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Nonequilibrium Properties of an Ising-Model Ferromagnet*

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The dynamical properties of a uniform-coupling Ising-model ferromagnet in contact with a thermal reservoir are studied using density-operator methods. We proceed from the reversible microscopic dynamics of the system reservoir to an irreversible equation of motion for the macroscopic magnetization probability density. The temporal evolution of the spin system is seen to depend upon details of the reservoir system, while the steady-state results yield the correct magnetic free energy and equation of state, independent of such details.

I. INTRODUCTION

The equilibrium properties of a ferromagnet have been extensively studied experimentally and various theoretical models have been suggested to account for the data.[1] For a dielectric crystal whose magnetic properties do not involve the orbital degrees of freedom of the magnetic ions, just the spin moments of the electrons contributing to the magnetic ordering, the Heisenberg model has served usefully by providing a Hamiltonian upon which many theoretical studies have been based. This model of the magnetic interactions of localized moments describes the interaction energy of the system as the sum of pair interactions each of which is proportional, through a single spatially varying exchange parameter, to the scalar product of the spin vectors of the two moments. Thus, in the isotropic Heisenberg model the different components of the spins of the two magnetic ions have equal interaction energies. This situation may be generalized somewhat by introducing different exchange parameters to couple the three components of each spin pair. An extreme limit of this extension of the Heisenberg model is the Ising model in which the two transverse coupling constants are made to vanish identically, thus leaving the interaction energy of each pair of spins proportional to a single spatially varying exchange parameter which couples only one component of the spin vectors of the pair.

The Ising model affords a significant simplification from the full Heisenberg-model Hamiltonian in the study of the equilibrium properties of the magnetic system in that all of the operators appearing in the Ising Hamiltonian commute with each other, thus allowing one to neglect the quantum-mechanical aspects of the problem and treat the system as though it were entirely classical. This simplification has, in fact, resulted in exact solutions[2] of aspects of the equilibrium statistical mechanics of this model for particular spatial lattices. Thus, although a great amount is known about the equilibrium properties of the Ising model, the nonequilibrium properties of this magnetic system have not been as thoroughly explored. In this paper we shall give a treatment of the nonequilibrium statistical dynamical properties of this system which yields the correct thermal equilibrium behavior as its steady-state (time-independent) solution. Our approach differs from that of Glauber,[3] whose well-known model has formed the principal basis for several studies[4] of dynamical properties of an Ising system, in that we proceed from reversible microscopic dynamics to an irreversible equation of motion for the magnetization probability density. This approach is also in contrast to recent studies[5] of critical phenomena which proceed to derive expressions for various moments of the probability distribution (e.g., correlation functions). Although there have been previous studies of the distribution function itself[3,6] based on assumed master equations of motion with associated transition probabilities, here we derive the equation of motion of the probability density from microscopic theory.

We shall make use of density-operator techniques similar to those that have previously been applied to study fluctuation phenomena in lasers.[7] Our general approach will be to consider the ferromagnetic spin system to be in contact with a second system which serves as a constant-temperature thermal reservoir. We shall obtain the equation of motion of the combined reservoir-ferromagnet-system density operator and then trace over reservoir states to obtain an equation of motion for the reduced density operator of the spin system. The latter operator contains all of the information needed for the calculation of the dynamical and fluctuation properties of the ferromagnet.

In Sec. II we present the model and derive the equation of motion for the reduced density operator of the spin system. We find it convenient to introduce an operator-ordering scheme and translate this density-operator equation of motion into a c-number differential-difference equation for the c-number magnetization quasiprobability density. In Sec. III we study the dynamical properties of the system near the ferromagnetic transition temperature and obtain a Fokker–Planck type of equation for the magnetization probability density. We show in Sec. IV that the exact equation of motion may be solved at steady state for all temperatures and that it yields the Bragg–Williams free energy. The results obtained are further discussed in Sec. V.

II. MODEL AND EQUATIONS OF MOTION

We take as our total Hamiltonian H the sum of the ferromagnetic-system Hamiltonian H_S, the reservoir-system Hamiltonian H_R, and an interaction V:

$$H = H_S + H_R + V . \tag{2.1}$$

We choose the Ising model for our magnetic system:

$$H_S = -2\sum_{i,j=1}^{N} J_{ij} S_i^z S_j^z - 2H_0 \sum_{j=1}^{N} S_j^z ; \tag{2.2}$$

that is, we imagine N spin-$\frac{1}{2}$ angular momenta, localized at fixed lattice sites and each having magnetic moment μ, to interact ferromagnetically ($J_{ij} \geqslant 0$) with each other and also with an externally applied uniform magnetic field H_0 which is taken to define the z direction of the system. The angular momentum operators obey the usual commutation relations (for a spin-$\frac{1}{2}$ total angular momentum):

$$[S_j^x, S_k^y] = i\delta_{jk} S_j^z \tag{2.3}$$

(δ_{jk} is the Kronecker δ function) and the two similar relations obtained from (2.3) by cyclically permitting the spacial indices x, y, and z.

We shall imagine that the spin system is in thermal contact with another system (such as the phonons in the crystal, or spin impurities) which forms the reservoir. The specific nature of the reservoir will be shown to be important in determining the dynamical behavior of the spin system. This will be clarified when particular examples are considered below. For now we shall take the reservoir Hamiltonian to represent a set of uncoupled boson modes:

$$H_R = \sum_\alpha \hbar\,\Omega_\alpha O_\alpha^\dagger O_\alpha ,$$
$$[O_\alpha, O_\beta] = 0, \quad [O_\alpha, O_\beta^\dagger] = \delta_{\alpha\beta} . \tag{2.4}$$

We take the following general form for the system-reservoir interaction V:

$$V = \sum_{j=1}^{N} \sum_\alpha g_\alpha(r_j)\,[S_j^- O_\alpha + S_j^+ O_\alpha^\dagger] , \tag{2.5}$$

where

$$S_j^\pm = S_j^x \pm i S_j^y \tag{2.6}$$

are the usual spin raising (+) and lowering (−) operators for the jth spin. For simplicity, we consider only a single effective coupling constant $g_\alpha(j)$ which couples the αth reservoir mode to the spin operators located at the various sites r_j. The spin system may exchange energy with the reservoir system by means of this interaction. We are neglecting here spin-spin interactions, so that the processes considered here will contribute only to the longitudinal relaxation time T_1 of the ferromagnet.

Having specified our total system, we proceed to consider an equation of motion for its density operator $\rho(t)$ in the interaction picture:

$$\dot{\rho}(t) = i\hbar^{-1}[V(t), \rho(t)] , \tag{2.7}$$

where

$$V(t) = e^{i\hbar^{-1}(H_S+H_R)t}\, V e^{-i\hbar^{-1}(H_S+H_R)t} \tag{2.8}$$

Using the interaction (2.5) and the reservoir Hamiltonian (2.4), we obtain

$$V(t) = \sum_{j=1}^{N}\sum_\alpha g_\alpha(r_j)\,[S_j^-(t)\,O_\alpha e^{-i\Omega_\alpha t} + S_j^+(t)\,O_\alpha^\dagger e^{i\Omega_\alpha t}] . \tag{2.9}$$

We shall use the interaction in this form in the density-operator equation of motion (2.7). We note that an equivalent form of (2.7) is the integral equation

$$\rho(t) = \rho(-\infty) - i\hbar^{-1}\int_{-\infty}^{t} dt'\,[V(t'), \rho(t')] , \tag{2.10}$$

so that, substituting (2.10) into (2.7) and tracing over the reservoir variables, we obtain an equation for the reduced density operator $\sigma(t)$, where $\sigma(t) \equiv \mathrm{Tr}_R\,\rho(t)$, of the spin system:

$$\dot{\sigma}(t) = -\hbar^{-2}\,\mathrm{Tr}_R \int_{-\infty}^{t} dt'\,[V(t), [V(t'), \rho(t')]] . \tag{2.11}$$

We shall assume that the reservoir remains for all time in thermal equilibrium and thus write

$$\rho(t) = \sigma(t) \otimes f_R , \tag{2.12}$$

where

$$f_R = Z_R^{-1} e^{-\beta H_R} , \tag{2.13}$$

$$\beta = (k_B T)^{-1} , \tag{2.14}$$

$$Z_R = \mathrm{Tr}_R\,(e^{-\beta H_R}) , \tag{2.15}$$

and thus obtain

$$\dot{\sigma}(t) = -\hbar^{-2}\int_{-\infty}^{t} dt'\,\mathrm{Tr}_R[V(t), [V(t'); \sigma(t') \otimes f_R]] \tag{2.16}$$

($\otimes$ signifies direct product).

