Model Hamiltonian

$$H = \varepsilon_1 n_1 + \varepsilon_2 n_2 + U_1 n_{1\uparrow} n_{1\downarrow} + U_2 n_{2\uparrow} n_{2\downarrow}$$
Orbital 1 and 2 with strong intra-level Coulomb repulsion $(\varepsilon_1 \le \varepsilon_2)$

$$+U_{12} n_1 n_2$$
Inter-level Coulomb repulsion

$$+\omega_0 b^\dagger b$$
Local phonon mode

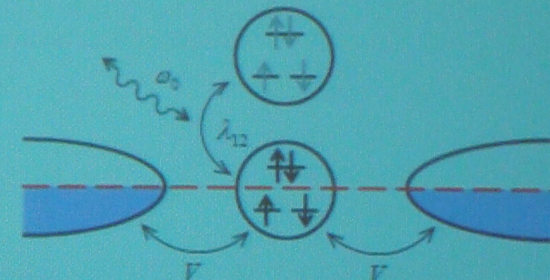
$$+\lambda_{12}(b^\dagger + b)\sum_\sigma (d_{1\sigma}^\dagger d_{2\sigma} + d_{2\sigma}^\dagger d_{1\sigma})$$
Phonon-assisted inter-orbital tunneling

$$+\sum_{k,\sigma} \varepsilon_k c_{k\sigma}^\dagger c_{k\sigma}$$
Leads

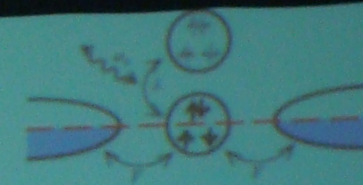
$$+\sum_{j=1,2} V_j \sum_{k,\sigma} (c_{k\sigma}^\dagger d_{j\sigma} + d_{j\sigma}^\dagger c_{k\sigma})$$
Tunneling between molecule and leads

Regime of interest

$$U_j > |\varepsilon_j|, \omega_0 \gg \Gamma_j = 2\pi \rho(E_F) V_j^2$$



E-e and e-ph competition



Define: $d_{e\sigma} = (d_{1\sigma} + d_{2\sigma}) / \sqrt{2}$ and $\tilde{b} = b - (\lambda / \omega_0)(n_e - n_o)$

$d_{o\sigma} = (d_{1\sigma} - d_{2\sigma}) / \sqrt{2}$

Terms involving phonon mode can be written as:

$$\omega_0 b^\dagger b + \lambda_{12}(b^\dagger + b)\sum_\sigma (d_{1\sigma}^\dagger d_{2\sigma} + d_{2\sigma}^\dagger d_{1\sigma}) = \omega_0 b^\dagger b + \lambda_{12}(b^\dagger + b)(n_e - n_o)$$

$$= \omega_0 \tilde{b}^\dagger \tilde{b} - \left(\lambda_{12}^2/\omega_0\right)\left(n_e - n_o\right)^2$$

