3

LINEAR RESPONSE THEORY

This chapter is devoted to a concise presentation of linear response theory, which provides a general framework for analysing the dynamical properties of a condensed-matter system close to thermal equilibrium. The dynamical processes may either be spontaneous fluctuations, or due to external perturbations, and these two kinds of phenomena are interrelated. Accounts of linear response theory may be found in many books, for example, des Cloizeaux (1968), Marshall and Lovesey (1971), and Lovesey (1986), but because of its importance in our treatment of magnetic excitations in rare earth systems and their detection by inelastic neutron scattering, the theory is presented below in adequate detail to form a basis for our later discussion.

We begin by considering the dynamical or *generalized susceptibility*, which determines the response of the system to a perturbation which varies in space and time. The *Kramers–Kronig relation* between the real and imaginary parts of this susceptibility is deduced. We derive the *Kubo formula* for the *response function* and, through its connection to the dynamic *correlation function*, which determines the results of a scattering experiment, the *fluctuation–dissipation theorem*, which relates the spontaneous fluctuations of the system to its response to an external perturbation. The energy absorption by the perturbed system is deduced from the susceptibility. The *Green function* is defined and its equation of motion established. The theory is illustrated through its application to the simple Heisenberg ferromagnet. We finally consider the calculation of the susceptibility in the *random-phase approximation*, which is the method generally used for the quantitative description of the magnetic excitations in the rare earth metals in this book.

3.1 The generalized susceptibility

A response function for a macroscopic system relates the change of an ensemble-averaged physical observable $\langle \hat{B}(t) \rangle$ to an external force $f(t)$. For example, $\hat{B}(t)$ could be the angular momentum of an ion, or the magnetization, and $f(t)$ a time-dependent applied magnetic field. As indicated by its name, the applicability of linear response theory is restricted to the regime where $\langle \hat{B}(t) \rangle$ changes linearly with the force. Hence we suppose that $f(t)$ is sufficiently weak to ensure that the response is linear. We further assume that the system is in thermal equilibrium *before* the external force is applied.

When the system is in thermal equilibrium, it is characterized by the density operator

$$\rho_0 = \frac{1}{Z} e^{-\beta \mathcal{H}_0} \qquad ; \quad Z = \mathrm{Tr}\, e^{-\beta \mathcal{H}_0}, \tag{3.1.1}$$

where $\mathcal{H}_0$ is the (effective) Hamiltonian, Z is the (grand) partition function, and $\beta = 1/k_B T$. Since we are only interested in the linear part of the response, we may assume that the weak external disturbance $f(t)$ gives rise to a linear time-dependent perturbation in the total Hamiltonian $\mathcal{H}$:

$$\mathcal{H}_1 = -\hat{A}\, f(t) \qquad ; \quad \mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1, \tag{3.1.2}$$

where $\hat{A}$ is a constant operator, as for example $\sum_i J_{zi}$, associated with the Zeeman term when $f(t) = g\mu_B H_z(t)$ (the circumflex over A or B indicates that these quantities are quantum mechanical operators). As a consequence of this perturbation, the density operator $\rho(t)$ becomes time-dependent, and so also does the ensemble average of the operator $\hat{B}$:

$$\langle \hat{B}(t) \rangle = \mathrm{Tr}\{\rho(t)\, \hat{B}\}. \tag{3.1.3}$$

The linear relation between this quantity and the external force has the form

$$\langle \hat{B}(t) \rangle - \langle \hat{B} \rangle = \int_{-\infty}^{t} \phi_{BA}(t - t')\, f(t') dt', \tag{3.1.4}$$

where $\langle \hat{B} \rangle = \langle \hat{B}(t = -\infty) \rangle = \mathrm{Tr}\{\rho_0\, \hat{B}\}$; here $f(t)$ is assumed to vanish for $t \to -\infty$. This equation expresses the condition that the differential change of $\langle \hat{B}(t) \rangle$ is proportional to the external disturbance $f(t')$ and the duration of the perturbation $\delta t'$, and further that disturbances at different times act independently of each other. The latter condition implies that the *response function* ϕ_{BA} may only depend on the time difference $t - t'$. In (3.1.4), the response is independent of any future perturbations. This causal behaviour may be incorporated in the response function by the requirement

$$\phi_{BA}(t - t') = 0 \qquad \text{for} \quad t' > t, \tag{3.1.5}$$

in which case the integration in eqn (3.1.4) can be extended from t to $+\infty$.

Because ϕ_{BA} depends only on the time difference, eqn (3.1.4) takes a simple form if we introduce the Fourier transform

$$f(\omega) = \int_{-\infty}^{\infty} f(t)\, e^{i\omega t} dt, \tag{3.1.6a}$$

and the reciprocal relation

$$f(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} f(\omega)\, e^{-i\omega t} d\omega. \tag{3.1.6b}$$

In order to take advantage of the causality condition (3.1.5), we shall consider the Laplace transform of $\phi_{BA}(t)$ (the usual s is replaced by $-iz$):

$$\chi_{BA}(z) = \int_0^{\infty} \phi_{BA}(t)\, e^{izt} dt. \tag{3.1.7a}$$

$z = z_1 + iz_2$ is a complex variable and, if $\int_0^{\infty} |\phi_{BA}(t)|\, e^{-\epsilon t} dt$ is assumed to be finite in the limit $\epsilon \to 0^+$, the converse relation is

$$\phi_{BA}(t) = \frac{1}{2\pi} \int_{-\infty+i\epsilon}^{\infty+i\epsilon} \chi_{BA}(z)\, e^{-izt} dz \qquad ; \quad \epsilon > 0. \tag{3.1.7b}$$

When $\phi_{BA}(t)$ satisfies the above condition and eqn (3.1.5), it can readily be shown that $\chi_{BA}(z)$ is an analytic function in the upper part of the complex z-plane ($z_2 > 0$).

In order to ensure that the evolution of the system is uniquely determined by $\rho_0 = \rho(-\infty)$ and $f(t)$, it is necessary that the external perturbation be turned on in a smooth, adiabatic way. This may be accomplished by replacing $f(t')$ in (4) by $f(t')\, e^{\epsilon t'}$, $\epsilon > 0$. This force vanishes in the limit $t' \to -\infty$, and any unwanted secondary effects may be removed by taking the limit $\epsilon \to 0^+$. Then, with the definition of the 'generalized' Fourier transform

$$\langle \hat{B}(\omega) \rangle = \lim_{\epsilon \to 0^+} \int_{-\infty}^{\infty} \Big(\langle \hat{B}(t) \rangle - \langle \hat{B} \rangle \Big)\, e^{i\omega t}\, e^{-\epsilon t} dt, \tag{3.1.8}$$

eqn (3.1.4) is transformed into

$$\langle \hat{B}(\omega) \rangle = \chi_{BA}(\omega)\, f(\omega), \tag{3.1.9a}$$

where $\chi_{BA}(\omega)$ is the boundary value of the analytic function $\chi_{BA}(z)$ on the real axis:

$$\chi_{BA}(\omega) = \lim_{\epsilon \to 0^+} \chi_{BA}(z = \omega + i\epsilon). \tag{3.1.9b}$$

$\chi_{BA}(\omega)$ is called the frequency-dependent or *generalized susceptibility* and is the Fourier transform, as defined by (3.1.8), of the response function $\phi_{BA}(t)$.

The mathematical restrictions (3.1.5) and (3.1.7) on $\phi_{BA}(t)$ have the direct physical significance that the system is respectively causal and stable against a small perturbation. The two conditions ensure that

$\chi_{BA}(z)$ has no poles in the upper half-plane. If this were not the case, the response $\langle\hat{B}(t)\rangle$ to a small disturbance would diverge exponentially as a function of time.

The absence of poles in $\chi_{BA}(z)$, when z_2 is positive, leads to a relation between the real and imaginary part of $\chi_{BA}(\omega)$, called the *Kramers–Kronig dispersion relation.* If $\chi_{BA}(z)$ has no poles within the contour $\mathcal{C}$, then it may be expressed in terms of the Cauchy integral along $\mathcal{C}$ by the identity

$$\chi_{BA}(z) = \frac{1}{2\pi i}\int_{\mathcal{C}} \frac{\chi_{BA}(z')}{z'-z}dz'.$$

The contour $\mathcal{C}$ is chosen to be the half-circle, in the upper half-plane, centred at the origin and bounded below by the line parallel to the z_1-axis through $z_2 = \epsilon'$, and z is a point lying within this contour. Since $\phi_{BA}(t)$ is a bounded function in the domain $\epsilon' > 0$, then $\chi_{BA}(z')$ must go to zero as $|z'| \to \infty$, whenever $z_2' > 0$. This implies that the part of the contour integral along the half-circle must vanish when its radius goes to infinity, and hence

$$\chi_{BA}(z) = \lim_{\epsilon'\to 0^+} \frac{1}{2\pi i}\int_{-\infty+i\epsilon'}^{\infty+i\epsilon'} \frac{\chi_{BA}(\omega'+i\epsilon')}{\omega'+i\epsilon'-z}d(\omega'+i\epsilon').$$

Introducing $z = \omega + i\epsilon$ and applying 'Dirac's formula':

$$\lim_{\epsilon\to 0^+} \frac{1}{\omega'-\omega-i\epsilon} = \mathcal{P}\frac{1}{\omega'-\omega} + i\pi\delta(\omega'-\omega),$$

in taking the limit $\epsilon \to 0^+$, we finally obtain the Kramers–Kronig relation ($\mathcal{P}$ denotes the principal part of the integral):

$$\chi_{BA}(\omega) = \frac{1}{i\pi}\mathcal{P}\int_{-\infty}^{\infty} \frac{\chi_{BA}(\omega')}{\omega'-\omega}d\omega', \tag{3.1.10}$$

which relates the real and imaginary components of $\chi(\omega)$.

