# Thermodynamics of the Mott-Hubbard metal-insulator transition in high dimensions

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## Outline

Introduction

Controversy: phase diagram for fully frustrated “Bethe” lattice

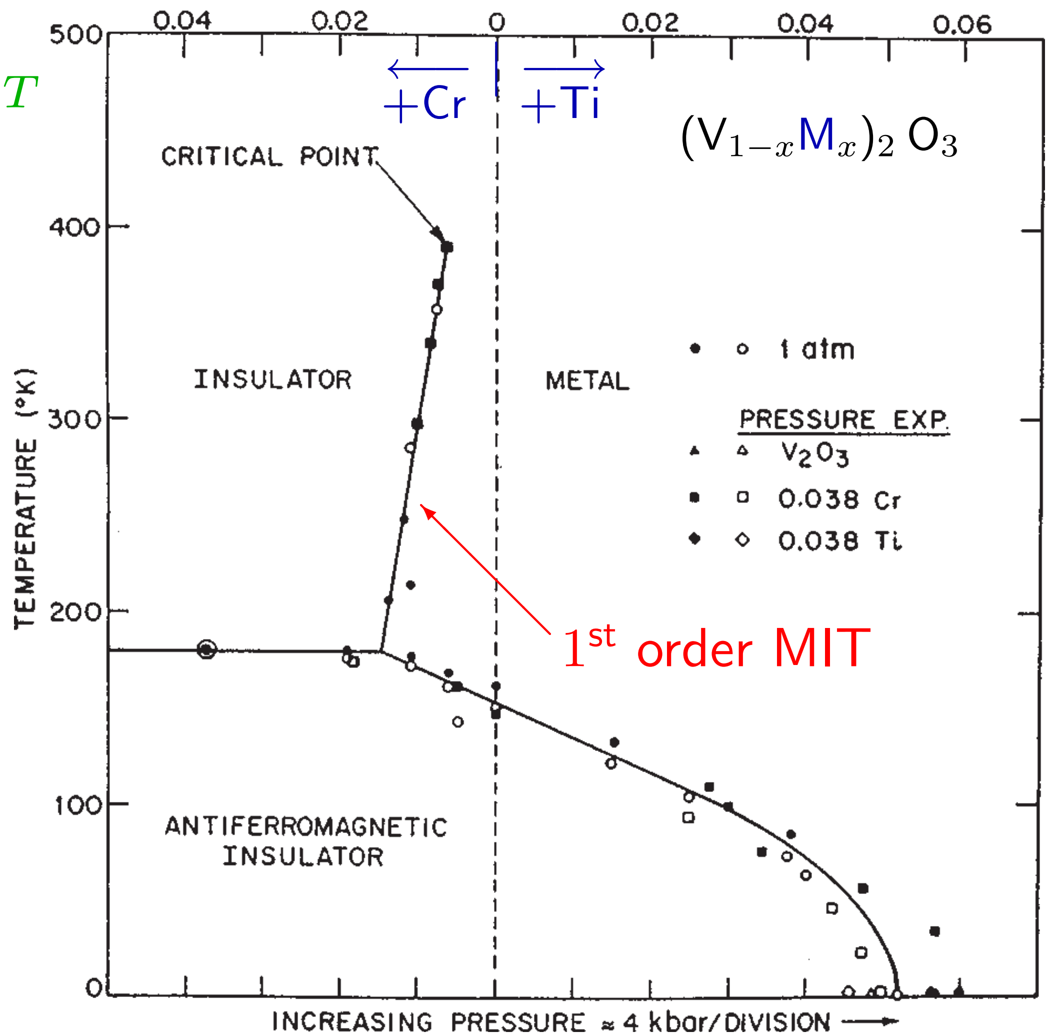
Construction of the coexistence phase diagram

Thermodynamic first-order phase transition line $U_c(T)$

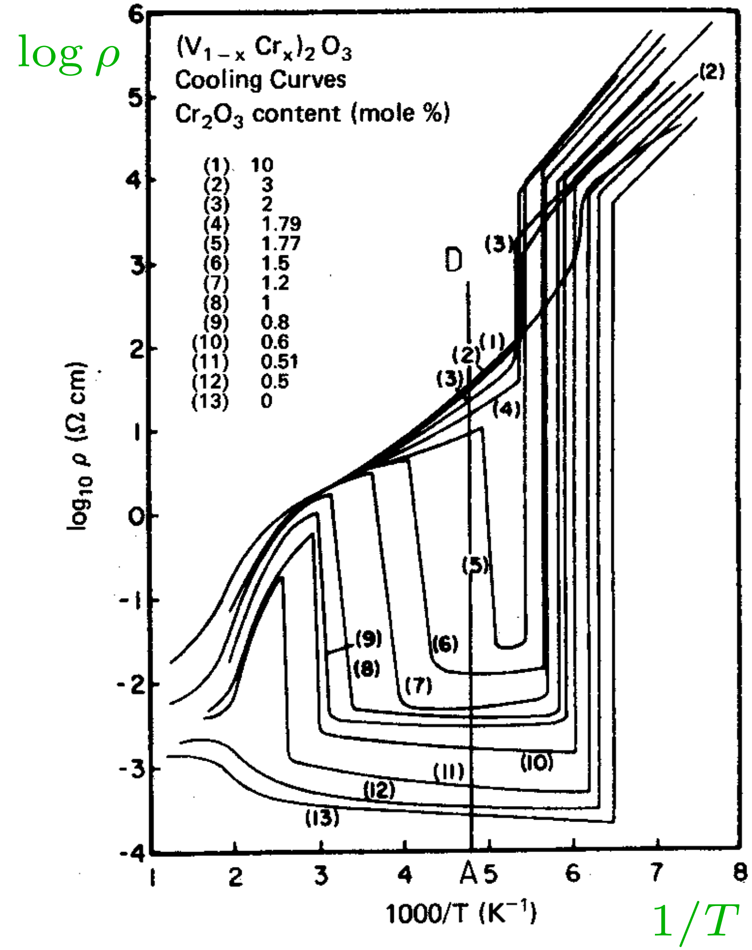
Conclusion

# Introduction

## Motivation: $V_2O_3$




McWhan et al, 1971




Kawamoto et al, 1980

MIT without LRO
resistivity $\rho$ increases by factor $10^3$
shift in lattice parameters

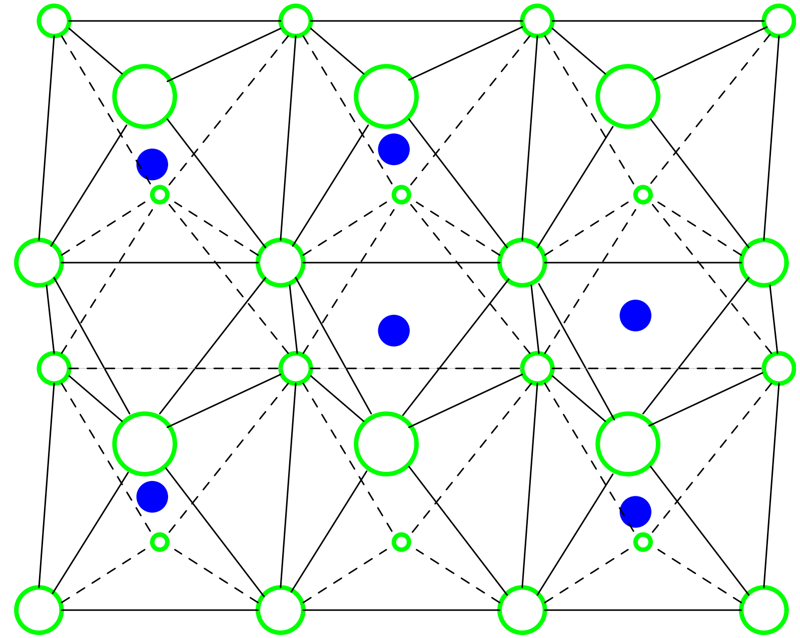
Corundum structure:

- hcp $O^{2-}$ lattice
- $V^{3+}$ fill 2/3 of octahedral vacancies

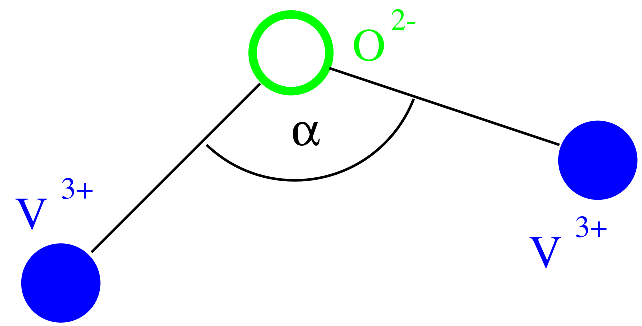
doping with Ti, Cr:

- (nearly) isovalent
- distorts lattice $\longrightarrow$ changes overlap
- drives MIT (like pressure)

Paramagnetic, bandwidth-controlled metal-insulator transition in $V_2O_3$
$\longrightarrow$ simple single-band model?

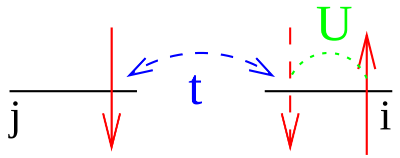


Corundum structure




$\alpha_{\mathbf{Cr}} < \alpha_{\mathbf{V}} < \alpha_{\mathbf{Ti}}$

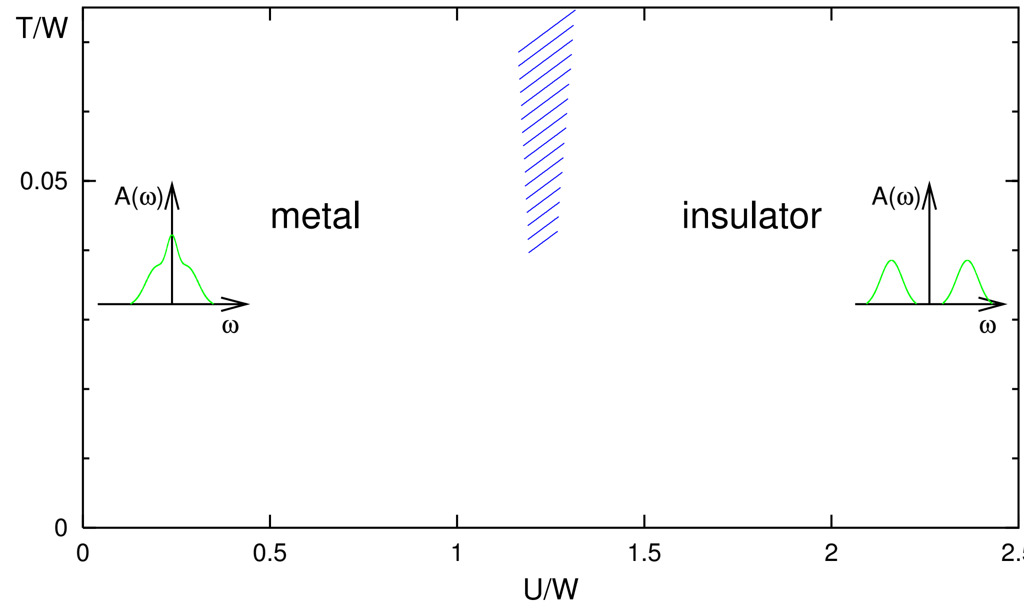
## Hubbard model



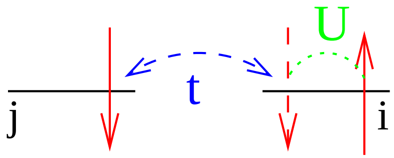

$$\hat{H} = \sum_{(i,j),\sigma} t_{ij} (\hat{c}^\dagger_{i\sigma} \hat{c}_{j\sigma} + \text{h.c.}) + U \sum_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}$$

Minimal model for correlated electrons

MIT/crossover at $U/W \approx 1$ (and half filling)



**Hubbard model**

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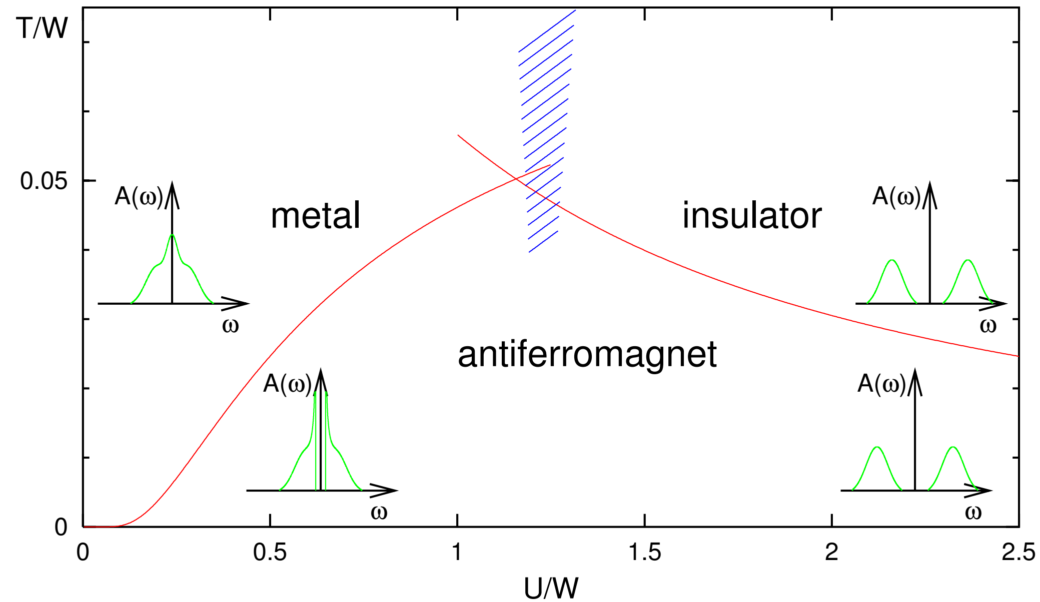
But: generically antiferromagnetism at low $T$



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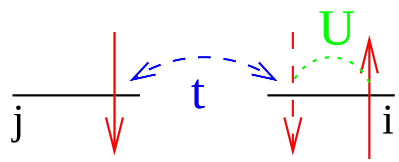


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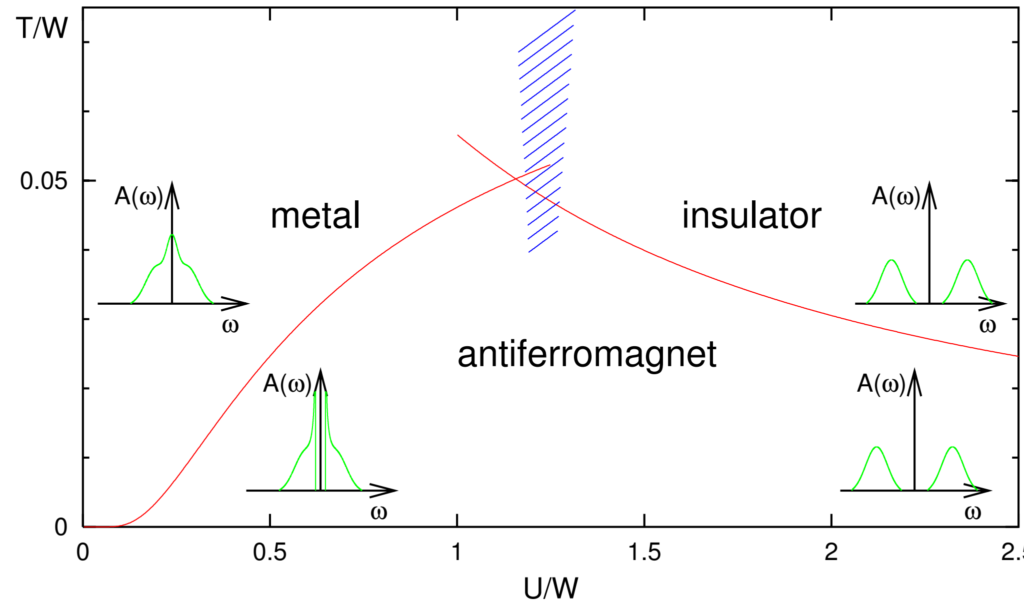
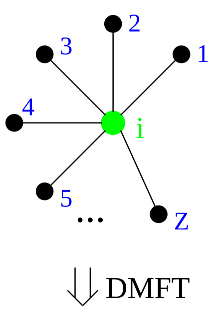
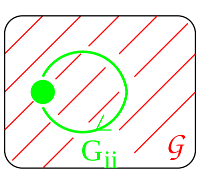



