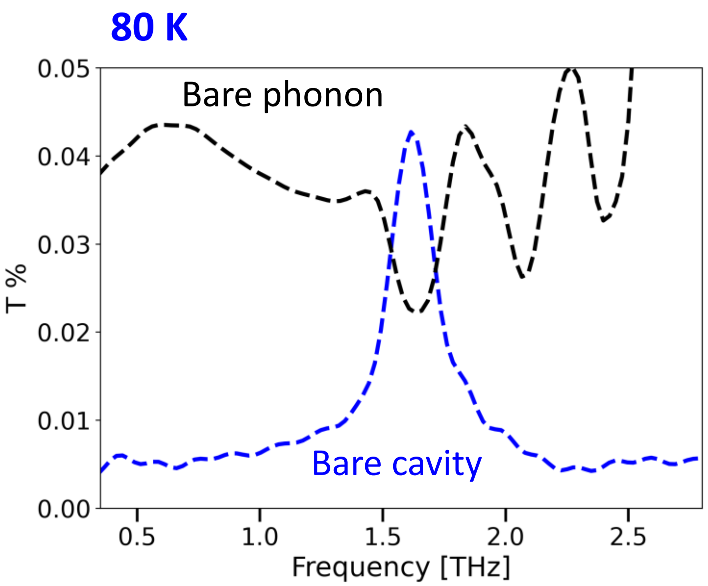
✓ **Sample metallicity**: phase transition observable



Resonant Coupling to phonons

✓ **Temperature dependent**

C-CDW phonon



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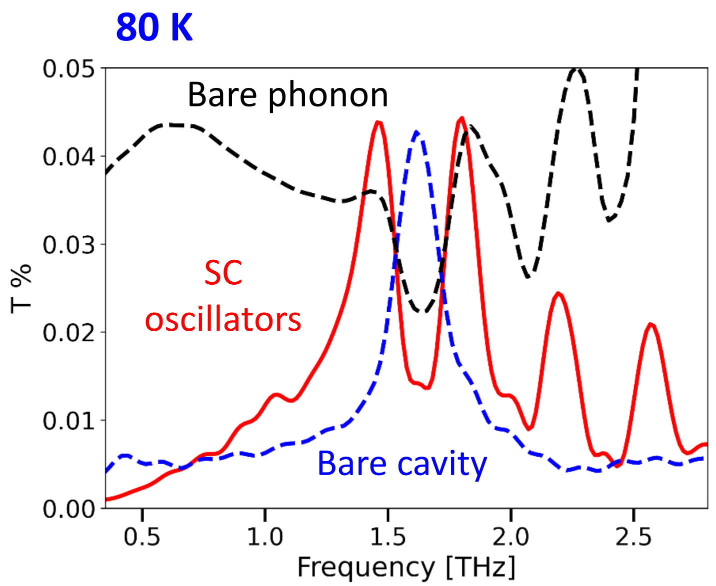

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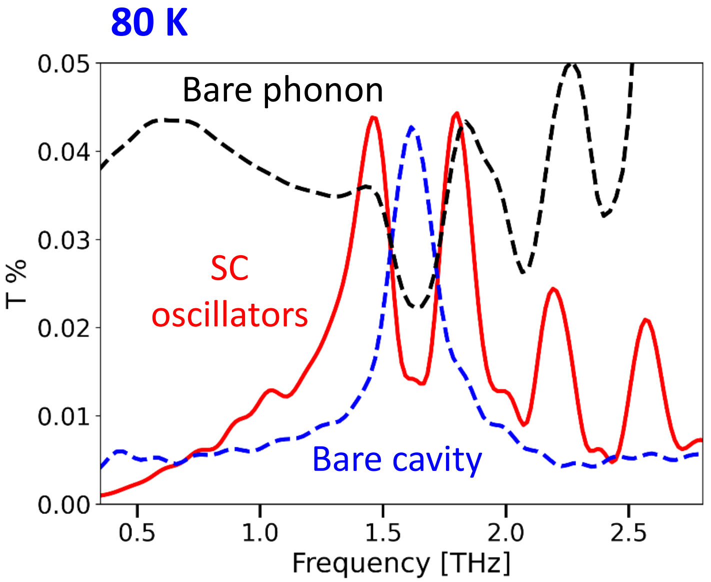

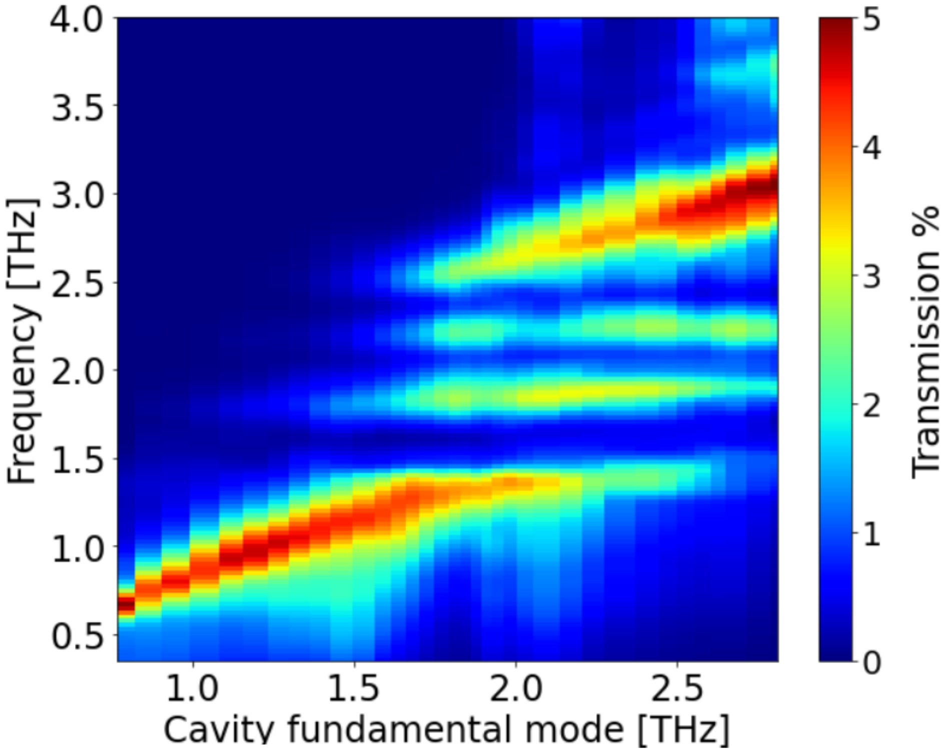

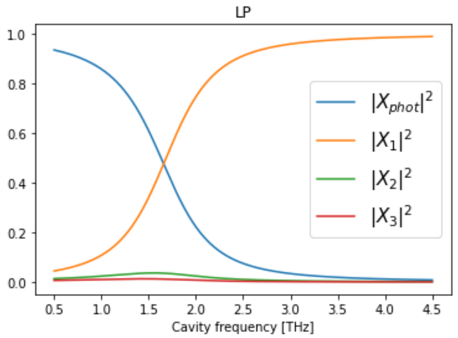

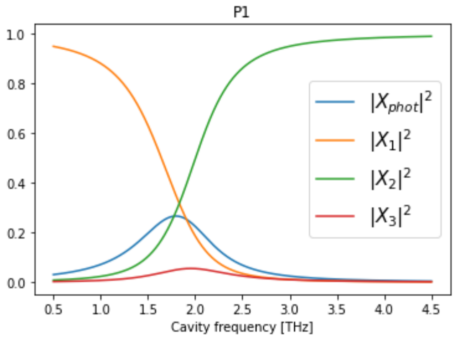

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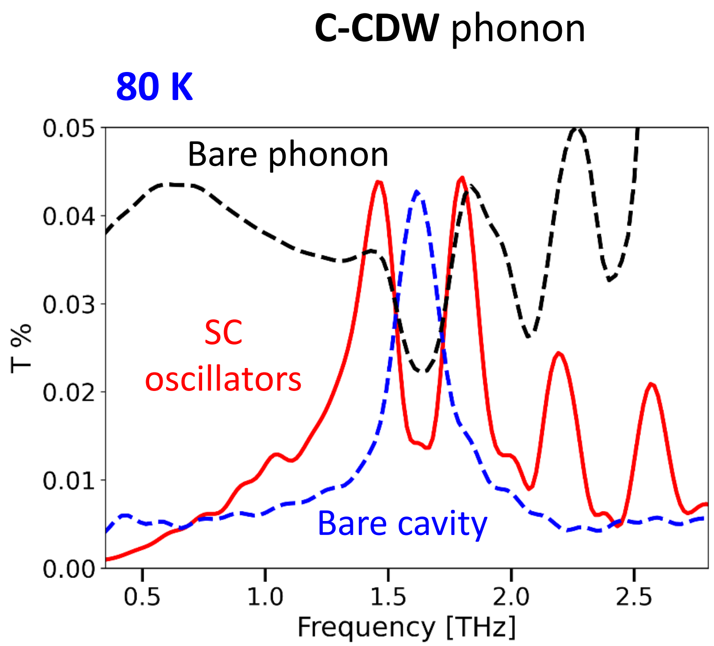
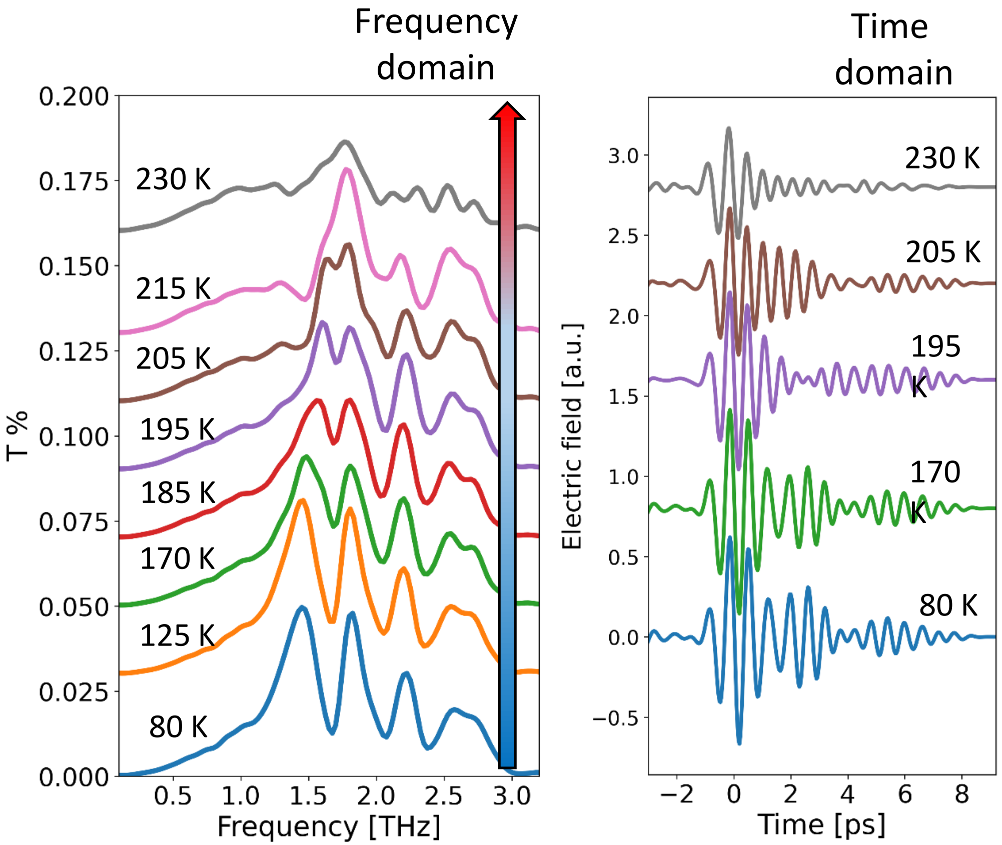
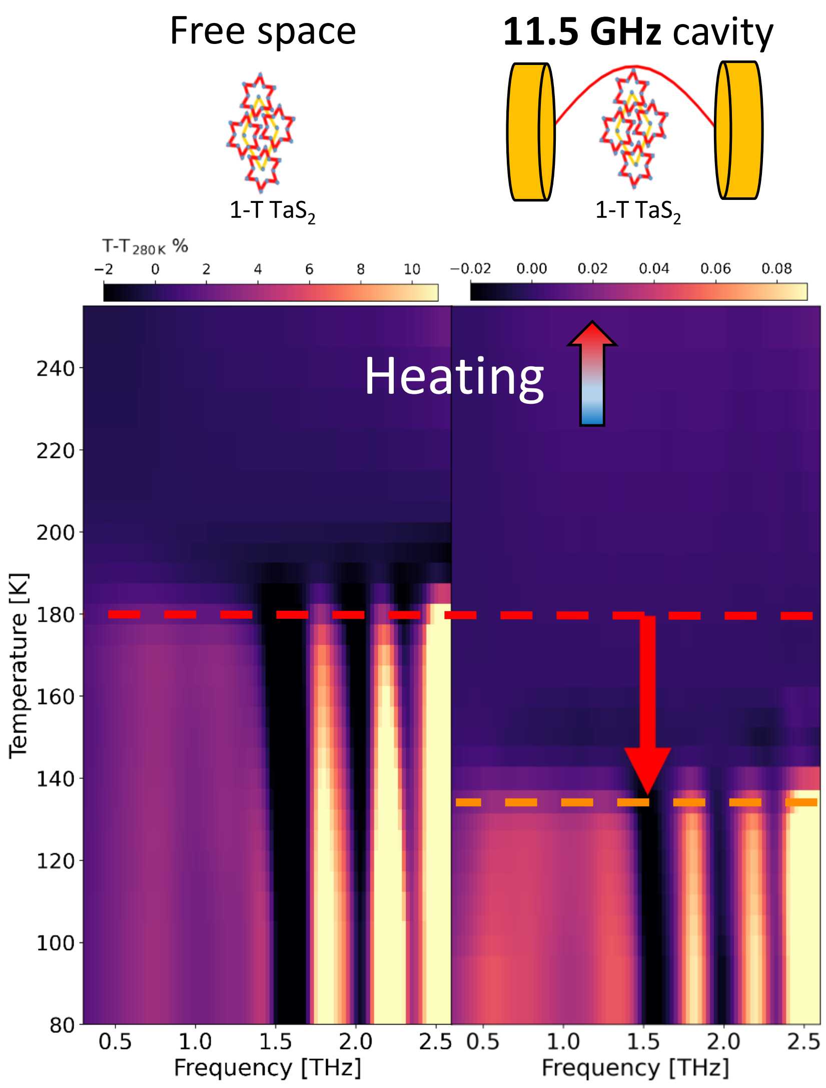