3.2 Response functions

In this section, we shall deduce an expression for the response function $\phi_{BA}(t)$, in terms of the operators $\hat{B}$ and $\hat{A}$ and the unperturbed Hamiltonian $\mathcal{H}_0$. In the preceding section, we assumed implicitly the use of the Schrödinger picture. If instead we adopt the Heisenberg picture, the wave functions are independent of time, while the operators become time-dependent. In the Heisenberg picture, the operators are

$$\hat{B}(t) = e^{i\mathcal{H}t/\hbar}\,\hat{B}\,e^{-i\mathcal{H}t/\hbar}, \tag{3.2.1}$$

corresponding to the equation of motion

$$\frac{d}{dt}\hat{B}(t) = \frac{i}{\hbar}[\,\mathcal{H}\,,\,\hat{B}(t)\,] \tag{3.2.2}$$

(assuming that $\hat{B}$ does not depend explicitly on time). Because the wave functions are independent of time, in the Heisenberg picture, the corresponding density operator ρ_H must also be. Hence we may write (3.1.3)

$$\langle\hat{B}(t)\rangle = \mathrm{Tr}\big\{\rho(t)\,\hat{B}\big\} = \mathrm{Tr}\big\{\rho_H\,\hat{B}(t)\big\}. \tag{3.2.3}$$

Introducing (3.2.1) into this expression, and recalling that the trace is invariant under a cyclic permutation of the operators within it, we obtain

$$\rho(t) = e^{-i\mathcal{H}t/\hbar}\,\rho_H\,e^{i\mathcal{H}t/\hbar},$$

or

$$\frac{d}{dt}\rho(t) = -\frac{i}{\hbar}[\,\mathcal{H}\,,\,\rho(t)\,]. \tag{3.2.4}$$

The equation of motion derived for the density operator, in the Schrödinger picture, is similar to the Heisenberg equation of motion above, except for the change of sign in front of the commutator.

The density operator may be written as the sum of two terms:

$$\rho(t) = \rho_0 + \rho_1(t) \qquad \text{with} \quad [\mathcal{H}_0\,,\,\rho_0] = 0, \tag{3.2.5}$$

where ρ_0 is the density operator (3.1.1) of the thermal-equilibrium state which, by definition, must commute with $\mathcal{H}_0$, and the additional contribution due to $f(t)$ is assumed to vanish at $t \to -\infty$. In order to derive $\rho_1(t)$ to leading order in $f(t)$, we shall first consider the following density operator, in the *interaction picture*,

$$\rho_I(t) \equiv e^{i\mathcal{H}_0 t/\hbar}\,\rho(t)\,e^{-i\mathcal{H}_0 t/\hbar}, \tag{3.2.6}$$

for which

$$\begin{aligned}\frac{d}{dt}\rho_I(t) &= e^{i\mathcal{H}_0 t/\hbar}\left\{\frac{i}{\hbar}[\mathcal{H}_0\,,\,\rho(t)\,] + \frac{d}{dt}\rho(t)\right\}e^{-i\mathcal{H}_0 t/\hbar}\\ &= -\frac{i}{\hbar}e^{i\mathcal{H}_0 t/\hbar}\,[\,\mathcal{H}_1\,,\,\rho(t)\,]\,e^{-i\mathcal{H}_0 t/\hbar}.\end{aligned}$$

Because $\mathcal{H}_1$ is linear in $f(t)$, we may replace $\rho(t)$ by ρ_0 in calculating the linear response, giving

$$\frac{d}{dt}\rho_I(t) \simeq -\frac{i}{\hbar}\big[\,e^{i\mathcal{H}_0 t/\hbar}\,\mathcal{H}_1\,e^{-i\mathcal{H}_0 t/\hbar}\,,\,\rho_0\,\big] = \frac{i}{\hbar}[\,\hat{A}_0(t)\,,\,\rho_0\,]f(t),$$

using (3.2.5) and defining

$$\hat{A}_0(t) = e^{i\mathcal{H}_0 t/\hbar}\, \hat{A}\, e^{-i\mathcal{H}_0 t/\hbar}.$$

According to (3.2.6), taking into account the boundary condition, the time-dependent density operator is

$$\begin{aligned} \rho(t) &= e^{-i\mathcal{H}_0 t/\hbar} \Big(\int_{-\infty}^{t} \frac{d}{dt'}\rho_I(t')dt' + \rho_0 \Big)\, e^{i\mathcal{H}_0 t/\hbar} \\ &= \rho_0 + \frac{i}{\hbar}\int_{-\infty}^{t} [\, \hat{A}_0(t'-t)\,,\, \rho_0\,]\, f(t')dt', \end{aligned} \tag{3.2.7}$$

to first order in the external perturbations. This determines the time dependence of, for example, $\hat{B}$ as

$$\begin{aligned} \langle \hat{B}(t)\rangle - \langle \hat{B}\rangle &= \mathrm{Tr}\big\{\, (\rho(t)-\rho_0)\ \hat{B}\big\} \\ &= \frac{i}{\hbar}\,\mathrm{Tr}\Big\{ \int_{-\infty}^{t} [\, \hat{A}_0(t'-t)\,,\, \rho_0\,]\, \hat{B}\, f(t')dt' \Big\} \end{aligned}$$

and, utilizing the invariance of the trace under cyclic permutations, we obtain, to leading order,

$$\begin{aligned} \langle \hat{B}(t)\rangle - \langle \hat{B}\rangle &= \frac{i}{\hbar}\int_{-\infty}^{t} \mathrm{Tr}\big\{\rho_0\, [\, \hat{B}\,,\, \hat{A}_0(t'-t)\,]\big\}\, f(t')dt' \\ &= \frac{i}{\hbar}\int_{-\infty}^{t} \langle\, [\, \hat{B}_0(t)\,,\, \hat{A}_0(t')\,]\,\rangle_0\, f(t')dt'. \end{aligned} \tag{3.2.8}$$

A comparison of this result with the definition (3.1.4) of the response function then gives

$$\phi_{BA}(t-t') = \frac{i}{\hbar}\theta(t-t')\langle\, [\, \hat{B}(t)\,,\, \hat{A}(t')\,]\,\rangle, \tag{3.2.9}$$

where the unit step function, $\theta(t) = 0$ or 1 when $t < 0$ or $t > 0$ respectively, is introduced in order to ensure that ϕ_{BA} satisfies the causality principle (3.1.5). In this final result, and below, we suppress the index 0, but we stress that both the variations with time and the ensemble average are thermal-equilibrium values determined by $\mathcal{H}_0$, and are unaffected by the external disturbances. This expression in terms of microscopic quantities, is called *the Kubo formula* for the response function (Kubo 1957, 1966).

The expression (3.2.9) is the starting point for introducing a number of useful functions:

$$K_{BA}(t) = \frac{i}{\hbar}\langle\, [\hat{B}(t)\,,\, \hat{A}\,]\,\rangle = \frac{i}{\hbar}\langle\, [\hat{B}\,,\, \hat{A}(-t)\,]\,\rangle \tag{3.2.10}$$

is also called a response function. $\hat{A}$ is a shorthand notation for $\hat{A}(t=0)$. The inverse response function $K_{AB}(t)$, which determines $\langle \hat{A}(t) \rangle$ caused by the perturbation $\mathcal{H}_1 = -f(t)\hat{B}$, is

$$K_{AB}(t) = \frac{i}{\hbar} \langle [\hat{A}(t)\,,\, \hat{B}\,] \rangle = -K_{BA}(-t),$$

and $K_{BA}(t)$ can be expressed in terms of the corresponding causal response functions as

$$K_{BA}(t) = \begin{cases} \phi_{BA}(t) & \text{for} \quad t > 0 \\ -\phi_{AB}(-t) & \text{for} \quad t < 0. \end{cases}$$

The susceptibility is divided into two terms, the reactive part

$$\chi'_{BA}(z) = \chi'_{AB}(-z^*) \equiv \frac{1}{2}\big\{\chi_{BA}(z) + \chi_{AB}(-z^*)\big\}, \qquad (3.2.11a)$$

and the absorptive part

$$\chi''_{BA}(z) = -\chi''_{AB}(-z^*) \equiv \frac{1}{2i}\big\{\chi_{BA}(z) - \chi_{AB}(-z^*)\big\}, \qquad (3.2.11b)$$

so that

$$\chi_{BA}(z) = \chi'_{BA}(z) + i\chi''_{BA}(z) \qquad (3.2.11c)$$

and, according to the Kramers–Kronig relation (3.1.10),

$$\chi'_{BA}(\omega) = \frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty} \frac{\chi''_{BA}(\omega')}{\omega' - \omega} d\omega' \quad ; \quad \chi''_{BA}(\omega) = -\frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty} \frac{\chi'_{BA}(\omega')}{\omega' - \omega} d\omega'. \qquad (3.2.11d)$$

In these equations, $\chi_{AB}(-\omega)$ is the boundary value obtained by taking $z = \omega + i\epsilon$, i.e. as $\lim_{\epsilon \to 0^+} \chi_{AB}(-z^* = -\omega + i\epsilon)$, corresponding to the condition that $\chi_{AB}(-z^*)$, like $\chi_{AB}(z)$, is analytic in the upper half-plane. The appropriate Laplace transform of $K_{BA}(t)$ with this property is

$$\begin{aligned} K_{BA}(z) &= \int_{-\infty}^{\infty} K_{BA}(t)\, e^{i(z_1 t + i z_2 |t|)} dt \\ &= \int_0^{\infty} \phi_{BA}(t)\, e^{izt} dt - \int_0^{\infty} \phi_{AB}(t)\, e^{-iz^* t} dt. \end{aligned}$$

Hence

$$K_{BA}(z) = 2i\,\chi''_{BA}(z). \qquad (3.2.12)$$

Next we introduce the dynamic *correlation function*, sometimes referred to as the *scattering function*. It is defined as follows:

$$S_{BA}(t) \equiv \langle \hat{B}(t)\,\hat{A} \rangle - \langle \hat{B} \rangle \langle \hat{A} \rangle = \langle \hat{B}\,\hat{A}(-t) \rangle - \langle \hat{B} \rangle \langle \hat{A} \rangle, \qquad (3.2.13)$$

and is related to the response function introduced earlier by

$$K_{BA}(t) = \frac{i}{\hbar}\big\{S_{BA}(t) - S_{AB}(-t)\big\}. \tag{3.2.14}$$

The different response functions obey a number of symmetry relations, due to the invariance of the trace under a cyclic permutation of the operators. To derive the first, we recall that the Hermitian conjugate of an operator is defined by

$$(<\alpha|\hat{B}|\alpha'>)^* = <\alpha'|\hat{B}^\dagger|\alpha> .$$

If we assume that a certain set of state vectors $|\alpha>$ constitutes a diagonal representation, i.e. $\mathcal{H}_0|\alpha> = E_\alpha|\alpha>$, then it is straightforward to show that

$$\langle\hat{B}(t)\,\hat{A}\rangle^* = \langle\hat{A}^\dagger(-t)\,\hat{B}^\dagger\rangle,$$

leading to the symmetry relations

$$K^*_{BA}(t) = K_{B^\dagger A^\dagger}(t)$$

and

$$\chi^*_{BA}(z) = \chi_{B^\dagger A^\dagger}(-z^*). \tag{3.2.15}$$

Another important relation is derived as follows:

$$\begin{aligned}\langle\hat{B}(t)\,\hat{A}\rangle &= \frac{1}{Z}\mathrm{Tr}\left\{e^{-\beta\mathcal{H}_0}\,e^{i\mathcal{H}_0 t/\hbar}\,\hat{B}\,e^{-i\mathcal{H}_0 t/\hbar}\,\hat{A}\right\}\\ &= \frac{1}{Z}\mathrm{Tr}\left\{e^{i\mathcal{H}_0(t+i\beta\hbar)/\hbar}\,\hat{B}\,e^{-i\mathcal{H}_0(t+i\beta\hbar)/\hbar}\,e^{-\beta\mathcal{H}_0}\,\hat{A}\right\}\\ &= \frac{1}{Z}\mathrm{Tr}\left\{e^{-\beta\mathcal{H}_0}\,\hat{A}\,\hat{B}(t+i\beta\hbar)\right\} = \langle\hat{A}\,\hat{B}(t+i\beta\hbar)\rangle,\end{aligned}$$

implying that

$$S_{BA}(t) = S_{AB}(-t-i\beta\hbar). \tag{3.2.16}$$

In any realistic system which, rather than being isolated, is in contact with a thermal bath at temperature T, the correlation function $S_{BA}(t)$ vanishes in the limits $t \to \pm\infty$, corresponding to the condition $\langle\hat{B}(t=\pm\infty)\,\hat{A}\rangle = \langle\hat{B}\rangle\langle\hat{A}\rangle$. If we further assume that $S_{BA}(t)$ is an analytic function in the interval $|t_2| \leq \beta$ of the complex t-plane, then the Fourier transform of (3.2.16) is

$$S_{BA}(\omega) = e^{\beta\hbar\omega}\,S_{AB}(-\omega), \tag{3.2.17}$$

which is usually referred to as being the *condition of detailed balance.* Combining this condition with the expressions (3.2.12) and (3.2.14), we get the following important relation between the correlation function and the susceptibility:

$$S_{BA}(\omega) = 2\hbar \frac{1}{1 - e^{-\beta\hbar\omega}} \chi''_{BA}(\omega), \tag{3.2.18}$$

which is called the *fluctuation–dissipation theorem.* This relation expresses explicitly the close connection between the spontaneous fluctuations in the system, as described by the correlation function, and the response of the system to external perturbations, as determined by the susceptibility.

The calculations above do not depend on the starting assumption that $\hat{B}$ (or $\hat{A}$) is a physical observable, i.e. that $\hat{B}$ should be equal to $\hat{B}^\dagger$. This has the advantage that, if the Kubo formula (3.2.9) is taken to be the starting point instead of eqn (3.1.4), the formalism applies more generally.

3.3 Energy absorption and the Green function

In this section, we first present a calculation of the energy transferred to the system by the external perturbation $\mathcal{H}_1 = -\hat{A}\, f(t)$ in (3.1.2), incidentally justifying the names of the two susceptibility components in (3.2.11). The energy absorption can be expressed in terms of $\chi_{AA}(\omega)$ and, without loss of generality, $\hat{A}$ may here be assumed to be a Hermitian operator, so that $\hat{A} = \hat{A}^\dagger$. In this case, $f(t)$ is real, and considering a harmonic variation

$$f(t) = f_0 \cos(\omega_0 t) = \tfrac{1}{2} f_0 \left(e^{i\omega_0 t} + e^{-i\omega_0 t} \right) \quad \text{with} \quad f_0^* = f_0,$$

then

$$f(\omega) = \pi f_0 \{\delta(\omega - \omega_0) + \delta(\omega + \omega_0)\}, \quad \text{as} \quad \int_{-\infty}^{\infty} e^{i(\omega-\omega_0)t} dt = 2\pi\delta(\omega - \omega_0),$$

and we have

$$\langle \hat{A}(t) \rangle - \langle \hat{A} \rangle = \tfrac{1}{2} f_0 \left\{ \chi_{AA}(-\omega_0)\, e^{i\omega_0 t} + \chi_{AA}(\omega_0)\, e^{-i\omega_0 t} \right\}.$$

The introduction of $\hat{A} = \hat{B} = \hat{A}^\dagger$ in (3.2.15), and in the definition (3.2.11), yields

$$\begin{aligned} \chi'_{AA}(\omega)^* &= \chi'_{AA}(\omega) = \chi'_{AA}(-\omega) \\ \chi''_{AA}(\omega)^* &= \chi''_{AA}(\omega) = -\chi''_{AA}(-\omega), \end{aligned} \tag{3.3.1}$$

and these symmetry relations allow us to write

$$\langle \hat{A}(t)\rangle - \langle \hat{A}\rangle = f_0 \left\{\chi'_{AA}(\omega_0)\cos(\omega_0 t) + \chi''_{AA}(\omega_0)\sin(\omega_0 t)\right\}.$$

The part of the response which is in phase with the external force is proportional to $\chi'_{AA}(\omega_0)$, which is therefore called the reactive component. The rate of energy absorption due to the field is

$$Q = \frac{d}{dt}\langle \mathcal{H}\rangle = \langle \partial\mathcal{H}/\partial t\rangle = -\langle \hat{A}(t)\rangle\, \partial f/\partial t,$$

which shows that the *mean* dissipation rate is determined by the out-of-phase response proportional to $\chi''_{AA}(\omega)$:

$$\overline{Q} = \tfrac{1}{2} f_0^2\, \omega_0\, \chi''_{AA}(\omega_0) \tag{3.3.2}$$

and $\chi''_{AA}(\omega)$ is therefore called the absorptive part of the susceptibility.

If the eigenvalues E_α and the corresponding eigenstates $|\alpha>$ for the Hamiltonian $\mathcal{H}(=\mathcal{H}_0)$ are known, it is possible to derive an explicit expression for $\chi_{BA}(\omega)$. According to the definition (3.2.10),

$$\begin{aligned}K_{BA}(t) = \frac{i}{\hbar}\frac{1}{Z}\mathrm{Tr}\Big\{ e^{-\beta\mathcal{H}}\,[\,e^{i\mathcal{H}t/\hbar}\,\hat{B}\,e^{-i\mathcal{H}t/\hbar}\,,\,\hat{A}\,]\Big\} = \\ \frac{i}{\hbar}\frac{1}{Z}\sum_{\alpha\alpha'} e^{-\beta E_\alpha}\left\{ e^{iE_\alpha t/\hbar} < \alpha|\hat{B}|\alpha'> e^{-iE_{\alpha'}t/\hbar} < \alpha'|\hat{A}|\alpha > \right. \\ \left. - < \alpha|\hat{A}|\alpha'> e^{iE_{\alpha'}t/\hbar} < \alpha'|\hat{B}|\alpha > e^{-iE_\alpha t/\hbar}\right\}.\end{aligned}$$

Interchanging α and α' in the last term, and introducing the population factor

$$n_\alpha = \frac{1}{Z}e^{-\beta E_\alpha} \quad ; \quad Z = \sum_{\alpha'} e^{-\beta E_{\alpha'}}, \tag{3.3.3a}$$

we get

$$K_{BA}(t) = \frac{i}{\hbar}\sum_{\alpha\alpha'} < \alpha|\hat{B}|\alpha'>< \alpha'|\hat{A}|\alpha > (n_\alpha - n_{\alpha'})\, e^{i(E_\alpha - E_{\alpha'})t/\hbar}, \tag{3.3.3b}$$

and hence

$$\begin{aligned}\chi_{BA}(\omega) &= \lim_{\epsilon\to 0^+}\int_0^\infty K_{BA}(t)\, e^{i(w+i\epsilon)t}dt \\ &= \lim_{\epsilon\to 0^+}\sum_{\alpha\alpha'}\frac{< \alpha|\hat{B}|\alpha'>< \alpha'|\hat{A}|\alpha >}{E_{\alpha'} - E_\alpha - \hbar\omega - i\hbar\epsilon}\,(n_\alpha - n_{\alpha'}),\end{aligned} \tag{3.3.4a}$$

or equivalently

$$\begin{aligned}\chi_{AB}(-\omega) &= \lim_{\epsilon\to 0^+} \chi_{AB}(-\omega+i\epsilon)\\ &= \lim_{\epsilon\to 0^+} \sum_{\alpha\alpha'} \frac{<\alpha|\hat{A}|\alpha'><\alpha'|\hat{B}|\alpha>}{E_{\alpha'}-E_\alpha+\hbar\omega-i\hbar\epsilon}\,(n_\alpha-n_{\alpha'}).\end{aligned} \tag{3.3.4b}$$

An interchange of α and α' shows this expression to be the same as (3.3.4a), with ϵ replaced by $-\epsilon$. The application of Dirac's formula then yields the absorptive part of the susceptibility (3.2.11b) as

$$\chi''_{BA}(\omega) = \pi \sum_{\alpha\alpha'} <\alpha|\hat{B}|\alpha'><\alpha'|\hat{A}|\alpha>\,(n_\alpha-n_{\alpha'})\,\delta\big(\hbar\omega-(E_{\alpha'}-E_\alpha)\big) \tag{3.3.5}$$

(equal to $K_{BA}(\omega)/2i$ in accordance with (3.2.12)), whereas the reactive part (3.2.11a) is

$$\chi'_{BA}(\omega) = \sum_{\alpha\alpha'}^{E_\alpha\neq E_{\alpha'}} \frac{<\alpha|\hat{B}|\alpha'><\alpha'|\hat{A}|\alpha>}{E_{\alpha'}-E_\alpha-\hbar\omega}\,(n_\alpha-n_{\alpha'}) + \chi'_{BA}(el)\,\delta_{\omega 0}, \tag{3.3.6a}$$

where

$$\delta_{\omega 0} \equiv \lim_{\epsilon\to 0^+} \frac{i\epsilon}{\omega+i\epsilon} = \begin{cases} 1 & \text{if} \quad \omega = 0\\ 0 & \text{if} \quad \omega\neq 0,\end{cases}$$

and the elastic term $\chi'_{BA}(el)$, which only contributes in the static limit $\omega = 0$, is

$$\chi'_{BA}(el) = \beta\Big\{\sum_{\alpha\alpha'}^{E_\alpha=E_{\alpha'}} <\alpha|\hat{B}|\alpha'><\alpha'|\hat{A}|\alpha>n_\alpha\ -\ \langle\hat{B}\rangle\langle\hat{A}\rangle\Big\}. \tag{3.3.6b}$$