## Dynamical Mean-field Theory (DMFT)



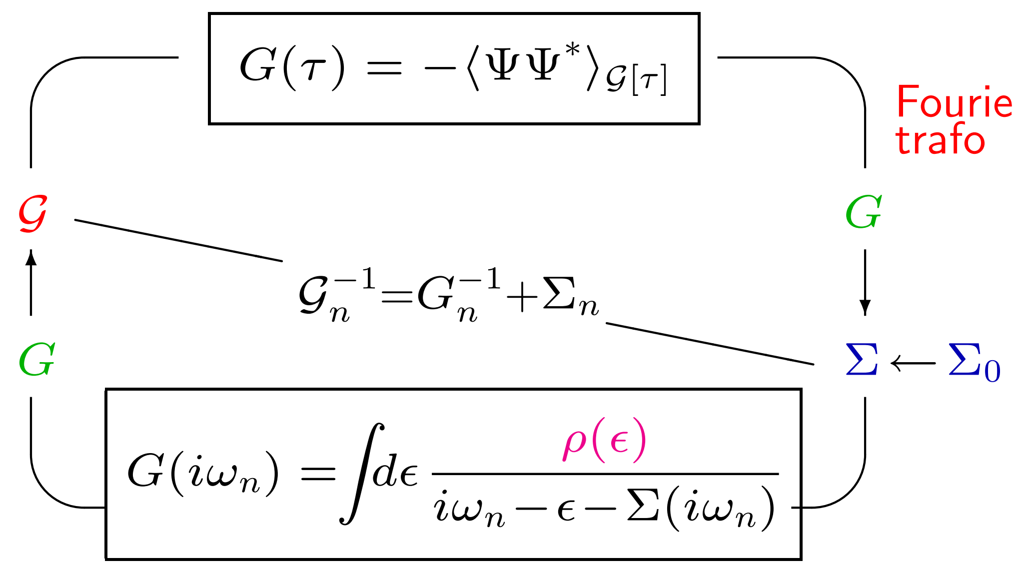




Mapping of lattice problem onto impurity model (SIAM) plus self-consistency condition

exact for coordination $Z \to \infty$

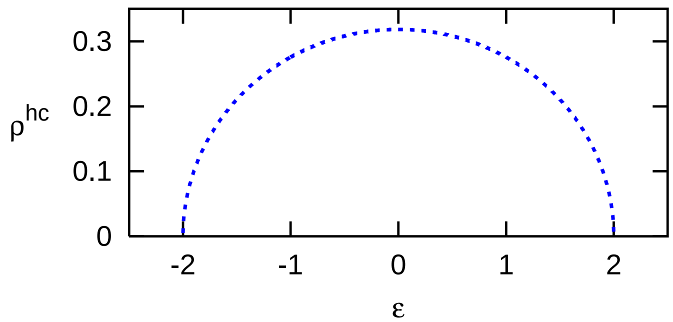
scaling of hopping $t \propto 1/\sqrt{Z}$

self-energy $\Sigma(\omega)$ local



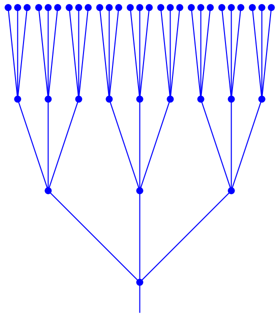

Local properties depend on lattice only via DOS $\rho(\epsilon)$ in paramagnetic phase

# Controversy: phase diagram for fully frustrated “Bethe” lattice



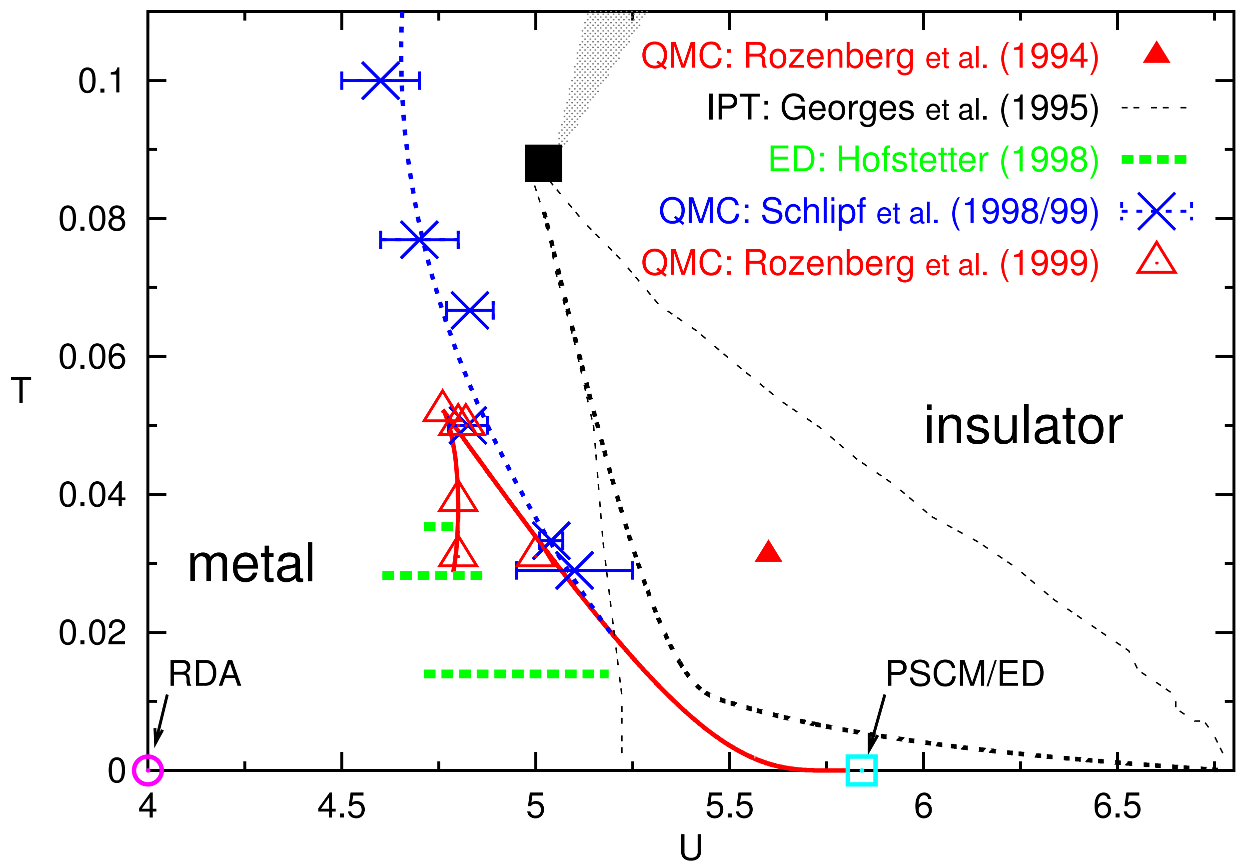

Convenient choice: semi-elliptic DOS

full frustration



Status as of 1999: coexistence?

- IPT, ED: yes!
- RDA: no! (much lower $U_c$)
- QMC (Schlipf et al.): no!
- QMC (Rozenberg et al.): yes!



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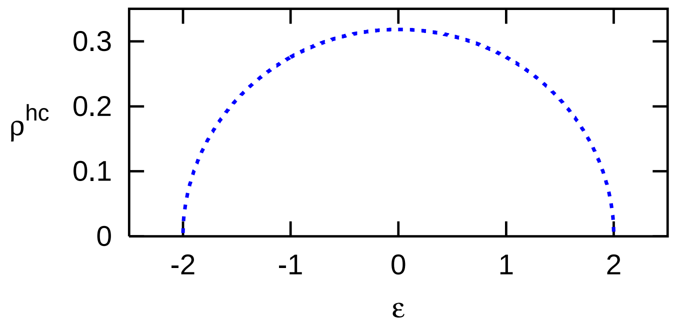


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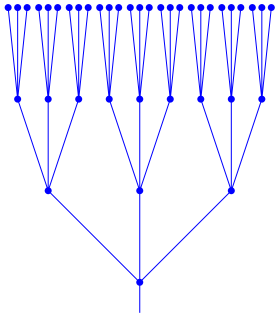

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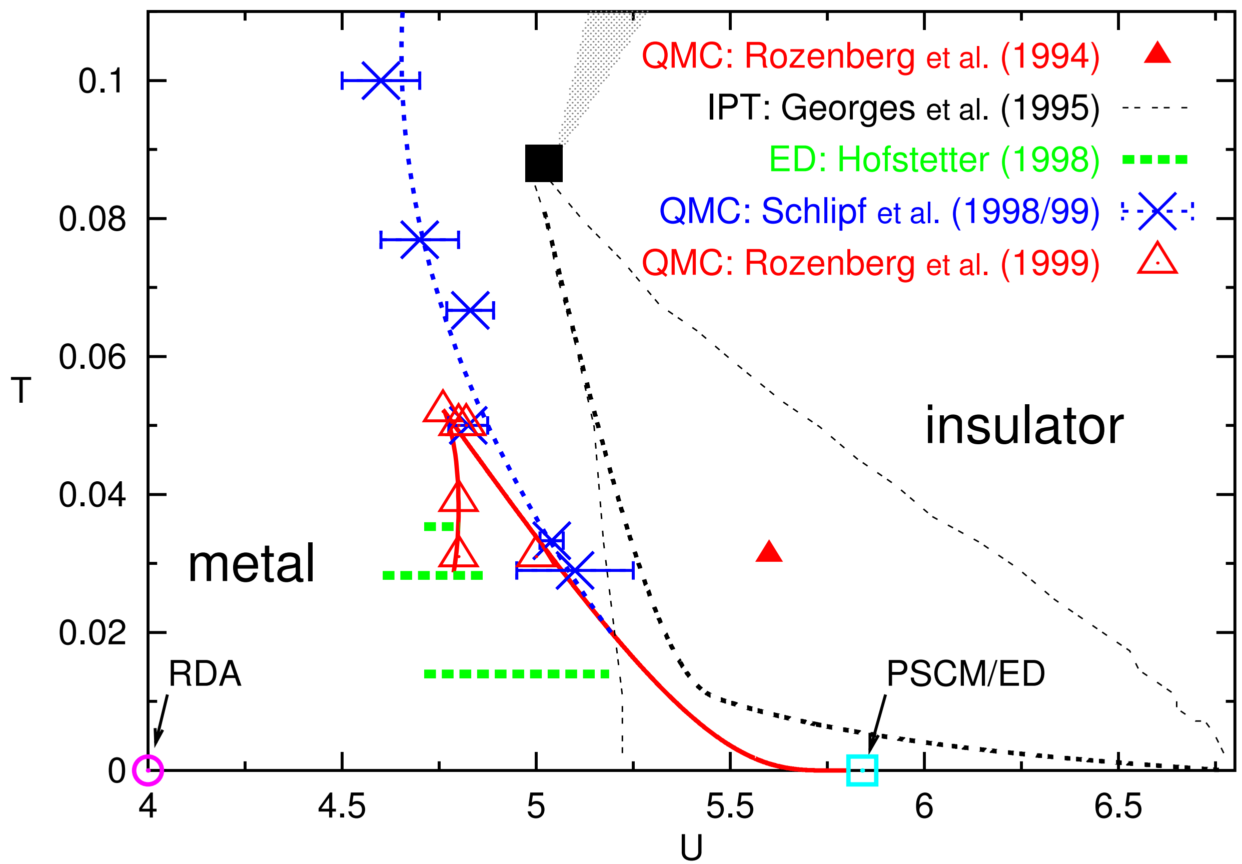



- Who is right / What went wrong?
- Precise coexistence phase diagram?
- Thermodynamic first order phase transition line?

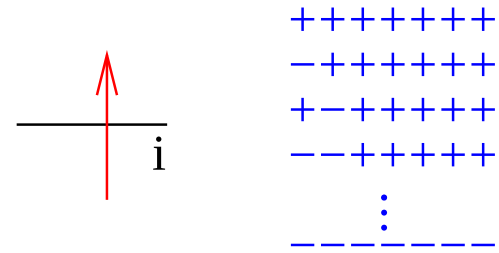
# Quantum Monte Carlo Algorithm

Trotter-Suzuki decomposition $e^{-\beta(\hat{A}+\hat{B})} = \left(e^{-\Delta\tau \hat{A}} e^{-\Delta\tau \hat{B}}\right)^{\Lambda} + \mathcal{O}(\Delta\tau)$; $\Lambda = \beta/\Delta\tau$

Hubbard-Stratonovich transformation, Wick's theorem:

$$G_{\sigma l l'} = \frac{\sum_{\{s\}} \left(\boldsymbol{M}_{\sigma}^{\{s\}}\right)^{-1}_{ll'} \det \boldsymbol{M}_{\uparrow}^{\{s\}} \det \boldsymbol{M}_{\downarrow}^{\{s\}}}{\sum_{\{s\}} \det \boldsymbol{M}_{\uparrow}^{\{s\}} \det \boldsymbol{M}_{\downarrow}^{\{s\}}},$$

$$M^{s_l}_{\sigma l l'} = (\Delta\tau)^2 \,(\boldsymbol{\mathcal{G}}_{\sigma}^{-1})_{ll'}\, e^{\lambda\sigma s_{l'}} + \delta_{ll'}\left(1 - e^{\lambda\sigma s_l}\right)$$




Monte-Carlo importance sampling of $\{s\}$, unknown prefactor in $\mathcal{Z}$ cancels

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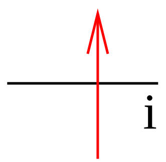


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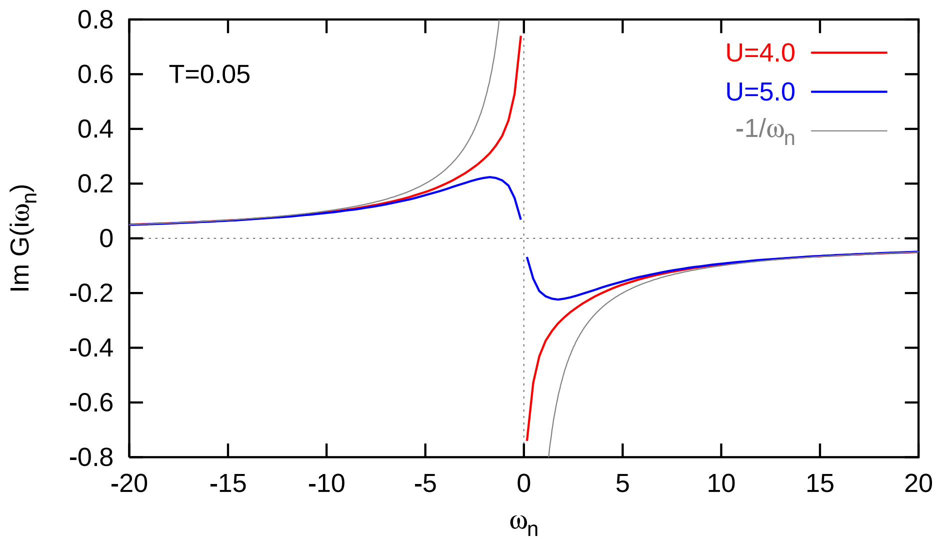
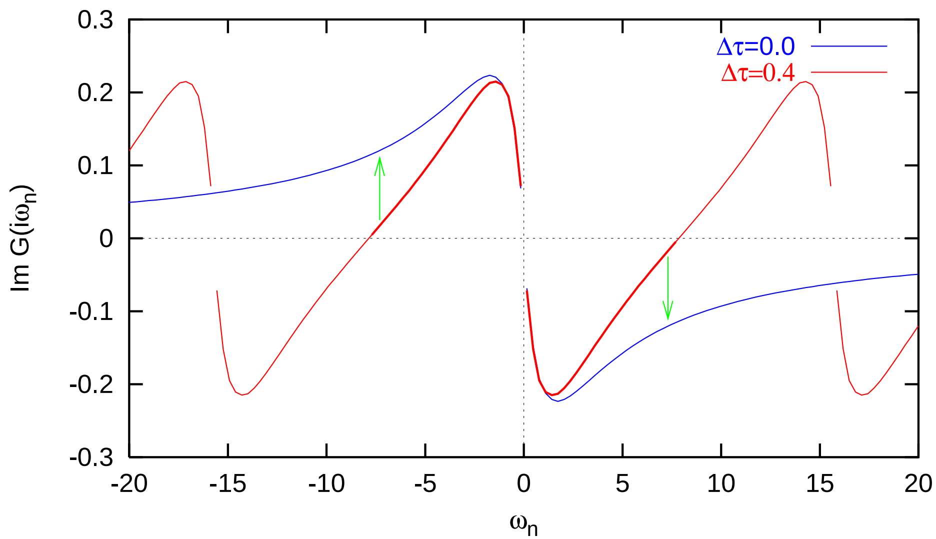
Monte-Carlo importance sampling of $\{s\}$, unknown prefactor in $\mathcal{Z}$ cancels