We note, recalling the interaction (2.9), that (2.16) is an expression involving spin operators and time correlation functions of reservoir operators. We shall assume that the time correlation functions of reservoir variables are nonvanishing only for time intervals much shorter than the characteristic times over which the spin system [and hence $\sigma(t)$] evolves appreciably. This will indeed turn out to be the case when the reservoir time correlation functions will be explicitly calculated below for two types of reservoir systems and will be shown to be δ functions in $t-t'$. Thus, we may rewrite (2.16) by replacing $\sigma(t')$ by $\sigma(t)$—thus making the master equation Markovian[8]:

$$\sigma(t)=-\hbar^{-2}\int_{-\infty}^{t} dt'\,\mathrm{Tr}_R[V(t),\,[V(t');\,\sigma(t)\otimes f_R]]. \tag{2.17}$$

Using the interaction (2.9) in (2.17), we obtain the following equation for the reduced-system density operator:

$$\begin{aligned}\dot{\sigma}(t)=-2\int_{-\infty}^{t} dt' \sum_{j=1}^{N} \{&S_j^{+}(t)S_j^{-}(t')\sigma(t)F_1(t,t')\\ &+S_j^{-}(t)S_j^{+}(t')\sigma(t)F_2(t,t')-S_j^{+}(t)\sigma(t)S_j^{-}(t')F_2(t',t)\\ &-S_j^{-}(t')\sigma(t)S_j^{+}(t')F_1(t',t)\}\,,\end{aligned} \tag{2.18}$$

where

$$F_1(t,t')=g^2\left\langle \sum_{\alpha} O_\alpha^{\dagger}(t)\sum_{\beta} O_\beta(t')\right\rangle_R\,, \tag{2.19}$$

$$F_2(t,t')=g^2\left\langle \sum_{\alpha} O_\alpha(t)\sum_{\beta} O_\beta^{\dagger}(t')\right\rangle_R\,. \tag{2.20}$$

In deriving (2.18) we have assumed that a second-order perturbation treatment of the system-reservoir coupling is adequate and also that each spin interacts with the reservoir independently of all the others; that is, we take

$$\begin{aligned}\sum_{\alpha} g_\alpha(r_j)g_\alpha(r_k)\,\mathrm{Tr}_R[O_\alpha O_\alpha^{\dagger} f_R] &=\sum_{\alpha} g_\alpha(r_j)g_\alpha(r_k)\langle O_\alpha O_\alpha^{\dagger}\rangle_R\\ &\propto g^2\delta(r_j-r_k)\,,\end{aligned} \tag{2.21}$$

where $g=g_\alpha(0)$.

We wish to focus our attention on the dynamical behavior of the z component of the total spin. It is convenient to introduce a c-number quasiprobability distribution function[9-11] for this quantity. We shall also introduce a transformation from spin operators to "atomic" operators associated with the two levels of each spin-$\frac{1}{2}$ system. We shall denote the two states of the jth spin by $|1,j\rangle$ and $|2,j\rangle$, these being the state of higher energy (but lower z component of spin, $\langle 1,j|S_j^z|1,j\rangle=-\frac{1}{2}$) and the state of lower energy (but higher z component of spin, $\langle 2,j|S_j^z|2,j\rangle=+\frac{1}{2}$), respectively. We note the following relations:

$$S_j^{-}\equiv |2,j\rangle\langle 1,j|\,, \tag{2.22}$$

$$S^{-}\equiv\sum_{j=1}^{N} S_j^{-}\,, \tag{2.23}$$

$$S_j^{+}\equiv |1,j\rangle\langle 2,j|\,, \tag{2.24}$$

$$S^{+}\equiv\sum_{j=1}^{N} S_j^{+}\,, \tag{2.25}$$

$$S_j^{z}\equiv\tfrac{1}{2}(|1,j\rangle\langle 1,j|-|2,j\rangle\langle 2,j|)\,, \tag{2.26}$$

$$S^{z}\equiv\sum_{j=1}^{N} S_j^{z}\,, \tag{2.27}$$

$$I_j\equiv |1,j\rangle\langle 1,j|+|2,j\rangle\langle 2,j|\,. \tag{2.28}$$

The two states of a single spin are taken to be normalized to unity and orthogonal to each other, while states of different spins are also taken to be orthogonal. We define operators N_1 and N_2 corresponding to the total populations of each of the two possible spin orientations by

$$N_1=\sum_{j=1}^{N} |1,j\rangle\langle 1,j|\,, \tag{2.29}$$

$$N_2=\sum_{j=1}^{N} |2,j\rangle\langle 2,j| \tag{2.30}$$

and note that because the total number of spins in the crystal is fixed to be N, one must have

$$N_1+N_2=N\,. \tag{2.31}$$

Instead of working directly with the reduced-system density operator $\sigma(t)$, we find it convenient to use a representation in which only c-number (not operator) equations appear. To this end, we next define a c-number quasiprobability distribution function[9-11] of c-number variables s, n_2, and s^*, by

$$\begin{aligned}P(s,n_2,s^*,t)\equiv \mathrm{Tr}[\sigma(t)\,\delta(s-S^{-})\,\delta(n_2-N_2)&\\ \times\delta(s^*-S^{+})]\,,&\end{aligned} \tag{2.32}$$

where the δ functions are defined formally by integral representations

$$\delta(n_2-N_2)\equiv(2\pi)^{-1}\int_{-\infty}^{\infty} dx\, e^{ix(n_2-N_2)}\,, \tag{2.33}$$

and similarly, for the other two.

We note again that the quantities s, n_2, and s^* are not operators, but rather are c numbers, so that the distribution function $P(s,n_2,s^*,t)$ is also a c-number function. The significant point is that in (2.32) we have chosen to order the (noncommuting) operators S^{-}, N_2, and S^{+} in a particular way; that is, in every term of (2.32) obtained by using the expansions of the δ functions, all of the S^{-} operators are to the left of all the N_2 operators, which in turn are to the left of the S^{+} operators. We have chosen this order arbitrarily; choosing a different order neither changes the final form of the equation that we solve nor simplifies the calculation noticeably.

The quasiprobability distribution function introduced here is closely related to the Glauber $P(\alpha)$ distribution[12] which was introduced in order to study statistical properties of the electromagnetic field. The mathematical properties of $P(\alpha)$ have been extensively studied.[13] The system being studied here requires three operators, so the distribution function (2.32) is a generalization of the $P(\alpha)$ function. The utility of introducing such a distribution comes from the fact that since we are dealing with a system involving the set (S^-, N_2, S^+) of noncommuting operators, we may use their commutation relations to put every product of operators which may occur into a particular chosen order (namely, every S^- to the left of every N_2, which in turn is to the left of every S^+). Having thus chosen a conventional ordering, we may associate with an arbitrary operator $\chi(S^-, N_2, S^+)$ (we envisage χ to be sums of products of the three basic operators) a c-number function $\chi^{(c)}(s, n_2, s^*)$ of the c-number variables s, n_2, and s^*,

$$\chi(S^-, N_2, S^+) = C\chi^{(c)}(s, n_2, s^*) \tag{2.34}$$

or

$$\chi(S^-, N_2, S^+) = \int ds\, dn_2 ds^* \chi^{(c)}(s, n_2, s^*)\,\delta(s - S^-) \times \delta(n_2 - N_2)\,\delta(s^* - S^+)\ , \tag{2.35}$$

where the δ functions are defined by (2.33). The operator C instructs one to replace in $\chi^{(c)}(s, n_2, s^*)$ each of the variables s, n_2, and s^* by the corresponding operators S^-, H_2, and S^+—keeping the operators in the chosen order by doing the integrations in (2.35) in sequence. One can similarly define a c-number distribution function associated with the density operator $\sigma(t)$ by (2.32) so that in order to calculate the average value of the operator $\chi(S^-, N_2, S^+)$ we need

$$\begin{aligned}\langle\chi(S^-, N_2, S^+)\rangle &= \mathrm{Tr}[\sigma(t)\chi(S^-, N_2, S^+)]\\ &= \mathrm{Tr}[\sigma(t) \int ds\, dn_2 ds^* \chi^{(c)}(s, n_2, s^*) \\ &\qquad \times \delta(s - S^-)\,\delta(n_2 - N_2)\,\delta(s^* - S^+)] \\ &= \int ds\, dn_2 ds^* \chi^{(c)}(s, n_2, s^*) \\ &\qquad \times \mathrm{Tr}[\sigma(t)\,\delta(s - S^-)\,\delta(n_2 - N_2)\,\delta(s^* - S^+)] \\ &= \int ds\, dn_2 ds^* \chi^{(c)}(s, n_2, s^*)\, P(s, n_2, s^*, t)\ .\end{aligned} \tag{2.36}$$

The interpretation of the $P(s, n_2, s^*, t)$ as a classical probability distribution may not always be justified. Studies[13] of the Glauber $P(\alpha)$ distribution have shown that it may possess singularities as well as take on negative values for some range of its arguments. However, we shall use (2.32) in expressions of the type (2.36) and shall see that in certain cases it does indeed have the properties of a classical distribution function.