We remark that $\chi'_{BA}(\omega)$ and $\chi''_{BA}(\omega)$ are often referred to respectively as the real and the imaginary part of $\chi_{BA}(\omega)$. This terminology is not valid in general, but only if the matrix-element products are real, as they are if, for instance, $\hat{B} = \hat{A}^\dagger$. The presence of the elastic term in the reactive response requires some additional consideration. There are no elastic contributions to $K_{BA}(t)$, nor hence to $\chi''_{BA}(\omega)$, because $n_\alpha - n_{\alpha'} \equiv 0$ if $E_\alpha = E_{\alpha'}$. Nevertheless, the appearance of an extra contribution at $\omega = 0$, not obtainable directly from $K_{BA}(t)$, is possible because the energy denominator in (3.3.4) vanishes in the limit $|\omega + i\epsilon| \to 0$, when $E_\alpha = E_{\alpha'}$. In order to derive this contribution, we consider the equal-time correlation function

$$\begin{aligned}S_{BA}(t=0) &= \langle(\hat{B}-\langle\hat{B}\rangle)(\hat{A}-\langle\hat{A}\rangle)\rangle\\ &= \sum_{\alpha\alpha'} <\alpha|\hat{B}|\alpha'><\alpha'|\hat{A}|\alpha>n_\alpha\ -\ \langle\hat{B}\rangle\langle\hat{A}\rangle\end{aligned} \tag{3.3.7a}$$

which, according to the fluctuation–dissipation theorem (3.2.18), should be

$$S_{BA}(t=0) = \frac{1}{2\pi}\int_{-\infty}^{\infty} S_{BA}(\omega)\,d\omega = \frac{1}{\pi}\int_{-\infty}^{\infty} \frac{1}{1-e^{-\beta\hbar\omega}}\,\chi''_{BA}(\omega)d(\hbar\omega). \tag{3.3.7b}$$

Introducing (3.3.5), the integration is straightforward, except in a narrow interval around $\omega = 0$, and we obtain

$$S_{BA}(t=0) = \sum_{\alpha\alpha'}^{E_\alpha \neq E_{\alpha'}} <\alpha\,|\,\hat{B}\,|\,\alpha'><\alpha'\,|\,\hat{A}\,|\,\alpha> n_\alpha + \lim_{\gamma\to 0^+}\int_{-\gamma}^{\gamma}\frac{\chi''_{BA}(\omega)}{\pi\beta\omega}d\omega$$

after replacing $1-e^{-\beta\hbar\omega}$ with $\beta\hbar\omega$ in the limit $\omega \to 0$. A comparison of this expression for $S_{BA}(t=0)$ with (3.3.7a) shows that the last integral has a definite value:

$$\lim_{\gamma\to 0^+}\int_{-\gamma}^{\gamma}\frac{\chi''_{BA}(\omega)}{\pi\beta\omega}d\omega = \sum_{\alpha\alpha'}^{E_\alpha = E_{\alpha'}} <\alpha\,|\,\hat{B}\,|\,\alpha'><\alpha'\,|\,\hat{A}\,|\,\alpha> n_\alpha \;-\; \langle\hat{B}\rangle\langle\hat{A}\rangle. \tag{3.3.8}$$

The use of the Kramers–Kronig relation (3.1.10), in the form of (3.2.11d), for calculating $\chi'_{BA}(0)$ then gives rise to the extra contribution

$$\chi'_{BA}(el) = \lim_{\gamma\to 0^+}\frac{1}{\pi}\int_{-\gamma}^{\gamma}\frac{\chi''_{BA}(\omega)}{\omega}d\omega \tag{3.3.9}$$

to the reactive susceptibility at zero frequency, as anticipated in (3.3.6b). The zero-frequency result, $\chi_{BA}(0) = \chi'_{BA}(0)$, as given by (3.3.6), is the same as the conventional isothermal susceptibility (2.1.18) for the magnetic moments, where the elastic and inelastic contributions are respectively the Curie and the Van Vleck terms. This elastic contribution is discussed in more detail by, for instance, Suzuki (1971).

The results (3.3.4–6) show that, if the eigenstates of the Hamiltonian are discrete and the matrix-elements of the operators $\hat{B}$ and $\hat{A}$ between these states are well-defined, the poles of $\chi_{BA}(z)$ all lie on the real axis. This has the consequence that the absorptive part $\chi''_{BA}(\omega)$ (3.3.5) becomes a sum of δ-functions, which are only non-zero when $\hbar\omega$ is equal to the *excitation* energies $E_{\alpha'} - E_\alpha$. In such a system, no spontaneous transitions occur. In a real macroscopic system, the distribution of states is continuous, and only the ground state may be considered as a well-defined discrete state. At non-zero temperatures, the parameters of the system are subject to fluctuations in space and time. The introduction of a non-zero probability for a spontaneous transition between the 'levels' α and α' can be included in a phenomenological way by replacing the energy difference $E_{\alpha'} - E_\alpha$ in (3.3.4) by $(E_{\alpha'} - E_\alpha) - i\,\Gamma_{\alpha'\alpha}(\omega)$, where the parameters, including the energy difference, usually depend on ω. According to the general stability and causality requirements, the poles of $\chi_{BA}(z)$ at $z = z_{\alpha'\alpha} = (E_{\alpha'} - E_\alpha) - i\,\Gamma_{\alpha'\alpha}$ must lie in the lower half-plane, implying that $\Gamma_{\alpha'\alpha}$ has to be positive (or zero). In the case where $|E_{\alpha'} - E_\alpha| \gg \Gamma_{\alpha'\alpha}$, the ω-dependence of these parameters is unimportant, and the δ-function in (3.3.5) is effectively replaced by a *Lorentzian*:

$$\begin{aligned} \chi''_{BA}(\omega) \simeq \sum_{\alpha\alpha'} \frac{< \alpha|\hat{B}|\alpha' >< \alpha'|\hat{A}|\alpha >}{(E_{\alpha'} - E_\alpha - \hbar\omega)^2 + \Gamma^2_{\alpha'\alpha}}\, \Gamma_{\alpha'\alpha}(n_\alpha - n_{\alpha'}) \\ + \frac{\hbar\omega\Gamma_0}{(\hbar\omega)^2 + \Gamma_0^2}\, \chi'_{BA}(el), \end{aligned} \tag{3.3.10}$$

with a *linewidth*, or more precisely FWHM (full width at half maximum), of $2\Gamma_{\alpha'\alpha}$. In (3.3.10), we have added the *quasi-elastic* response due to a pole at $z = -i\,\Gamma_0$, which replaces the one at $z = 0$. The corresponding reactive part of the susceptibility is

$$\begin{aligned} \chi'_{BA}(\omega) \simeq \sum_{\alpha\alpha'} \frac{< \alpha|\hat{B}|\alpha' >< \alpha'|\hat{A}|\alpha >}{(E_{\alpha'} - E_\alpha - \hbar\omega)^2 + \Gamma^2_{\alpha'\alpha}} (E_{\alpha'} - E_\alpha - \hbar\omega)(n_\alpha - n_{\alpha'}) \\ + \frac{\Gamma_0^2}{(\hbar\omega)^2 + \Gamma_0^2}\, \chi'_{BA}(el). \end{aligned} \tag{3.3.11}$$

The non-zero linewidth corresponds to an exponential decay of the oscillations in the time dependence of, for instance, the correlation function:

$$S_{BA}(t) \sim e^{-iz_{\alpha'\alpha}t/\hbar} = e^{-i(E_{\alpha'} - E_\alpha)t/\hbar}\, e^{-\Gamma_{\alpha'\alpha}t/\hbar}.$$

The absorption observed in a *resonance* experiment is proportional to $\chi''_{AA}(\omega)$. A peak in the absorption spectrum is interpreted as an *elementary* or *quasi-particle excitation*, or as a *normal mode* of the dynamic variable $\hat{A}$, with a *lifetime* $\tau = \hbar/\Gamma_{\alpha'\alpha}$. A pole at $z = -i\Gamma_0$ is said to represent a *diffusive mode*. Such a pole is of particular importance for those transport coefficients determined by the low-frequency or hydrodynamic properties of the system. Kubo (1957, 1966) gives a detailed discussion of this subject. As we shall see later, the differential scattering cross-section of, for example, neutrons in the Born-approximation is proportional to a correlation function, and hence to $\chi''(\omega)$. This implies that the presence of elementary excitations in the system leads to peaks in the intensity of scattered neutrons as a function of the energy transfer. Finally, the dynamic correlation-functions are related directly to various thermodynamic second-derivatives, such as the compressibility and the magnetic susceptibility, and thereby indirectly to the corresponding first-derivatives, like the specific heat and the magnetization. Consequently, most physical properties of a macroscopic system near equilibrium may be described in terms of the correlation functions.

As a supplement to the response function $\phi_{BA}(t-t')$, we now introduce the *Green function*, defined as

$$\begin{aligned} G_{BA}(t-t') &\equiv \langle\langle \hat{B}(t)\,;\,\hat{A}(t')\rangle\rangle \\ &\equiv -\frac{i}{\hbar}\theta(t-t')\langle\,[\,\hat{B}(t)\,,\,\hat{A}(t')\,]\,\rangle = -\phi_{BA}(t-t'). \end{aligned} \tag{3.3.12}$$

This Green function is often referred to as the *double-time* or the *retarded* Green function (Zubarev 1960), and it is simply our previous response function, but with the opposite sign. Introducing the Laplace transform $G_{BA}(z)$ according to (3.1.7), we find, as before, that the corresponding Fourier transform is

$$\begin{aligned} G_{BA}(\omega) \equiv \langle\langle \hat{B}\,;\,\hat{A}\rangle\rangle_\omega &= \lim_{\epsilon\to 0^+} G_{BA}(z=\omega+i\epsilon) \\ &= \lim_{\epsilon\to 0^+}\int_{-\infty(0)}^{\infty} G_{BA}(t)\, e^{i(\omega+i\epsilon)t}dt = -\chi_{BA}(\omega). \end{aligned} \tag{3.3.13}$$

We note that, if $\hat{A}$ and $\hat{B}$ are dimensionless operators, then $G_{BA}(\omega)$ or $\chi_{BA}(\omega)$ have the dimensions of inverse energy.