✓ **Temperature dependent light-matter coupling** revealed by THz spectroscopy

Control of Tc in long wavelength cavities



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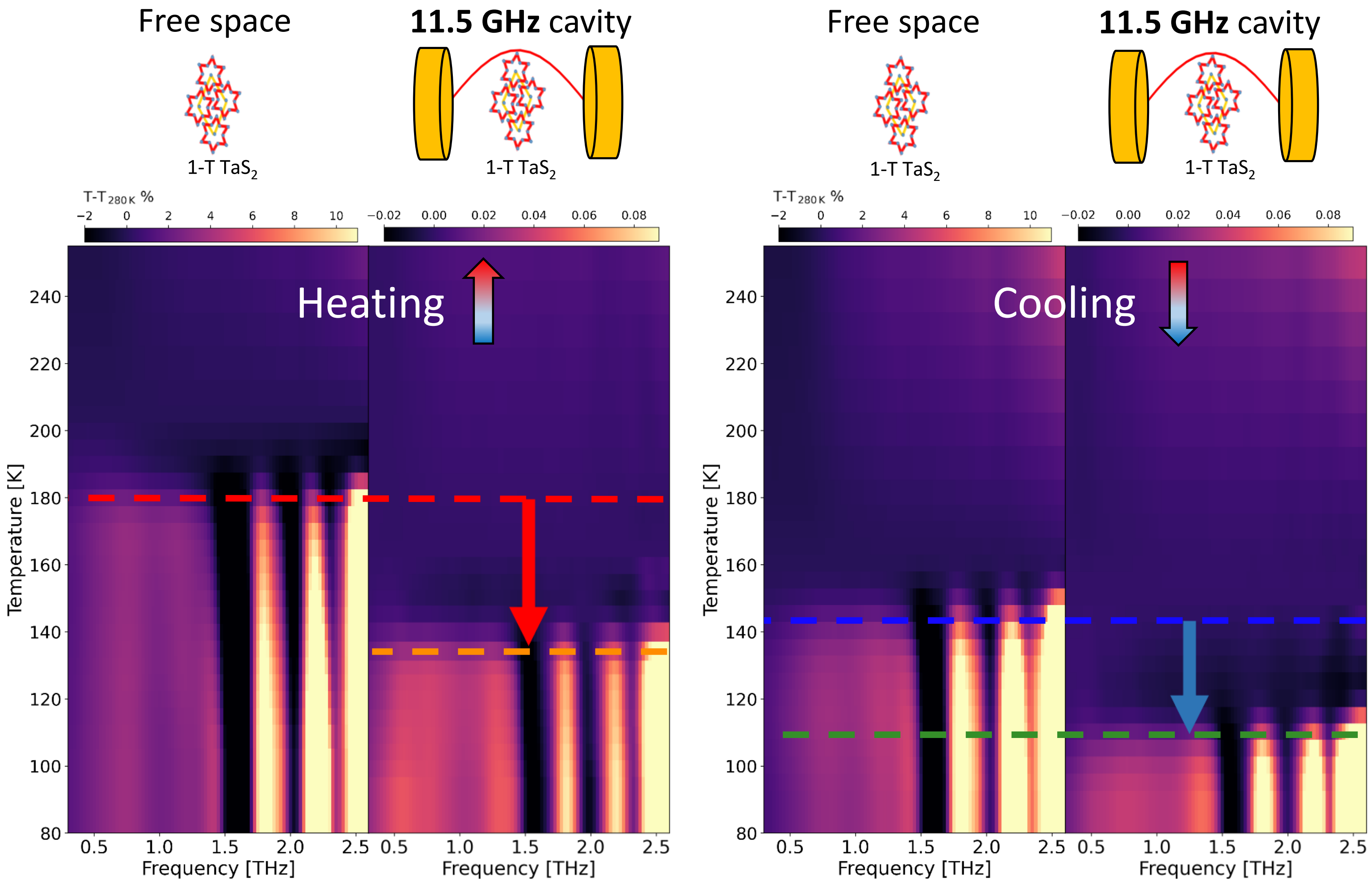

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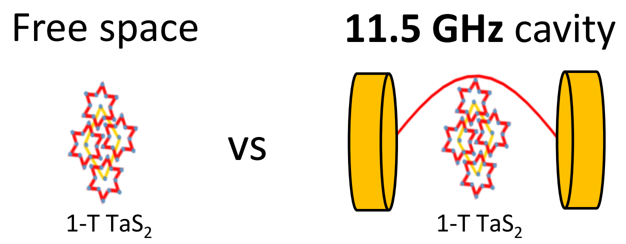
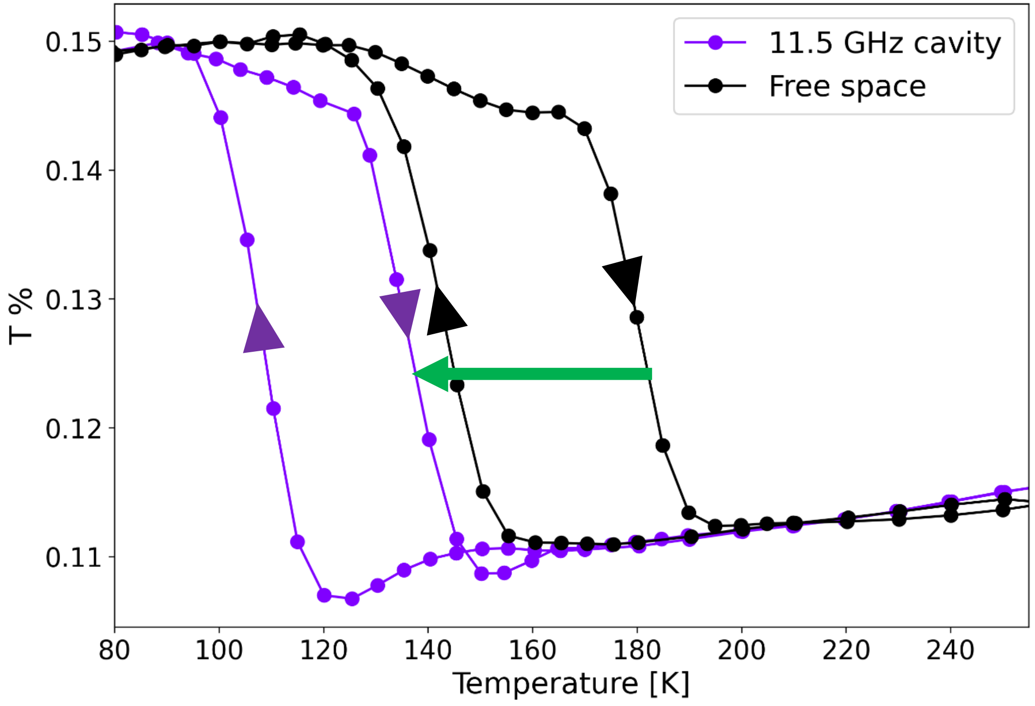
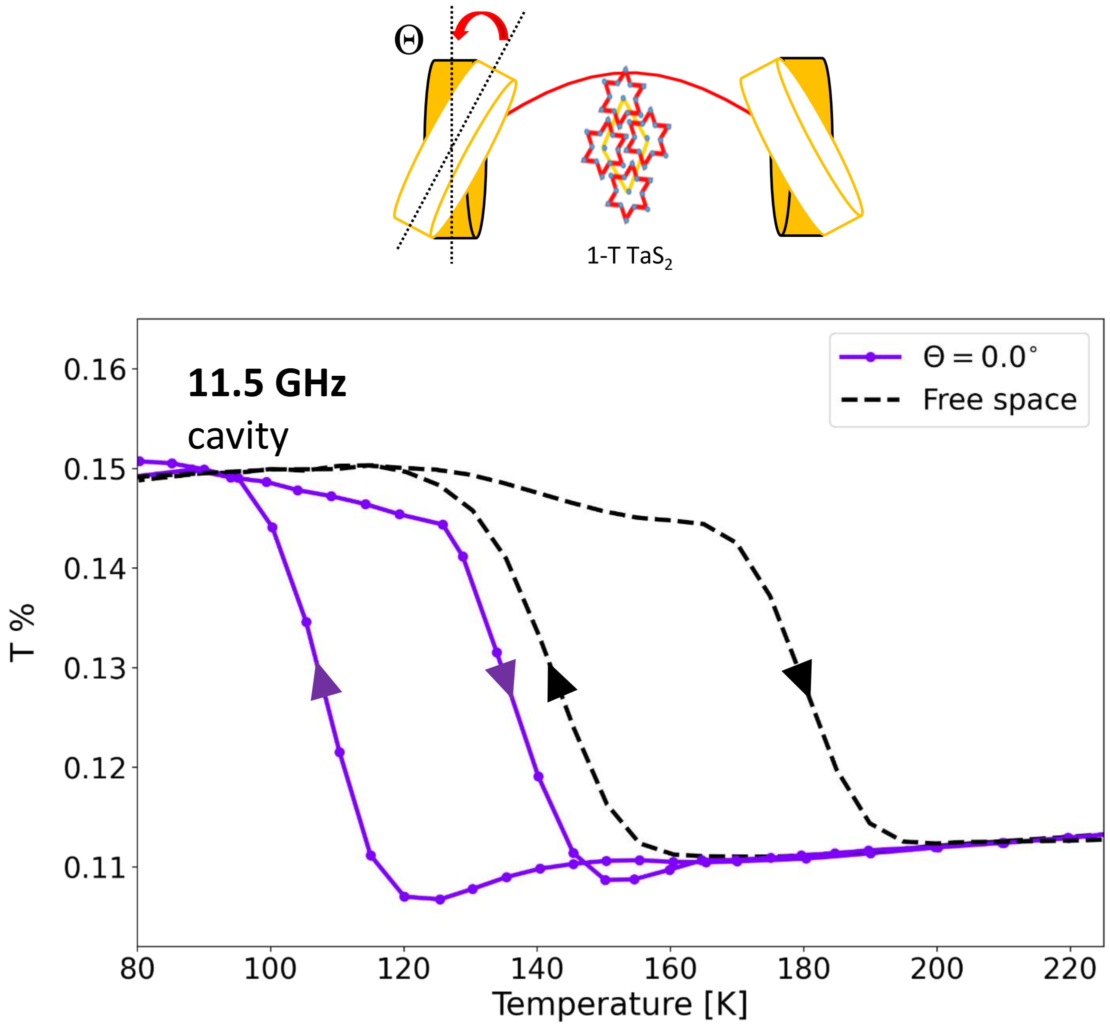




✓ **Cavity-mediated** renormalization of the **phase transition temperature**

https://arxiv.org/abs/2210.02346

Dependence on the Cavity geometry



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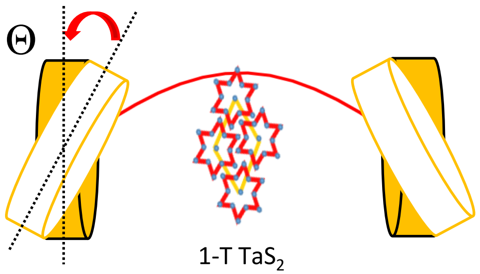

