

Thermal and magnetic properties of spin ladder and chain compounds

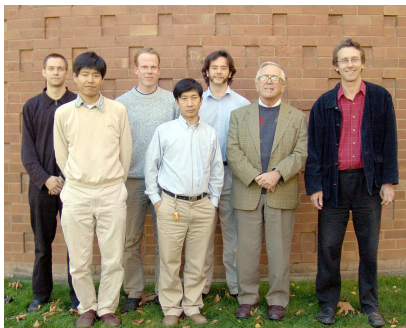
Application of the TBA and QTM to realistic models

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We have now had more than a half century since the work of Hans Bethe (1931) in which the exact solution of the Heisenberg magnet was found. Over the past few decades considerable effort has been invested in exactly solved models in statistical mechanics. Various methods for the integrability approach have been established, e.g. quantum inverse scattering method(**QISM**), thermodynamic Bethe ansatz(**TBA**), quantum transfer matrix method(**QTM**), conformal field theory, etc. Recently, there has been a revival of interest in the integrable systems which describe real physical models in condensed matter physics, quantum field theory, low-dimensional spin systems, Bose-Einstein condensates, quantum degenerate gases of ultracold atoms, etc.

- ▶ Haldane conjecture in 1D antiferromagnetic Heisenberg chains(similarity has been found in spin ladder models)
- ▶ Luttinger liquids: strongly correlated electron systems, quantum degenerate gases,
- ▶ Long range interaction: Calogero-Sutherland models, Gaudin magnet, BCS model

Outline

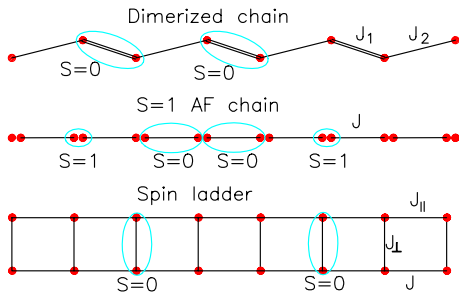
Theory: TBA and QTM

Spin- $\frac{1}{2}$ Ladders

- ▶ CuHpCl
- ▶ B5i2aT
- ▶ BPCB

Spin-1 Chains:

- ▶ NENC
- ▶ NBYC
- ▶ NDPK



- ▶ dimerized chains
- ▶ spin-1 chain
- ▶ spin ladders

1D Hamiltonians:

$$\text{su(N) Chain } H = J \sum_{i=1}^L P_{i,i+1}^{(N)} + \sum_{i=1}^L \mu_i$$

$$\text{Spin-}\frac{1}{2}\text{ Chain } H = J \sum_{i=1}^L (\vec{\mathbf{s}}_i \cdot \vec{\mathbf{s}}_{i+1}) - \sum_{i=1}^L \mu_{BG} H S_i^z$$

$$\text{Spin-1 Chain } H = J \sum_{i=1}^L (\vec{\mathbf{s}}_i \cdot \vec{\mathbf{s}}_{i+1} + (\vec{\mathbf{s}}_i \cdot \vec{\mathbf{s}}_{i+1})^2) + \sum_{i=1}^L D (S_i^z)^2 + E (S_i^{x2} - S_i^{y2}) - \mu_{BG} H S_i^{x/z}$$

$$\text{Spin-}\frac{1}{2}\text{ Ladder } H = J \sum_{i=1}^L \left(\left(\vec{\mathbf{s}}_i \cdot \vec{\mathbf{s}}_{i+1} + \frac{1}{4} \right) \left(\vec{\mathbf{T}}_i \cdot \vec{\mathbf{T}}_{i+1} + \frac{1}{4} \right) \right) + \sum_{i=1}^L J_{\perp} \vec{\mathbf{s}}_i \cdot \vec{\mathbf{T}}_i - \mu_{BG} H (S_i^z + T_i^z)$$

Interaction → **Exact Solution from the Bethe Ansatz**
On-Site → **Model Physics of the Real Compound**

Spin- $\frac{1}{2}$ ladders: theory

$$\mathcal{H} = J_{\parallel} \sum_{j=1}^L (\vec{S}_j \cdot \vec{S}_{j+1} + \vec{T}_j \cdot \vec{T}_{j+1}) + J_{\perp} \sum_{j=1}^L \vec{S}_j \cdot \vec{T}_j - \mu_B g H \sum_{j=1}^L (S_j^z + T_j^z)$$

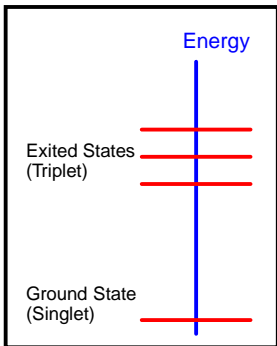
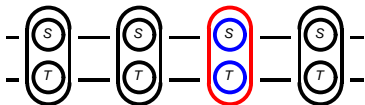
▶ **spin- $\frac{1}{2}$ Heisenber ladder model**

