Effect of dephasing and local vibrations in the persistence of quantum coherence and emergence of decoherence in molecular junctions

Presented by: Gabriela Valencia Guzmán
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December 6, 2022

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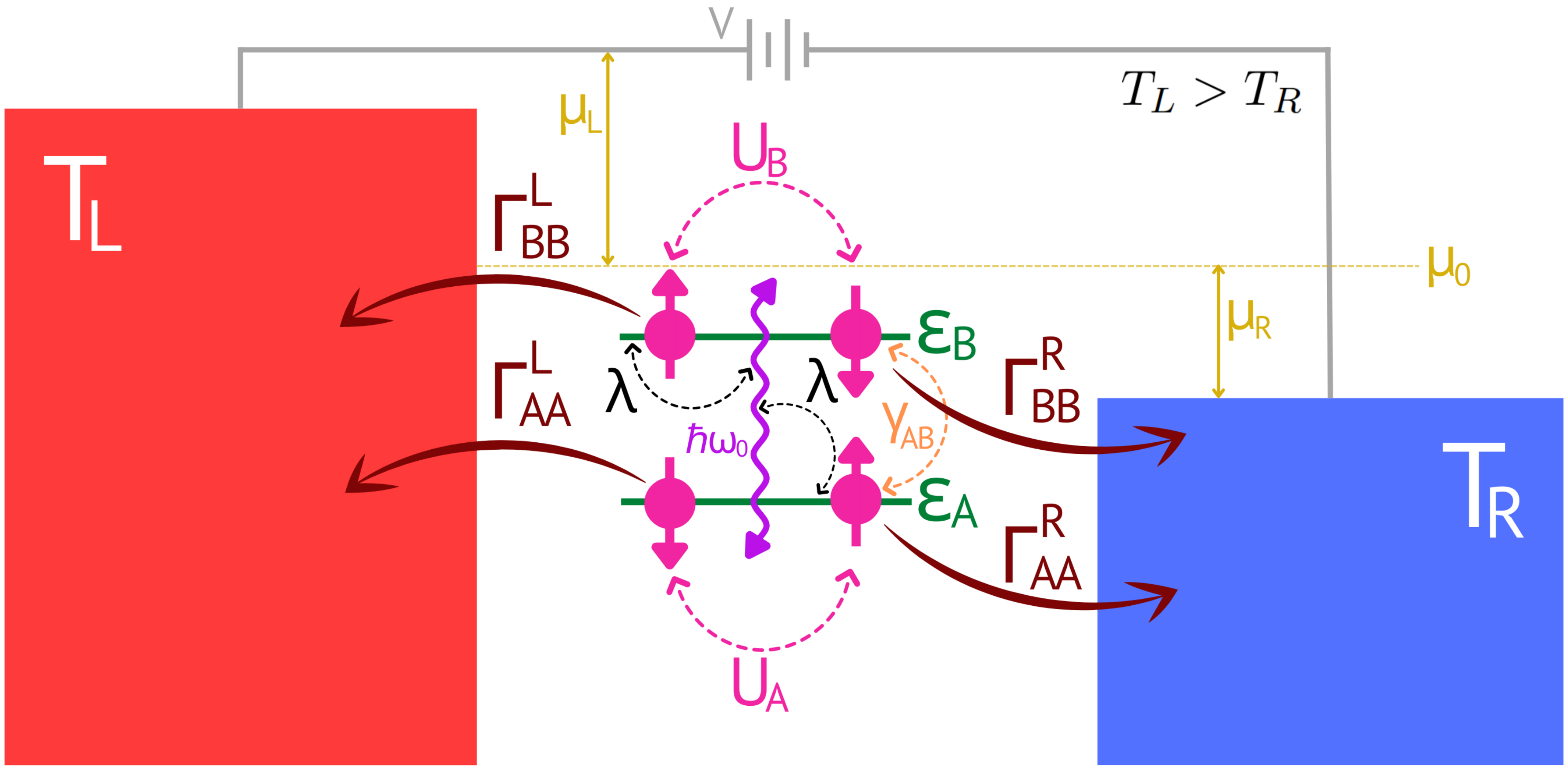
[1] J. D. Vasquez Jaramillo, *Probing Magnetism at the Atomic Scale: Non-Equilibrium Statistical Mechanics Theoretical Treatise* (2018).

What is the physical system?



$$\hat{H}_{juntion} = \hat{H}_{mol} + \sum_{\alpha=1}^{2}\left[\hat{H}^{(\alpha)}_{lead} + \hat{H}^{(\alpha)}_{T}\right] \begin{cases} \underset{(\vec{k}\in\alpha)}{\hat{H}^{(\alpha)}_{Lead}} = \sum_{\vec{k}\sigma} \varepsilon_{\vec{k}\sigma}\hat{C}^{\dagger}_{\vec{k}\sigma}\hat{C}_{\vec{k}\sigma} \\ \hat{H}_T = \sum_{\vec{k}\sigma m}\left(V_{\vec{k}\sigma m\alpha}\hat{C}^{\dagger}_{\vec{k}\sigma\alpha}\hat{d}_{m\sigma} + V^{*}_{\vec{k}\sigma m\alpha}\hat{d}^{\dagger}_{m\sigma}\hat{C}_{\vec{k}\sigma\alpha}\right) \end{cases}$$

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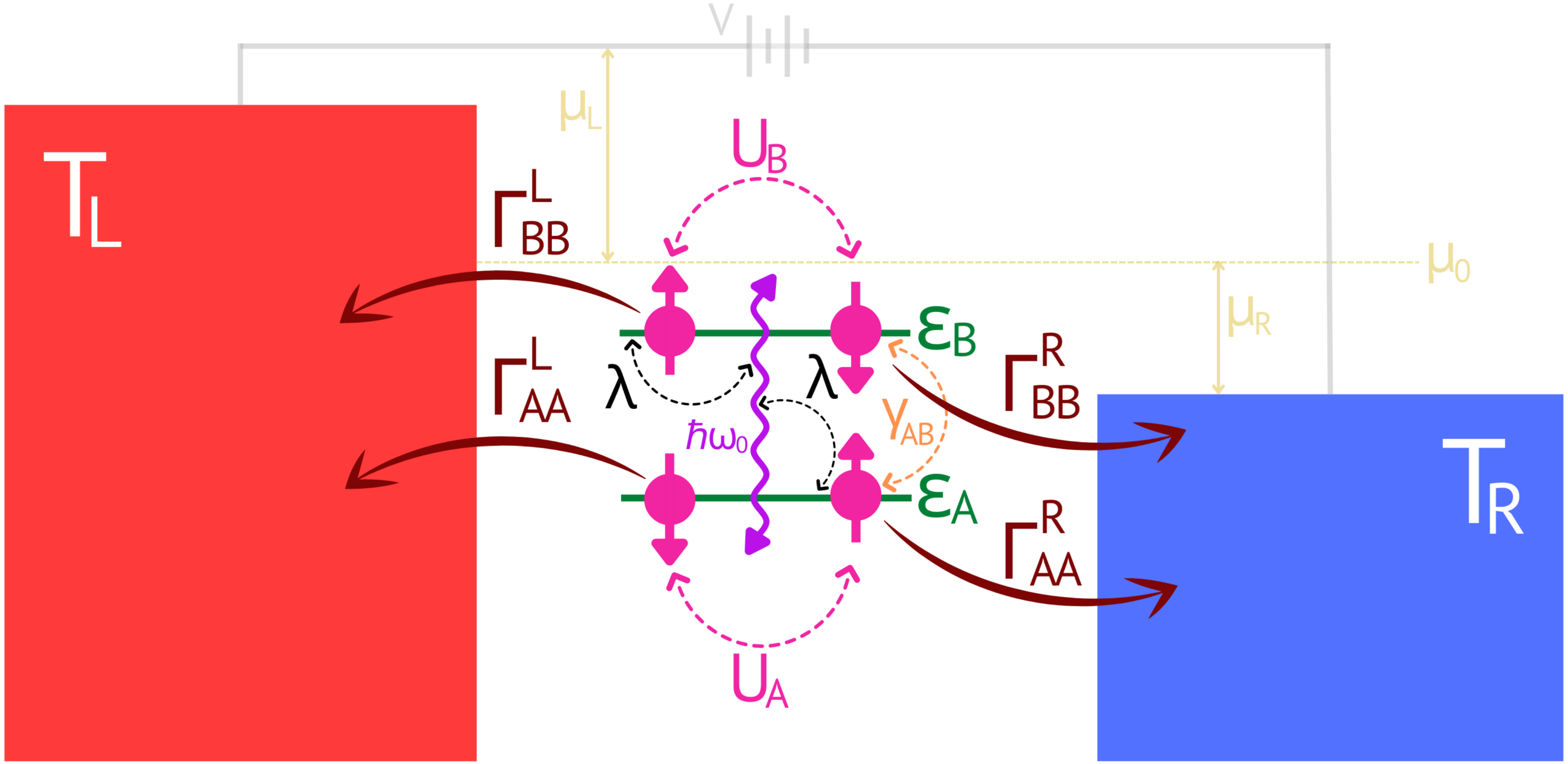

$$\hat{H}_{mol} = \sum_{m\sigma} \varepsilon_{m\sigma} \hat{d}^\dagger_{m\sigma} \hat{d}_{m\sigma} + \sum_m U_m \hat{n}_{m\uparrow} \hat{n}_{m\downarrow} + \sum_{mn\sigma} \gamma_{mn} \hat{d}^\dagger_{m\sigma} \hat{d}_{n\sigma} + \underbrace{\lambda \sum_{m\sigma} \hat{d}^\dagger_{m\sigma} \hat{d}_{m\sigma} (\hat{a}^\dagger + \hat{a})}_{\text{Fermion-Boson Coupling}}$$

$(m = A, B)$

$$+\hbar\omega_0 \left(\hat{a}^\dagger \hat{a} + \frac{1}{2} \right) \longrightarrow \text{Boson Field Hamiltonian}$$

