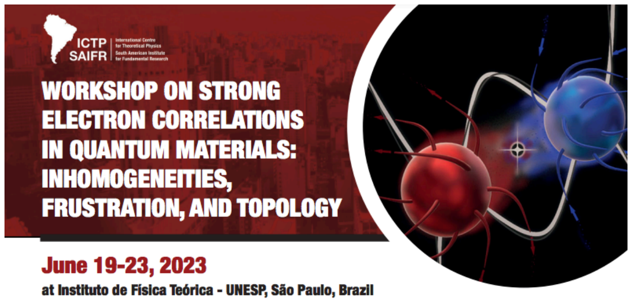
# Geometrical effects on thermopower properties of correlated electrons

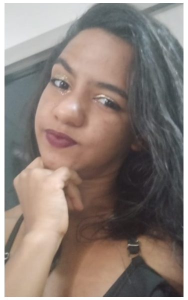
Thereza Paiva

Quantum Matter

UFRJ



# Collaborators



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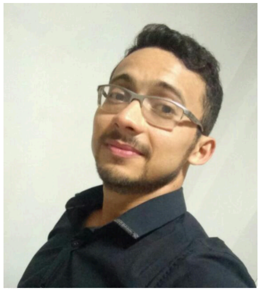
Natanael C Costa



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THE OHIO STATE UNIVERSITY

**ArXiv:2303.16291**

**Effects of lattice geometry on thermopower properties of the repulsive Hubbard model**


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# Summary

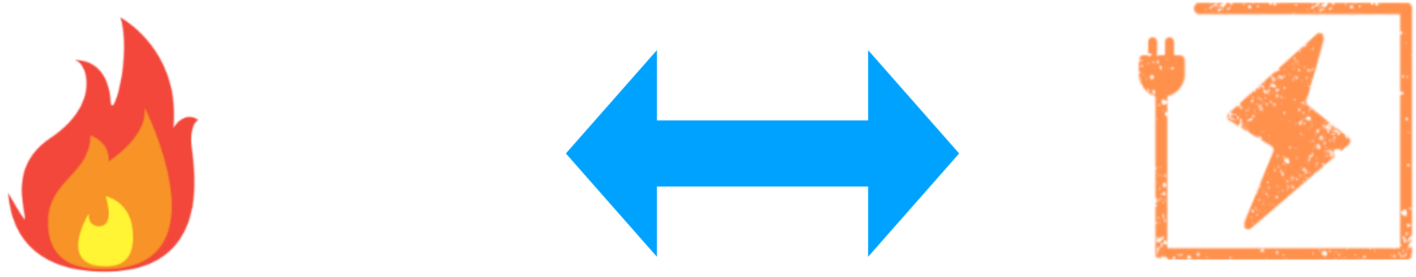
Thermoelectric materials
and thermopower

Hubbard Model

Seebeck coefficient and Power factor

Conclusions

# Thermoelectric effects



Thermic energy

Electric energy

Thermoelectric materials: induced voltage in the presence of temperature gradient

# Thermoelectric materials

Figure of merit

$$ZT = \frac{S^2 \sigma T}{\kappa}$$

$S$ → Thermopower or Seebeck coefficient

Conversion efficiency from thermal to electrical energy

$\sigma$ → Conductivity

$\kappa$ → Thermal conductivity

G. Mahan, B. Sales and J. Sharp, Physics Today 50, 3, 42 (1997)

# Thermoelectric materials

Figure of merit

$$ZT = \frac{S^2 \sigma T}{\kappa}$$

$S$ → Thermopower or Seebeck coefficient

Conversion efficiency from thermal to electrical energy

$\sigma$ → Conductivity

$\kappa$ → Thermal conductivity

Metals → Low $S$

Insulators → Low $\sigma$

Doped insulators → Best choice?

# Thermoelectric materials

## Figure of merit

$$ZT = \frac{S^2 \sigma T}{\kappa}$$

$S$ → Thermopower or Seebeck coefficient

Conversion efficiency from thermal to electrical energy

$\sigma$ → Conductivity

$\kappa$ → Thermal conductivity

## Power Factor

$$PF = S^2 \sigma$$

# Correlated materials

$Na_xCoO_2$ → Large Seebeck coefficient

Wissgott et al PRB82 (10), Wissgott et al PRB 84 (11)

Cuprates → Sign change of Seebeck coefficient near optimum doping

Obertelli et al PRB 46 (92), Tallon et al PRB 51 (95)

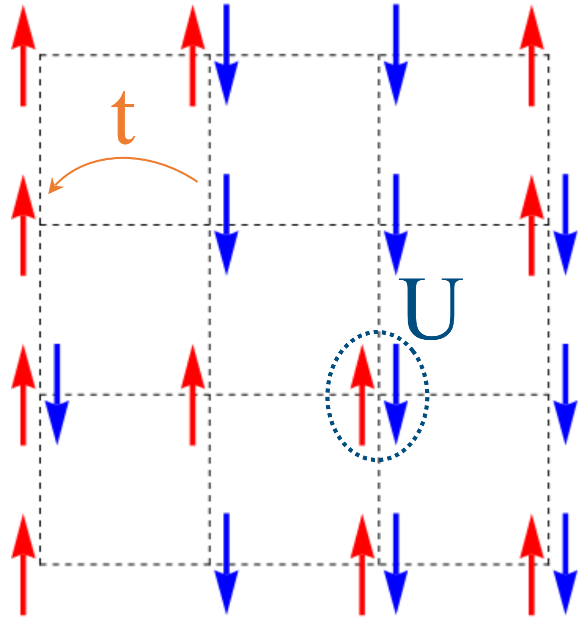
How is the thermopower affected by geometry?

How is thermopower affected by correlations?

# Hubbard Model

$$\mathcal{H} = -\mathrm{t} \sum_{\langle i,j \rangle,\sigma} \left( \hat{c}^{\dagger}_{i,\sigma} \hat{c}_{j,\sigma} + h.c. \right)$$

$$+\mathrm{U} \sum_{i} \left( \hat{n}_{i\uparrow} - \frac{1}{2} \right) \left( \hat{n}_{i\downarrow} - \frac{1}{2} \right) - \mu \hat{N}$$




Coulomb repulsion (U>0)

Hopping (t)

chemical potential ($\mu$)

No known analytic solution in 2D

QMC to study the Hubbard Model on square, triangular and honeycomb lattices

# Some details on our QMC simulations

- 100 sites
- 144 sites
- 162 sites

Each run:

2000 warm up sweeps,
5000 measurement sweeps

$\Delta\tau < 0.1$

$0 \leq U \leq 10$

Sweeps through density: 500 jobs for each temperature and interaction strength

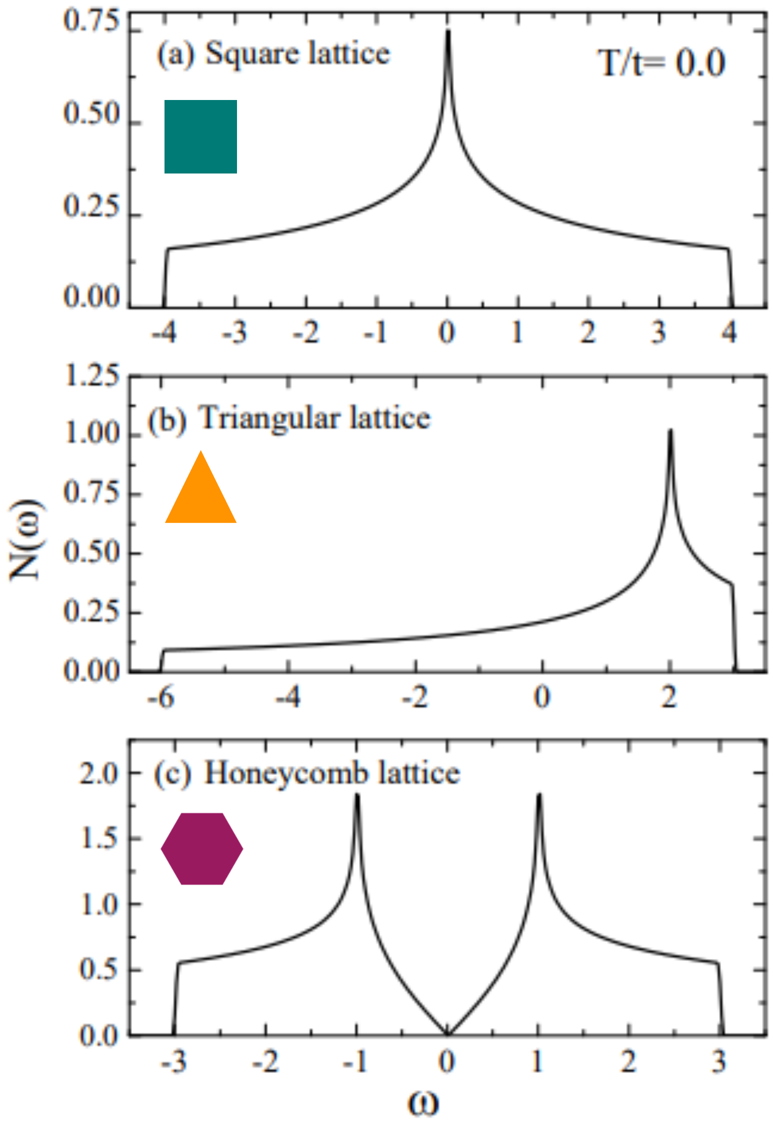
# Non-interacting Density of states

Dispersion relations

$$E(k) = -2t(\cos k_x + \cos k_y)$$

$$E(k) = -2t[\cos(\frac{k_x + \sqrt{3}k_2}{2})\cos(\frac{k_x - \sqrt{3}k_2}{2}) + \cos(k_x)]$$