Fourier transformations $\{G(\tau_l)\}_{l=0}^{\Lambda} \leftrightarrow G(i\omega_n)$




naive discrete Fourier trafo + correction

Krauth: spline of $G(\tau)$, analytic FT

Jarrell: spline of $G(\tau) - G_0(\tau)$

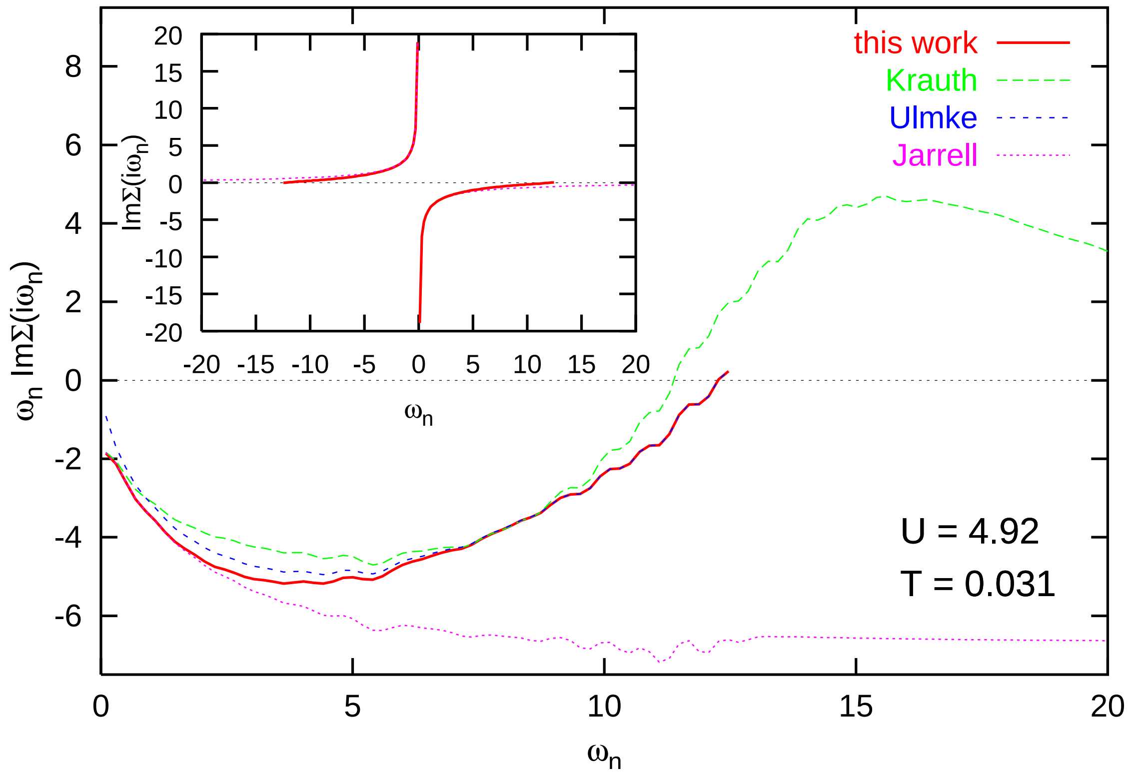
different approach: Ulmke smoothing

$$\tilde{G}(i\omega_n) = \Delta\tau \sum_{l=0}^{\Lambda-1} e^{i\omega_n \tau_l} G(\tau_l)$$

$$G(i\omega_n) = \frac{\Delta\tau}{\ln(1 + \Delta\tau/\tilde{G}(i\omega_n))}$$

large $\Delta\tau$ dependence at small $\omega$

Results of FT $G(\tau) \to G(i\omega_n)$ (plus inverted Dyson eq.)




Frequency dependent smoothing correction: $\Delta\tau \longrightarrow \left(1 - (\omega_n \Delta\tau/\pi - 1)^8\right)\Delta\tau$

changes $G(i\omega_n)$ only for $\omega_n \approx \omega_{\text{Nyquist}} = \pi/\Delta\tau$

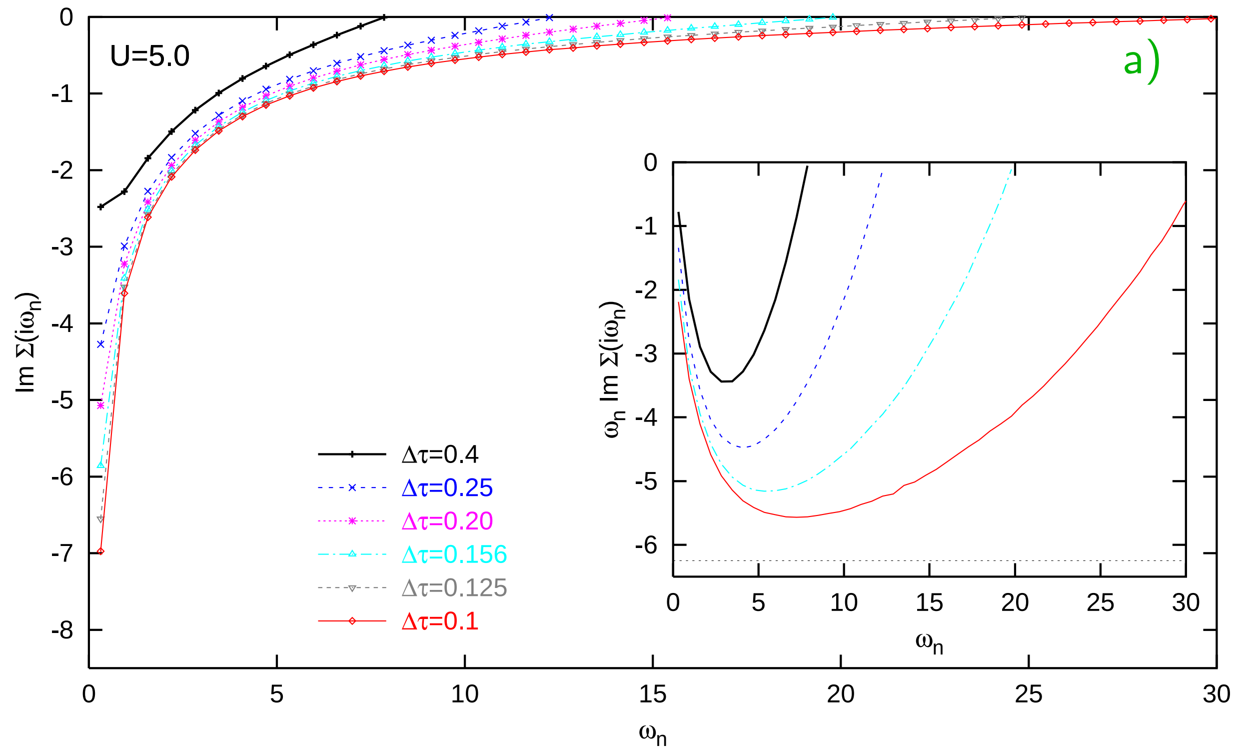
correct for small $\omega$ already at finite $\Delta\tau$

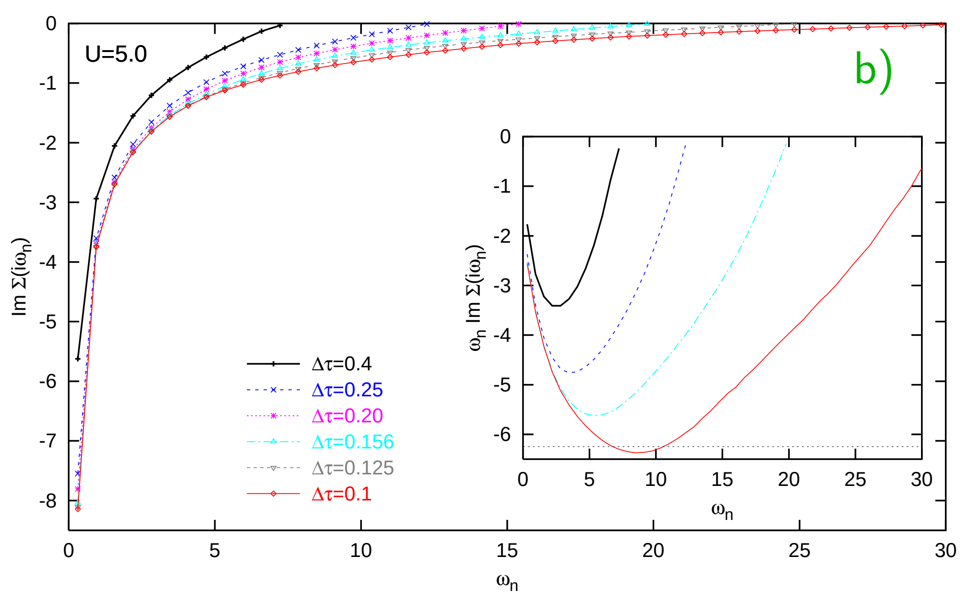
Quantitative comparison of Fourier transformation schemes:
self-energy ($T = 0.1$, $U = 5.0$)

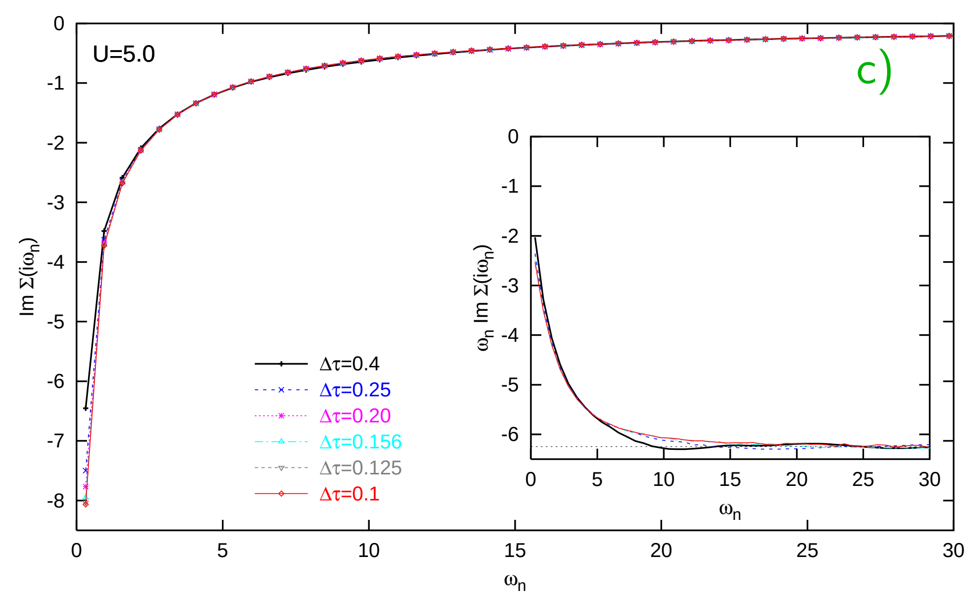
a) "Ulmke Smoothing",
b) improved "Smoothing",
c) spline plus analytic high frequency corrections

Stronger effects at lower $T$

Low-frequency part of $\Sigma(\omega)$ well captured in b) und c)