If $t'=0$, the derivative of the Green function with respect to t is

$$\begin{aligned} \frac{d}{dt}G_{BA}(t) &= -\frac{i}{\hbar}\Big(\delta(t)\langle\,[\,\hat{B}(t)\,,\,\hat{A}\,]\,\rangle + \theta(t)\langle\,[\,d\hat{B}(t)/dt\,,\,\hat{A}\,]\,\rangle\Big) \\ &= -\frac{i}{\hbar}\Big(\delta(t)\langle\,[\,\hat{B}\,,\,\hat{A}\,]\,\rangle - \frac{i}{\hbar}\theta(t)\langle\,[\,[\,\hat{B}(t)\,,\,\mathcal{H}\,]\,,\,\hat{A}\,]\,\rangle\Big). \end{aligned}$$

A Fourier transformation of this expression then leads to the *equation of motion* for the Green function:

$$\hbar\omega\langle\langle \hat{B}\,;\,\hat{A}\rangle\rangle_\omega - \langle\langle\,[\,\hat{B}\,,\,\mathcal{H}\,]\,;\,\hat{A}\rangle\rangle_\omega = \langle\,[\,\hat{B}\,,\,\hat{A}\,]\,\rangle. \tag{3.3.14a}$$

The suffix ω indicates the Fourier transforms (3.3.13), and $\hbar\omega$ is shorthand for $\hbar(\omega+i\epsilon)$ with $\epsilon\to 0^+$. In many applications, $\hat{A}$ and $\hat{B}$ are the same (Hermitian) operator, in which case the r.h.s. of (3.3.14a) vanishes and one may proceed to the second derivative. With the condition that $\langle\,[\,[\,[\,\hat{A}(t)\,,\,\mathcal{H}\,]\,,\,\mathcal{H}\,]\,,\,\hat{A}\,]\,\rangle$ is $-\langle\,[\,[\,\hat{A}(t)\,,\,\mathcal{H}\,]\,,\,[\,\hat{A}\,,\,\mathcal{H}\,]\,]\,\rangle$, the equation of motion for the Green function $\langle\langle\,[\,\hat{A}\,,\,\mathcal{H}\,]\,;\,\hat{A}\rangle\rangle_\omega$ leads to

$$(\hbar\omega)^2\langle\langle \hat{A}\,;\,\hat{A}\rangle\rangle_\omega + \langle\langle\,[\,\hat{A}\,,\,\mathcal{H}\,]\,;\,[\,\hat{A}\,,\,\mathcal{H}\,]\,\rangle\rangle_\omega = \langle\,[\,[\,\hat{A}\,,\mathcal{H}\,]\,,\,\hat{A}\,]\,\rangle. \tag{3.3.14b}$$

The pair of equations (3.3.14) will be the starting point for our application of linear response theory.

According to the definition (3.2.10) of $K_{BA}(t)$, and eqn (3.2.12),

$$K_{BA}(\omega) = 2i\chi''_{BA}(\omega) = -2iG''_{BA}(\omega).$$

We may write

$$\frac{i}{\pi}\int_{-\infty}^{\infty} \chi''_{BA}(\omega)\, e^{-i\omega t} d\omega = \frac{i}{\hbar}\langle\, [\, \hat{B}(t)\, ,\, \hat{A}\,]\, \rangle \tag{3.3.15}$$

and, setting $t = 0$, we obtain the following *sum rule*:

$$\frac{\hbar}{\pi}\int_{-\infty}^{\infty} \chi''_{BA}(\omega) d\omega = \langle\, [\, \hat{B}\, ,\, \hat{A}\,]\, \rangle, \tag{3.3.16}$$

which may be compared with the value obtained for the equal-time correlation function $\langle \hat{B}\, \hat{A}\rangle - \langle \hat{B}\rangle\langle \hat{A}\rangle$, (3.3.7). The Green function in (3.3.14*a*) must satisfy this sum rule, and we note that the thermal averages in (3.3.14*a*) and (3.3.16) are the same. Equation (3.3.16) is only the first of a whole series of sum rules.

The nth time-derivative of $\hat{B}(t)$ may be written

$$\frac{d^n}{dt^n}\hat{B}(t) = \left(\frac{i}{\hbar}\right)^n \mathcal{L}^n \hat{B}(t) \qquad \text{with} \quad \mathcal{L}\hat{B}(t) \equiv [\, \mathcal{H}\, ,\, \hat{B}(t)\,].$$

Taking the nth derivative on both sides of eqn (3.3.15), we get

$$\frac{i}{\pi}\int_{-\infty}^{\infty} (-i\omega)^n \chi''_{BA}(\omega)\, e^{-i\omega t} d\omega = \left(\frac{i}{\hbar}\right)^{n+1} \langle\, [\, \mathcal{L}^n \hat{B}(t)\, ,\, \hat{A}\,]\, \rangle.$$

Next we introduce the normalized *spectral weight function*

$$F_{BA}(\omega) = \frac{1}{\chi'_{BA}(0)}\frac{1}{\pi}\frac{\chi''_{BA}(\omega)}{\omega}, \quad \text{where} \quad \int_{-\infty}^{\infty} F_{BA}(\omega) d\omega = 1. \tag{3.3.17a}$$

The normalization of $F_{BA}(\omega)$ is a simple consequence of the Kramers–Kronig relation (3.2.11*d*). The nth order moment of ω, with respect to the spectral weight function $F_{BA}(\omega)$, is then defined as

$$\langle\, \omega^n \rangle_{BA} = \int_{-\infty}^{\infty} \omega^n F_{BA}(\omega) d\omega, \tag{3.3.17b}$$

which allows the relation between the nth derivatives at $t = 0$ to be written

$$\chi'_{BA}(0)\, \langle (\hbar\omega)^{n+1} \rangle_{BA} = (-1)^n \langle\, [\, \mathcal{L}^n \hat{B}\, ,\, \hat{A}\,]\, \rangle. \tag{3.3.18a}$$

These are the sum rules relating the spectral frequency-moments with the thermal expectation-values of operators obtainable from $\hat{B}$, $\hat{A}$, and $\mathcal{H}$. If $\hat{B} = \hat{A} = \hat{A}^\dagger$, then (3.3.1) shows that$F_{BA}(\omega)$ is even in ω, and all the odd moments vanish. In this case, the even moments are

$$\chi'_{AA}(0)\,\langle(\hbar\omega)^{2n}\rangle_{AA} = -\langle\,[\,\mathcal{L}^{2n-1}\hat{A}\,,\,\hat{A}\,]\,\rangle. \qquad (3.3.18b)$$

3.4 Linear response of the Heisenberg ferromagnet

In this section, we shall illustrate the use of linear response theory by applying it to the case of the three-dimensional Heisenberg ferromagnet, with the Hamiltonian

$$\mathcal{H} = -\frac{1}{2}\sum_{i\neq j}\mathcal{J}(ij)\,\mathbf{S}_i\cdot\mathbf{S}_j, \qquad (3.4.1)$$

where $\mathbf{S}_i$ is the spin on the ith ion, placed in a Bravais lattice at the position $\mathbf{R}_i$. The spatial Fourier transform of the exchange coupling, with the condition $\mathcal{J}(ii)\equiv 0$, is

$$\mathcal{J}(\mathbf{q}) = \frac{1}{N}\sum_{ij}\mathcal{J}(ij)\,e^{-i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)} = \sum_{j}\mathcal{J}(ij)\,e^{-i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)}, \qquad (3.4.2a)$$

and conversely

$$\mathcal{J}(ij) = \frac{1}{N}\sum_{\mathbf{q}}\mathcal{J}(\mathbf{q})\,e^{i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)} = \frac{V}{N(2\pi)^3}\int\mathcal{J}(\mathbf{q})\,e^{i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)}d\mathbf{q}, \qquad (3.4.2b)$$

depending on whether $\mathbf{q}$, defined within the primitive Brillouin zone, is considered to be a discrete or a continuous variable (we shall normally assume it to be discrete). N is the total number of spins, V is the volume, and the inversion symmetry of the Bravais lattice implies that $\mathcal{J}(\mathbf{q}) = \mathcal{J}(-\mathbf{q}) = \mathcal{J}^*(\mathbf{q})$. The maximum value of $\mathcal{J}(\mathbf{q})$ is assumed to be $\mathcal{J}(\mathbf{q}=\mathbf{0})$, in which case the equilibrium state at zero temperature, i.e. the ground state, is the ferromagnet:

$$\langle\mathbf{S}_i\rangle = S\,\hat{\mathbf{z}} \qquad \text{at} \quad T = 0, \qquad (3.4.3)$$

where $\hat{\mathbf{z}}$ is a unit vector along the z-axis, which is established as the direction of magnetization by an infinitesimal magnetic field. This result is exact, but as soon as the temperature is increased above zero, it is necessary to make a number of approximations. As a first step, we introduce the thermal expectation-values $\langle \mathbf{S}_i \rangle = \langle \mathbf{S} \rangle$ in the Hamiltonian which, after a simple rearrangement of terms, can be written

$$\mathcal{H} = \sum_i \mathcal{H}_i - \frac{1}{2} \sum_{i \neq j} \mathcal{J}(ij) (\mathbf{S}_i - \langle \mathbf{S} \rangle) \cdot (\mathbf{S}_j - \langle \mathbf{S} \rangle), \tag{3.4.4a}$$

with

$$\mathcal{H}_i = -S_i^z \, \mathcal{J}(\mathbf{0}) \langle S^z \rangle + \tfrac{1}{2} \mathcal{J}(\mathbf{0}) \langle S^z \rangle^2, \tag{3.4.4b}$$

and $\langle \mathbf{S} \rangle = \langle S^z \rangle \, \hat{\mathbf{z}}$. In the mean-field approximation, discussed in the previous chapter, the dynamic correlation between spins on different sites is neglected. This means that the second term in (3.4.4*a*) is disregarded, reducing the original many-spin Hamiltonian to a sum of N independent single-spin Hamiltonians (3.4.4*b*). In this approximation, $\langle S^z \rangle$ is determined by the self-consistent equation

$$\langle S^z \rangle = \sum_{M=-S}^{+S} M \, e^{\beta M \mathcal{J}(\mathbf{0}) \langle S^z \rangle} \Big/ \sum_{M=-S}^{+S} e^{\beta M \mathcal{J}(\mathbf{0}) \langle S^z \rangle} \tag{3.4.5a}$$

(the last term in (3.4.4*b*) does not influence the thermal average) which, in the limit of low temperatures, is

$$\langle S^z \rangle \simeq S \, - \, e^{-\beta S \mathcal{J}(\mathbf{0})}. \tag{3.4.5b}$$

In order to incorporate the influence of two-site correlations, to leading order, we consider the Green function

$$G^{\pm}(ii', t) = \langle\langle S_i^+(t) \, ; \, S_{i'}^- \rangle\rangle. \tag{3.4.6}$$

According to (3.3.14*a*), the variation in time of $G^{\pm}(ii', t)$ depends on the operator

$$[\, S_i^+ \, , \, \mathcal{H} \,] = -\frac{1}{2} \sum_j \mathcal{J}(ij) \left(-2 S_i^+ S_j^z + 2 S_i^z S_j^+ \right).$$