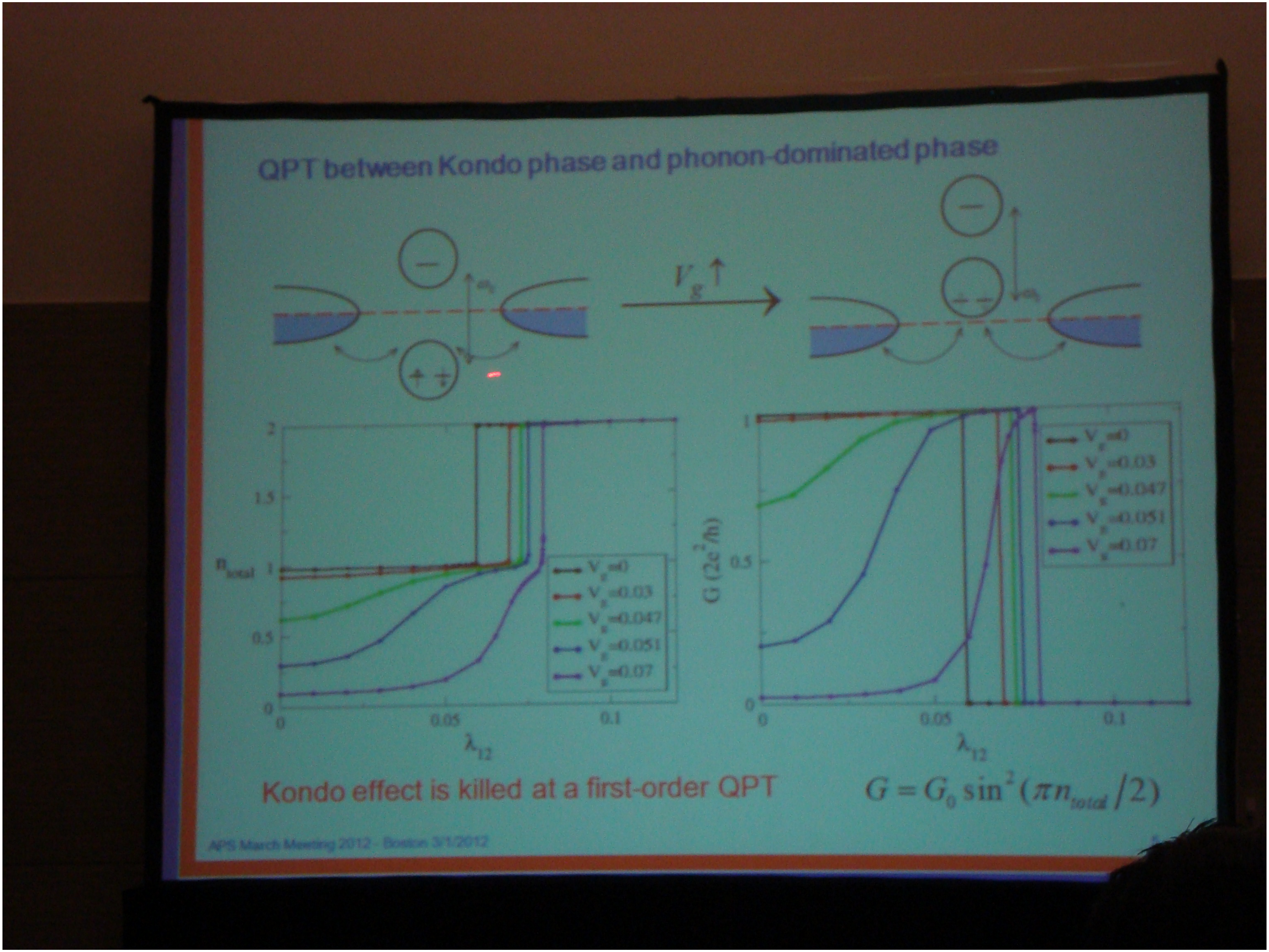
Increasing λ_{12} changes ground state from single-occupied to double-occupied:

$n_e = 2 \;\; n_o = 0$ or $n_e = 0 \;\; n_o = 2$

Gain in phonon energy should balance Coulomb cost of double occupation around

$4\lambda_{12}^2/\omega_0 = O(U_j)$

Does the ground state change continuously or abruptly?


QPT between Kondo phase and phonon-dominated phase
$V_g \uparrow$
n_{total}
λ_{12}
$G\ (2e^2/h)$
V_g=0
V_g=0.03
V_g=0.047
V_g=0.051
V_g=0.07
Kondo effect is killed at a first-order QPT
$G = G_0 \sin^2(\pi n_{total}/2)$
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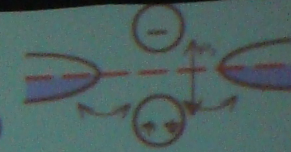
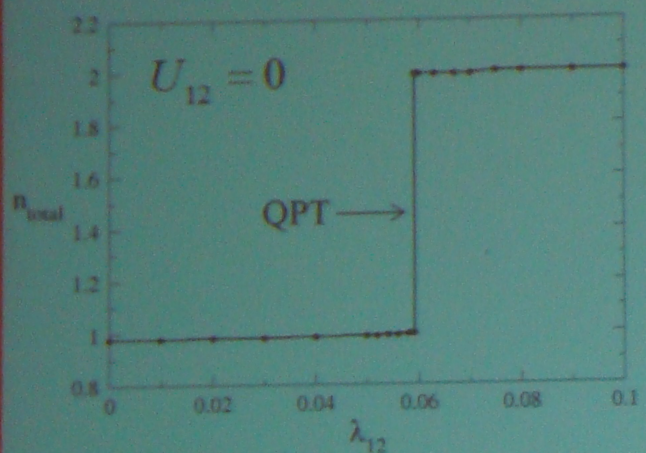
Why is there a QPT?