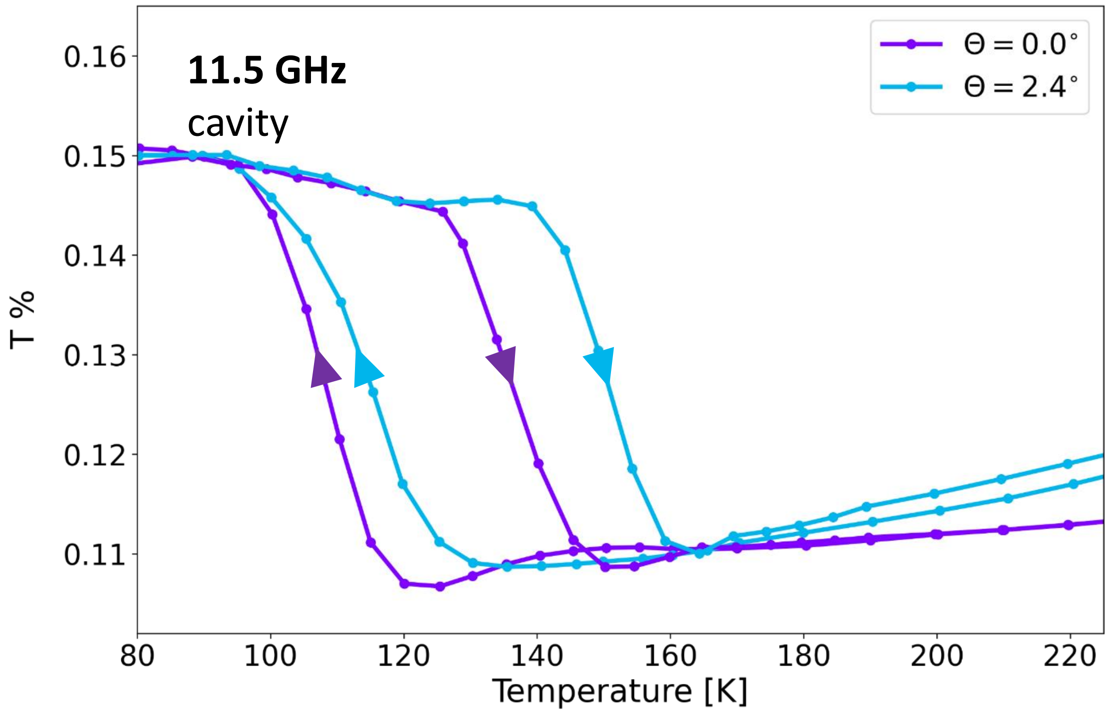

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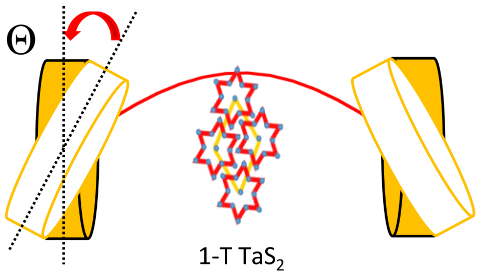
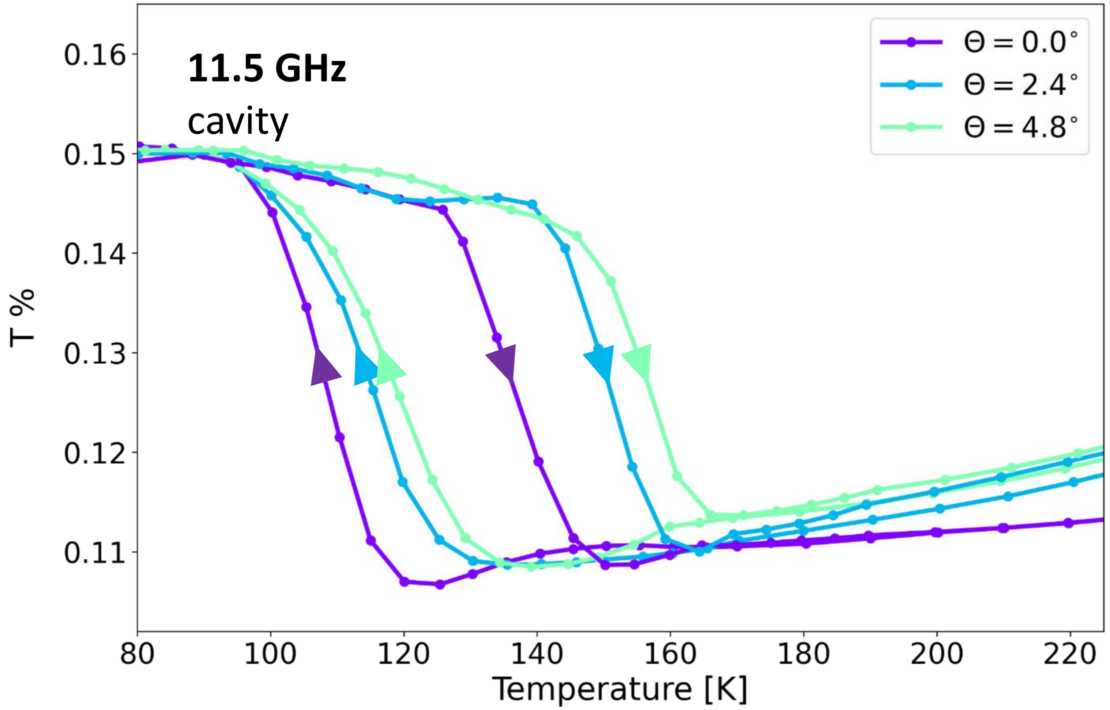
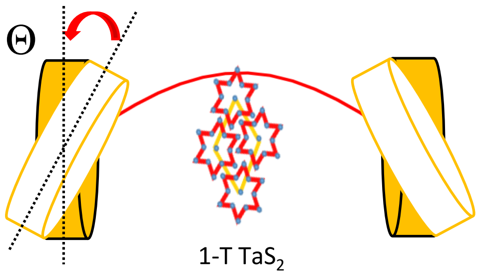
Dependence on the Cavity geometry

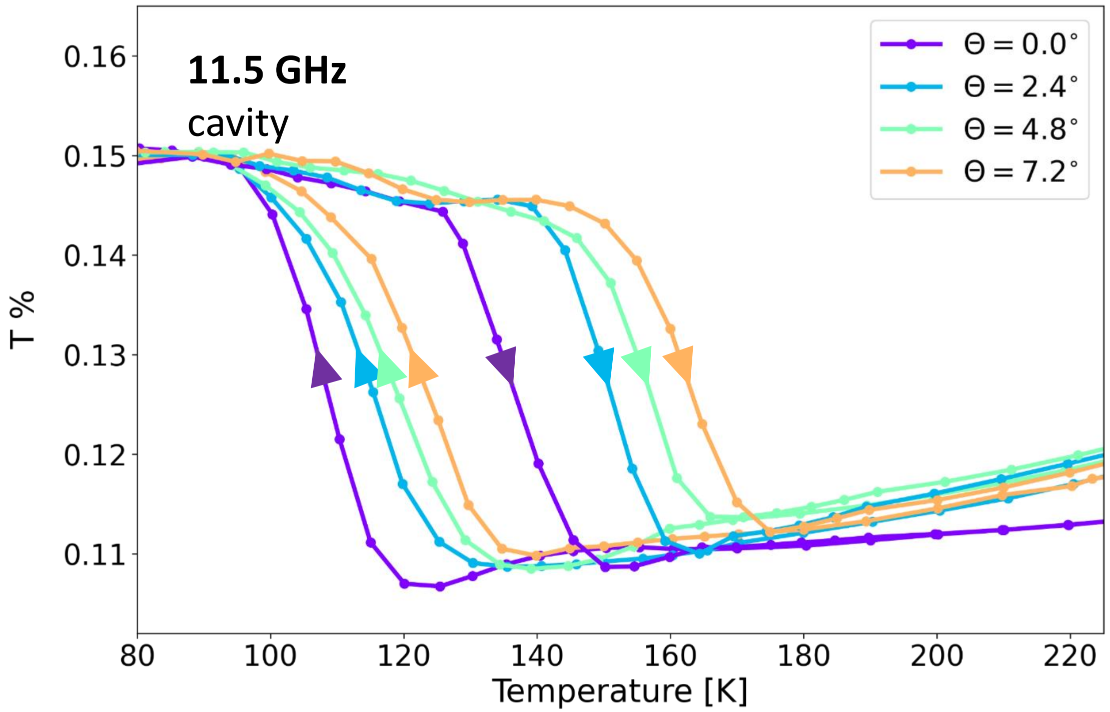




https://arxiv.org/abs/2210.02346

Dependence on the Cavity geometry





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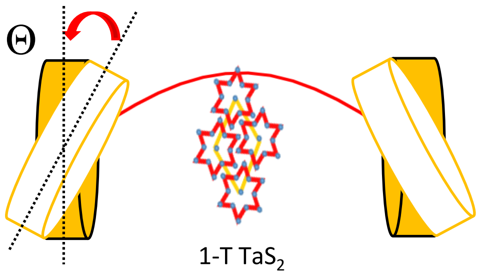

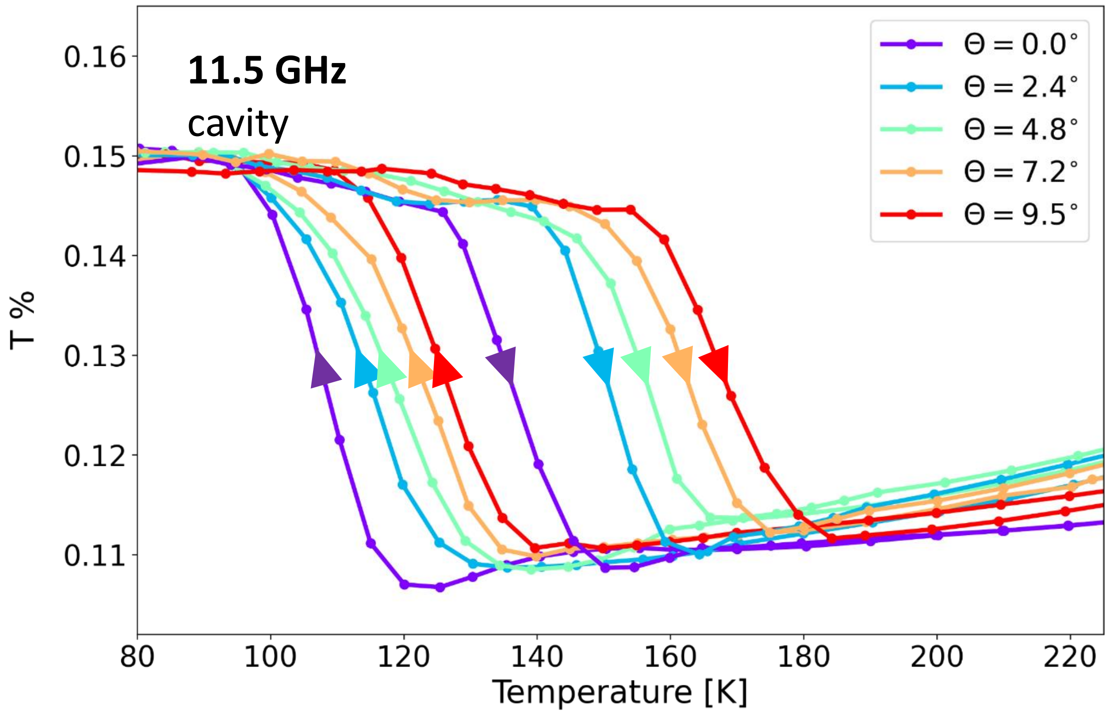

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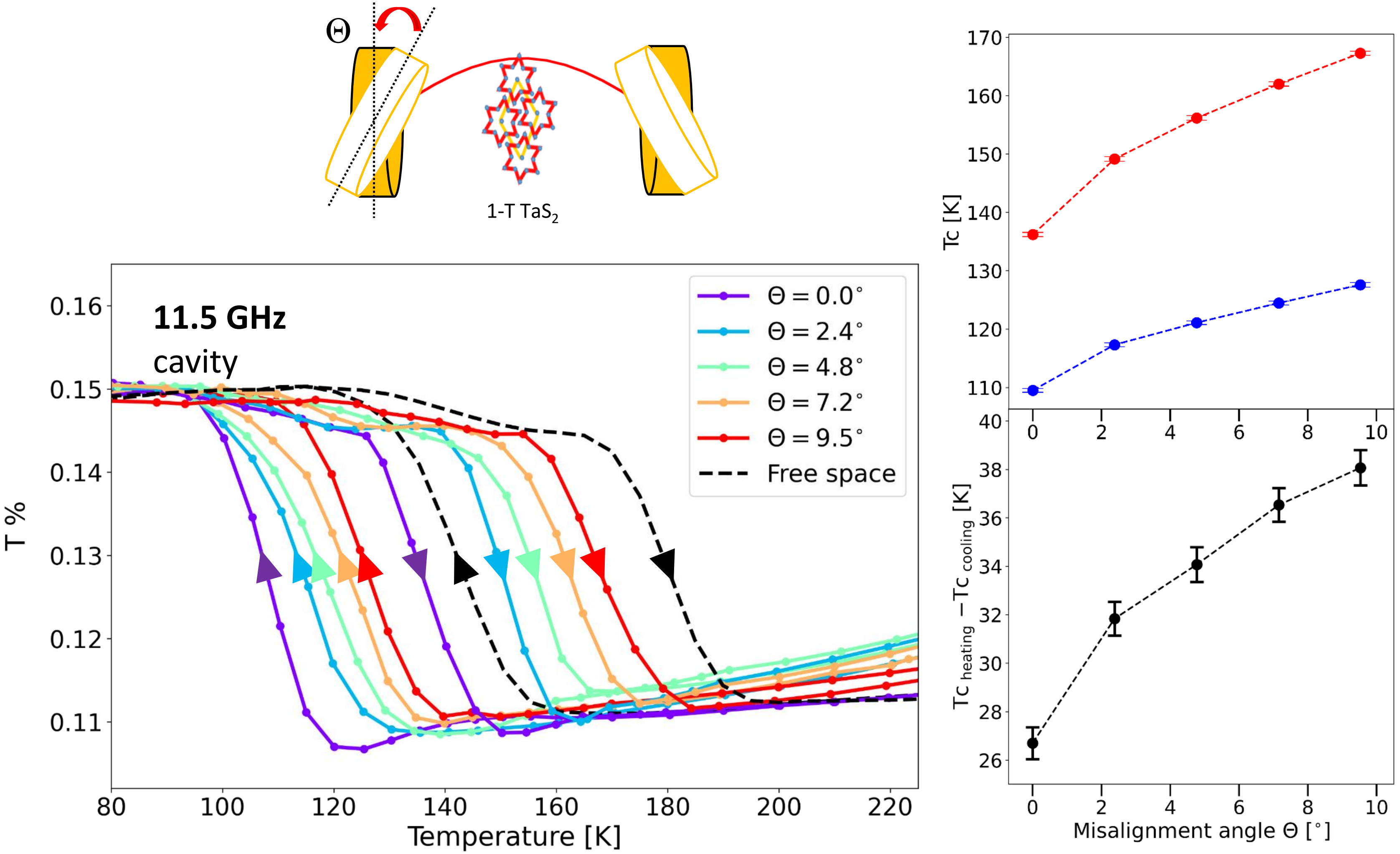

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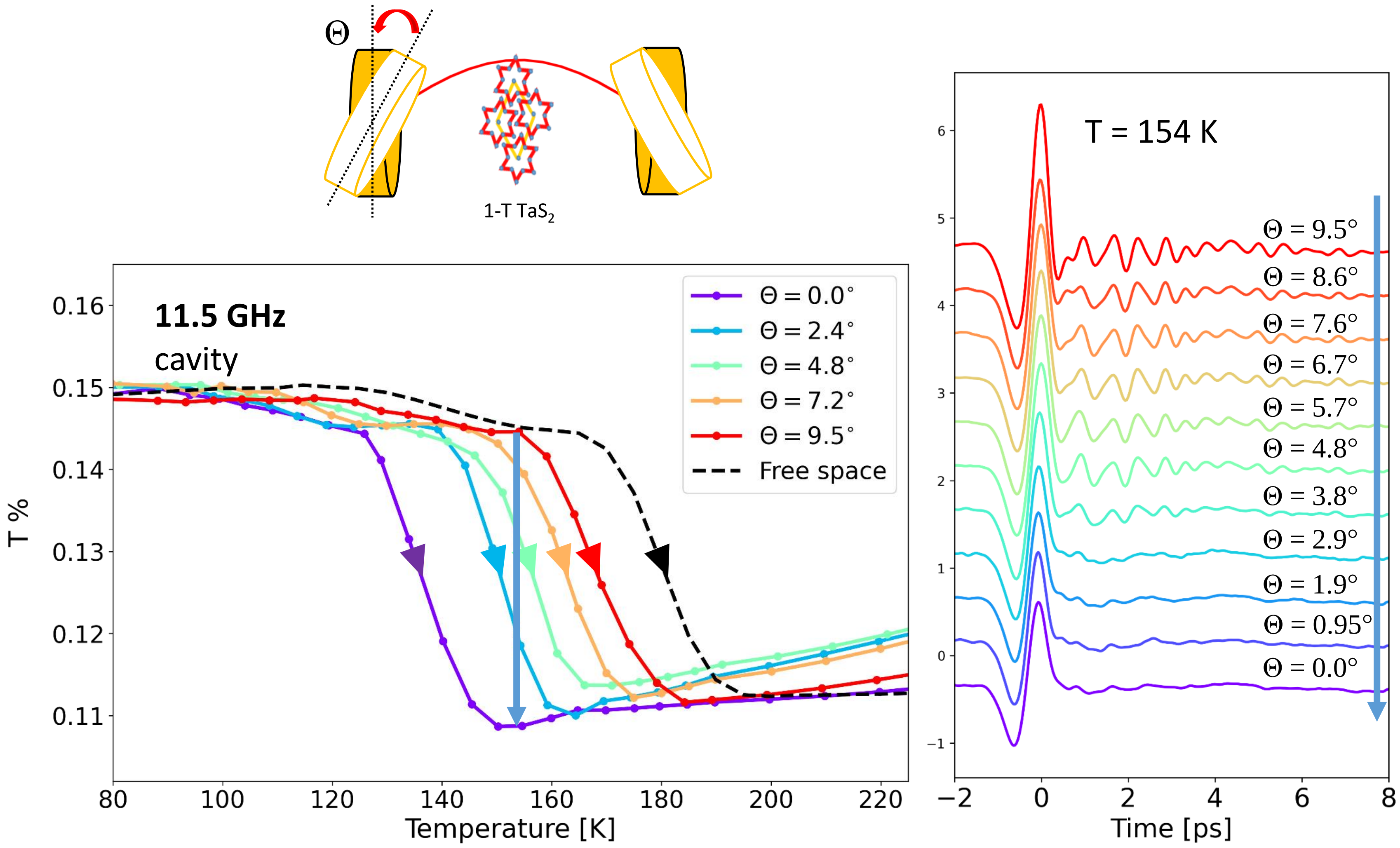

