

# THE VARIATIONAL METHOD FOR SYSTEMS WITH DISCRETE ENERGY LEVELS

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The variational method of calculating the thermodynamic properties of quantum systems having a discrete energy levels system is discussed. The method is a certain modification of Sawada's variational method. The method permits one to calculate all the correlation functions occurring in the problem by self-consistent means without using additional approximations constituent of the decoupling of higher correlation functions. The effectiveness of the method was illustrated by several examples taken from the theory of magnetism.

## 1. Introduction

The variational methods based on thermodynamic inequalities have received increasing attention in recent years [1, 2, 3]. These methods are employed to calculate the poles of thermodynamic Green's function or just thermodynamic correlation functions. Sawada [2] postulates the existence of a model Hamiltonian  $H_0$  for a quantum system described by Hamiltonian  $H$ , which fulfills for the assumed set of operators  $\{C_\alpha\}$  the following commutation relations

$$[H_0, C_\alpha]_- = \sum_{\alpha'} \varepsilon_{\alpha\alpha'} C_{\alpha'}, \quad (1.1)$$

where coefficients  $\varepsilon_{\alpha\alpha'}$  can be found through minimization of the free energy defined by the well known Bogolubov inequality [4]. This method yielding the equation

$$\langle [C_\alpha, [H - H_0, C_{\alpha'}^+]_-]_{\pm} \rangle_0 + \langle [[C_\alpha, H - H_0]_-, C_{\alpha'}^+]_{\pm} \rangle_0 = 0. \quad (1.2)$$

The choice of a commutator or anticommutator in equation (1.2) for the calculation of parameters  $\varepsilon_{\alpha\alpha'}$  depends on the type of commutation rules for the set  $\{C_\alpha\}$ . The form of the variational equations (1.2) suggests that the model Hamiltonian  $H_0$  has been chosen so that the first moments of the correlation functions calculated with the Hamiltonian  $H_0$  and  $H$  were equal to each other. The correspondence with the method employed in [5, 6] is visible.

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This method was used [3, 7] to calculate the correlation functions of magnetics described by Heisenberg's Hamiltonians, however in that case, beside the approximation connected with the minimization of free energy, use was made of decouplings of the higher order correlation functions [3] which could not be calculated in self-consistent manner.

Here we define the variational method sufficiently accurately as to be able to calculate precisely all the correlation functions. The only approximation used is connected with the minimization of free energy and selection of a base operator set. The results of the method are exemplified by the isotropic Heisenberg ferromagnet. It was also shown how to use the method in order to get the equation for the directions of magnetization vector in more complicated systems.

## 2. Formalism of the variational method

Let us assume that for the given physical system described by the Hamiltonian  $H$  there exists such a model Hamiltonian  $H_0$ , which together with operators of the given set  $\{O_\alpha^i\}$  fulfills the following commutation relations

$$[H_0, O_\alpha^i]_- = \sum_{j\alpha'} \varepsilon_{\alpha\alpha'}^{ij} O_{\alpha'}^j. \quad (2.1)$$

The operators of the set  $\{O_\alpha^i\}$  can be chosen in such a way that they would form a set of eigen-operators [8] of a certain part of the superoperator  $H^x = [H, \dots]_-$ . Indices  $\alpha$  and  $\alpha'$  can stand for e. g. a number of the empty lattice site or vectors of the reciprocal lattice while indices  $i, j$  are numbers of operators at fixed  $\alpha, \alpha'$ . Now we minimize the right-hand side of the Bogolubov inequality

$$F[H] \leq F_1[H_0] = -\frac{1}{\beta} \ln \text{Tr} e^{-\beta H_0} - \langle H - H_0 \rangle_0, \quad (2.2)$$

relative to the parameters  $\varepsilon_{\alpha\alpha'}^{ij}$ . The average value  $\langle \dots \rangle_0$  is defined by

$$\langle \dots \rangle_0 = \text{Tr} (e^{-\beta H_0} \dots) / \text{Tr} e^{-\beta H_0}, \quad (2.3)$$