- ▶ \vec{S}_j and \vec{T}_j : spin- $\frac{1}{2}$ operators acting on the site j of the upper and lower leg
- ▶ J_{\parallel} and J_{\perp} : the intrachain and interchain coupling strength
- ▶ H : an external magnetic field; μ_B : the Bohr magneton; g : the Landé factor

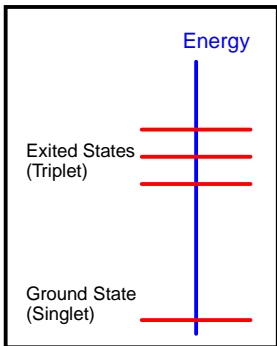
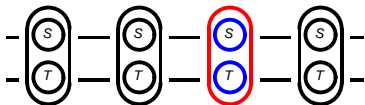
strong coupling: energy gap $\Delta \approx J_{\perp} - J_{\parallel}$; susceptibility $\chi \propto e^{-\Delta/T} / \sqrt{T}$; critical fields: $H_{c1} \approx J_{\perp} - J_{\parallel}$, $H_{c2} \approx J_{\perp} + 2J_{\parallel}$

weak coupling: energy gap: $\Delta \approx J_{\perp}/2$, existence of long-range order

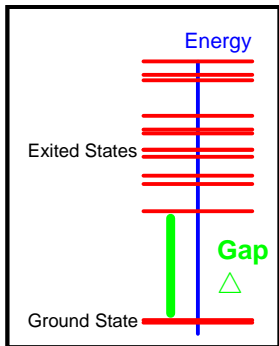
reference: Dagotto, Rep. Prog. Phys. 62(99)2525; S. Sachdev Science 288(00)475; Troyer et al. PRB 50(94)13515; Johnston Cond-mat/0001147; Tsvelik et al.; Affleck, Chaboussant et al.; Sierra; White; Rice; Milla; . . .



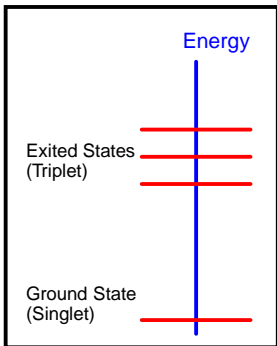
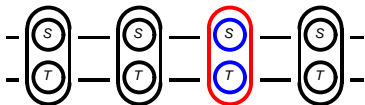
Take single rung:
 two coupled Spin- $\frac{1}{2}$: $J_{\perp} \vec{S} \cdot \vec{T}$



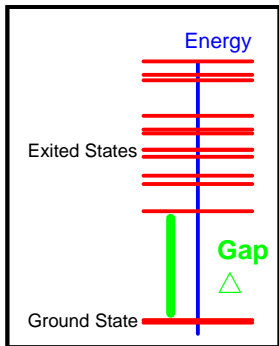
Take single rung:
two coupled Spin- $\frac{1}{2}$: $J_{\perp} \vec{S} \cdot \vec{T}$



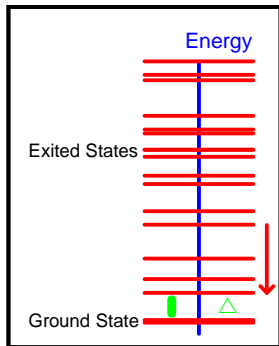
Energy spectrum of whole ladder



Take single rung:
two coupled Spin- $\frac{1}{2}$: $J_{\perp} \vec{S} \cdot \vec{T}$



Energy spectrum of whole ladder



Closing of gap Δ by external
parameter, e.g. magnetic field H

Spin- $\frac{1}{2}$ ladders: theory

integrable spin ladder $\mathcal{H} = J_{\parallel} \mathcal{H}_{\text{leg}} + J_{\perp} \sum_{j=1}^L \vec{S}_j \cdot \vec{T}_j - \mu_B g H \sum_{j=1}^L (S_j^z + T_j^z)$

Wang, PRB 60(99)9236 $\mathcal{H}_{\text{leg}} = \sum_{j=1}^L \left(\vec{S}_j \cdot \vec{S}_{j+1} + \vec{T}_j \cdot \vec{T}_{j+1} + 4\vec{S}_j \cdot \vec{S}_{j+1} \vec{T}_j \cdot \vec{T}_{j+1} \right)$

eigenbasis $|1\rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$, $|2\rangle = |\uparrow\uparrow\rangle$,
 $|3\rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$, $|4\rangle = |\downarrow\downarrow\rangle$,

energy $\mathcal{E} = -J_{\parallel} \sum_{i=1}^{M_1} \frac{1}{(v_i^{(1)})^2 + \frac{1}{4}} - J_{\perp} N_0 - \mu_B g (N_+ - N_-) H$