We shall focus our attention only on the z component of the magnetization and therefore we need not use a distribution of the three components of the magnetization, as defined by (2.32). Although the Ising Hamiltonian does not involve transverse components of spin operators, the interaction V with the reservoir does. It is not apparent that the density-operator equation may be fully represented only in terms of the z component of magnetization; a more general equation of motion for $P(s, n_2, s^*, t)$ has been derived as noted below. We thus define

$$P(n_2, t) \equiv \mathrm{Tr}[\sigma(t)\,\delta(n_2 - N_2)] \tag{2.37}$$

and note that the equation of motion of this quantity is clearly

$$\dot{P}(n_2, t) = \mathrm{Tr}[\mathring{\sigma}(t)\,\delta(n_2 - N_2)]\ . \tag{2.38}$$

We may use (2.18) for the ratio of change of the reduced-system density operator and thus obtain the following equation:

$$\begin{aligned}\mathring{P}(n_2, t) = -2\int_{-\infty}^{t} dt' \sum_{j=1}^{N} \mathrm{Tr}\{\sigma(t)\,[F_1(t, t')\,\delta(n_2 - N_2) S_j^+(t) S_j^-(t') + F_2(t, t')\,\delta(n_2 - N_2) S_j^-(t) S_j^+(t') \\ - F_2(t', t) S_j^-(t')\,\delta(n_2 - N_2) S_j^+(t) - F_1(t', t) S_j^+(t')\,\delta(n_2 - N_2) S_j^-(t)]\}.\end{aligned} \tag{2.39}$$

To evaluate the time dependence of the spin operators in (2.39) we shall consider here only the case of the uniform-coupling Ising model in which each spin interacts with the same strength with every other spin in the system. That is, we set $J_{ij} = J$ for all i, j in the system Hamiltonian (2.2), which then may be written

$$H_S = -2J(S^z)^2 - 2\mu H_0 S^z\ , \tag{2.40}$$

From (2.8) and (2.40) we then obtain the following results for the spin operators:

$$\begin{aligned}S_j^+(t) &= e^{-i\hbar^{-1}\alpha^- t} S_j^+(0) \\ &= S_j^+(0) e^{-i\hbar^{-1}\alpha^+ t}\ ,\end{aligned} \tag{2.41a}$$

$$\begin{aligned}S_j^-(t) &= e^{i\hbar^{-1}\alpha^+ t} S_j^-(0) \\ &= S_j^-(0)\, e^{i\hbar^{-1}\alpha^- t}\ ,\end{aligned} \tag{2.41b}$$

where

$$\alpha^- \equiv 4JS^z - 2J + 2\mu H_0 , \tag{2.42a}$$

$$\alpha^+ \equiv 4JS^z + 2J + 2\mu H_0 . \tag{2.42b}$$

Using these results and writing the equation in terms of the atomic operators (2.22)–(2.26), Eq. (2.39) becomes

$$\begin{aligned}\dot{P}(n_2, t) = -2\int_{-\infty}^{t} dt' \,\mathrm{Tr}\Big[\sigma(t)\Big(& F_1(t, t')e^{-i\hbar^{-1}\alpha^-(t-t')}\,\delta(n_2-N_2)N_1 + F_2(t,t')\,e^{i\hbar^{-1}\alpha^+(t-t')}\,\delta(n_2-N_2)N_2 \\ & - \sum_{j=1}^{N} F_2(t', t)\, e^{i\hbar^{-1}\alpha^+ t'}\,|2, j\rangle\langle 1, j\,|\,\delta(n_2-N_2)\,|\,1, j\rangle\langle 2, j\,|e^{i\hbar^{-1}\alpha^- t} \\ & - \sum_{j=1}^{N} F_1(t', t)e^{-i\hbar^{-1}\alpha^- t'}\,|1, j\rangle\langle 2, j\,|\delta(n_2-N_2)\,|\,2, j\rangle\langle 1, j\,|e^{-i\hbar^{-1}\alpha^-}\Big)\Big] .\end{aligned} \tag{2.43}$$

Further reduction of (2.43) is carried out in the Appendix. The final result is the following equation of motion for the magnetization distribution function, which we write in terms of the variable m defined by

$$m \equiv \tfrac{1}{2}N - n_2 : \tag{2.44}$$

$$\begin{aligned}\dot{P}(m, t) = \{&\tfrac{1}{2}N[(e^{-\partial/\partial m} - 1)\,\boldsymbol{\Gamma}_2(m) + (e^{\partial/\partial m} - 1)\,\boldsymbol{\Gamma}_1(m)] \\ &+ [(e^{\partial/\partial m} - 1)\boldsymbol{\Gamma}_1(m) - (e^{-\partial/\partial m} - 1) \\ &\times \boldsymbol{\Gamma}_2(m)]m\}P(m, t) .\end{aligned} \tag{2.45}$$

Here the exponentials of the differential operators imply that (2.45) is an infinite-order differential equation. The two damping functions $\boldsymbol{\Gamma}_1(m)$ and $\boldsymbol{\Gamma}_2(m)$ are defined by Eqs. (A5) and (A6) of the Appendix in general terms. We shall consider specific models of reservoir systems for which these functions will be explicitly calculated. We note, as is shown in the Appendix, that these two quantities obey a detailed balance relation in general.

Equation (2.45) is a special case of a somewhat more general equation obtained by one of the authors in his thesis.[14] The full distribution function $P(s, n_2, s^*, t)$, defined by (2.32), we found to satisfy the following equation of motion:

$$\begin{aligned}\dot{P}(s, n_2, s^*, t) = {} & \frac{\partial}{\partial s^*}\,[\tfrac{1}{2}(\boldsymbol{\Gamma}_1 + \boldsymbol{\Gamma}_2)s^*] + \frac{\partial}{\partial s}\,[\tfrac{1}{2}(\boldsymbol{\Gamma}_1 + \boldsymbol{\Gamma}_2)\,s] + (e^{-\partial/\partial n_2} - 1)\,\boldsymbol{\Gamma}_1 n_1 + (e^{\partial/\partial n_2} - 1)\,\boldsymbol{\Gamma}_2 n_2 \\ & + \frac{\partial^2}{\partial s\partial s^*}\,\boldsymbol{\Gamma}_1 N + \frac{\partial}{\partial s^*}\,(e^{-\partial/\partial n_2} - 1)\,\boldsymbol{\Gamma}_1 s^* + \frac{\partial}{\partial s}\,(e^{-\partial/\partial n_2} - 1)\,\boldsymbol{\Gamma}_1 s + \frac{\partial^3}{\partial s\partial s^{*2}}\,\boldsymbol{\Gamma}_1 s^* + \frac{\partial^3}{\partial s^2\partial s^*}\,\boldsymbol{\Gamma}_1 s \\ & + \frac{\partial^4}{\partial s^2\partial s^{*2}}\,e^{\partial/\partial n_2}\,\boldsymbol{\Gamma}_1 n_2\, P(s, n_2, s^*, t) .\end{aligned}$$

We may obtain (2.45) from this equation by integrating over the transverse variables, that is,

$$P(n_2, t) = \int ds\, ds^* P(s, n_2, s^*, t) ,$$

and then changing to the variable m.

III. CRITICAL FLUCTUATIONS

A useful approximation to the full equation of motion (2.45) for the longitudinal component of the magnetization may be formulated: There is ample evidence[15,16] that large fluctuations exist in the magnetization for temperatures near the Curie temperature T_C, above which the system is paramagnetic. This means that near T_C the function $P(m)$ of (2.45) should be a relatively slowly varying function of m so as to increase the fluctuations, of which the mean square fluctuation

$$\sigma_m^2 = \langle m^2\rangle - \langle m\rangle^2 \tag{3.1}$$

is one measure. Therefore, in this region we expect that the higher-order derivatives, obtained by expanding the exponential operators in (2.45), will rapidly become smaller then the lowest-order terms. It is thus reasonable to approximate the complete equation by retaining only the lowest-order terms.