$$E_{\pm}(k) = \pm t\sqrt{3 + 2\cos(k_x) + 2\cos(k_y) + 2\cos(k_x + k_y)}$$

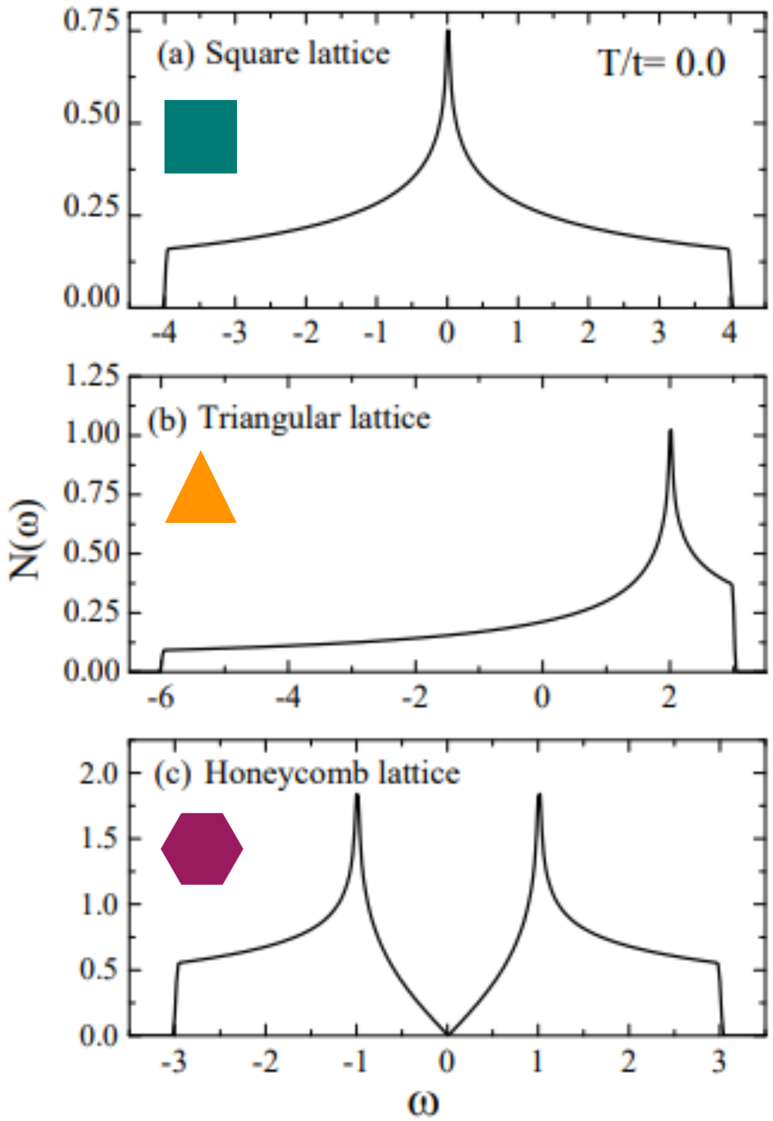


# Non-interacting Density of states

Van Hove singularity

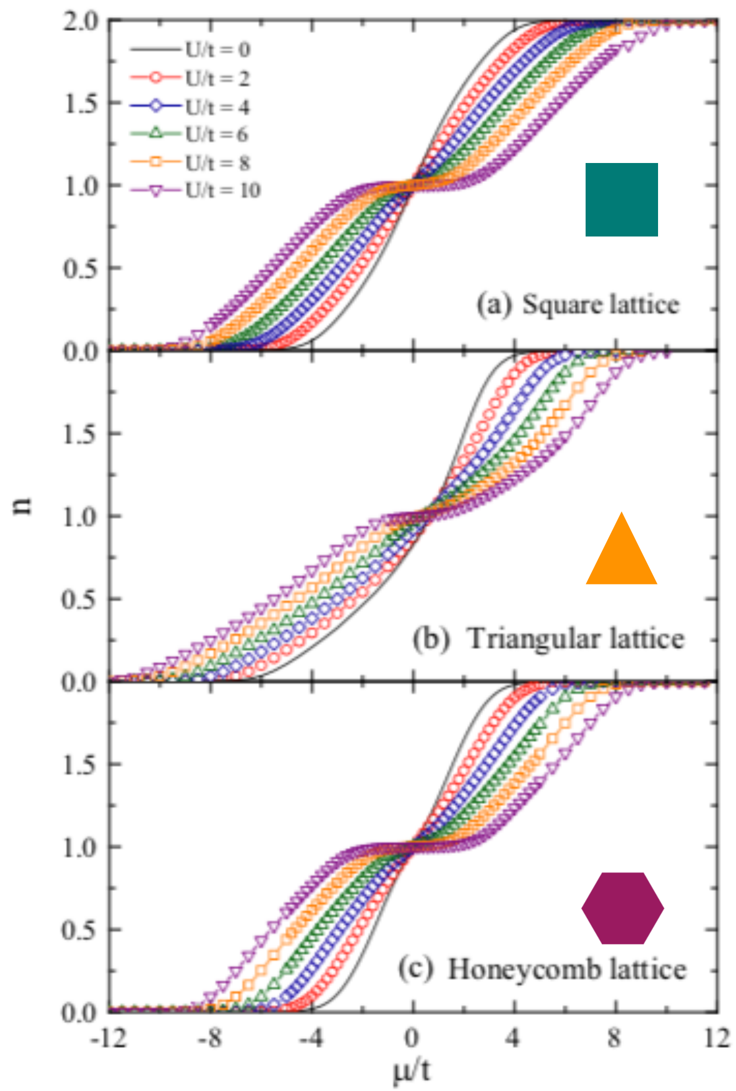
Particle-hole symmetry

Particle-hole symmetry



# Density

T/t=0.5




$n(-\mu) = 2 - n(\mu)$ → Particle-hole symmetry

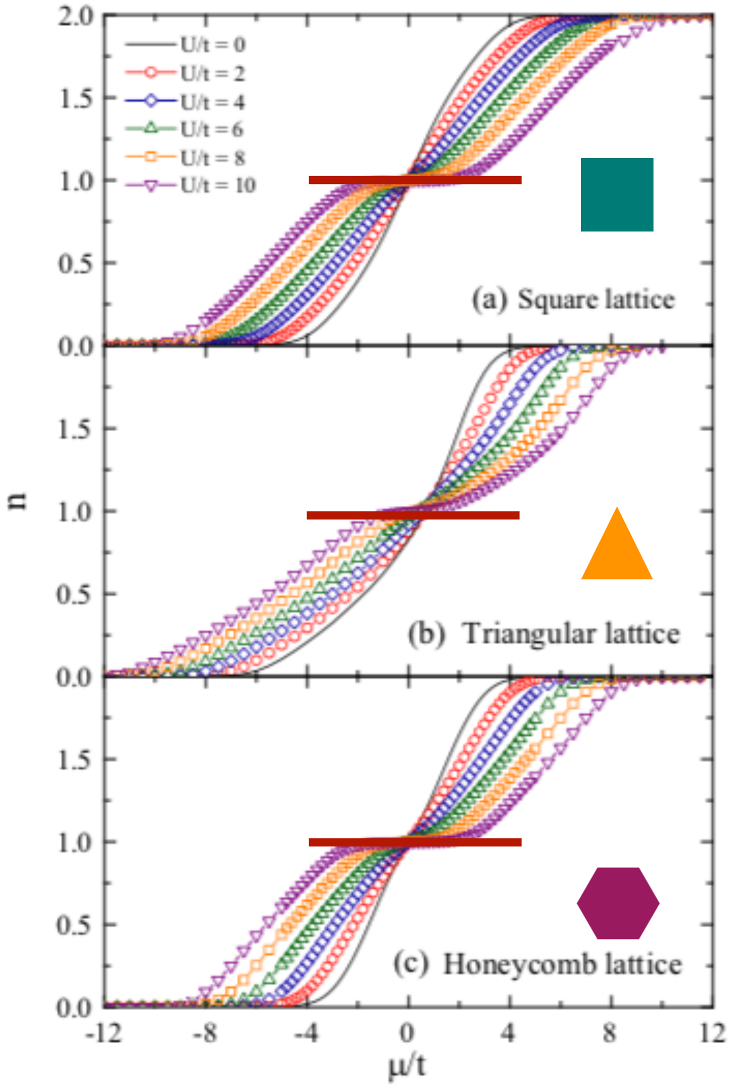
$n(-\mu) \neq 2 - n(\mu)$ → Non-bipartite

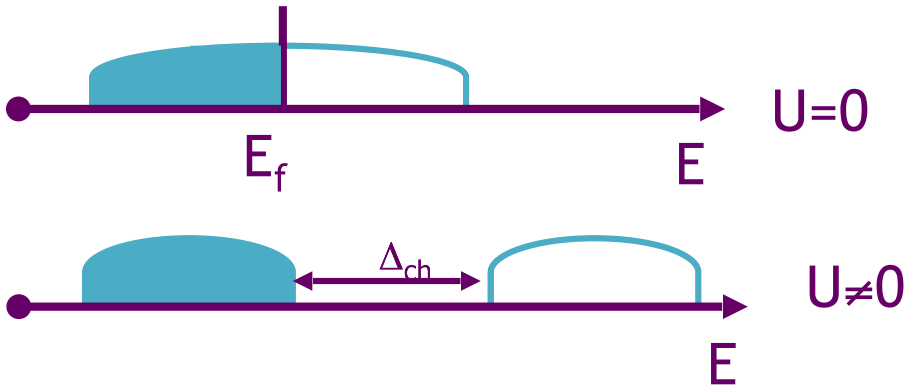
$n(-\mu) = 2 - n(\mu)$ → Particle-hole symmetry

# Density

T/t=0.5

Mott transition



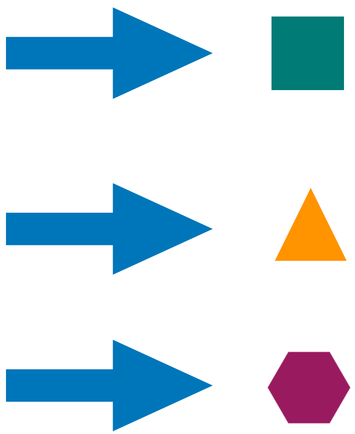

$U/t > 0$

$U_c/t \approx 7-8$

Yoshioka PRL 09,
Shirakawa PRB 17

$U_c/t \approx 3.8$

Otsuka PRX 16

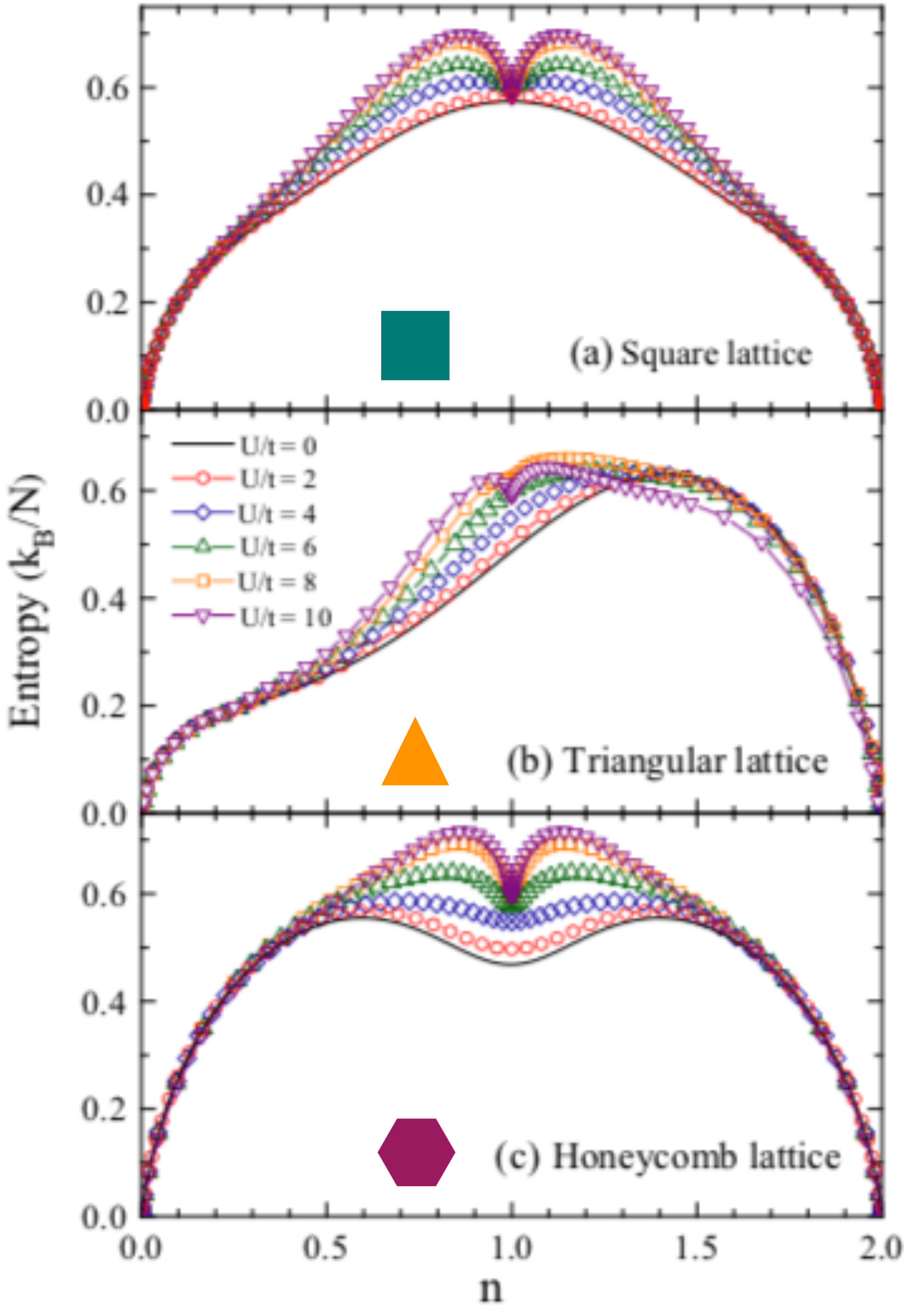


# Entropy

$$s(\mu, T) = \int_{-\infty}^{\mu} d\mu \frac{\partial n}{\partial T}\Big|_{\mu}$$

in units of $k_B$

T/t=0.5

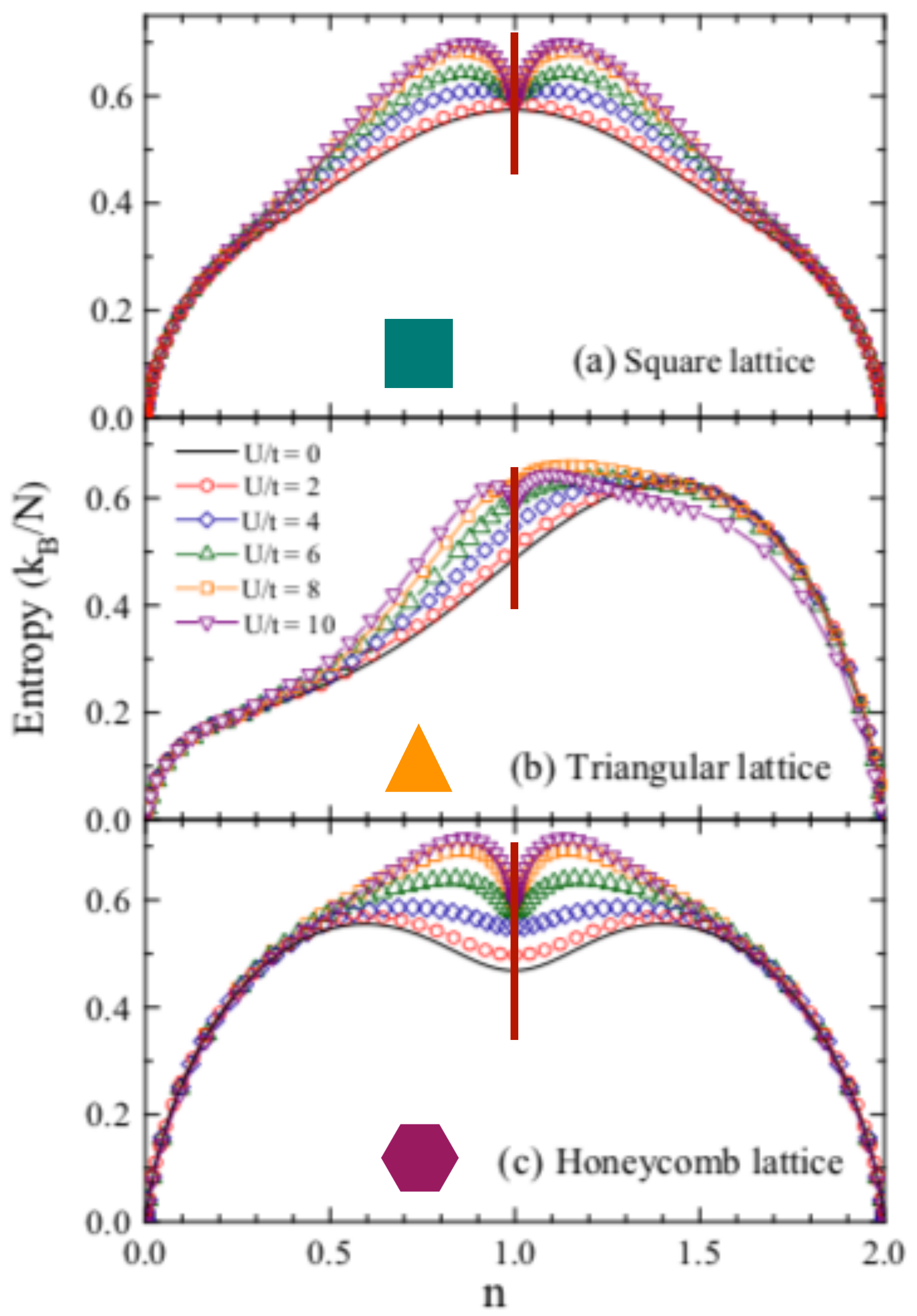


# Entropy

$$s(\mu, T) = \int_{-\infty}^{\mu} d\mu \frac{\partial n}{\partial T}\Big|_{\mu}$$

Correlations only play a role in a geometry dependent region around half-filling

Mott insulator has lower entropy than surrounding metal



## Density of States

## Conductivity

$$G(\mathbf{r}=0,\tau) = \int_{-\infty}^{\infty} d\omega \, \frac{e^{-\omega\tau}}{1+e^{-\beta\omega}} \, N(\omega)$$

$$\Lambda(\mathbf{q}=0,\tau) = \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \frac{e^{-\omega\tau}}{1-e^{-\beta\omega}} \, \mathrm{Im}\,\Lambda(\mathbf{q}=0,\omega)$$

## Key quantities obtained without inverting Laplace transforms

$$N(\omega=0,T) = \frac{dn}{d\mu} = n^2\kappa(T)$$

$$\sigma_{dc} \approx \frac{\beta^2}{\pi}\Lambda_{xx}(\mathbf{q}=\mathbf{0},\tau=\beta/\mathbf{2})$$