Bethe ansatz equations

$$\prod_{i=1}^{M_{k-1}} \frac{v_j^{(k)} - v_i^{(k-1)} + \frac{i}{2}}{v_j^{(k)} - v_i^{(k-1)} - \frac{i}{2}} = \prod_{\substack{l=1 \\ l \neq j}}^{M_k} \frac{v_j^{(k)} - v_l^{(k)} - i}{v_j^{(k)} - v_l^{(k)} + i} \prod_{l=1}^{M_{k+1}} \frac{v_j^{(k)} - v_l^{(k+1)} - \frac{i}{2}}{v_j^{(k)} - v_l^{(k+1)} + \frac{i}{2}}$$

Spin- $\frac{1}{2}$ ladders: TBA

ground state

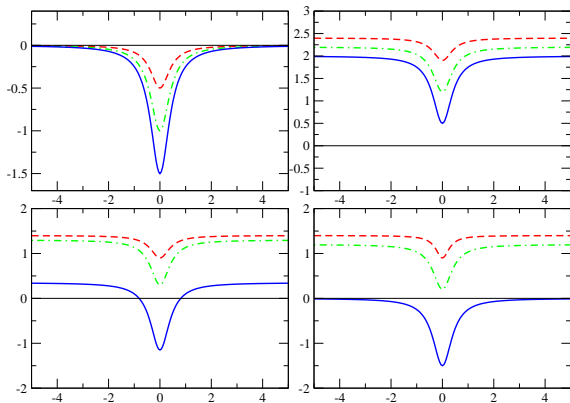
$$\begin{aligned}\epsilon^{(1)} &= g_1^{(1)} - a_2 * \epsilon^{(1)-} + a_1 * \epsilon^{(2)-} \\ \epsilon^{(2)} &= g_1^{(2)} - a_2 * \epsilon^{(2)-} + a_1 * [\epsilon^{(1)-} + \epsilon^{(3)-}] \\ \epsilon^{(3)} &= g_1^{(3)} - a_2 * \epsilon^{(3)-} + a_1 * \epsilon^{(2)-}\end{aligned}$$

$$\mathbf{H} < \mathbf{J}_\perp \quad g_1^{(1)} = -J_\parallel 2\pi a_1 + J_\perp - \mu_B g H$$

$$g_1^{(2)} = g_1^{(3)} = \mu_B g H$$

$$\mathbf{H} > \mathbf{J}_\perp \quad g_1^{(1)} = -J_\parallel 2\pi a_1 - J_\perp + \mu_B g H$$

$$g_1^{(2)} = J_\perp, \quad g_1^{(3)} = \mu_B g H$$



Spin- $\frac{1}{2}$ ladders: theory

critical fields

$$H_{c1} = (J_{\perp} - 4J_{\parallel})/\mu_B g$$

$$H_{c2} = (J_{\perp} + 4J_{\parallel})/\mu_B g$$

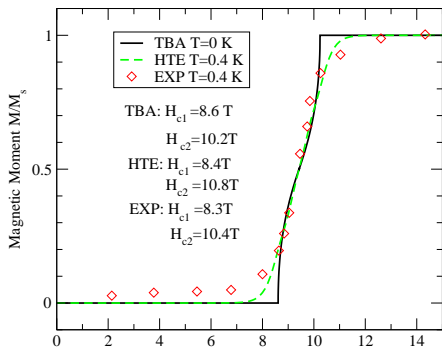
$$H_{IP} = J_{\perp}/\mu_B g$$

in the vicinity of H_{c1}

$$M^z \approx \frac{1}{\pi} \sqrt{\frac{\mu_B g (H - H_{c1})}{J_{\parallel}}}$$

in the vicinity of H_{c2}

$$M^z \approx 1 - \frac{1}{\pi} \sqrt{\frac{\mu_B g (H_{c2} - H)}{J_{\parallel}}}$$



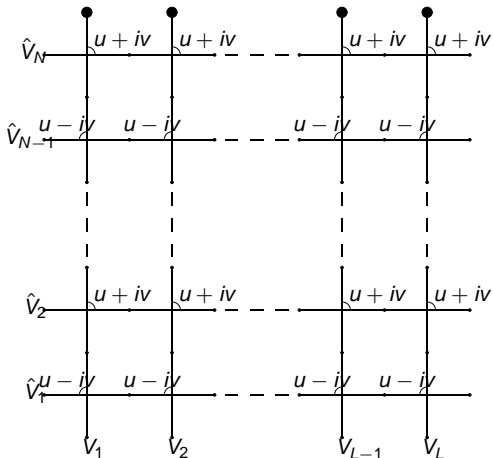
Magnetization versus magnetic field

- ▶ **B5i2aT** (Landee et al PRB 63(01)100402)
- ▶ TBA and HTE prediction: $J_{\perp} = 13.3$ K, $J_{\parallel} = 0.2875$ K
- ▶ inflection point $H = J_{\perp}/\mu_B g$