The introduction of this commutator in the equation of motion (3.3.14*a*) leads to a relation between the original Green function and a new, more elaborate Green function. Through its equation of motion, this new function may be expressed in terms of yet another. The power of the exchange coupling in the Green functions which are generated in this way is raised by one in each step, and this procedure leads to an infinite hierarchy of coupled functions. An approximate solution may be obtained by utilizing the condition that the expectation value of S_i^z is close to its saturation value at low temperatures. Thus, in this limit,

S_i^z must be nearly independent of time, i.e. $S_i^z \simeq \langle S^z \rangle$. In this *random-phase approximation* (RPA) the commutator reduces to

$$[\, S_i^+ \,,\, \mathcal{H} \,] \simeq -\sum_j \mathcal{J}(ij)\langle S^z\rangle \left(S_j^+ - S_i^+\right),$$

and the equations of motion lead to the following linear set of equations:

$$\begin{aligned} \hbar\omega G^{\pm}(ii',\omega) + \sum_j \mathcal{J}(ij)\langle S^z\rangle \, \left\{G^{\pm}(ji',\omega) - G^{\pm}(ii',\omega)\right\} \\ = \langle\, [\, S_i^+ \,,\, S_{i'}^- \,]\,\rangle = 2\langle S^z\rangle \, \delta_{ii'}. \end{aligned} \tag{3.4.7}$$

The infinite set of RPA equations is diagonal in reciprocal space. Introducing the Fourier transform

$$G^{\pm}(\mathbf{q},\omega) = \sum_{i'} G^{\pm}(ii',\omega)\, e^{-i\mathbf{q}\cdot(\mathbf{R}_i - \mathbf{R}_{i'})}, \tag{3.4.8}$$

we obtain

$$\hbar\omega G^{\pm}(\mathbf{q},\omega) + \langle S^z\rangle \, \left\{\mathcal{J}(\mathbf{q})G^{\pm}(\mathbf{q},\omega) - \mathcal{J}(\mathbf{0})G^{\pm}(\mathbf{q},\omega)\right\} = 2\langle S^z\rangle,$$

or

$$G^{\pm}(\mathbf{q},\omega) = \lim_{\epsilon\to 0^+} \frac{2\langle S^z\rangle}{\hbar\omega + i\hbar\epsilon - E_{\mathbf{q}}}, \tag{3.4.9}$$

where the *dispersion relation* is

$$E_{\mathbf{q}} = \langle S^z\rangle \left\{\mathcal{J}(\mathbf{0}) - \mathcal{J}(\mathbf{q})\right\}. \tag{3.4.10}$$

Introducing the susceptibility $\chi_{+-}(\mathbf{q},\omega) = -G^{\pm}(\mathbf{q},\omega)$, we obtain

$$\chi_{+-}(\mathbf{q},\omega) = \frac{2\langle S^z\rangle}{E_{\mathbf{q}} - \hbar\omega} + i\pi\, 2\langle S^z\rangle\, \delta(\hbar\omega - E_{\mathbf{q}}). \tag{3.4.11a}$$

Defining $\chi_{-+}(\mathbf{q},\omega)$ analogously to $\chi_{+-}(\mathbf{q},\omega)$, but with S^+ and S^- interchanged, we obtain similarly, or by the use of the symmetry relation (3.2.15),

$$\chi_{-+}(\mathbf{q},\omega) = \frac{2\langle S^z\rangle}{E_{\mathbf{q}} + \hbar\omega} - i\pi\, 2\langle S^z\rangle\, \delta(\hbar\omega + E_{\mathbf{q}}), \tag{3.4.11b}$$

so that the absorptive susceptibility is

$$\chi''_{+-}(\mathbf{q},\omega) = -\chi''_{-+}(\mathbf{q},-\omega) = 2\pi\, \langle S^z\rangle\, \delta(\hbar\omega - E_{\mathbf{q}}). \tag{3.4.11c}$$

The above susceptibilities do not correspond directly to physical observables but, for instance, $\chi_{xx}(\mathbf{q},\omega)$ (where S^+ and S^- are both replaced by S_x) does. It is straightforward to see (by symmetry or by direct verification) that $\chi_{++}(\mathbf{q},\omega) = \chi_{--}(\mathbf{q},\omega) \equiv 0$, and hence

$$\chi_{xx}(\mathbf{q},\omega) = \chi_{yy}(\mathbf{q},\omega) = \frac{1}{4}\big\{\chi_{+-}(\mathbf{q},\omega) + \chi_{-+}(\mathbf{q},\omega)\big\}.$$

The presence of two-site correlations influences the thermal average $\langle S^z\rangle$. A determination of the correction to the MF result (3.4.5*b*) for $\langle S^z\rangle$, leading to a *self-consistent RPA* result for the transverse susceptibility, requires a relation between $\langle S^z\rangle$ and the susceptibility functions deduced above. The spin commutator-relation, $[\,S_i^+\,,\,S_{i'}^-\,] = 2S^z\,\delta_{ii'}$, turns out to be satisfied identically, and thus leads to no additional conditions. Instead we consider the *Wortis expansion*

$$S_i^z = S - \frac{1}{2S}S_i^-S_i^+ - \frac{1}{8S^2(S-\frac{1}{2})}(S_i^-)^2(S_i^+)^2 - \cdots \tag{3.4.12}$$

for which the matrix elements between the p lowest single-spin (or MF) levels are correct, where $p \le 2S+1$ is the number of terms in the expansion. Using (3.4.11), we find from the fluctuation–dissipation theorem (3.2.18):

$$\begin{aligned}\langle S_i^-S_i^+\rangle =& \frac{1}{N}\sum_{\mathbf{q}} S_{-+}(\mathbf{q},t=0)\\ =& \frac{1}{N}\sum_{\mathbf{q}}\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{1}{1-e^{-\beta\hbar\omega}}\chi''_{-+}(\mathbf{q},\omega)d(\hbar\omega) = 2\langle S^z\rangle\,\Phi,\end{aligned} \tag{3.4.13a}$$

with

$$\Phi = \frac{1}{N}\sum_{\mathbf{q}} n_{\mathbf{q}} \qquad ; \quad n_{\mathbf{q}} = \frac{1}{e^{\beta E_{\mathbf{q}}}-1}, \tag{3.4.13b}$$

where $n_{\mathbf{q}}$ is the population factor for bosons of energy $E_{\mathbf{q}}$. If $S = \frac{1}{2}$, then S^z is determined by the two first terms of (3.4.12), and

$$\langle S^z\rangle = S - \Phi\langle S^z\rangle/S,$$

or

$$\langle S^z\rangle = S^2/(S+\Phi) \simeq \tfrac{1}{2} - \Phi + 2\Phi^2 - \cdots$$

In general one may use a 'Hartree–Fock decoupling', $\langle(S_i^-)^2(S_i^+)^2\rangle \simeq 2(\langle S_i^-S_i^+\rangle)^2$, of the higher-order terms in (3.4.13) in order to show that

$$\langle S^z\rangle = S - \Phi + (2S+1)\Phi^{2S+1} - \cdots \simeq S - \frac{1}{N}\sum_{\mathbf{q}} n_{\mathbf{q}}, \tag{3.4.14}$$

where the *kinematic correction*, of the order Φ^{2S+1}, due to the limited number of single-spin states, which is neglected in this expression, is unimportant when $S \geq 1$. Utilizing the Hartree–Fock decoupling once more to write $\langle S_i^z S_j^z \rangle_{(i \neq j)} \simeq \langle S^z \rangle^2 \simeq S^2 - 2\langle S^z \rangle \Phi$, we find the internal energy to be

$$\begin{aligned} U = \langle \mathcal{H} \rangle &= -\tfrac{1}{2} N \mathcal{J}(\mathbf{0}) S^2 + \sum_{\mathbf{q}} E_{\mathbf{q}} \, n_{\mathbf{q}} \\ &= -\tfrac{1}{2} N \mathcal{J}(\mathbf{0}) S(S+1) + \sum_{\mathbf{q}} E_{\mathbf{q}} (n_{\mathbf{q}} + \tfrac{1}{2}). \end{aligned} \tag{3.4.15}$$

The second form, expressing the effect of the zero-point motion, is derived using $\mathcal{J}(ii) = \frac{1}{N} \sum_{\mathbf{q}} \mathcal{J}(\mathbf{q}) \equiv 0$.

The thermodynamic properties of the Heisenberg ferromagnet are determined by (3.4.10), (3.4.14), and (3.4.15), which are all valid at low temperatures. In a cubic crystal, the energy dispersion $E_{\mathbf{q}}$ is isotropic and proportional to q^2 in the long wavelength limit, and (3.4.14) then predicts that the magnetization $\langle S^z \rangle$ decreases from its saturation value as $T^{3/2}$. The specific heat is also found to be proportional to $T^{3/2}$. The thermodynamic quantities have a very different temperature dependence from the exponential behaviour (3.4.5*b*) found in the MF approximation. This is due to the presence of elementary excitations, which are easily excited thermally in the long wavelength limit, since $E_{\mathbf{q}} \to 0$ when $\mathbf{q} \to \mathbf{0}$ in the RPA. These normal modes, which are described as *spin waves*, behave in most aspects (disregarding the kinematic effects) as non-conserved Bose-particles, and they are therefore also called *magnons*.

We shall not present a detailed discussion of the low-temperature properties of the Heisenberg ferromagnet. Further details may be found in, for instance, Marshall and Lovesey (1971), and a quite complete treatment is given by Tahir-Kheli (1976). The RPA model is correct at $T = 0$ where $\langle S^z \rangle = S$, but as soon as the temperature is increased, the magnons start to interact with each other, giving rise to finite lifetimes, and the temperature dependence of the excitation energies is modified (or *renormalized*). The temperature dependence of $E_{\mathbf{q}} = E_{\mathbf{q}}(T)$ is responsible for the leading order 'dynamic' corrections to $\langle S^z \rangle$ and to the heat capacity. A more accurate calculation, which we will present in Section 5.2, adds an extra term to the dispersion:

$$E_{\mathbf{q}} = \langle S^z \rangle \{\mathcal{J}(\mathbf{0}) - \mathcal{J}(\mathbf{q})\} + \frac{1}{N} \sum_{\mathbf{k}} \{\mathcal{J}(\mathbf{k}) - \mathcal{J}(\mathbf{k}+\mathbf{q})\} \, n_{\mathbf{k}}, \tag{3.4.16}$$

from which the heat capacity of this non-interacting Bose-gas can be determined as

$$C = \partial U / \partial T = \sum_{\mathbf{q}} E_{\mathbf{q}} \, dn_{\mathbf{q}} / dT. \tag{3.4.17}$$

We note that there are corrections to U, given by (3.4.15), of second order in Φ. The low-temperature properties, as determined by (3.4.14), (3.4.16), and (3.4.17), agree with the systematic expansion performed by Dyson (1956), including the leading-order dynamical correction of fourth power in T (in the cubic case), except for a minor kinematic correction which is negligible for $S \geq 1$.

