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High-temperature series expansions with cyclic exchanges on a triangular lattice: Application to two-dimensional solid ^{3}He

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High-temperature series expansions for the specific heat and spin correlations are performed up to sixth order with a generalized Heisenberg Hamiltonian including the most important n-particle exchanges on a triangular lattice. These series provide a rigorous framework to describe the thermodynamic properties of ^{3}He films adsorbed on graphite where cyclic ring exchanges are expected to play an important role, as already observed in bulk solid ^{3}He. [S0163-1829(97)50530-3]

Low-temperature measurements of the susceptibility and specific heat on two solid layers of ^{3}He adsorbed on grafoil have revealed striking behavior. While the contribution to the susceptibility of the dense first layer is essentially of Curie type, the behavior of the second layer changes from antiferromagnetic to ferromagnetic as its density increases.[1,2] Different theoretical interpretations have been proposed. The experimental evidence that, just at solidification, the second layer is commensurate with the first layer with a density ratio 4/7 has provided a lot of excitement: a possible candidate for the commensurate phase has atoms B on potential maxima and atoms A on shoulders forming a *kagomé* lattice. The conjecture by Elser[3] that pair exchange between two A atoms could be stronger than between one A and one B atoms stimulated much work on the antiferromagnetic Heisenberg model for a *kagomé* lattice. Various models have tried to interpret the change from antiferromagnetic to ferromagnetic behavior at higher densities. Indirect exchange between the solid second layer and a fluid third layer[4,5] could play an important role in some density range. There is also a vast literature[1] about the influence of defects such as ''ground-state vacancies.''

A more unified view is that, just like in the three-dimensional solid,[6] cyclic n-particle exchanges play an important role. The principles of a general exchange Hamiltonian have been laid down a long time ago:[7,8]

$$\mathcal{H}_{ex}=-\sum_{P}(-1)^{p}J_{P}P, \tag{1}$$

where the sum is extended over all permutations P (with parity p) of the symmetric group acting on spin variables. Usually only pair transpositions are retained: this corresponds to the Heisenberg Hamiltonian. Thouless was the first to stress the importance of higher-order interactions, as cyclic three and four-particle exchange, in a hard core quantum solid. He also emphasised that even permutations like cyclic three-particle exchange generally lead to ferromagnetism while odd permutations like two or cyclic four-particle exchange favor antiferromagnetism (i.e., all J_P are positive).[8] Delrieu predicted the predominance of three-particle exchange leading to ferromagnetism in closed packed lattices like the three-dimensional (3D) hcp phase of solid ^{3}He or the 2D triangular lattice at high density.[9] However, at low densities, close to the commensurate phase, the interatomic distance (4.2 Å) is much larger than that corresponding to the melting of the 3D bcc phase (3.7 Å) and higher-order antiferromagnetic exchanges like cyclic four- and six-spin exchange should compete with ferromagnetic three-spin exchange, just like in the loose packed 3D bcc phase. The thermodynamic properties of the antiferromagnetic commensurate phase and the change to a ferromagnetic behavior at higher densities can be qualitatively interpreted within the frame of this multiple-spin exchange (MSE) model.[10] Recent *ab initio* Monte Carlo calculations of various exchange frequencies have corroborated this picture:[11] for an interatomic distance $a=3.84$ Å, the relevant processes are cyclic exchanges J_n involving the most symmetric rings of n nearest neighbors, with $n=3,2,4,6,5$ by decreasing amplitude (Fig. 1). The calculation applies to a monolayer, with the potential of graphite. We expect the same processes to be relevant for the second layer, although the weaker potential may affect the relative magnitude of the exchange constants.

We need reliable estimates of various thermodynamic quantities from a MSE model. Exact diagonalizations performed on 4×4 finite clusters[10,11] have provided interesting semiquantitative information. However, in the high-temperature range where accurate experimental data are now available, high-temperature series expansions (HTSE) are most suitable. Precise information on thermodynamic quantities for the Heisenberg model have recently been obtained through HTSE up to order 13 in $\beta=1/T$.[12] With a MSE Hamiltonian, the same degree of complexity is reached at order 6. This is, however, sufficient to compare with experimental data at $T\gtrsim 5$ mK.