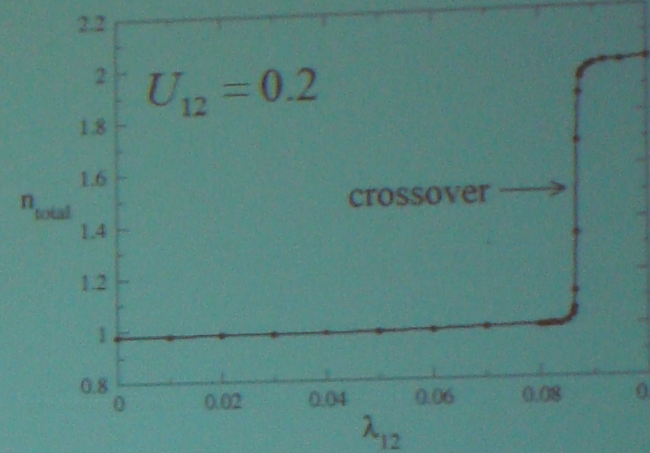


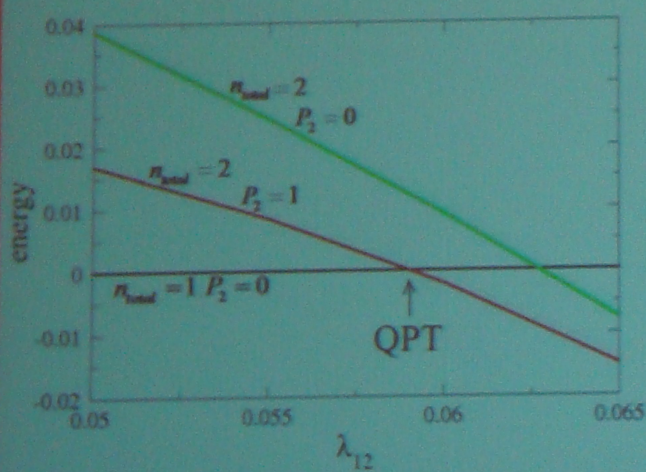
For $\Gamma_2 = 0$, n_2 changes only under $\lambda_{12}(b^\dagger + b)\sum_\sigma (d_{1\sigma}^\dagger d_{2\sigma} + d_{2\sigma}^\dagger d_{1\sigma})$

$P_2 = (n_2 + n_b) \bmod 2$ is conserved.

