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ЛАБОРАТОРИЯ ТЕОРЕТИЧЕСКОЙ ФИЗИКИ

EFFECT OF CRYSTAL-FIELD ANISOTROPY ON CURIE TEMPERATURE OF HEISENBERG FERROMAGNET. HTS EXPANSION METHOD

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EFFECT OF CRYSTAL-FIELD ANISOTROPY ON CURIE TEMPERATURE OF HEISENBERG FERROMAGNET. HTS EXPANSION METHOD

I. Introduction

Recently, the effect of crystal field anisotropy on the Curie temperature (T_c) of a Heisenberg ferromagnet has been extensively studied in the approximations both of the molecular-field theory (MFT) [/1/] and of different Green-function decoupling schemes [/2,3,4,5/]. The discrepancy in the results, obtained in the MFA on the one hand and with the aid of Green-function technique on the other, is especially great in the evaluation of the sensitivity of T_c to the anisotropy parameter D. In the case of large crystal field anisotropy with easy Z-axis, the best Green function theory prediction [/5/] for the asymptotic value of $T_c(D)$ as $D \to \infty$ is the same as the MFT result, though it seems more likely in this case the Curie temperature to approach the Ising model one, than the too high MFT value (see [/4/]).

As, in the absence of exact results, the most accurate estimations of T_c are available from the exact high temperature series expansion (HTSE) for zero-field susceptibility, it seems of interest to analyze the problem by this method.

It should be noted, that HTSE method has been applied in recent investigations of the critical behaviour of generalized Ising model, including a crystal-field anisotropy term of the form[6,7]:

$$\mathcal{H}_{cf} = -\frac{D}{S^2}\sum_i (S_i^z)^2. \quad (1)$$

Although fewer terms of the zero-field susceptibility series have been derived, than for the pure Ising model, the estimates of $T_c(D)$ are fairly confident. The sensitivity of the extrapolated Curie temperature to D was found to be very close to the MFT one (see[7]).

In the present discission we retain the form (1) for H_{cf} but assume Heisenberg (anisotropic) exchange interaction, which, in contrast to the Ising one, does not commute with the so-chosen H_{cf} operator.

We restrict ourselves to derivation of HTSE for the longitudinal component $\chi^0_{\parallel}$ of the initial susceptibility tensor χ^0_{rr}, and the case of positive D is considered only. In the case $D<0$, the other components of χ^0_{rr} should be taken in consideration too (see[1]).

II. HTSE for the Zero-Field Susceptibility

The Hamiltonian for the system is:

$$\mathcal{H} = \mathcal{H}_z + \mathcal{H}_{ex} + \mathcal{H}_{cf}, \quad (2)$$

where

$$\mathcal{H}_z = -\frac{\mu}{S} H \sum_i S_i^z ,$$

$$\mathcal{H}_{ex} = -\frac{1}{S^2} \sum_{ij} J_{ij} [S_i^z S_j^z + \frac{1}{2} g(S_i^- S_j^+ + S_i^+ S_j^-)],$$

and H_{cf} is given by (1).

The exchange interaction J_{ij} we assume restricted to the nearest neighbours. In the sum $\sum_{ij}$, i and j run over all spins in the lattice.

The longitudinal component $\chi_{||}^0$ of the initial susceptibility is given by:

$$\frac{kTS^2}{\mu^2} \chi_{||}^0 = \frac{1}{N} \sum_{ij} \frac{Tr\{S_i^z S_j^z \; exp(-\beta\mathcal{H}_{ex} - \beta\mathcal{H}_{cf})\}}{Tr\{exp(-\beta\mathcal{H}_{ex} - \beta\mathcal{H}_{cf})\}} , \qquad (3)$$

where $\beta = \frac{1}{kT}$.

As we place no restrictions on the relative magnitude of D and $kT = \frac{1}{\beta}$, the expansion for $\chi_{||}^0$ will be carried out in powers of βH_{ex} only. Since H_{cf} and H_{ex} do not commute, the following expression is used:

$$exp(-\beta\mathcal{H}_{ex} - \beta\mathcal{H}_{cf}) =$$

$$= \{1 + \sum_{n=1}^{\infty} (-\beta)^n \int_0^1 d\tau_1 \dots \int_0^{\tau_{n-1}} d\tau_n \mathcal{H}_{ex}(\tau_n) \dots \mathcal{H}_{ex}(\tau_1) exp(-\beta\mathcal{H}_{cf})\}, \qquad (4)$$

where

$$\mathcal{H}_{ex}(\tau) = exp(-\beta\mathcal{H}_{cf}\tau)\mathcal{H}_{ex} exp(\beta\mathcal{H}_{cf}\tau).$$

By substitution of (4) into (3) we obtain susceptibility expansion of the form:

$$\frac{S^2}{\beta\mu^2}\chi_{||}^0 = \sum_{n=0}^{\infty} a_n (\beta D)\left(\frac{\beta J}{S^2}\right)^n. \qquad (5)$$

The first two terms in the series (5) coincide with the corresponding ones of the case of Ising exchange interaction [7]. We have calculated in addition a_2, a_3 and a_4 for all cubic lattices, general spin value and exchange anisotropy, described by parameter g . The explicit expressions for these terms are given in the Appendix.

At D=0, we have checked our coefficients by comparing with those given by Dalton and Rimmer [8] for the anisotropic Heisenberg model.

Our method of analysis of the susceptibility functional series (5) and the estimation of the Curie point has been discussed in [7], which paper we refer to for details. We remind here, that d'Alambert criterion is used for estimation of the singularity point of the series (5), and the successive approximations to the Curie temperature ($T_c^{(n)}$) are calculated from the equation:

$$\frac{a_n\,(D/kT_c^{(n)})}{S^2 a_{n-1}(D/kT_c^{(n)})} = \frac{kT_c^{(n)}}{J}. \qquad (6)$$

For n=1 equation (6) yields the MFT result, i.e. $T_c^{(1)}=T_c^{MFA}$ (see [4,5]).