Expanding the exponential factors up to second derivatives only, we obtain from (2.45)

$$\begin{aligned}\dot{P}(m, t) = \Big(&\frac{\partial}{\partial m}\,\{\tfrac{1}{2}N[\boldsymbol{\Gamma}_1(m) - \boldsymbol{\Gamma}_2(m)] + [\boldsymbol{\Gamma}_1(m) + \boldsymbol{\Gamma}_2(m)]m\} \\ &+ \frac{1}{2}\,\frac{\partial^2}{\partial m^2}\,\{\tfrac{1}{2}N[\boldsymbol{\Gamma}_1(m) + \boldsymbol{\Gamma}_2(m)] \\ &+ [\boldsymbol{\Gamma}_1(m) - \boldsymbol{\Gamma}_2(m)]m\}\Big)\,P(m, t) .\end{aligned} \tag{3.2}$$

Equations of the form

$$\frac{\partial}{\partial t} F(x, t) = \left(-\frac{\partial}{\partial x} A + \frac{\partial^2}{\partial x^2} B \right) F(x, t) \qquad (3.3)$$

are referred to as Fokker–Planck equations[17] with drift coefficient A and diffusion coefficient B. It is seen that our equation is of this general type, but with nonconstant drift and diffusion coefficients. From the definition

$$\langle \dot{m}(t) \rangle = \int dm\, m \dot{P}(m, t) \,, \qquad (3.4)$$

we obtain from Eq. (3.2) the equation of motion for the average magnetization,

$$\langle \dot{m}(t) \rangle = \langle \tfrac{1}{2} N [\mathbf{\Gamma}_2(m) - \mathbf{\Gamma}_1(m)] \rangle - \langle [\mathbf{\Gamma}_1(m) + \mathbf{\Gamma}_2(m)] m \rangle . \qquad (3.5)$$

Here we have integrated by parts and assumed that the distribution function and its derivative vanish at the extreme limits of integration.

To proceed further we must know what the functions $\mathbf{\Gamma}_1$ and $\mathbf{\Gamma}_2$ are. Although, as stated in (A7), the ratio of these quantities is known from the general forms (A5) and (A6), their explicit forms depend upon the details of the reservoir chosen to represent the thermal bath to which the ferromagnetic spin system is coupled. We therefore introduce two specific reservoir models and study the dynamical properties of the spin system as it interacts with each one in turn.

Oscillator-Reservoir Case

We first consider the reservoir to be composed of an infinite array of harmonic oscillators of a broad distribution of frequencies whose density operator is the canonical-ensemble thermal equilibrium density operator. Thus, we identify the reservoir operators O_α, $O_\alpha^\dagger$ of (2.4) with the boson absorption and creation operators b_j, $b_j^\dagger$ which obey the commutation relations

$$[b_j, b_{j'}^\dagger] = \delta_{jj'}, \quad [b_j, b_{j'}] = [b_j^\dagger, b_{j'}^\dagger] = 0. \qquad (3.6)$$

Thus the reservoir Hamiltonian is

$$H_R = \sum_j \hbar\omega_j\, b_j^\dagger b_j \,. \qquad (3.7)$$

Taking the thermal equilibrium reservoir density operator (2.14) and the general formula (A6) for $\mathbf{\Gamma}_2$, we have

$$\tfrac{1}{2}\mathbf{\Gamma}_2(\omega) = g^2 \operatorname{Re} \int_{-\infty}^{t} dt' \sum_{j=1}^{N} [\bar{n}(\omega_j) + 1] e^{i(\omega - \omega_j)(t - t')} , \qquad (3.8)$$

where

$$\bar{n}(\omega_j) = (e^{\beta\hbar\omega_j} - 1)^{-1} \qquad (3.9)$$

is the average thermal occupation number of phonons in the jth mode. We assume that the number of degrees of freedom (modes) of the reservoir is large enough to enable us to convert the sum in (3.8) into an integral by means of a density-of-states function $\Omega(\omega_j)$:

$$\begin{aligned} \tfrac{1}{2}\mathbf{\Gamma}_2(\omega) &= g^2 \operatorname{Re} \int_{-\infty}^{t} dt' \int_0^\infty d\omega_j\, \Omega(\omega_j) \\ &\quad \times [\bar{n}(\omega_j) + 1]\, e^{i(\omega - \omega_j)(t - t')} \\ &= g^2 \operatorname{Re} \int_{-\infty}^{t} dt' \int_{\omega}^{-\infty} (-du)\, \Omega(\omega - u) \\ &\quad \times [\bar{n}(\omega - u) + 1] e^{iu(t - t')} \\ &\simeq g^2 \operatorname{Re} \int_{-\infty}^{t} dt' \int_{-\infty}^{\infty} du\, \Omega(\omega - u) \\ &\quad \times [\bar{n}(\omega - u) + 1] e^{iu(t - t')} . \end{aligned} \qquad (3.10)$$

Arguing that the density-of-states function and the thermal occupation number are slowly varying functions of their arguments compared to the exponential function in (3.10), we approximately calculate the integral by extracting those two functions from under the integral sign—evaluated at the point of least rapid variation of the exponential, which occurs for $u = 0$—and thus obtain

$$\mathbf{\Gamma}_2(\omega) = \gamma^{(a)} [\bar{n}(\omega) + 1] \,, \qquad (3.11)$$

where we denote by $\gamma^{(a)}$ the spontaneous emission rate.

We obtain $\mathbf{\Gamma}_1$ in a similar calculation from its defining relation (A5):

$$\mathbf{\Gamma}_1(\omega) = \gamma^{(a)} \bar{n}(\omega) \,. \qquad (3.12)$$

We may now return to the equation of motion for the magnetization distribution function. However, first we note the following connection between the two-level energy separation $\hbar\omega$ and the magnetization which follows from the Hamiltonian (2.40):

$$\hbar\omega = 2\mu (H_0 + \lambda M) \,, \qquad (3.13)$$

where H_0 is the uniform external field, λ is the Weiss internal-field constant, and M is the (correctly dimensioned) magnetization given by

$$M = 2\mu V^{-1} m \,, \qquad (3.14)$$

where μ is the magnetic moment of each spin and V is the volume of the system. Using these relations in (3.2), we obtain

$$\dot{P}^{(a)}(M, t) = \left[\frac{\partial}{\partial M} \gamma^{(a)} \left(-\frac{N\mu}{V} + M \coth[\beta\mu(H_0 + \lambda M)] \right) + \frac{\mu}{V} \frac{\partial^2}{\partial M^2} \gamma^{(a)} \left(\frac{N\mu}{V} \coth[\beta\mu(H_0 + \lambda M)] - M \right) \right] P(M, t) \,. \qquad (3.15)$$

We have used (3. 9) to obtain

$$\Gamma_1(\omega) - \Gamma_2(\omega) = -\gamma^{(a)} , \qquad \Gamma_1(\omega) + \Gamma_2(\omega) = \gamma^{(a)} \coth(\tfrac{1}{2}\beta\hbar\omega) . \tag{3. 16}$$

From (3. 15) we see that the mean magnetization satisfies the equation of motion

$$\frac{1}{\gamma^{(a)}} \frac{\partial}{\partial t} \langle M(t)\rangle = N\mu/V - \langle M(t)\rangle \times \coth\{\beta\mu[H_0 + \lambda \langle M(t)\rangle]\} , \tag{3. 17}$$

where we have neglected fluctuation effects in this equation by writing the average of a product of $M(t)$ factors as the product of the average value $\langle M(t)\rangle$. We note at once that the steady-state (time-independent) solution of (3. 17) yields the molecular-field equation of state:

$$0 = N\mu/V - \langle M\rangle \coth[\beta\mu(H_0 + \lambda\langle M\rangle)] , \qquad \langle M\rangle = (N/V)\tanh[\beta\mu(H_0 + \lambda\langle M\rangle)] . \tag{3. 18}$$

