The Overhauser Instability

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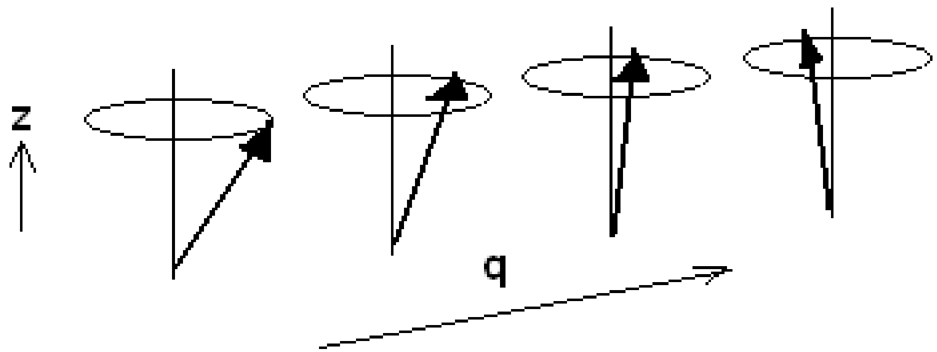
Introduction

- Motivation: Noncollinear spins and Spiral Spin Density Waves
- Problem: The Overhauser Instability in the Homogeneous Electron Gas
- Method: Generalized Hartree-Fock theory
- Results
- Interpretation

Noncollinear spins

- *Collinear*: definite spin (up or down) with respect to global quantization axis.
- Particles are distinguishable, simplifies calculation.
- *Noncollinear*: spin directions that are not parallel to the global quantization axis and the spin direction can vary with position.
- Wavefunction depends fully on position and spin coordinates.
- *Ab Initio* methods: some DFT codes. CASINO does VMC for specific problems.

- *Spin density* or *Magnetization density*: net magnetic moment due to spin. Vector function of position. Noncollinear example:



Spiral Spin Density Wave
(magnetization wavevector $\mathbf{q}$)

Neutron scattering experiments: SSDW is ground state of several systems

- Motivation: Need trial wavefunction for QMC study of such system.

Overhauser Instability

- What is ground state of Homogeneous Electron Gas in Hartree-Fock theory?

- High-density (low r_s) limit: paramagnet. Low-density (high r_s): ferromagnet. Collinear states.

- Overhauser: paramagnet is never ground state, but instead a Spiral Spin Density Wave is.

- Proof and analytical solution for 1D with repulsive δ-function interaction. (A. W. Overhauser, *Giant spin density waves*, Phys. Rev. Lett. **4**, p.462 (1960))

- Proof of existence for 3D with Coulomb interaction. (A. W. Overhauser, *Spin density waves in an electron gas*, Phys. Rev. **128**, p.1437 (1962))

Generalized Hartree-Fock theory

- HF theory of noncollinear spins. Orbitals have full spin dependence.

- Can write orbitals as function of space-spin coordinates $\psi(\mathbf{x})$. Equivalently, can write orbitals as two-component spinors: $\underline{\psi}(\mathbf{r}) = \begin{pmatrix} \psi_1(\mathbf{r}) \\ \psi_2(\mathbf{r}) \end{pmatrix}$.

- Wavefunction is determinant of spinors: $\Psi = \frac{1}{\sqrt{N!}} \det |\underline{\psi}_i(\mathbf{r}_j)|$.

- Hamiltonian: $\hat{H} = \sum_i -\frac{1}{2}\nabla_i^2 + \sum_i U(\mathbf{r}_i) + \frac{1}{2}\sum_{i,j\neq i} V(\mathbf{r}_i, \mathbf{r}_j) + \lambda$

  V could be Coulomb or Ewald interaction.

- Total energy is $E = \langle \Psi | \hat{H} | \Psi \rangle$. Evaluate variation with respect to orbitals, subject to orthonormality constraint.

- Gives single-particle HF equation (depending on exact form of $\hat{H}$) such as:

$$(\hat{\mathbf{K}} + \hat{\mathbf{U}} + \hat{\mathbf{V}} - \hat{\mathbf{J}})\underline{\psi}_k(\mathbf{r}) = \epsilon_k \underline{\psi}_k(\mathbf{r}) \tag{1}$$

- This is a 2x2 matrix equation. $\hat{\mathbf{K}}$ (kinetic energy), $\hat{\mathbf{U}}$ (external potential), $\hat{\mathbf{V}}$ (direct term) and $\hat{\mathbf{J}}$ (exchange term) are 2x2 matrices of spatial operators.

- For spin independent $\hat{H}$, $\hat{\mathbf{J}}$ *can still have off-diagonal components*, giving rise to noncollinearity.

- Collinear case is special case with $\underline{\psi}(\mathbf{r}) = \phi(\mathbf{r})\underline{\chi}$, where $\underline{\chi}$ is a spin eigenstate. Reduces problem to Unrestricted HF theory.

- HF equation needs to be solved self-consistently.

Hartree-Fock theory of Homogeneous Electron Gas

- Hamiltonian: $\hat{H} = \sum_i -\frac{1}{2}\nabla_i^2 + \frac{1}{2}\sum_{i,j\neq i} V(\mathbf{r}_i, \mathbf{r}_j)$

- Single-particle HF equation is self-consistently solved by SSDW orbitals:

$$\underline{\psi}_{\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} \begin{pmatrix} \cos(\frac{1}{2}\theta_{\mathbf{k}})\, e^{-i\frac{1}{2}\mathbf{q}\cdot\mathbf{r}} \\ \sin(\frac{1}{2}\theta_{\mathbf{k}})\, e^{+i\frac{1}{2}\mathbf{q}\cdot\mathbf{r}} \end{pmatrix}$$

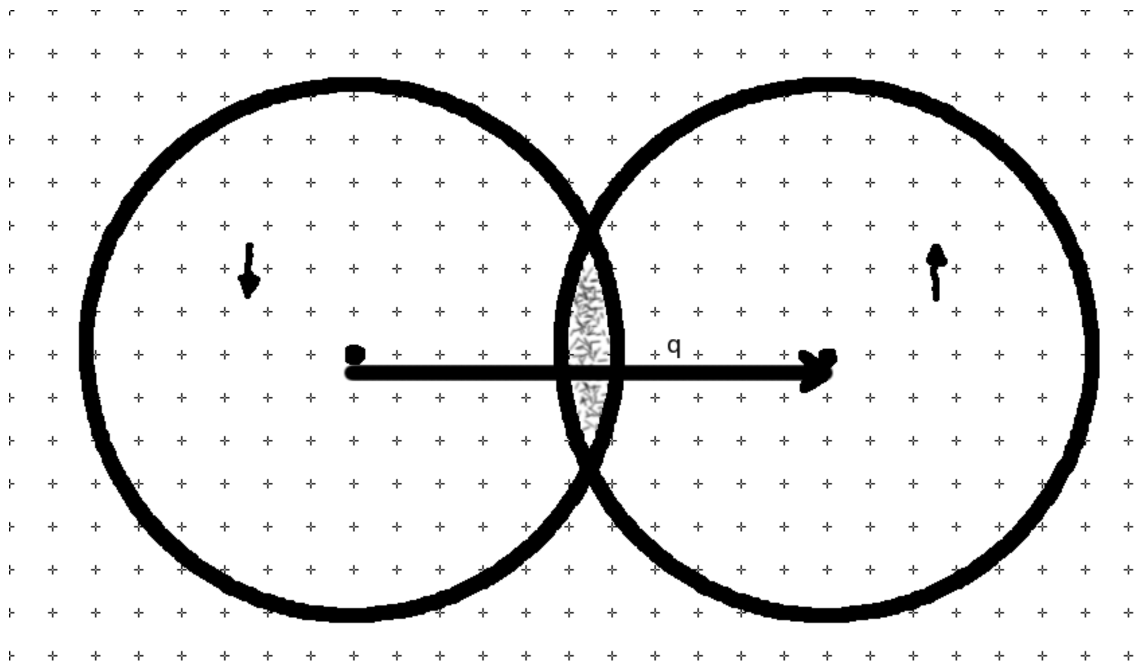
- $\mathbf{k}$ is plane-wave vector, $\mathbf{q}$ is magnetization wave vector (constant), and the orbital has its spin pointing in $(\theta, \mathbf{q}\cdot\mathbf{r})$ direction (spiral in space).