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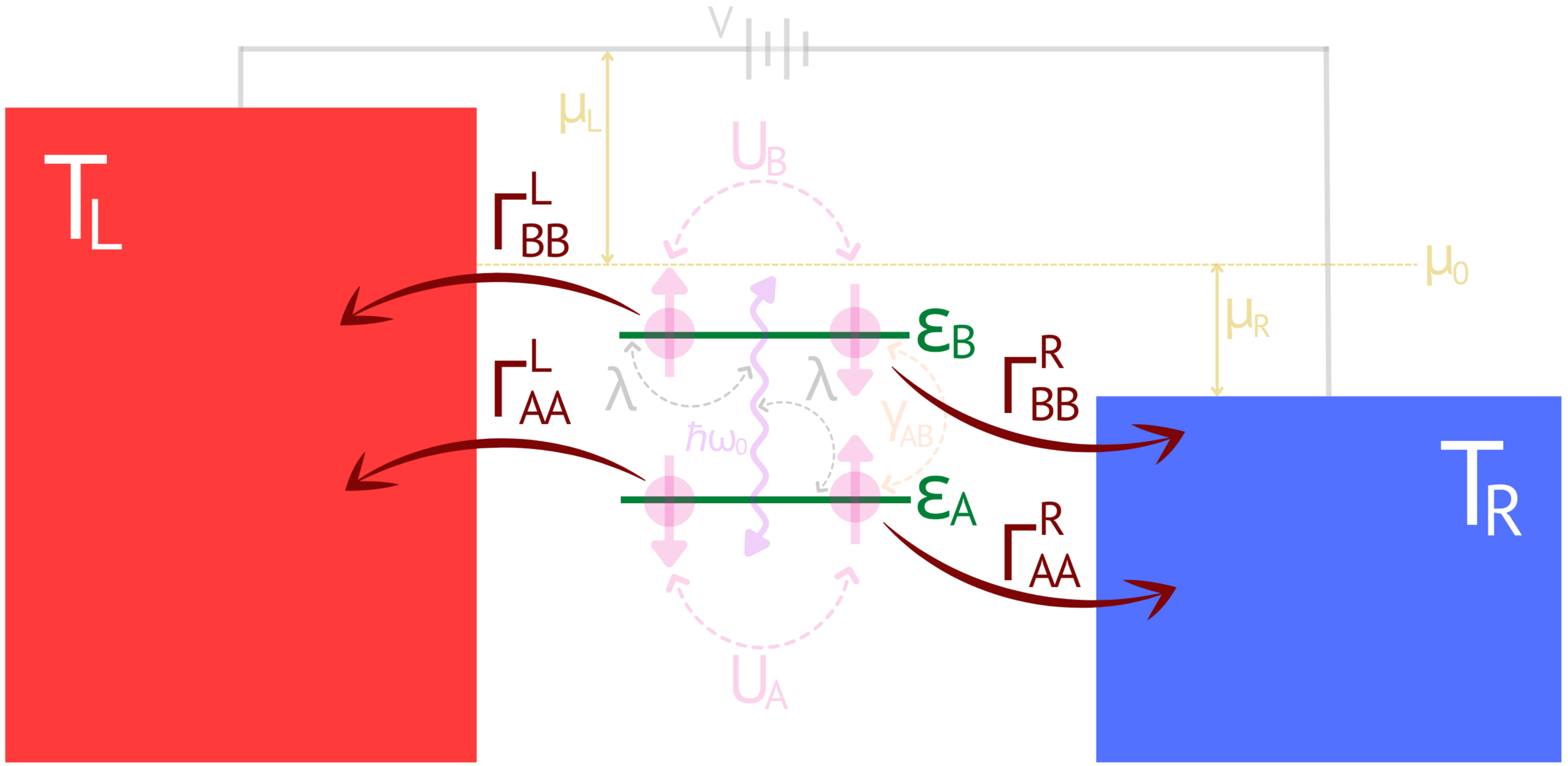

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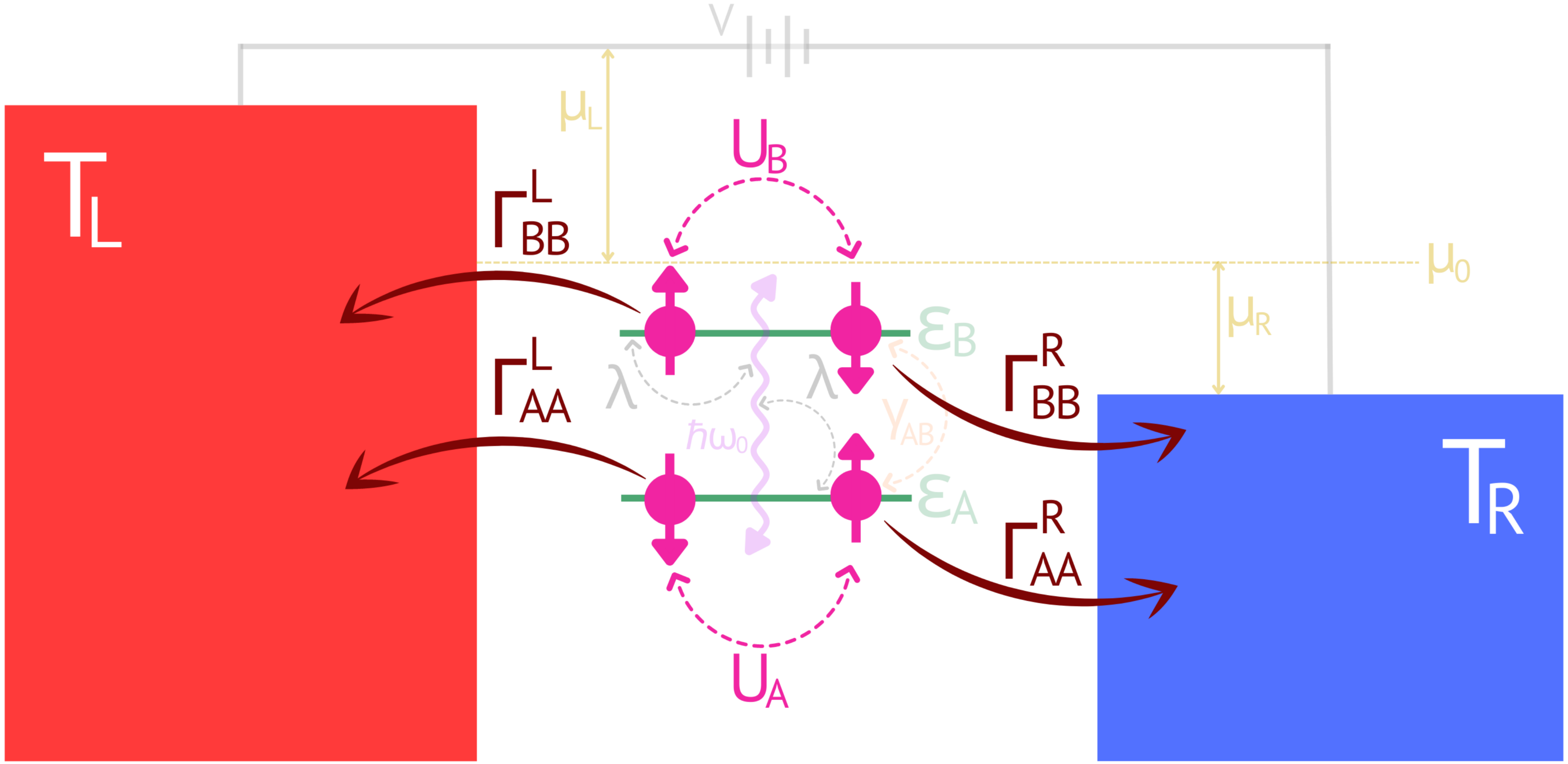

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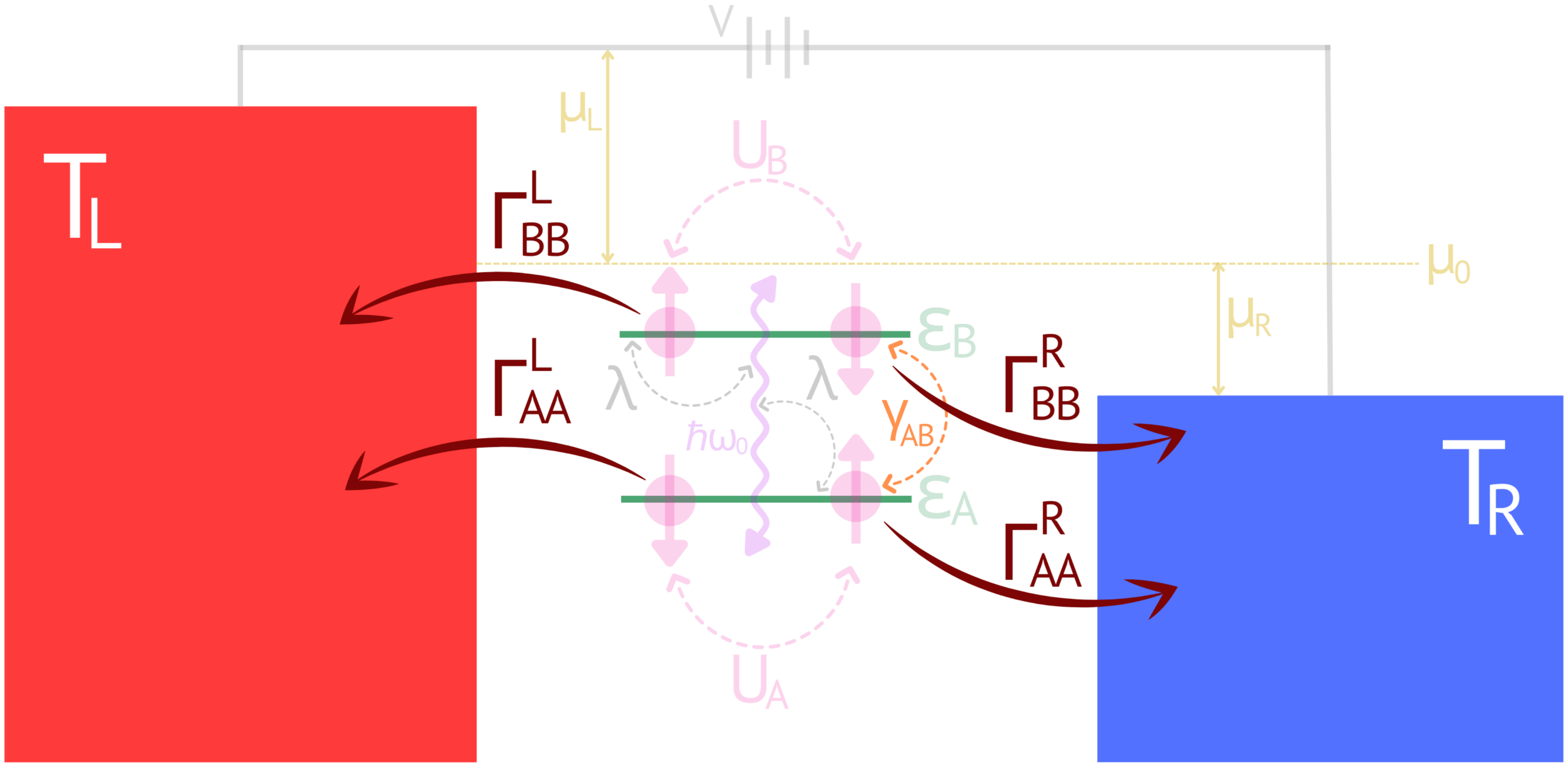