where  $\beta = (kT)^{-1}$ ,  $k$  is the Boltzmann constant,  $T$  is temperature. Near the minimum of free energy  $F_1[H_0]$  the relation

$$F_1[H_0 + \delta H_0] = F_1[H_0] \quad (2.4)$$

must hold, where  $\delta H_0$  is a small variation of the operator  $H_0$ . Let us further assume that the variation  $\delta H_0$  is hermitian like the Hamiltonian  $H_0$  and its average value equals zero at every temperature. Let us expand the left-hand side of (2.4) with respect to small  $\delta H_0$ . As a result we get

$$\left\langle \int_0^\beta d\beta' e^{\beta' H_0} \delta H_0 e^{-\beta' H_0} (H - H_0) \right\rangle_0 = 0. \quad (2.5)$$

One of the possible forms of the variation  $\delta H_0$  under the assumed conditions is

$$\delta H_0 = \sum_{l,m} \sum_{ij} \sum_{\alpha\alpha'} \omega_{\alpha\alpha'}^{ij} (G_{\alpha\alpha'}^{ij})_{l,m} \frac{\beta(\varepsilon_l - \varepsilon_m)}{e^{\beta/2(\varepsilon_l - \varepsilon_m)} - e^{-\beta/2(\varepsilon_l - \varepsilon_m)}} |l\rangle \langle m|, \quad (2.6)$$

where  $\varepsilon_l, \varepsilon_m$  are eigenvalues of the model Hamiltonian

$$H_0|l\rangle = \varepsilon_l|l\rangle, \quad (2.7)$$

and  $(G_{\alpha\alpha'}^{ij})_{lm}$  is a matrix element of the hermitian operator  $G_{\alpha\alpha'}^{ij}$  defined as follows

$$\begin{aligned} G_{\alpha\alpha'}^{ij} = & -\frac{1}{2} e^{\beta/2H_0} [O_\alpha^i (O_{\alpha'}^j)^+ + O_{\alpha'}^j (O_\alpha^i)^+] e^{-\beta/2H_0} \\ & -\frac{1}{2} e^{-\beta/2H_0} [O_\alpha^i (O_{\alpha'}^j)^+ + O_{\alpha'}^j (O_\alpha^i)^+] e^{\beta/2H_0} + e^{\beta/2H_0} (O_{\alpha'}^j)^+ e^{-\beta H_0} O_\alpha^i e^{\beta/2H_0} \\ & + e^{\beta/2H_0} (O_\alpha^i)^+ e^{-\beta H_0} O_{\alpha'}^j e^{\beta/2H_0}. \end{aligned} \quad (2.8)$$

Equation (2.5) can be significantly simplified if one takes the above defined variation (2.6). We get

$$\sum_{ij} \sum_{\alpha\alpha'} \omega_{\alpha\alpha'}^{ij} \{ \langle O_\alpha^i [H - H_0, (O_{\alpha'}^j)^+]_- \rangle_0 + \langle [O_\alpha^i, H - H_0]_- (O_{\alpha'}^j)^+ \rangle_0 \} = 0. \quad (2.9)$$

The variation  $\delta H_0$  must be arbitrary in such a sense that it is an arbitrary operator in a space  $\{|l\rangle \langle m|\}$ , hence the coefficients  $\omega_{\alpha\alpha'}^{ij}$  are arbitrary and take on independent values. Taking into account equation (2.9) it appears hence that the basic relations of the variational method are of the form

$$\langle \langle O_\alpha^i [H - H_0, (O_{\alpha'}^j)^+]_- \rangle_0 + \langle [O_\alpha^i, H - H_0]_- (O_{\alpha'}^j)^+ \rangle_0 = 0, \quad (2.10)$$

and are fulfilled for every  $i, j$  and  $\alpha, \alpha'$ . Now, it is enough to assume that

$$\{O_\alpha^i\} = \{(O_\alpha^i)^+\}, \quad (2.11)$$

to get the relations (2.10) in a more clear form

$$\langle [O_\alpha^i, [H - H_0, (O_{\alpha'}^j)^+]_- ]_\pm \rangle_0 + \langle [[O_\alpha^i, H - H_0]_-, (O_{\alpha'}^j)^+]_\pm \rangle_0 = 0, \quad (2.12)$$