III. Results and Discussion

Here results for $T_c(D)$ are given in the case of isotropic exchange interaction (g =1).

Evidently, the number of terms at our disposal is insufficient for decisive conclusions to be drawn, especially about the critical exponent γ . Nevertheless, as far as $T_c(D)$ is concerned, we think that our estimates can provide some useful quantitative information about the effect of the crystal-field term (1) on the Curie temperature.

The accuracy of extrapolation is illustrated by Table I, where our estimates of $\theta_c = \frac{kT_c}{2zJ}$ at $D/J=0$ are compared with the corresponding figures θ_c^{RW} obtained from the formula given by Rushbrooke and Wood/9/ as an approximation to the Curie point of pure Heisenberg ferromagnet.

Table 1

		fcc	bcc	sc
S = 1	$\theta_c(0)$	0.501	0.480	0.421
	θ_c^{RW}	0.501	0.478	0.456
$S = \frac{3}{2}$	$\theta_c(0)$	0.434	0.419	0.375
	θ_c^{RW}	0.427	0.408	0.388

The situation for $D/J > 0$ can be visualized on Fig.1, where our successive approximations $\theta_c^{(n)}$ are plotted against $1/n$ for a few D/J and three cubic lattices (S=3/2). The extrapolated values θ_c^{ex} are shown on the kT_C axis. Since the convergence of the series does not change strongly, we may expect for $D/J > 0$ not much worse accuracy than at $D/J = 0$.

Further we concentrate our attention on the f.c.c. lattices, for which convergence is most rapid.

In Table 2 the values of $\theta_c(D/J)$ and the extrapolated values $\theta_c^{ex}(D/J)$ are given for S=1, 3/2, f.c.c. lattice and several D/J .

Fig.2 contains plots of $\theta_c^{(n)}$ in the cases of Heisenberg (solid line) and Ising (broken line) exchange interaction for S=1 and some values of the ratio D/J.

In Fig.3 the estimated Curie temperature as a function of D/J is plotted (H) in comparison with the generalized Ising model *(I)* and the molecular-field approximation (MFA) results (S=3/2).

There is one notable feature apparent on Figs.2 and 3, namely, the asymptotic $(D/J \to \infty)$ coincidence of the Ising and Heisenberg model Curie temperature with the S=1/2 Ising model one /10/. Indeed, for $D/J \gg 1$, only the doublet with $S_z = \pm S$ is expected to be appreciably populated at $T \sim T_C$ and effectively we have a spin 1/2 Ising system. This statement can readily be proved by taking the limit $D \to \infty$ in the coefficients $a_n(D/kT)$, given in the Appendix.

The sensitivity of the Curie temperature to the ratio D/J is illustrated in Fig.4, where the curves for the generalized Ising model (I) and the MFA result are plotted too.

In Fig.5 the curve $T_c(D/J)/T_c(0)$ obtained from our series (f) is compared with the results of different Green-function decoupling schemes, all given for b.c.c. lattice and S=1. The other curves represent respectively: (a) Narath's result/2/; (b) Anderson and Callen's result/3/; (c) Lines's result/4/; (d) Devlin's result/5/; (e) MFT result.

It is evident that our curve, although showing stronger dependence of $T_c(D/J)$ on D/J than the MFT one, is in sharp contrast with the results of the Green-function decoupling schemes. This is not too surprising since the only one of the latter, predicting finite value for $T_c(D/J)$ as $D/J \to \infty$, namely Devlin's result (d), gives $\theta_c = 1$ at $D/J = \infty$, i.e. the MFT result (see Fig.3).

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References

1. G.J.Bowden, D.St.P.Bunbury, M.A.H.McCausland. J.Phys.C., Solid St.Phys., 4, 1840 (1971).

2. A.Narath. Phys.Rev., 140, A854 (1965).
3. F.B.Anderson, H.B.Callen. Phys.Rev., 136, A1068 (1964).
4. M.E.Lines. Phys.Rev., 156, 534 (1967).
5. J.F.Devlin. Phys.Rev., B4, 136 (1971).
6. J.Oitmaa. J.Phys.C., Solid St.Phys., 5, 435 (1972).
7. J.G.Brankov, J.Przystawa, E.Praveczki. JINR, E4-6238, Dubna, 1971.
8. N.W.Dalton, D.E.Rimmer. Phys.Lett., 29A,611 (1969).
9. G.S.Rushbrooke, P.J.Wood. Mol.Phys., 1, 257 (1958).
10. C.Domb, M.F.Sykes. Proc.Roy.Soc., A240, 214 (1957).

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Appendix

Expressions for the susceptibility coefficients $a_n(\beta\mathcal{D})$

$$a_n(\beta\mathcal{D}) = a_n^{I} + a_n^{XY}$$

a_n^{I} correspond to the case of Ising exchange interaction /7/ (for completeness they are reproduced, too), and a_n^{XY} to the case of XY model.