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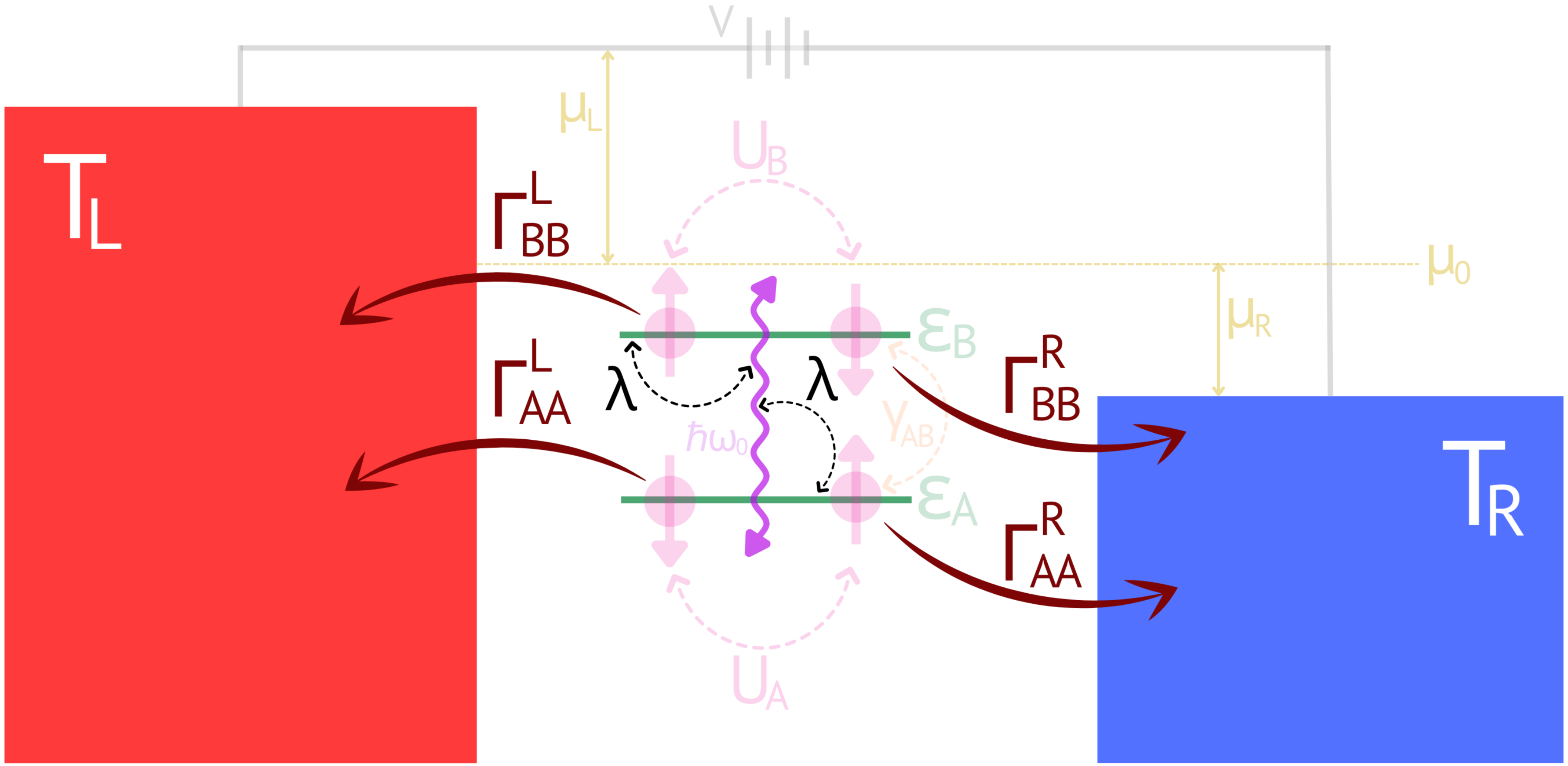

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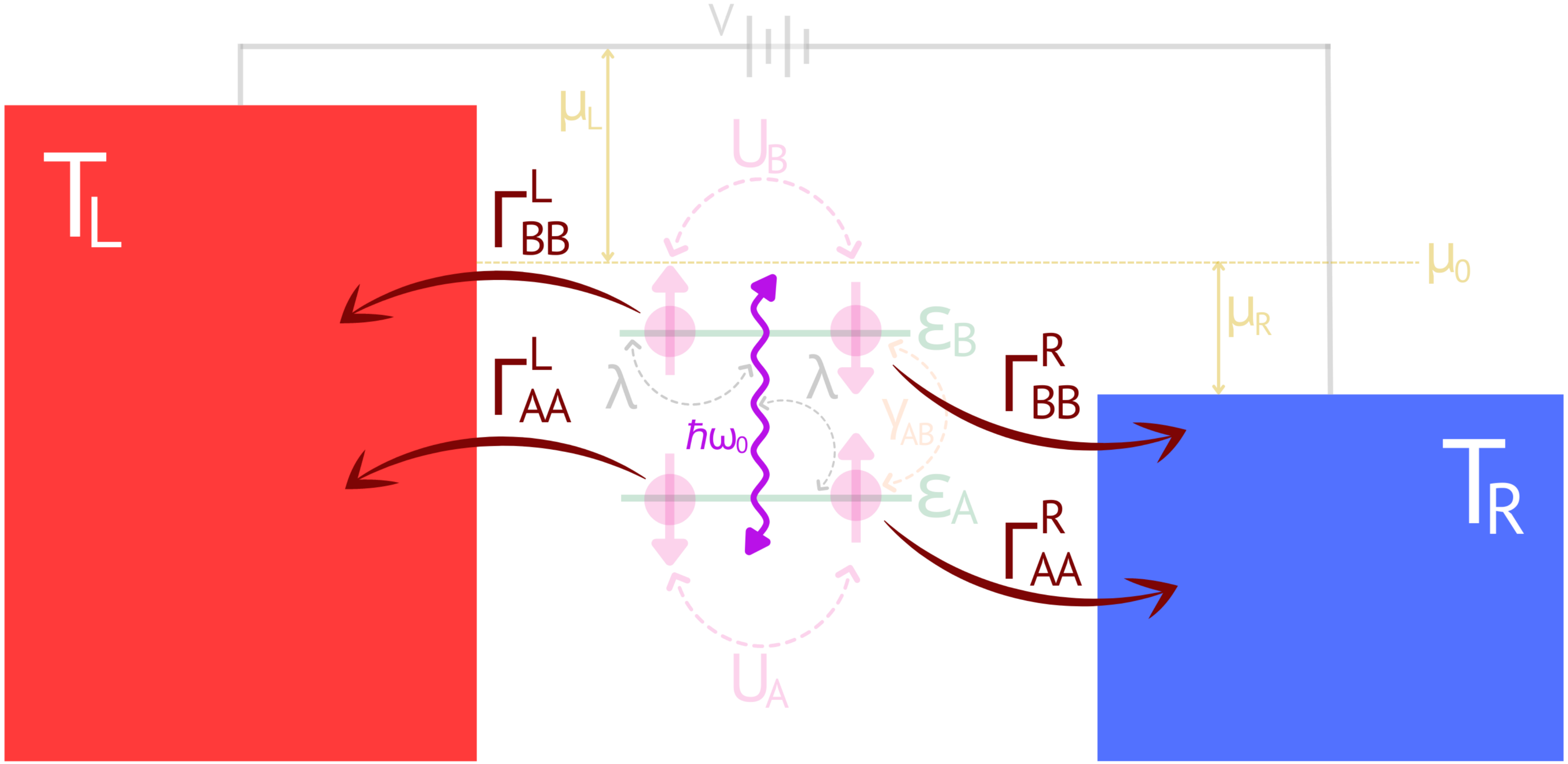