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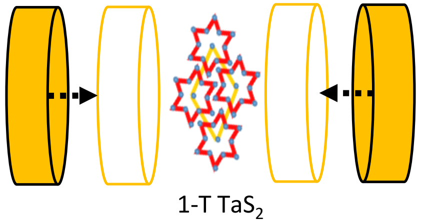
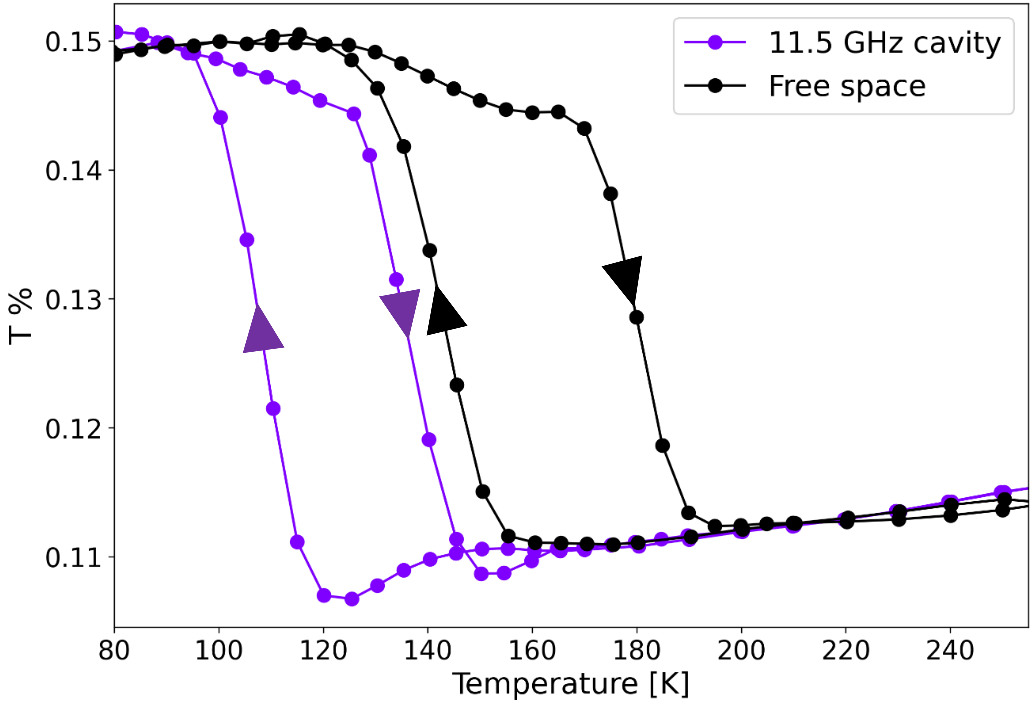
Dependence on the Cavity frequency



Cavity fundamental frequency tuning ?



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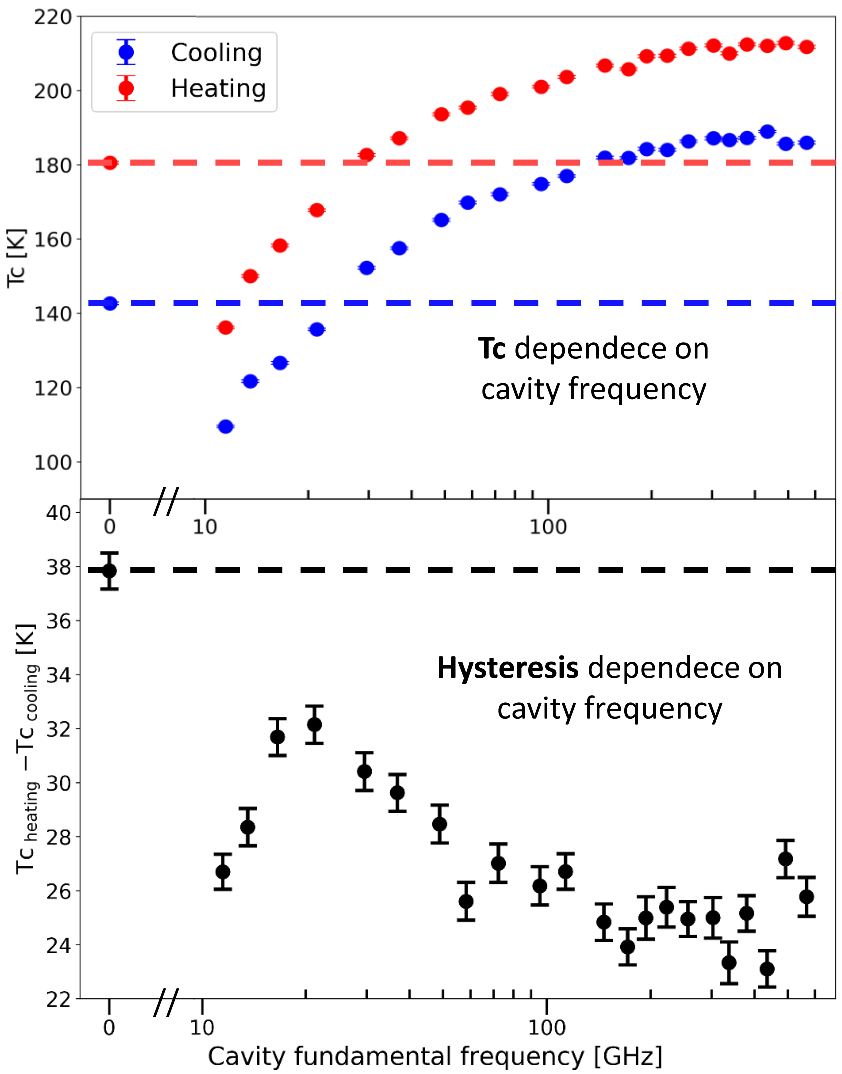
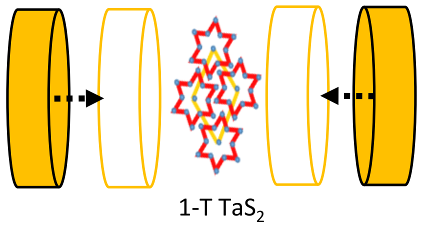
Dependence on the Cavity frequency

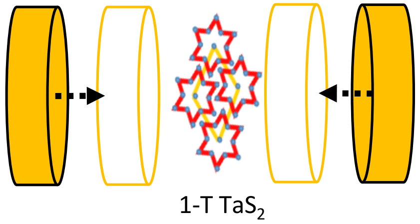




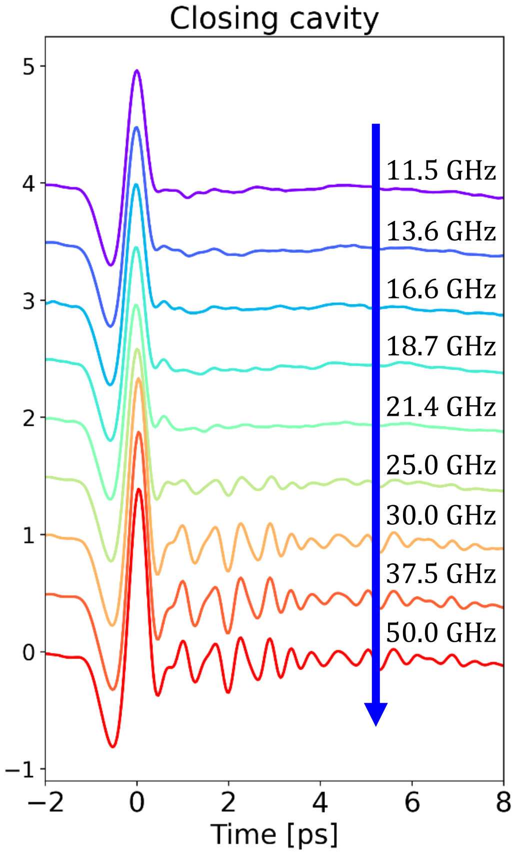
- ✓ **Metallic (NC-CDW)** phase stabilization upon cavity frequency reducing
- ✓ **Cavity-mediated** hysteresis change

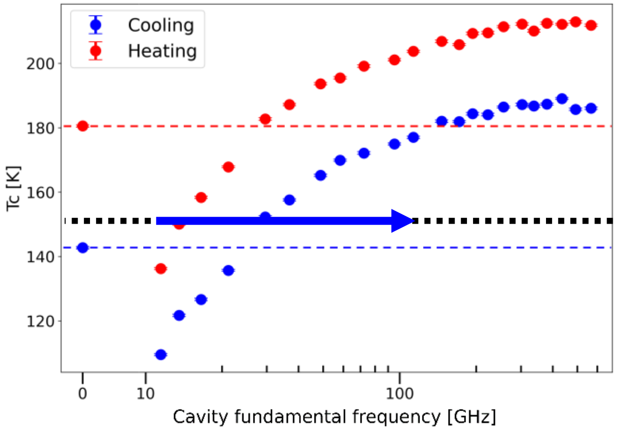
https://arxiv.org/abs/2210.02346

Reversible control of M-I transition



T = 150 K





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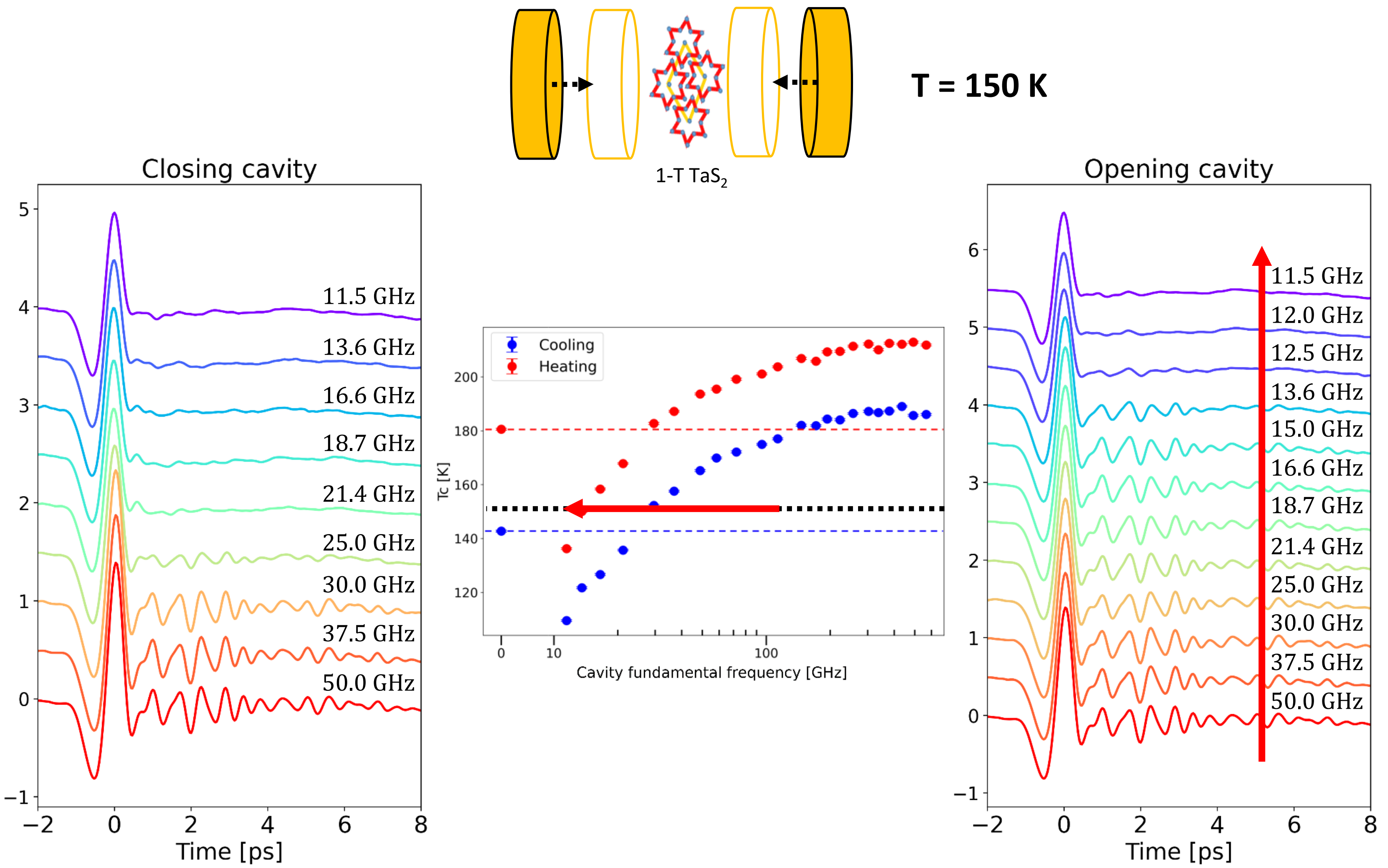

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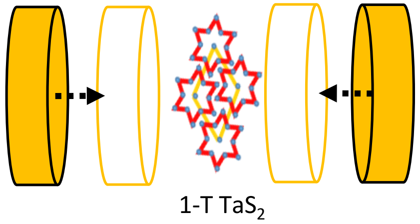