Spin- $\frac{1}{2}$ ladders: QTM

1D quantum model \rightarrow 2D classical model :

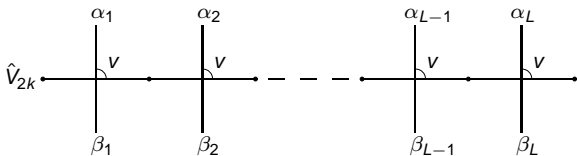
$$Z = \text{tr}_{V_1 \otimes \dots \otimes V_L} e^{-\beta H} = \lim_{N \rightarrow \infty} \text{tr}_{V_1 \otimes \dots \otimes V_L} e^{-\beta H_{ch}} (t(u_N) \tilde{t}(u_N))^{\frac{N}{2}}$$



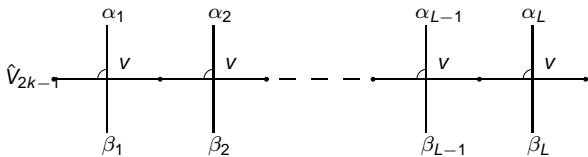
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 Suzuki, Nucl Phys B487
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 Klümper Z. Physik
 B91(93)507;
 Tsuboi JPA 37(04)1747;
 Tsuboi and M. Takahashi**;
 Kuniba, Nakanishi and
 Suzuki, Int. J. Mod. Phys.
 A 9(94)5215;
 9(94)5267

Spin- $\frac{1}{2}$ ladders: QTM

$$t(v) = \text{tr}_{\hat{V}_{2k}} (R_{2k,L}(v) \cdots R_{2k,2}(v) R_{2k,1}(v)) \quad k = 1, 2, \dots, \frac{N}{2}$$



$$\tilde{t}(v) = \text{tr}_{\hat{V}_{2k-1}} (\tilde{R}_{2k-1,L}(v) \cdots \tilde{R}_{2k-1,2}(v) \tilde{R}_{2k-1,1}(v)) \quad k = 1, 2, \dots, \frac{N}{2}$$



Spin- $\frac{1}{2}$ ladders: QTM

NLIE $T_1^{(a)}(v) = Q_1^{(a)} + \oint_{C^{(a)}} \frac{dy}{2\pi i} \frac{T_1^{(a-1)}(y + \beta_1^{(a)} - \frac{i}{2}) T_1^{(a+1)}(y + \beta_1^{(a)} - \frac{i}{2})}{(v - y - \beta_1^{(a)}) T_1^{(a)}(y + \beta_1^{(a)} - i)}$

$$\oint_{\bar{C}^{(a)}} \frac{dy}{2\pi i} \frac{T_1^{(a-1)}(y - \beta_1^{(a)} + \frac{i}{2}) T_1^{(a+1)}(y - \beta_1^{(a)} + \frac{i}{2})}{(v - y + \beta_1^{(a)}) T_1^{(a)}(y - \beta_1^{(a)} + i)}$$

for $a \in \{1, 2, \dots, r\}$

HTE $T_1^{(a)}(v) = \exp\left(\sum_{n=0}^{\infty} b_n^{(a)}(v) \left(\frac{J_{\parallel}}{T}\right)^n\right), \quad a \in \{1, 2, \dots, r\}$

$$b_n^{(a)}(v) = \sum_{j=0}^{n-1} \frac{c_{n,j}^{(a)} v^{2j}}{(v^2 + \frac{(a+1)^2}{4})^n}$$

freeenergy $-\frac{1}{T} f_{\text{HTE}}(T, H) = \ln Q_1^{(1)} + C_{1,0}^1 \left(\frac{J}{T}\right) + C_{2,0}^1 \left(\frac{J}{T}\right)^2 + C_{3,0}^1 \left(\frac{J}{T}\right)^3 + \dots$

- ▶ High Temperature Expansion (HTE) predicts **free energy** $f_{\text{HTE}}(T, H)$ per site
- ▶ Obtain thermodynamical properties:

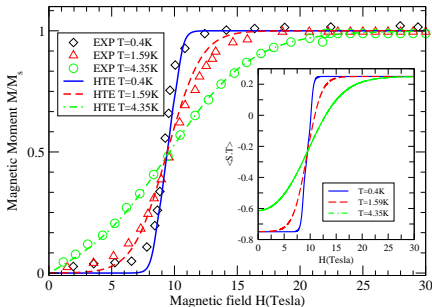
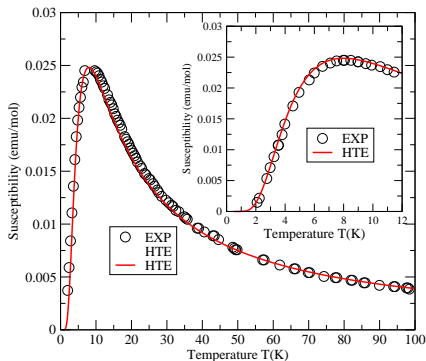
$$\text{Magnetization: } M(H) = -\frac{\partial}{\partial H} f_{\text{HTE}}(T, H)|_T$$

$$\text{(magnetic) Susceptibility: } \chi(H) = -\frac{\partial^2}{\partial H^2} f_{\text{HTE}}(T, H)|_T$$

$$\text{(magnetic) Specific Heat: } C(T) = -T \frac{\partial^2}{\partial T^2} f_{\text{HTE}}(T, H)|_H$$