We consider the following Hamiltonian:

$$\mathcal{H}_{ex}=J\sum^{(2)}\mathcal{P}_2+J_4\sum^{(4)}\mathcal{P}_4-J_5\sum^{(5)}\mathcal{P}_5+J_6\sum^{(6)}\mathcal{P}_6. \tag{2}$$

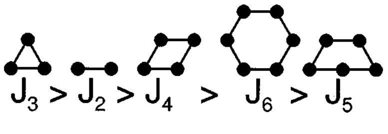

FIG. 1. Relevant exchange processes in 2D solid ^{3}He.

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TABLE I. Coefficients $a_{n,\lambda\mu\nu}$ of the HTSE for the logarithm of the partitions function $\ln(Z)$ and spin correlations $S(\mathbf{Q})$.

$n\ \lambda\mu\nu$	$\ln(Z)$	$S(0)=\chi(0)T$	$S(\mathbf{Q}_A)$	$n\ \lambda\mu\nu$	$\ln(Z)$	$S(0)=\chi(0)T$	$S(\mathbf{Q}_A)$
1 000		-6	3	1 100		-72	9
1 010		240	-48	1 001		-60	-6
2 000	9	48	12	2 100	180	1512	-72
2 200	1188	9696	-1104	2 010	-504	-6672	600
2 110	-7344	-80832	10896	2 020	13320	153024	-21792
2 001	72	2112	-120	2 101	864	27168	-3072
2 011	-3744	-98112	11616	2 002	2316	9216	864
3 000	-18	-408	-102	3 100	-1404	-21960	-5724
3 200	-18792	-355392	-51048	3 300	-68688	-1675584	-78768
3 010	7128	115344	16704	3 110	176256	3471552	250272
3 210	974592	23395392	24480	3 020	-346032	-7857792	-197280
3 120	-3865536	-102057408	3096864	3 030	4791456	139847424	-7054752
3 001	-1728	-50976	2232	3 101	-53568	-1422144	74016
3 201	-377568	-9212832	757440	3 011	215136	5783040	-330048
3 111	2904768	74336256	-6399360	3 021	-5188752	-146210976	13713264
3 002	-11664	-756720	55512	3 102	-186624	-10170144	986256
3 012	730080	39199104	-4006944	3 003	-37296	-2333664	494784
4 000	-306	3600	-864	4 100	-9936	261216	14688
4 200	-41616	7340736	1163424	4 300	538560	83930112	12480768
4 400	2368224	325475328	33894912	4 010	576	-1505664	-249984
4 110	-1080576	-81457920	-12469248	4 210	-22336128	-1314766848	-158057472
4 310	-96083712	-6444610560	-504608256	4 020	4542048	216124800	25013184
4 120	139631616	6556824576	550909440	4 220	837309312	46067851776	2269527168
4 030	-228183552	-10274293248	-536262144	4 130	-2622509568	-139462419456	-3380014080
4 040	2710582848	151195478016	646692096	4 001	23616	885504	76224
4 101	1104192	41720064	2873472	4 201	15269760	616104192	26640768
4 301	63226368	2817314304	44429568	4 011	-4867776	-197801088	-8023488
4 111	-132744960	-5661014016	-96526848	4 211	-821693952	-37991725056	271300608
4 021	281258496	12387512832	-10515072	4 121	3433895424	163496083968	-3676967424
4 031	-4523777280	-226619745792	7813627392	4 002	534816	36576576	-2045088
4 102	16265088	1006741248	-51985920	4 202	103700736	6712886016	-443600640
4 012	-69085440	-4089654528	218410752	4 112	-871925760	-54252519936	3658833408
4 022	1667052288	108405573888	-7685870208	4 003	3439872	323328000	-23077248
4 103	49641984	4331157504	-290834688	4 013	-142147584	-17138701824	1380599040
4 004	-64908768	955397376	-254661120				
5 000	3240	-42336	26496	5 100	339120	-3219360	2141472
5 200	9106560	-112170240	39054528	5 300	97107840	-2152942080	189421824
5 400	412571520	-19765470720	-525029376	5 500	438439680	-66680527872	-4380128256
5 010	-1553760	16334400	-6603936	5 110	-68653440	1258041600	-120170880
5 210	-939744000	36459152640	1384774272	5 310	-4359674880	429619399680	33024617472
5 410	-2906519040	1730736998400	141697625088	5 020	94095360	-3735598080	-170534784