# Construction of the Coexistence Phase Diagram

## Observables

a) double occupancy

$$D = \langle \hat{n}_{i\downarrow} \hat{n}_{i\uparrow} \rangle$$

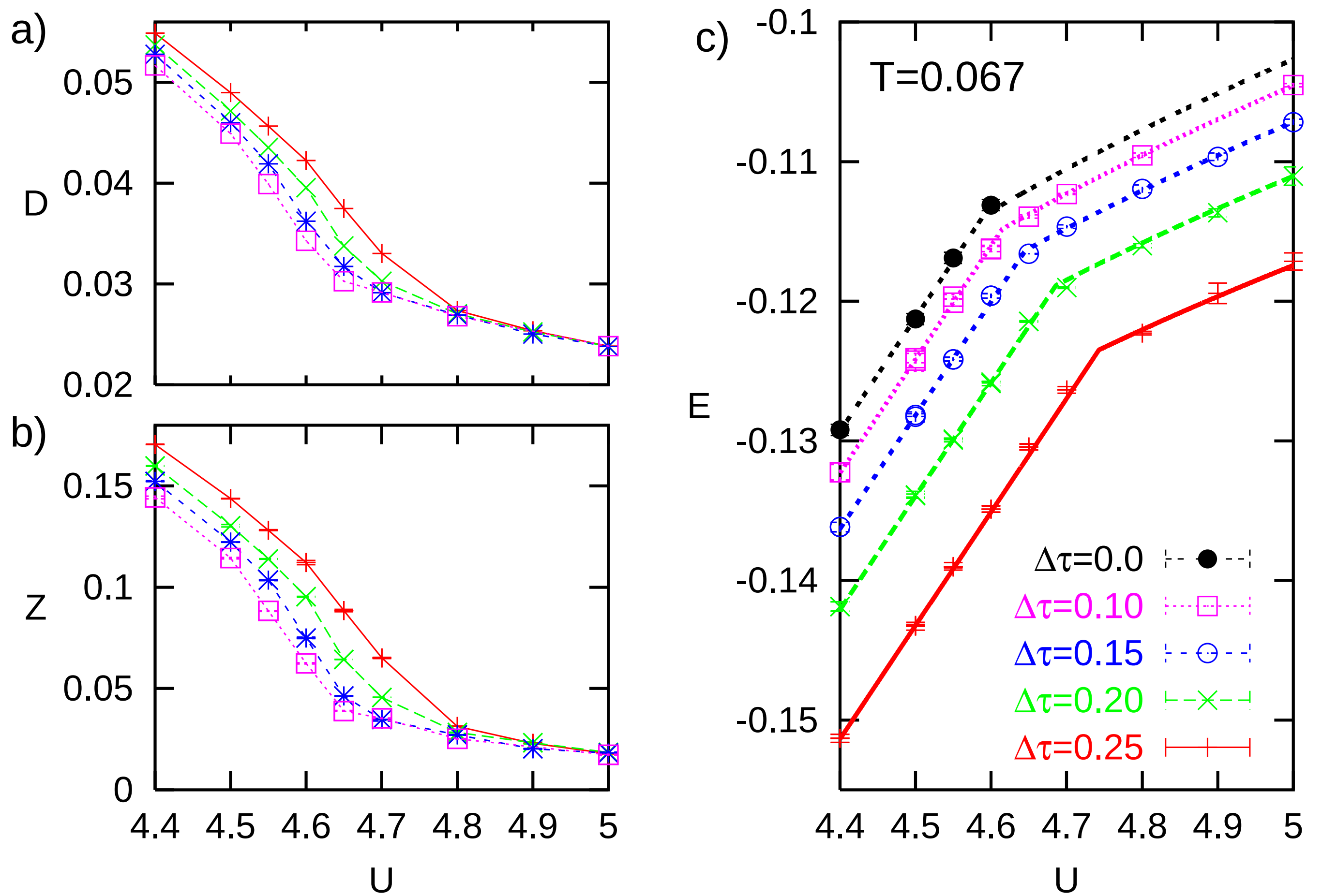
b) quasiparticle weight

$$Z^{-1} = 1 - \frac{d\,\mathrm{Re}\,\Sigma(\omega)}{d\omega}$$

$$\approx 1 - \frac{\mathrm{Im}\,\Sigma(i\pi T)}{\pi T}$$

c) energy

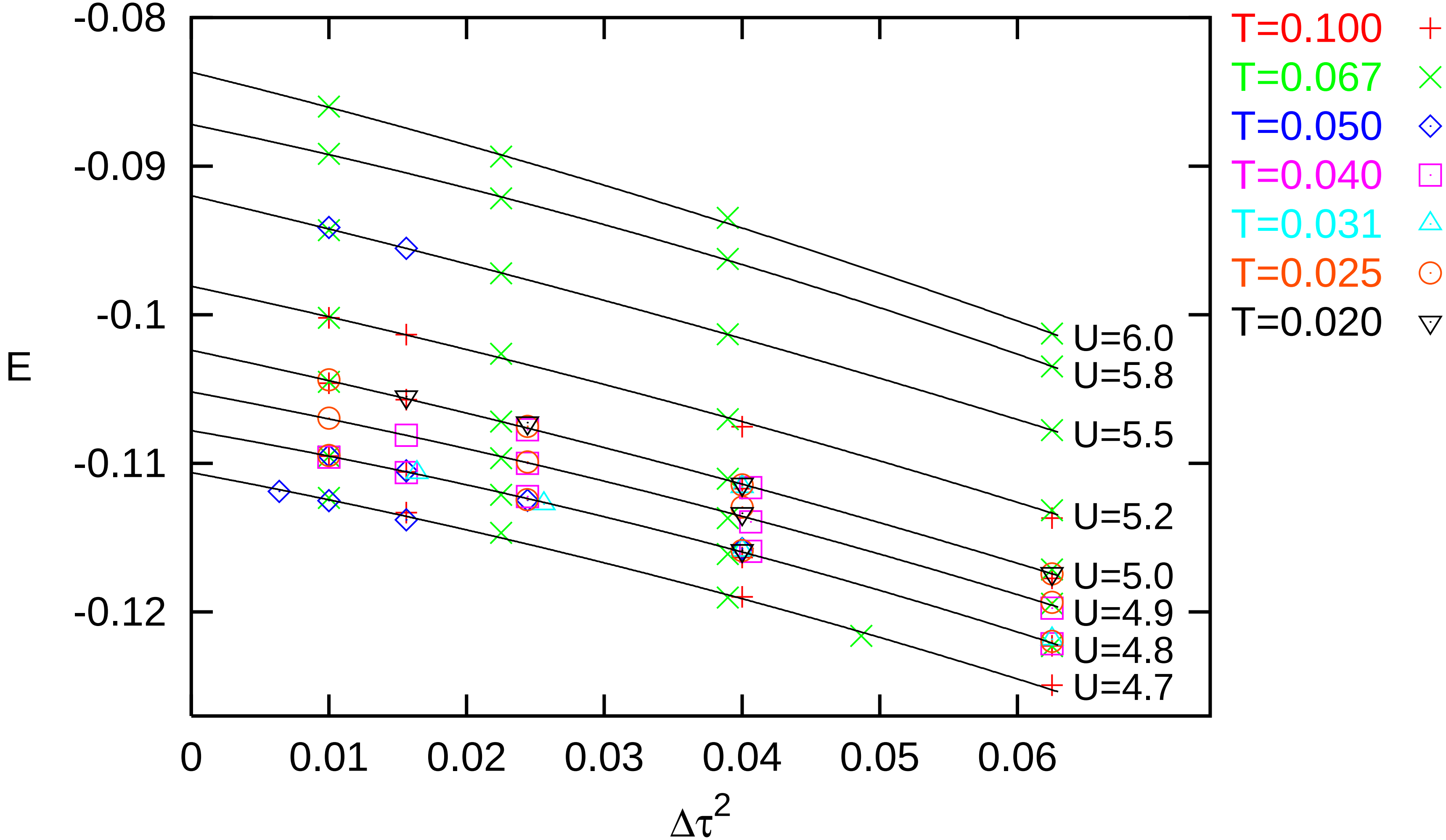
$$E = UD + E_{\text{kin}}$$




$$E_{\text{kin}} = E^0_{\text{kin}} + \int_{-\infty}^{\infty} d\epsilon\, \epsilon \rho(\epsilon)\, 2T \sum_{n=-\infty}^{\infty} \big(G_\epsilon(i\omega_n) - G^0_\epsilon(i\omega_n)\big); \qquad E^0_{\text{kin}} = 2 \int_{-\infty}^{\infty} d\epsilon\, \epsilon\, \rho(\epsilon)/(e^{\beta\epsilon}+1)$$

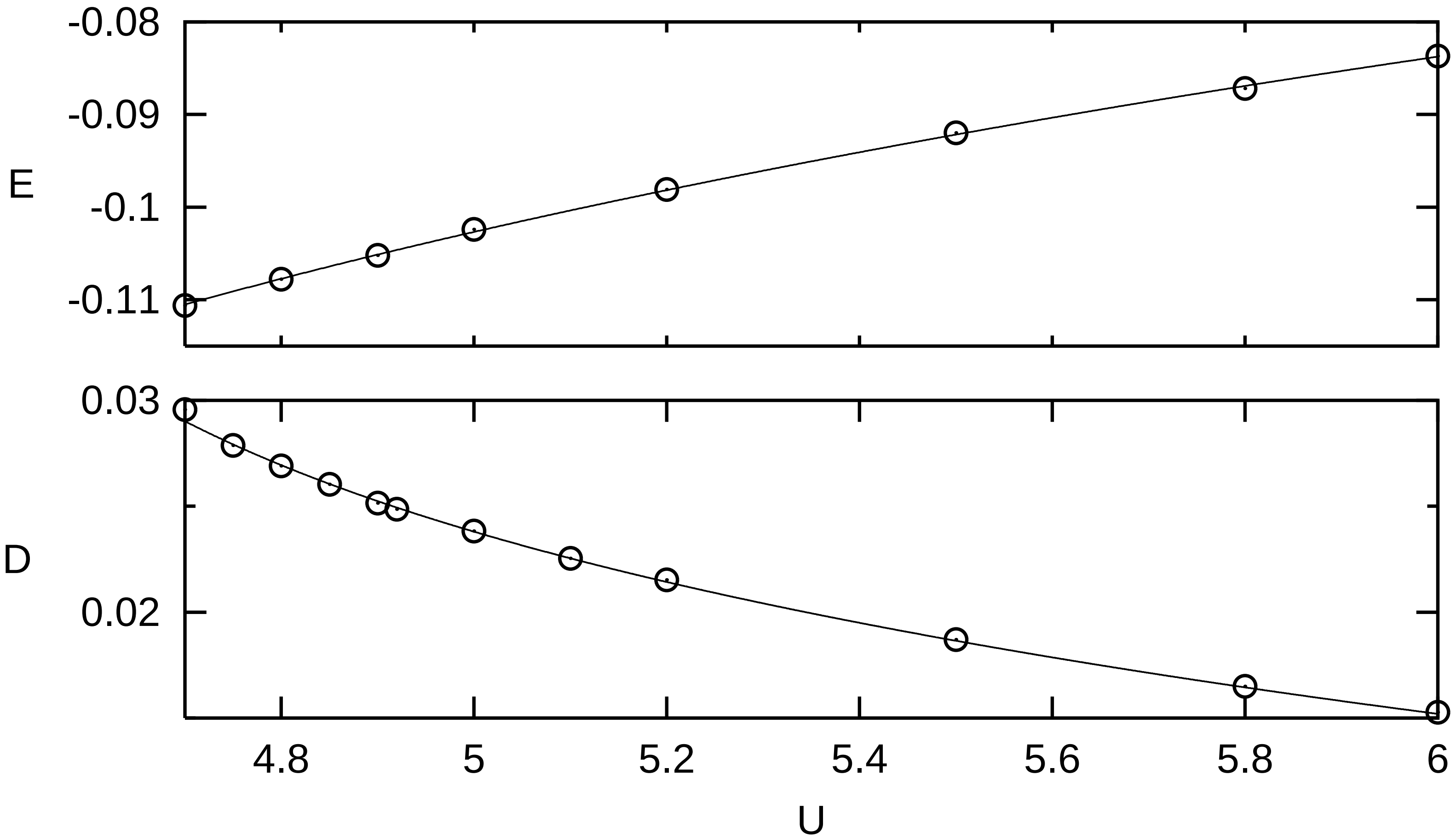
energy: ideal observable for localizing MIT:
- regular (quadratic) $\Delta\tau$ dependence within each phase
- nearly zero curvature within each phase

In insulating phase: $E(U,T) \equiv E(U)$, $D(U,T) \equiv D(U)$




very precise extrapolations $\Delta\tau \longrightarrow 0$ for energy $E$

no significant $\Delta\tau$ error for double occupancy $D$ (not shown)



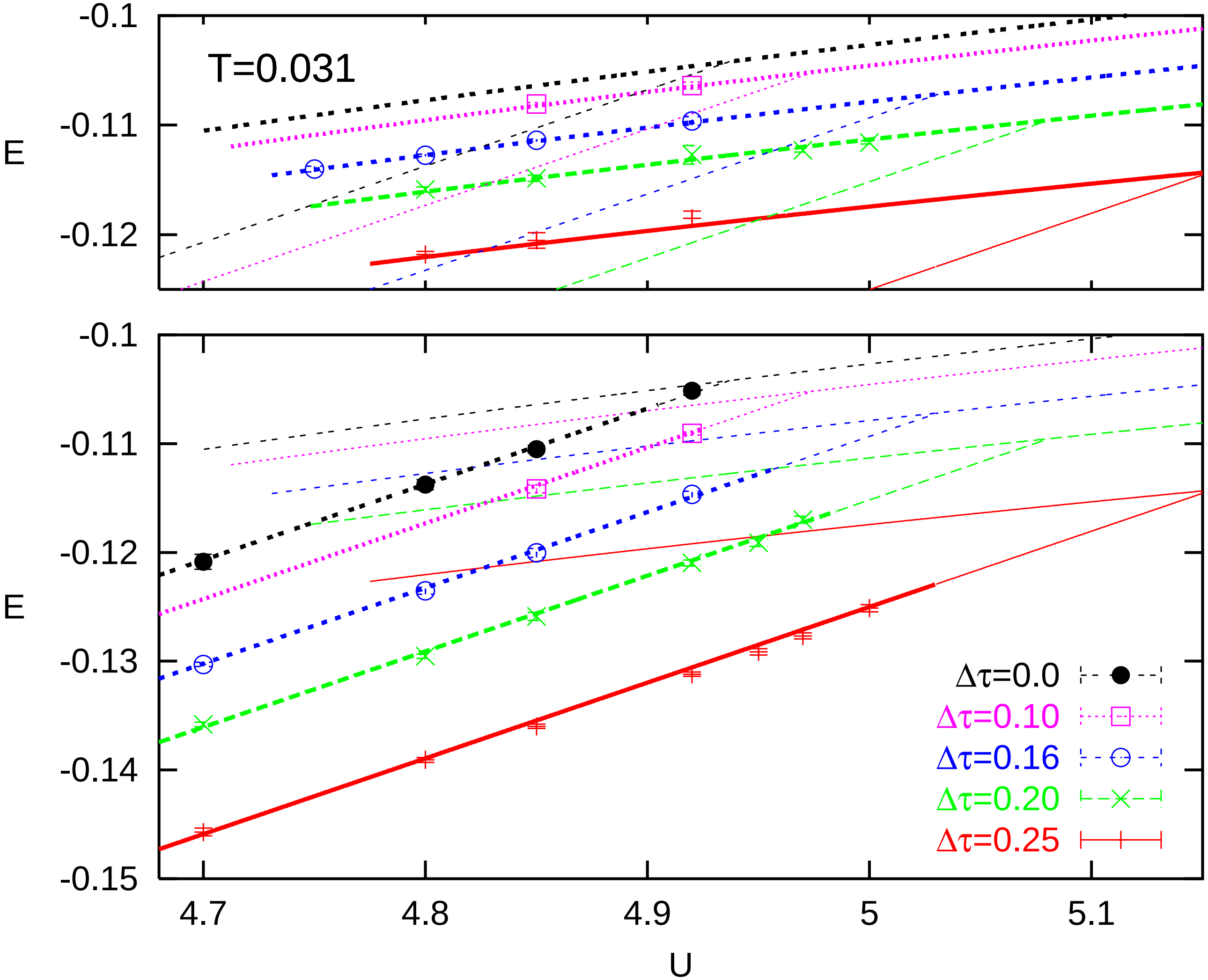

Consistent fits for ground state properties (at $\Delta\tau = 0$)

$$E_{\mathsf{i}}(U) \quad = \quad -\frac{0.515}{U} - \frac{0.0027}{U-3.95} - 0.0071 + 0.00325\; - 0.00025\,U^2$$

$$(4.7 \lesssim U \lesssim 6.0)$$

$$D_{\mathsf{i}}(U) \quad = \quad \frac{0.515}{U^2} + \frac{0.0027}{(U-3.95)^2} + 0.00325 - 0.0005\,U$$