$$a_0^{I} = m_2 \qquad a_0^{XY} = 0$$

$$a_1^{I} = 2z m_2^2 \qquad a_1^{XY} = 0$$

$$a_2^{I} = 4z^2 m_2^3 \left[1 - \frac{1}{2z}\left(3 - \frac{m_4}{m_2^2}\right)\right]$$

$$a_2^{XY} = 2z^2 g\left[-m_2 V_2(0,0) + V_2(2,0) + V_2(1,1)\right]$$

$$a_3^{I} = 8z^3 m_2^4\left\{1 - \frac{1}{z}\left(3 - \frac{m_4}{m_2^2}\right) + \frac{1}{2z^2}\left(3 - \frac{m_4}{m_2^2}\right)\left[\frac{1}{3}\left(3 - \frac{m_4}{m_2^2}\right) - x\right]\right\}$$

$$a_3^{XY} = 8z^2 m_2 g^2\left[-m_2 V_2(0,0) + V_2(2,0) + V_2(1,1)\right] +$$

$$+4zg^2\left[m_2^2 V_2(0,0) - 2m_2 V_2(2,0) - 3m_2 V_2(1,1) + V_2(3,1) + V_2(2,2) +\right.$$

$$\left.+m_2 V_3(0,0) - 2m_2 V_3(1,0) - V_3(2,0) - V_3(1,1) + V_3(3,0) + V_3(2,1)\right] +$$

$$+2zxg^3\left[-3m_2 T_3(0,0,0) + T_3(\{2,0,0\}) + 2T_3(\{1,1,0\})\right]$$

$$a_4^{I} = 16z^4 m_2^5\left\{1 - \frac{3}{2z}\left(3 - \frac{m_4}{m_2^2}\right) + \frac{1}{4z^2}\left[24 - \frac{35}{2}\frac{m_4}{m_2^2} + \frac{7}{3}\frac{m_4^2}{m_2^4} + \frac{1}{2}\frac{m_6}{m_2^3} -\right.\right.$$

$$\left.\left.-4x\left(3 - \frac{m_4}{m_2^2}\right)\right] + \frac{1}{8z^3}\left(3 - \frac{m_4}{m_2^2}\right)\left[-6 + 5\frac{m_4}{m_2^2} - \frac{1}{3}\frac{m_6}{m_2^3} + 4x\left(3 - \frac{m_4}{m_2^2}\right) - 4y\right]\right\}$$

$$a_4^{XY} = 24 m_2^2 z^3 g^2\left[-m_2 V_2(0,0) + V_2(2,0) + V_2(1,1)\right] +$$

$$+2z^2 g^2\left\{(28m_2^3 - 4m_4 m_2)V_2(0,0) - 36m_2^2 V_2(1,1) -\right.$$

$$-(38m_2^2 - 2m_4)V_2(2,0) + 2m_2\left[V_2(4,0) + 5V_2(2,2) + 6V_2(1,3)\right] +$$

$$+8m_2^2 V_3(0,0) + 4(m_2^2 - m_4)V_3(1,0) - 8m_2\left[V_3(1,1) + V_3(2,0)\right] +$$

$$+4m_2\left[V_3(2,1) + V_3(3,0)\right] - (20m_2^2 - 4m_4)V_4(0,0) + 8m_2\left[V_4(1,1) + V_4(2,0)\right] +$$

$$+g^2\Big[4V_2(0,0)\big(m_2V_2(0,0)-V_2(2,0)-V_2(1,1)\big)-3m_2G(0,0,0)+$$

$$+G(\{2,0,0\})+2G(\{1,1,0\})\Big]\Big\}+$$

$$+8m_2z^2xg^3\big[-3m_2T_3(0,0,0)+T_3(\{2,0,0\})+2T_3(\{1,1,0\})\big]+$$

$$+2zg^2\Big\{(-12m_2^3+2m_4m_2)V_2(0,0)+(24m_2^2-2m_4)V_2(2,0)+$$

$$+18m_2^2V_2(1,1)-2m_2\big[V_2(4,0)+6V_2(2,2)+6V_2(3,1)\big]+$$

$$+2\big[V_2(3,3)+V_2(4,2)\big]-4m_2^2V_3(0,0)-(12m_2^2-4m_4)V_3(1,0)+m_2\big[8V_3(2,0)+$$

$$+12V_3(1,1)\big]-4m_2\big[V_3(3,0)+3V_3(2,1)\big]-4\big[V_3(2,2)+V_3(3,1)+4\big[V_3(4,1)+$$

$$+V_3(3,2)\big]+(20m_2^2-4m_4-4m_2)V_4(0,0)+16m_2V_4(1,0)-16m_2V_4(2,0)-$$

$$-8\big[V_4(3,0)+V_4(2,1)\big]+4\big[V_4(2,0)+V_4(1,1)+V_4(4,0)-V_4(2,2)\big]+$$

$$+g^2\Big[2V_2(0,0)\big(-m_2V_2(0,0)+V_2(2,0)+V_2(1,1)\big)-m_2H(0,0)+H(2,0)+$$

$$+H(1,1)+3m_2G(0,0,0)-G(\{2,0,0\})-2G(\{1,1,0\})\Big]\Big\}+$$

$$+4zxg^2\Big\{2m_2^3V_2(0,0)-(9m_2^2-m_4)V_2(1,1)-4m_2^2V_2(2,0)+$$

$$+2m_2\big[V_2(2,2)+V_2(3,1)\big]-(10m_2^2-2m_4)V_3(1,0)+2m_2\big[V_3(3,0)+V_3(2,1)\big]+$$

$$+(10m_2^2-2m_4)V_4(0,0)-4m_2\big[V_4(2,0)+V_4(1,1)\big]+g\big[6m_2^2T_3(0,0,0)-$$

$$-11m_2T_3(\{1,1,0\})-4m_2T_3(\{2,0,0\})+T_3(\{3,1,0\})+2T_3(\{2,2,0\})+$$

$$+5T_3(\{2,1,1\})+6m_2T_4(0,0,0)-6m_2T_4([1,0],0)-2T_4(\{2,0,0\})-$$

$$-4T_4(\{1,1,0\})+2T_4([3,0],0)+2\big[T_4([2,1],0)+T_4([1,0],2)\big]+4T_4([2,0],1)\Big\}+$$

$$+2zyg^4\Big\{-4m_2K(0,0,0,0)+K(\{2,0,0,0\})+2K(\{1,1,0,0\})\Big\},$$

where:

	s.c.	b.c.c.	f.c.c.
x	0	0	4
y	4	12	22
z	6	8	12

$$m_n=\langle S_z^n\rangle_0\ .$$

We denote: $\int_0^1 d\tau_1 \int_0^{\tau_1} d\tau_2 \ldots \int_0^{\tau_{n-1}} d\tau_n \, F(\tau_2-\tau_1,\ldots,\tau_n-\tau_1) \equiv \hat{I}_n F(\tau_2,\ldots,\tau_n)$

then:

$$V_n(k,\ell) = \hat{I}_n \langle S^+(\tau_2) S^- S_z^k \rangle_0 \langle S^-(\tau_2) S^+ S_z^\ell \rangle_0 \qquad (n = 2,3,4)$$

$$T_{3,4}(k,\ell,q) = \hat{I}_{3,4} \langle S^+(\tau_{3,4}) S^- S_z^k \rangle_0 \langle S^-(\tau_{3,4}-\tau_2) S^+ S_z^\ell \rangle_0 \langle S^-(\tau_2) S^+ S_z^q \rangle_0$$

$$\begin{aligned} H(k,\ell) = \hat{I}_4 \{ & \langle S^+(\tau_4) S^+(\tau_3) S^-(\tau_2) S^- S_z^k \rangle_0 \langle S^-(\tau_4) S^-(\tau_3) S^+(\tau_2) S^+ S_z^\ell \rangle_0 + \\ & + \langle S^+(\tau_4) S^-(\tau_3) S^+(\tau_2) S^- S_z^k \rangle_0 \langle S^-(\tau_4) S^+(\tau_3) S^-(\tau_2) S^+ S_z^\ell \rangle_0 + \\ & + \langle S^+(\tau_4) S^-(\tau_3) S^-(\tau_2) S^+ S_z^k \rangle_0 \langle S^-(\tau_4) S^+(\tau_3) S^+(\tau_2) S^- S_z^\ell \rangle_0 \} \end{aligned}$$

$$\begin{aligned} G(k,\ell,q) = \hat{I}_4 \{ & \langle S^-(\tau_4) S^-(\tau_3) S^+(\tau_2) S^+ S_z^q \rangle_0 \left[\langle S^+(\tau_4-\tau_2) S^- S_z^k \rangle_0 \langle S^+(\tau_3) S^- S_z^\ell \rangle_0 + \right. \\ & \left. + \langle S^+(\tau_4) S^- S_z^k \rangle_0 \langle S^+(\tau_3-\tau_2) S^- S_z^\ell \rangle_0 \right] + \\ & + \langle S^-(\tau_4) S^+(\tau_3) S^-(\tau_2) S^+ S_z^q \rangle_0 \left[\langle S^+(\tau_4-\tau_2) S^- S_z^k \rangle_0 \langle S^+(\tau_2) S^- S_z^\ell \rangle_0 + \right. \\ & \left. + \langle S^+(\tau_4) S^- S_z^k \rangle_0 \langle S^-(\tau_3-\tau_2) S^+ S_z^\ell \rangle_0 \right] + \\ & + \langle S^-(\tau_4) S^+(\tau_3) S^+(\tau_2) S^- S_z^q \rangle_0 \left[\langle S^+(\tau_4-\tau_2) S^- S_z^k \rangle_0 \langle S^-(\tau_3) S^+ S_z^\ell \rangle_0 + \right. \\ & \left. + \langle S^+(\tau_4-\tau_3) S^- S_z^k \rangle_0 \langle S^-(\tau_2) S^+ S_z^\ell \rangle_0 \right] \} \end{aligned}$$

$$\begin{aligned} K(k,\ell,r,q) = & \\ = \hat{I}_4 \{ & \langle S^+(\tau_4-\tau_3) S^- S_z^k \rangle_0 \langle S^+(\tau_3-\tau_2) S^- S_z^\ell \rangle_0 \langle S^+(\tau_2) S^- S_z^r \rangle_0 \langle S^-(\tau_4) S^+ S_z^q \rangle_0 + \\ & + \langle S^+(\tau_4-\tau_3) S^- S_z^k \rangle_0 \langle S^+(\tau_3) S^- S_z^\ell \rangle_0 \langle S^-(\tau_2) S^+ S_z^r \rangle_0 \langle S^-(\tau_4-\tau_2) S^+ S_z^q \rangle_0 + \\ & + \langle S^+(\tau_4-\tau_2) S^- S_z^k \rangle_0 \langle S^+(\tau_3) S^- S_z^\ell \rangle_0 \langle S^-(\tau_3-\tau_2) S^+ S_z^r \rangle_0 \langle S^-(\tau_4) S^+ S_z^q \rangle_0 \} \end{aligned}$$

$$F(\{k_1,\ldots k_n\}) \equiv \sum_{\left\{ \substack{\text{all different} \\ \text{orders of } k_1,\ldots,k_n} \right\}} F(k_1,\ldots k_n)$$

$$T([k,\ell],q) \equiv T(k,\ell,q) - T(\ell,k,q)$$

$$\langle A \rangle_0 = \mathrm{Tr}\{A \exp(\beta D S_z^2/S^2)\} / \mathrm{Tr}\{\exp(\beta D S_z^2/S^2)\}$$

Table 2.