5 120	1869569280	-206622263040	-17953163520	5 220	2785985280	-3449384305920	-302410773504
5 320	-61076459520	-17627712545280	-1377657781248	5 030	482993280	376263897600	34311121536
5 130	61679900160	11853558205440	924717871104	5 230	614452124160	86836590412800	5600092950528
5 040	-124997103360	-14548827409920	-894264929280	5 140	-1889588229120	-205354655646720	-9878319204864
5 050	1910432816640	186465394053120	6012647662080	5 001	150480	-9846720	-1870128
5 101	5060160	-765713280	-114536064	5 201	-1632960	-20218849920	-2660238720
5 301	-686131200	-218216102400	-24183489024	5 401	-2747900160	-819256465920	-73254035712
5 011	12222720	4423776000	472710912	5 111	1388920320	217639011840	20943239424
5 211	25752211200	3342177016320	266680068096	5 311	115062474240	16058898063360	1001943567360
5 021	-5299692480	-547060506240	-38561776320	5 121	-163886538240	-16178221570560	-897789933312
5 221	-1049886489600	-113120583390720	-4595844858624	5 031	283863997440	24949573570560	904147562496
5 131	3540882908160	340694663270400	7911565406208	5 041	-4046842137600	-371637535503360	-3452335744512
5 002	-16240320	-1156707840	-50605536	5 102	-668695680	-51494288640	-1200656832
5 202	-9547096320	-734823536640	-10504106880	5 302	-43131939840	-3349026762240	-24089767680
5 012	2857841280	231975782400	2283173760	5 112	81077483520	6529659801600	7198436352
5 212	541403066880	44139361536000	-260946639360	5 022	-172812625920	-14119809273600	122687626368
5 122	-2268367395840	-188958837312000	2998347068160	5 032	3126156503040	263675667901440	-6231467586048
5 003	-235771200	-25216573440	566975616	5 103	-5718343680	-696964331520	24234610176
5 203	-34190461440	-4721349020160	189169603584	5 013	22810233600	2871982840320	-114107132160
5 113	281034293760	38622029783040	-1728215382528	5 023	-609212033280	-78044062494720	3626813356800
5 004	-2246791680	-174376646400	5945703936	5 104	-23692631040	-2332451366400	87042324480
5 014	82690122240	9284983910400	-350952437760	5 005	-3091868160	-467857036800	-30567638016
6 000	49176	781728		6 100	1103328	67023936	
6 200	-77076576	2218227840		6 300	-2681399808	44007459840	
6 400	-31074475392	594183329280		6 500	-149858864640	4547055495168	
6 600	-232143856128	13824881688576		6 001	-12631680	58785408	
6 101	-850566528	7443283968		6 201	-21595092480	348452278272	
6 301	-247094641152	7244092339200		6 401	-1256881397760	67185011718144	

TABLE I. (*Continued*).

$n\ \lambda\mu\nu$	ln(*Z*)	$S(0)=\chi(0)T$	$S(\mathbf{Q}_A)$	$n\ \lambda\mu\nu$	ln(*Z*)	$S(0)=\chi(0)T$	$S(\mathbf{Q}_A)$
6 501	-2176839014400	226858379968512		6 002	-65223360	21507604992	
6 102	-434363904	1505352259584		6 202	46375621632	36920708241408	
6 302	843290293248	380469864600576		6 402	3805549876224	1394947834269696	
6 003	8764084224	1261127904768		6 103	381286503936	55117668722688	
6 203	5534907061248	780261401991168		6 303	26047428212736	3581254619037696	
6 004	138704085504	18515614371840		6 104	3556631503872	516562257272832	
6 204	25276398243840	3516948906190848		6 005	901594312704	113580991346688	
6 105	11416926302208	1543767870210048		6 006	15547926117888	222996679464960	