- Two orthogonal orbitals are possible at each $\mathbf{k}$-point: $\theta \longrightarrow \theta + \pi$.

- *Mixing* up spin at $\mathbf{k} - \frac{1}{2}\mathbf{q}$ with down spin at $\mathbf{k} + \frac{1}{2}\mathbf{q}$.

- Paramagnet and ferromagnet are special cases. They correspond to particular choices of occupation and $\theta_{\mathbf{k}}$.

- Example: paramagnet is two overlapping spheres in $\mathbf{k}$-space:



- Overhauser instability: driven by exchange. Arises as noncollinearity near $\mathbf{k} = 0$ and accompanying reoccupation of k-space.

Analytical part

- Total energy:

$$E = \sum_{\mathbf{k}} \frac{1}{2} \left\{ \mathbf{k}^2 + \frac{1}{4}\mathbf{q}^2 - \mathbf{k}\cdot\mathbf{q}\cos\theta_{\mathbf{k}} \right\} - \frac{1}{2}\frac{1}{\Omega} \sum_{\mathbf{k},\mathbf{k}'\neq\mathbf{k}} \frac{4\pi}{|\mathbf{k}-\mathbf{k}'|^2} \cos^2\frac{1}{2}(\theta_{\mathbf{k}} - \theta_{\mathbf{k}'}) + \frac{1}{2}N\xi \tag{2}$$

(ξ is Ewald self-image term)

- Single-particle HF equation:

$$\left[\begin{pmatrix} K_{\mathbf{k}1} & 0 \\ 0 & K_{\mathbf{k}2} \end{pmatrix} - \begin{pmatrix} J_{\mathbf{k}1} & J_{\mathbf{k}o} \\ J_{\mathbf{k}o} & J_{\mathbf{k}2} \end{pmatrix} \right] \begin{pmatrix} \cos(\frac{1}{2}\theta_{\mathbf{k}}) \\ \sin(\frac{1}{2}\theta_{\mathbf{k}}) \end{pmatrix} = \epsilon_{\mathbf{k}} \begin{pmatrix} \cos(\frac{1}{2}\theta_{\mathbf{k}}) \\ \sin(\frac{1}{2}\theta_{\mathbf{k}}) \end{pmatrix} \tag{3}$$

Numerical part

- **Aim** Find occupation of $\mathbf{k}$-space and form of $\theta_{\mathbf{k}}$ self-consistently. Find value of q that gives lowest energy.

- Parameters: r_s and q.

- Start by choosing initial occupation of orbitals and $\theta_{\mathbf{k}}$.

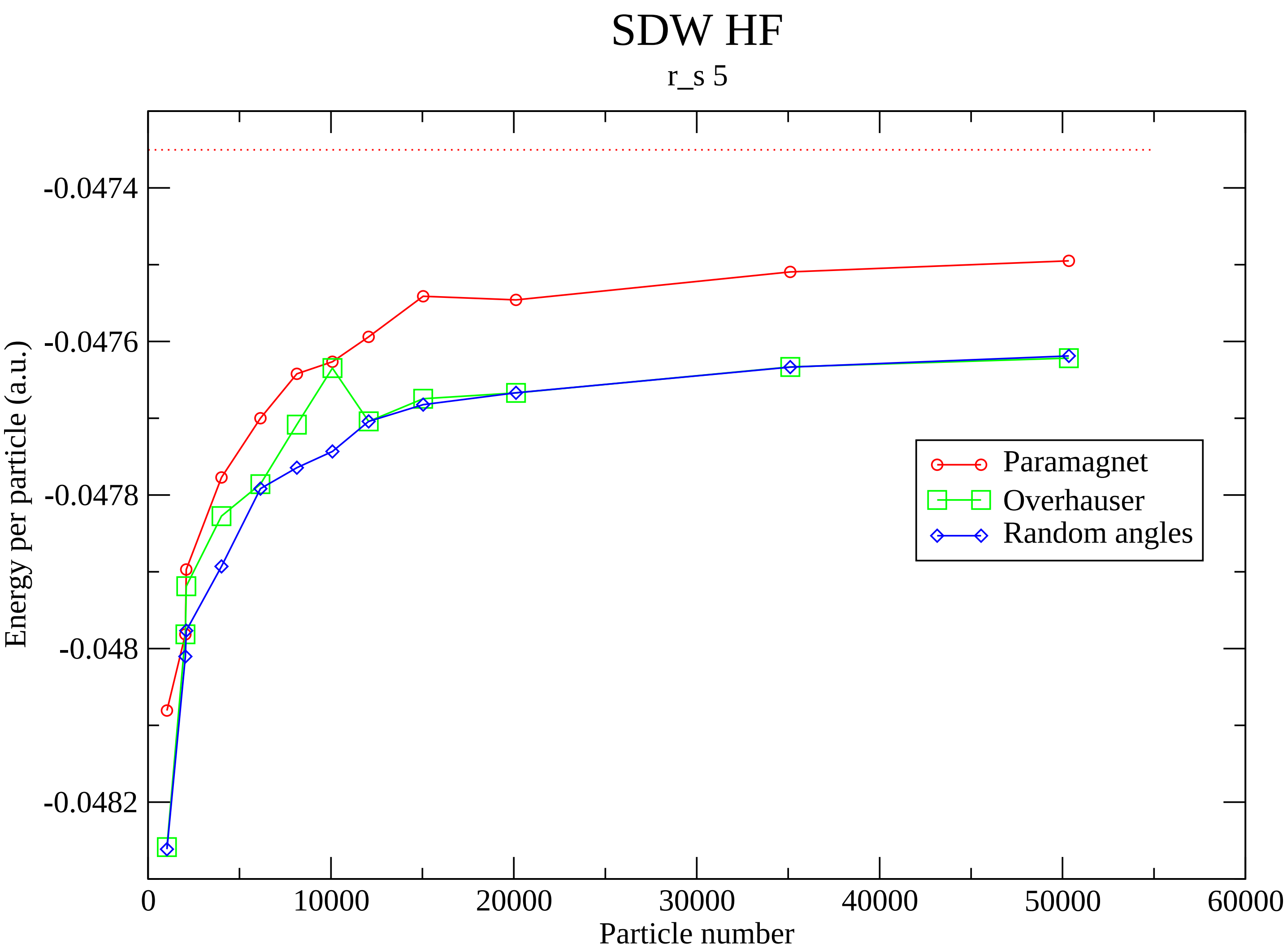
- Several schemes: Best guess at solution ("Overhauser-like"). Paramagnetic occupation with randomized angles. Combination of the two. Starting from converged result of another calculation.