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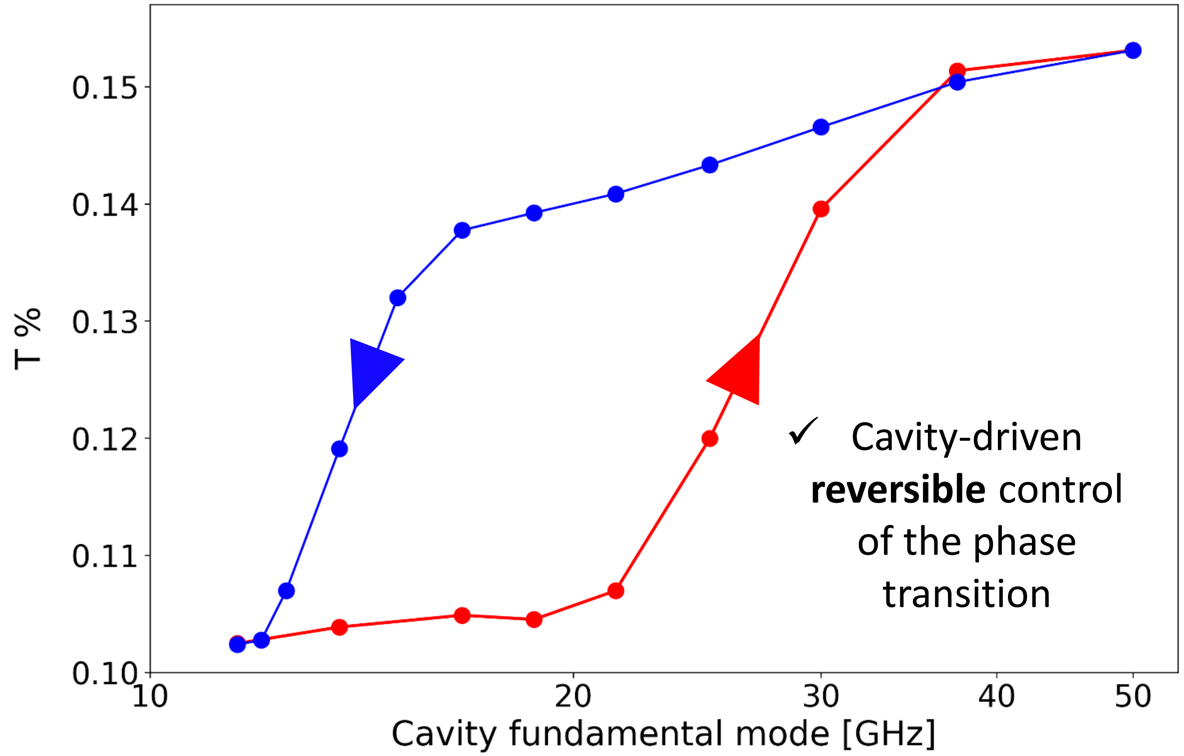

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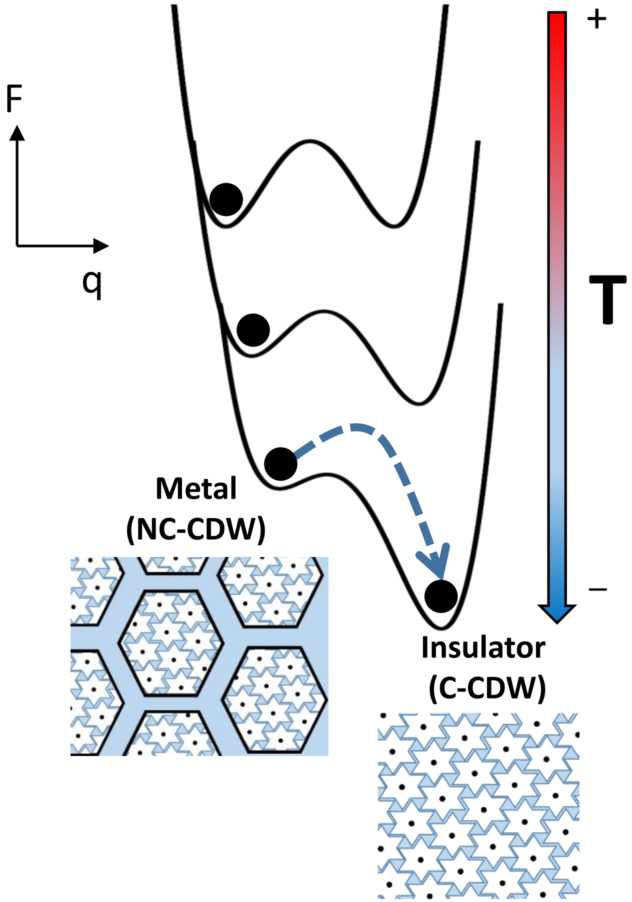
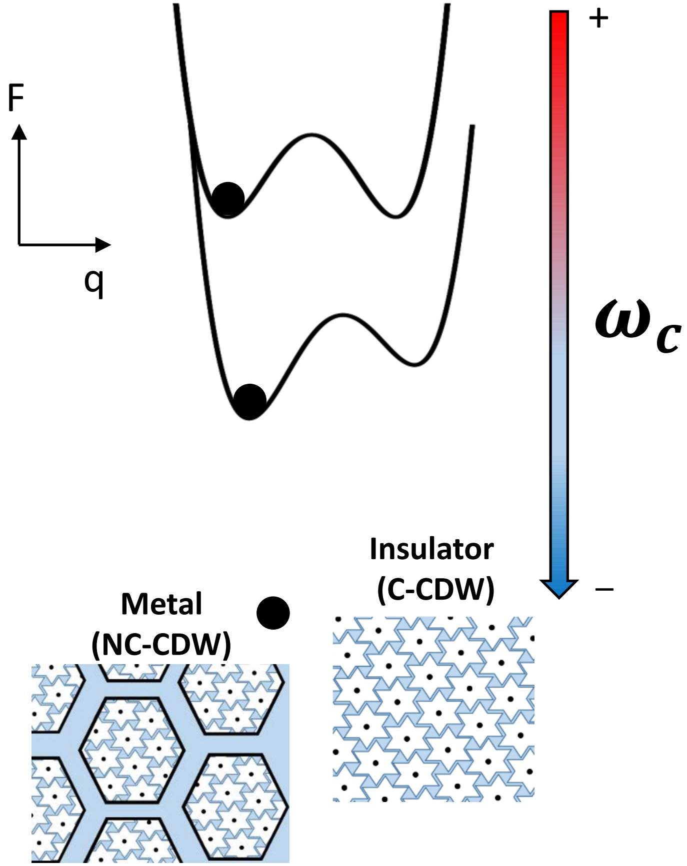
Possible mechanisms #1

Freezing of domain boundaries fluctuations
(dominant in the **GHz-THz** spectrum)



PRB 14, 1496 (1976), J.Phys_Soc_Jap 45,4 (1978)

Charge domain boundary fluctuation in the metallic phase couple to the cavity
→ Renormalization of the free energy of the metallic phase



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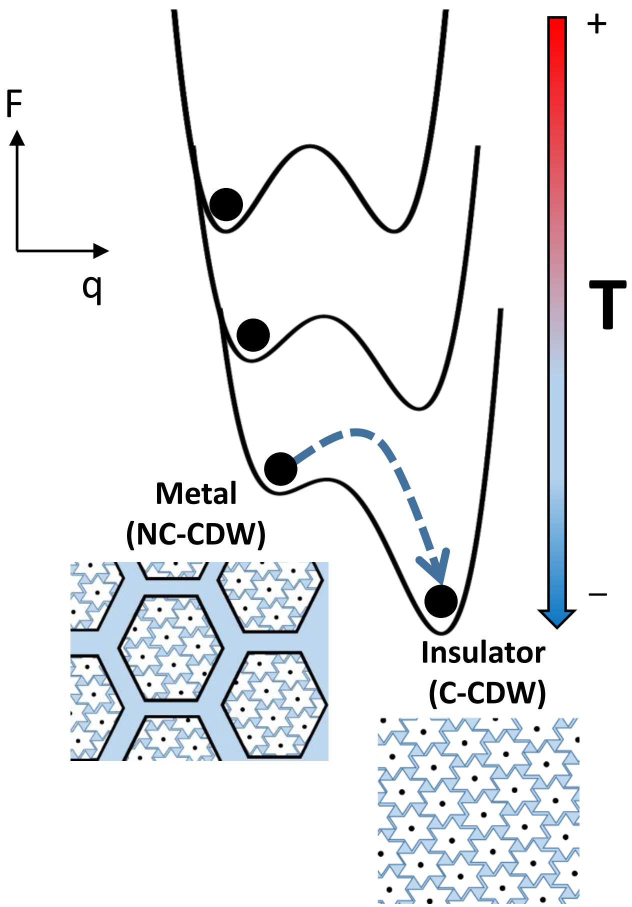

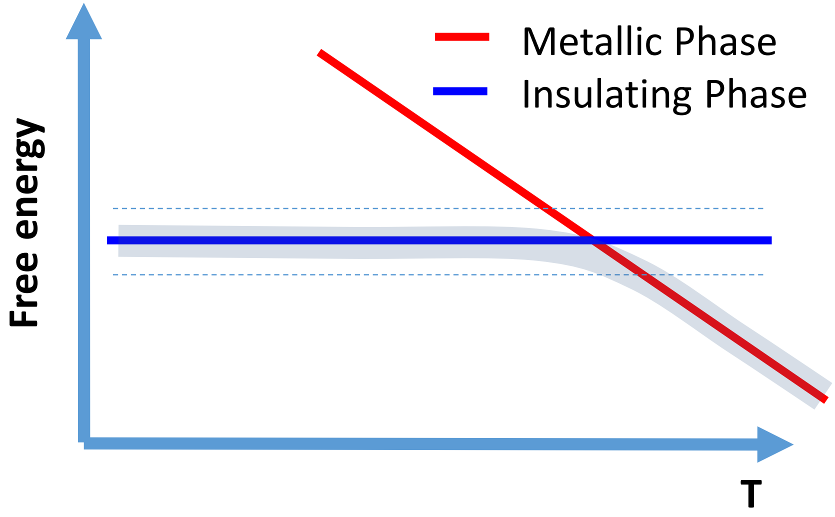

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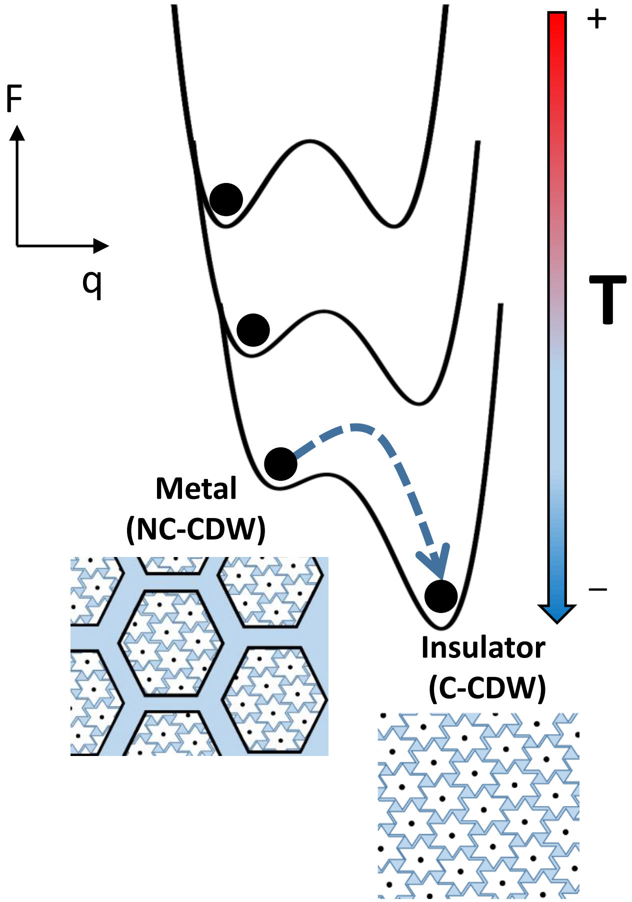

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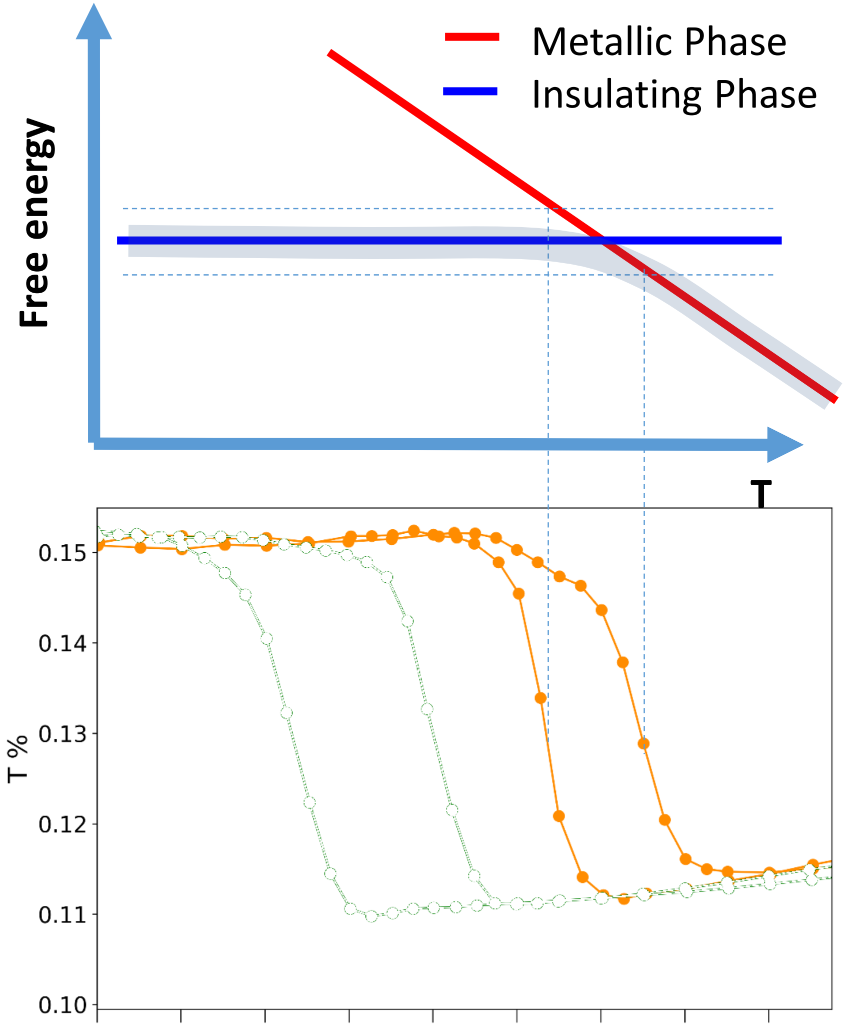