Since, for spins 1/2, three-particle exchange can be expressed as pair exchange,[8] both processes are incorporated in an effective pair exchange constant: $J=J_2-2J_3$ (first sum), the other sums are extended to cyclic 4, 5, and 6 permutation operators (Fig. 1) acting on spin variables. Note that, if interactions between the solid second layer and the fluid third layer play a role in some density range, the dominant part of them can be reduced to an effective Heisenberg interaction J_{RKKY} between first neighbors in the second layer.[4,5] They are easily incorporated in our Hamiltonian with $J=J_2-2J_3-J_{RKKY}$.

Using the cluster method,[13,14] we have generated HTSE for the free energy $F=-T\ln Z$ $(Z=\mathrm{Tr}\{\exp[-\beta\mathcal{H}_{ex}]\})$ and spin correlation functions: $S(\mathbf{Q})=\Sigma_j\langle\sigma_i^z\sigma_j^z\rangle\exp[i\mathbf{Q}\cdot(\mathbf{R}_i-\mathbf{R}_j)]$ for $\mathbf{Q}=0$ $[S(0)=T\chi(0)$ provides the zero-field susceptibility] and $\mathbf{Q}=\mathbf{Q}_A$, corresponding to the classical antiferromagnetic ground state for the Heisenberg model (three sublattices with spins lying at 120° to each other). The results are expressed in Table I. For each quantity A, we define coefficients $a_{n,\lambda\mu\nu}$ by

$$\frac{A}{N}=\sum a_{n,\lambda\mu\nu}\left[\frac{J}{2}\right]^{n-\lambda-\mu-\nu}\left[\frac{J_4}{8}\right]^{\lambda}\left[\frac{J_5}{16}\right]^{\mu}\left[\frac{J_6}{32}\right]^{\nu}\frac{\beta^n}{n!}.$$

The general expansion is performed up to order $n=5$. This involves diagrams including up to 25 nodes with about 10^9 embeddings on the lattice [same order of magnitude as for the Heisenberg model at order 13 (Ref. 12)]. The expansion has been carried further to 6th order with $J_5=0$. The calculation took three weeks of CPU time on a Sun Ultra2 workstation. We used ''the brute force,'' calculating the traces for each geometry (classification of diagrams according to their topologies is here a formidable task that we avoided).

The series are analyzed by Padé approximants.[13] A $[L,M]$ Padé approximant to the series $A(\beta)$ is a rational fraction P_M/Q_L, with P and Q, polynomials of degree L and M in β, satisfying: $A(\beta)=P_M/Q_L+O(\beta^{L+M+1})$. It is first essential to test the convergence of Padé approximants. Figure 2 represents Padé approximants to the specific heat C_v and ferromagnetic correlation function $T\chi(0)$ with $J_5=J_6=0$ and various ratios J_4/J (since J is dominated by three-spin exchange, J_4/J is negative). The temperature is scaled with J_c that represents the leading term in the specific heat: $C_v/N=9(J_c\beta)^2/4$. We have observed a better behavior of the near diagonal Padé approximant after a Euler transformation $\beta=\beta'/(1-\beta')$. The [2,3] and [3,3] Padé approximants provide an excellent description of the specific heat down to $T/J_c\approx 2.5$. The [3,2] and [4,2] Padé approximants to $T\chi(0)$ are reliable down to $T/J_c\approx 2$. Figure 3 represents isotherms of C_v at $T/J_c=3$ as a function of J_4/J. The first isotherm is for $J_6=J_5=0$, the second curve for $J_5=0$ and $J_6=J_4$. Various Padé approximants are compared with the results of exact diagonalizations on a finite 4×4 cluster with periodic boundary conditions.[10] The finite-size results give a poor estimate of the thermodynamic limit for small J_4. Surprisingly, it improves for larger values. Both isotherms show