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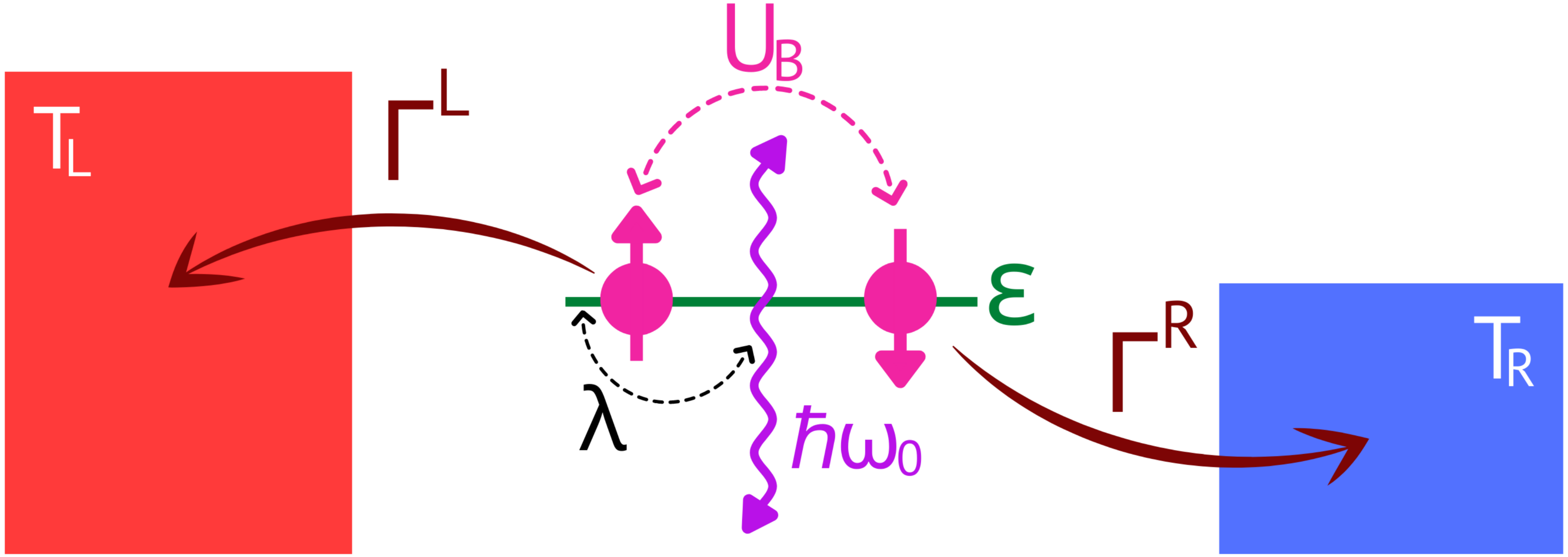
One single energy level physical system



Using the Baker-Campbell-Hausdorff identity, the term of the Hamiltonian containing the Fermion-Boson coupling is eliminated. After the process, the system constants have changed:

$$\overline{\hat{H}_{mol}} = \sum_{\sigma} \overline{\varepsilon_\sigma} \hat{d}'^{\dagger}_{\sigma} \hat{d}'_{\sigma} + \overline{U} \hat{n}'_{\uparrow} \hat{n}'_{\downarrow} + \hbar\omega_0 \hat{a}^{\dagger} \hat{a} \longrightarrow \overline{\varepsilon_\sigma} = \varepsilon_\sigma - \frac{\lambda^2}{\hbar\omega_0} \text{ and } \overline{U} = U - 2\frac{\lambda^2}{\hbar\omega_0}$$

Important: How does transformation affect operators?

$$\hat{d}'_\sigma = \hat{d}_\sigma \hat{\chi} \longrightarrow \underbrace{\hat{\chi} = e^{-\frac{\lambda}{\hbar\omega_0}(\hat{a}^{\dagger} - \hat{a})}}_{\text{Moment traslation operator}}$$

What about Green's Functions?

$$G(t,t') = \frac{-i}{\hbar}\left\langle \hat{T}_\tau \hat{d}_\sigma(t)\hat{d}^\dagger_{\sigma'}(t')\right\rangle \longrightarrow \text{Electron + Vibrational}$$

$$G(t,t') = \frac{-i}{\hbar}\left\langle \hat{T}_\tau \overline{\hat{d}}_\sigma(t)\hat{\chi}(t)\overline{\hat{d}^\dagger}_{\sigma'}(t')\hat{\chi}(t')\right\rangle \longrightarrow \text{So difficult}$$

$$\xrightarrow{\text{approximation}} \quad G(t,t') = \underbrace{\frac{-i}{\hbar}\left\langle \hat{T}_\tau \overline{\hat{d}}_\sigma(t)\overline{\hat{d}^\dagger}_{\sigma'}(t')\right\rangle}_{\text{Electronic Green's Function}} \underbrace{\left\langle \hat{\chi}(t)\hat{\chi}(t')\right\rangle}_{A(t,t')}$$

Vibrational Correction

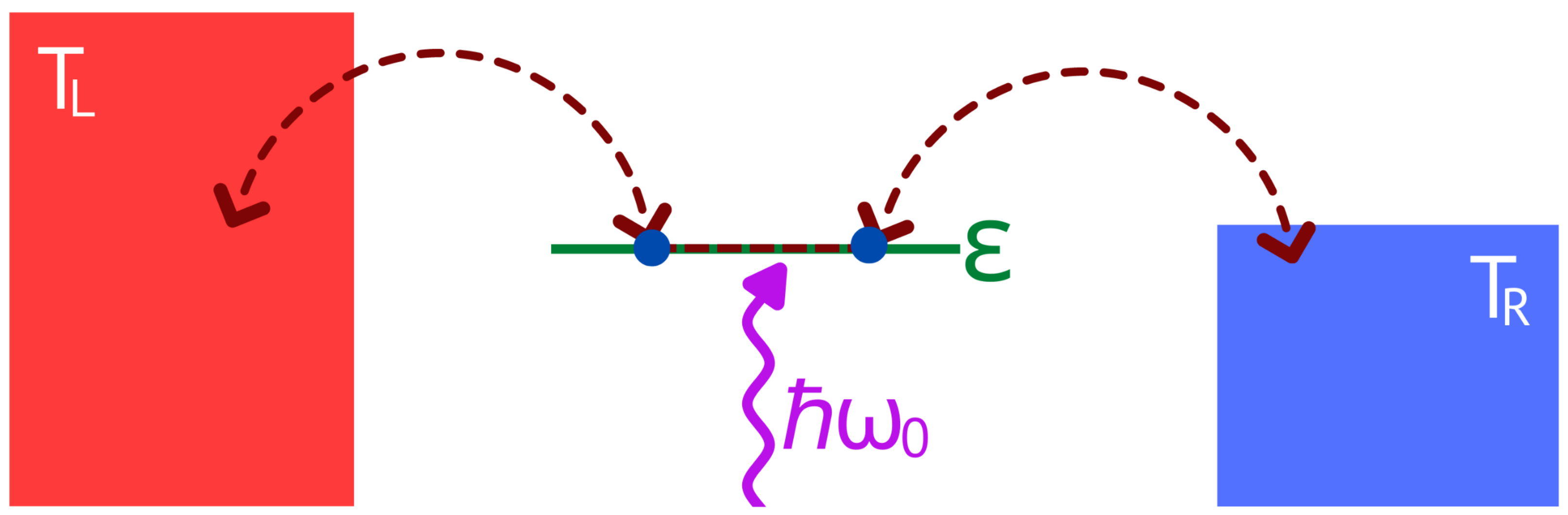
In the weak coupling limit $(\lambda < \hbar\omega_0)$

$$G_{\sigma\sigma'}(\omega) = e^{-\left(\frac{\lambda}{\hbar\omega_0}\sqrt{1+2n_B(\omega_0)}\right)^2} \sum_{n=-\infty}^{+\infty} e^{-n\omega_0\beta\hbar/2} I_n\left(2\left(\frac{\lambda}{\hbar\omega_0}\right)^2 \sqrt{n_B(\omega_0)\left(n_B(\omega_0)+1\right)}\right) \overline{G_{\sigma\sigma'}}\left(\omega\tau + n\omega_0\right)$$

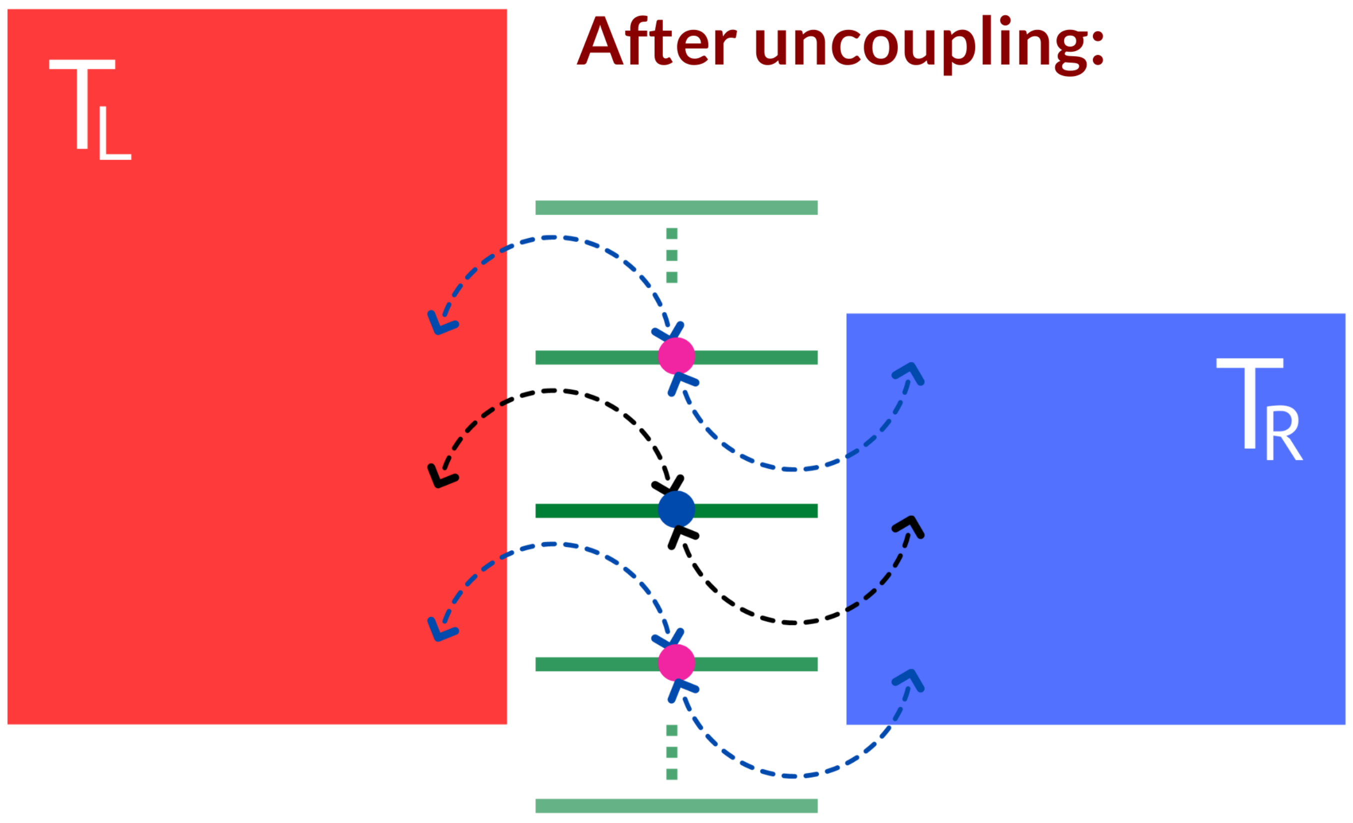
$$\tau = t - t', \quad n_B(\omega_0) = \frac{1}{e^{\beta\hbar\omega_0} - 1} \longrightarrow \text{Bose-Einstein Distribution}$$