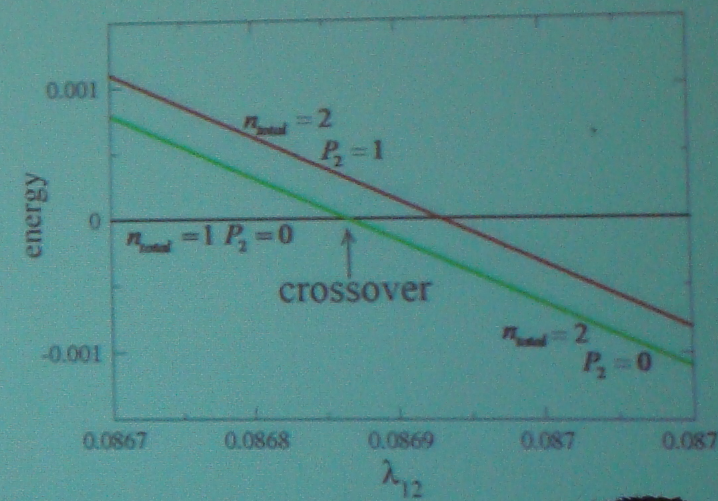


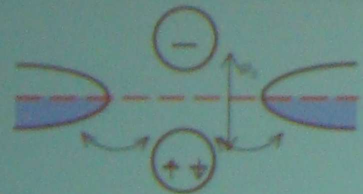
QPT vs. crossover



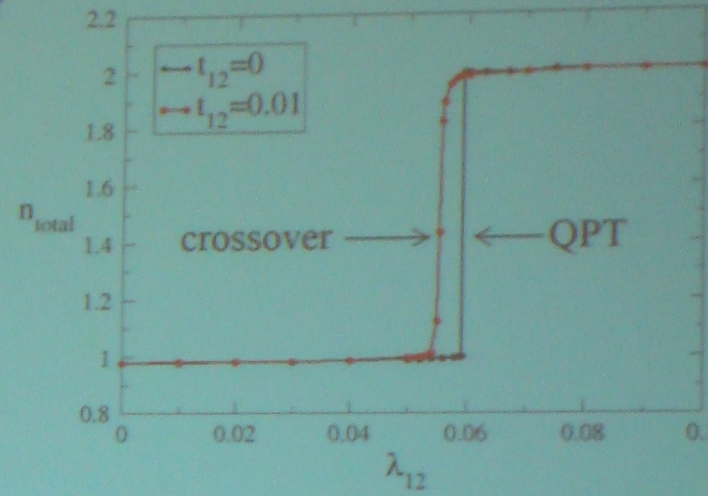


Lowest energies of isolated molecule





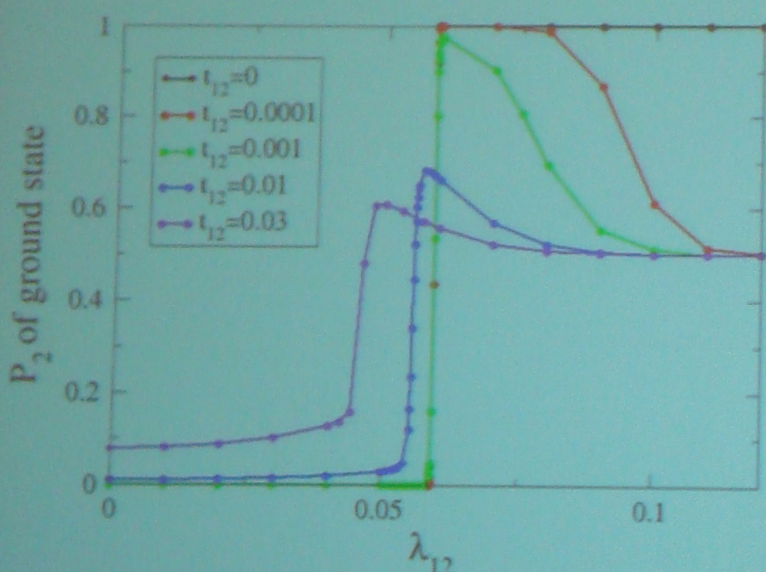
QPT or crossover?



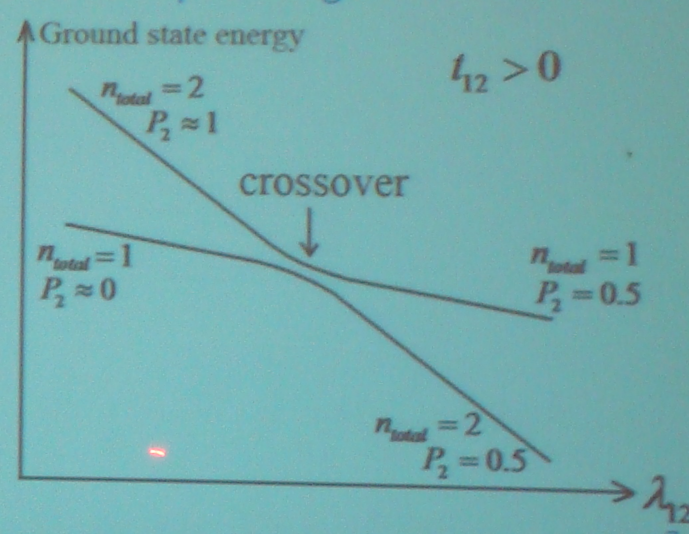
Turning on direct inter-level tunneling

$$t_{12}\sum_{\sigma}(d^{\dagger}_{1\sigma}d_{2\sigma}+d^{\dagger}_{2\sigma}d_{1\sigma})$$