# Coexistence region: energy $E$ in insulating and metallic phases

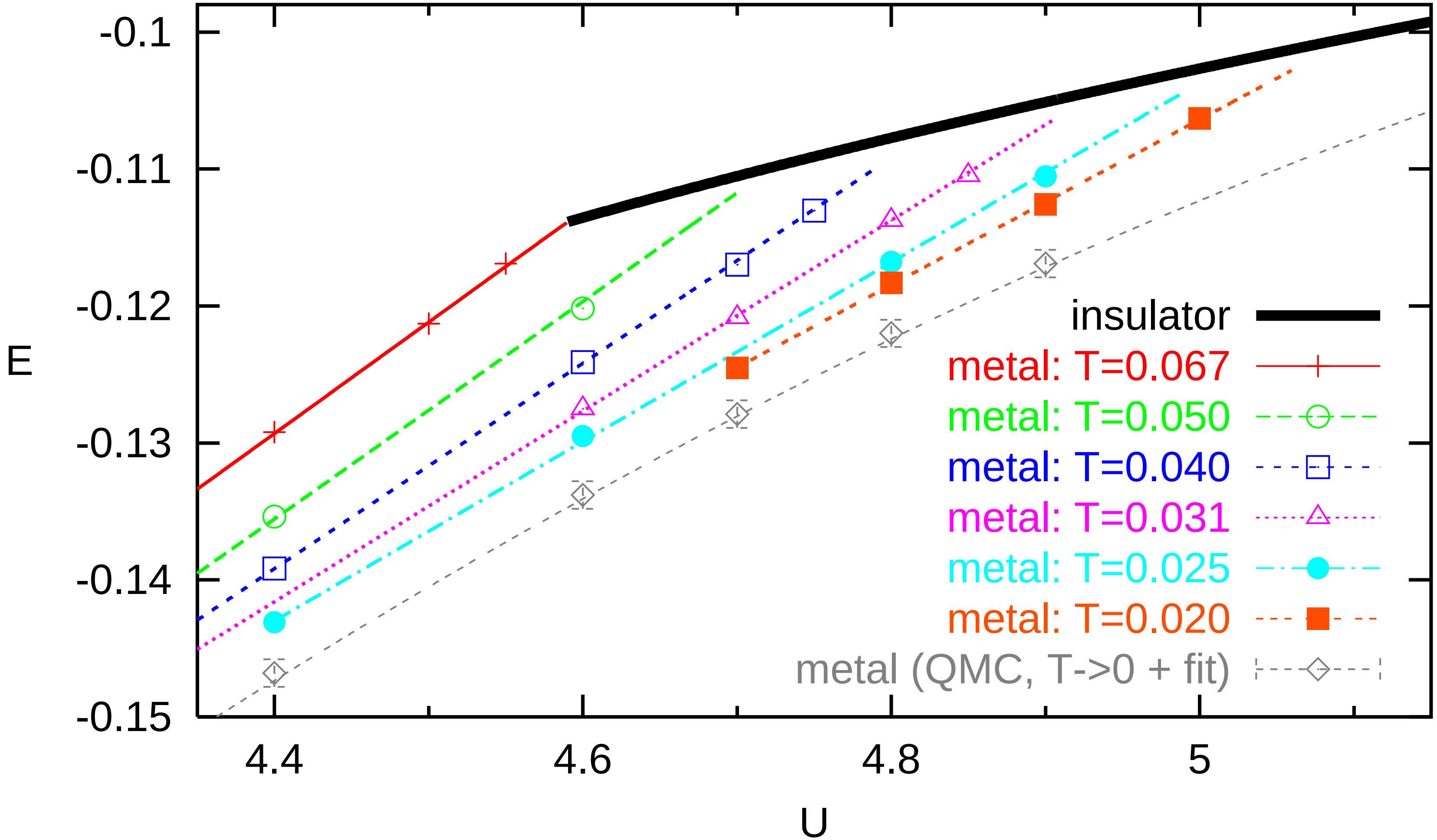



broad coexistence at $T = \frac{1}{32}$

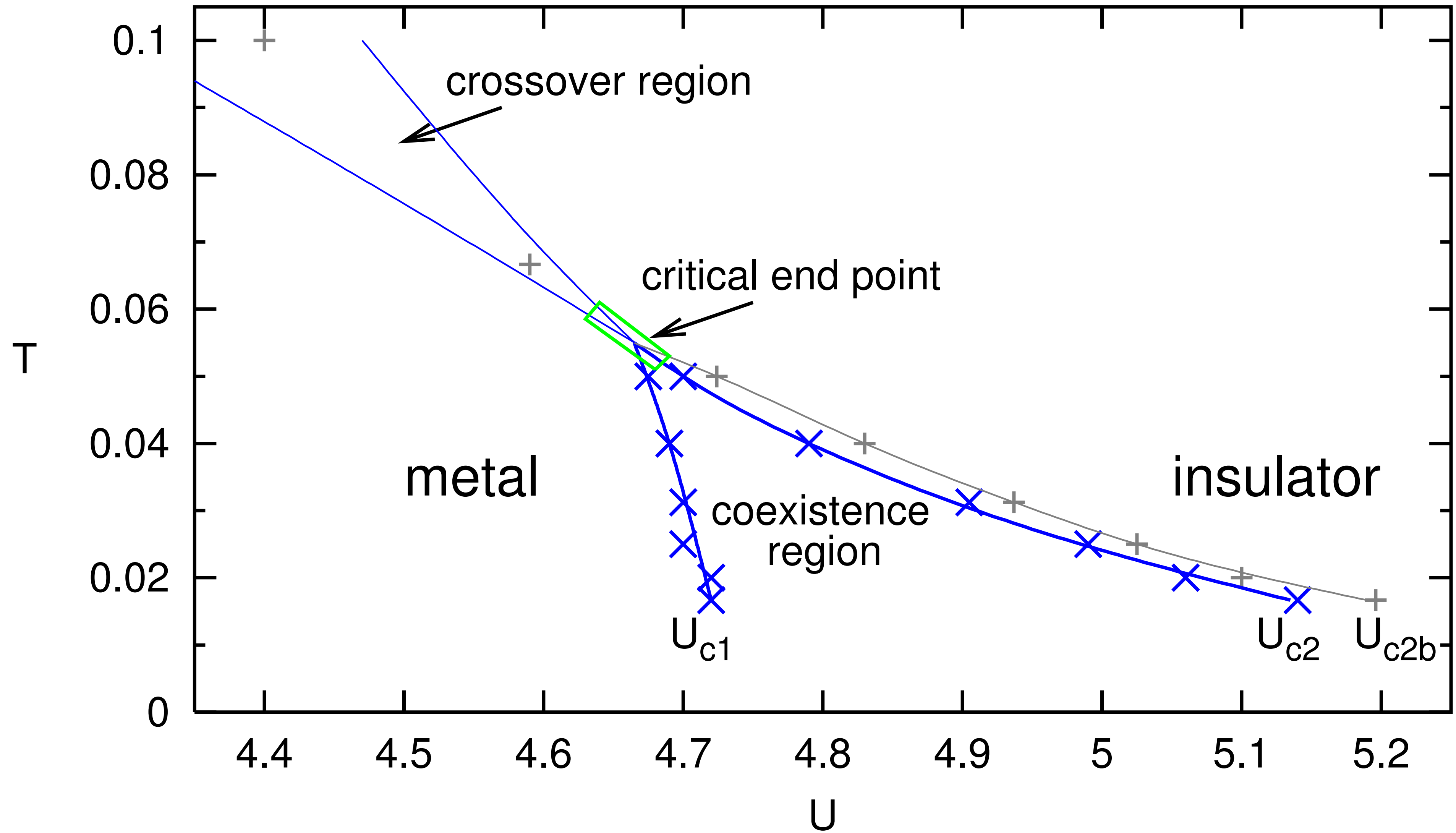
energy linear (in $U$) in metallic phase

metallic phase almost stable up to $U_{c2b}$, where extrapolated energy matches $E_i(U)$

Energy, extrapolated to $\Delta\tau = 0$



# Final coexistence phase diagram



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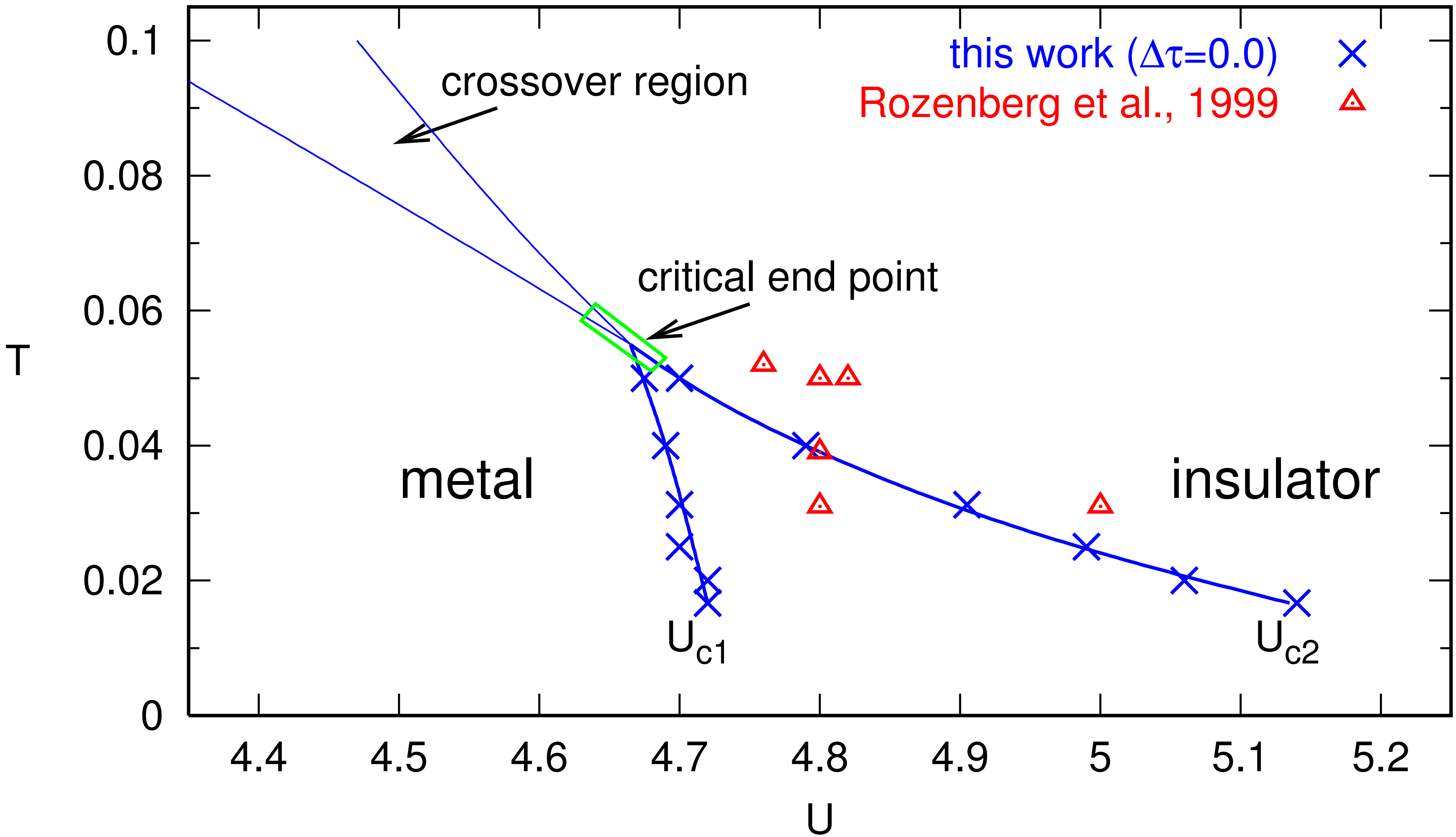

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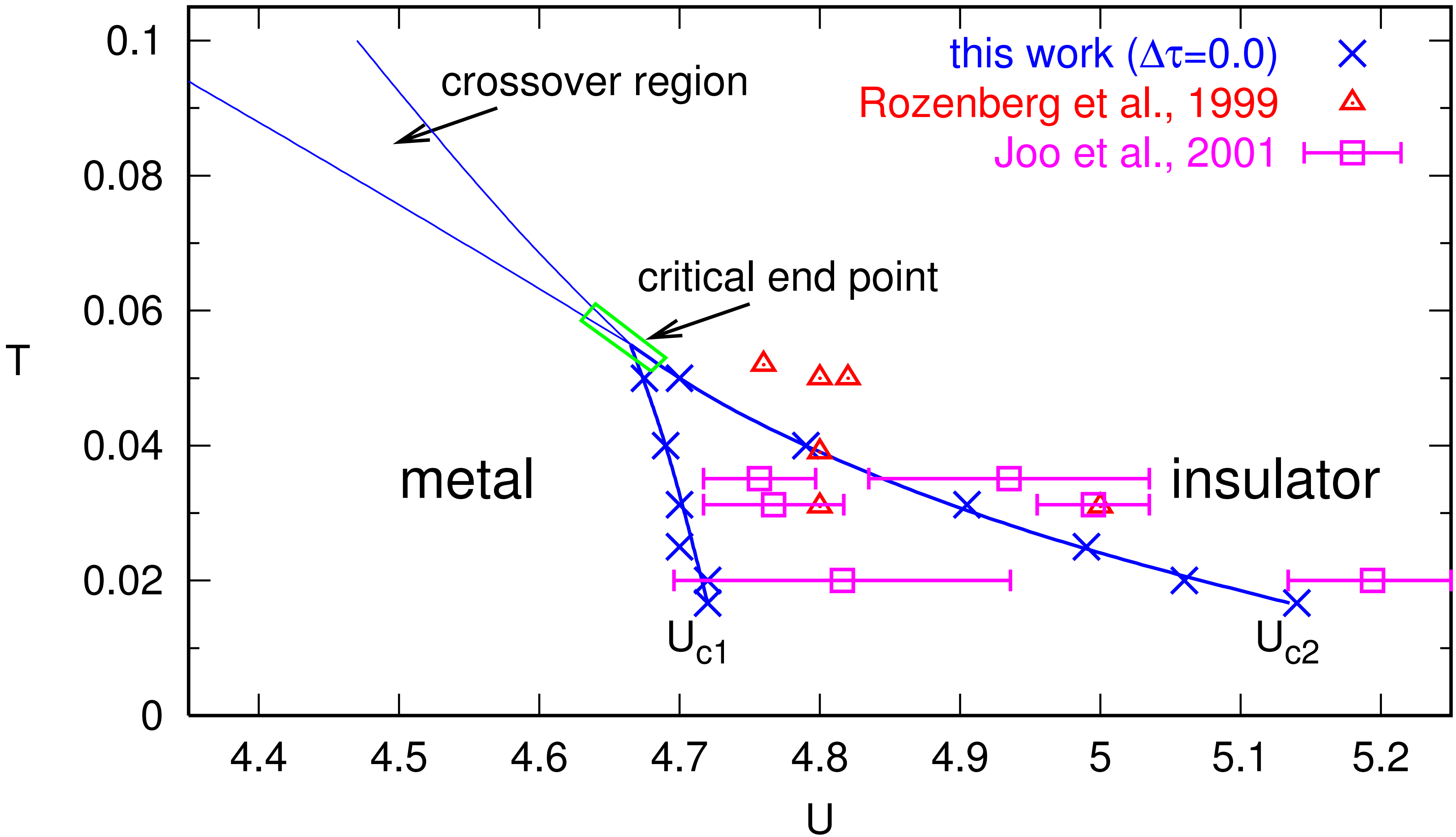

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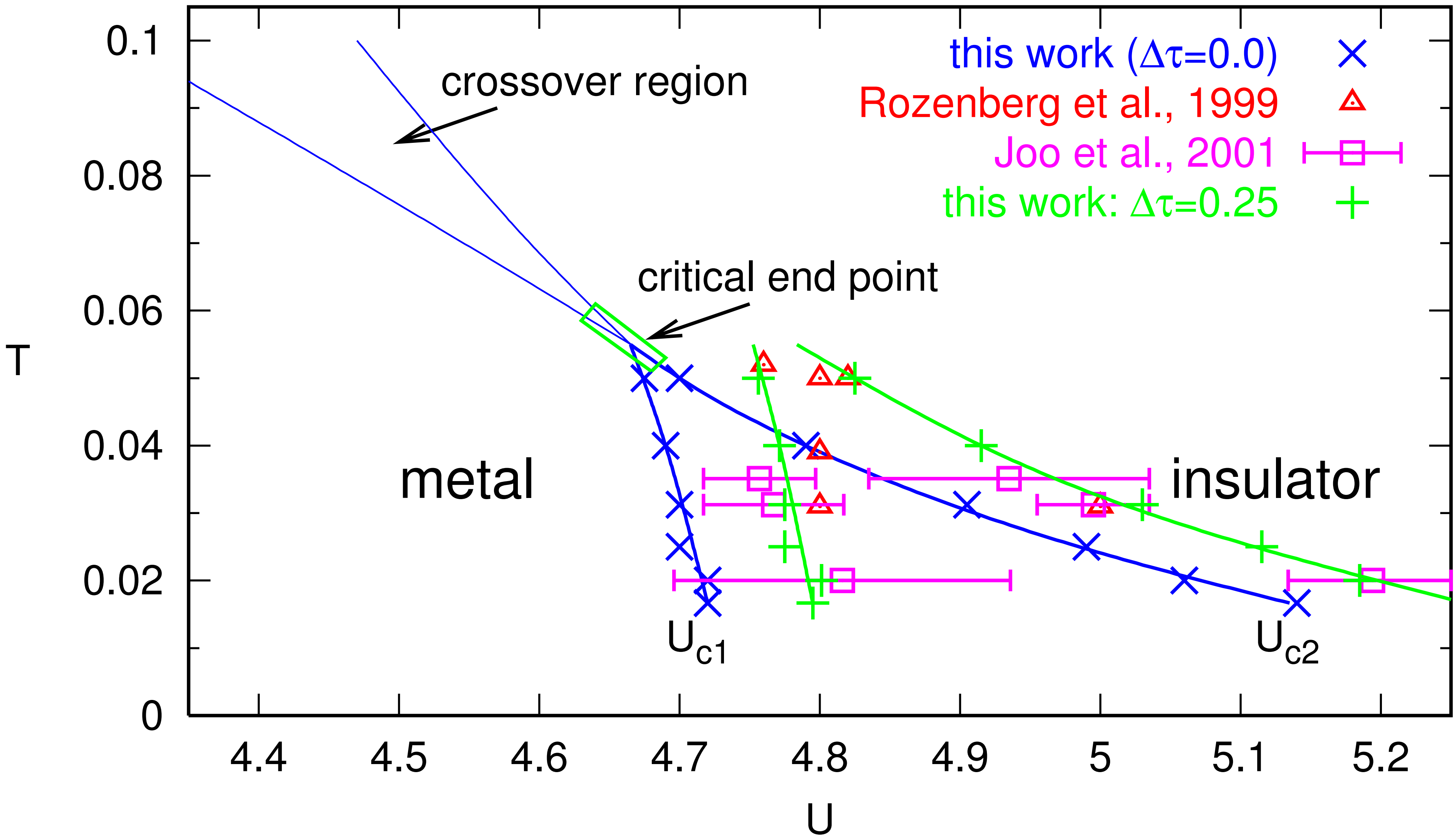


extrapolation $\Delta\tau \to 0$ is quantitatively important

# Thermodynamic first-order phase transition line $U_c(T)$

## Differential equation for $U_c(T)$

MIT line determined by free energy $F$,

$$F = E - TS; \qquad dF = -S\,dT + D\,dU\,.$$

Problem: free energy $F$ and entropy $S$ cannot be computed within QMC (prefactor in $\mathcal{Z}$)

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Using

$$\left.\frac{\partial \beta F}{\partial \beta}\right|_U = F - T\left.\frac{\partial F}{\partial T}\right|_U = F + TS = E$$