We define a dimensionless magnetization $x(t)$ by

$$x(t) \equiv M(t)\,(N\mu/V)^{-1} . \tag{3. 19}$$

We may similarly define a dimensionless magnetic field and inverse temperature by

$$h \equiv H_0[\lambda(N\mu/V)]^{-1}, \quad \tau \equiv T_c T^{-1}, \tag{3. 20}$$

where the transition temperatures T_C is given by

$$K_B T_C = \lambda N \mu^2 V^{-1} . \tag{3..21}$$

Returning to (3. 15), which we write in terms of the dimensionless variables (3. 19) and (3. 20) as

$$\dot{P}^{(a)}(x, t) = \left(\frac{\partial}{\partial x} \gamma^{(a)}\{x \coth[\tau(h+x)] - 1\} + N^{-1} \frac{\partial^2}{\partial x^2} \gamma^{(a)}\{\coth[\tau(h+x)] - x\}\right) \times P^{(a)}(x, t) , \tag{3. 22}$$

we may easily obtain its steady-state (time-independent) solution. Calling the drift and diffusion coefficients of (3. 22) A and B, respectively, as in (3. 3), we see that the steady-state solution $P^{(a)}(x)$ satisfies

$$0 = \frac{\partial}{\partial x}\left(-AP^{(a)}(x) + \frac{\partial}{\partial x} BP^{(a)}(x)\right). \tag{3. 23}$$

We note that the Fokker–Planck equation in general has the form of a conservation probability law:

$$\frac{\partial P}{\partial t} + \frac{\partial J}{\partial x} = 0 . \tag{3. 24}$$

Here J is a probability current. At steady state, provided we are restricted to a finite range of the variable (in our case $|x| \leqslant 1$), we must choose $J = 0$ in order not to violate this general conservation requirement.[17] Thus, the integration constant from (3. 23) must vanish, and we obtain the steady-state solution

$$P^{(a)}(x) = CB^{-1}(x) \exp[\int_0^x dx' A(x')B^{-1}(x')] \tag{3. 25}$$

$$= \frac{NC[\gamma^{(a)}]^{-1}}{\coth[\tau(h+x)] - x} \times \exp\left(-N\int_0^x dx' \frac{x'\coth[\tau(h+x')] - 1}{\coth[\tau(h+x')] - x'}\right), \tag{3. 26}$$

where C is a normalization constant (depending upon N, h, and τ). We note that we may write (2. 26) in the form

$$P^{(a)}(x) = C \exp\left[-N\left(\int_0^x dx' \frac{x'\coth[\tau(h+x)] - 1}{\coth[\tau(h+x')] - x'} - \frac{1}{N}\ln(\gamma^{(a)}\{\coth[\tau(h+x)] - x\})\right)\right]. \tag{3. 27}$$

Since N is an extremely large number ($N \sim 10^{23}$), it is a reasonable approximation to neglect the second term in (3. 27) in comparison with the first, so we take the steady-state solution of (3. 22) to be

$$P^{(a)}(x) = \dot{C} \exp\left(-N\int_0^x dx' \frac{x'\coth[\tau(h+x')] - 1}{\coth[\tau(h+x')] - x'}\right). \tag{3. 28}$$

We note that the first derivative of this function vanishes at points $x = x_0$, where x_0 satisfies the molecular-field equation of state (3. 18). Because of the great numerical value of N, the peak of the maximum is extremely sharp for low temperatures. However, since we neglected higher derivatives in obtaining our Fokker–Planck equation, this solution is not expected to have any validity in that region where it is so sharply peaked. However, for temperatures near $T_C(\tau \sim 1)$, we may expand the solution (2. 13) for small x (since the peak of the distribution occurs at the molecular-field value of x which will be small near T_C) to obtain

$$P^{(a)}(x) \simeq c \exp(-N\int_0^x dx' \{(1-\tau)x' + [\tau(1-\tau) + \tfrac{1}{3}\tau^3](x')^3\}) = c \exp\{-(K_B T)^{-1}[c(T - T_C)\tfrac{1}{2}m^2 + d'T\,\tfrac{1}{4}m^4]\} , \tag{3. 29}$$

where

$$c = K_B(N\mu^2)^{-1}, \qquad d' = [\tau^3 + 3\tau(1-\tau)]d, \qquad d = K_B(3N^3\mu^4)^{-1} . \tag{3. 30}$$

The thermodynamic theory of fluctuations[18,19] predicts that the probability density for the fluctuations of a thermodynamic variable η is given by

$$P(\eta)=C\,e^{-\phi(\eta)(K_BT)^{-1}}\,, \tag{3.31}$$

where $\phi(\eta)$ is the appropriate thermodynamic potential for the situation considered. Besides having this general form, (3. 29) also has the form for the free energy which the Landau theory of a second-order phase transition[18] predicts; namely, Landau theory predicts that near such a phase transition $\phi(\eta)$ is of the form

$$\phi(\eta)=\phi_0+\alpha(T-T_C)\eta^2+\Delta\eta^4\,, \tag{3.32}$$

where η is the order parameter of the transition. That is, η is a thermodynamic variable which is assumed to vanish above the transition temperature T_C (corresponding to the disordered state) and be nonzero below T_C (in the ordered state). In the case of a ferromagnet the order parameter is the magnetic moment M. The precise values of the constants α and Δ (α is independent of temperature, while Δ may be a function of temperature) are not predicted by the Landau theory. However, in fact the constants c and d are precisely those which come from expanding the free energy, which leads one to the molecular-field equation of state. The appropriate free energy is that obtained from the Bragg–Williams[20] approximation, which corresponds to making a mean-field approximation. Thus the solution of the Fokker–Planck equation (3. 15), or (3. 22), for the distribution function of the magnetization yields the correct free energy of the system as its steady-state value in the region where this approximation to the exact equation (2. 45) is expected to be valid.

Spin-Reservoir Case

We turn next to a different model for the thermal reservoir with which the ferromagnetic system is in contact. For the reservoir we take an assembly of two-level systems possessing a broad distribution of energy-level splittings $\hbar\omega_K$. The reservoir Hamiltonian H_R now becomes

$$H_R=\sum_K \hbar\omega_K \mathcal{S}_K^z\,, \tag{3.33}$$

where the spin-$\frac{1}{2}$ angular momentum operators of the reservoir obey the usual commutation relations

$$[\mathcal{S}_K^+,\ \mathcal{S}_{K'}^z]=-\delta_{KK'}\mathcal{S}_K^+\,, \tag{3.34}$$

$$[\mathcal{S}_K^-,\ \mathcal{S}_{K'}^z]=\delta_{KK'}\mathcal{S}_K^-\,, \tag{3.35}$$

$$\mathcal{S}_K^\pm\equiv\mathcal{S}_K^x\pm i\mathcal{S}_K^y\,. \tag{3.36}$$

Once again it is convenient to relate these to atomic operators via

$$\mathcal{S}_K^+=(\,|2\rangle\langle 1|\,)_K\,,\quad \mathcal{S}_K^-=(\,|1\rangle\langle 2|\,)_K\,, \tag{3.37}$$

where the energies of the two states obey

$$\epsilon_2(K)-\epsilon_1(K)=\hbar\omega_K\geqslant 0\,. \tag{3.38}$$

We thus identify the reservoir operators O_α, $O_\alpha^\dagger$ of (2. 4) with $\mathcal{S}_K^-$, $\mathcal{S}_K^+$ and take the reservoir density operator $f_0(H_R)$ to be

$$\begin{aligned} f_0(H_R)&=e^{-\beta H_R}(\mathrm{Tr}_R e^{-\beta H_R})^{-1}\\ &=\prod_K \frac{1}{z_K}\exp\{-\beta[\epsilon_1(K)\,|1,K\rangle\\ &\qquad\times\langle 1,K|+\epsilon_2(K)\,|2,K\rangle\langle 2,K|]\}\,, \end{aligned} \tag{3.39}$$

where

$$z_K=e^{-\beta\epsilon_1(K)}+e^{-\beta\epsilon_2(K)}\,. \tag{3.40}$$

Thus, from (1. 6) we may calculate $\mathbf{\Gamma}_2$:

$$\begin{aligned} \mathbf{\Gamma}_2(\omega)&=2g^2\,\mathrm{Re}\int_{-\infty}^{t}dt'\\ &\quad\times\int_0^\infty d\omega_K\,\Omega(\omega_K)\bar{n}_1(\omega_K)e^{i(\omega-\omega_K)(t-t')}\,, \end{aligned} \tag{3.41}$$

where

$$\begin{aligned} \bar{n}_1(\omega_K)&=e^{-\beta\epsilon_2(K)}[(e^{-\beta\epsilon_1(K)}+e^{-\beta\epsilon_2(K)})]^{-1}\\ &=(1+e^{\beta\hbar\omega_K})^{-1}\,. \end{aligned} \tag{3.42}$$

Once again assuming that the reservoir density of states $\Omega(\omega_K)$ and $\bar{n}_1(\omega_K)$ are slowly varying in comparison with the exponential function in (3. 41), we obtain

$$\mathbf{\Gamma}_2(\omega)=2\pi g^2\Omega(\omega)\bar{n}_1(\omega)=\gamma^{(b)}\bar{n}_1(\omega)\,, \tag{3.43}$$

where $\gamma^{(b)}$ is the spontaneous decay rate.