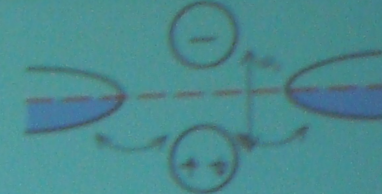
Conservation of $(n_2+n_b)\bmod 2=P_2$
is broken, QPT is replaced by crossover



Schematic plot of ground state energy

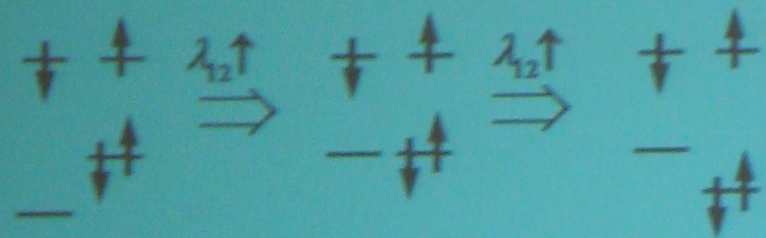


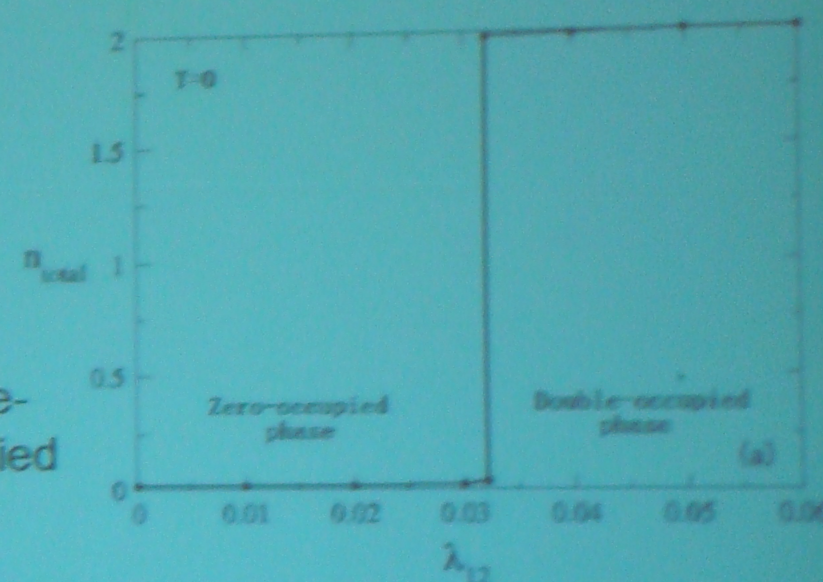
Interplay of two phonon effects



$$\lambda_{12}\sum_{\sigma}(d^{\dagger}_{1\sigma}d_{2\sigma}+d^{\dagger}_{2\sigma}d_{1\sigma})(b^{\dagger}+b)$$ Phonon-assisted inter-level tunneling