- Iterate to self-consistency.

- Consistency of result: total energy vs. sum of eigenvalues.

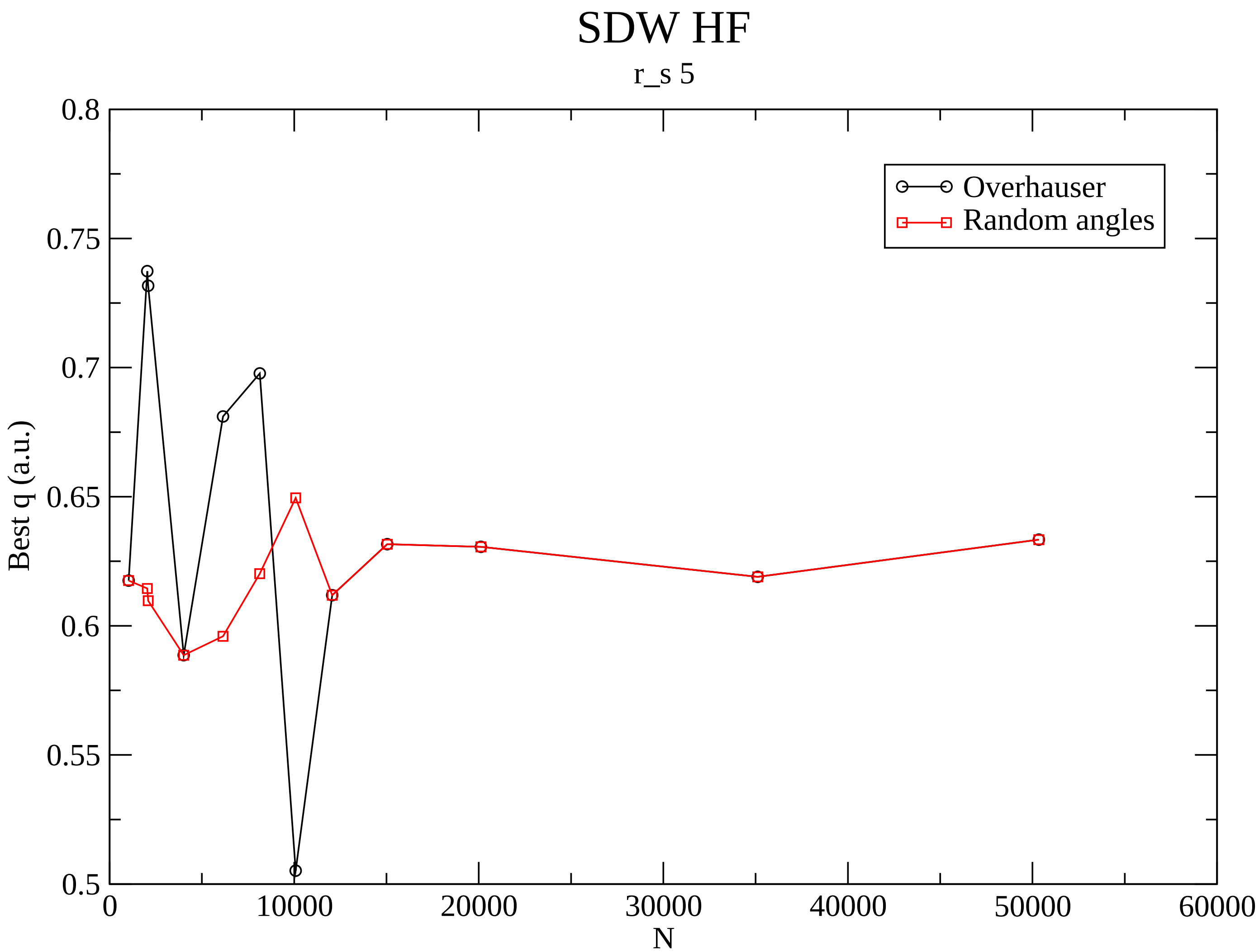
- Numerically tricky, false local minima, lack of convergence.