since the algebra of the given set  $\{O_\alpha^i\}$  is also very well defined. Employing relations (2.12) one can easily calculate all parameters  $e_{\alpha\alpha'}^{ij}$  of equation (2.1). The problem of existence of the model Hamiltonian  $H_0$  which would fulfill simultaneously equations (2.1) and (2.12) is not clear yet. The main part of the difficulties in proving the existence of  $H_0$  comes from the fact that in the variational method only the relations of  $H_0$  with the given set  $\{O_\alpha^i\}$  (2.1) is taken into account in (2.12) and not its explicit form.

### 3. Thermodynamics of the isotropic Heisenberg ferromagnet

The Hamiltonian of the electron spins interacting with an external magnetic field and among themselves is of the form

$$H = -\mu h \sum_f S_f^z - \sum_{f \neq g} J_{fg} (S_f^z S_g^z + S_f^+ S_g^-). \quad (3.1)$$

The correlation function and a value of magnetization of the isotropic Heisenberg ferromagnet can be calculated applying equations of the variational method (2.10) with a model Hamiltonian  $H_0$  which together with operators of the set

$$\{O_f^i\} = \{S_f^-, S_f^+, S_f^- S_f^z, S_f^z S_f^+\}, \quad (3.2)$$

fulfills the commutation relations

$$[H_0, O_f^i] = \sum_g \varepsilon_{fg}^i O_g^i, \quad (3.3)$$

where indices  $f, g$  denote the lattice sites. Let us note that if we want the model Hamiltonian to describe correctly the thermodynamics of the interacting spin system it should exhibit at least principal features of the symmetry of the Hamiltonian (3.1). Let us assume that  $H_0$  is translationally invariant and also the rotation around the third axis does not change its form. From these assumptions it follows that the relation

$$\begin{aligned} \langle [H, [O_f^i, (O_g^j)^+]_-]_- \rangle_0 &= \langle [[O_f^i, H - H_0]_-, (O_g^j)^+]_- \rangle_0 \\ &+ \langle [O_f^i, [(O_g^j)^+, H - H_0]_-]_- \rangle_0 = 0 \end{aligned} \quad (3.4)$$

holds. Relations (3.4) for  $f \neq g$  are trivial, while there are two cases for  $f = g$ . In the first case, when the commutator  $[O_f^i, (O_f^j)^+]_-$  is a linear combination of operators  $1, S_f^z, \dots, (S_f^z)^{2S}$  equation (3.4) becomes obvious when we make use of the translational invariance of Hamiltonian (3.1). In the second one, when  $[O_f^i, (O_f^j)^+]_-$  is an operator  $(S_f^-)^2$  or  $(S_f^+)^2$  we can prove the correctness of relation (3.4) employing the invariance of  $H$  with respect to the rotation around the third axis.

Combining equations (3.4) and (2.12) we get

$$\langle [O_f^i, [H - H_0, (O_g^j)^+]_-]_- \rangle_0 = 0, \quad (3.5)$$

from which we can determine the coefficients  $\varepsilon_{fg}^i$  for the isotropic ferromagnet. To calculate numbers  $\varepsilon_{f,g}^1$  defined by

$$[H_0, S_f^-] = \sum_g \varepsilon_{f,g}^1 S_g^-, \quad (3.6)$$

we shall make use of two from among equations (3.5). These are

$$\langle [[S_f^-, H - H_0]_-, S_g^+]_- \rangle_0 = 0, \quad (3.7)$$

and

$$\langle [[S_f^-, H - H_0]_-, S_g^z S_g^+]_- \rangle_0 = 0. \quad (3.8)$$

We get the explicit form of equations (3.7) and (3.8) employing known spin operator commutation rules. Performing the Fourier transformation [9] we get

$$\varepsilon_k^1 = \mu h + 2(J_0 - J_k)f + 2(J_0 - J_k) \frac{X}{\sigma}, \quad (3.9)$$