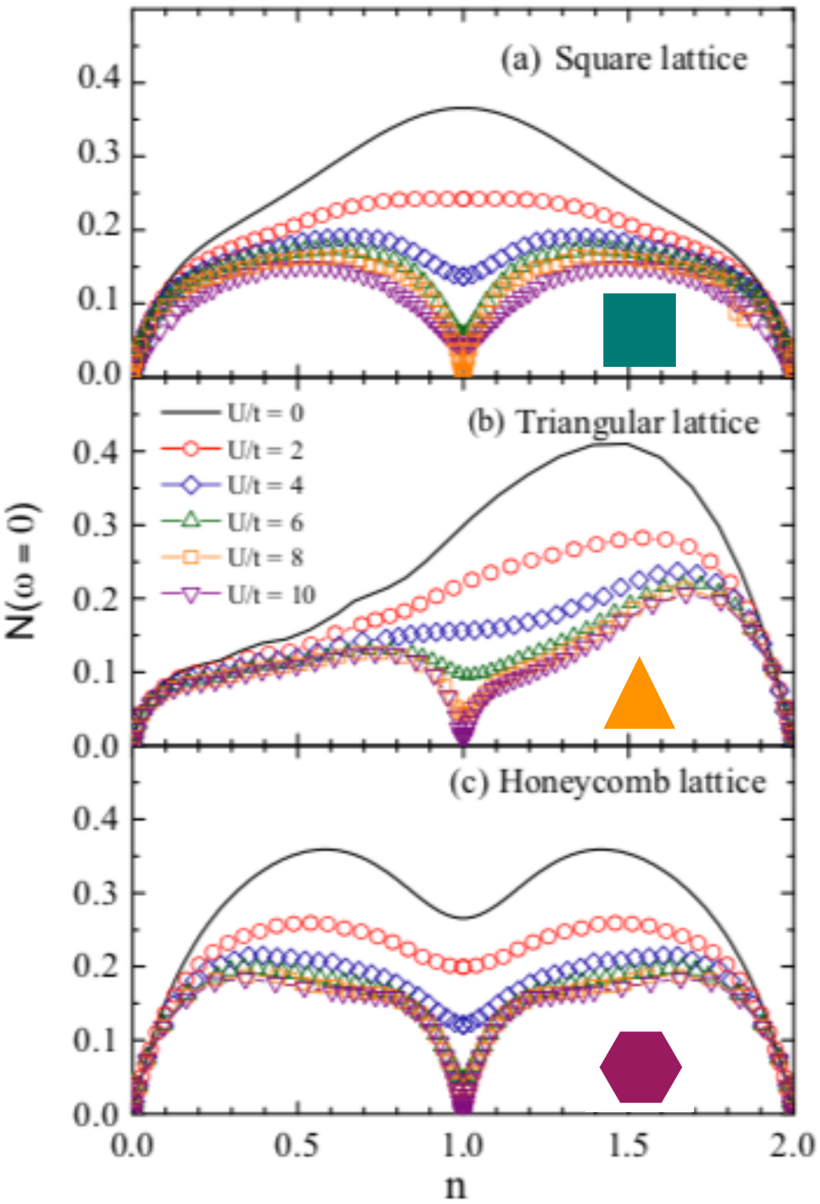
Current-current correlation function

$$\Lambda_{xx}(\mathbf{q},\tau) = \langle j_x(\mathbf{q},\tau) j_x(-\mathbf{q},0)\rangle$$

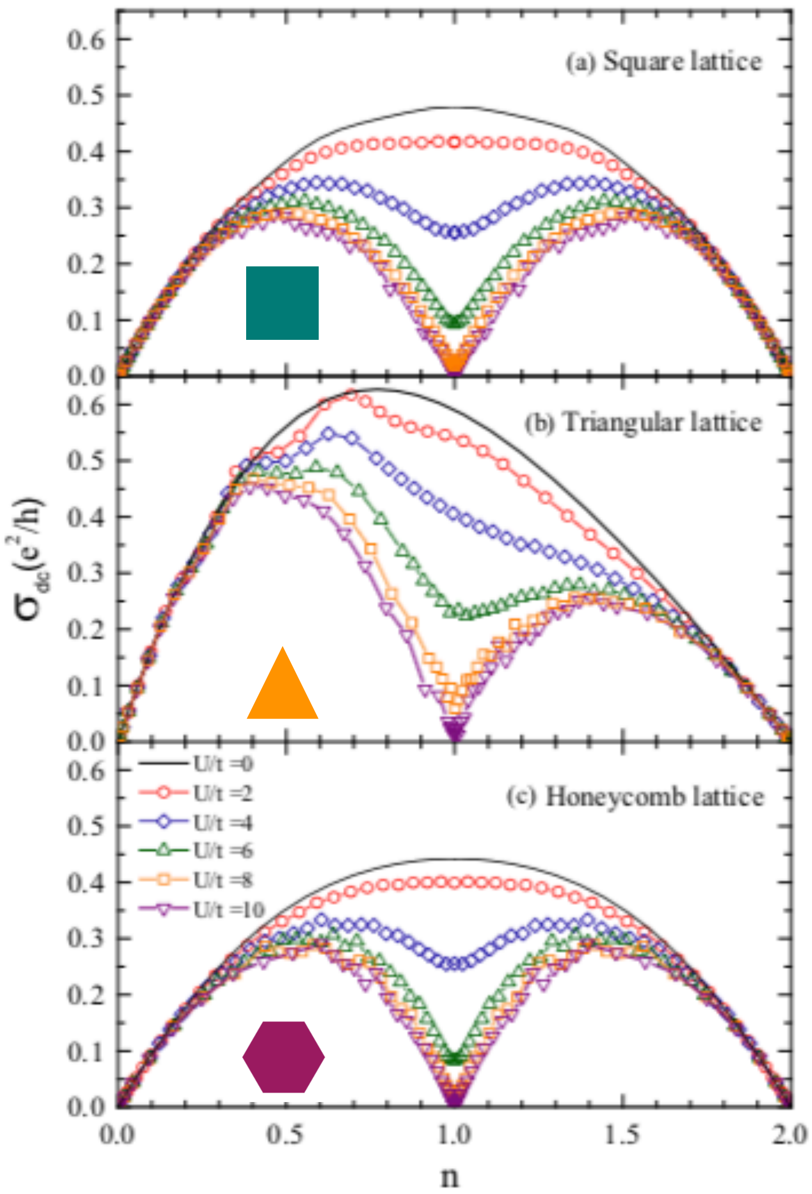
Unequal time current operator

$$j_x(\mathbf{i},\tau) = \mathrm{e}^{\tau\mathcal{H}}\left[it\sum_{\sigma}\left(c^{\dagger}_{\mathbf{i}+\mathbf{x},\sigma}c_{\mathbf{i},\sigma} - c^{\dagger}_{\mathbf{i},\sigma}c_{\mathbf{i}+\mathbf{x},\sigma}\right)\right]\mathrm{e}^{-\tau\mathcal{H}}$$

## Density of States




## Conductivity



# Seebeck coefficient

Kevin formula

$$S_{Kelvin} = -\frac{1}{e}\frac{\partial \mu}{\partial T}\Big|_{V,n} = \frac{\partial s}{\partial n}\Big|_{T,V}$$

Low frequency: $\hbar\omega < U$

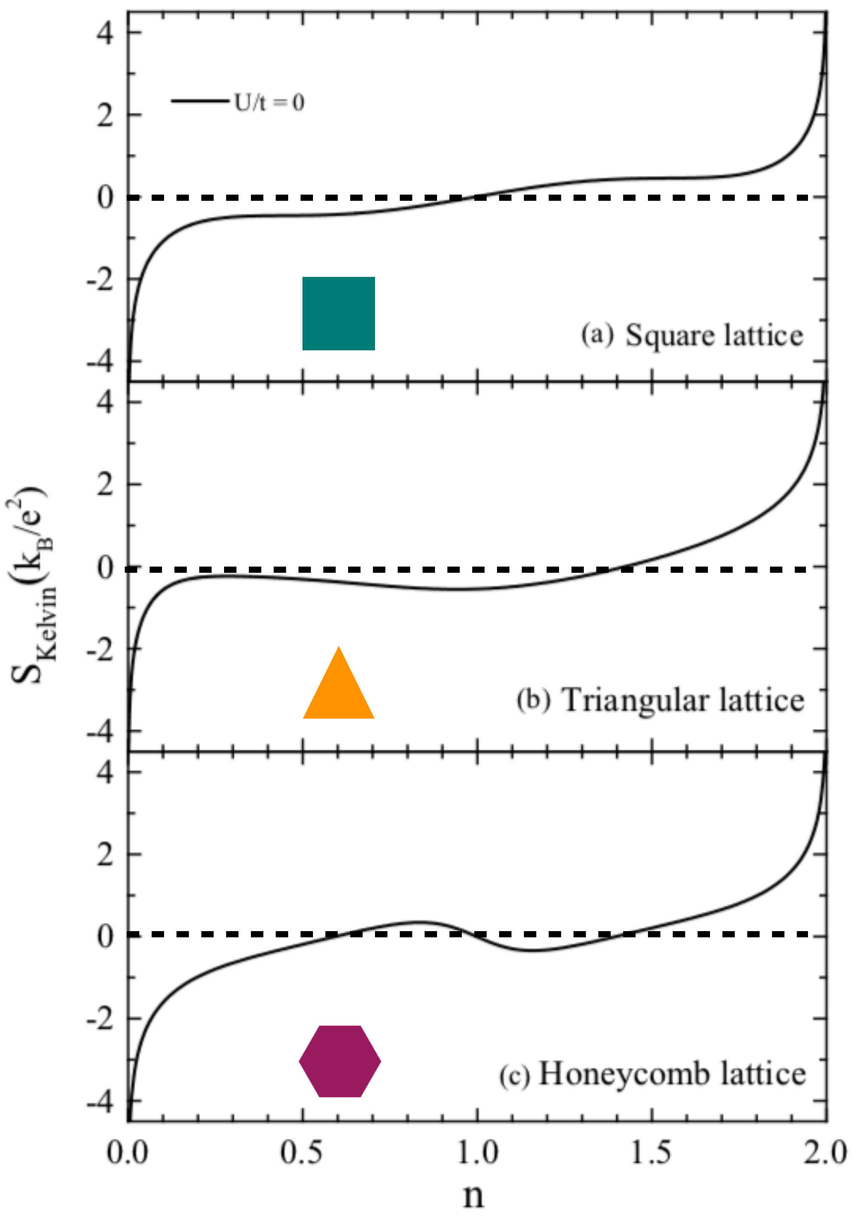
Sign is related to carrier:

negative for holes + and

positive for electrons -

# Seebeck coefficient

Non-interacting case



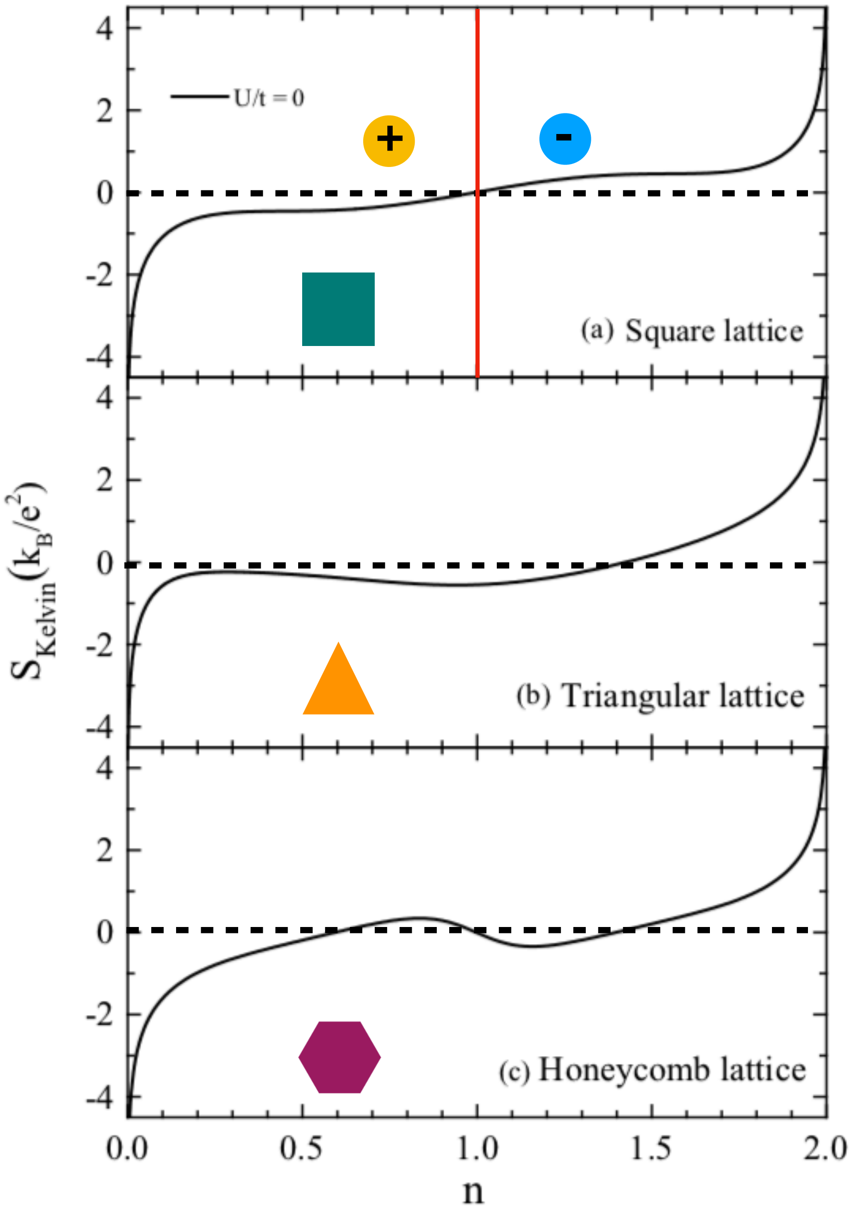

Geometry effects

T/t=0.5

Change of sign: change of carrier

# Seebeck coefficient

Non-interacting case




Geometry effects

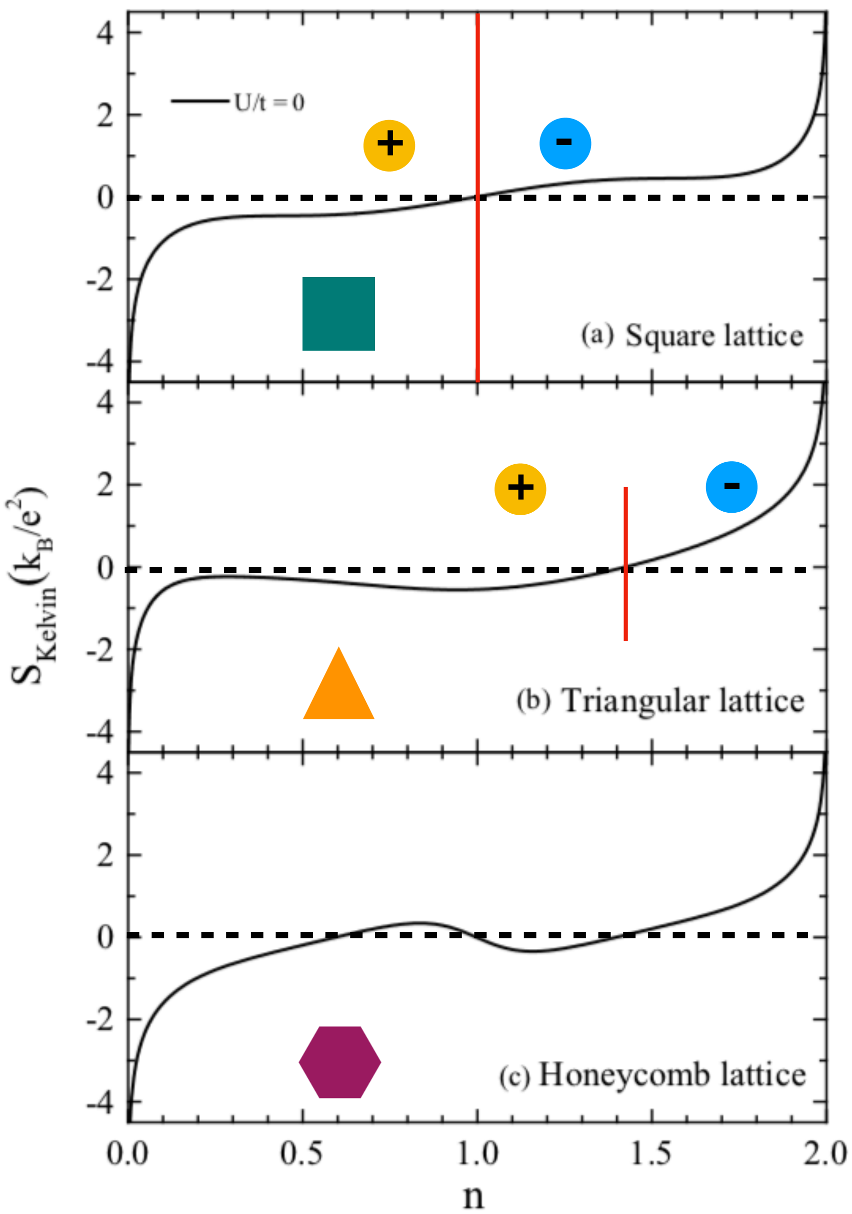
T/t=0.5

Change of sign: change of carrier

 Half-filling

# Seebeck coefficient

Non-interacting case




Geometry effects

T/t=0.5

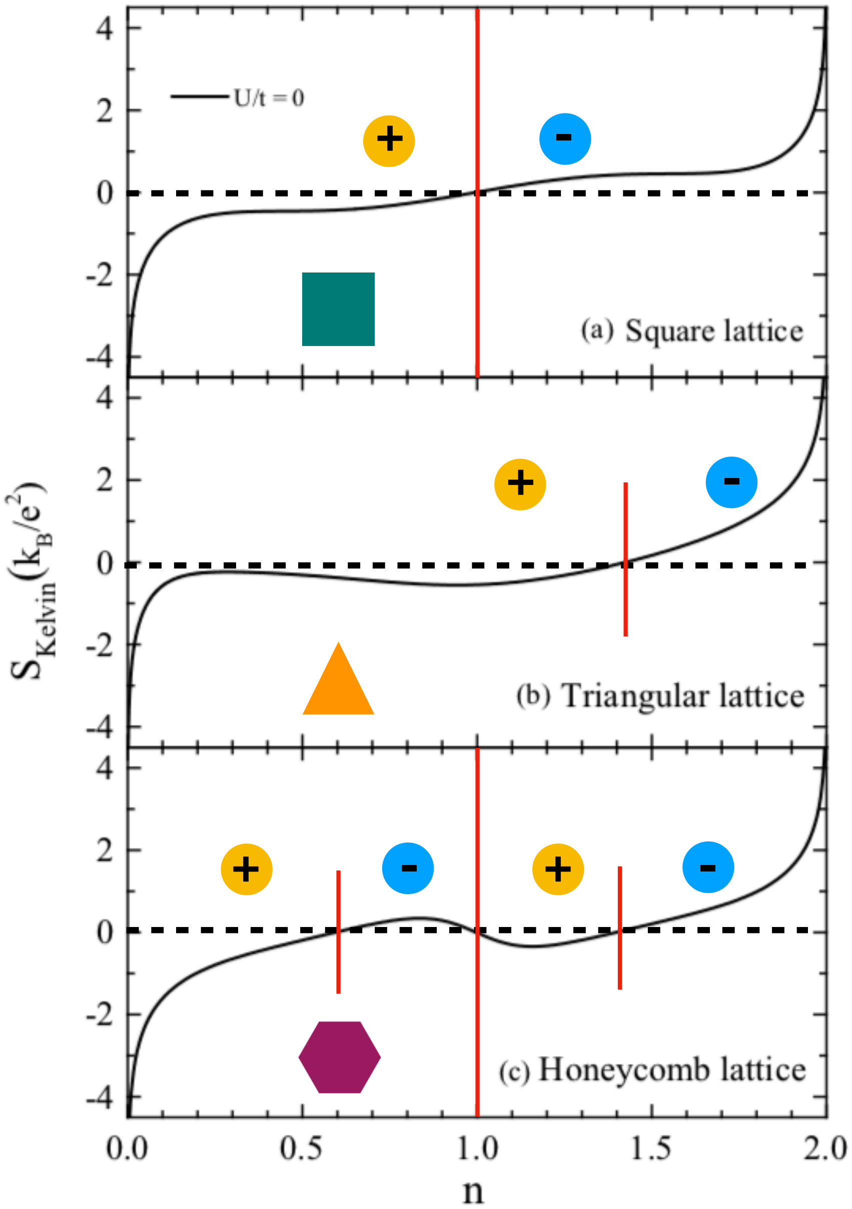
Change of sign: change of carrier

Half-filling

n=1.42

# Seebeck coefficient

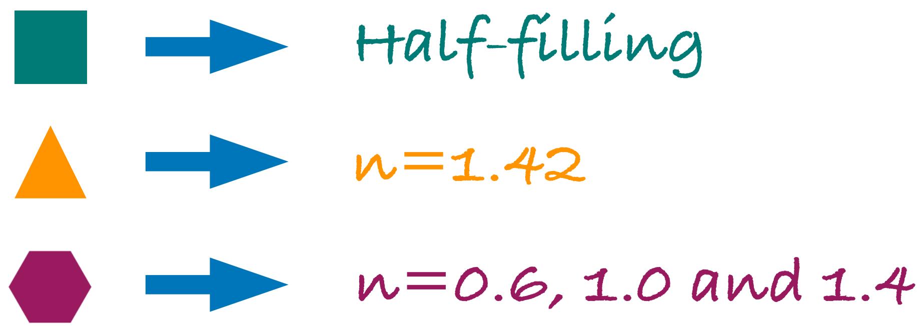
Non-interacting case



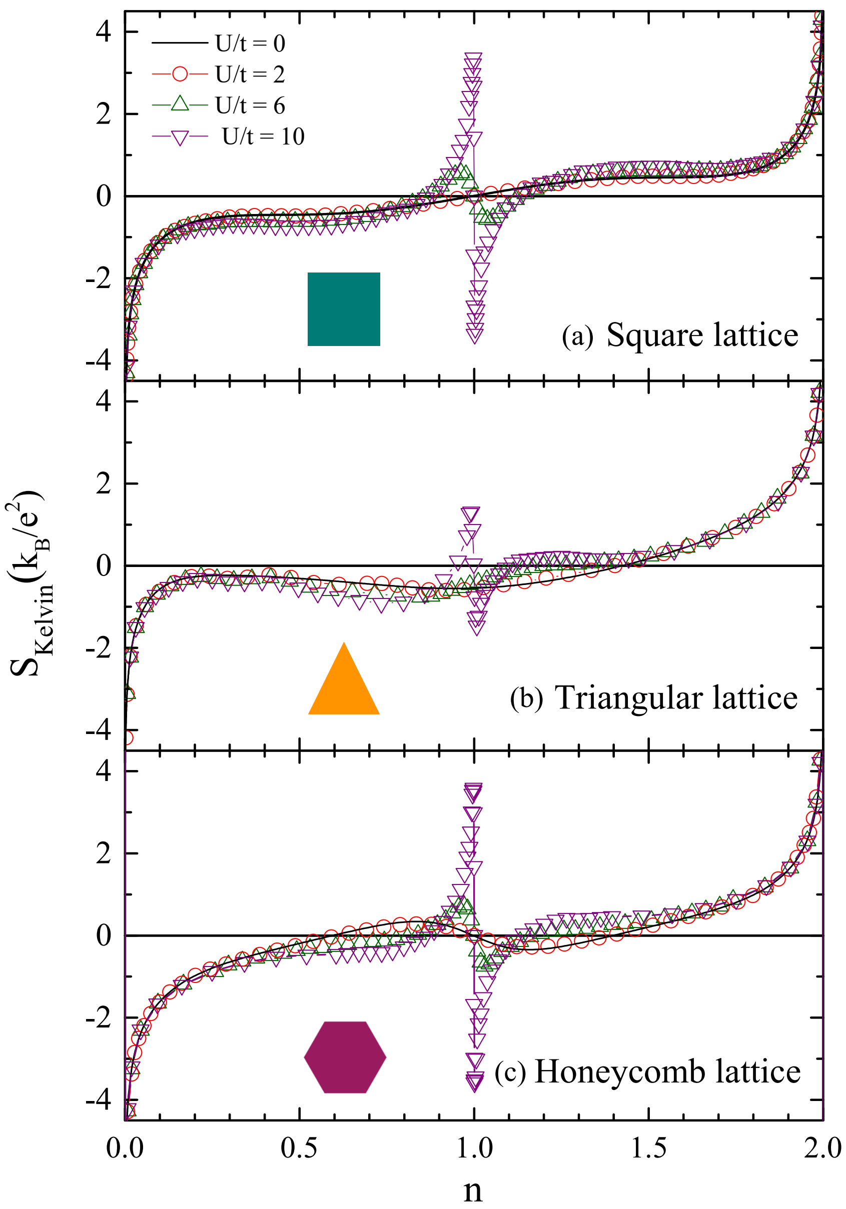

Geometry effects

T/t=0.5

Change of sign: change of carrier



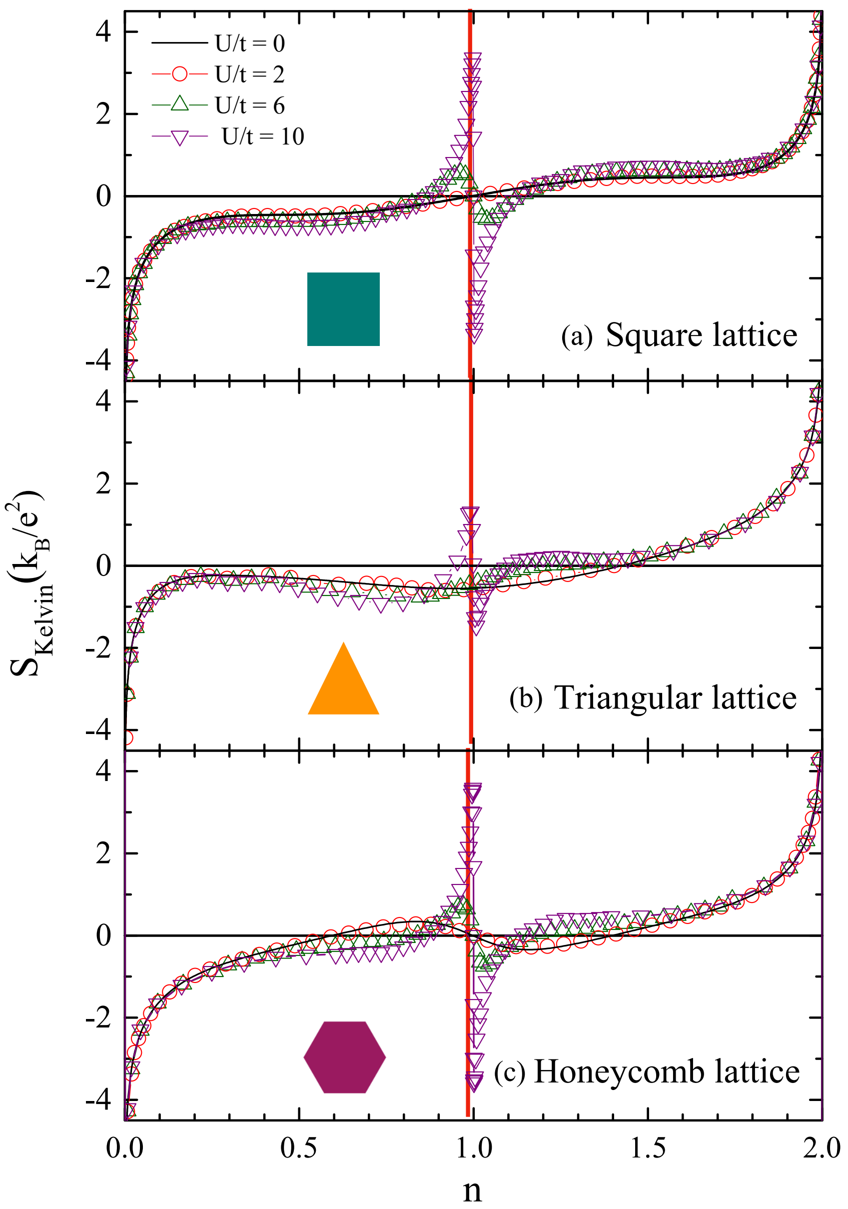