Results: Energy vs. System size



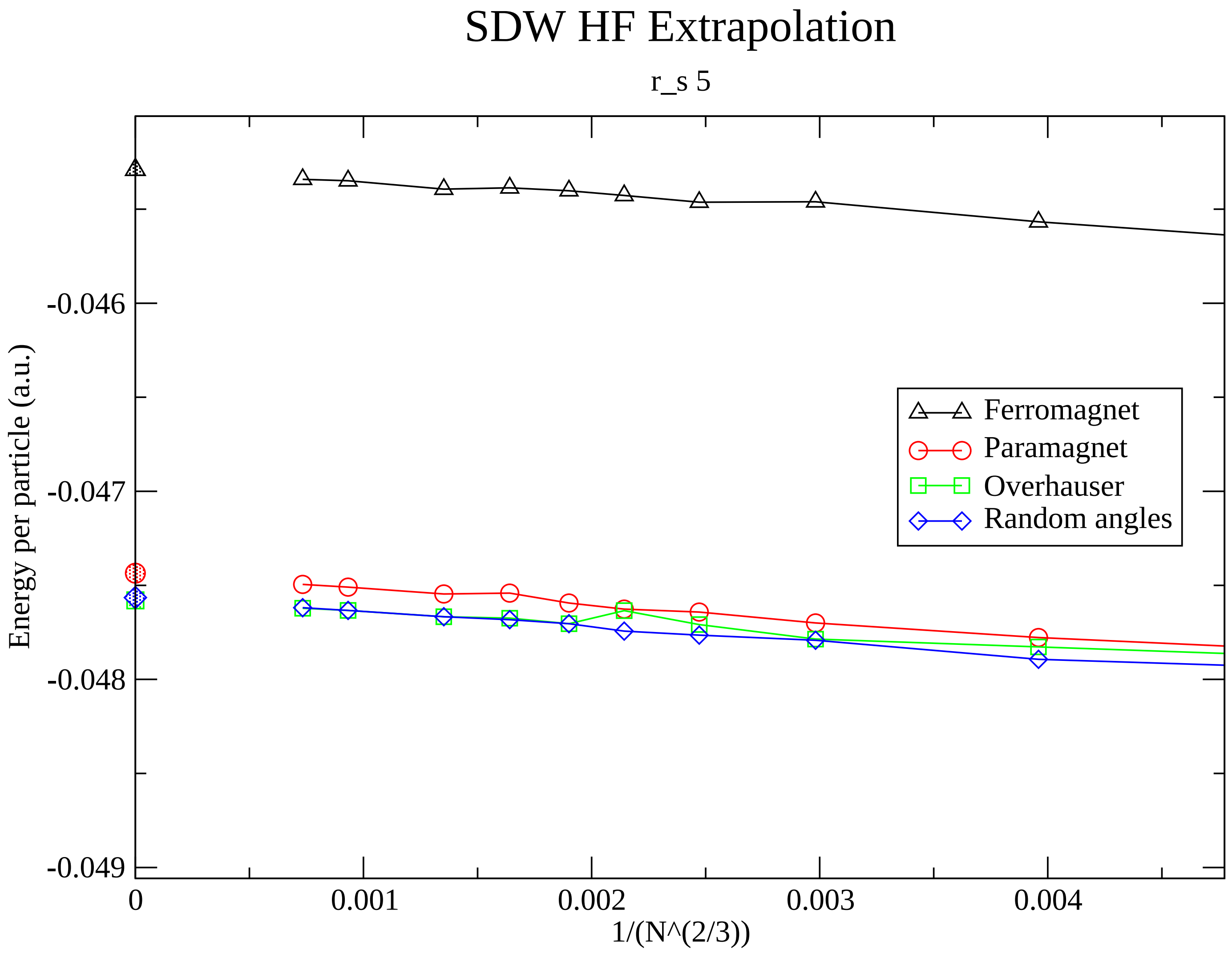
Energy from best value of q

Results: Best Q



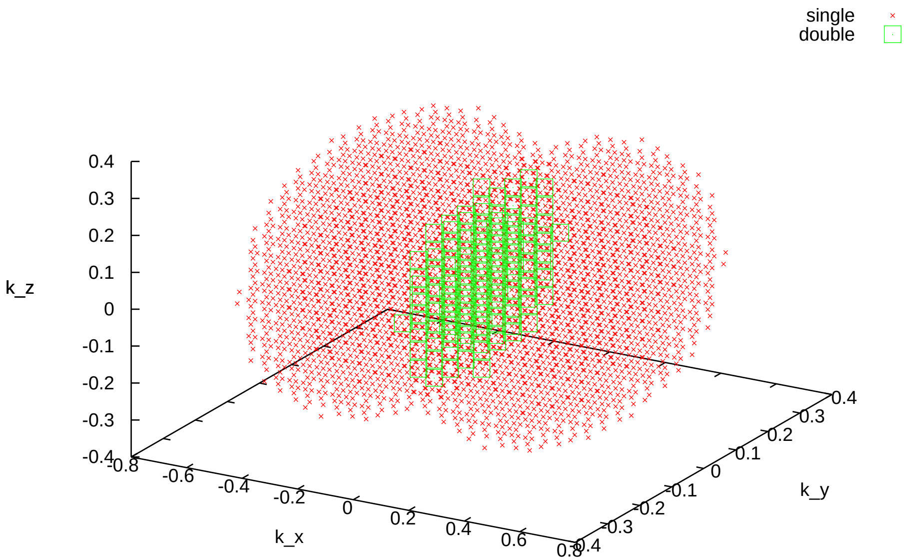
Best value of q converges. $2k_F = 0.768$.

Results: Energy vs. System size

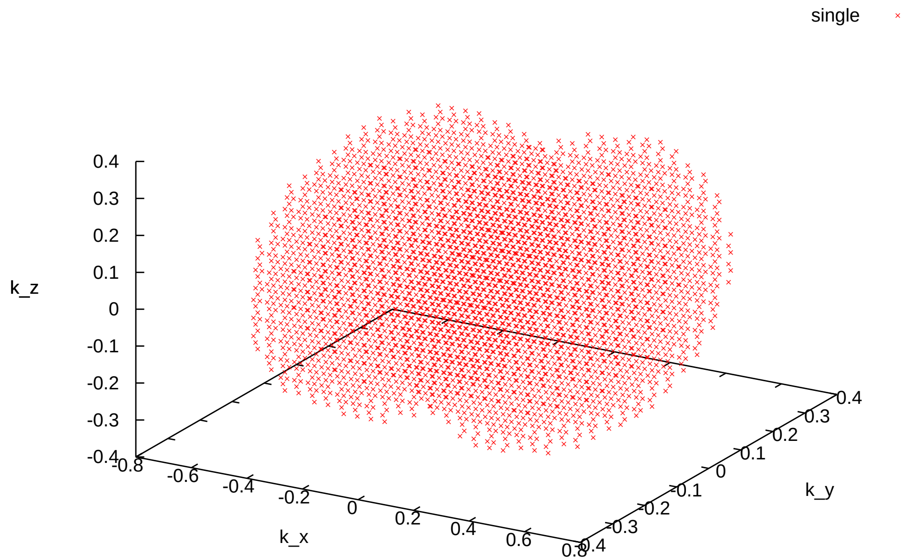


Extrapolating to infinite system size

Nature of the instability

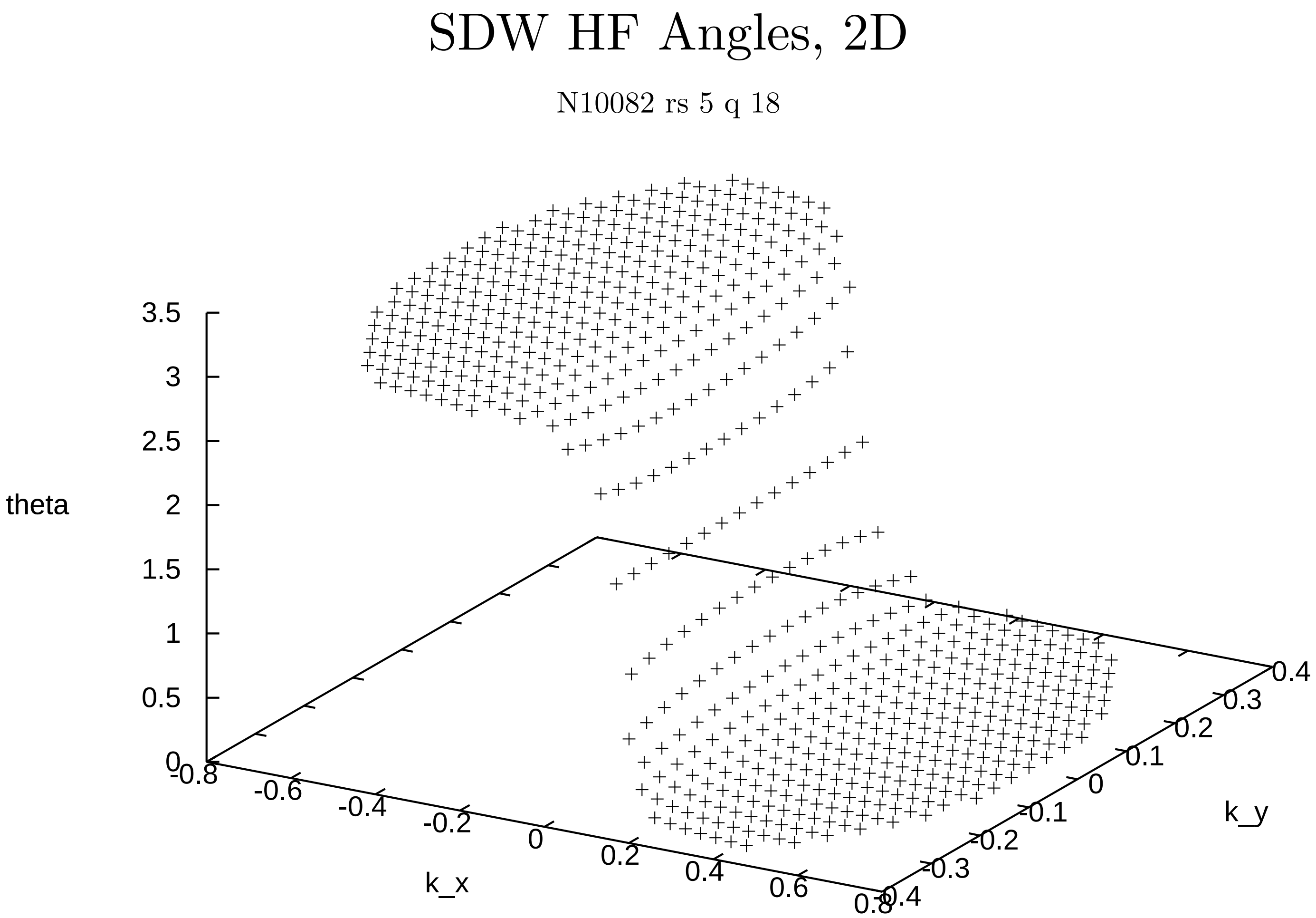


Occupation in $\mathbf{k}$-space at start
(paramagnet-like starting scheme)