and from (3.8)

$$\varepsilon_k^1 = \mu h + 2(J_0 - J_k)f + 2(J_0 - J_k) \frac{3Y + X - S(S+1)\sigma}{\sigma + \lambda}, \quad (3.10)$$



where

$$\varepsilon_k^1 = \sum_{f=g} e^{ik(f-g)} \varepsilon_{fg}^1,$$

$$\sigma = \langle S_f^z \rangle_0, \quad \lambda = \langle 3(S_f^z)^2 - S(S+1) \rangle_0, \quad (3.11)$$

and

$$X = \frac{1}{N} \sum_k \frac{J_k}{J_0} \langle S_k^z S_{-k}^z \rangle_0, \quad (3.12)$$

$$Y = \frac{1}{N} \sum_k \frac{J_k}{J_0} \langle S_k^z (S^z)_{-k}^2 \rangle_0, \quad (3.13)$$

$$f = \frac{1}{N} \sum_k \frac{J_k}{J_0} (e^{\beta \varepsilon_k^1} - 1)^{-1}. \quad (3.14)$$

Comparison of (3.9) with (3.10) yields the first equation connecting the correlation functions  $X$  and  $Y$

$$-\lambda X + 3\delta Y = S(S+1)\sigma^2. \quad (3.15)$$

Now we assume  $f = g$ ,  $i = j$ , and  $O_f^i = S_f^-$  in equation (3.5) and afterwards we perform a summation over every direct lattice site. Hence we obtain

$$\sum_k \varepsilon_k^1 \langle S_k^- S_{-k}^+ \rangle_0 = \sum_f \langle [S_f^-, H] - S_f^+ \rangle_0. \quad (3.16)$$

When all commutators of the right-hand side of (3.16) are calculated, one can apply the identity for spin operators

$$S_f^- = S(S+1) - S_f^- S_f^+ - (S_f^z)^2 \quad (3.17)$$

to obtain equation (3.16) in the form

$$(1 + 2\varphi - 2f)X + Y = S(S+1)\sigma - (\lambda + \sigma)f - 2f(\varphi - f)\delta, \quad (3.18)$$

where

$$\varphi = \frac{1}{N} \sum_k (\exp \beta \varepsilon_k^1 - 1)^{-1}. \quad (3.19)$$

From the set of equations (3.15) and (3.18) we calculate the correlation functions  $X$  and  $Y$  and then, applying their explicit form we obtain the excitation energy

$$\varepsilon_k^1 = \mu h + 2(J_0 - J_k)SR, \quad (3.20)$$

where

$$SR = \frac{S(S+1)\sigma - f\lambda}{S(S+1) - 3f\sigma}. \quad (3.21)$$

For spin  $S = \frac{1}{2}$  we obtain

$$\varepsilon_k^1 = \mu h + \frac{J_0 - J_k}{1 + 2\varphi - 2f}, \quad (3.22)$$

which coincides with the result derived in [7] and [10]. We can calculate the  $N$ -th power of the  $z$ -component of the spin vector using the formula [11]

$$\langle (S^z)^N \rangle_0 = \left[ S^N + \sum_{k=1}^{2S} (S-k)^N \left( \frac{\varphi}{1+\varphi} \right)^k \right] \left[ \sum_{k=0}^{2S} \left( \frac{\varphi}{1+\varphi} \right)^k \right]^{-1}. \quad (3.23)$$

Near the phase transition point we expand the magnetization  $\sigma$  and factor  $\lambda = \langle 3(S^z)^2 - S(S+1) \rangle$  with respect to small  $(1+2\varphi)^{-1}$  applying formula (3.23). Hence we obtain

$$\sigma = \frac{2S(S+1)}{3(1+2\varphi)} + O(\varphi^{-2}), \quad (3.24)$$

$$\lambda = \frac{2S(S+1)(2S-1)(2S+3)}{15(1+2\varphi)^2} + O(\varphi^{-3}). \quad (3.25)$$