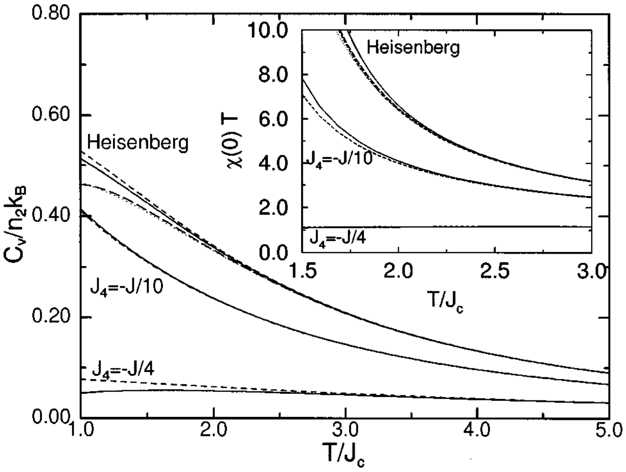

FIG. 2. Padé approximants to the specific heat for $J_5=0$, $J_6=0$, after performing the Euler transformation to the variable $\beta'=\beta/(1+\beta)$: [3,3] (solid line), [2,3] (short dashed), [4,4] (dotted), [6,6] (long dashed). The inset shows Padé approximants to $\chi(0)T$: [6,6] (long dashed), [4,2] (full line), [3,2] (short dashed).

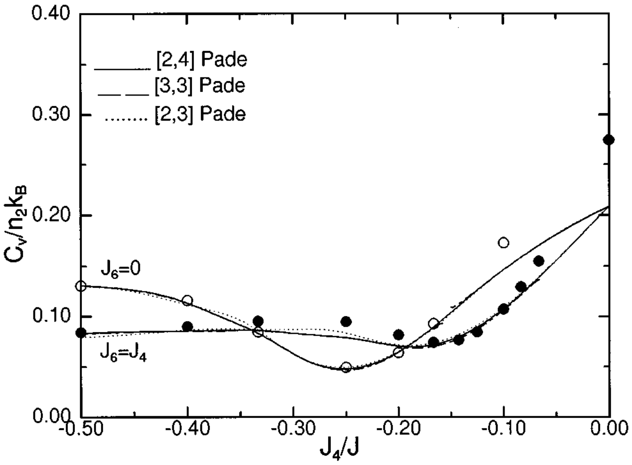

FIG. 3. Specific-heat isotherms at $T/J_c=3$ for $J_5=0$. The exact results for a 4×4 finite cluster (circles) are compared to the thermodynamic limit obtained through HTSE.

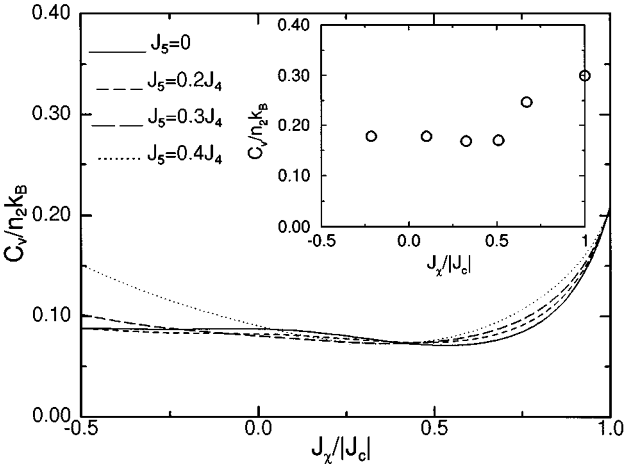

FIG. 4. [2,3] Padé approximant to the specific heat at $T/J_c=3$ for $J_6=J_4$ and various values of J_5/J_4, as a function of $J_\chi/|J_c|$. The inset compares the experimental isotherm at $T/J_c=1$ (Ref. 2).