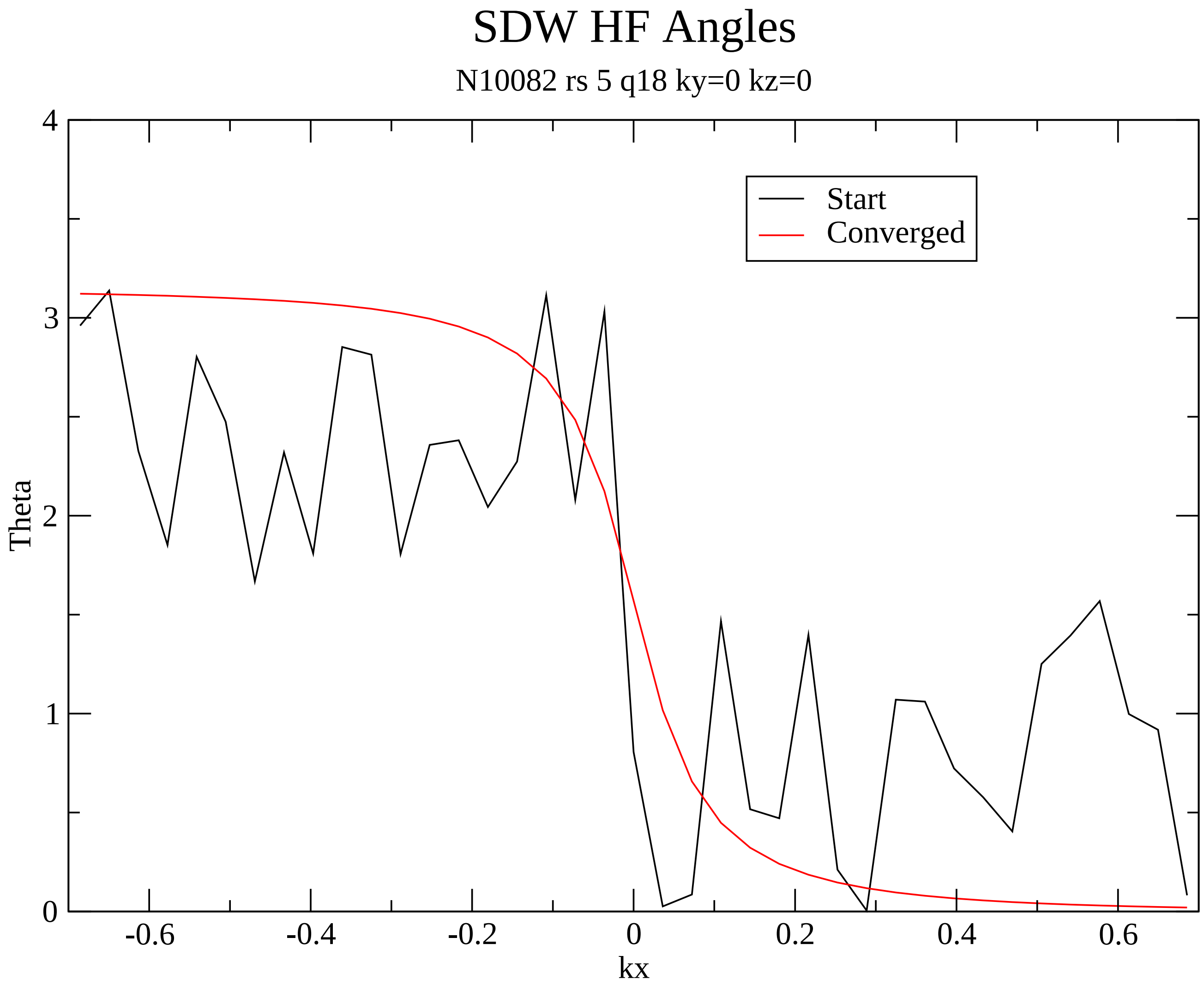


Occupation in $\mathbf{k}$-space at convergence

Nature of the instability

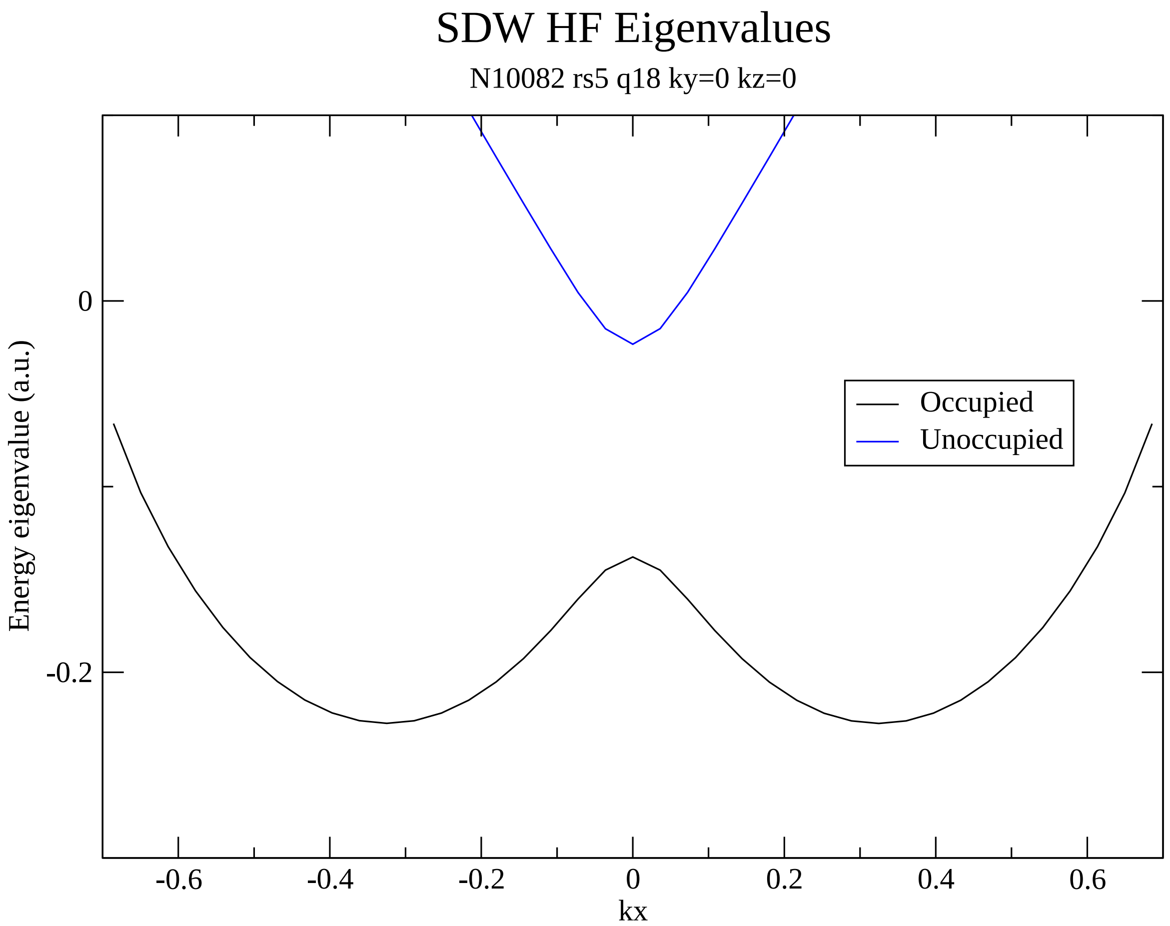


Nature of the instability

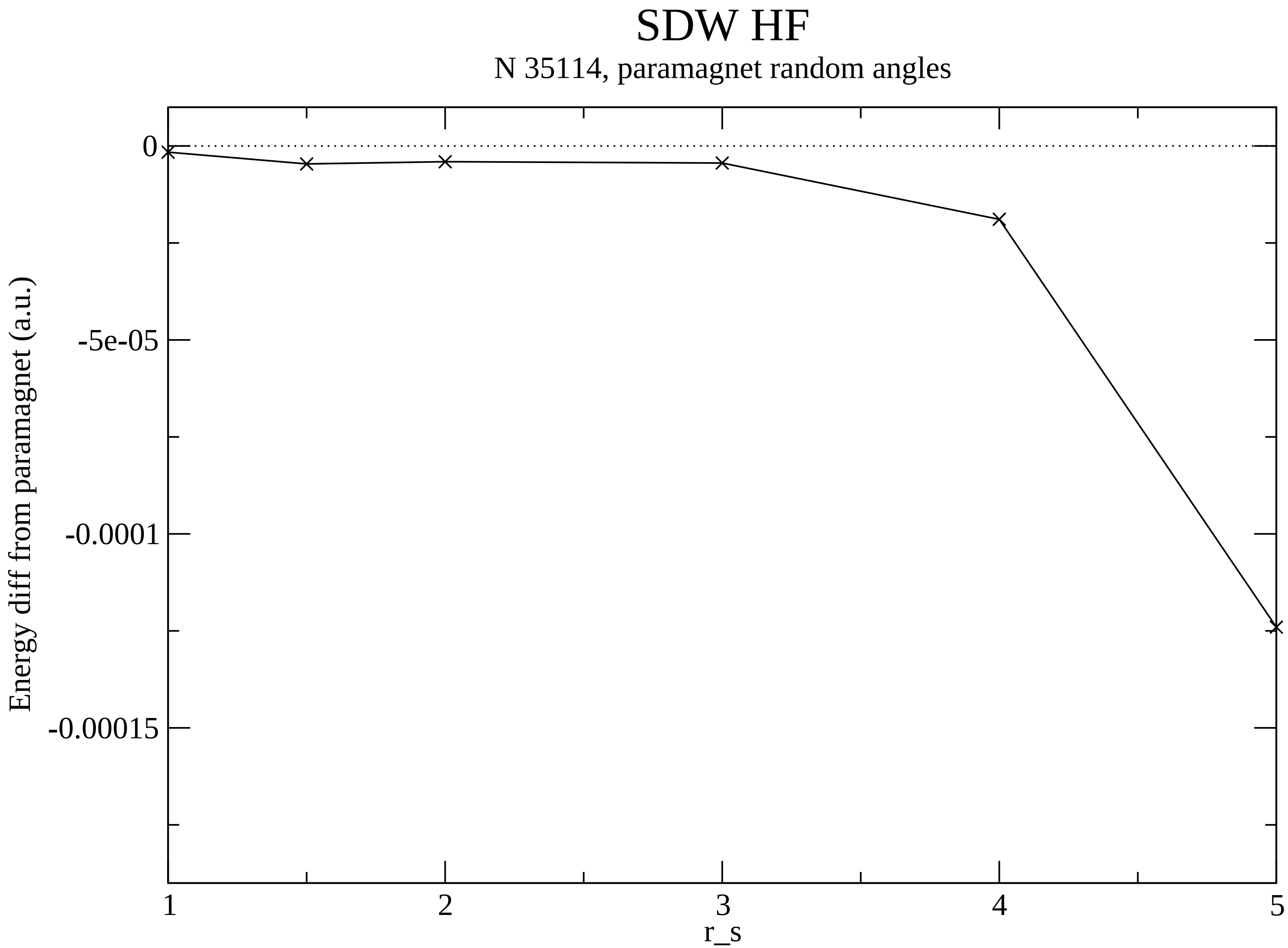


Nature of the instability

Instability is associated with formation of an energy gap. The spiral spin density is a short-wavelength periodic structure.



Results: Energy vs. r_s



Effect gets smaller at higher density, questionable at $r_s = 1.0$.