- ▶ Concentrate on these 3 properties because **experimental data widely available** for many compounds

Spin- $\frac{1}{2}$ ladders: thermal and magnetic properties



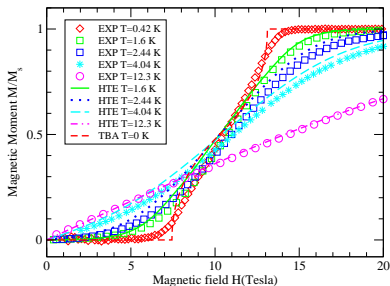
Susceptibility versus temperature

- ▶ **B5i2aT** (Landee et al PRB 63(01)100402)
- ▶ coupling constants $J_{\perp} = 13.3$ K, $J_{\parallel} = 0.2875$ K, $g = 2.1$

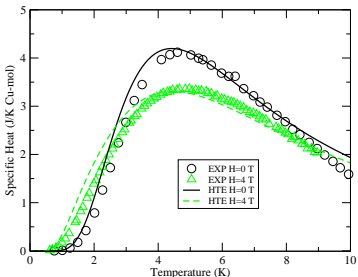
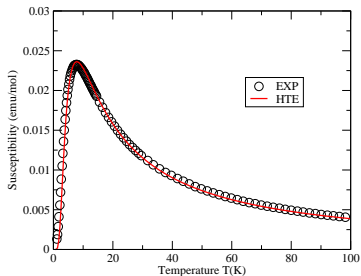
Magnetization versus magnetic field

- ▶ TBA: $H_{c1} = 8.6$ T, $H_{c2} = 10.02$ T
- ▶ EXP: $H_{c1} = 8.3$ T, $H_{c2} = 10.4$ T

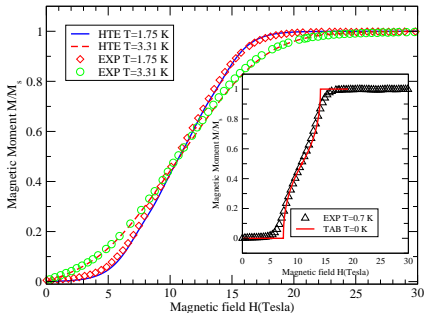
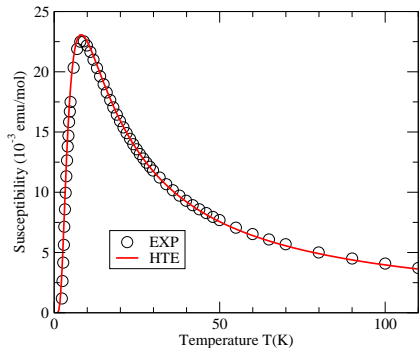
Spin- $\frac{1}{2}$ ladders: thermal and magnetic properties



- ▶ **CHpCl** (Chaboussant et al PRL 79(98)925; PRL 80(98)2713; Hagiwara et al PRB 62(00)1051)
- ▶ coupling constants $J_{\perp} = 14.5\text{K}$, $J_{\parallel} = 1.0\text{K}$, $g = 2.1$
- ▶ TBA: $H_{c1} = 7.4\text{T}$, $H_{c2} = 13.1\text{T}$
- ▶ EXP: $H_{c1} = 7.5\text{T}$, $H_{c2} = 13.0\text{T}$



Spin- $\frac{1}{2}$ ladders: thermal and magnetic properties



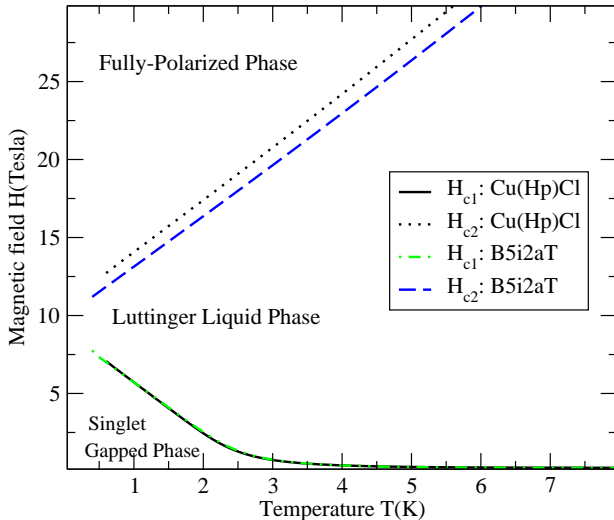
Susceptibility versus temperature

- ▶ $(\text{C}_5\text{H}_{12}\text{N})_2\text{CuBr}_4$ (Watson et al PRL 86(01)5168)
- ▶ coupling constants $J_{\perp} = 15.4$ K, $J_{\parallel} = 1.2$ K, $g = 2.1$