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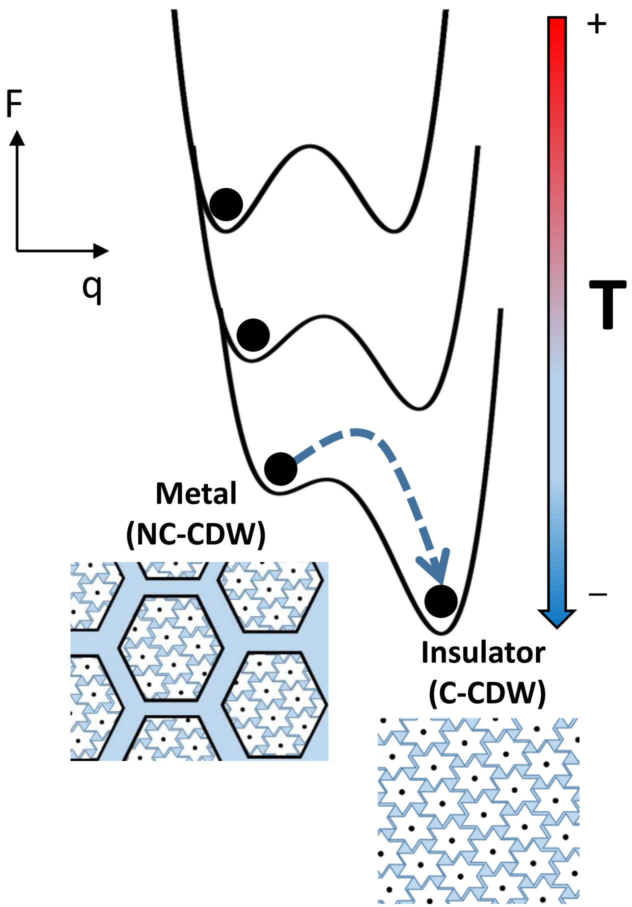

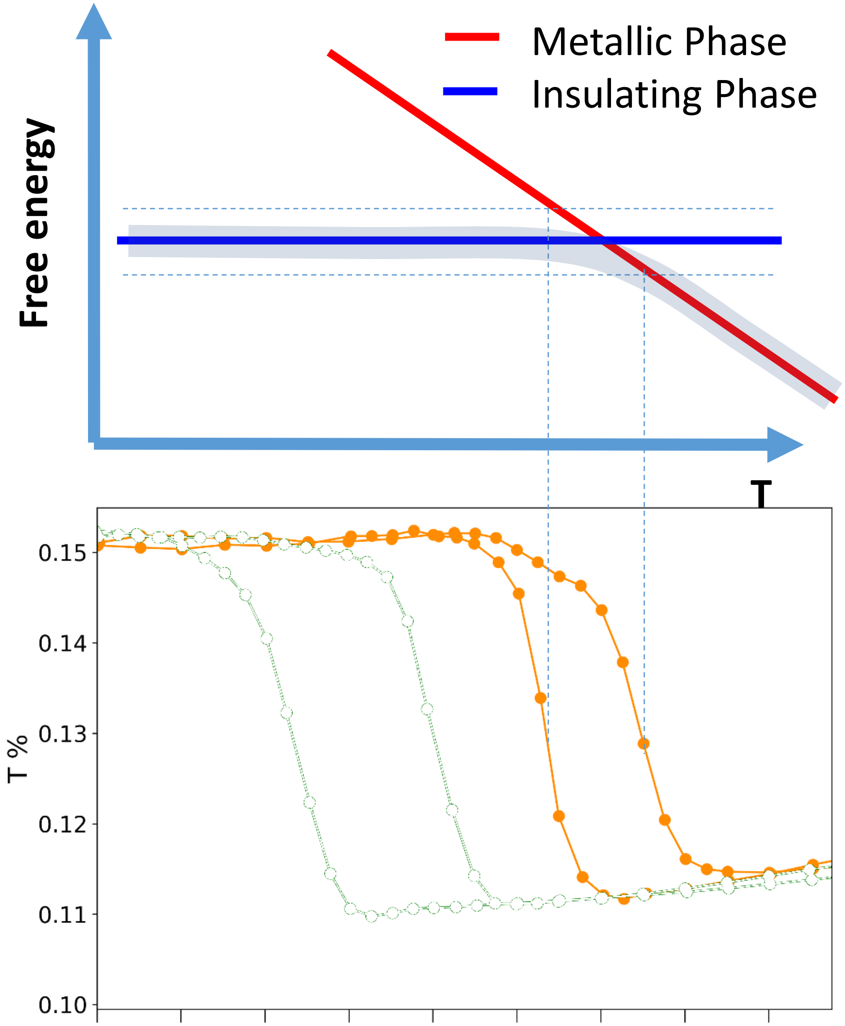

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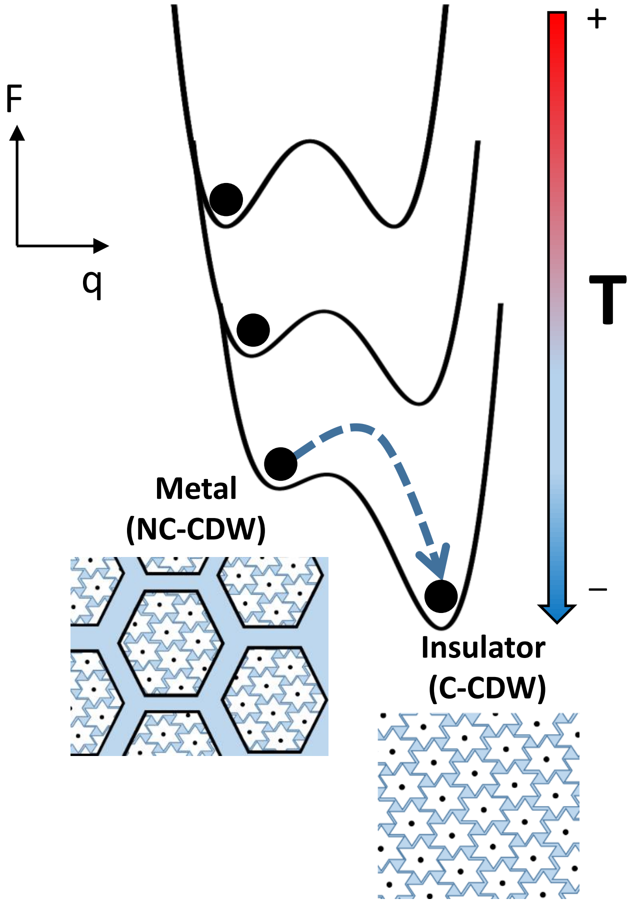

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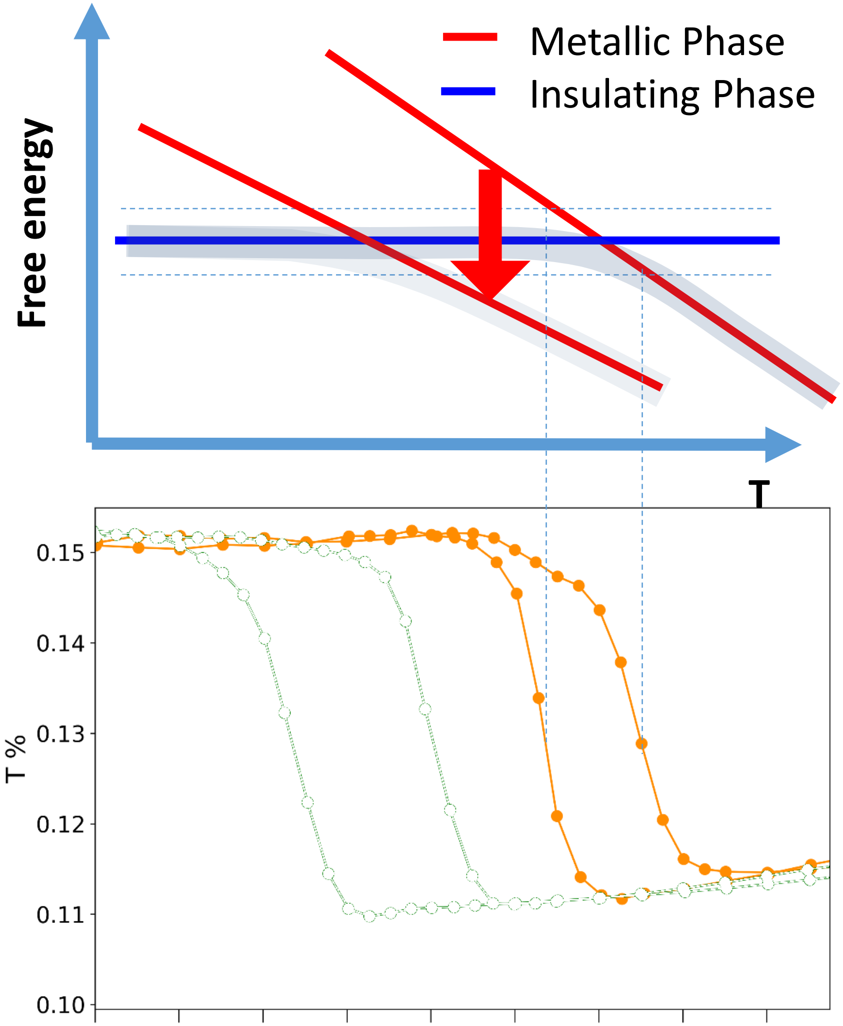

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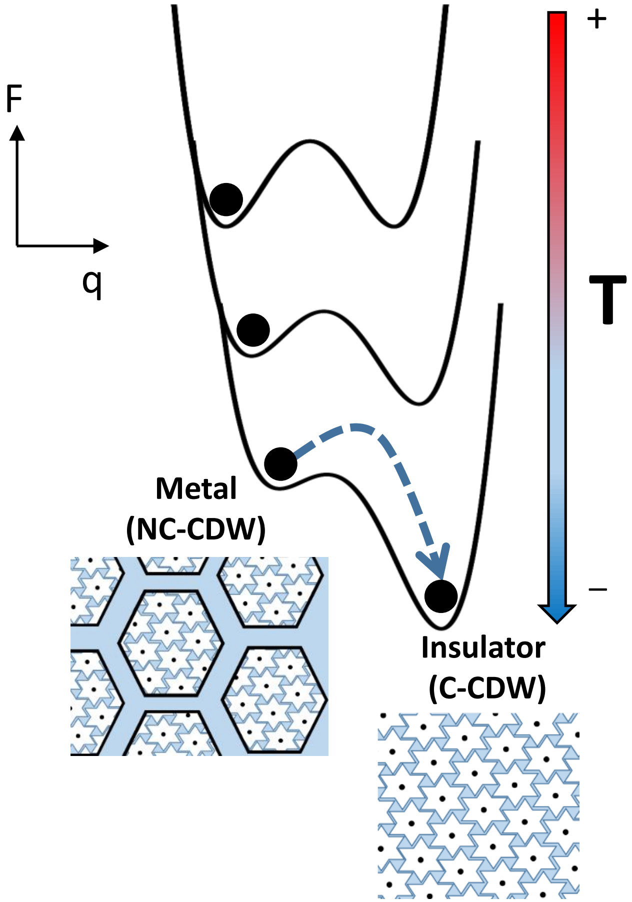

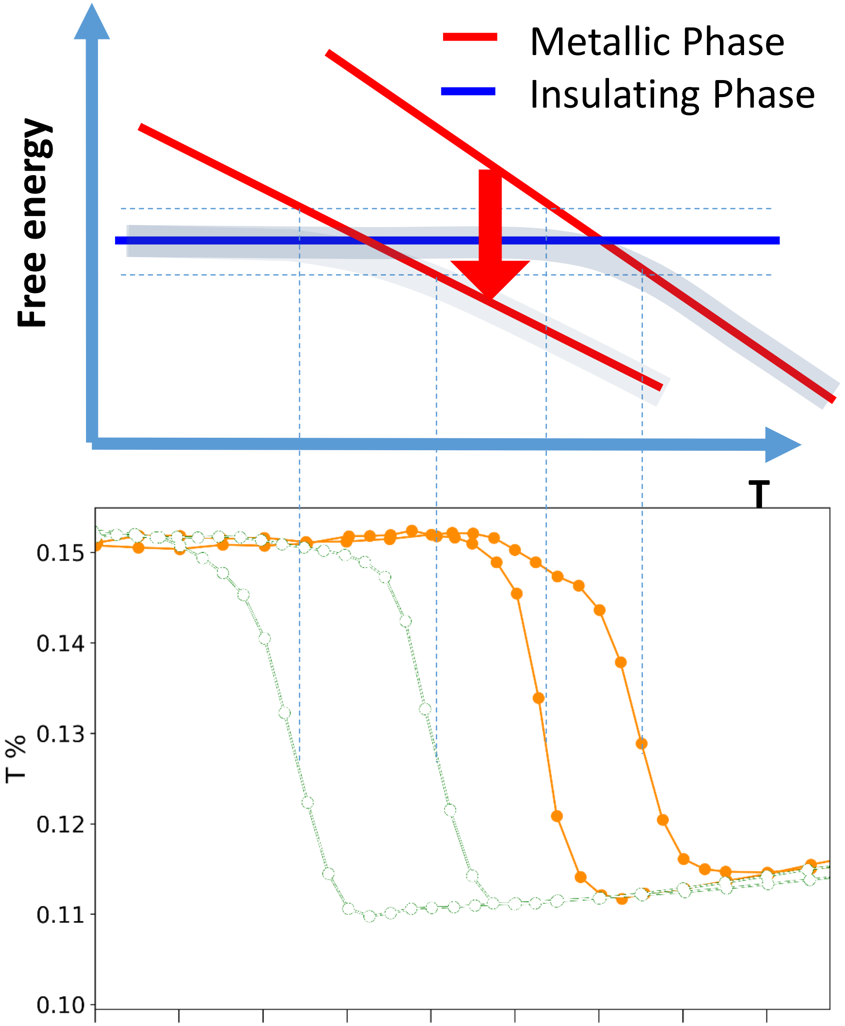

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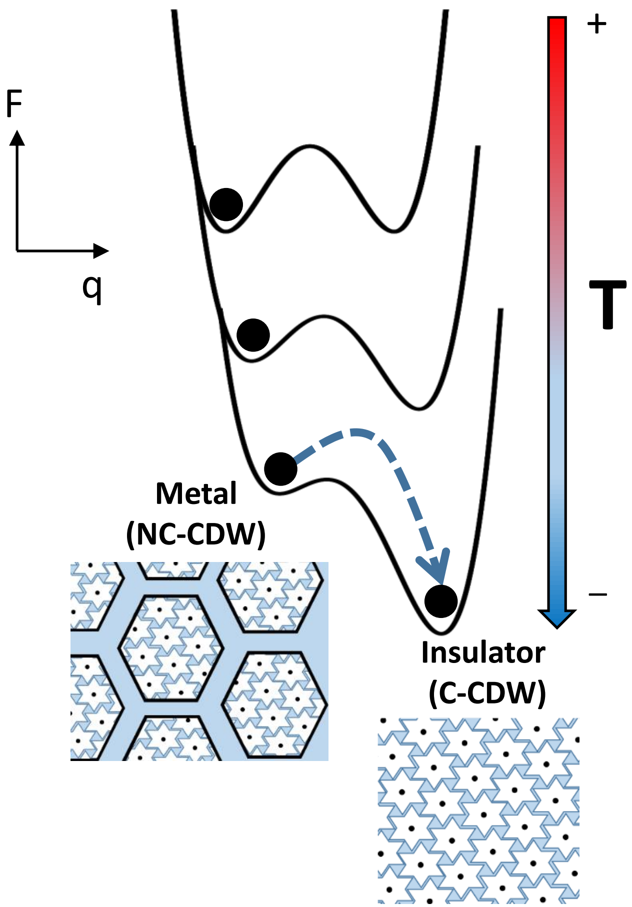