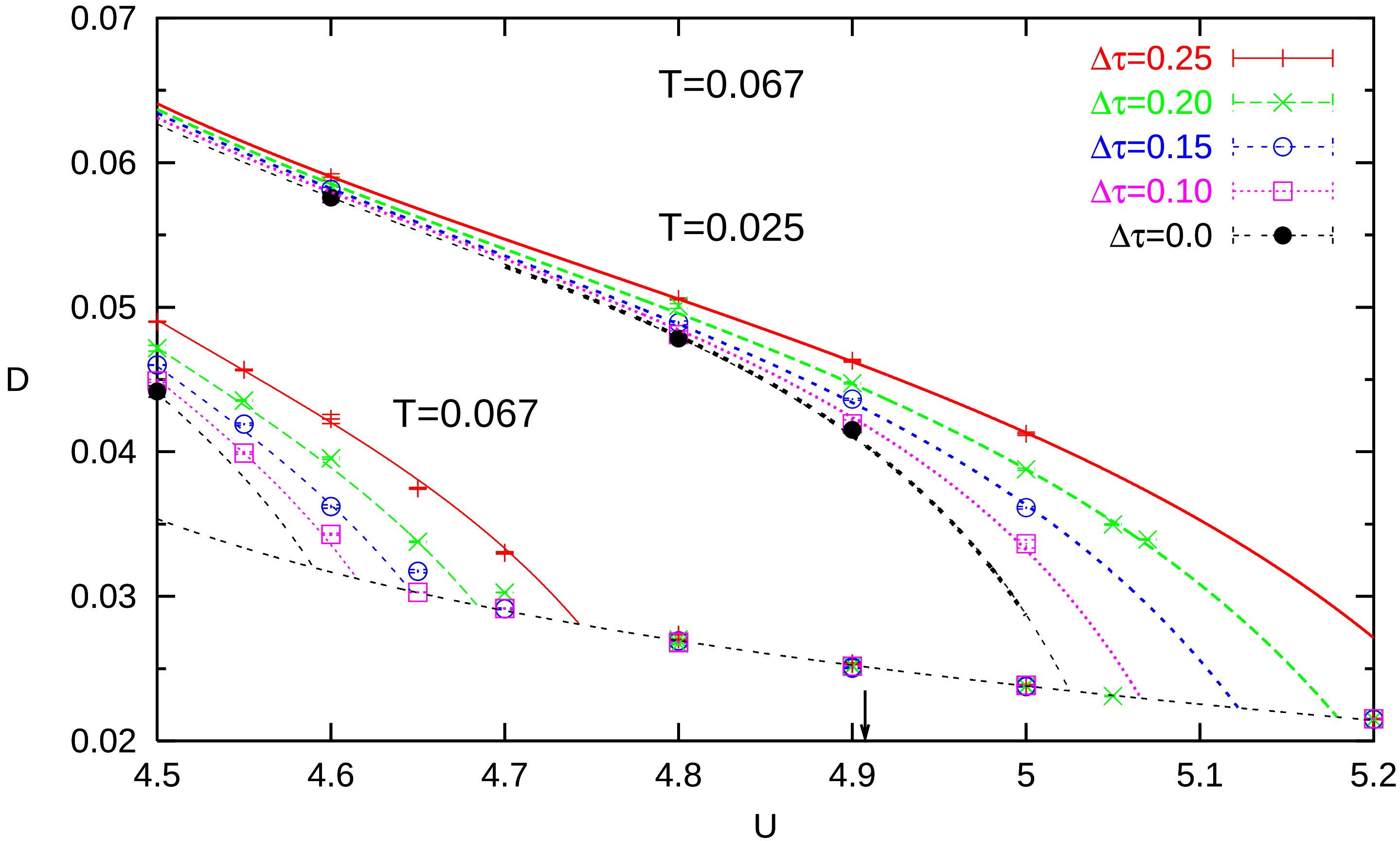
write differential for metal/insulator

$$\mathrm{d}\big(\beta F_{\mathrm{m/i}}(\beta, U)\big) = E_{\mathrm{m/i}}(\beta, U)\,\mathrm{d}\beta + \beta D_{\mathrm{m/i}}(\beta, U)\,\mathrm{d}U\,.$$

$d\big(\beta \Delta F(\beta, U)\big) = 0$ on smooth first-order MIT line (denoting $\Delta F \equiv F_{\mathrm{m}} - F_{\mathrm{i}}$ etc.)

$$\Rightarrow \quad \boxed{\frac{\mathrm{d}U_c(T)}{\mathrm{d}T} = f(T, U_c(T)); \;\; f(T,U) := \frac{\Delta E(T,U)}{T\,\Delta D(T,U)}}$$

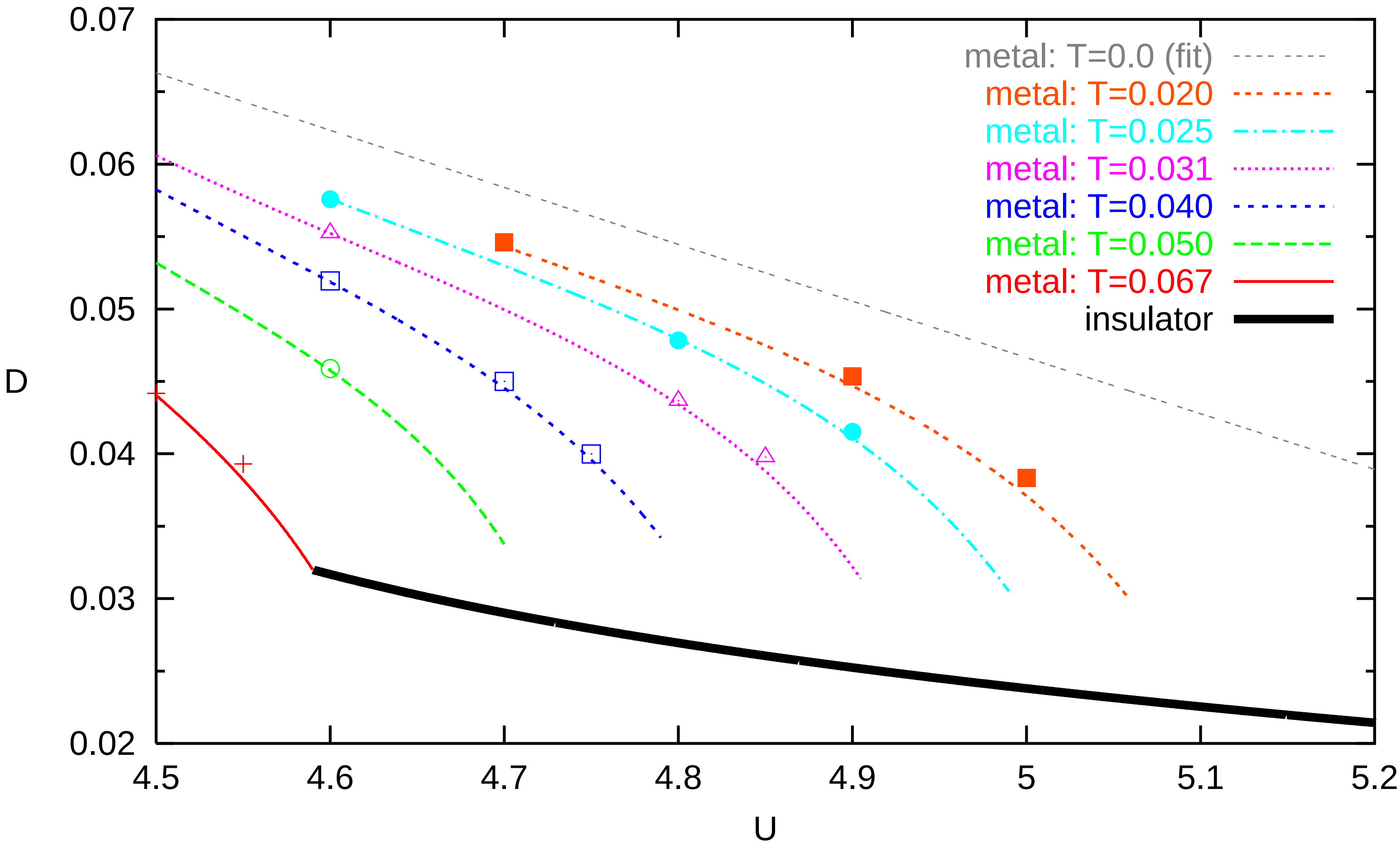
Extrapolation $\Delta\tau \to 0$ for double occupancy $D$



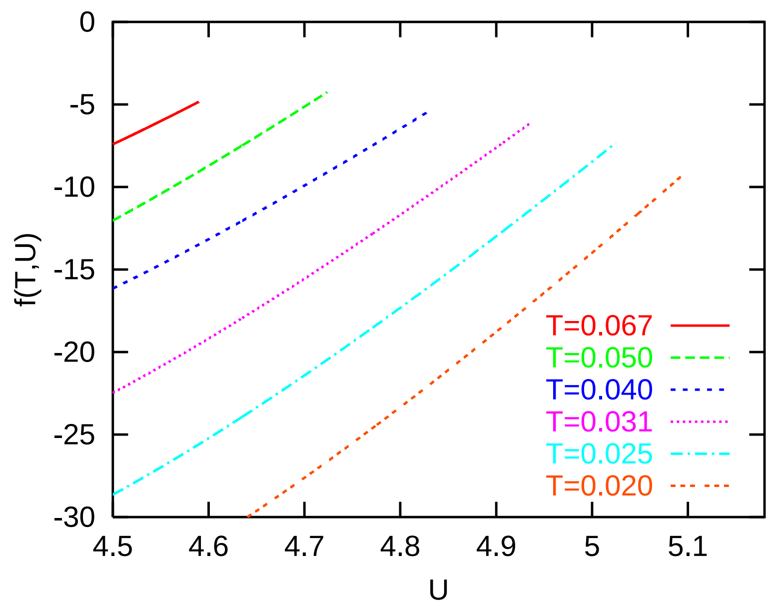

fits use extrapolations for energy ($\rightsquigarrow U_{\text{c2b}}$)

$$D_{\text{m}}(T, U, \Delta\tau) = D_i(U) + \frac{A_{T,\Delta\tau}\left[U_{\text{c2b}}(T, \Delta\tau) - U\right]}{1 + B_{T,\Delta\tau}[U_{\text{c2b}}(T, \Delta\tau) - U]}$$

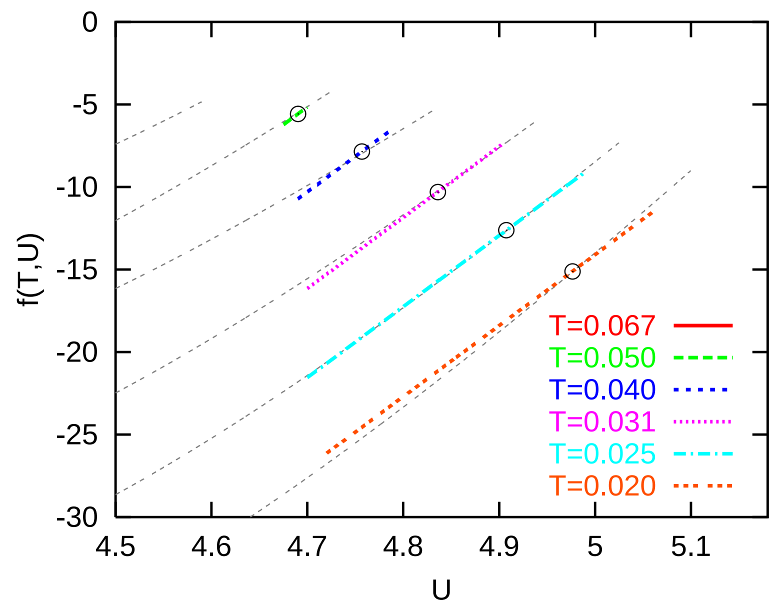
Double occupancy, extrapolated to $\Delta\tau = 0$




$$\Rightarrow \quad f(T,U) = \frac{\Delta E(T,U)}{T\,\Delta D(T,U)} \text{ known on grid}$$



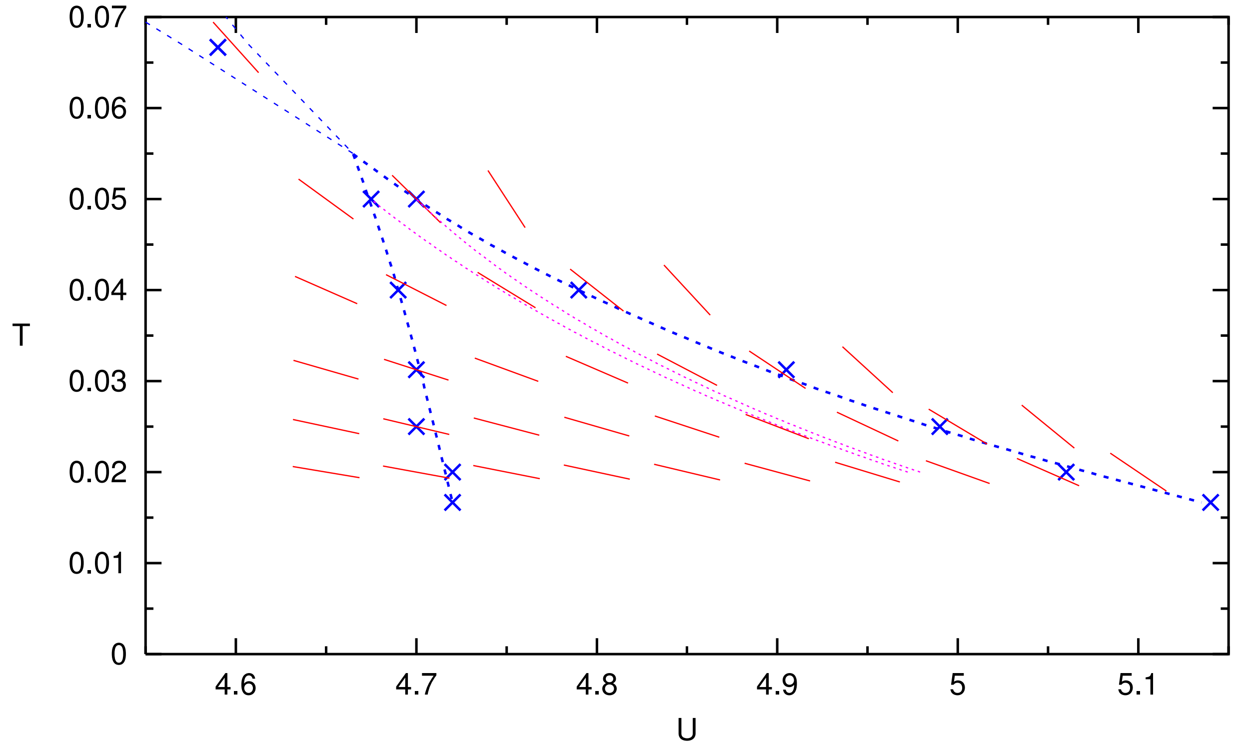

linearization $\longrightarrow$ in $U$




$$f(T,U) = A(T) + 43U$$

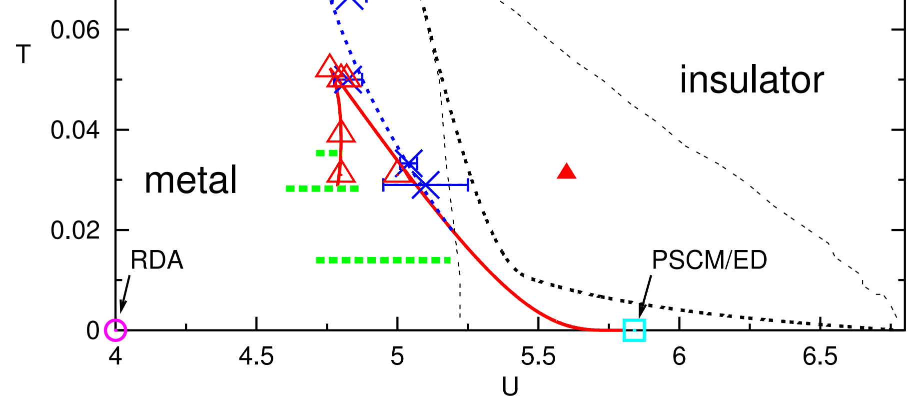
$\longrightarrow$ gradient field

stable integration of $U_c(T)$ for $T \gtrsim 0.02$



## Low-temperature asymptotics of $U_c(T)$

$U_c^0 \equiv U_{c2}(T=0) = 5.85 \pm 0.1$ (PSCT, ED, NRG)