The problems

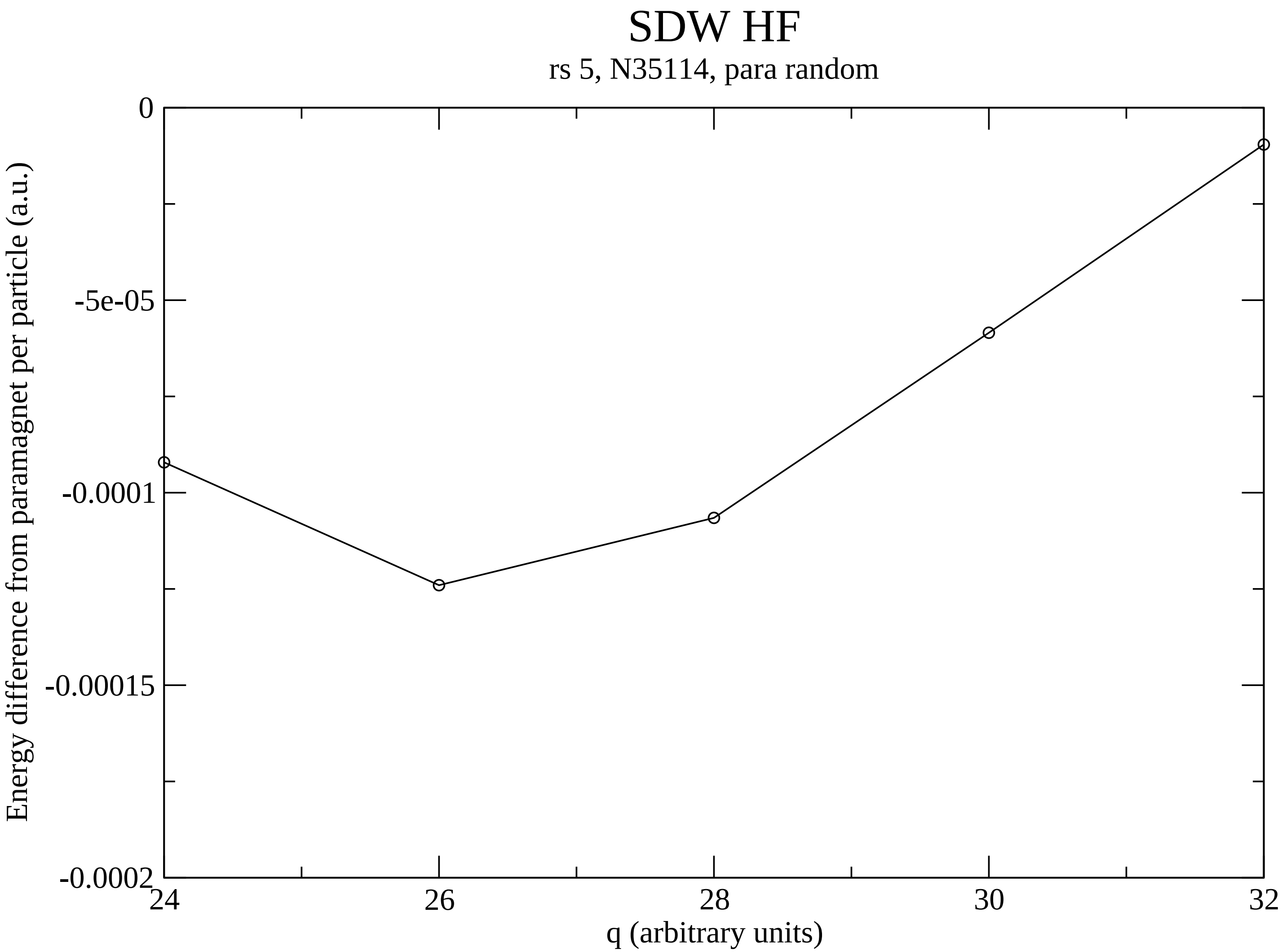
- Converged results at $r_s = 1.0$ are paramagnetic like. (Double occupancy, θ_k step-like)

- At $r_s = 1.0$, particle number 50346, all calculations converge to paramagnet!

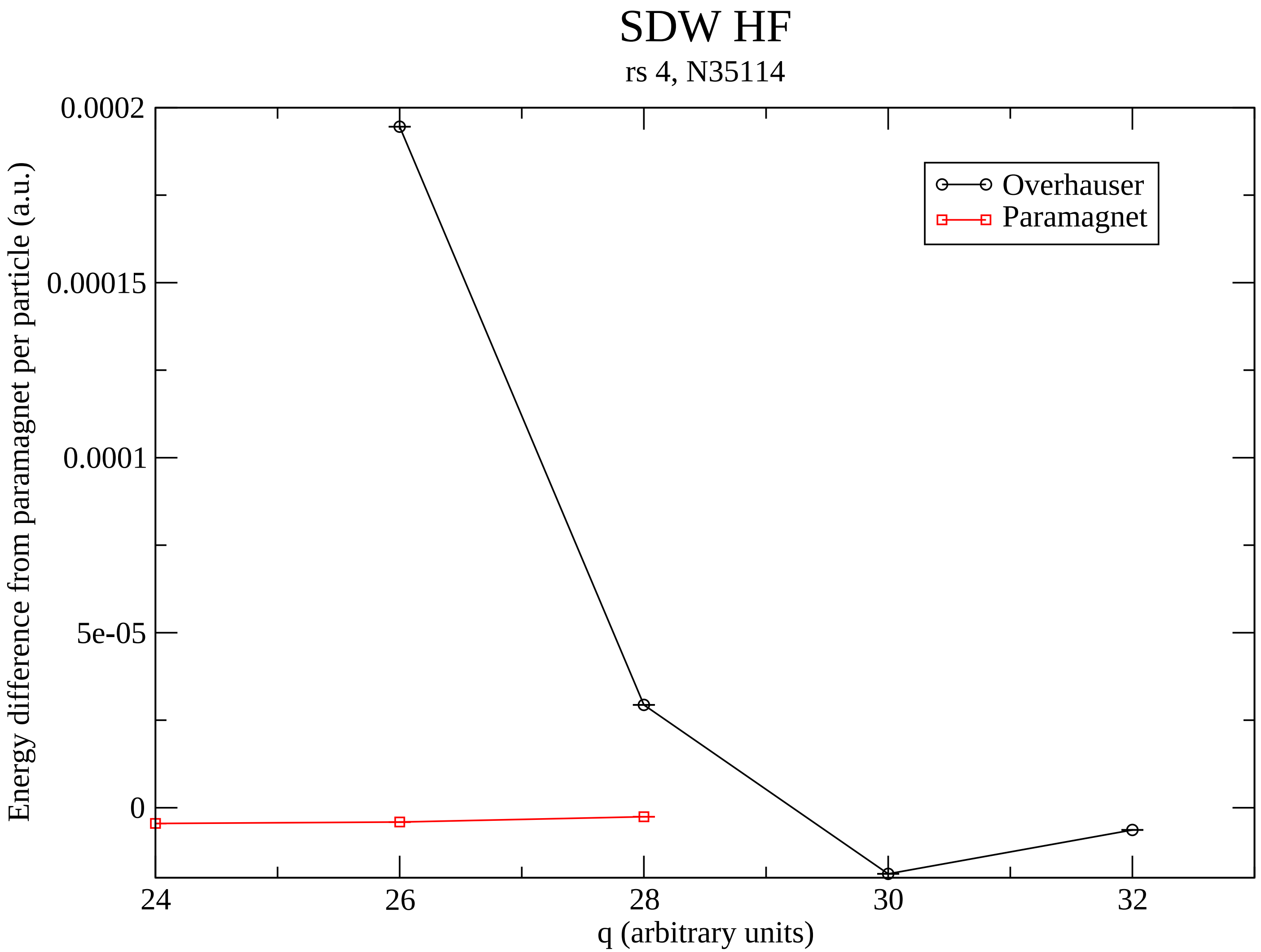
- At $r_s = 4.0$, for some values of q, converges to states with energy higher than paramagnet. This is independent of starting schemes.

- Thorough look at what's happening at $r_s = 4.0$: Local minima!