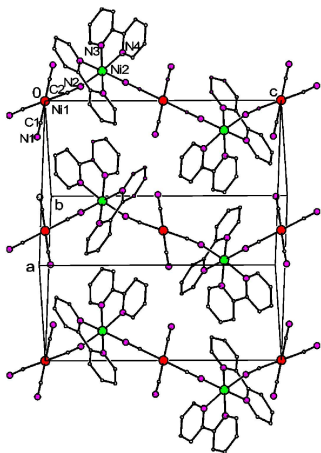
Magnetization versus magnetic field

- ▶ TBA: $H_{c1} = 7.4$ T, $H_{c2} = 14.1$ T
- ▶ EXP: $H_{c1} = 6.6$ T, $H_{c2} = 14.6$ T

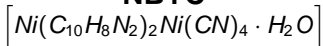
Spin- $\frac{1}{2}$ ladders: thermal and magnetic properties



Spin-1 chains

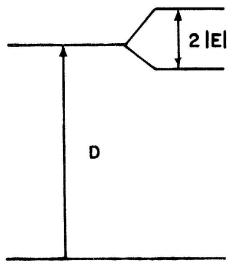


NBYC



- ▶ NENC similar structure $[Ni(C_2H_8N_2)_2Ni(CN)_4]$
- ▶ paramagnetic Nickel Ni^{2+} ion (●)
- ▶ diamagnetic Nickel Ni^{2+} ion (●)
- ▶ Simplifications
 - only red Ni^{2+} active in chain, carry Spin-1
 - indirect superexchange via green/violet cluster
 - *antiferromagnetic* coupling between red Ni^{2+}
 - very weak intra-chain interaction \rightarrow 1D structure
- ▶ **Experimentalists Conclusion:**
Both Antiferromagnetic Spin-1 Chains

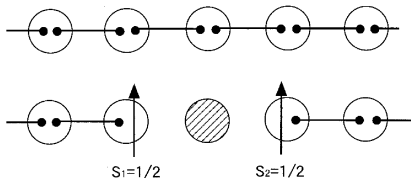
Spin-1 chains



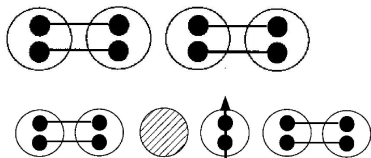
- ▶ Gapped phase induced by crystal field anisotropies
- ▶ Model Hamiltonian: anisotropies

$$\mathcal{H}_1 = D \sum_{j=1}^L (S_j^z)^2 + E \sum_{j=1}^L \left((S_j^x)^2 - (S_j^y)^2 \right)$$

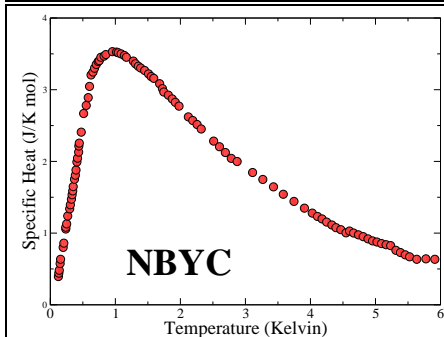
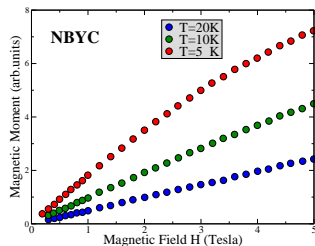
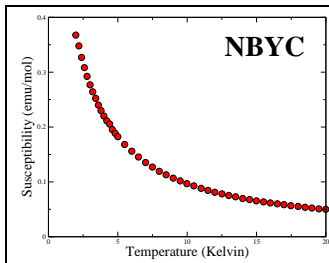
VBS state



dimerized state



Spin-1 chains



$$\mathcal{H} = J \sum_{i=1}^L \vec{\mathbf{s}}_i \cdot \vec{\mathbf{s}}_{i+1} + D \sum_{i=1}^L (S_i^z)^2 + E \sum_{i=1}^L \left[(S_i^x)^2 - (S_i^y)^2 \right] - \mu_B g H \sum_{i=1}^L S_i^z$$