Functions  $\varphi$  and  $f$  are to be expanded with respect to temperature

$$1 + 2\varphi \approx \frac{I(1)}{SR\beta_c J_0}, \quad (3.26)$$

$$f \approx \frac{I(1)-1}{2SR\beta_c J_0}. \quad (3.27)$$

The renormalization factor  $SR$ , defined in (3.21), behaves for high temperatures as

$$SR = \frac{2S(S+1)}{3(1+2\varphi)} \left[ I(1) - (I(1)-1) \frac{(2S+3)(2S-1)}{10S(S+1)} \right]. \quad (3.28)$$

Combining expressions (3.24), (3.26) and (3.28) we obtain the following formula for the phase transition temperature

$$kT_c = \frac{2S(S+1)}{3} J_0 \left[ 1 - \frac{I(1)-1}{I(1)} \frac{(2S+3)(2S-1)}{10S(S+1)} \right], \quad (3.29)$$

and for  $S = \frac{1}{2}$  we get the result following from the molecular field theory. The expression

$$1 + 2\varphi = \frac{1}{N} \sum_k \operatorname{cth} \frac{\beta \varepsilon_k^1}{2} \approx \frac{1}{N} \sum_k \left( \frac{2}{\beta \varepsilon_k^1} + \frac{3}{2} \beta \varepsilon_k^1 \right), \quad T < T_c \quad (3.30)$$

permits one to write down the magnetization behaviour as

$$\sigma \sim \left( 1 - \frac{T}{T_c} \right)^{1/2}, \quad (3.31)$$

and the magnetic susceptibility

$$\chi \sim \left( \frac{T}{T_c} - 1 \right)^{-2} \quad (3.32)$$

near the critical point, which means that the critical indices assume values  $\beta = \frac{1}{2}$  and  $\gamma = 2$ . At low temperatures, the magnetization and renormalization factor  $SR$  behave like

$$\sigma = S - \varphi + (2S+1)\varphi^{2S+1} + \dots, \quad (3.33)$$

$$SR = S - f + \varphi + \dots, \quad (3.34)$$

whereas the expansions of the functions  $\varphi$  and  $f$  with respect to temperature are

$$\varphi = z\left(\frac{3}{2}\right) \left(\frac{\tau}{R}\right)^{3/2} + \frac{3\pi}{4} z\left(\frac{5}{2}\right) \left(\frac{\tau}{R}\right)^{5/2} + \frac{33\pi^2}{32} z\left(\frac{7}{2}\right) \left(\frac{\tau}{R}\right)^{7/2} + \dots, \quad (3.35)$$

$$f = z\left(\frac{3}{2}\right) \left(\frac{\tau}{R}\right)^{3/2} - \frac{\pi}{4} z\left(\frac{5}{2}\right) \left(\frac{\tau}{R}\right)^{5/2} + \dots, \quad (3.36)$$

where

$$\tau = \frac{3kT}{4\pi J_0 S} \quad \text{and} \quad z(\alpha) = \sum_{n=1}^{\infty} n^{-\alpha} e^{-\mu h \beta n}.$$

Substituting (3.36), (3.35) into (3.34) we get

$$SR = S - \pi z\left(\frac{5}{2}\right) \tau^{5/2} + \dots, \quad (3.37)$$

and

$$\sigma = S - z\left(\frac{3}{2}\right) \tau^{3/2} - \frac{3\pi}{4} z\left(\frac{5}{2}\right) \tau^{5/2} - \frac{33\pi^2}{32} z\left(\frac{7}{2}\right) \tau^{7/2} - \frac{3\pi}{2S} z\left(\frac{3}{2}\right) z\left(\frac{5}{2}\right) \tau^4 + \dots, \quad (3.38)$$

Within that the term proportional to  $T^3$  in the expression for  $\sigma$  appears in the case of  $S = \frac{1}{2}$  only. The obtained results (3.37) and (3.38) coincides with those of Dyson [12].

#### 4. The equations for the direction of magnetization

We restrict our considerations to the case when a ferromagnet is isotropic in such a sense that every site of its crystalline lattice responds in the same way to an external magnetic field and internal interactions. We rotate the coordination system in such a way that a new third axis coincides with the magnetization axis.