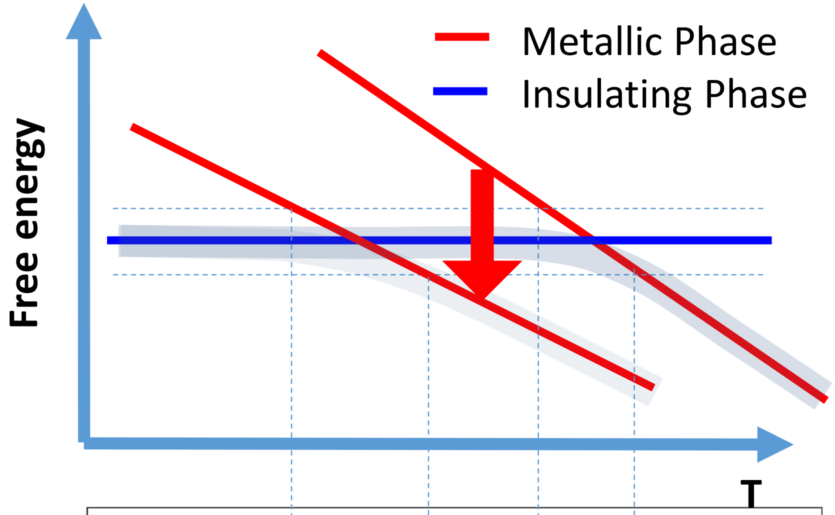

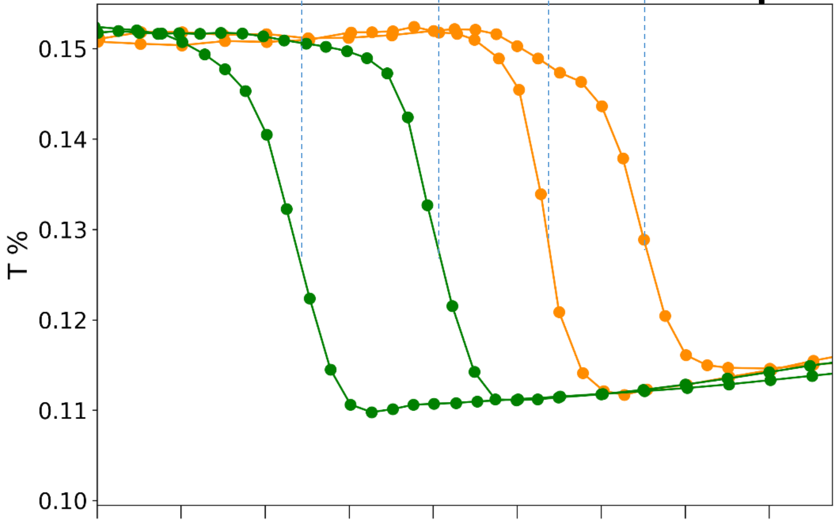

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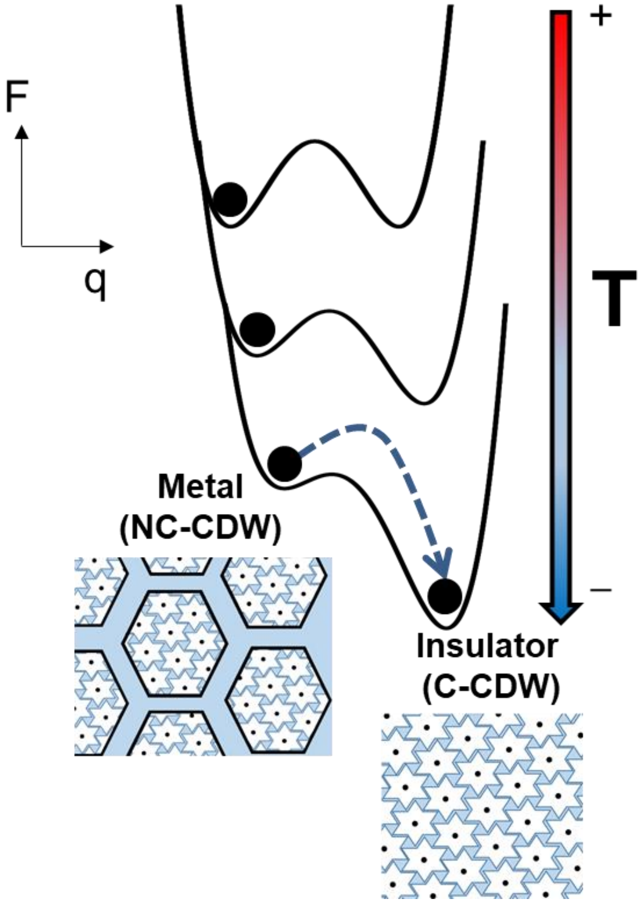

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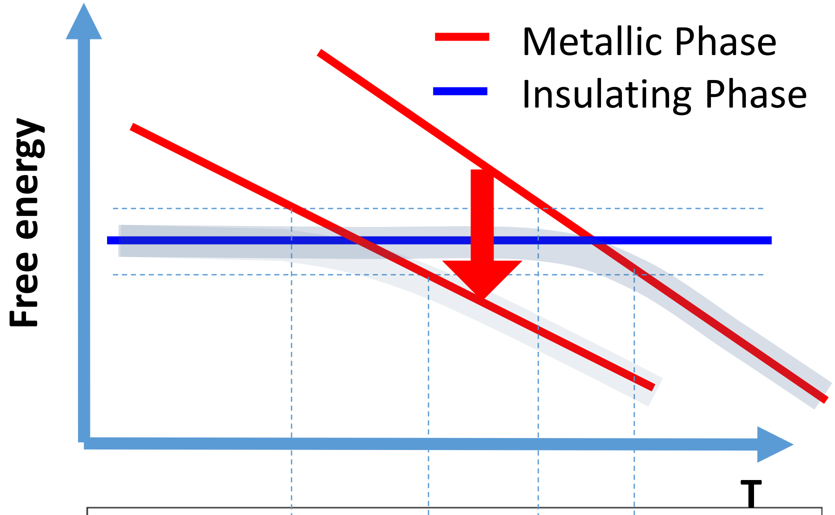

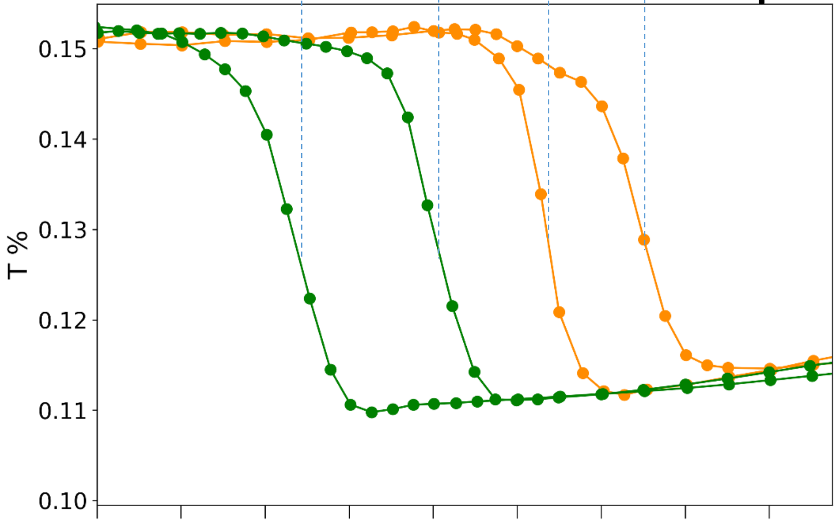

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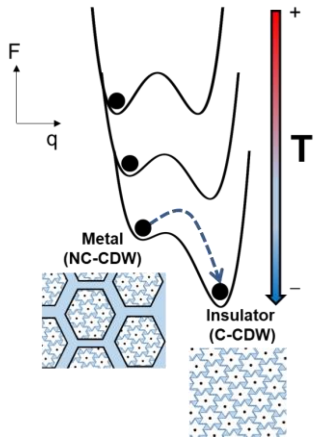

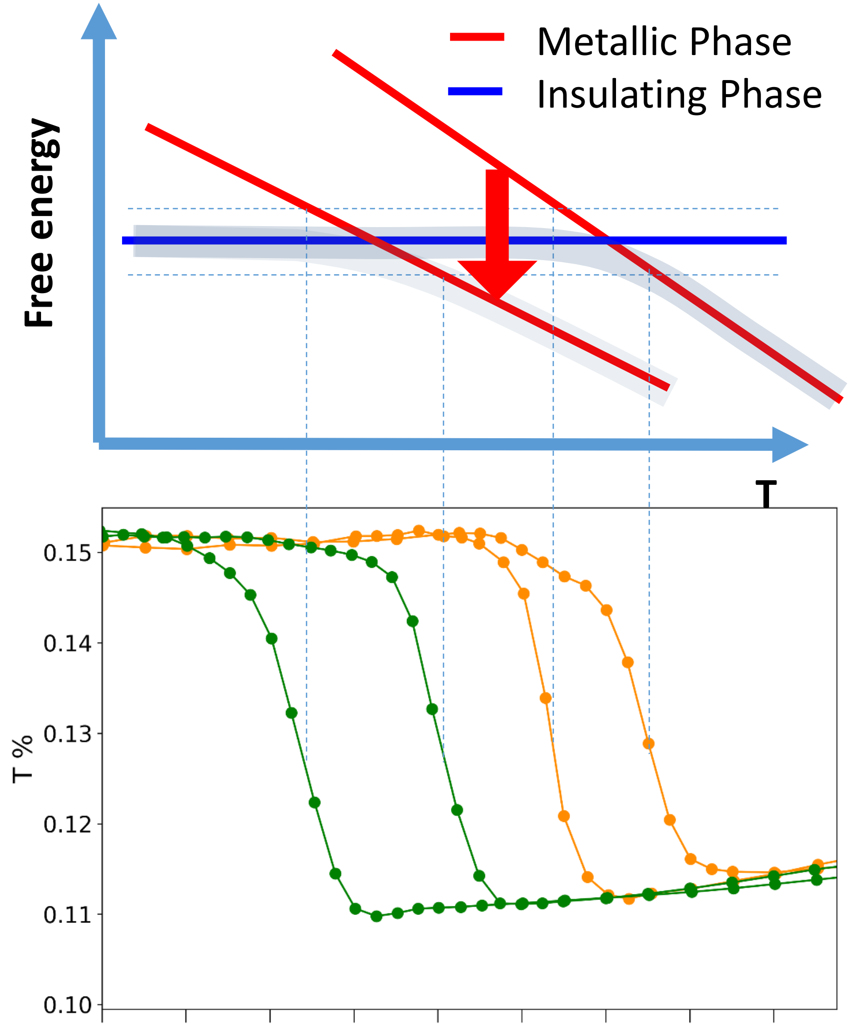

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Freezing of domain boundaries fluctuations
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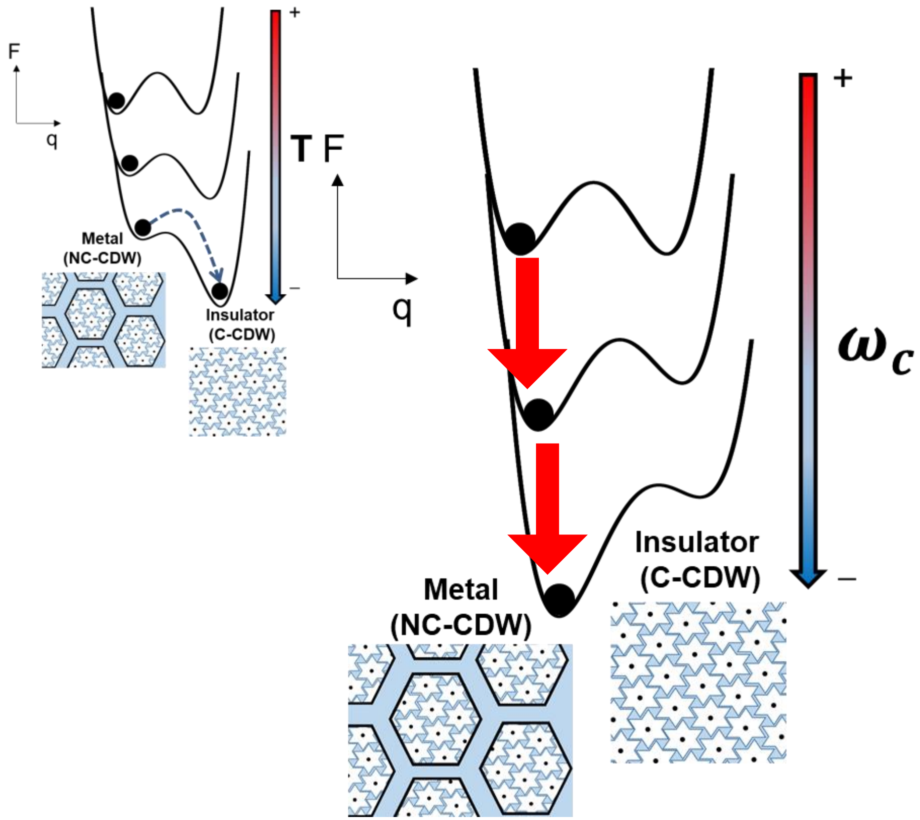