# Seebeck coefficient




**Role of Interactions**

Strong increase near half-filling

# Seebeck coefficient

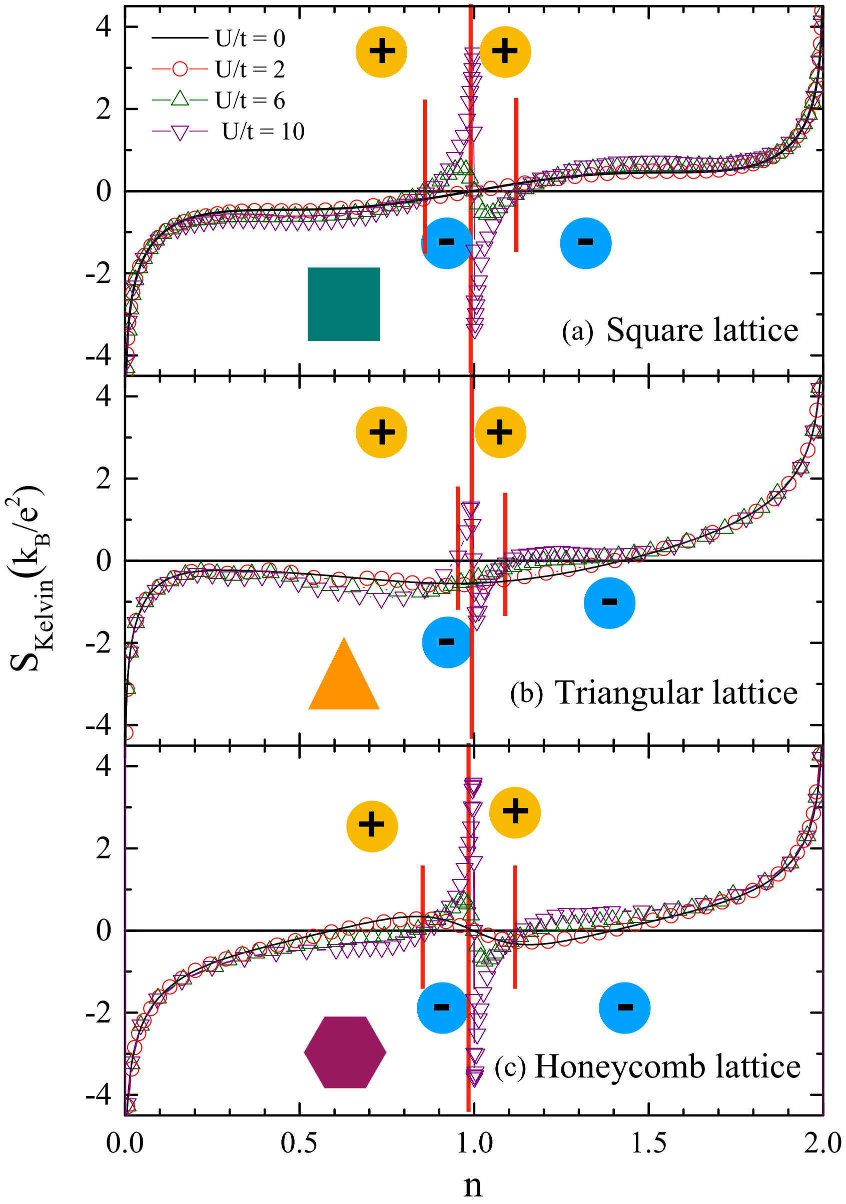



Role of Interactions

Strong increase near half-filling

Triangular lattice at half-filling: change of sign → Mott transition

# Seebeck coefficient



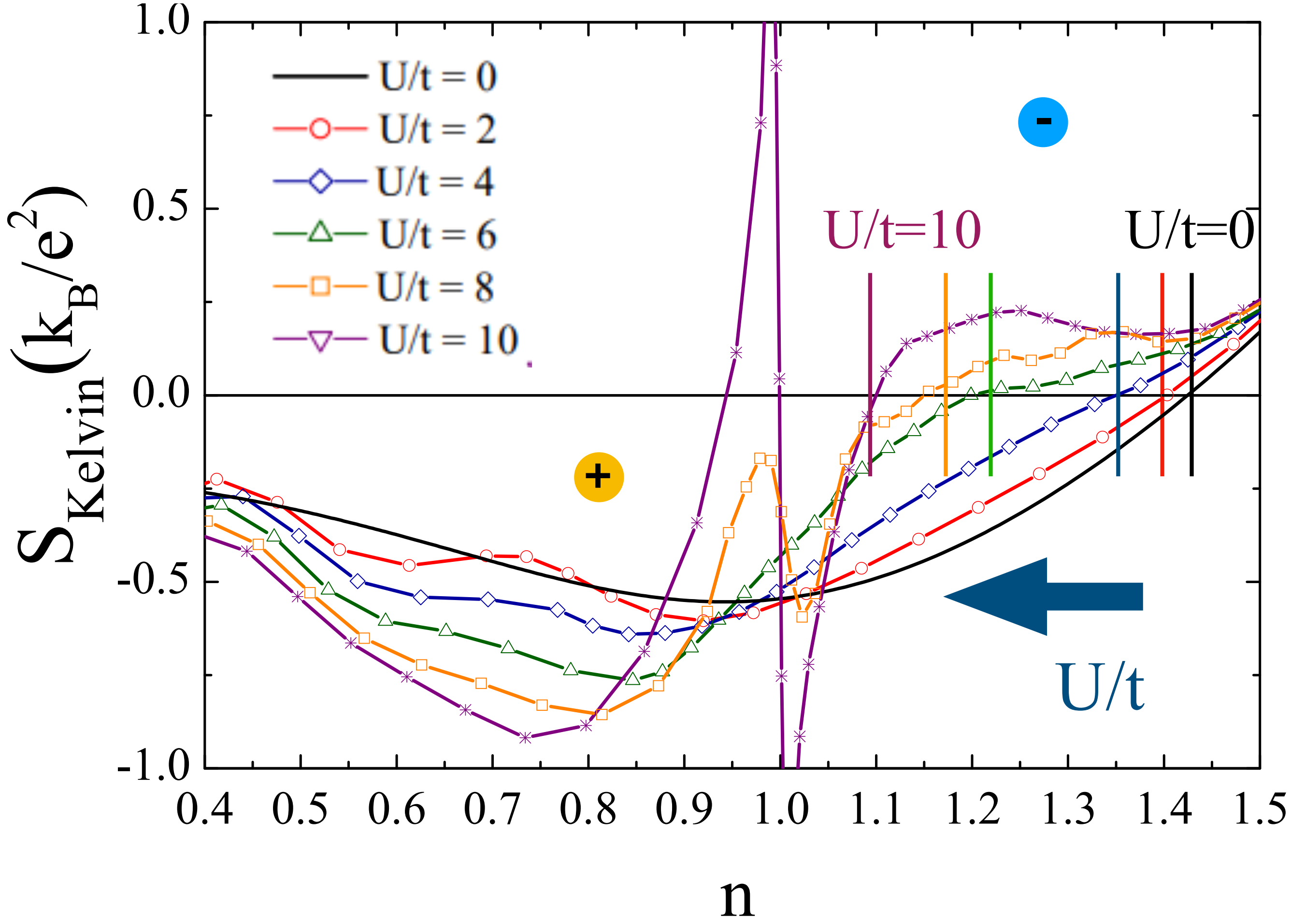

Role of Interactions

Strong increase near half-filling

Triangular lattice at half-filling: change of sign → Mott transition

Additional sign changing densities

# Seebeck coefficient for the triangular lattice

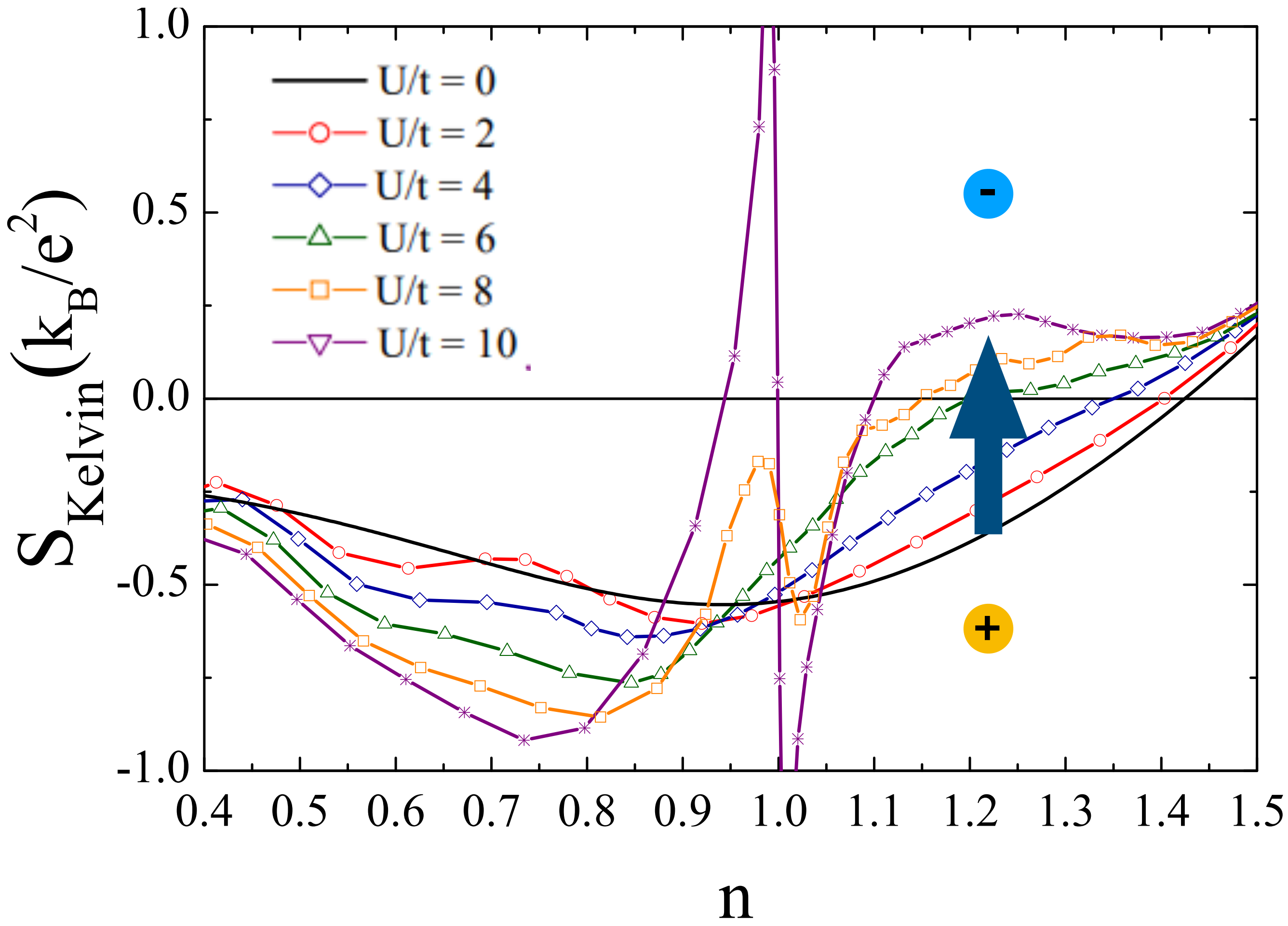



U/t dependent number of crossings

Large U/t → strong increase near half-filling

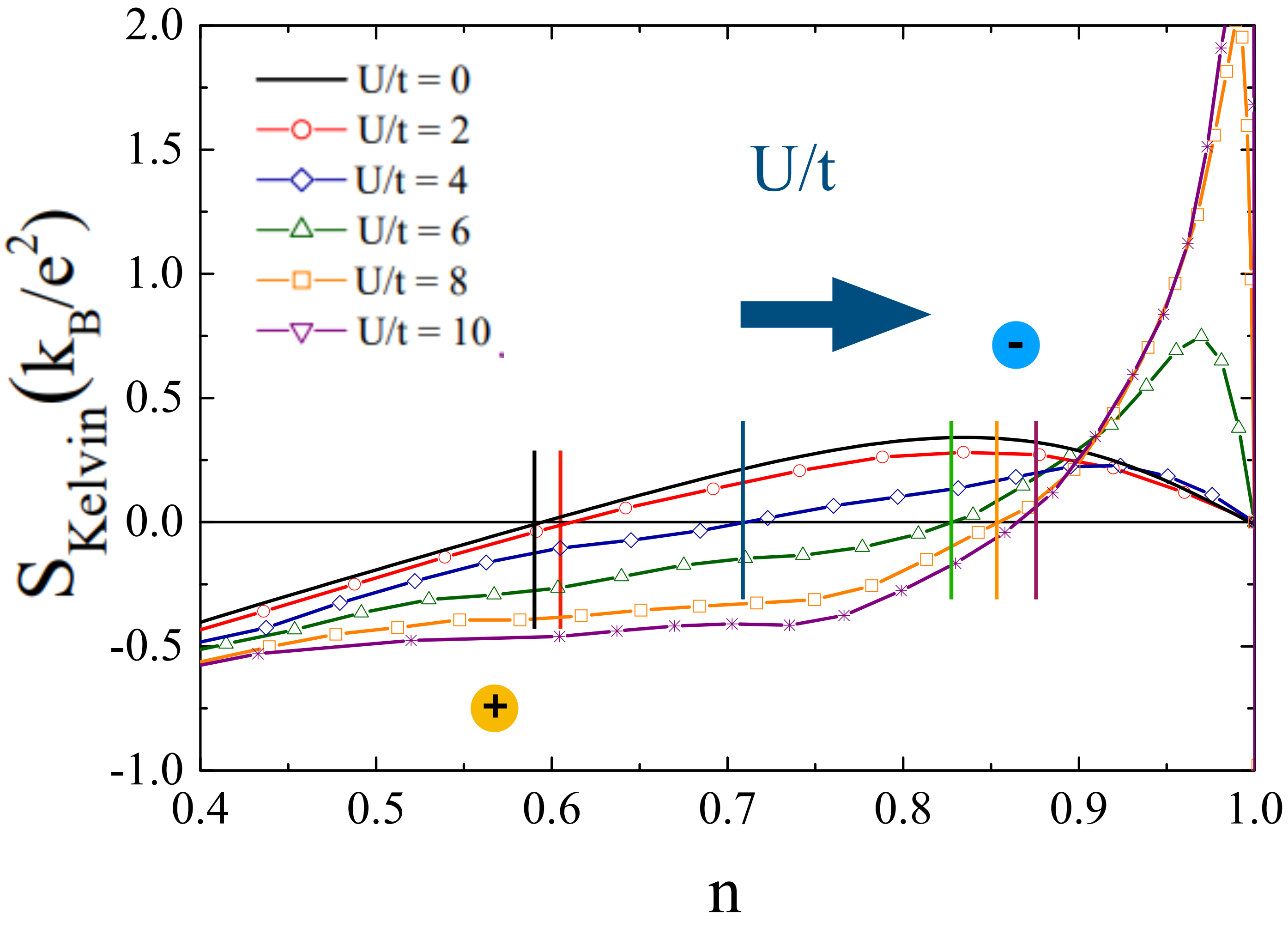
Sign changes move to lower densities as U/t increases