Fermi liquid theory:
$$E_m(T,U) = E_m^0(U) + \tfrac{1}{2}\gamma(U)T^2; \quad S_m(T,U) = \gamma(U)T$$
$$E_i(T,U) = E_i^0(U); \quad S_i(T,U) = S_0$$

PSCT:
$$E_i^0(U) - E_m^0(U) = \frac{a}{2}(U - U_c^0)^2; \quad \gamma(U) = \frac{\gamma_0}{U_c^0 - U}$$

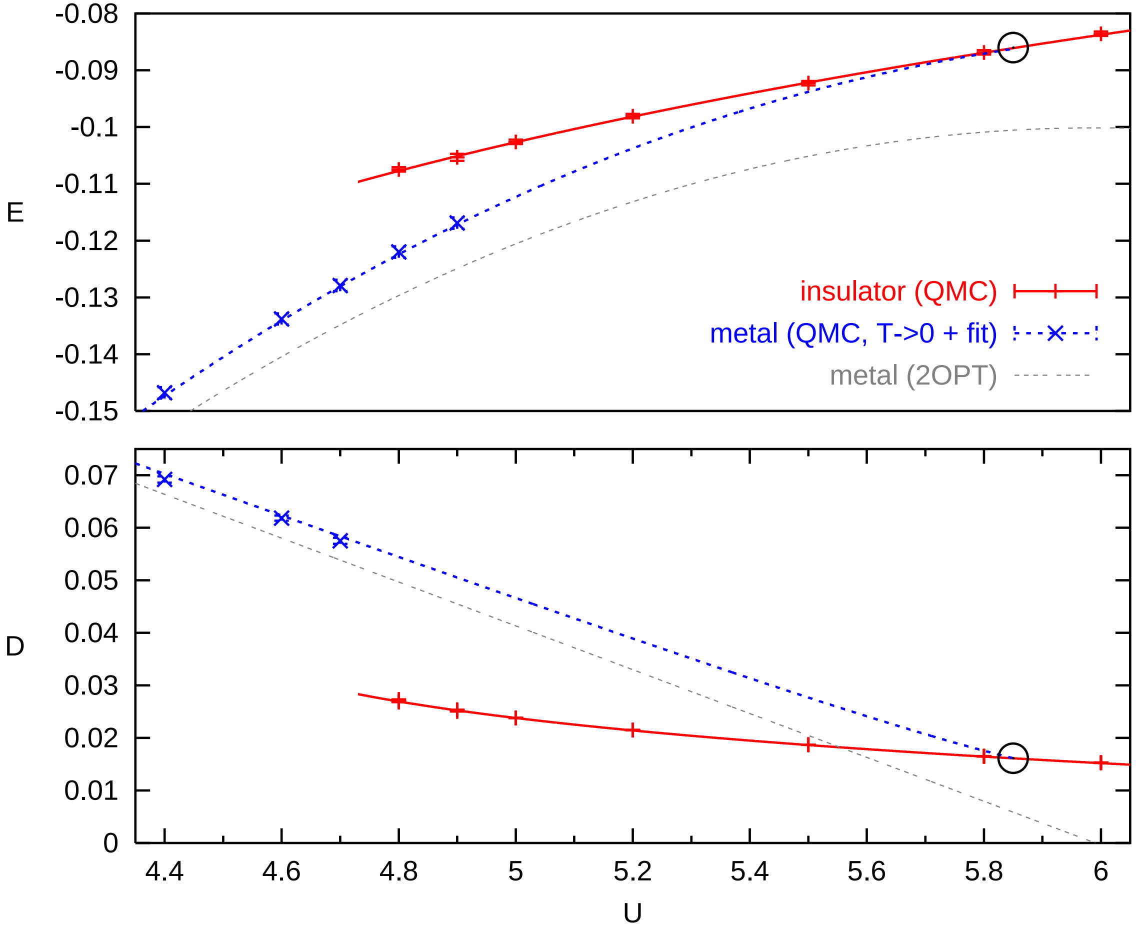
Equate free energies:
$$0 = \Delta F(T, U_c) \approx \frac{a}{2}(U - U_c^0)^2 + \frac{1}{2}\gamma(U)T^2 - TS_0$$

for low-$T$ solution
$$U_c(T) = U_c^0 - \sqrt{\frac{2S_oT}{a} + \frac{\gamma_0}{4S_0}T} + \mathcal{O}(T^{3/2})$$

Consequently: $\dfrac{\mathrm{d}U_c(T)}{\mathrm{d}T} = -\sqrt{\dfrac{S_o}{2aT}} + \mathcal{O}(1), \qquad A(T) = -\sqrt{\dfrac{S_o}{2a}}\,T^{-1/2} + \mathcal{O}(T^0)$

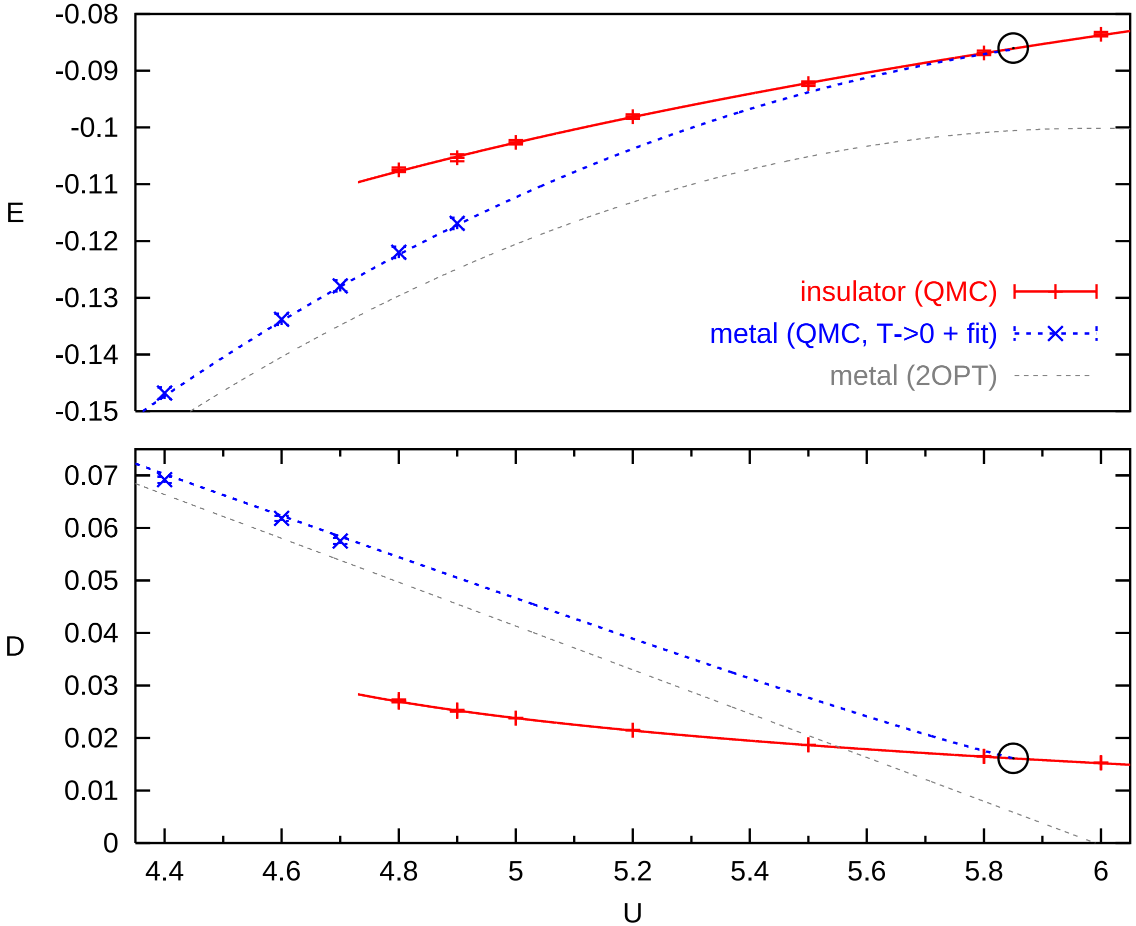
But: no reliable (PSCT) estimate for $D_m(U)$ and $a = \Delta(dD/dU)$

# Second order perturbation theory and beyond

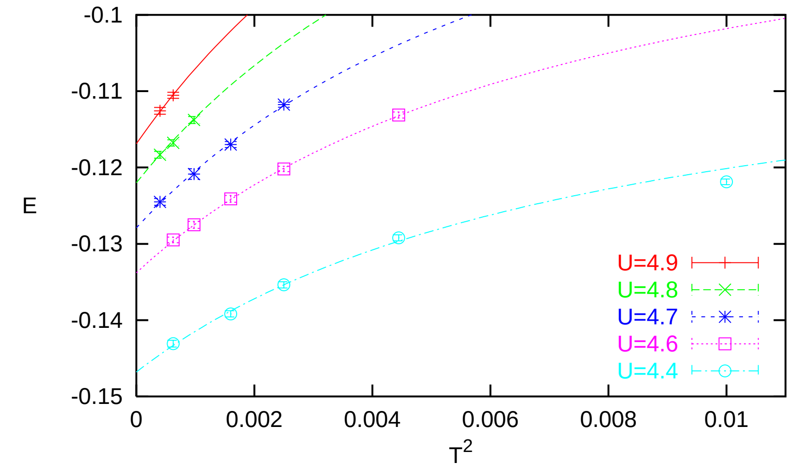



$$a \approx 0.0207 \quad \Rightarrow \quad A(T) = -4.1\, T^{-1/2} + \mathcal{O}(T^0)$$

# Second order perturbation theory and beyond



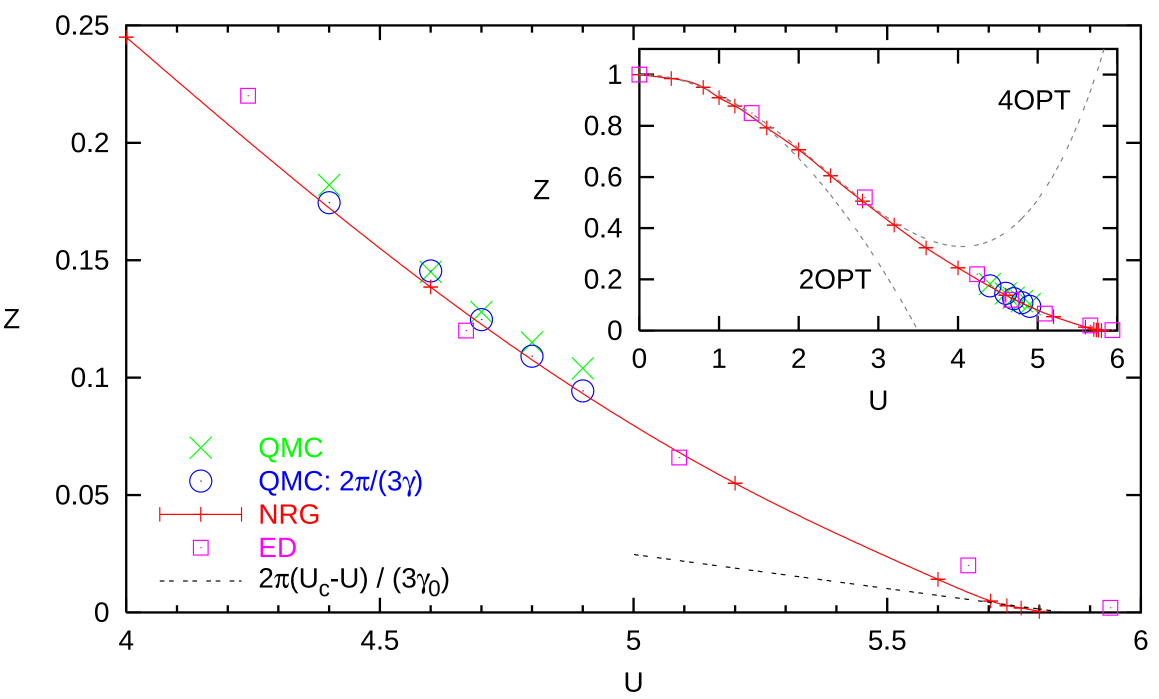

Extrapolation of QMC data $E(U,T)$

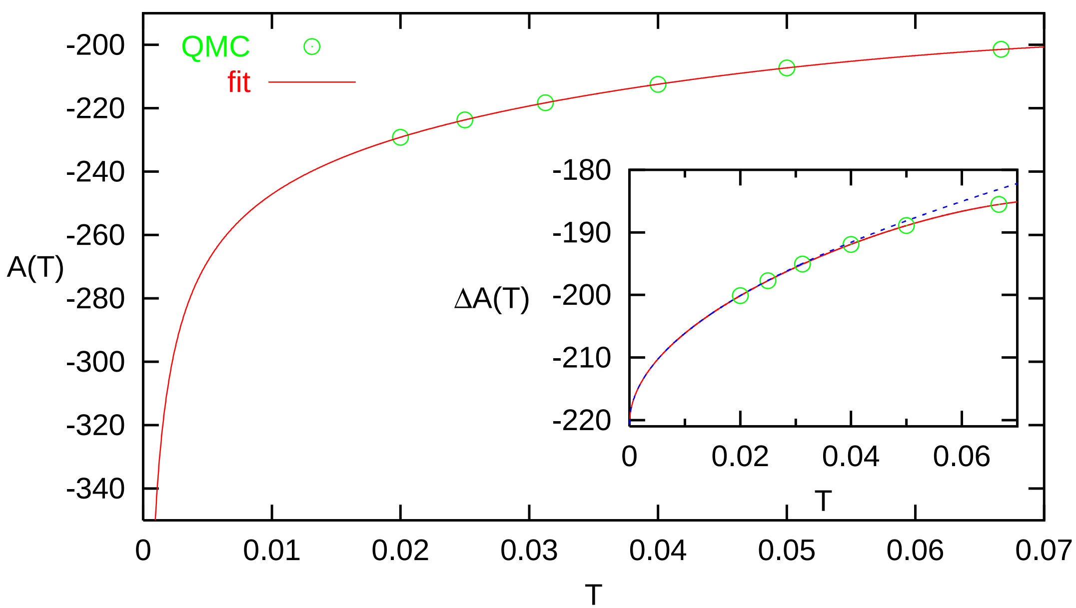



$\rightsquigarrow E(U, T=0),\ \gamma(U)$

$$a \approx 0.0207 \quad \Rightarrow \quad A(T) = -4.1\, T^{-1/2} + \mathcal{O}(T^0)$$