What is the effect of vibration on an energy level?

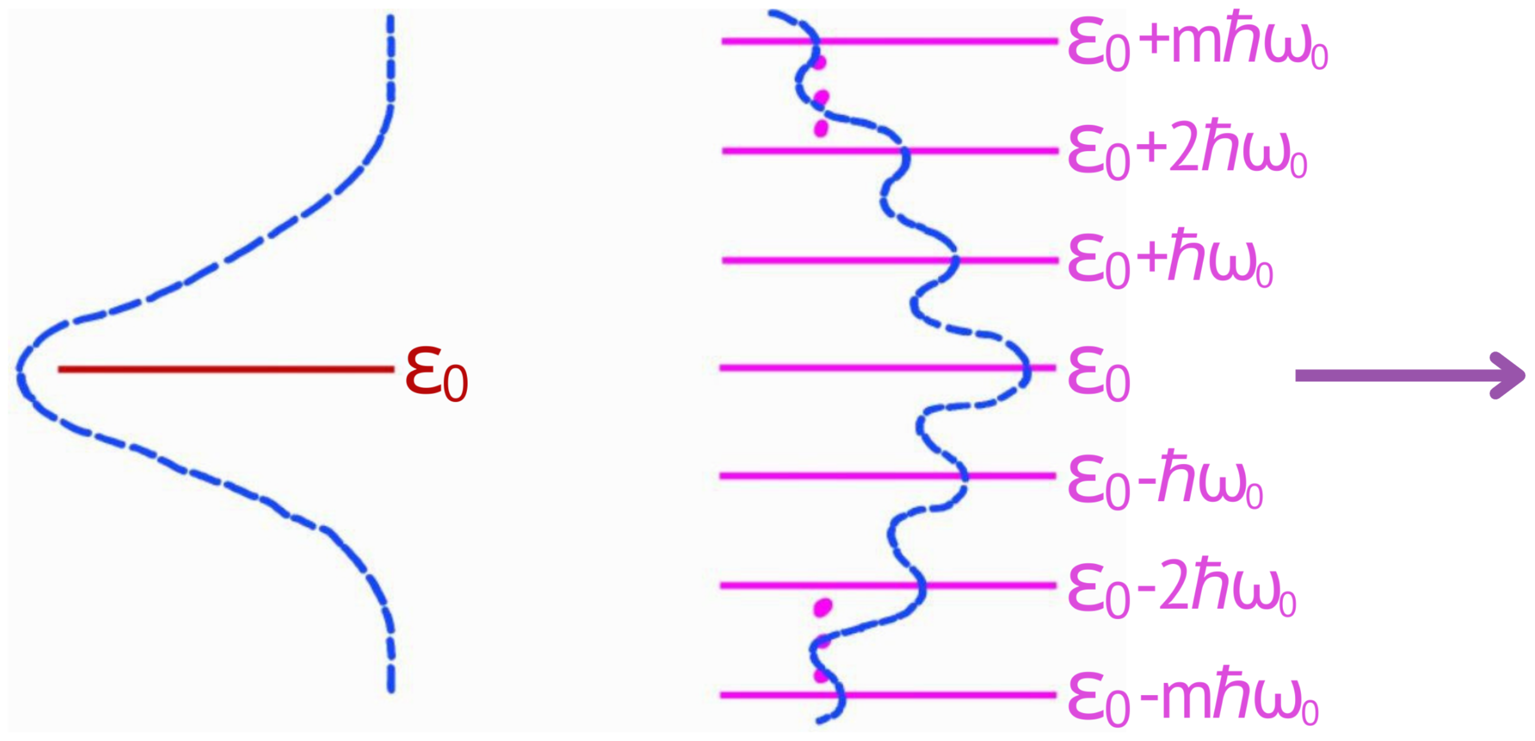
Before uncoupling:

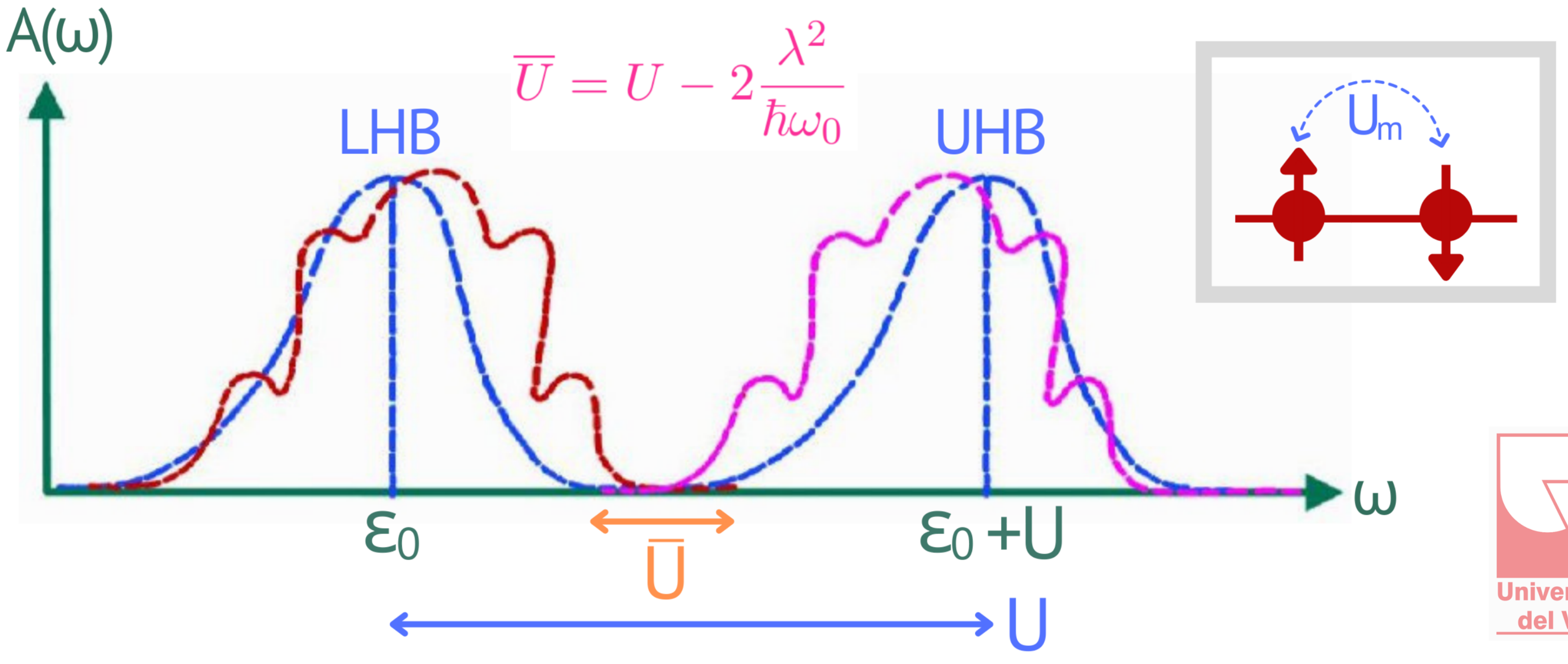


After uncoupling:

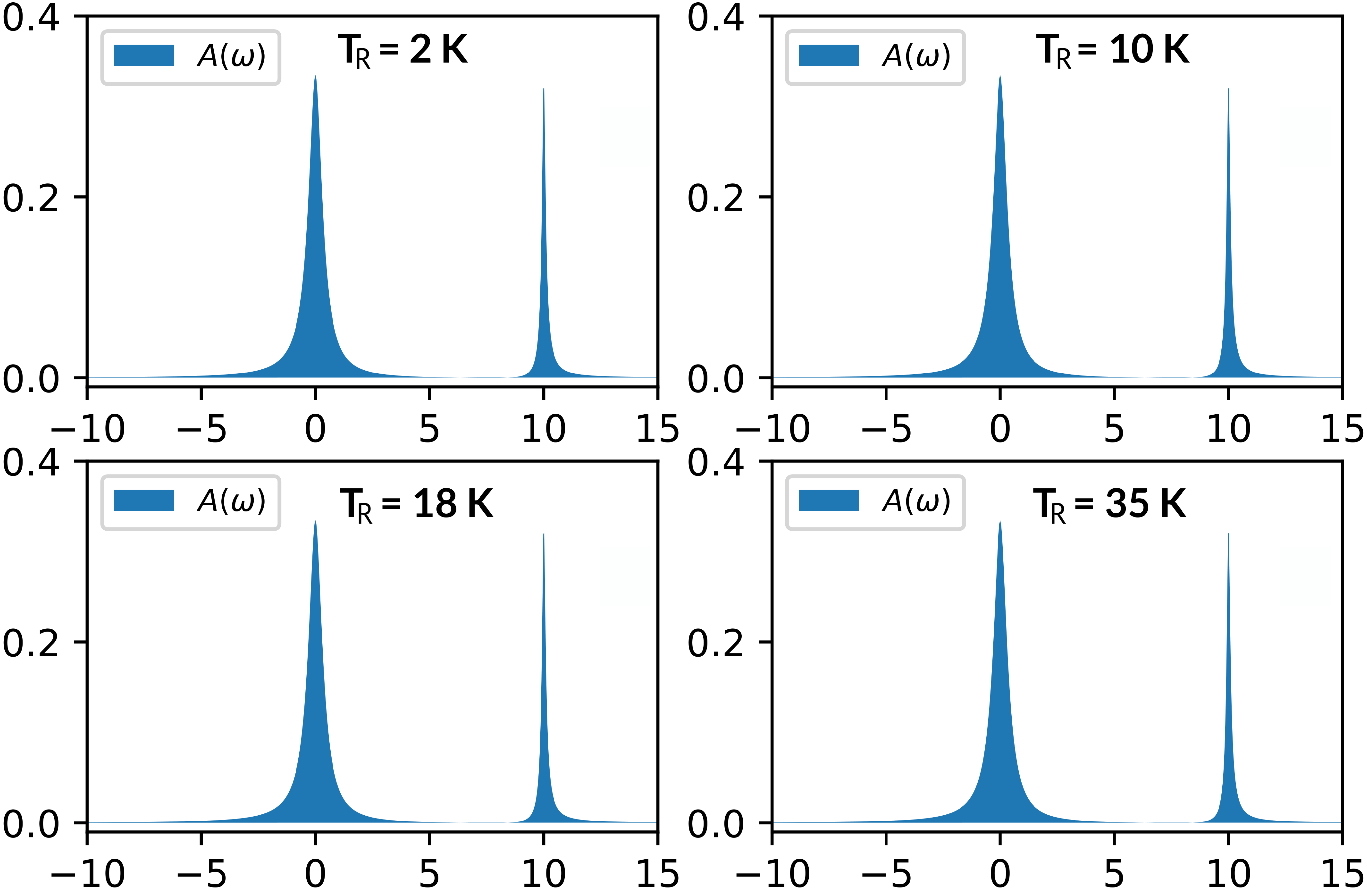


Eliminating Electron-Vibration coupling is equivalent to multiplying the energy level. In that case, how is the density of states affected?



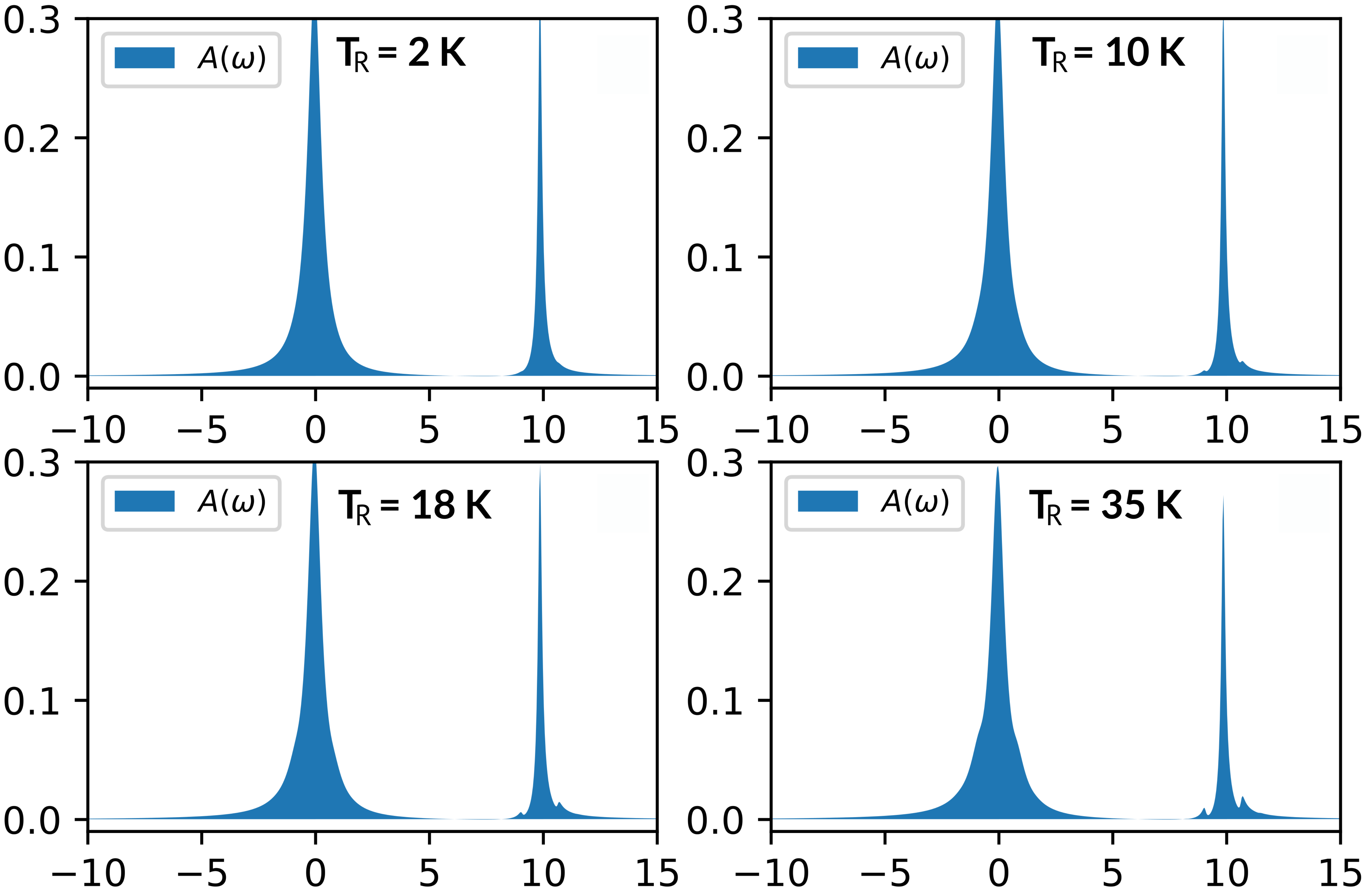


Density of states for an energy level ($\hbar\omega_0$ = 0.8 meV, U = 10 meV, λ = 0 meV, T_L = 3 K)



Taken from code made by Juan David V. Jaramillo and Pedro Luis Artunduaga.

Density of states for an energy level ($\hbar\omega_0$ = 0.8 meV, U = 10 meV, λ = 0.2 meV, T_L = 3 K)



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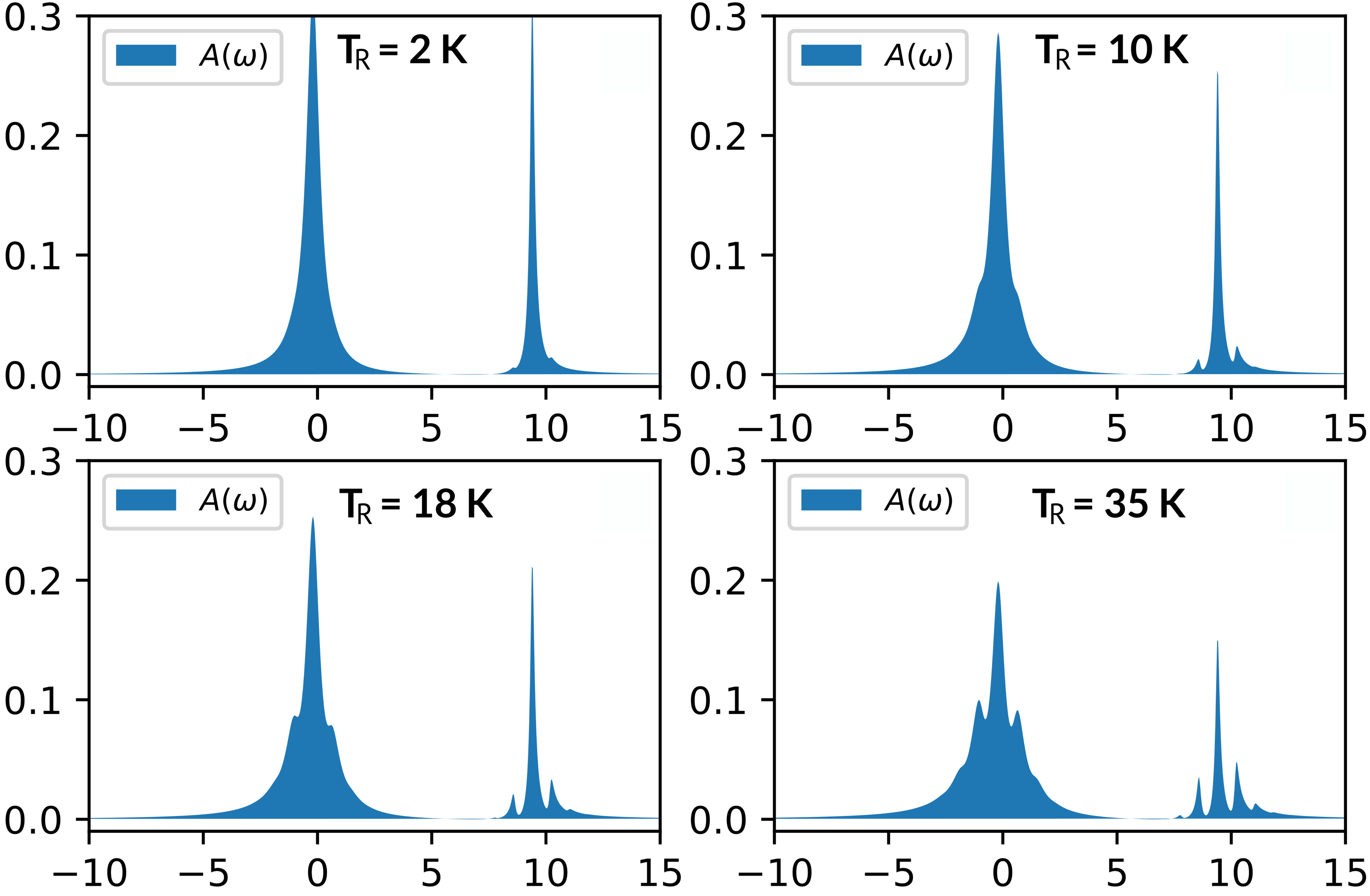
Density of states for an energy level ($\hbar\omega_0$ = 0.8 meV, U = 10 meV, λ = 0.4 meV, T_L = 3 K)