# Seebeck coefficient for the triangular lattice




Increasing U/t changes carriers from holes to electrons

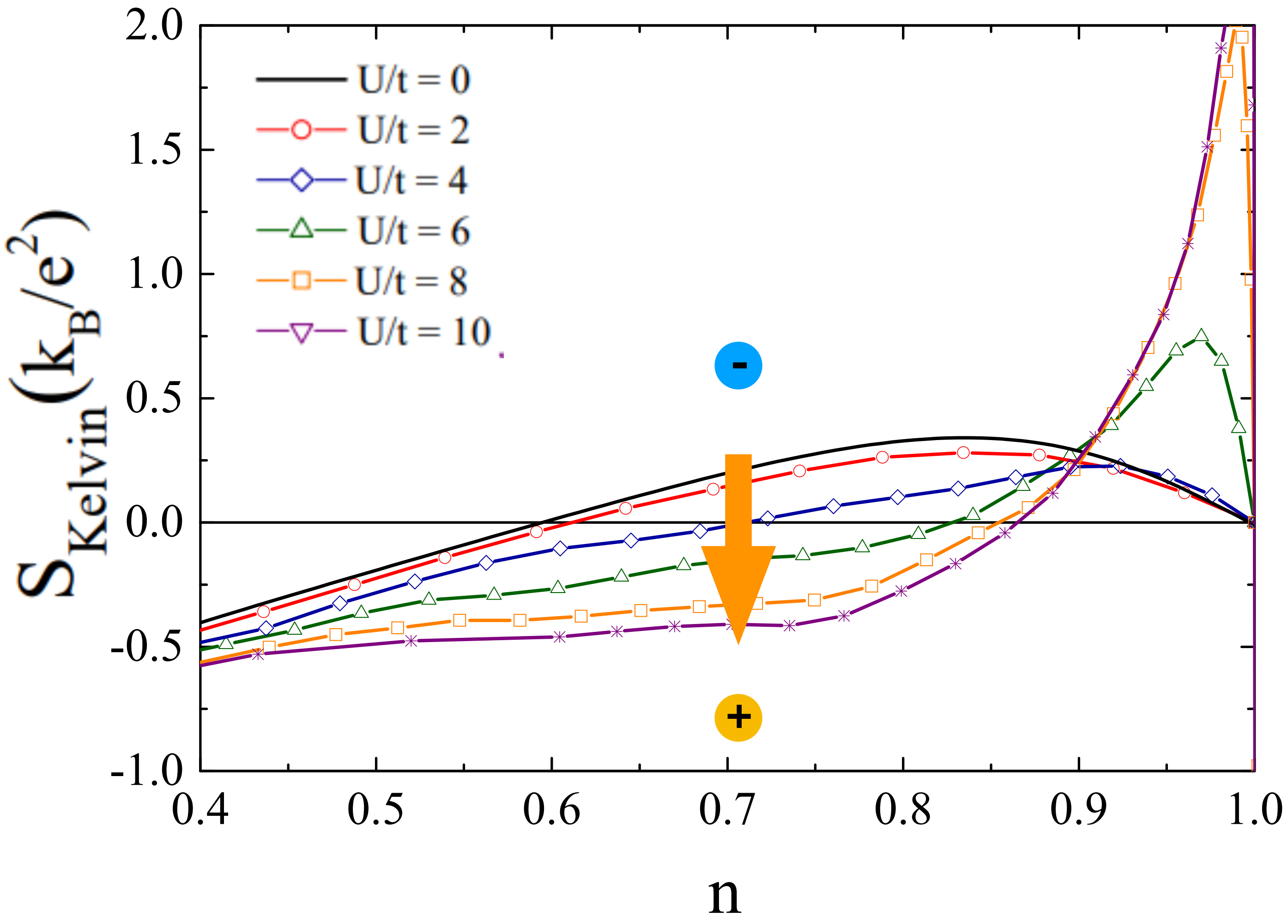
# Seebeck coefficient for the Honeycomb lattice




Large U/t → Strong increase near half-filling

Sign changes move to higher (lower) densities below (above) half-filling as U/t increases

# Seebeck coefficient for the Honeycomb lattice

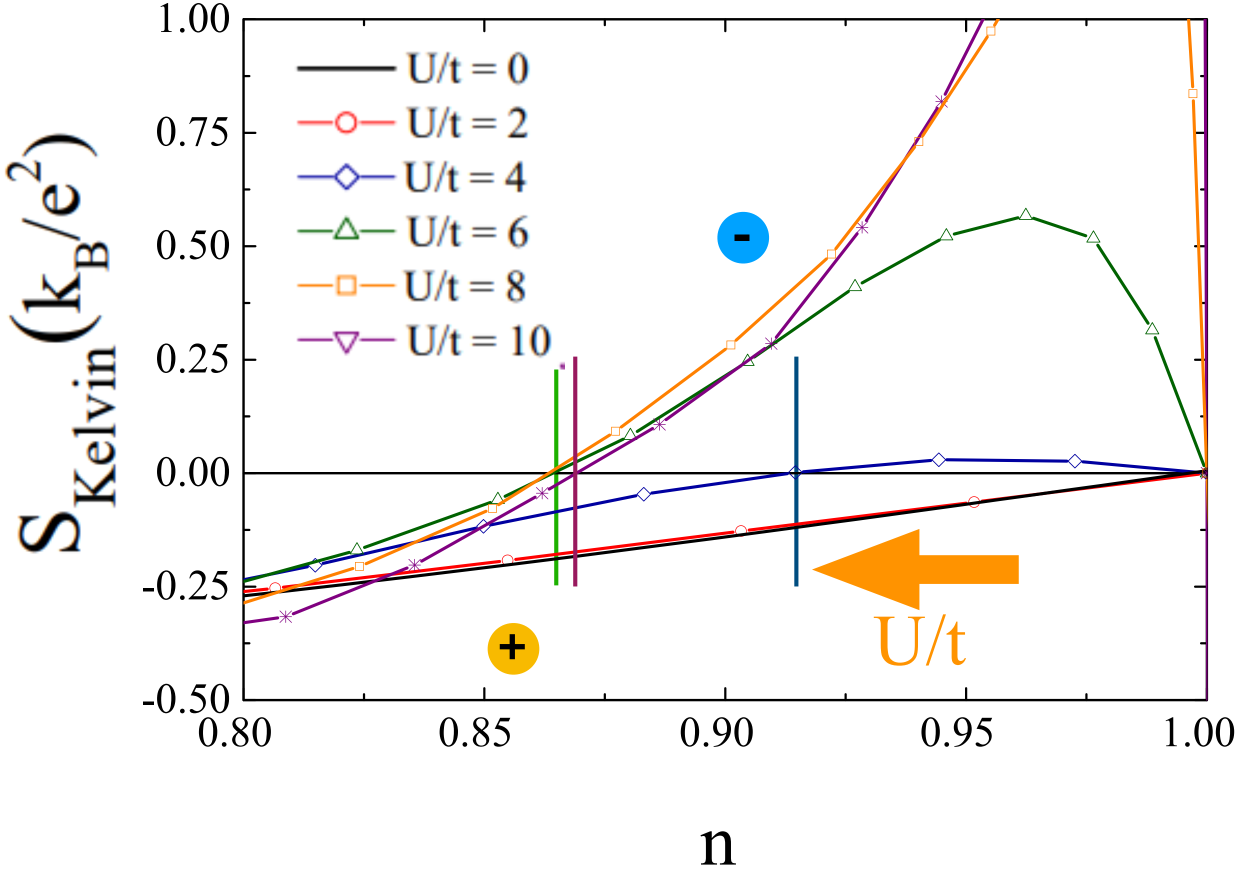



Increasing U/t changes carriers from electrons to holes for $0.6 < n < 0.85$

P-h symmetry

Increasing U/t changes carriers from holes to electrons for $1.2 < n < 1.7$.

# Seebeck coefficient for square lattice

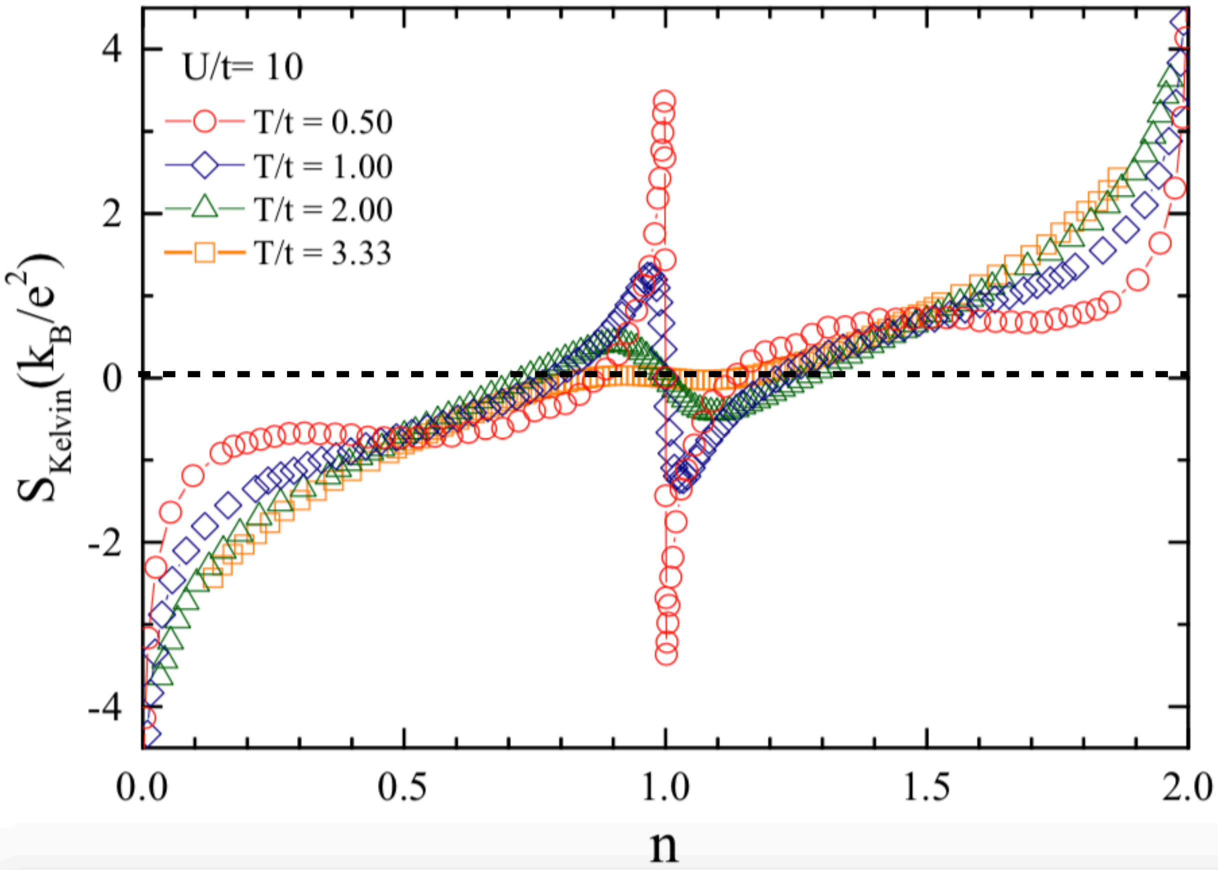



Large U/t → Strong increase near half-filling

Sign changes move to lower (higher) densities below (above) half-filling as U/t increases