For our Hamiltonian we derive a new model one which for a given set of operators  $\{O_\alpha^i\}$  fulfills the commutation relations

$$[H_0, O_\alpha^i]_- = \sum_{i'\alpha'} \epsilon_{\alpha\alpha'}^{ii'} O_{\alpha'}^{i'}. \quad (4.1)$$

Parameters  $\varepsilon_{\alpha\alpha'}^{ii'}$  are to be calculated employing equations of the variational method. These numbers obviously depend on an angle of rotation of the coordination system. Now we define the set of Green functions

$$\langle\langle O_{\alpha}^i(t) | O_{\beta}^j \rangle\rangle = -i\theta(t) \langle [O_{\alpha}^i(t), O_{\beta}^j]_- \rangle_0, \quad (4.2)$$

where  $\langle \dots \rangle_0$  is a thermodynamic average value being calculated with the model Hamiltonian  $H_0$  and  $A(t)$  is the operator in a Heisenberg representation involving also the model Hamiltonian  $H_0$ . For these Green functions there holds an identity

$$\langle\langle O_{\alpha}^i | O_{\beta}^j \rangle\rangle_E = \langle\langle O_{\beta}^j | O_{\alpha}^i \rangle\rangle_{-E}, \quad (4.3)$$

where  $\langle\langle | \rangle\rangle_E$  is the Fourier transform of function (4.2). If the parameters  $\varepsilon_{\alpha\alpha'}^{ii'}$  are known then it is easy to find the explicit form of Green's functions (4.2) and to make use of the equation (4.3). Each of the equations (4.3) defines in a unique way an angle by which the system of coordinates should be rotated to get the third axis coinciding with the magnetization one. It means that equation (4.3) is the equation for the direction of magnetization. Notice that for the isotropic Heisenberg ferromagnet the rotation angle sought for is exactly that formed by the external magnetic field with an old third coordination system axis. In other words the magnetization in its own direction superimposes the external magnetic field.

### 5. Discussion

A new possibility of employing the variational method has been indicated. The variation  $\delta H_0$  was defined in such a way that the calculation of the correlation functions for a system with a discrete energy levels system can be done in a selfconsistent way. It was found that the reasonability of results obtained by the method at hand depends also on an appropriate choice of the set of operators  $\{O_{\alpha}^i\}$ . We propose to choose these operators in such a way that the obtained set is a solution of the respective eigenproblem [8] for a part of the Hamiltonian of a system described.

The variational method has been verified for the isotropic Heisenberg ferromagnet. It was shown that the low temperature results agree with those of Dyson [12] and others [7, 10] and the high temperature ones with those of Oguchi [7]. The phase transition temperature as calculated using formula (3.29) relates to the other results as follows

$$T_c^{\text{RPA}} < T_c^{(3.29)} = T_c^{\text{Tahir-Kheli}} = T_c^{\text{Oguchi}} = T_c^{\text{MFA}},$$

for  $S = \frac{1}{2}$ , and

$$T_c^{\text{RPA}} < T_c^{(3.29)} < T_c^{\text{Tahir-Kheli}} = T_c^{\text{Oguchi}} = T_c^{\text{MFA}},$$

for  $S \geq 1$ .

It appears that the correlation function  $X = \frac{1}{N} \sum_k \frac{J_k}{J_0} \langle S_k^z S_{-k}^z \rangle_0$  near the phase transition temperature as calculated by the method presented here changes its sign from



positive to negative regardless of the choice of the basic set of operators. However this fault cannot be avoided by this method. Therefore the correctness of results obtained by the use of the variational method as well as other ones [6, 7] for high temperature is doubtful. The result obtained for low temperature are very correct and conformable to [12, 10]. For  $T < T_c$  the new method of deriving equations for direction of magnetization was proposed. Since results obtained for simple spin Hamiltonians are rather trivial, we did not present the calculations restricting ourselves to the presentation of the problem itself.

Study of the magnetization directions in magnetics displaying anisotropy of the crystal-line field or exchange type are to be continued.

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