In a similar fashion we may obtain $\mathbf{\Gamma}_1$ from (1. 5):

$$\begin{aligned} \mathbf{\Gamma}_1(\omega)&=2g^2\,\mathrm{Re}\int_{-\infty}^{t}dt'\\ &\quad\times\int_0^\infty d\omega_K\,\Omega(\omega_K)\bar{n}_2(\omega_K)e^{i(\omega_K-\omega)(t-t')}\,, \end{aligned} \tag{3.44}$$

where

$$\bar{n}_2(\omega_K)=e^{-\beta\epsilon_2(K)}(e^{-\beta\epsilon_1(K)}+e^{-\beta\epsilon_2(K)})^{-1}=(1+e^{-\beta\hbar\omega_K})^{-1}\,. \tag{3.45}$$

Thus we obtain, from (3.44),

$$\mathbf{\Gamma}_1(\omega)=2\pi g^2\Omega(\omega)\bar{n}_2(\omega)=\gamma^{(b)}\bar{n}_2(\omega)\,. \tag{3.46}$$

Returning to the general equation of motion for the magnetization distribution function (3. 2), we obtain for this spin reservoir, using (3. 43) and (3. 46) and the molecular-field-like form (3. 13) for $\hbar\omega$,

$$\begin{aligned} \dot{P}^{(b)}(M,t)=\bigg[&\frac{\partial}{\partial M}\gamma^{(b)}\left(M-\frac{N\mu}{V}\tanh[\beta\mu(H_0+\lambda M)]\right)\\ &+\frac{\mu}{V}\frac{\partial^2}{\partial M^2}\gamma^{(b)}\\ &\times\left(\frac{N\mu}{V}-M\tanh[\beta\mu(H_0+\lambda M)]\right)\bigg]P^{(b)}(M,t)\,, \end{aligned} \tag{3.47}$$

where we have used (3. 14) to define the magnetization M and the results for this reservoir

$$\Gamma_1(\omega)-\Gamma_2(\omega)=-\gamma^{(b)}\tanh(\tfrac{1}{2}\beta\hbar\omega)\,, \tag{3.48}$$

$$\Gamma_1(\omega)+\Gamma_2(\omega)=\gamma^{(b)}\,. \tag{3.49}$$

We see at once from (3. 47) that the equation of motion for the average magnetization is given for this case by

$$\frac{1}{\gamma^{(b)}}\frac{\partial}{\partial t}\langle M(t)\rangle=\frac{N\mu}{V}\tanh\{\beta\mu[H_0+\lambda\langle M(t)\rangle]\}-\langle M(t)\rangle\,, \tag{3.50}$$

where we have neglected fluctuation effects in this mean equation of motion by writing the average of a product of factors of $M(t)$ as the product of the average value $\langle M(t)\rangle$. We see immediately that the steady-state solution of (3. 50) is the molecular-field equation of state (3. 18). Therefore, although the equation of motion for the average magnetization for this thermal reservoir is different from the corresponding equation of motion for the phonon reservoir, Eq. (3. 17), both equations have the same steady-state solution.

In terms of the dimensionless variables x, h, and τ defined by (3. 19) and (3. 20), the Fokker–Planck equation (3. 47) may be written

$$\overset{\circ}{P}{}^{(b)}(x,t)=\left(\frac{\partial}{\partial x}\gamma^{(b)}\{x-\tanh[\tau(h+x)]\}+\frac{1}{N}\frac{\partial^2}{\partial x^2}\gamma^{(b)}\{1-x\tanh[\tau(h+x)]\}\right)\times P^{(b)}(x,t)\,. \tag{3.51}$$

The steady-state solution, from (3. 25), is therefore

$$P^{(b)}(x)=\frac{NC[\gamma^{(b)}]^{-1}}{1-x\tanh[\tau(h+x)]}\times\exp\left(-N\int_0^x dx'\,\frac{x'-\tanh[\tau(h+x')]}{1-x'\tanh[\tau(h+x')]}\right)\,, \tag{3.52}$$

where C is a normalization constant. We write (3. 52) in the form

$$P^{(b)}(x)=C\exp\left[-N\left(\int_0^x dx'\,\frac{x'-\tanh[\tau(h+x')]}{1-x'\tanh[\tau(h+x')]}-\frac{1}{N}\ln(\gamma^{(b)}\{1-x\tanh[\tau(h+x)]\})\right)\right]\,. \tag{3.53}$$

Since N is such an enormous number ($\sim 10^{23}$), we may neglect the second term in comparison to the first one in (3. 53). Thus we take as the steady-state solution to the equation of motion of the magnetization distribution function, Eq. (3. 47) or (3. 51),

$$P^{(b)}(x)=C\exp\left(-N\int_0^x dx'\,\frac{x'-\tanh[\tau(h+x')]}{1-x\tanh[\tau(h+x')]}\right)\,. \tag{3.54}$$

We note that, although the equations of motion for the magnetization distribution function are different for the two types of reservoirs, the steady-state solutions are in fact the same, as comparison of (3. 54) with (3. 28) immediately shows. Thus, although the time-dependent properties of the ferromagnetic spin system depend upon the detailed nature of the thermal reservoir with which it is in contact, the steady-state properties are the same. The mean magnetization at steady state for both types of reservoirs satisfies the molecular-field equation of state, and the steady-state magnetization distributions agree with the Landau form of the magnetic free energy for temperatures near the Curie temperature, and identically with the Bragg–Williams free energy when it is approximated near the transition temperature.

IV. DISCRETE REPRESENTATION AND BRAGG–WILLIAMS FREE ENERGY

As we have noted earlier, exponential operators such as those found in the full equation of motion for the magnetization distribution function, Eq. (2. 45), are in effect displacement operators acting on the functions of the variable m that are placed on their right-hand side. Noting that these include the damping functions $\Gamma_1(m)$ and $\Gamma_2(m)$, and the factor m itself as well as $P(m,t)$, we carry out the displacements to get

$$\begin{aligned}\overset{\circ}{P}(m,t)={}&\Gamma_2(m-1)\,[\tfrac{1}{2}N-(m-1)]\,P(m-1,t)\\&-\Gamma_2(m)\,[\tfrac{1}{2}N-m]P(m,t)\\&-\Gamma_1(m)\,[\tfrac{1}{2}N+m]P(m,t)\\&+\Gamma_1(m+1)\,[\tfrac{1}{2}N+m+1]P(m+1,t)\,.\end{aligned} \tag{4.1}$$

We have indicated explicitly that Γ_1 and Γ_2 depend on the variable m, since from their definitions (A5) and (A6) they are shown to depend on this quantity as well as the external field H_0. The displacement operators in (2. 45) thus act on Γ_1 and Γ_2 also and yield (4. 1).

The solution of (4. 1) corresponding to steady-state [$P(m)$ independent of time] may be easily found to be

$$\frac{P(m)}{P(m-1)}=\frac{\Gamma_2(m-1)}{\Gamma_1(m)}\,\frac{\tfrac{1}{2}N-(m-1)}{\tfrac{1}{2}N+m}\,, \tag{4.2a}$$

$$\frac{P(m+1)}{P(m)}=\frac{\Gamma_2(m)}{\Gamma_1(m+1)}\,\frac{\tfrac{1}{2}N-m}{\tfrac{1}{2}N+m+1}\,, \tag{4.2b}$$

where (4. 2a) corresponds to the balance of the first and third terms of (4. 1) and (4. 2b) to the balance of the second and fourth terms. It is seen that (4.2a) and (4.2b) are identical, in fact.