good consistency $\gamma \leftrightarrow Z$



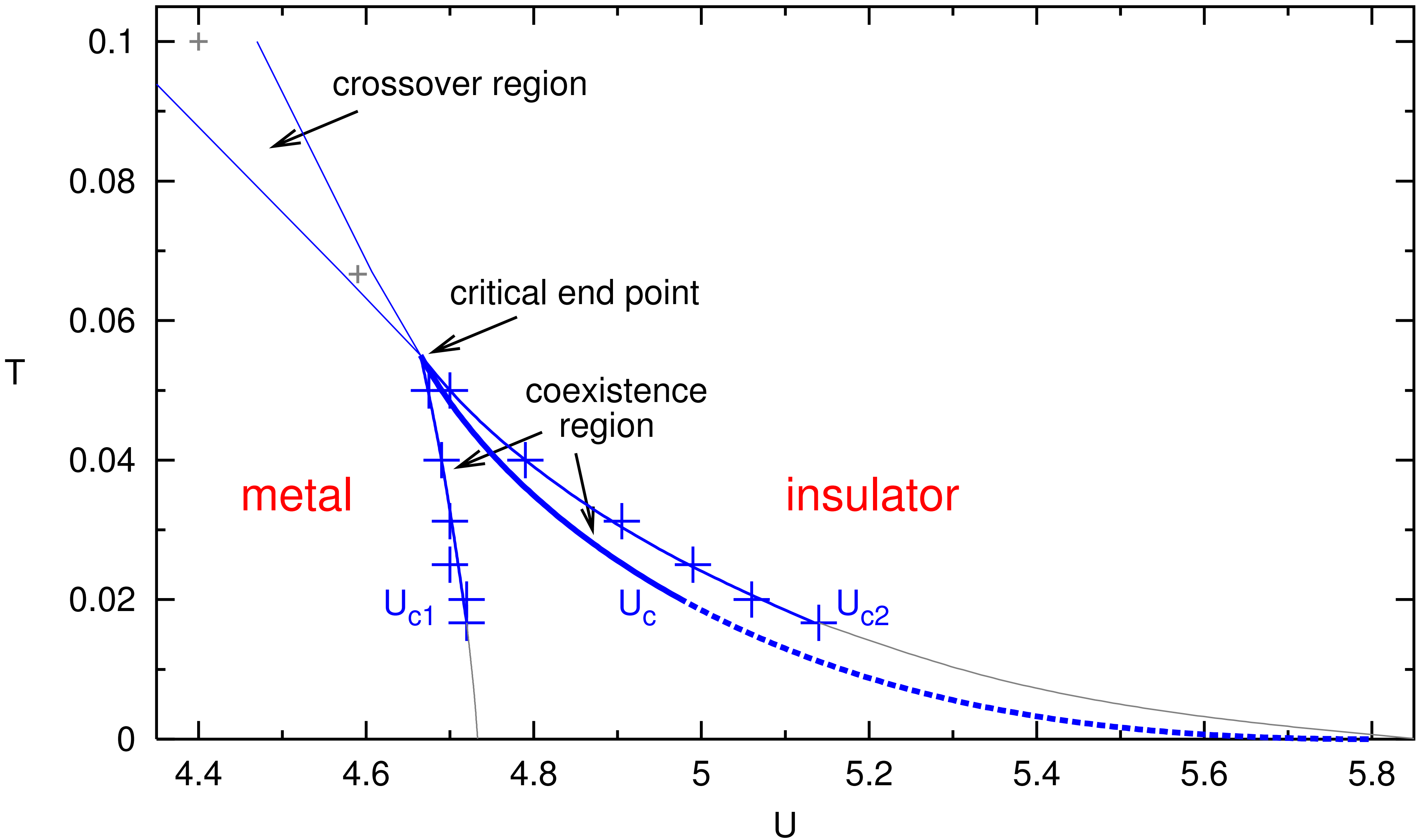

differential equation fully determined:

$$\frac{\mathrm{d}U_c(T)}{\mathrm{d}T} = f(T, U_c(T))$$

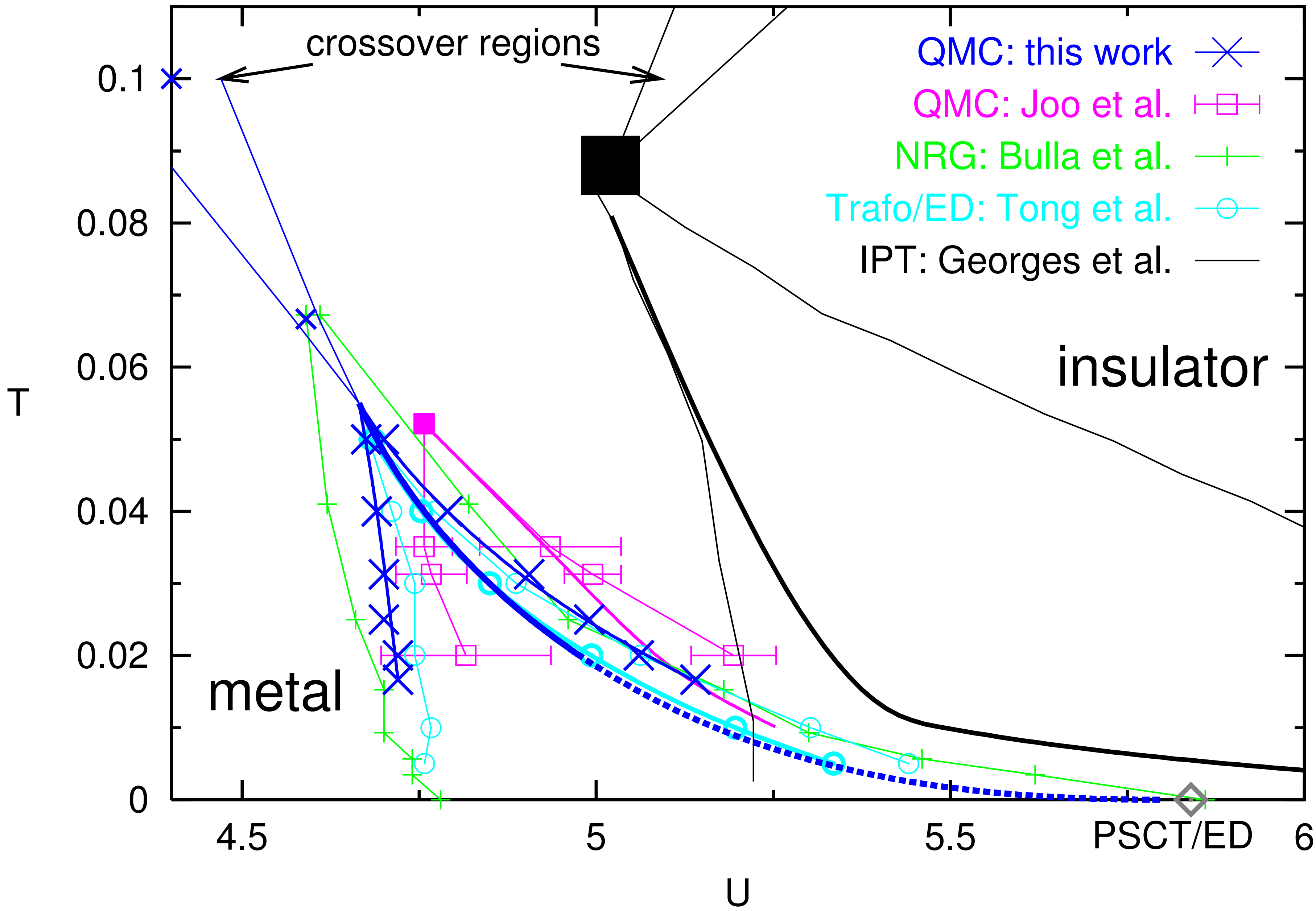
$$f(T,U) = A(T) + 43U$$

$$A(T) = -4.1T^{-1/2} + A_1 + (A_2T)^{1/2} + (A_3T)^4$$

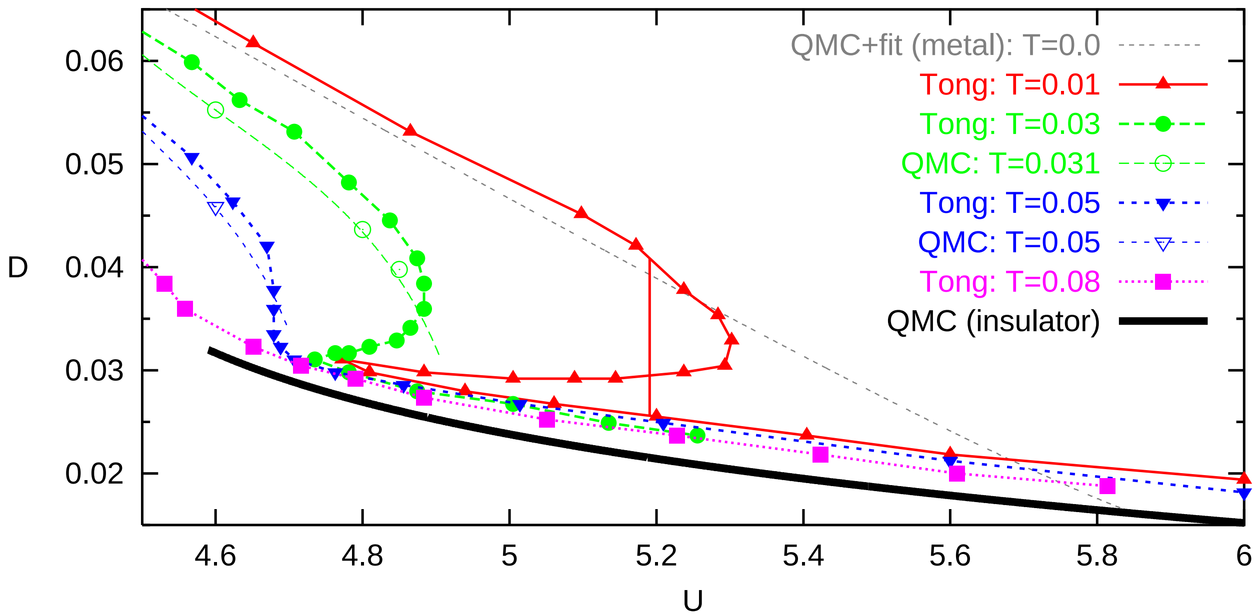
## Full MIT phase diagram



## Comparison with literature



# Transformation technique [Tong et al., PRB **64**, 235109 (2001)]




$U \longrightarrow \tilde{U} = U - c\, G(\tau = \beta/2)$

$\rightsquigarrow$ continuous curve $D(\tilde{U})$

back trafo $\rightsquigarrow$ s-shaped curves

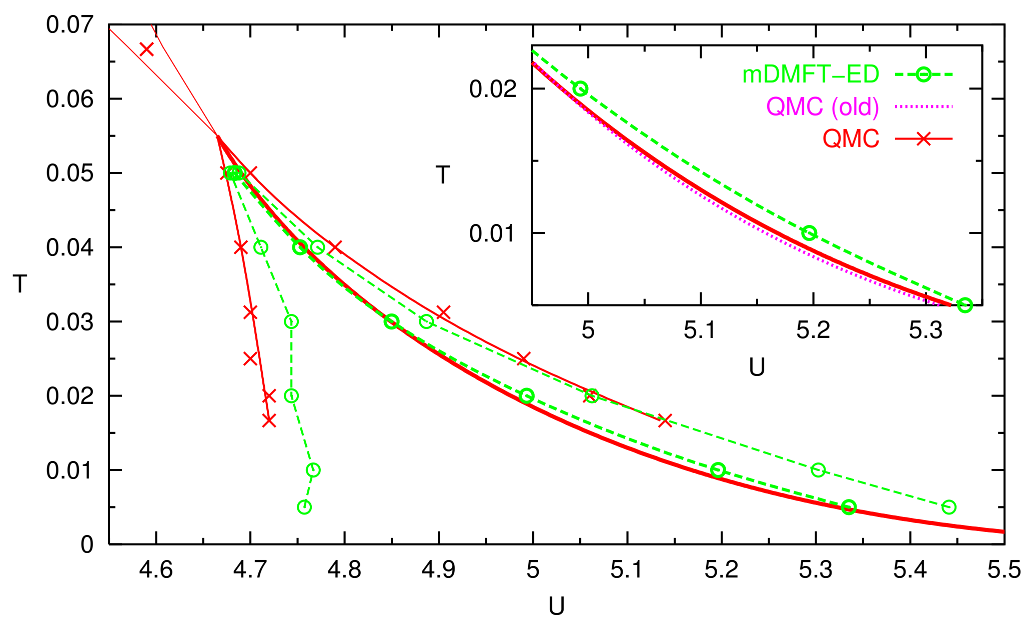
Maxwell construction $\rightsquigarrow$ $U_c$

scheme is approximate:

$$\Sigma(U,\omega) = \frac{U^2}{4\omega} + \mathcal{O}(\omega^{-2}) \neq \Sigma(\tilde{U},\omega)$$

$\rightsquigarrow$ wrong second derivative of $G(\tau)$

still: very good agreement for $U_c(T)$



# Conclusion

Inaccuracy of Schlipf et al.'s results traced back to non-optimal Fourier transformation scheme (larger $\Delta\tau$ error in insulator)

Improved FT algorithm as accurate (at small/moderate $\omega$) as good spline algorithms

Very precise determination of coexistence phase diagram

First controlled determination of thermodynamic first-order MIT line

- differential equation for $U_c(T)$
- ground state properties $E(U)$, $D(U)$ in insulating phase
- inclusion of Fermi liquid properties
- fit guided by second-order PT and PSCT/ED/NRG result at $T = 0$
- numerous checks of consistency

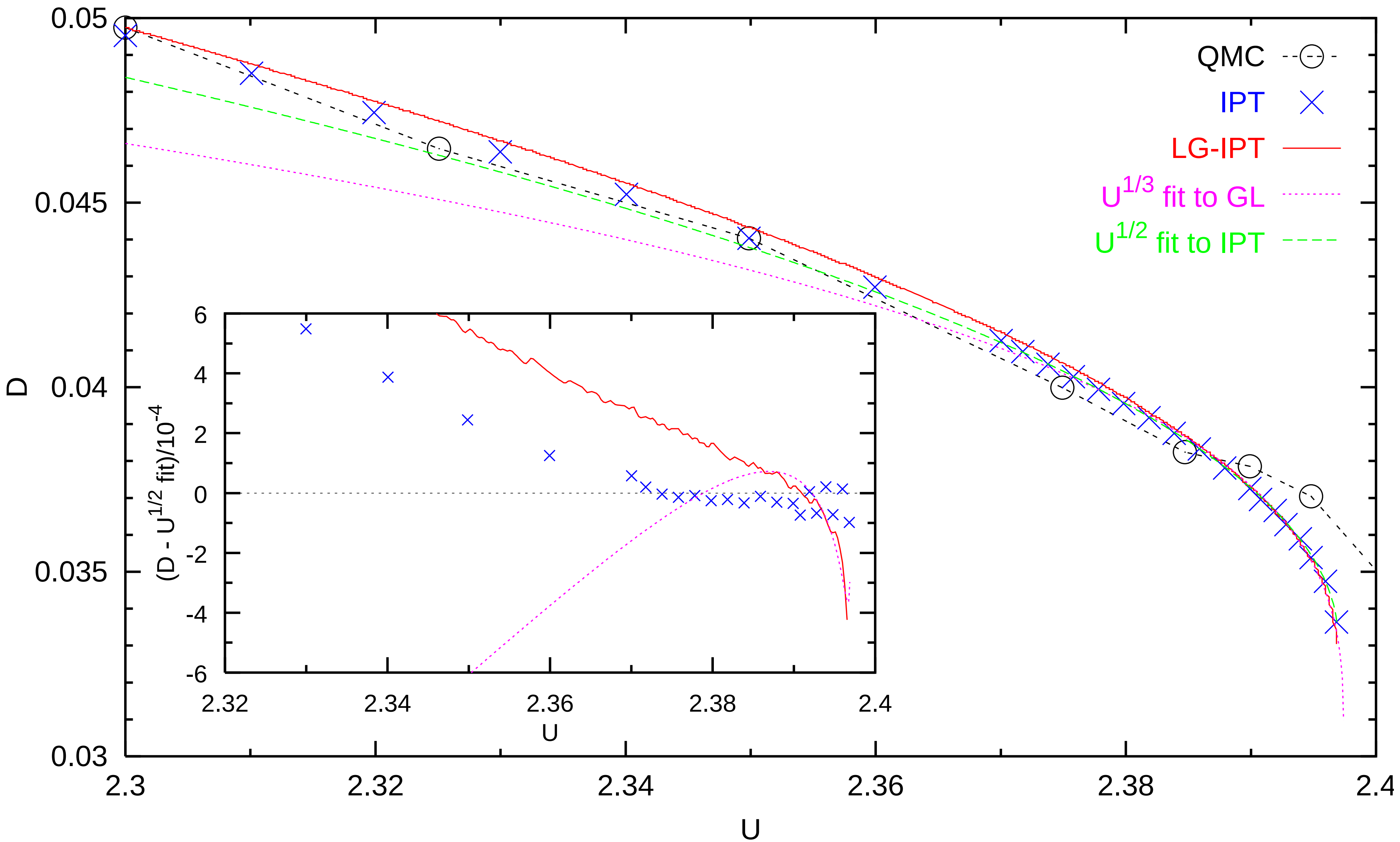
Critical behavior barely observable within QMC (difficult even within IPT)

Remaining mystery: RDA results

Advanced spline FT scheme with analytic high-energy corrections (Knecht/Blümer)

Direct computations of free energy differences from Ginzburg-Landau functional

# Critical behavior: Landau theory [Kotliar et al., PRL **84**, 5180 (2000)]




Asymptotic exponent of IPT is $1/2$, not $1/3$!