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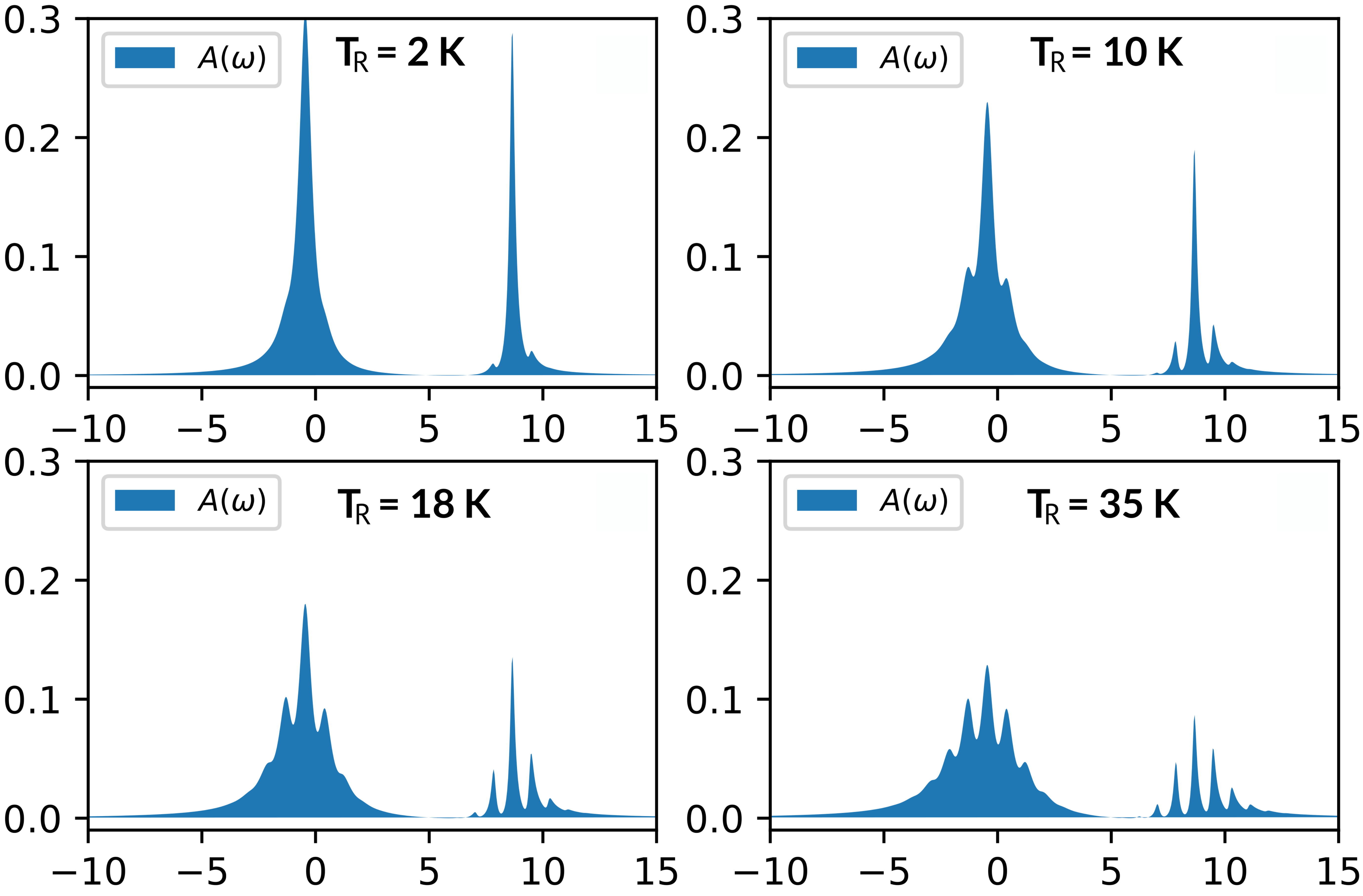
Density of states for an energy level ($\hbar\omega_0$ = 0.8 meV, U = 10 meV, λ = 0.6 meV, T_L = 3 K)



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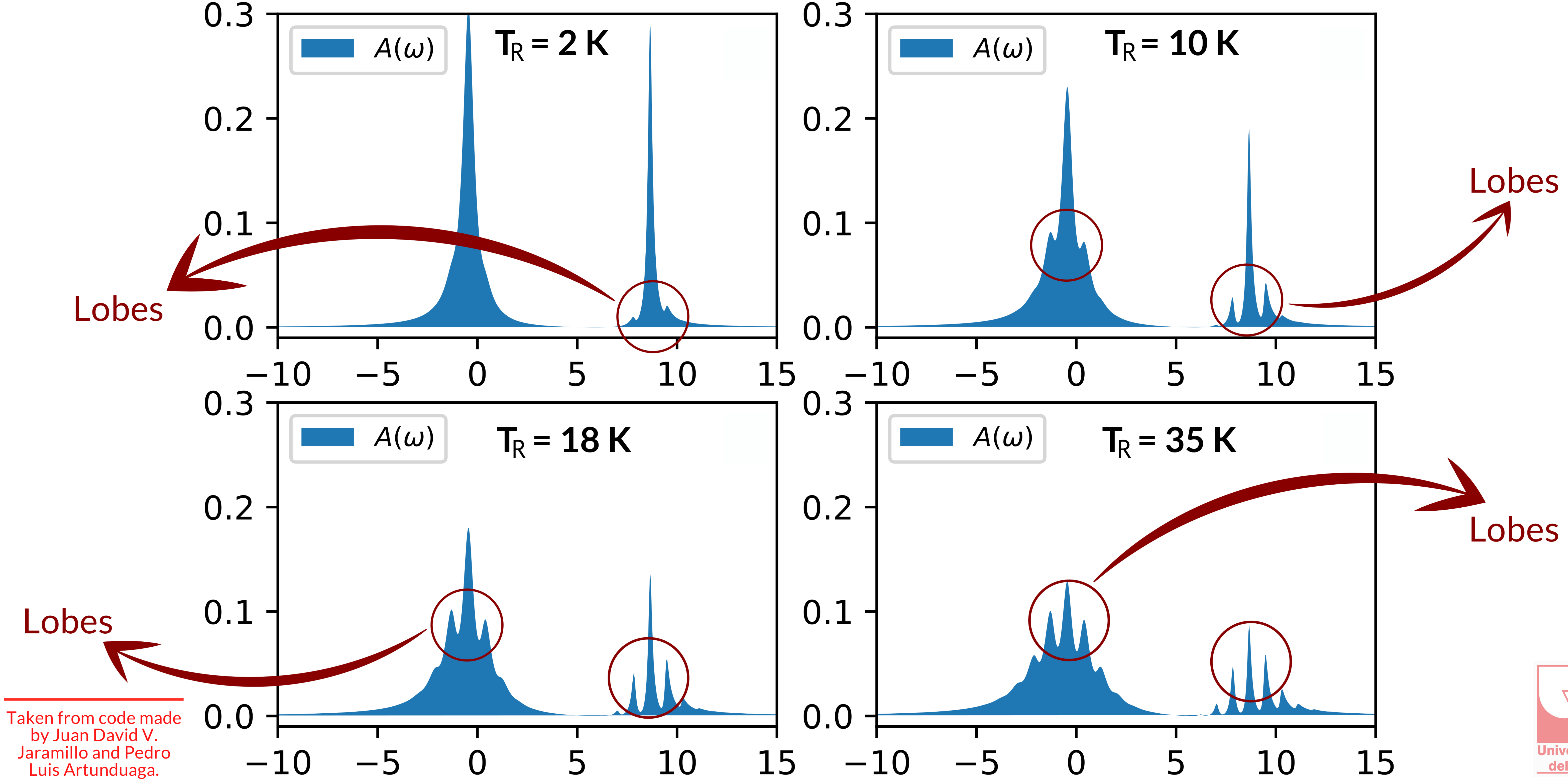