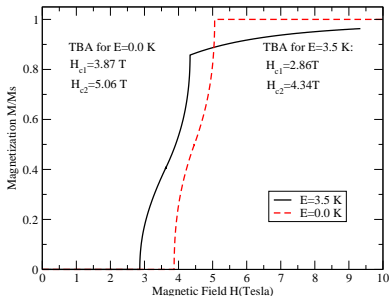
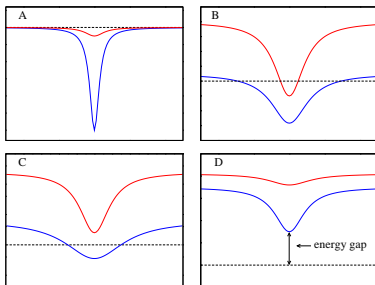
- ▶ J : spin-spin coupling strength
- ▶ D : single-ion anisotropy
- ▶ E : in-plane anisotropy

Spin-1 chains: TBA

ground state

$$\epsilon^{(1)} = g_1 + a_1 * \epsilon^{(2)} - a_2 * \epsilon^{(1)}$$

$$\epsilon^{(2)} = g_2 + a_1 * \epsilon^{(1)} - a_2 * \epsilon^{(2)}$$



- ▶ **dashed line:** gapped phase with Pokrovsky-Talapov transition
- ▶ **solid line:** gapped phase with transition

$$M_{H>H_{c2}}(H) = \frac{\mu_B g H}{\sqrt{E^2 + (\mu_B g H)^2}}$$

Critical fields and energy gap

$$\Delta = H_{c1} = \sqrt{(D - 4J)^2 - E^2} / \mu_B g$$

$$H_{c2} = \sqrt{(D + 4J)^2 - E^2} / \mu_B g$$

Spin-1 chains: QTM

free energy

$$f(T, H) = -T \left\{ \ln Q_1^{(1)} + C_{1,0}^1 \left(\frac{J}{T} \right) + C_{2,0}^1 \left(\frac{J}{T} \right)^2 + C_{3,0}^1 \left(\frac{J}{T} \right)^3 + \dots \right\}$$

Expansion coefficients

$$C_{1,0}^{(1)} = 2 \frac{Q_1^{(2)}}{Q_1^{(1)2}}$$

$$C_{2,0}^{(1)} = 3 \frac{Q_1^{(2)}}{Q_1^{(1)2}} - 6 \frac{Q_1^{(2)2}}{Q_1^{(1)4}} + 3 \frac{Q_1^{(3)}}{Q_1^{(1)3}}$$

$$C_{3,0}^{(1)} = \frac{10}{3} \frac{Q_1^{(2)}}{Q_1^{(1)2}} - 18 \frac{Q_1^{(2)2}}{Q_1^{(1)4}} + \frac{80}{3} \frac{Q_1^{(2)3}}{Q_1^{(1)6}} \\ + 8 \frac{Q_1^{(3)}}{Q_1^{(1)3}} - 24 \frac{Q_1^{(2)} Q_1^{(3)}}{Q_1^{(1)5}}$$

Q-System:

$$Q_1^{(1)} = e^{-\beta\mu_1} + e^{-\beta\mu_2} + e^{-\beta\mu_3}$$

$$Q_1^{(2)} = e^{-\beta\mu_1 - \beta\mu_2} + e^{-\beta\mu_1 - \beta\mu_3} + e^{-\beta\mu_2 - \beta\mu_3}$$

$$Q_1^{(3)} = e^{-\beta\mu_1 - \beta\mu_2 - \beta\mu_3}$$

Spin-1 chains: QTM

Powdered Samples

Synthesizing single crystals sometimes impossible

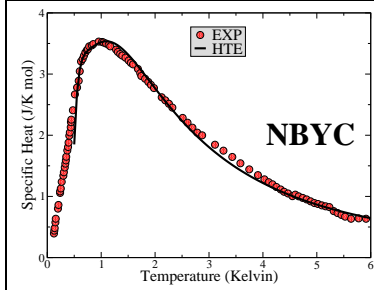
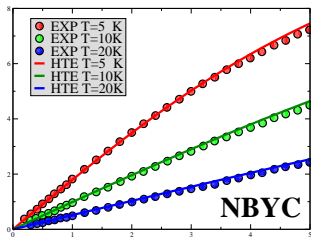
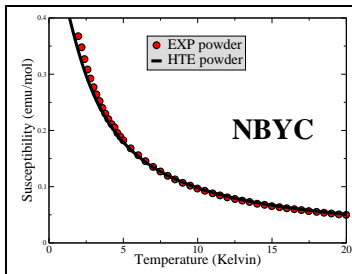
Powdered samples in all directions (naive averaging):

$$\chi_{\text{powder}} = \frac{2}{3}\chi_{\perp} + \frac{1}{3}\chi_{\parallel}$$

Chemical potential part:

$$-\mu_B g H_z \sum_{j=1}^L S_j^z + D \sum_{j=1}^L (S_j^x)^2 + E \sum_{j=1}^L ((S_j^z)^2 - (S_j^y)^2)$$