a minimum at some J_4/J, corresponding to the "highest frustration." Six-spin exchange J_6 raises this minimum, leading to an essentially flat curve in a large domain: $-0.5<J_4/J\lesssim-0.12$ and an abrupt increase for $-0.12\lesssim J_4/J<0$. The influence of five-spin exchange is analyzed in Fig. 4. Four isotherms at $T/J_c=3$ with $J_6=J_4$ and $J_5/J_4=0,0.2,0.3,0.4$ are plotted as a function of $\phi=J_\chi/J_c$, where $J_\chi=\theta/3=-J-3J_4+5J_5-5J_6/8$ (θ is the Curie-Weiss constant). For $J_5/J_4\lesssim0.3$ the influence of J_5 is small. It is more important for $J_5/J_4\approx1/2$ for the following reason: five-spin exchange can be recast in terms of pair and four-spin exchanges (just like three-spin exchange is expressed in terms of pair exchange),[8] this leads to an effective four-spin exchange interaction between first neighbors $K=J_4-2J_5$ plus other four-spin interactions between atoms that are not all first neighbors, and K cancels precisely at $J_5/J_4=1/2$. The inset shows the specific-heat isotherm at $T/J_c=1$ observed experimentally.[2] There is a remarkable similarity to the theoretical curves at $T/J_c=3$: a very flat region for $-0.5<\phi\lesssim0.6$ and an abrupt rise for $0.6\lesssim\phi<1$. The parameter ratios $J_6/J_4\approx1$ and $J_5/J_4\lesssim0.4$ are consistent with Monte Carlo calculations.[11]

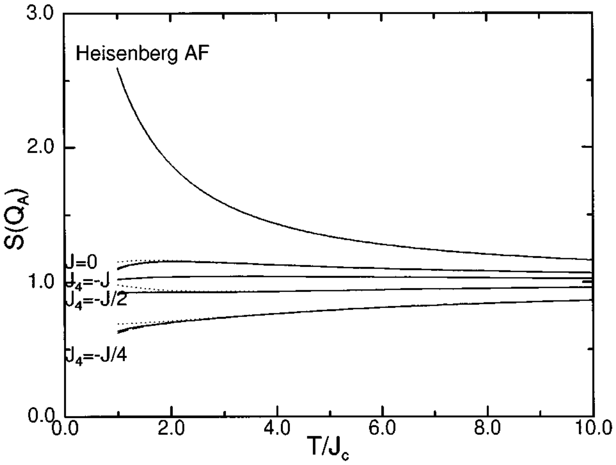

FIG. 5. Antiferromagnetic correlations $S(\mathbf{Q}_A)$ corresponding to a three-sublattice ground state for $J_5=J_6=0$ and $J_4/J=-0.25$, $-0.5,-1,-\infty$. Padé approximant [2,3] (solid line), [3,2] (long dashed), and [2,2] (dotted). For comparison, the upper curve represents $S(\mathbf{Q}_A)$ for the antiferromagnetic Heisenberg model with $J<0$ (Ref. 12).

At $T=0$, the ground state of the Heisenberg model with $J<0$ is ferromagnetic. The ferromagnetic state becomes unstable with respect to a spin-wave state if $B=-J-4J_4+10J_5-2J_6$ is negative (note that $B\neq J_\chi$). For $B<0$, the nature of the ground state at $T=0$ is a puzzling question. In Fig. 5 the antiferromagnetic correlation function $S(\mathbf{Q}_A)$, $\mathbf{Q}_A$, corresponding to the classical antiferromagnetic ground state for the antiferromagnetic Heisenberg model $(J>0)$,[12] is compared to $S(\mathbf{Q}_A)$ for MSE models with $J_5=J_6=0$ and various J_4/J. There is no evidence for an increase of this correlation function, even with a Hamiltonian containing only four-spin exchange $(J=0)$. If there is an ordered state, it should be more complicated.

We have provided a rigorous framework for the analysis of thermodynamic data on 2D solid ^{3}He, which could contribute to a clear answer concerning competing theoretical interpretations. We encourage further *ab initio* Monte Carlo calculations of exchange frequencies and experimental work on this system.

I am indebted to J. Saunders for his suggestion to carry out this calculation and for stimulating discussions and correspondences. I thank B. Bernu, J. M. Delrieu, and H. Godfrin for valuable discussions.

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