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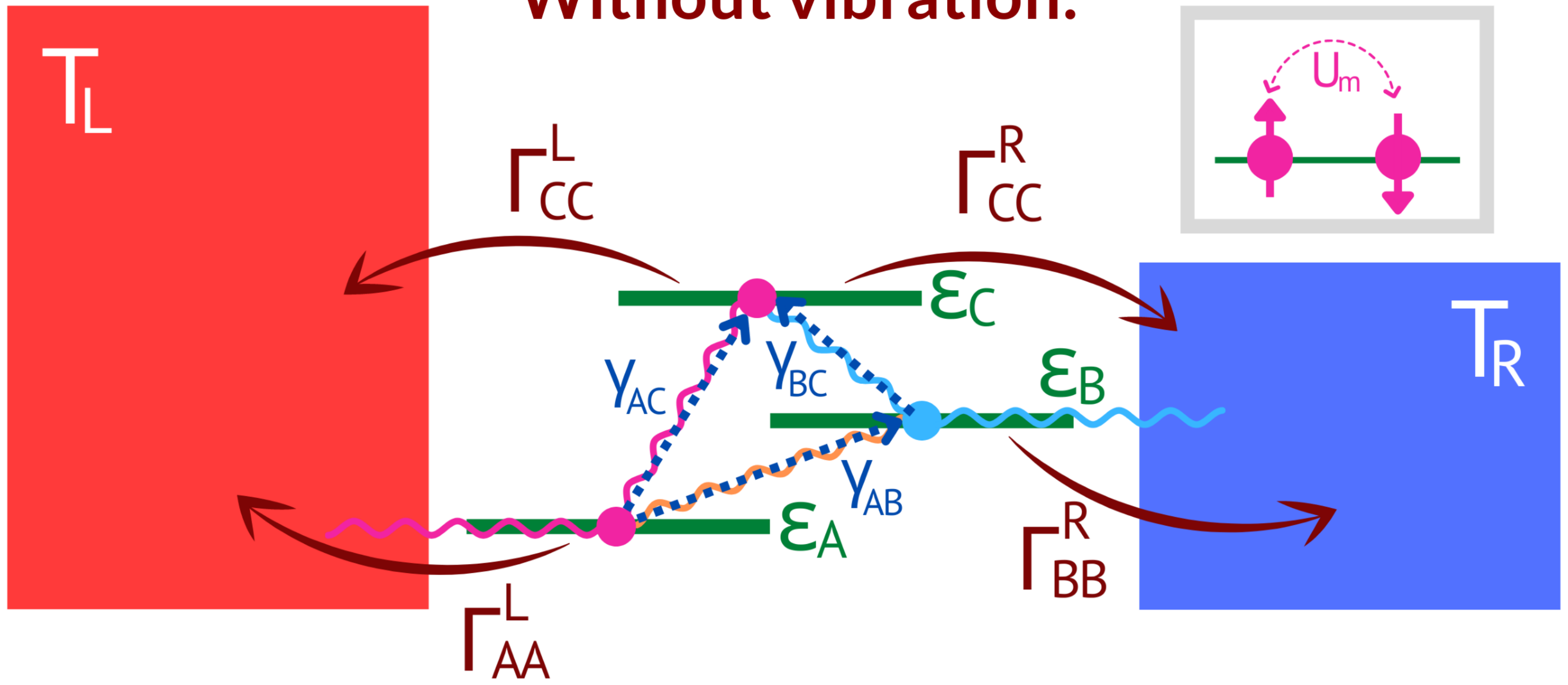
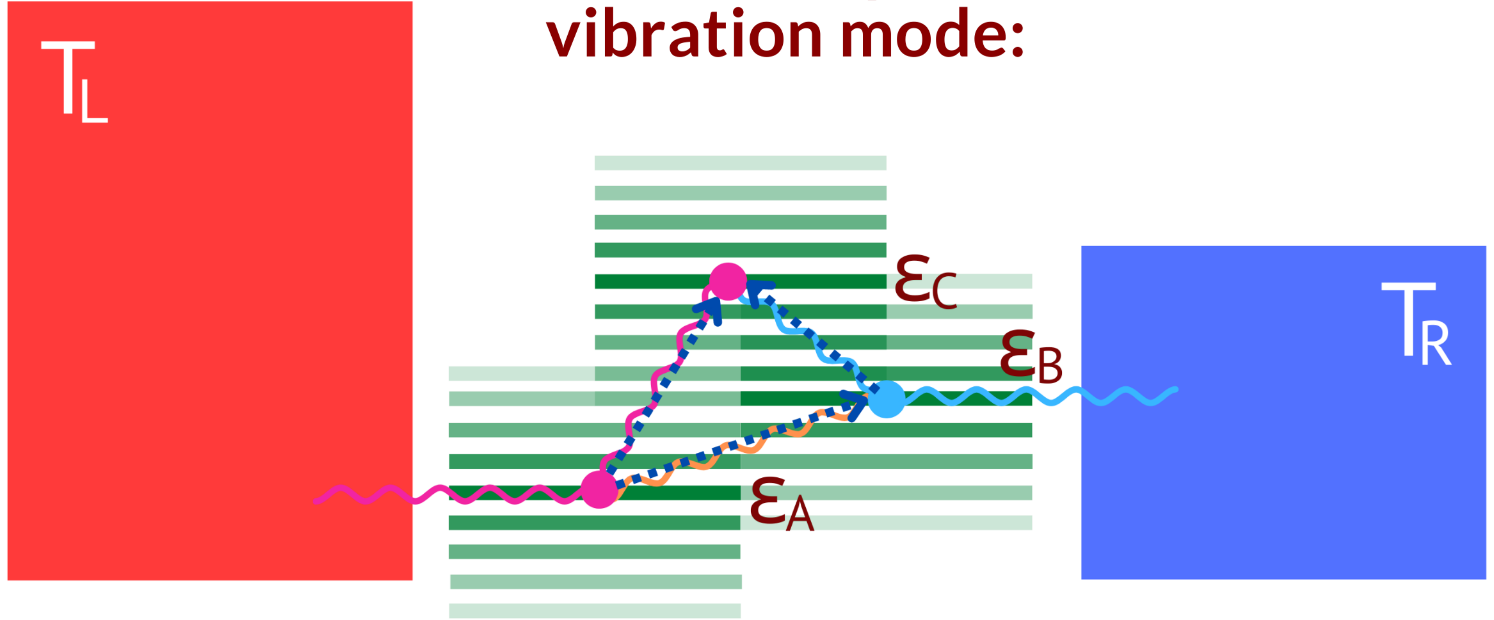
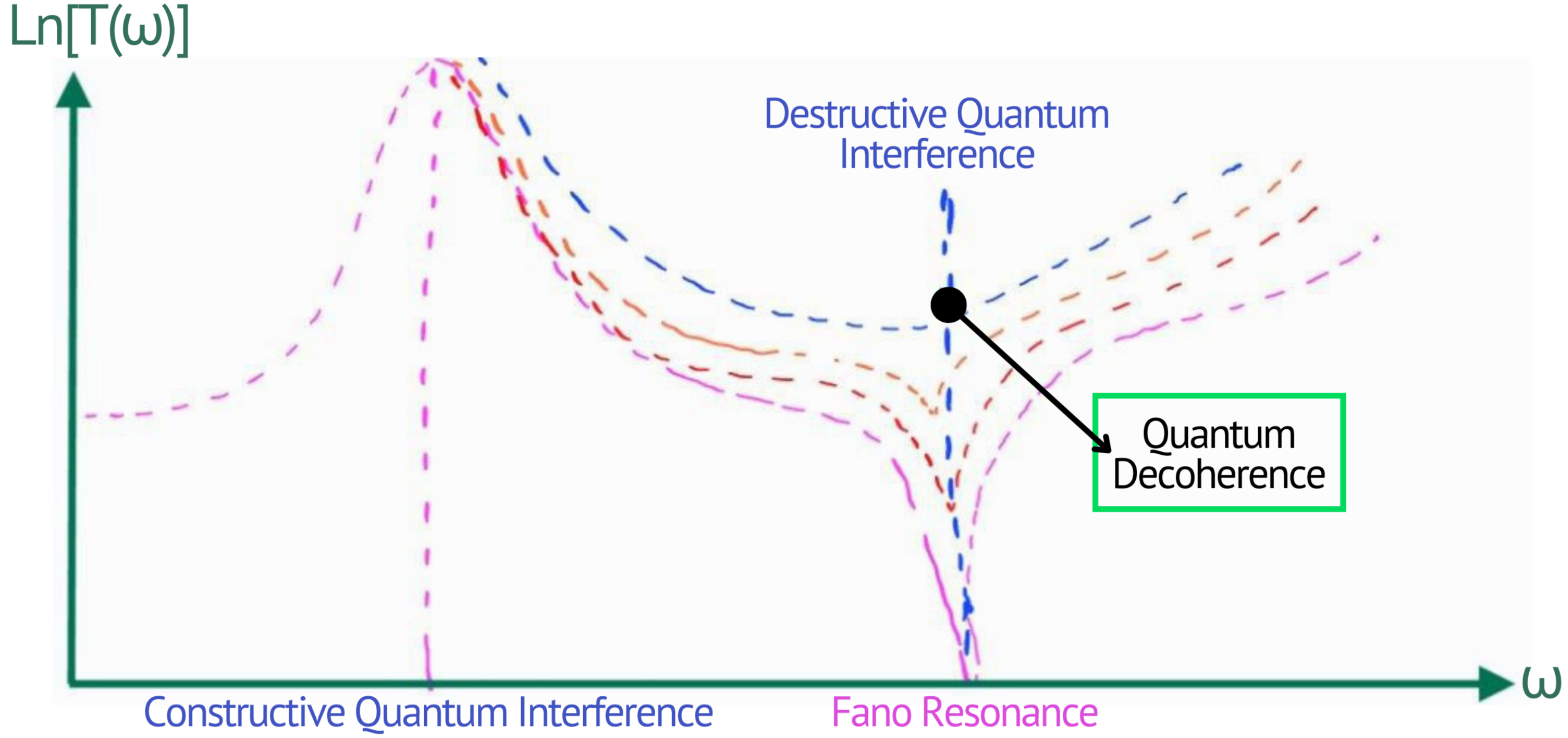
What happens for multilevel molecules? (Three Level System)

Without vibration:



After uncouple the vibration mode:





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[2] K. G. Pedersen, et al., *“Illusory Connection between Cross-Conjugation and Quantum Interference”*, Journal of Physical Chemistry C, (2015).
[3] D. Lovey and R. Romero, *“Conductance and electronic structure of conjugated organic molecules”*, Anales AFA, (2013).

The end

Thank you very much for your attention. Questions?

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