S	D/J	0	1	2	5	10	20	50	100
S=1	θ_c^{MFA}	0.667	0.680	0.693	0.727	0.774	0.843	0.947	0.993
	$\theta_c^{(2)}$	0.590	0.605	0.618	0.654	0.702	0.770	0.869	0.908
	$\theta_c^{(3)}$	0.559	0.575	0.589	0.626	0.675	0.744	0.840	0.875
	$\theta_c^{(4)}$	0.543	0.560	0.574	0.613	0.663	0.732	0.825	0.860
	θ_c^{ex}	0.501	0.518	0.534	0.574	0.625	0.694	0.784	0.814
S=3/2	θ_c^{MFA}	0.556	0.570	0.584	0.621	0.675	0.757	0.899	0.980
	$\theta_c^{(2)}$	0.500	0.515	0.530	0.568	0.622	0.703	0.835	0.902
	$\theta_c^{(3)}$	0.477	0.493	0.508	0.548	0.603	0.683	0.810	0.871
	$\theta_c^{(4)}$	0.465	0.482	0.498	0.538	0.593	0.673	0.797	0.855
	θ_c^{ex}	0.434	0.452	0.468	0.510	0.565	0.644	0.762	0.813

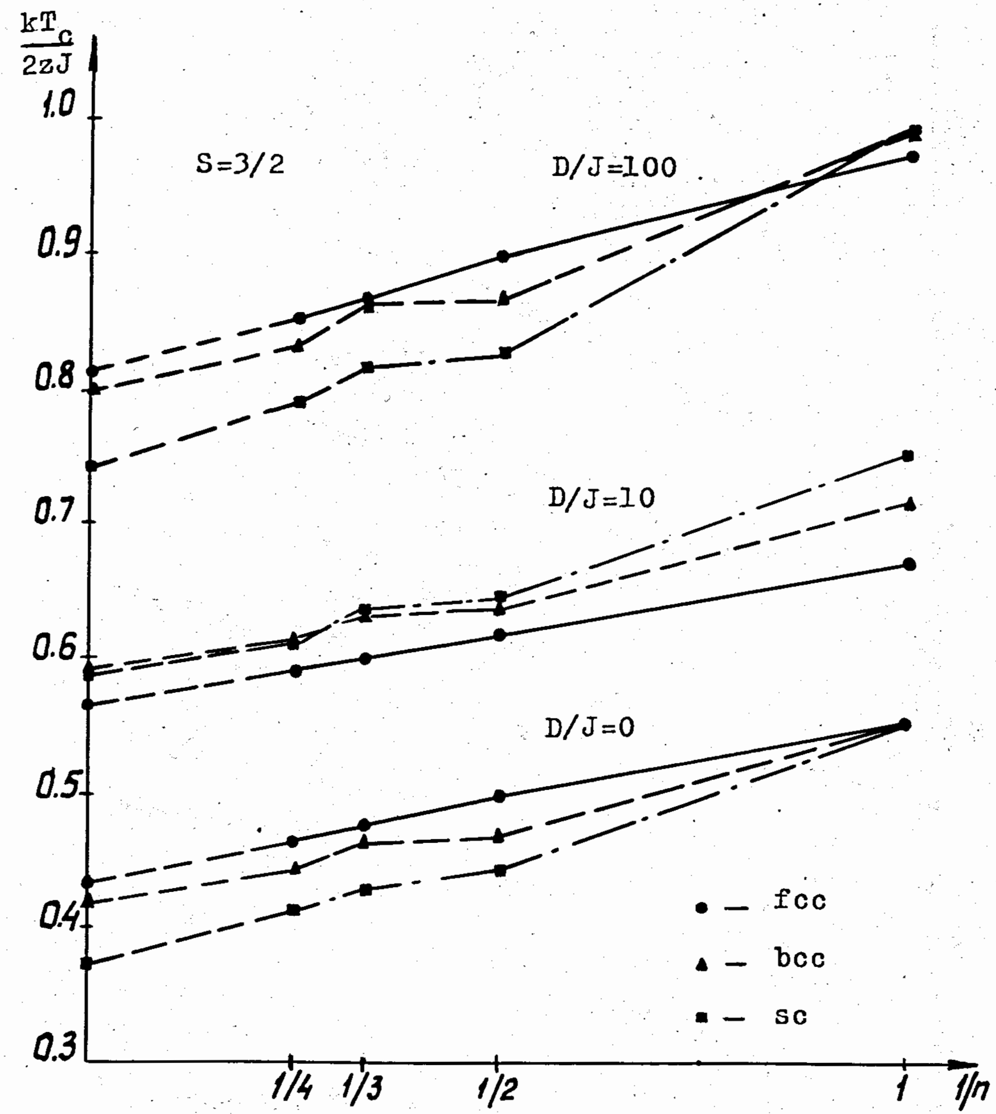
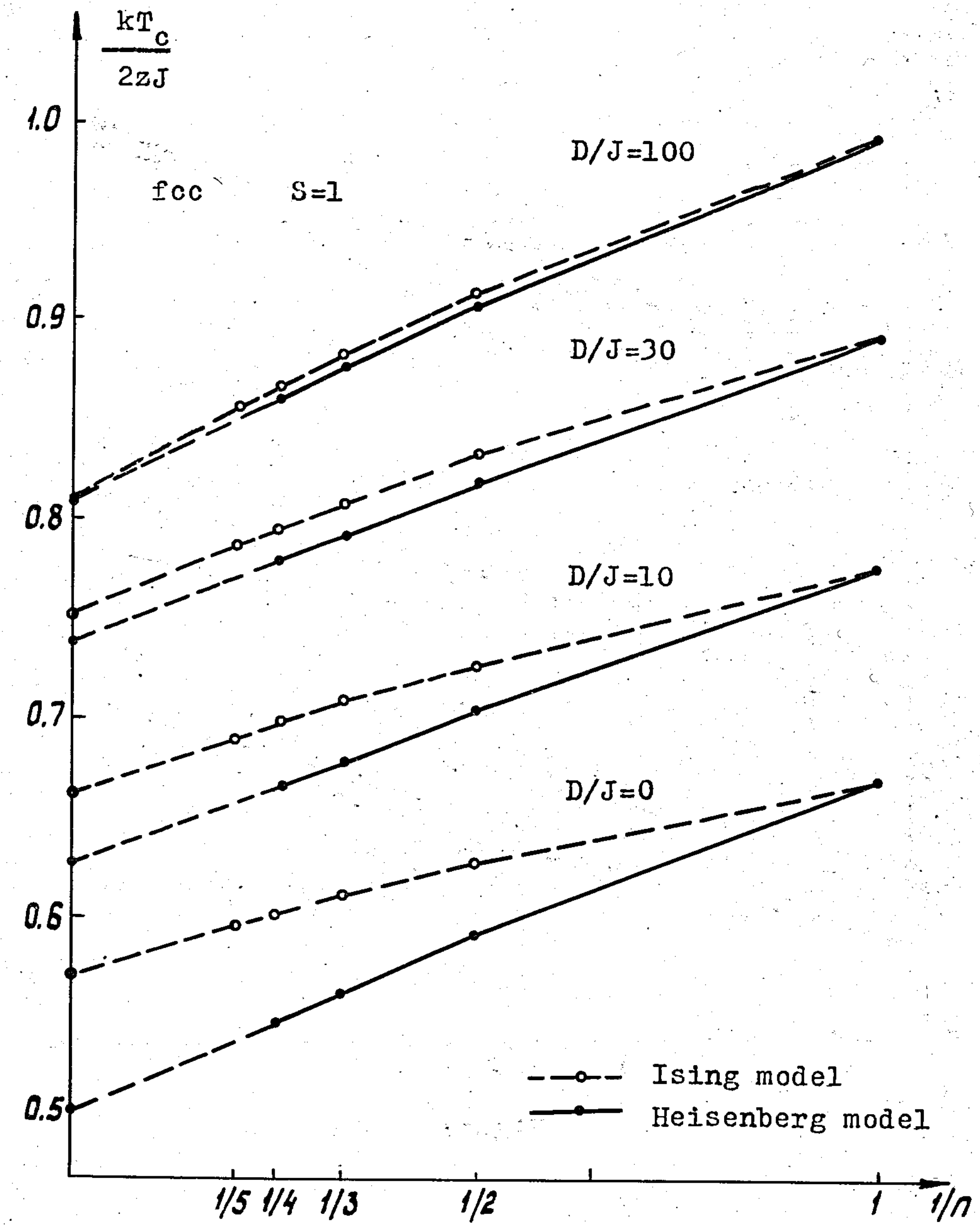
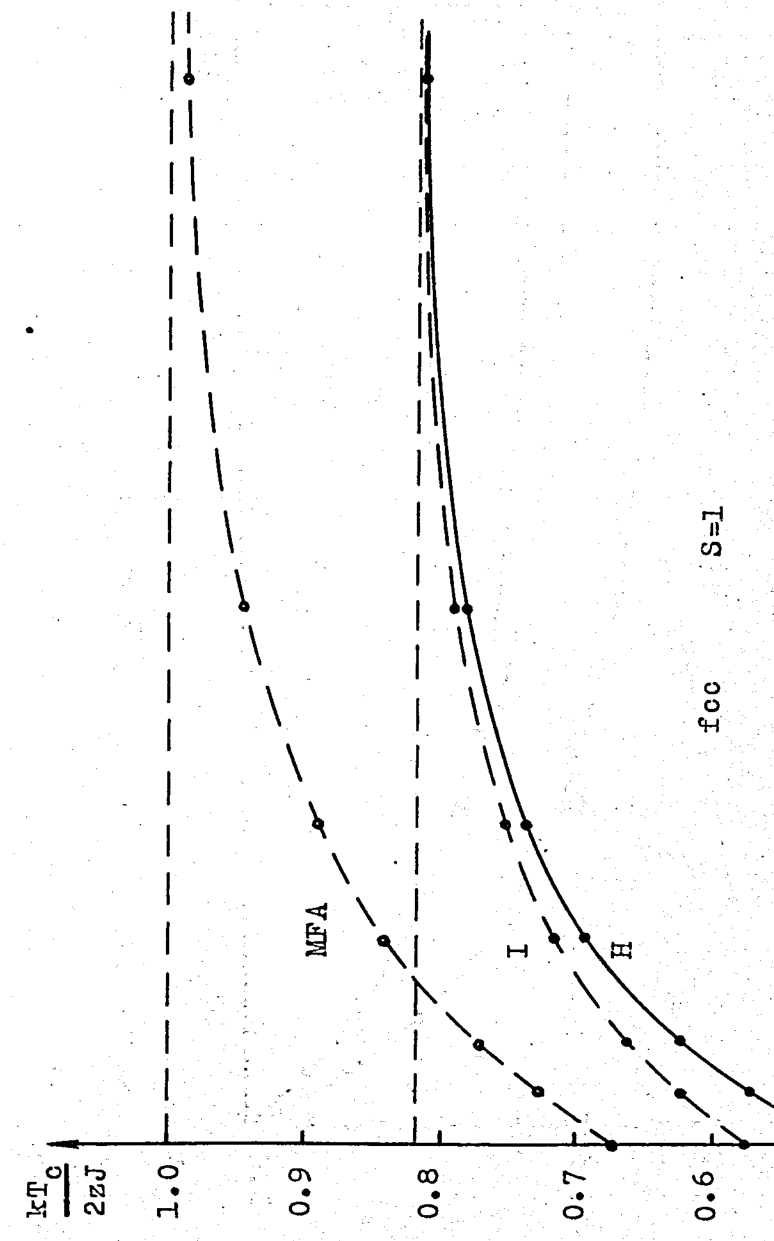


Figure 1.