We imagine that m is a discrete variable which, by (2.44) and the fixed total number of spins, obeys the inequality

$$-\tfrac{1}{2}N \leq m \leq +\tfrac{1}{2}N \;. \tag{4.3}$$

We first consider the case $m > 0$. Thus, (4.2a) gives

$$P(m) = e^{2\beta(2Jm+\mu H_0-J)}\,[\tfrac{1}{2}N-m+1][\tfrac{1}{2}N+m]^{-1}$$

$$\times P(m-1)\;, \tag{4.4}$$

where we have used the detailed balance relation (A7) connecting Γ_1 and Γ_2:

$$\Gamma_2(m-1)\,[\Gamma_1(m)]^{-1} = e^{\beta[E(m-1)-E(m)]}\;, \tag{4.5}$$

where the energy of the system $E(m)$ is found for the uniform coupling version (2.40) of the Ising Hamiltonian to be

$$E(m) = -2Jm^2 - 2\mu H_0 m\;. \tag{4.6}$$

Thus, using (4.4) repeatedly, we obtain

$$P(m) = e^{\beta[4Jm+2\mu H_0-2J]}\,e^{\beta[4J(m-1)+2\mu H_0-2J]}\cdots\frac{\frac{1}{2}N-(m-1)}{\frac{1}{2}N+m}\cdots\frac{\frac{1}{2}N-0}{\frac{1}{2}N+1}\,P_0\;,$$

where P_0 is the value of $P(m=0)$. Thus

$$P(m) = \exp\left[\beta J\left(\sum_{n=1}^{m} n\right)\right]e^{\beta(\mu H_0-J/2)m}\,\frac{(\frac{1}{2}N)!\,(\frac{1}{2}N)(\frac{1}{2}N-1)\cdots[\frac{1}{2}N-(m-1)][\frac{1}{2}N-m]!}{[\frac{1}{2}N+m]\cdots[\frac{1}{2}N+1](\frac{1}{2}N)![\frac{1}{2}N-m]!}\,P_0\;.$$

Using

$$\sum_{n=1}^{m} n = \tfrac{1}{2}m(m+1)\;,$$

we thus obtain

$$P(m) = e^{\beta Jm^2/2+\beta\mu H_0 m}\,[(\tfrac{1}{2}N)!]^2[(\tfrac{1}{2}N+m)!]^{-1}$$

$$\times[(\tfrac{1}{2}N-m)!]^{-1}P_0\;. \tag{4.7}$$

Thus we have the complete solution. The constant P_0 (which is independent of m but will depend on the external field H_0 and the temperature β) can be obtained from the normalization requirement

$$\sum_{m=-N/2}^{+N/2} P(m) = 1\;. \tag{4.8}$$

Using the solution (4.7), if we compute

$$F(m) - F_0 = -\beta^{-1}\ln P(m)\;, \tag{4.9}$$

we find that $F(m)$ is exactly the free energy of an Ising-model ferromagnet as computed from the Bragg–Williams method[20] (we include all terms independent of m in F_0):

$$F(m) = 2Jm^2 - 2\mu H_0 m + NK_BT$$

$$\times\{\tfrac{1}{2}(1+2mN^{-1})\ln[\tfrac{1}{2}(1+2mN^{-1})]$$

$$+\tfrac{1}{2}(1-2mN^{-1})\ln[\tfrac{1}{2}(1-2mN^{-1})]\}\;. \tag{4.10}$$

Noting that our variable m still corresponds to a spin-$\frac{1}{2}$ system, we may change variables to model a two-level system with arbitrary value μ of magnetic moment by defining

$$M = 2\mu mV^{-1} = \mu V^{-1}(n_1-n_2)\;. \tag{4.11}$$

Defining a dimensionless variable x as the ratio of M to its saturation value $N\mu V^{-1}$, we obtain, from (4.10),

$$F(x) = -\tfrac{1}{2}JN^2x^2 - \mu H_0Nx + NK_BT\{\tfrac{1}{2}(1+x)$$

$$\times\ln[\tfrac{1}{2}(1+x)] + \tfrac{1}{2}(1-x)\ln[\tfrac{1}{2}(1-x)]\}\;, \tag{4.12}$$

$$x = M(N\mu)^{-1} = 2N^{-1}m\;. \tag{4.13}$$

Minimizing the free energy $F(x)$ with respect to x and solving for x, we obtain the conventional molecular-field equation of state

$$x = (N\mu/V)^{-1}M$$

$$= \tanh\{(K_BT)^{-1}[\mu H_0 + NJ(N\mu/V)^{-1}M]\}\;. \tag{4.14}$$

Referring to (4.16), if we set $H_0 = 0$ and assume that $x \ll 1$, we may expand the logarithmic factors in order to approximate the expression. Thus, keeping terms up to the fourth degree, we get

$$F(x) \simeq \tfrac{1}{2}(NK_BT - JN^2)x^2 + (12)^{-1}NK_BT\,x^4$$

$$= NK_B\,(N\mu^2)^{-1}\,(T - K_B^{-1}NJ)\,\tfrac{1}{2}M^2$$

$$+\tfrac{1}{3}NK_B(N^4\mu^4)^{-1}\tfrac{1}{4}M^4 \tag{4.15}$$

using the definition (4.13) for x. If we make the identification

$$T_C = K_B^{-1}NJ\;, \tag{4.16}$$

which in view of the definition (3.21) of T_C relates the Weiss internal-field constant λ to the exchange energy J via

$$\lambda V^{-1}\mu^2 = J\;, \tag{4.17}$$

we see that the Bragg–Williams free energy becomes, near T_C,

$$F(m)=[K_B(N\mu^2)^{-1}](T-T_C)\,\tfrac{1}{2}M^2 + [K_B(3N^2\mu^4)^{-1}]\,T\,\tfrac{1}{4}M^4 = c(T-T_C)\,\tfrac{1}{2}M^2 + dT\,\tfrac{1}{4}M^4\,, \tag{4.18}$$

which is exactly like the expression found by expanding the solutions of the Fokker–Planck equations near T_C, Eq. (3.29) [the constant d differs negligibly from the constant d' of (3.30) for $\tau \sim 1$].

Although we have not obtained the complete time-dependent solution of the equation of motion of the magnetization distribution function, Eq. (4.1), we may consider the equation for the mean magnetization,

$$\langle \dot{m}(t)\rangle = \sum_{m=-N/2}^{+N/2} m\dot{P}(m,t)\,. \tag{4.19}$$

Using the equation of motion (4.1), we may easily evaluate the various terms implied by (4.19).

Hence, we obtain from (4.19)

$$\langle \dot{m}(t)\rangle = \tfrac{1}{2}N\langle \Gamma_2(m)-\Gamma_1(m)\rangle - \langle m[\Gamma_2(m)+\Gamma_1(m)]\rangle\,. \tag{4.20}$$

Thus we see that the equation of motion (3.5) of the average magnetization deduced from the Fokker–Planck equation for the distribution function agrees exactly with the equation for the mean motion (4.20) which followed from the complete equation of motion for the distribution function.

We have therefore shown that the steady-state solution of the full equation of motion for the magnetization distribution function, Eq. (2.45), yields the same free energy (for all temperatures) as the Bragg–Williams approximation. We note that nowhere in our equation-of-motion method did we have to use any of the statistical counting procedures usually employed in the derivation of the Bragg–Williams free energy.

V. CONCLUSION

We have treated the problem of a uniform-coupling Ising-model ferromagnet coupled to a thermal reservoir using methods previously applied to problems in laser physics. Beginning with a reversible density-operator equation of motion, we have derived an irreversible master equation for the reduced-system density operator. The operator master equation was then transformed into a c-number differential-difference equation for the magnetization quasiprobability distribution function. For temperatures near the ferromagnetic transition temperature, this equation reduced to an equation of the Fokker–Planck type with drift and diffusion terms which depended upon the details of the reservoir system to which the ferromagnetic spin system was coupled. The steady-state solution of the Fokker–Planck equation was shown to yield the Landau form of the magnetic free energy, while the difference equation was then shown to yield the Bragg–Williams free energy at all temperatures below T_C. Thus, the steady-state results—which were shown to be independent of the details of the reservoir—yielded the molecular-field magnetic equation of state, a result known to be correct for the uniform-coupling Ising model.

The dynamical evolution of the system was found to depend upon the details of the reservoir system with which it is in thermal contact. The damping functions reflect this reservoir dependence. Two general reservoir models were studied in which these functions were explicitly found, aside from zero-temperature transition rates which depend upon such specific reservoir details as density-of-states functions. In particular, it may be seen from Eqs. (3.17) and (3.50) that the zero-temperature transition rates $\gamma^{(a)}$ and $\gamma^{(b)}$ must be, respectively, odd and even functions of the magnetization (for $H_0 \equiv 0$); otherwise the equations do not have the correct symmetry properties under reflections through the xy plane ($M \to -M$). The dependence of the damping functions upon the instantaneous magnetization of the system leads to nonlinear equations for the time evolution of the mean magnetization.