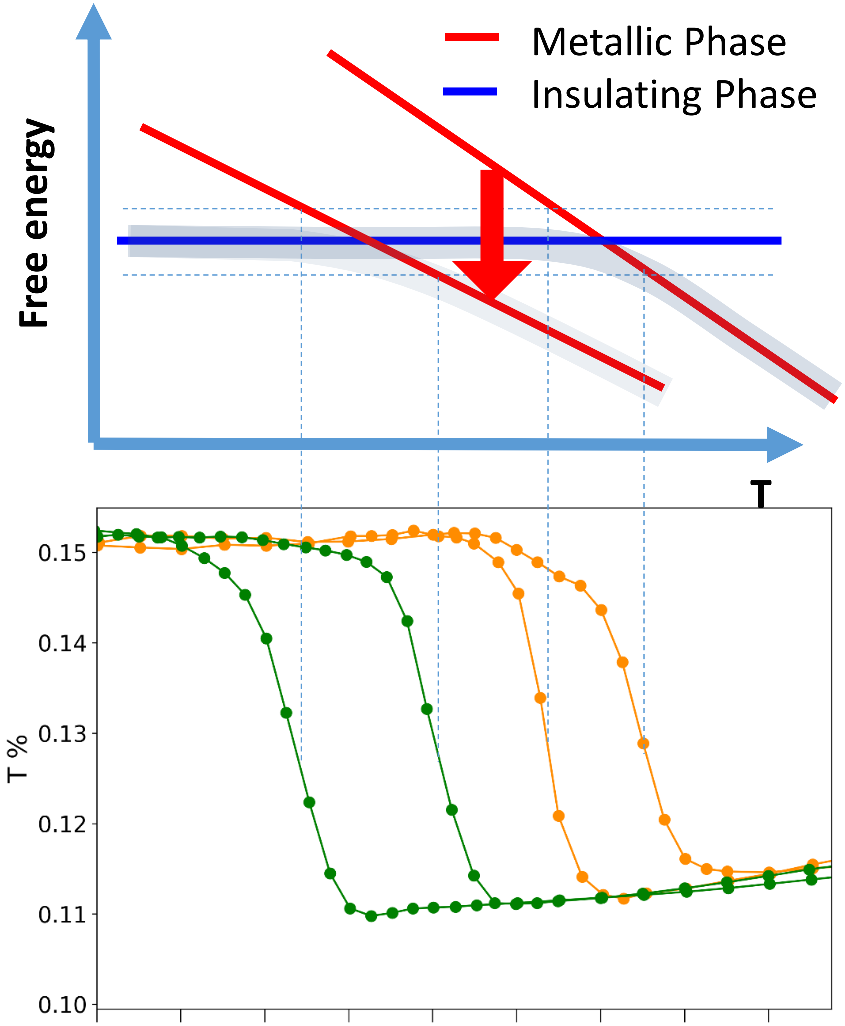

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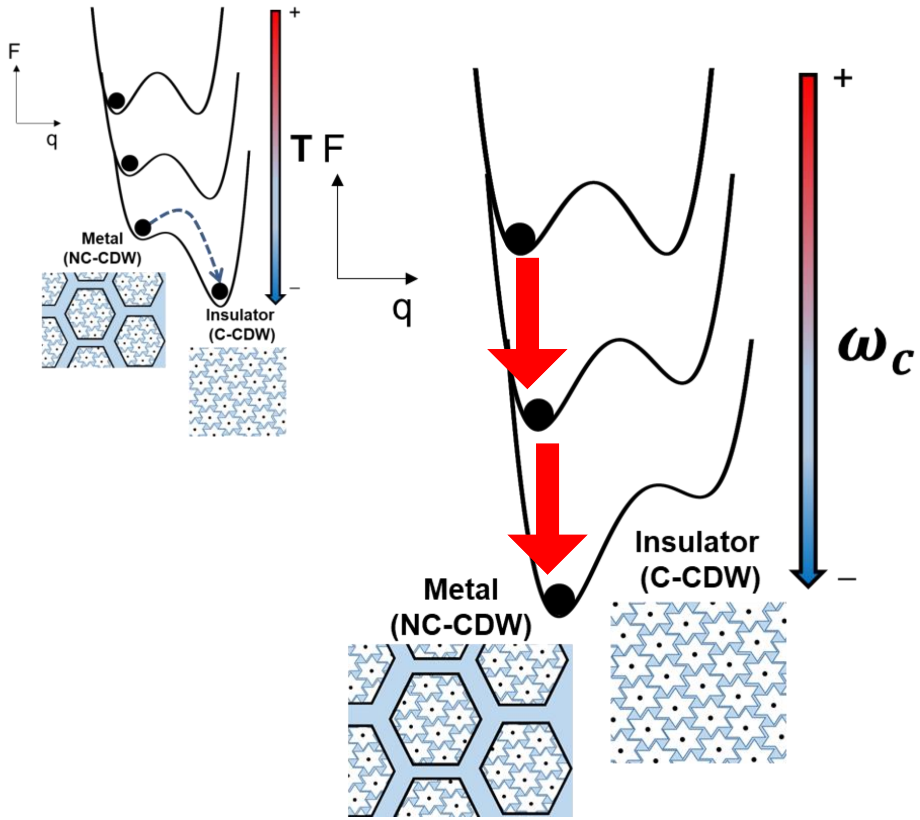

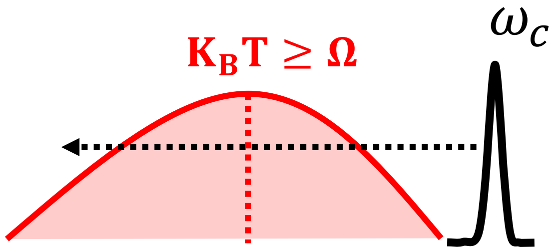

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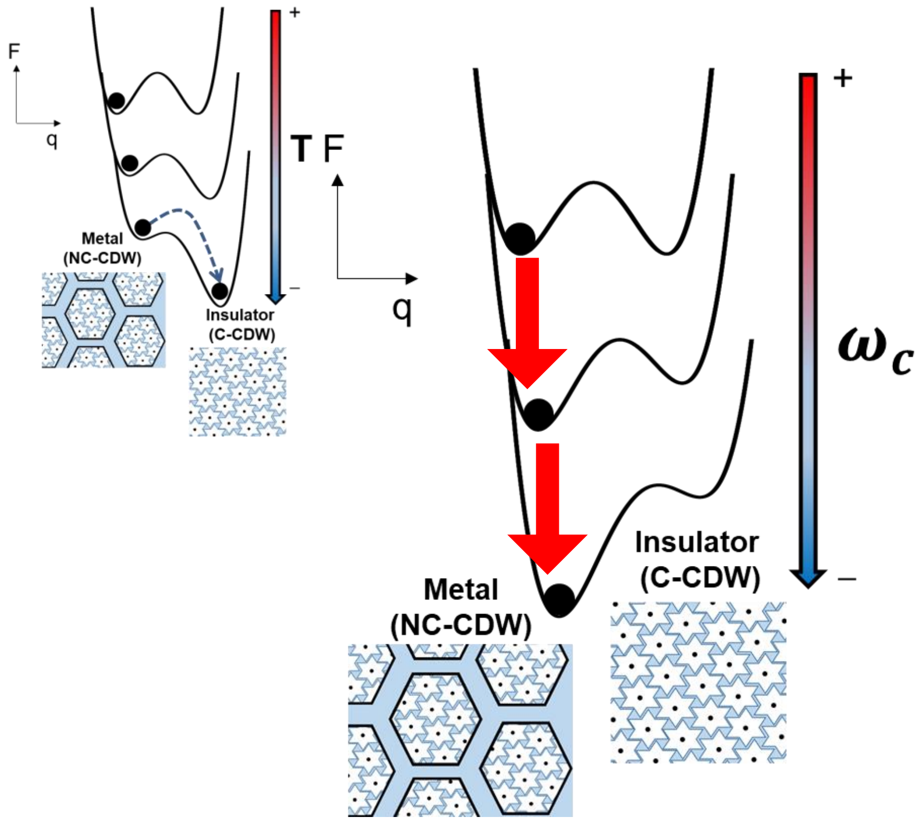
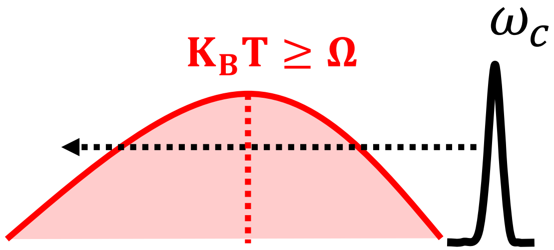
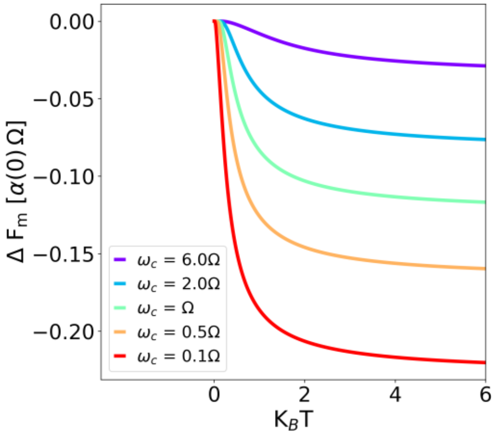




Tunable cavity mode

$+ \longrightarrow \Delta F_m(\omega_c, T)$

Broad Mode
(domain wall fluctuations)



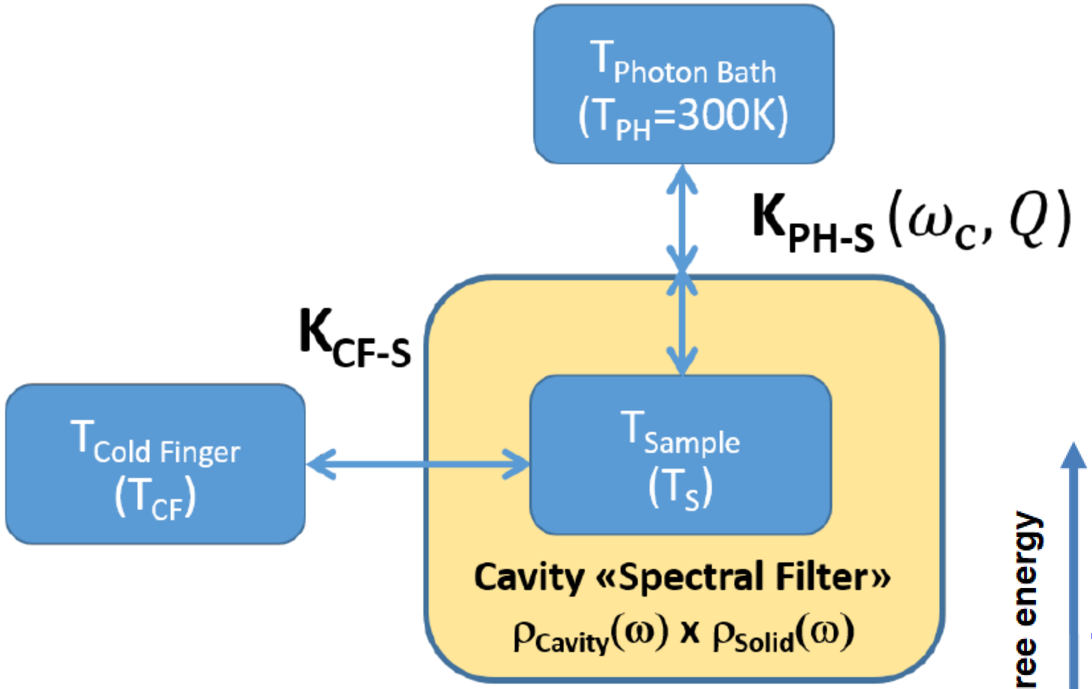
PRB 14, 1496 (1976), J.Phys_Soc_Jap 45,4 (1978)

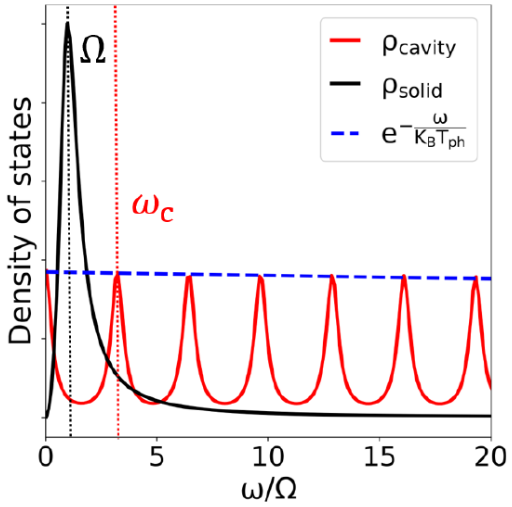
https://arxiv.org/abs/2210.02346

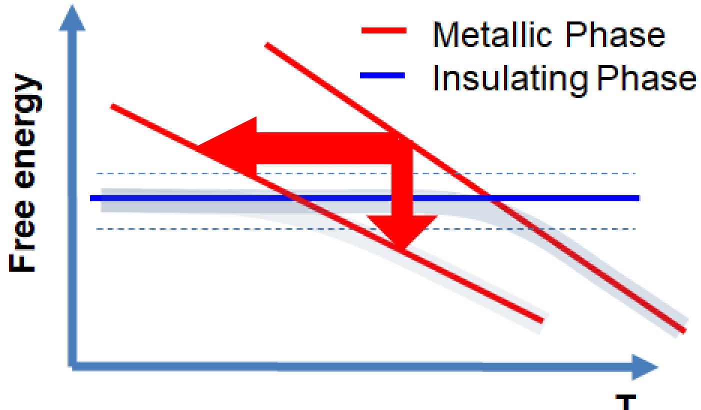
Possible mechanisms #2

→ No cavity modification of the samlpe properties

→ Modification of the temperature (or the population of electronic ex.)







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