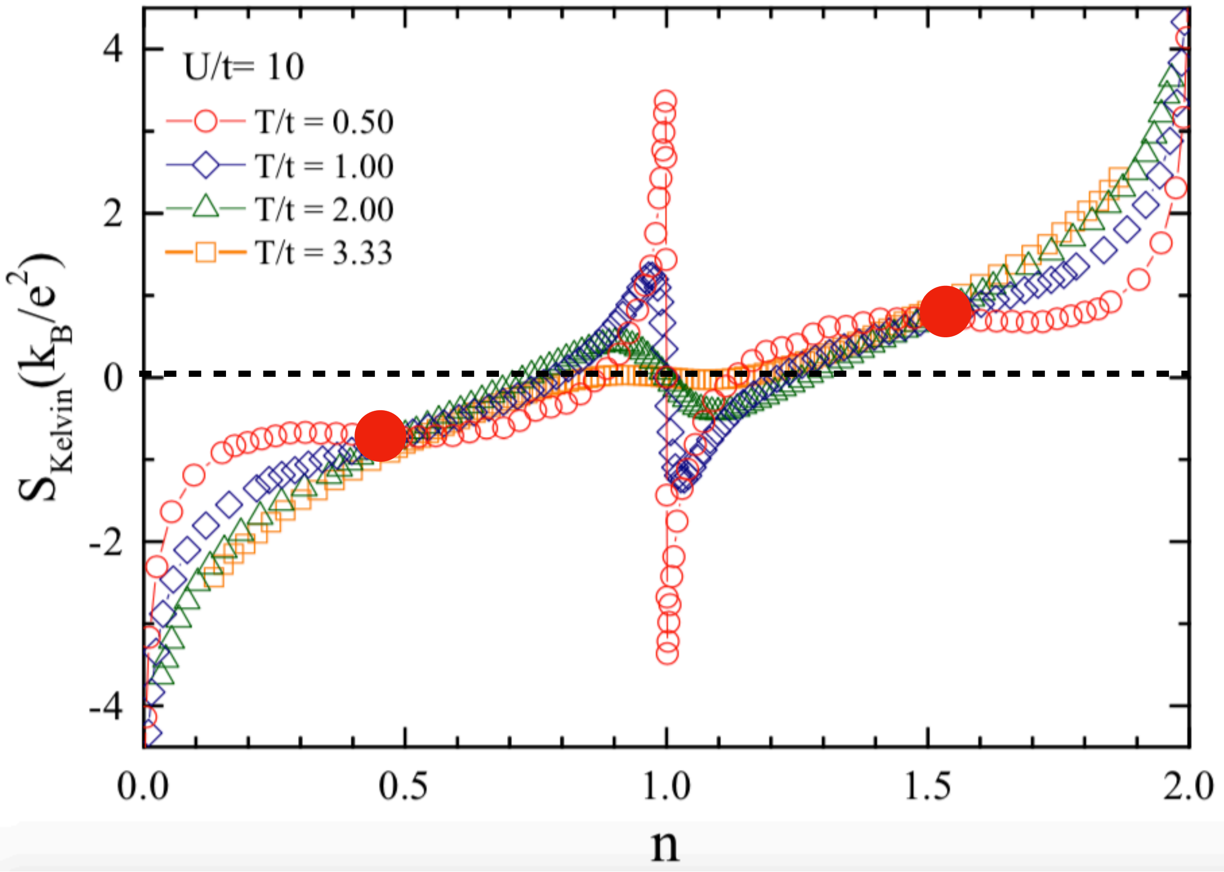
Change of sign pushed to smaller U/t

# Effect of temperature for the square lattice




Decreasing temperature increases Seebeck effect near half-filling

# Effect of temperature for the square lattice

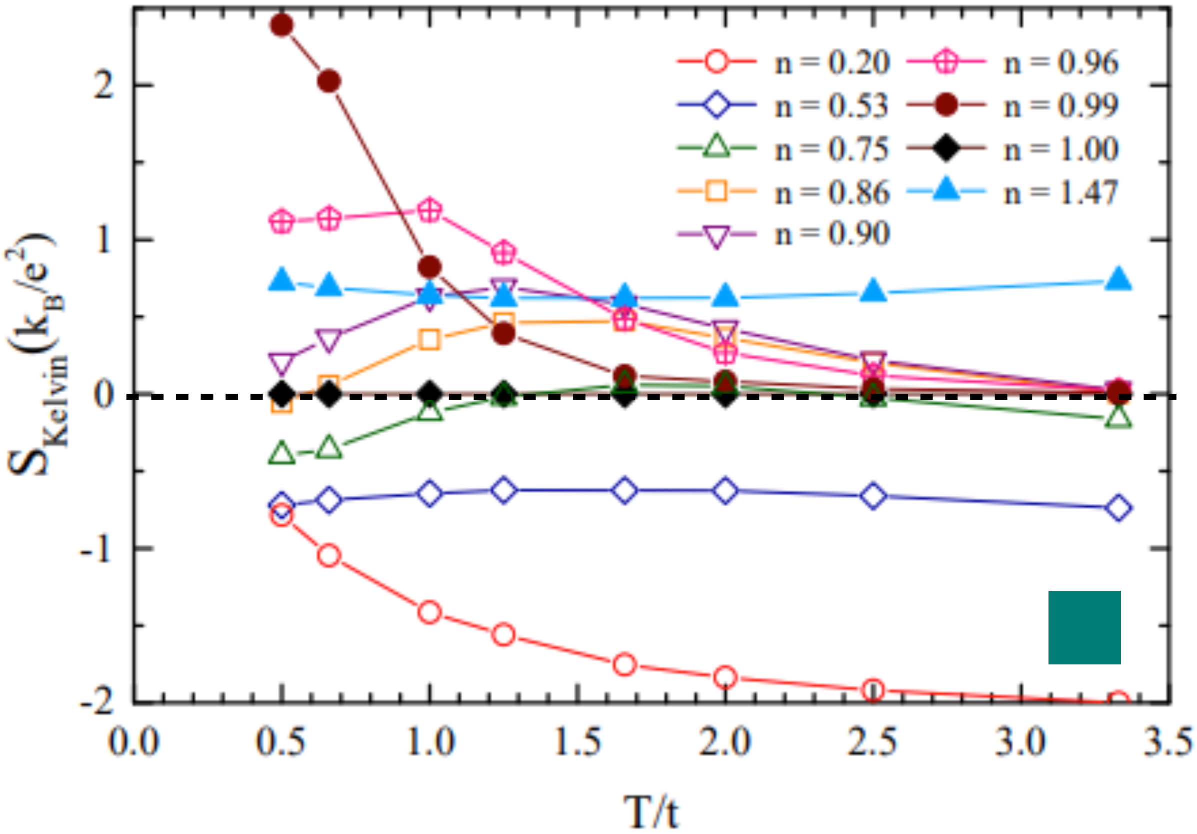



Decreasing temperature increases Seebeck effect near half-filling

Temperature independent densities n =0.5 and 1.5

Isosbestic points

# Effect of temperature for the square lattice

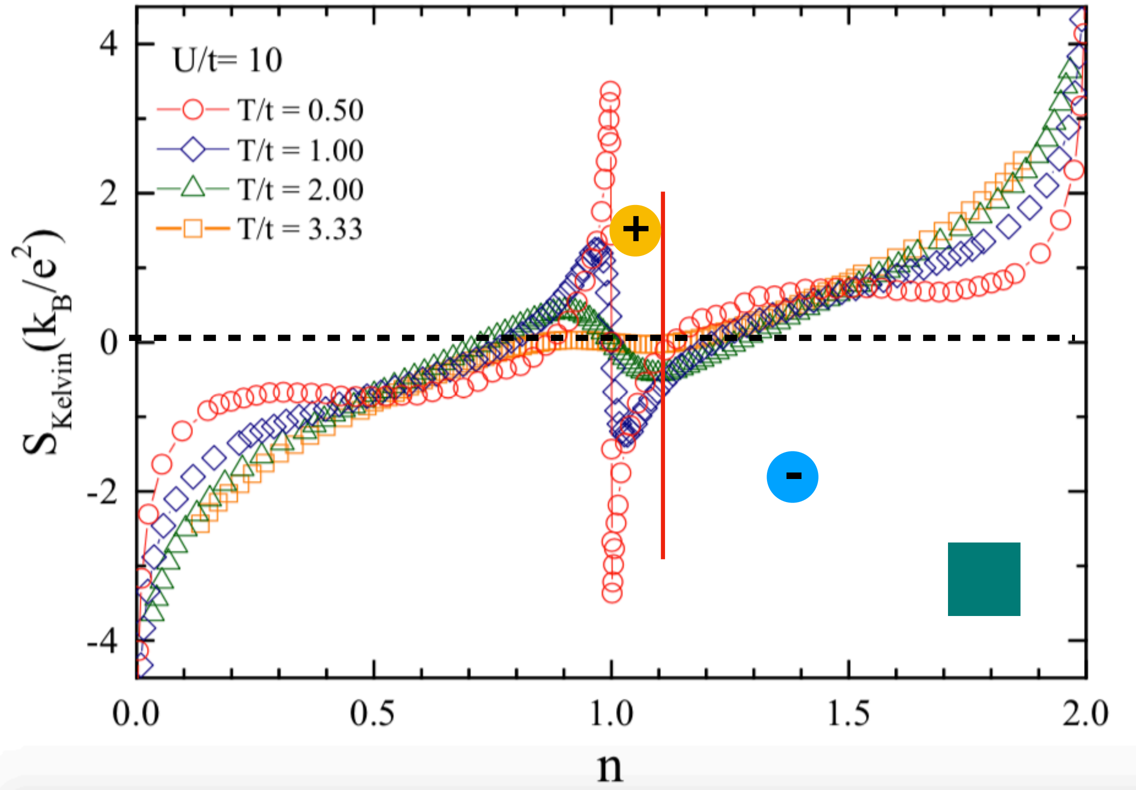


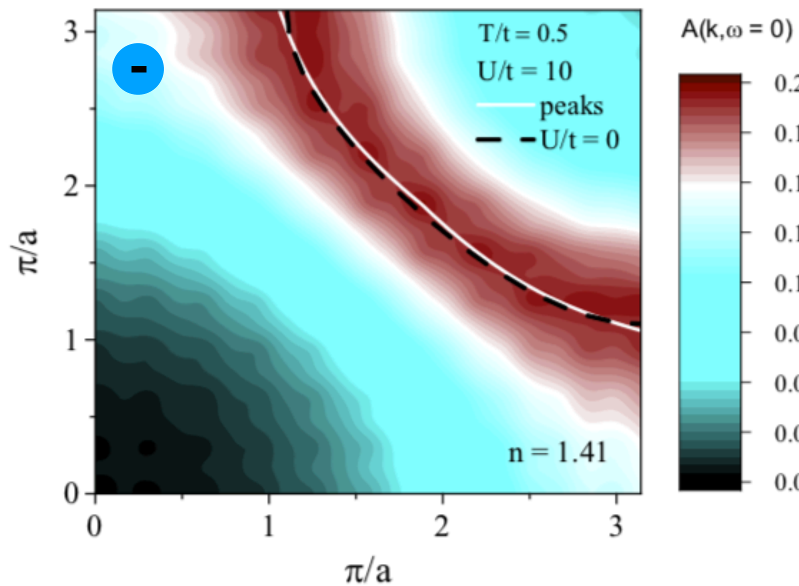

n = 1.47

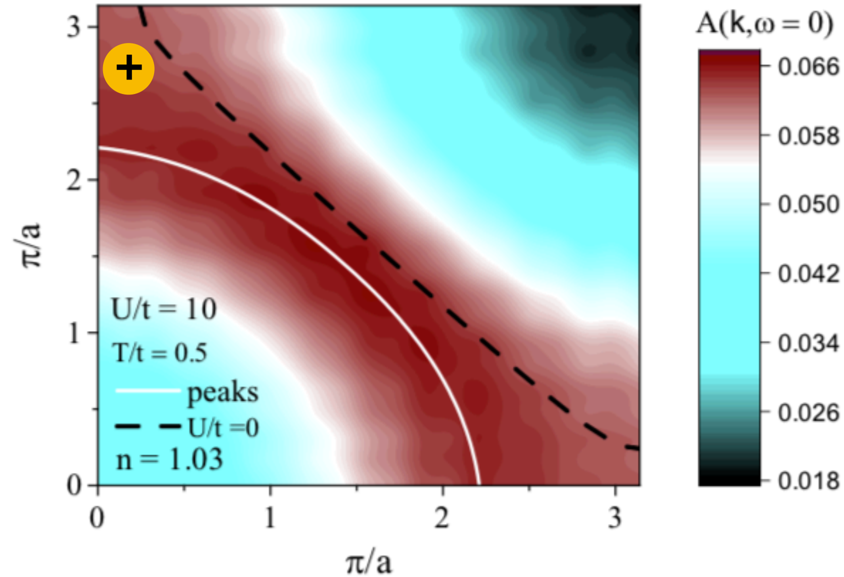
n =1.0

n =0.53

# Fermi surface reconstruction

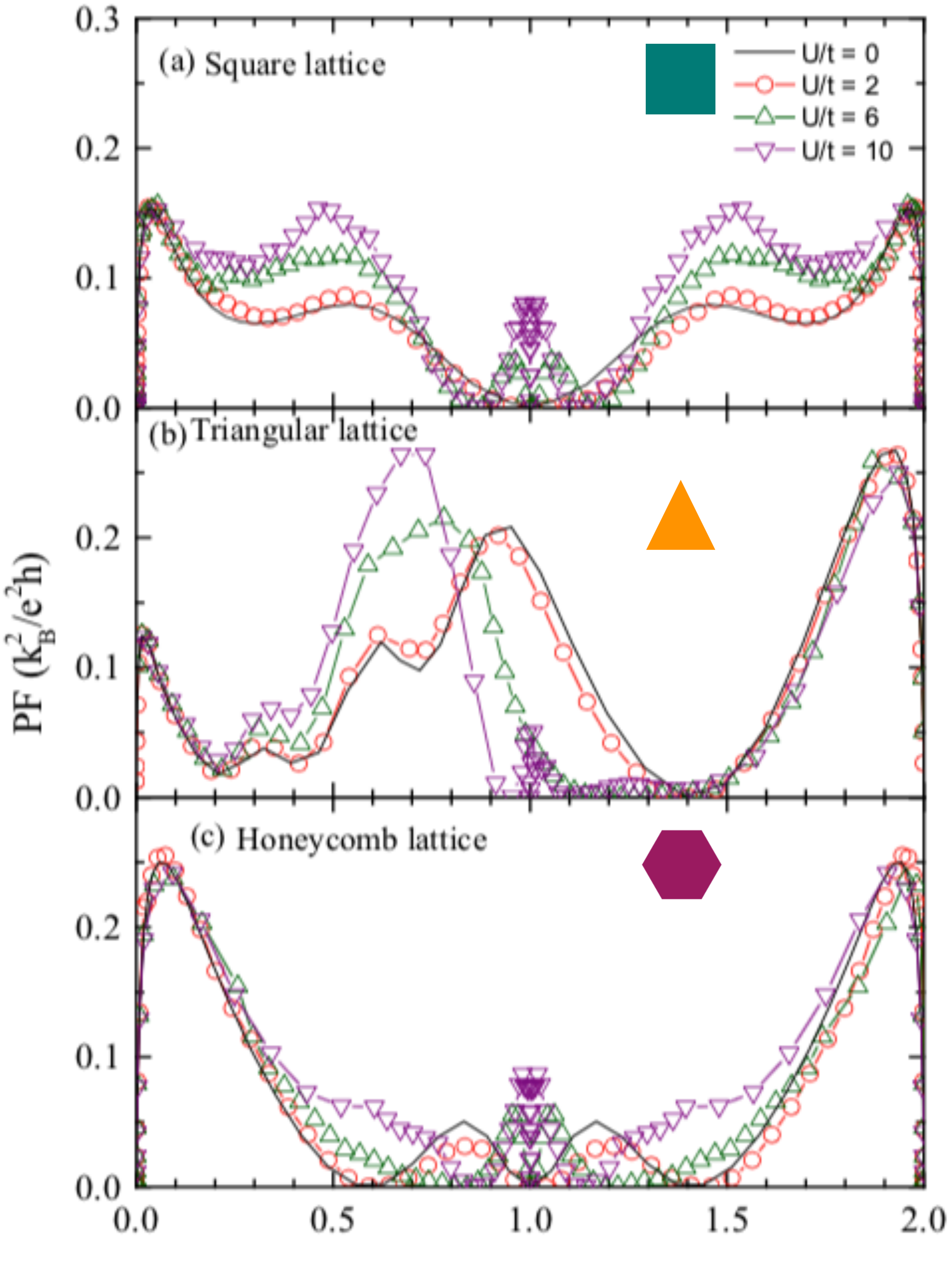


# Power factor

$$PF = S^2\sigma$$




Increased by correlations in the vicinity of half-filling

Can be tuned by interactions

At intermediate densities (around $n = 0.4 - 0.6$ and $n = 1.4 - 1.6$) the peaks in PF have a strong contribution from the conductivity with positions strongly dependent on geometry.

# Conclusions

Anomalous Seebeck effect near half-filling: change in sign signals the Fermi surface reconstruction

Anomaly intensified by temperature reduction and increased correlations

Away from half-filling, at intermediate densities (around $n = 0.4 - 0.6$ and $n = 1.4 - 1.6$) the peaks in PF have a strong contribution from the conductivity with positions strongly dependent on geometry.