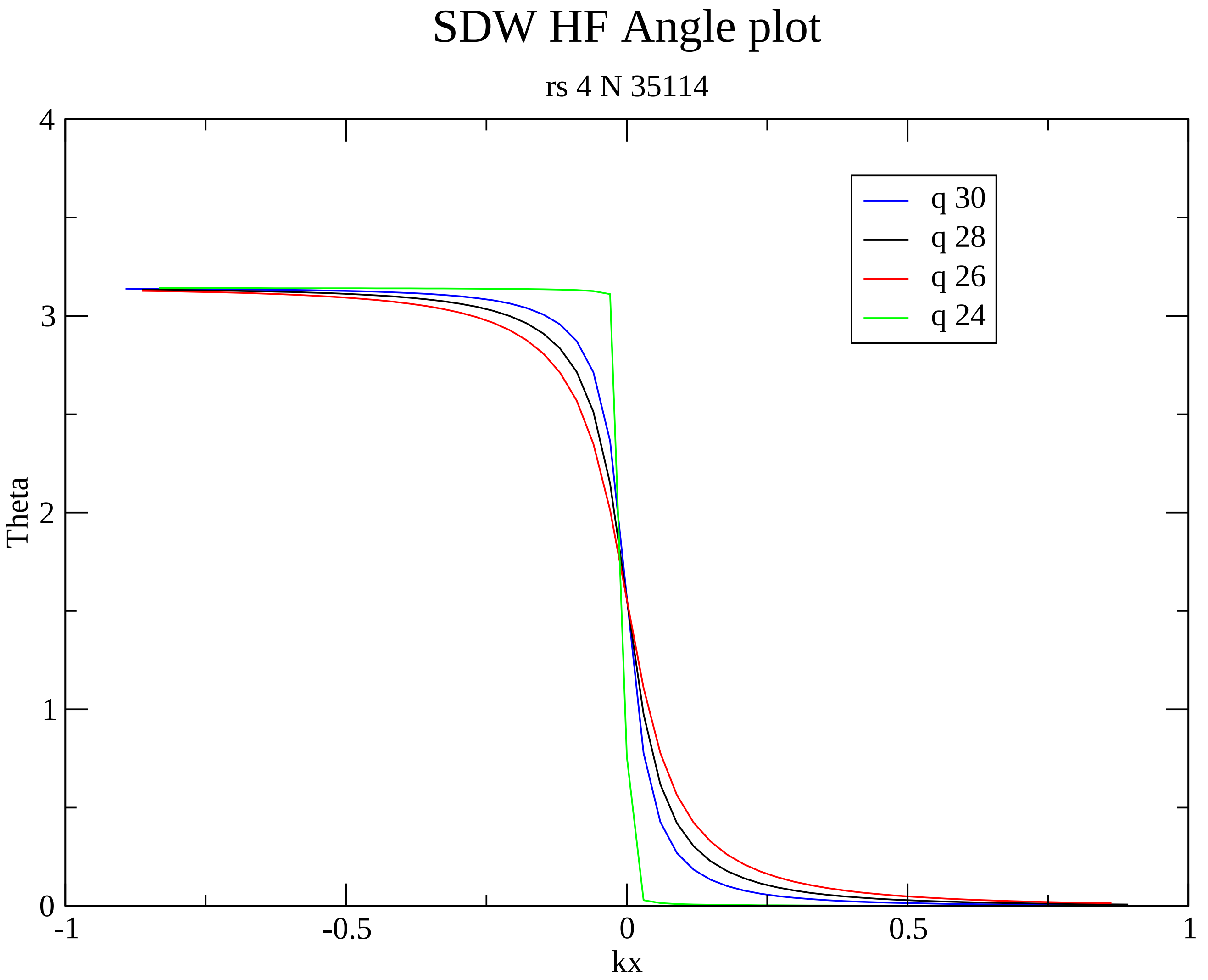
Local minima: $r_s = 5.0$



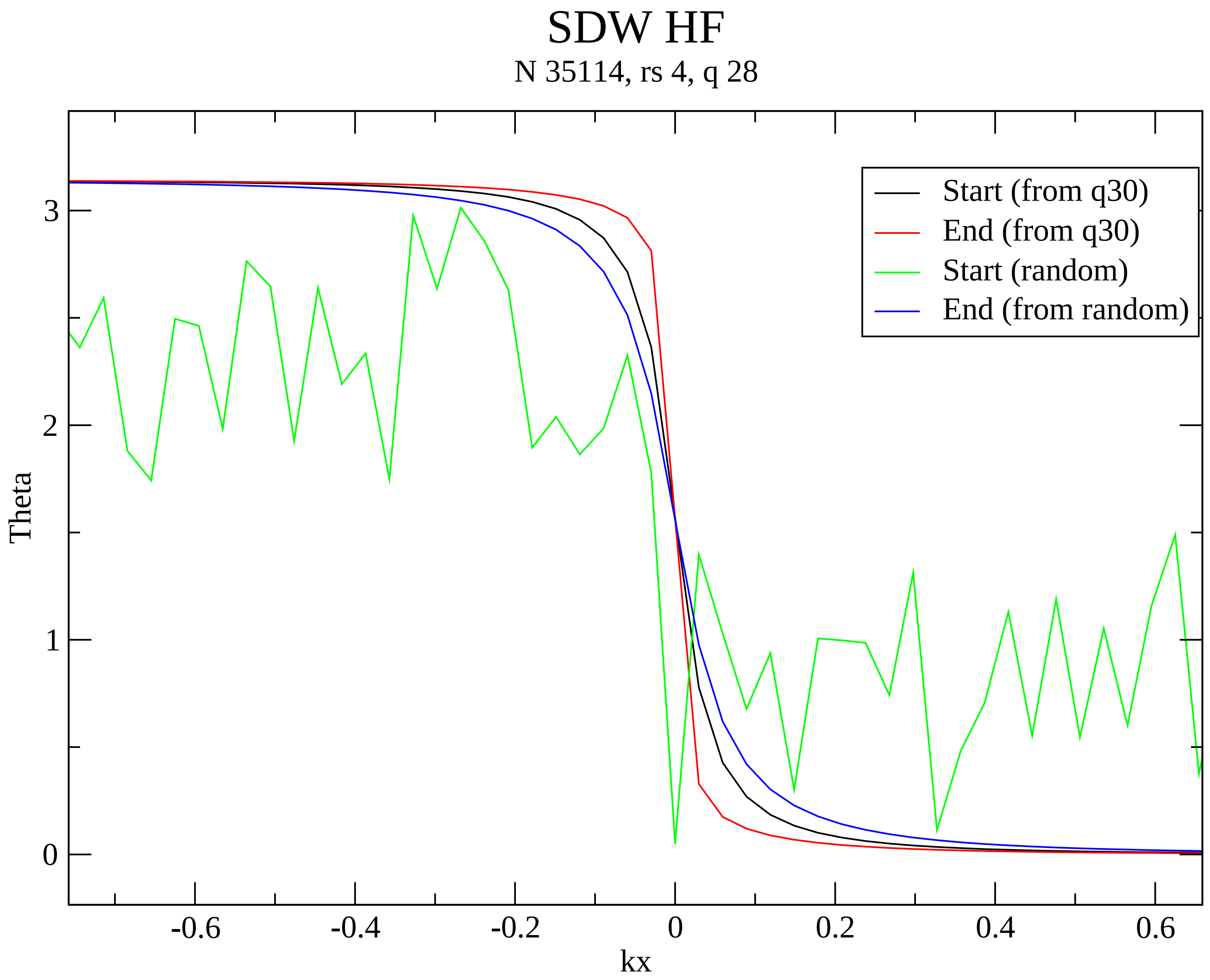
LOCAL MINIMA: $r_s = 4.0$



Local minima: Theta vs. q



Local minima: Different starting states



Conclusions

- Demonstrated the existence of the instability for some densities. Its nature is qualitatively as predicted by Overhauser.

- Converged occupation of $\mathbf{k}$-space and shape of θ_k consistent with each other. Can clearly see instability when it's present.

- For high densities, may get slightly lower energy than paramagnet but it's not Overhauser instability.

- Starting choice of θ_k more important than starting occupation of $\mathbf{k}$-space in determining what minimum the calculation converges to.

Acknowledgments

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