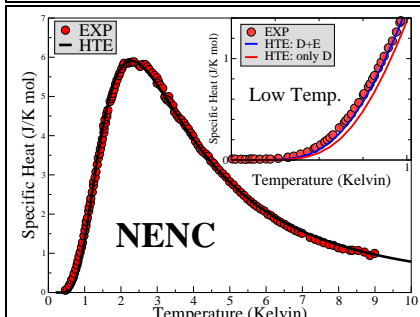
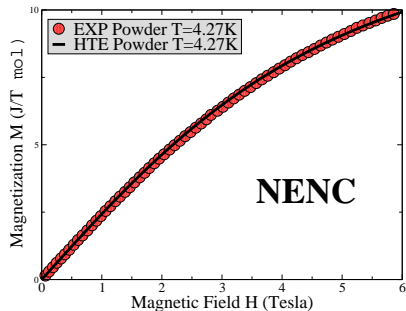
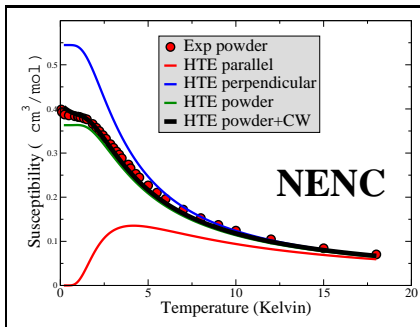
Spin-1 chains: thermal and magnetic properties



comparison

- ▶ HTE $J = 0.35\text{K}$ $D = 2.62\text{K}$ $E = 1.49$
- ▶ Heisenberg chain $J = 0.2\text{K}$
 $D = 2.55\text{K}$ $E = 1.5$
- ▶ TBA H_{c1} N/A, $H_{c2} = 2.7\text{T}$

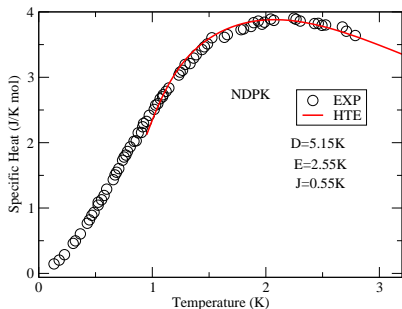
Spin-1 chains: thermal and magnetic properties



comparison:

- ▶ HTE $J = 0.17\text{K}$ $D = 6.4\text{K}$ $E = 0$
- ▶ Heisenberg chain $J = 0.65\text{K}$
 $D = 6.15\text{K}$ $E = 0.7$
- ▶ TBA $H_{c1} = 3.8\text{T}$, $H_{c2} = 4.7\text{T}$

Spin-1 chains: thermal and magnetic properties



comparison

- ▶ HTE $J = 0.55\text{K}$ $D = 5.15\text{K}$ $E = 2.55$
- ▶ Heisenberg chain $J = 0.96\text{K}$
 $D = 5.0\text{K}$ $E = 2.8$
- ▶ TBA $H_{c1} = 1.0\text{T}$, $H_{c2} = 4.66\text{T}$

Conclusion and discussion

- ▶ **TBA & QTM** for spin- $\frac{1}{2}$ ladder model with strong rung coupling
- ▶ **TBA & QTM** for spin-1 chain with large single-anisotropy
- ▶ **thermal & magnetic properties** for real spin ladder and chain compounds

- ▶ **Spin- $\frac{1}{2}$ Ladders:**

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- ▶ **Spin-1 \otimes Spin- $\frac{1}{2}$ Ladders:**

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- ▶ **review on spin ladder models:**

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- ▶ **Spin-1 Chain:**

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- ▶ **Spin-orbital model:**

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▶ **Spin-Orbital Model**

- ▶ 2-leg ladder with different Lande factors $g_S \neq g_T$, S=Spin , T=orbital momentum
- ▶ via su(4) model

▶ **Mixed (Spin-1) \otimes (Spin- $\frac{1}{2}$) ladder**

- ▶ 2-leg ladder: one leg carries Spin-1 , the other Spin- $\frac{1}{2}$ operators
- ▶ via su(6) model
- ▶ in strong rung-coupling case: describes alternating legs case as well!

▶ **Integrable Spin- $\frac{3}{2}$ chain**

- ▶ chain carrying Spin- $\frac{3}{2}$ operators
- ▶ via su(4) model

▶ **(Spin-1) \otimes (Spin-1)**

- ▶ 2-leg ladder: both leg carrying Spin-1
- ▶ via su(9) model

▶ **many more models possible, have to find realization in nature yet...**

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■ See references therein, especially to

- ▶ Bethe Ansatz
- ▶ Functional relations, e.g. T-System
- ▶ Non Linear Integral Equations etc.

■ All experimental data taken from the referenced papers, see the above publications

▶ Spin-1 Chains

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▶ Spin- $\frac{1}{2}$ Ladders

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- ▶ **T-System, Q-System:**
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