3.5 The random-phase approximation

Earlier in this chapter, we have demonstrated that many experimentally observable properties of solids can be expressed in terms of two-particle correlation functions. Hence it is of great importance to be able to calculate these, or the related Green functions, for realistic systems. We shall therefore consider the determination of the generalized susceptibility for rare earth magnets, using the random-phase approximation which was introduced in the last section, and conclude the chapter by applying this theory to the simple Heisenberg model, in which the single-ion anisotropy is neglected.

3.5.1 The generalized susceptibility in the RPA

The starting point for the calculation of the generalized susceptibility is the (effective) Hamiltonian for the angular momenta which, as usual, we write as a sum of single- and two-ion terms:

$$\mathcal{H} = \sum_i \mathcal{H}_{\mathrm{J}}(\mathbf{J}_i) - \tfrac{1}{2} \sum_{i \neq j} \mathcal{J}(ij)\, \mathbf{J}_i \cdot \mathbf{J}_j. \tag{3.5.1}$$

For our present purposes, it is only necessary to specify the two-ion part and, for simplicity, we consider only the Heisenberg interaction. As in Section 2.2, we introduce the thermal expectation values $\langle \mathbf{J}_i \rangle$ in the Hamiltonian, which may then be written

$$\mathcal{H} = \sum_i \mathcal{H}_{\mathrm{MF}}(i) - \tfrac{1}{2} \sum_{i \neq j} \mathcal{J}(ij)\, (\mathbf{J}_i - \langle \mathbf{J}_i \rangle) \cdot (\mathbf{J}_j - \langle \mathbf{J}_j \rangle), \tag{3.5.2}$$

where

$$\mathcal{H}_{\mathrm{MF}}(i) = \mathcal{H}_{\mathrm{J}}(\mathbf{J}_i) - \big(\mathbf{J}_i - \tfrac{1}{2}\langle \mathbf{J}_i \rangle\big) \cdot \sum_j \mathcal{J}(ij) \langle \mathbf{J}_j \rangle. \tag{3.5.3}$$

From the mean-field Hamiltonians $\mathcal{H}_{\mathrm{MF}}(i)$, we may calculate $\langle \mathbf{J}_i \rangle$ as before. The Hamiltonian (3.5.3) also determines the dynamic susceptibility of the ith ion, in the form of a Cartesian tensor $\overline{\overline{\chi}}{}_i^{o}(\omega)$, according to eqns (3.3.4–6), with $\hat{A}$ and $\hat{B}$ set equal to the angular-momentum components $J_{i\alpha}$. We wish to calculate the linear response $\langle \mathbf{J}_i(t) \rangle$ of the system to a small perturbative field $\mathbf{h}_j(t) = g\mu_B \mathbf{H}_j(t)$ (the Zeeman term due to a stationary field is taken as included in $\mathcal{H}_{\mathrm{J}}(\mathbf{J}_i)$). From (3.5.2), we may extract all terms depending on $\mathbf{J}_i$ and collect them in an effective Hamiltonian $\mathcal{H}_i$, which determines the time-dependence of $\mathbf{J}_i$. Transformed to the Heisenberg picture, this Hamiltonian is

$$\mathcal{H}_i(t) = \mathcal{H}_{\mathrm{MF}}(i,t) - \big(\mathbf{J}_i(t) - \langle \mathbf{J}_i \rangle\big) \cdot \Big(\sum_j \mathcal{J}(ij)(\mathbf{J}_j(t) - \langle \mathbf{J}_j \rangle) + \mathbf{h}_i(t) \Big). \tag{3.5.4}$$

We note that a given site i appears twice in the second term of (3.5.2), and that the additional term $\langle \mathbf{J}_i \rangle \cdot \mathbf{h}_i$ has no consequences in the limit when $\mathbf{h}_i$ goes to zero. The differences $\mathbf{J}_j(t) - \langle \mathbf{J}_j(t) \rangle$ fluctuate in a virtually uncorrelated manner from ion to ion, and their contribution to the sum in (3.5.4) is therefore small. Thus, to a good approximation, these fluctuations may be neglected, corresponding to replacing $\mathbf{J}_j(t)$ in (3.5.4) by $\langle \mathbf{J}_j(t) \rangle$ (when $j \neq i$). This is just the random-phase approximation (RPA), introduced in the previous section, and so called on account of the assumption that $\mathbf{J}_j(t) - \langle \mathbf{J}_j(t) \rangle$ may be described in terms of a random phase-factor. It is clearly best justified when the fluctuations are small, i.e. at low temperatures, and when many sites contribute to the sum, i.e. in three-dimensional systems with long-range interactions. The latter condition reflects the fact that an increase in the number of (nearest) neighbours improves the resemblance of the sum in (3.5.4) to an ensemble average. If we introduce the RPA in eqn (3.5.4), the only dynamical variable which remains is $\mathbf{J}_i(t)$, and the Hamiltonian becomes equivalent to $\mathcal{H}_{\mathrm{MF}}(i)$, except that the probing field $\mathbf{h}_i(t)$ is replaced by an effective field $\mathbf{h}_i^{\mathrm{eff}}(t)$. With $\langle \mathbf{J}_i(\omega) \rangle$ defined as the Fourier transform of $\langle \mathbf{J}_i(t) \rangle - \langle \mathbf{J}_i \rangle$, then, according to eqn (3.1.9),

$$\langle \mathbf{J}_i(\omega) \rangle = \overline{\overline{\chi}}{}_i^{o}(\omega) \mathbf{h}_i^{\mathrm{eff}}(\omega),$$

where the effective field is

$$\mathbf{h}_i^{\mathrm{eff}}(\omega) = \mathbf{h}_i(\omega) + \sum_j \mathcal{J}(ij) \langle \mathbf{J}_j(\omega) \rangle. \tag{3.5.5}$$

This may be compared with the response determined by the two-ion susceptibility functions of the system, defined such that

$$\langle \mathbf{J}_i(\omega) \rangle = \sum_j \overline{\overline{\chi}}(ij,\omega)\, \mathbf{h}_j(\omega). \tag{3.5.6}$$

The two ways of writing the response should coincide for all $\mathbf{h}_j(\omega)$, which implies that, within the RPA,

$$\overline{\overline{\chi}}(ij,\omega) = \overline{\overline{\chi}}{}_i^{o}(\omega) \Big(\delta_{ij} + \sum_{j'} \mathcal{J}(ij')\, \overline{\overline{\chi}}(j'j,\omega) \Big). \tag{3.5.7}$$

This self-consistent equation may be solved under various conditions. For convenience, we shall consider here only the uniform case of a ferro- or paramagnet, where $\mathcal{H}_{\mathrm{MF}}(i)$ is the same for all the ions, i.e. $\langle \mathbf{J}_i \rangle = \langle \mathbf{J} \rangle$ and $\overline{\overline{\chi}}{}_i^o(\omega) = \overline{\overline{\chi}}{}^o(\omega)$, in which case we get the final result

$$\overline{\overline{\chi}}(\mathbf{q}, \omega) = \left\{ 1 - \overline{\overline{\chi}}{}^o(\omega) \mathcal{J}(\mathbf{q}) \right\}^{-1} \overline{\overline{\chi}}{}^o(\omega). \tag{3.5.8}$$

Here 1 is the unit matrix, and we have used the Fourier transform (3.4.2) of $\mathcal{J}(ij)$

$$\mathcal{J}(\mathbf{q}) = \sum_j \mathcal{J}(ij)\, e^{-i\mathbf{q}\cdot(\mathbf{R}_i - \mathbf{R}_j)}. \tag{3.5.9}$$

In the RPA, the effects of the surrounding ions are accounted for by a time-dependent molecular field, which self-consistently enhances the response of the isolated ions. The above results are derived from a kind of hybrid MF-RPA theory, as the single-ion susceptibility $\overline{\overline{\chi}}{}_i^o(\omega)$ is still determined in terms of the MF expectation values. A self-consistent RPA theory might be more accurate but, as we shall see, gives rise to further problems. At high temperatures (or close to a phase transition), the description of the dynamical behaviour obtained in the RPA is incomplete, because the thermal fluctuations introduce damping effects which are not included. However, the static properties may still be described fairly accurately by the above theory, because the MF approximation is correct to leading order in $\beta = 1/k_B T$.

The RPA, which determines the excitation spectrum of the many-body system to leading order in the two-ion interactions, is simple to derive and is of general utility. Historically, its applicability was appreciated only gradually, in parallel with the experimental study of a variety of systems, and results corresponding to eqn (3.5.8) were presented independently several times in the literature in the early 1970s (Fulde and Perschel 1971, 1972; Haley and Erdös 1972; Purwins *et al.* 1973; Holden and Buyers 1974). The approach to this problem in the last three references is very similar, and we will now present it, following most closely the account given by Bak (1974).