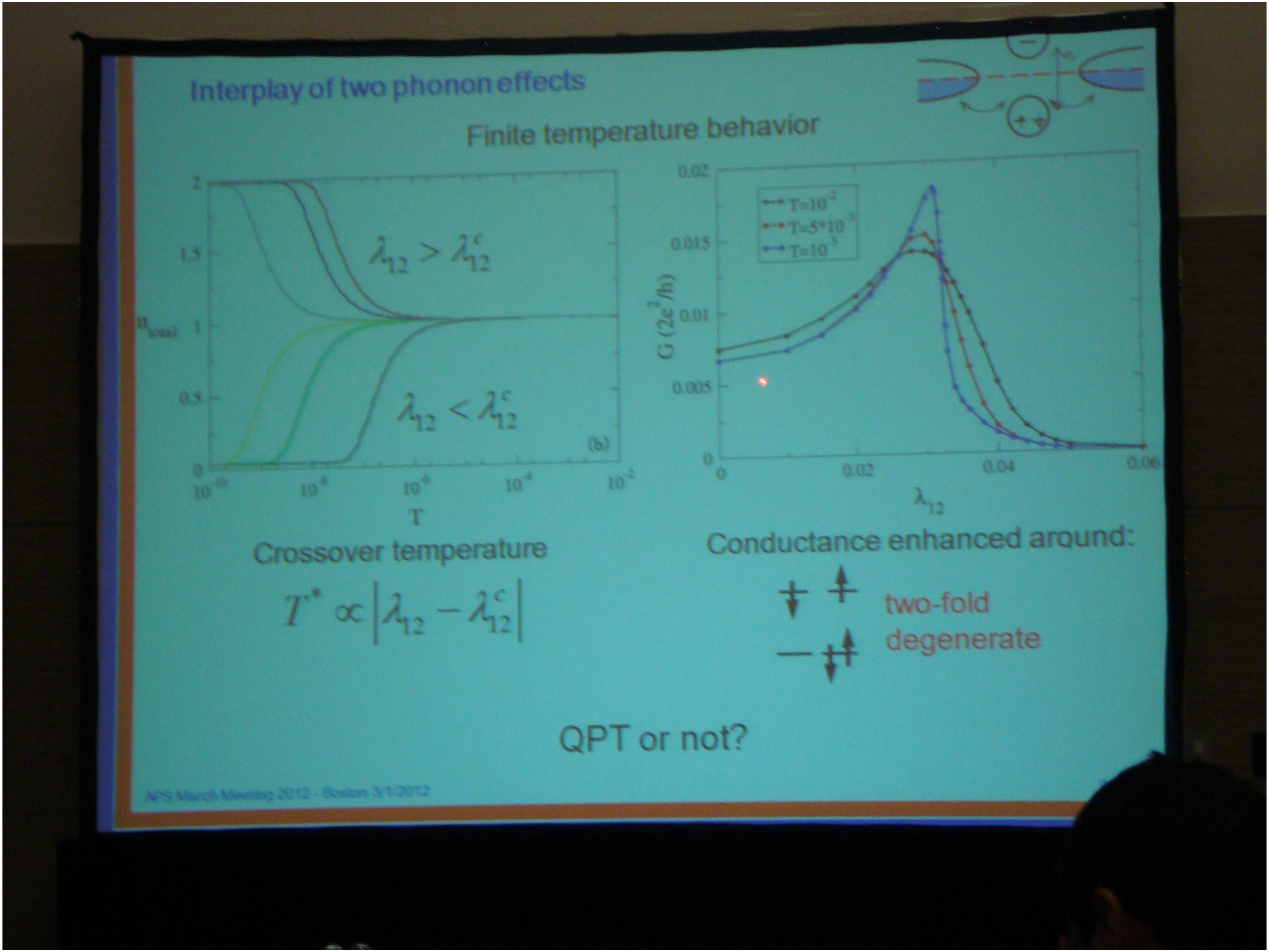
$$-\lambda_{1}(n_{1}-1)(b^{\dagger}+b)$$ Holstein-type phonon coupling to orbital-1





zero-occupied ⟹ degenerate ⟹ double-occupied

Does ground state change abruptly or continuously?


Interplay of two phonon effects
Finite temperature behavior
$\lambda_{12} > \lambda_{12}^c$
$\lambda_{12} < \lambda_{12}^c$
n_{total}
T
(b)
T=10^-2
T=5*10^-3
T=10^-3
G (2e^2/h)
λ_{12}
Crossover temperature
$T^* \propto |\lambda_{12} - \lambda_{12}^c|$
Conductance enhanced around:
two-fold
degenerate
QPT or not?
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Summary

- Have studied phonon-assisted inter-level tunneling in a single-molecule device.

- Competition between electron-electron and electron-phonon interactions can produce a first-order QPT between Kondo phase and phonon-dominated phase.

- The QPT arises from a special symmetry of the problem. Weak symmetry breaking leads to a sharp crossover instead.

- With Holstein phonon coupling, find a QPT or sharp crossover between two different phonon-dominated regimes.