The thermoelectric Power Factor displays a competition between the Seebeck coefficient and the conductivity

# Thank you!


CNPq

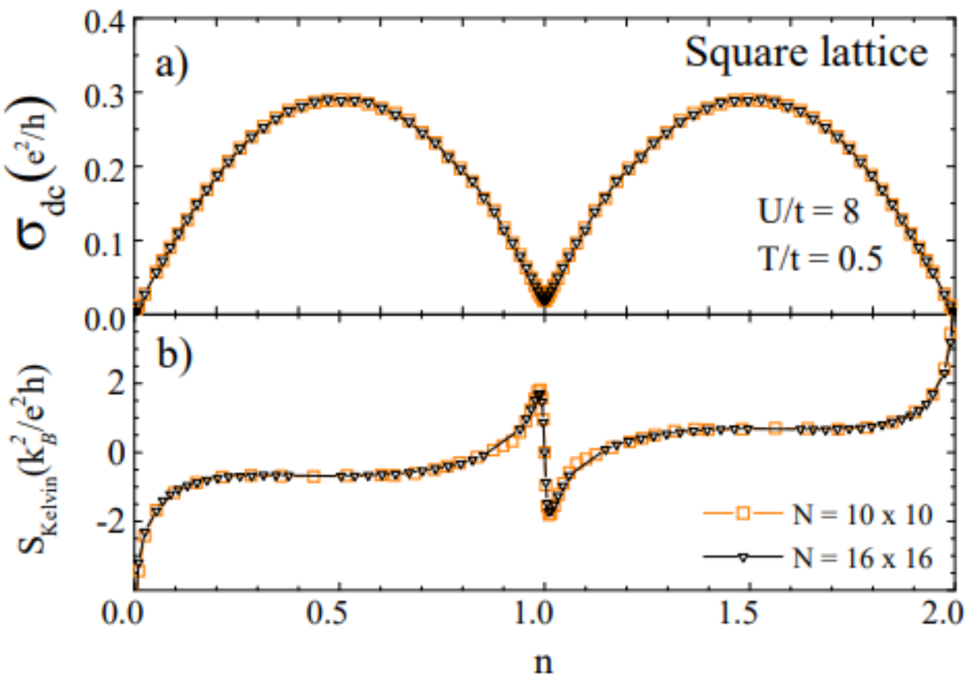
CAPES

FAPERJ
Fundação Carlos Chagas Filho de Amparo à Pesquisa do Estado do Rio de Janeiro

INCT-IQ

# Feliz aniversário, Eduardo!

# Size effects




T/t=0.5

Irrelevant at this temperature

## Density of States

$$N(\omega = 0) \approx \frac{\beta}{\pi} G(|\mathbf{i} - \mathbf{j}| = \mathbf{0}, \tau = \beta/\mathbf{2})$$

$$\tau = \beta/2$$

$$\beta = 1/k_B T$$

## Conductivity

$$\sigma_{dc} \approx \frac{\beta^2}{\pi} \Lambda_{xx}(\mathbf{q} = \mathbf{0}, \tau = \beta/\mathbf{2})$$

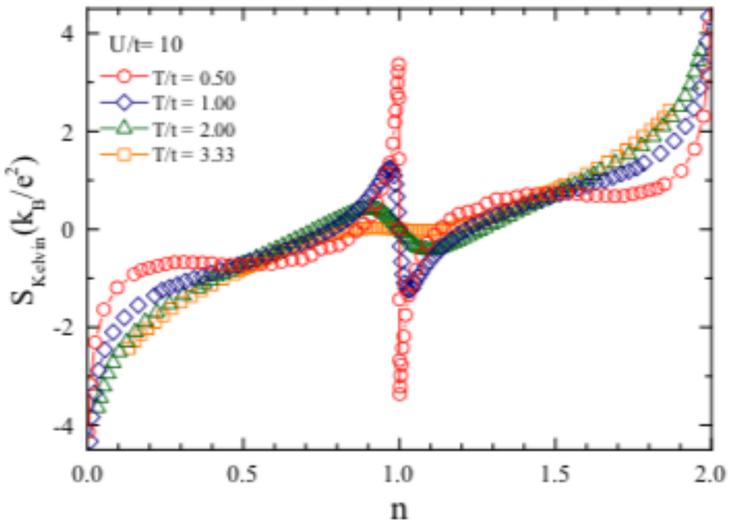
$$\Lambda_{xx}(\mathbf{q}, \tau) = \langle j_x(\mathbf{q}, \tau) j_x(-\mathbf{q}, 0) \rangle$$

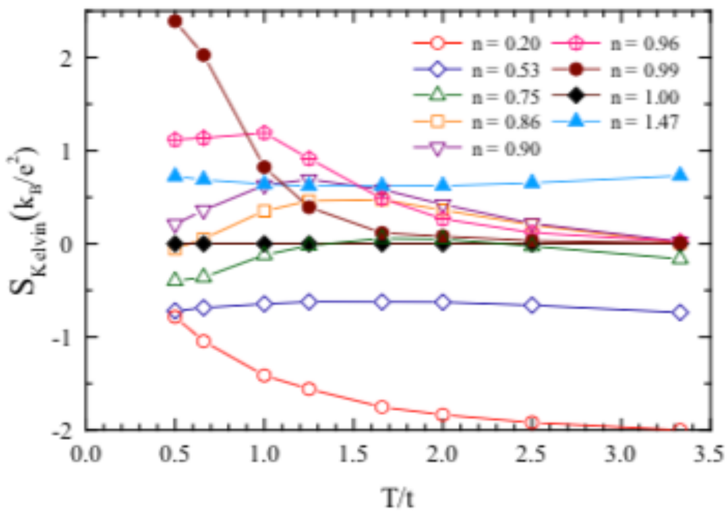
$$j_x(\mathbf{i}, \tau) = \mathrm{e}^{\tau \mathcal{H}} \left[ it \sum_\sigma \left( c^\dagger_{\mathbf{i}+\mathbf{x},\sigma} c_{\mathbf{i},\sigma} - c^\dagger_{\mathbf{i},\sigma} c_{\mathbf{i}+\mathbf{x},\sigma} \right) \right] \mathrm{e}^{-\tau \mathcal{H}}$$

# Seebeck coefficient

Temperature effects

Square lattice




Decreasing temperature increases Seebeck effect near half-filling

Temperature independent densities n =0.5 and 1.5

# Power factor

Figure of merit

$$ZT = \frac{S^2 \sigma T}{\kappa}$$

$S$ → Thermopower or Seebeck coefficient

$\sigma$ → Conductivity

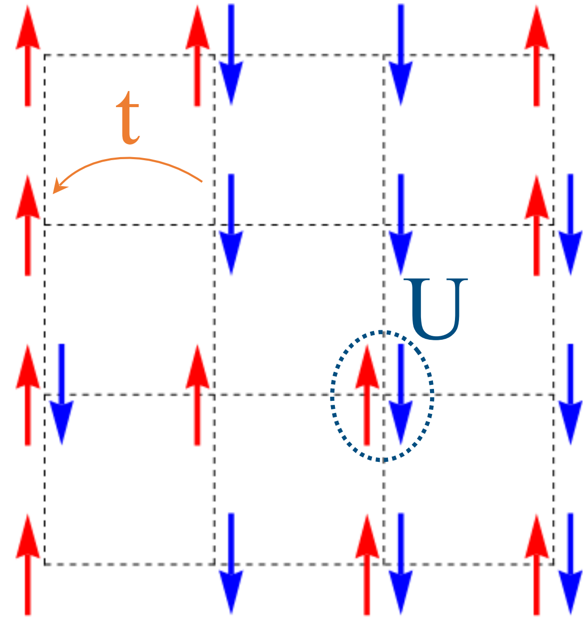
$\kappa$ → Thermal conductivity

$$PF = S^2 \sigma$$

Competition between Seebeck coefficient and conductivity

# Hubbard Model

$$\mathcal{H} = -t \sum_{\langle i,j \rangle,\sigma} \left( \hat{c}^{\dagger}_{i,\sigma} \hat{c}_{j,\sigma} + h.c. \right) + U \sum_{i} \left( \hat{n}_{i\uparrow} - \frac{1}{2} \right) \left( \hat{n}_{i\downarrow} - \frac{1}{2} \right) - \mu \hat{N}$$




Coulomb repulsion (U>0)

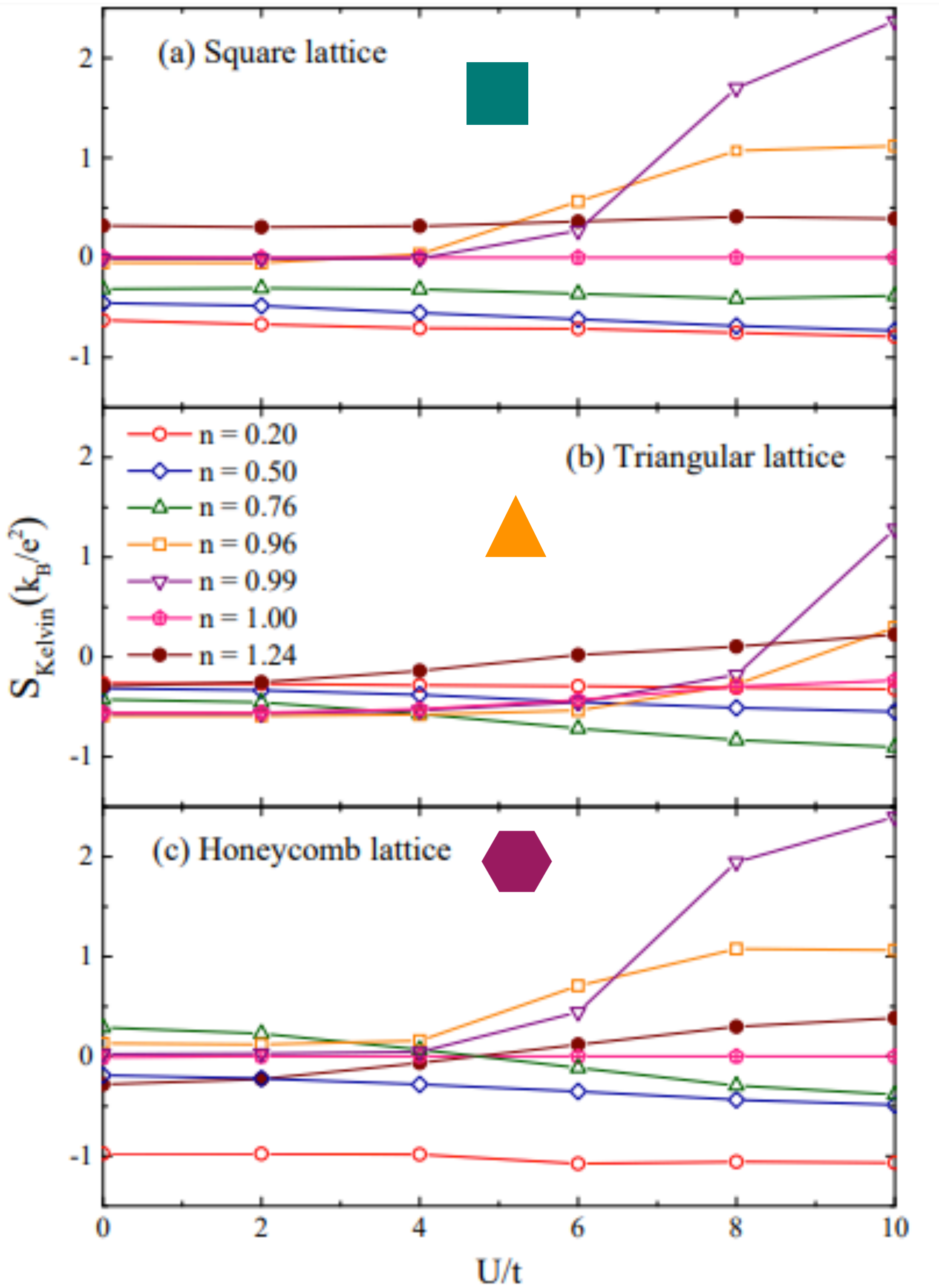
Hopping (t)

chemical potential ($\mu$)

No known analytic solution in 2D

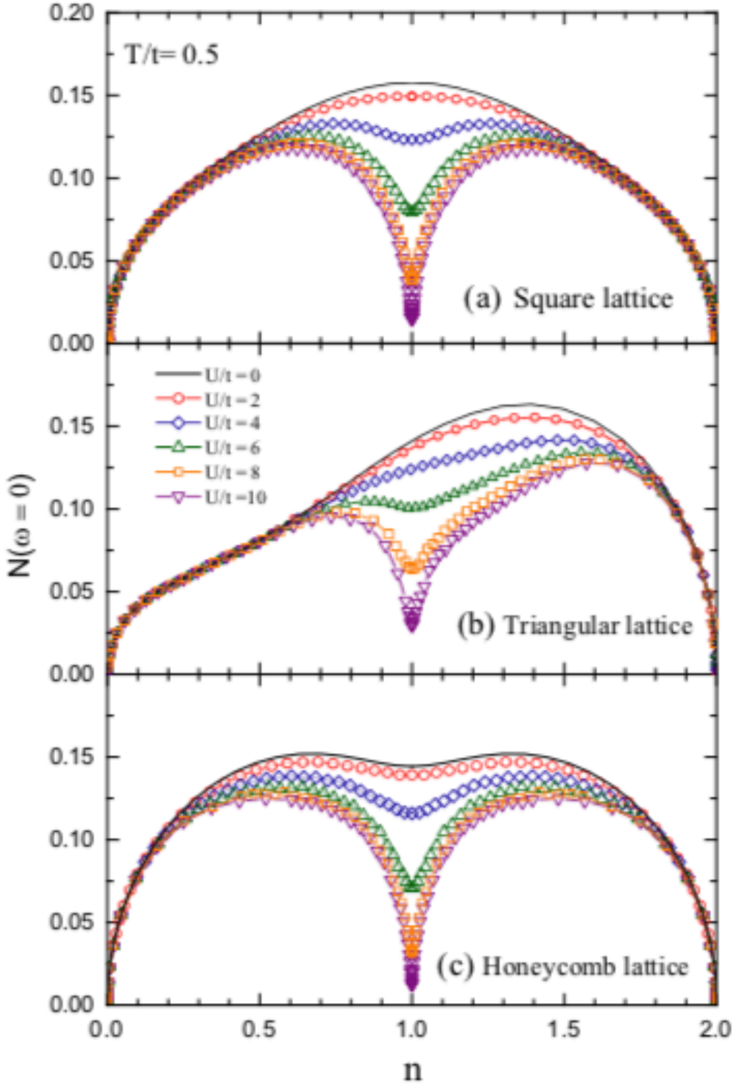
# Effect of correlations

T/t=0.5



# Density of States

$$N(\omega = 0, T) = \frac{dn}{d\mu} = n^2 \kappa(T)$$




$$N(\omega = 0) \approx \frac{\beta}{\pi} G(|\mathbf{i} - \mathbf{j}| = \mathbf{0}, \tau = \beta/\mathbf{2})$$