APPENDIX

We shall give here details of the manipulation used in obtaining Eq. (2.45) from (2.43). First we examine the operator part of the first term of (2.43):

$$t_1 = \mathrm{Tr}[\sigma(t)\,e^{-i\hbar^{-1}2J(N-2n_2)(t-t')}\,\delta(n_2-N_2)(N-n_2)]\,. \tag{A1}$$

We rewrite the δ function as

$$\delta(n_2-N_2)=e^{-N_2(\partial/\partial n_2)}\,\delta(n_2) \tag{A2}$$

and replace the operator N_2 by the c number n_2' and a δ function:

$$t_1=\int dn_2'\,\mathrm{Tr}\{\sigma(t)\,e^{-i\hbar^{-1}2J(N-2n_2')(t-t')} \times e^{-n_2'(\partial/\partial n_2)}\,\delta(n_2)(N-n_2')\,\delta(n_2'-N_2)\} = \int dn_2'\,e^{-i\hbar^{-1}2J(N-2n_2')(t-t')}\,\delta(n_2'-n_2)(N-n_2') \times \mathrm{Tr}[\sigma(t)\,\delta(n_2'-N_2)]\,. \tag{A3}$$

Evidently the last term yields $P(n_2', t)$ by definition, and doing the n_2' integration yields

$$t_1=e^{-i\hbar^{-1}2J(N-2n_2)}(N-n_2)\,P(n_2,t)\,. \tag{A4}$$

We now define the damping function Γ_1 by

$$\tfrac{1}{2}\Gamma_1[2J(N-2n_2)-2J+2\mu H_0]=\mathrm{Re}\int_{-\infty}^{t}dt'\,F_1(t,t')\exp\{-i\hbar^{-1}[2J(N-2n_2)-2J+2\mu H_0](t-t')\}$$

$$=g^2\,\mathrm{Re}\int_{-\infty}^{t}dt'\Big\langle\sum_\alpha O_\alpha(t)\sum_\beta O_\beta^\dagger(t')\Big\rangle_R\exp\{-i\hbar^{-1}[2J(N-2n_2)-2J+2\mu H_0](t-t')\}\ . \qquad (A5)$$

Evidently the reduction of the second term in (2.43) proceeds analogously and leads to definition of the following damping function:

$$\tfrac{1}{2}\Gamma_2[2J(N-2n_2)+2J+2\mu H_0]=\mathrm{Re}\int_{-\infty}^{t}dt'\,F_2(t,t')\exp\{i\hbar^{-1}[2J(N-2n_2)+2J+2\mu H_0](t-t')\}$$

$$=g^2\,\mathrm{Re}\int_{-\infty}^{t}dt'\Big\langle\sum_\alpha O_\alpha^\dagger(t)\sum_\beta O_\beta^\dagger(t')\Big\rangle_R\exp\{i\hbar^{-1}[2J(N-2n_2)+2J+2\mu H_0](t-t')\}\ . \qquad (A6)$$

These damping functions are functions of the total z component of the magnetization $m=\frac{1}{2}N-n_2$. From the general definitions one can easily show that the following detailed balance relation holds:

$$\Gamma_1(m)[\Gamma_2(m-1)]^{-1}=e^{\beta[E(m)-E(m-1)]}\ , \qquad (A7)$$

where, from the uniform-coupling Ising Hamiltonian (2.40),

$$E(m)=-2Jm^2-2\mu H_0 m\ . \qquad (A8)$$

To treat the third term of Eq. (2.43) we consider the following object:

$$t_3=\sum_{j=1}^{N}|2,j\rangle\langle 1,j|\delta(n_2-N_2)|1,j\rangle\langle 2,j|\ . \qquad (A9)$$

Using (A2), we have

$$t_3=\sum_j|2,j\rangle\langle 1,j|e^{-N_2(\partial/\partial n_2)}|1,j\rangle\langle 2,j|\delta(n_2)$$

$$=\sum_j|2,j\rangle\langle 1,j|\prod_K T_K|1,j\rangle\langle 2,j|\delta(n_2)\ , \qquad (A10)$$

where

$$T_K=e^{-|2,K\rangle\langle 2,K|(\partial/\partial n_2)}\ . \qquad (A11)$$

Since the operators for different spins commute, we may write (A10) as

$$t_3=\prod_{K\neq j}T_K\sum_j|2,j\rangle\langle 1,j|T_j|1,j\rangle\langle 2,j|\delta(n_2)$$

$$=\prod_{K\neq j}T_K\sum_j\{|2\rangle\langle 1|e^{-|2\rangle\langle 2|(\partial/\partial n_2)}|1\rangle$$

$$\times\langle 2|e^{|2\rangle\langle 2|(\partial/\partial n_2)}\}_j\,e^{|2,j\rangle\langle 2,j|(\partial/\partial n_2)\delta(n_2)}\ . \qquad (A12)$$

Expanding the exponential factors in (A12), one has

$$e^{\pm|2\rangle\langle 2|(\partial/\partial n_2)}=1+|2\rangle\langle 2|(e^{\pm(\partial/\partial n_2)}-1)\ . \qquad (A13)$$

Carrying out these operations, one finally obtains

$$t_3=e^{\partial/\partial n_2}N_2\,\delta(n_2-N_2)\ . \qquad (A14)$$

The remaining part of the third term of (2.43) is simple, since only the operator N_2 is involved. Proceeding as in t_1 above, we easily obtain this term as well as the fourth term. Collecting these results, we obtain Eq. (2.45) of the text.

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Self-Consistent Calculation of Energy Bands in Ferromagnetic Nickel*

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A self-consistent calculation of energy bands in ferromagnetic nickel has been made using the tight-binding method. Eighty-nine points in $\frac{1}{48}$th of the Brillouin zone were used to determine the charge density in the final stages of an iterative procedure. The Kohn–Sham–Gaspar exchange was employed. The approximately self-consistent potential was used to compute energy levels at 505 regularly spaced points in $\frac{1}{48}$th of the Brillouin zone plus certain extra points close to the Fermi surface. Results are given for the density of states, the Fermi surface, the charge density, and the spin density. A magneton number of 0.58 was determined.

I. INTRODUCTION

This paper is an extension of a previous calculation of energy bands in ferromagnetic nickel using the tight-binding method[1] (Paper I). The work described in I was based on a crystal potential constructed from a superposition of overlapping neutral-atom charge densities, the atoms being in the $d^9 s^1$ configuration. The present calculation obtains an approximately self-consistent potential based on a sampling of the charge density at 89 points in $\frac{1}{48}$th of the Brillouin zone. The procedure for achieving self-consistency has been described elsewhere.[2] Exchange has been included according to the $X\alpha$ method of Slater, Wilson, and Wood,[3] however, it was found that the Kohn–Sham–Gaspar[4,5] value of the coefficient $\alpha(\frac{2}{3})$ appeared to yield the most satisfactory results for the Fermi surface and other properties. This result is in marked contrast to the non-self-consistent calculation, in which it was found that a value of α close to that proposed by Slater ($\alpha = 1$)[6] was more satisfactory.

Separate exchange potentials are obtained for electrons of majority and minority (↑ and ↓) spins and energy bands are computed separately for the two spin states. This calculation is a test of the ability of such a procedure (the spin-polarized method) to account for the magnetic and electronic properties of a ferromagnetic metal. The results are found to be in reasonable agreement with a variety of different experiments.

II. METHOD

The calculation employed the tight-binding method as reformulated by Lafon and Lin.[7] The following set of basis functions was used: Atomic wave functions for all states except 3*d* (e.g., 1*s*, 2*s*, 3*s*, 4*s*, 2*p*, 3*p*, and 4*p*) were represented by the linear combinations of Gaussian-type orbitals (GTO) determined by Wachters from a self-consistent-field calculation for the free nickel atom.[8] It appears to be important to allow the *d* wave functions sufficient freedom to distort in the crystalline environment. To accomplish this, we used a set of five separate radial GTO for each type of angular dependence of the *d* functions. The orbital exponents used in defining these functions were the same as used by Wachters.[8]

This choice of basis functions leads to a 38×38 matrix problem at a general point of the Brillouin zone. The *d-d* portion is 25×25, the *p-p* portion 9×9, and the *s-s* portion 4×4. With matrices of this size it is possible to obtain energy levels at a reasonably large number of points in the zone. Two such matrices, which refer to ↑ and ↓ spin, are obtained at each point.

The energy levels and wave functions from our previous non-self-consistent calculation[1] were