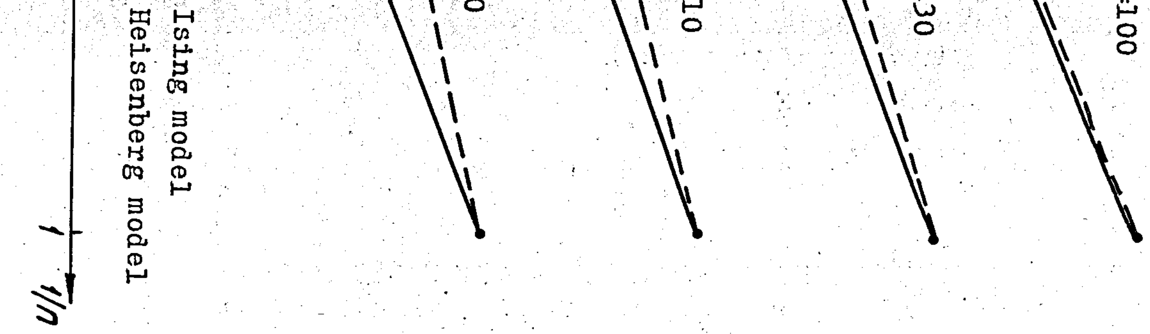


kT_c / 2zJ
fcc
S=1
D/J=100
D/J=30
D/J=10
D/J=0
Ising model
Heisenberg model
1.0
0.9
0.8
0.7
0.6
0.5
1/5
1/4
1/3
1/2
1
1/n

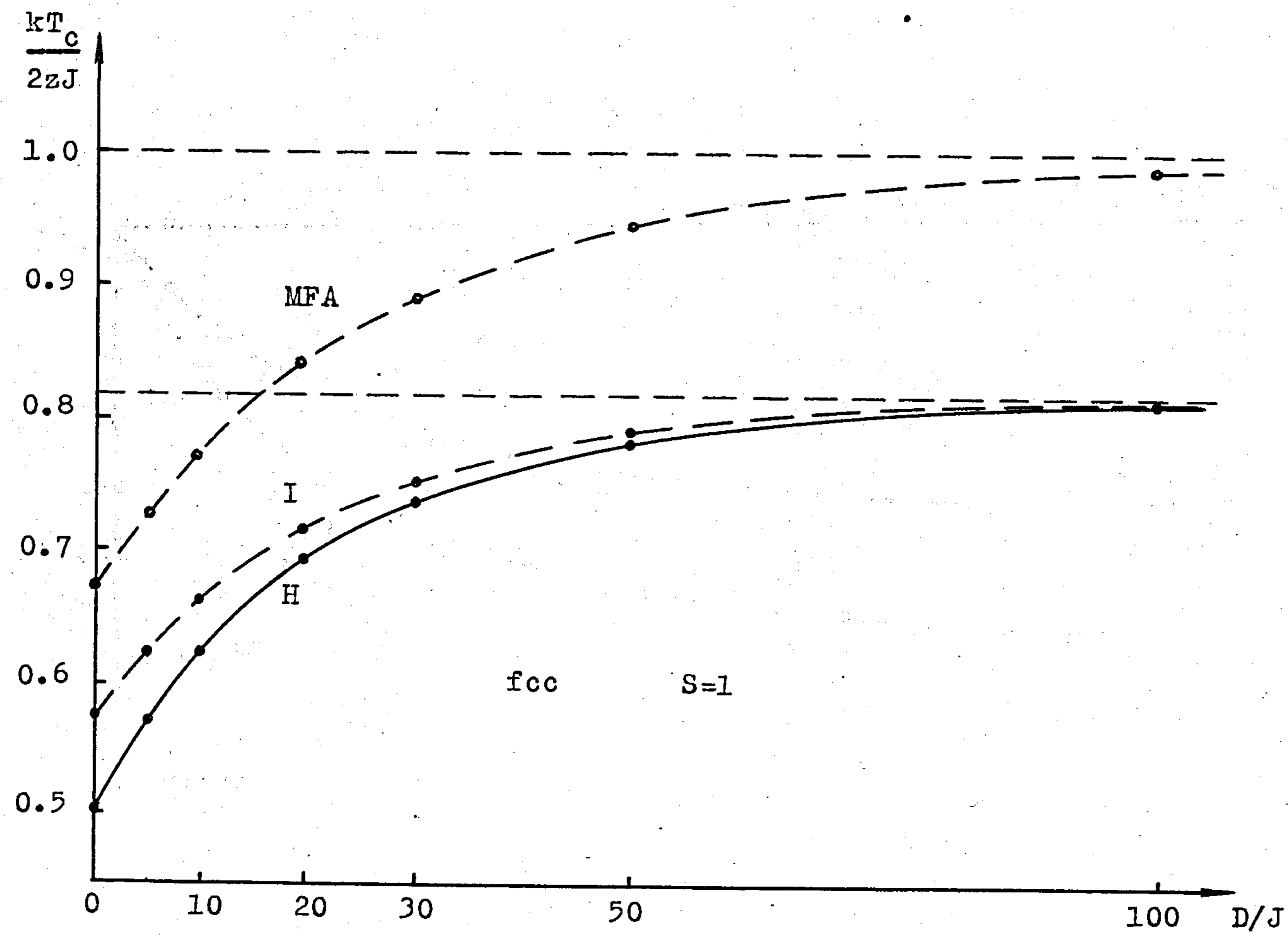
Figure 2.



S=1
fcc
MFA
I
H
kT_c / 2zJ
1.0
0.9
0.8
0.7
0.6



Ising model
Heisenberg model
1/n



kTc / 2zJ
1.0
0.9
0.8
0.7
0.6
0.5
MFA
I
H
fcc
S=1
0
10
20
30
50
100
D/J

Figure 3.

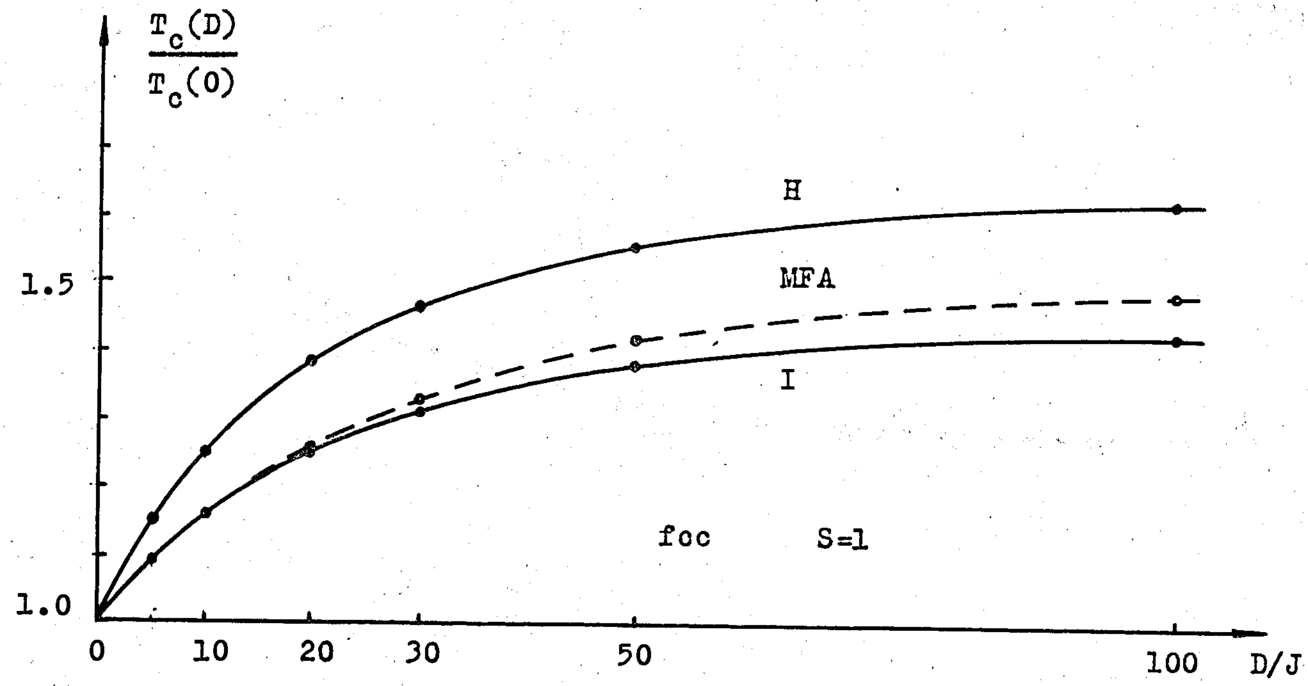


Figure 4.

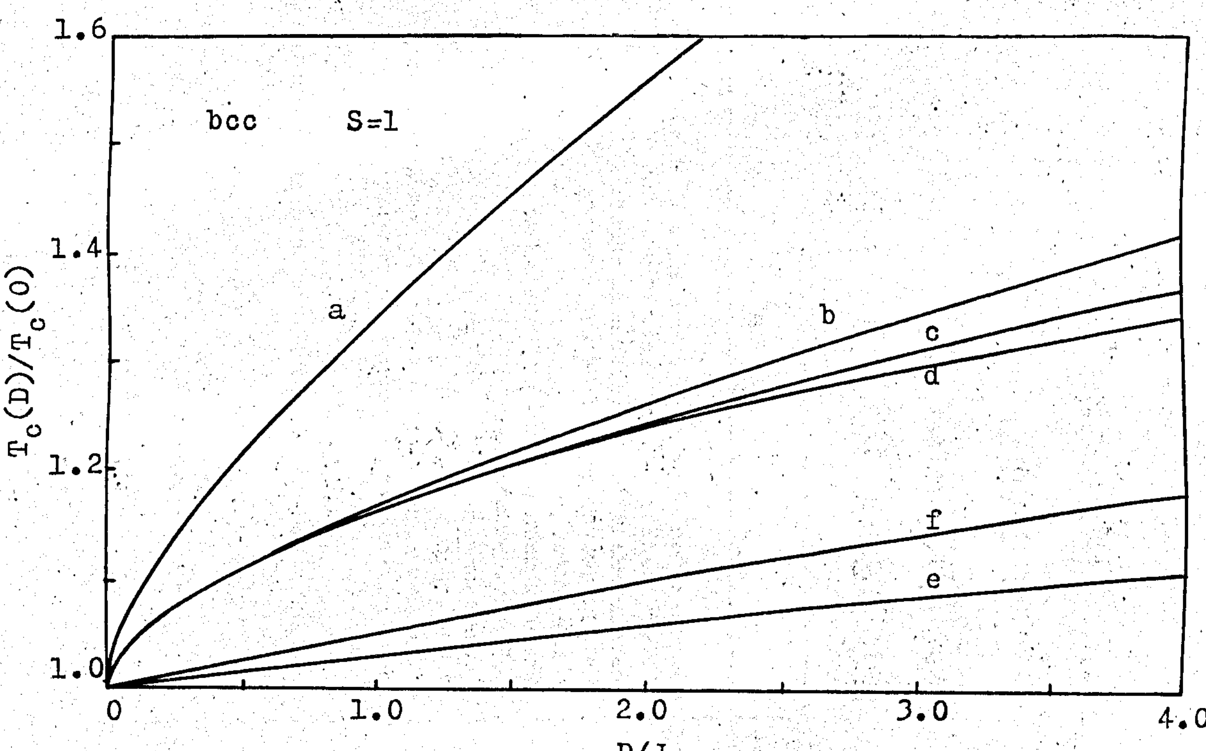


Figure 5.