We start by considering the MF Hamiltonian defined by (3.5.3). The basis in which $\mathcal{H}_{\mathrm{MF}}(i)$ is diagonal is denoted $|\nu_i>$; $\nu = 0, 1, \ldots, 2J$, and we assume that $\mathcal{H}_{\mathrm{MF}}(i)$ is the same for all the ions:

$$\mathcal{H}_{\mathrm{MF}}(i) |\nu_i> = E_\nu |\nu_i>, \tag{3.5.10}$$

with E_ν independent of the site index i . The eigenvalue equation defines the *standard-basis* operators

$$a_{\nu\mu}(i) = |\nu_i>< \mu_i|, \tag{3.5.11}$$

in terms of which $\mathcal{H}_{\rm MF}(i) = \sum_\nu E_\nu a_{\nu\nu}(i)$. Defining the matrix-elements

$$\mathbf{M}_{\nu\mu} = < \nu_i | \mathbf{J}_i - \langle \mathbf{J}_i \rangle | \mu_i >, \tag{3.5.12}$$

we may write

$$\mathbf{J}_i - \langle \mathbf{J}_i \rangle = \sum_{\nu\mu} \mathbf{M}_{\nu\mu}\, a_{\nu\mu}(i),$$

and hence

$$\mathcal{H} = \sum_i \sum_\nu E_\nu\, a_{\nu\nu}(i) - \frac{1}{2} \sum_{ij} \sum_{\nu\mu} \sum_{\nu'\mu'} \mathcal{J}(ij)\, \mathbf{M}_{\nu\mu} \cdot \mathbf{M}_{\nu'\mu'}\, a_{\nu\mu}(i)\, a_{\nu'\mu'}(j). \tag{3.5.13}$$

We have expressed $\mathcal{H}$ in terms of the standard-basis operators, as we now wish to consider the Green functions $G_{\nu\mu,rs}(ii',\omega) = \langle\langle a_{\nu\mu}(i)\, ; \, a_{rs}\,(i')\rangle\rangle$. According to (3.3.14), their equations of motion are

$$\hbar\omega\, G_{\nu\mu,rs}(ii',\omega) - \langle\langle\, [\, a_{\nu\mu}(i)\, , \, \mathcal{H}\,]\, ; \, a_{rs}(i')\rangle\rangle = \langle\, [\, a_{\nu\mu}(i)\, , \, a_{rs}(i')\,]\, \rangle. \tag{3.5.14}$$

The MF basis is orthonormal, and the commutators are

$$[\, a_{\nu\mu}(i)\, , \, a_{rs}(i')\,] = \delta_{ii'} \{ \delta_{\mu r} a_{\nu s}(i) - \delta_{s\nu} a_{r\mu}(i) \},$$

so we obtain

$$\begin{aligned} &\{\hbar\omega - (E_\mu - E_\nu)\} G_{\nu\mu,rs}(ii',\omega) \\ &\quad + \sum_j \mathcal{J}(ij) \sum_{\xi\nu'\mu'} \langle\langle \{ a_{\nu\xi}(i) \mathbf{M}_{\mu\xi} - a_{\xi\mu}(i) \mathbf{M}_{\xi\nu} \} \cdot \mathbf{M}_{\nu'\mu'}\, a_{\nu'\mu'}(j)\, ; \, a_{rs}(i')\rangle\rangle \\ &\qquad = \delta_{ii'} \langle \delta_{\mu r}\, a_{\nu s}(i) - \delta_{s\nu}\, a_{r\mu}(i) \rangle. \end{aligned} \tag{3.5.15}$$

In order to solve these equations, we make an *RPA decoupling* of the higher-order Green functions:

$$\begin{aligned} &\langle\langle a_{\nu\xi}(i)\, a_{\nu'\mu'}(j)\, ; \, a_{rs}(i')\rangle\rangle_{i\neq j} \simeq \\ &\quad \langle a_{\nu\xi}(i)\rangle \langle\langle a_{\nu'\mu'}(j)\, ; \, a_{rs}(i')\rangle\rangle + \langle a_{\nu'\mu'}(j)\rangle \langle\langle a_{\nu\xi}(i)\, ; \, a_{rs}(i')\rangle\rangle. \end{aligned} \tag{3.5.16}$$

This equation is correct in the limit where two-ion correlation effects can be neglected, i.e. when the ensemble averages are determined by the MF Hamiltonian. The decoupling is equivalent to the approximation made above, when $\mathbf{J}_j(t)$ in (3.5.4) was replaced by $\langle \mathbf{J}_j(t)\rangle$. The thermal expectation value of a single-ion quantity $\langle a_{\nu\mu}(i)\rangle$ is independent of i, and to leading order it is determined by the MF Hamiltonian:

$$\langle a_{\nu\mu}\rangle \simeq \langle a_{\nu\mu}\rangle_0 = \frac{1}{Z}\, {\rm Tr}\Big\{ e^{-\beta\mathcal{H}({\rm MF})}\, a_{\nu\mu} \Big\} = \delta_{\nu\mu}\, n_\nu, \tag{3.5.17}$$

and correspondingly $\langle \mathbf{J} \rangle$ in (3.5.12) is assumed to take the MF value $\langle \mathbf{J} \rangle_0$. Here Z is the partition function of the MF Hamiltonian, and thus n_ν is the population factor of the νth MF level. With the two approximations (3.5.16) and (3.5.17), and the condition that $\sum_{\nu'\mu'} \langle \mathbf{M}_{\nu'\mu'} a_{\nu'\mu'}(j) \rangle_0 = \langle \mathbf{J}_j - \langle \mathbf{J}_j \rangle_0 \rangle_0 = 0$ by definition, (3.5.15) is reduced to a closed set of equations by a Fourier transformation:

$$\begin{aligned}&\{\hbar\omega - (E_\mu - E_\nu)\} G_{\nu\mu,rs}(\mathbf{q},\omega)\\ &\quad + \sum_{\nu'\mu'} \mathcal{J}(\mathbf{q})(n_\nu - n_\mu)\mathbf{M}_{\mu\nu} \cdot \mathbf{M}_{\nu'\mu'}\, G_{\nu'\mu',rs}(\mathbf{q},\omega) = (n_\nu - n_\mu)\,\delta_{\mu r}\delta_{\nu s}.\end{aligned} \tag{3.5.18}$$

We now show that these equations lead to the same result (3.5.8) as found before. The susceptibility, expressed in terms of the Green functions, is

$$\overline{\overline{\chi}}(\mathbf{q},\omega) = -\sum_{\nu\mu,rs} \mathbf{M}_{\nu\mu}\mathbf{M}_{rs} G_{\nu\mu,rs}(\mathbf{q},\omega). \tag{3.5.19}$$

$\mathbf{M}_{\nu\mu}\mathbf{M}_{rs}$ is the dyadic vector-product, with the $(\alpha\beta)$-component given by $(\mathbf{M}_{\nu\mu}\mathbf{M}_{rs})_{\alpha\beta} = (M_{\nu\mu})_\alpha (M_{rs})_\beta$. Further, from eqns (3.3.4–6), the MF susceptibility is

$$\overline{\overline{\chi}}^{\,o}(\omega) = \sum_{\nu\mu}^{E_\nu \neq E_\mu} \frac{\mathbf{M}_{\nu\mu}\mathbf{M}_{\mu\nu}}{E_\mu - E_\nu - \hbar\omega}(n_\nu - n_\mu) + \sum_{\nu\mu}^{E_\nu = E_\mu} \mathbf{M}_{\nu\mu}\mathbf{M}_{\mu\nu}\beta\, n_\nu\, \delta_{\omega 0}. \tag{3.5.20}$$

Multiplying (3.5.18) by $\mathbf{M}_{\nu\mu}\mathbf{M}_{rs}/(E_\mu - E_\nu - \hbar\omega)$, and summing over $(\nu\mu, rs)$, we get (for $\omega \neq 0$)

$$\overline{\overline{\chi}}(\mathbf{q},\omega) - \overline{\overline{\chi}}^{\,o}(\omega)\mathcal{J}(\mathbf{q})\,\overline{\overline{\chi}}(\mathbf{q},\omega) = \overline{\overline{\chi}}^{\,o}(\omega), \tag{3.5.21}$$

in accordance with (3.5.8). Special care must be taken in the case of degeneracy, $E_\mu = E_\nu$, due to the resulting singular behaviour of (3.5.18) around $\omega = 0$. For $\omega \neq 0$, $G_{\nu\mu,rs}(\mathbf{q},\omega)$ vanishes identically if $E_\mu = E_\nu$, whereas $G_{\nu\mu,rs}(\mathbf{q},\omega = 0)$ may be non-zero. The correct result, in the zero frequency limit, can be found by putting $E_\mu - E_\nu = \delta$ in (3.5.18), so that $n_\nu - n_\mu = n_\nu(1 - e^{-\beta\delta}) \simeq \beta n_\nu \delta$. Dividing (3.5.18) by δ, and taking the limit $\delta \to 0$, we obtain in the degenerate case $E_\nu = E_\mu$:

$$-G_{\nu\mu,rs}(\mathbf{q},0) - \beta \sum_{\nu'\mu'} \mathcal{J}(\mathbf{q}) n_\nu \mathbf{M}_{\nu\mu} \cdot \mathbf{M}_{\nu'\mu'}\, G_{\nu'\mu',rs}(\mathbf{q},0) = \beta n_\nu\, \delta_{\mu r}\, \delta_{\nu s}. \tag{3.5.22}$$

Since $\overline{\overline{\chi}}(\mathbf{q},\omega)$ does not depend on the specific choice of state-vectors in the degenerate case, (3.5.22) must also apply for a single level, i.e. when $\mu = \nu$. It then follows that (3.5.18), when supplemented with (3.5.22), ensures that (3.5.21) is also valid at $\omega = 0$, as (3.5.22) accounts for the elastic contributions due to $\overline{\overline{\chi}}^{o}(\omega)$, proportional to $\delta_{\omega 0}$. This zero-frequency modification of the equations of motion was derived in this context in a slightly different way by Lines (1974a).

Although eqns (3.5.18) and (3.5.22) only lead to the result (3.5.8), derived previously in a simpler manner, the equations of motion clarify more precisely the approximations made, and they contain more information. They allow us to keep track in detail of the different transitions between the MF levels, which may be an advantage when performing actual calculations. Furthermore, the set of Green functions $G_{\nu\mu,rs}(\mathbf{q},\omega)$ is complete, and hence any magnetic single- or two-ion response function may be expressed as a linear combination of these functions.

In the derivation of the RPA result, we utilized two approximate equations, (3.5.16) and (3.5.17). The two approximations are consistent, as both equations are correct if two-ion correlation effects are negligible. However, the RPA Green functions contain implicitly two-ion correlations and, according to (3.3.7), we have in the linear response theory:

$$\begin{aligned}\langle a_{\nu\mu}(i)\, a_{rs}(j)\rangle - \langle a_{\nu\mu}(i)\rangle\langle a_{rs}(j)\rangle &= \\ \frac{1}{N}\sum_{\mathbf{q}} e^{i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)}\,\frac{1}{\pi}\int_{-\infty}^{\infty} &\frac{-1}{1-e^{-\beta\hbar\omega}} G''_{\nu\mu,rs}(\mathbf{q},\omega)d(\hbar\omega),\end{aligned} \qquad (3.5.23)$$

where, by the definition (3.2.11*b*),

$$G''_{\nu\mu,rs}(\mathbf{q},\omega) = \frac{1}{2i}\lim_{\epsilon\to 0^+}\big\{G_{\nu\mu,rs}(\mathbf{q},\omega+i\epsilon) - G_{rs,\nu\mu}(-\mathbf{q},-\omega+i\epsilon)\big\}.$$

Equation (3.5.23), with $i = j$, might be expected to give a better estimate of the single-ion average $\langle a_{\nu\mu}\rangle$ than that afforded by the MF approximation used in (3.5.17). If this were indeed the case, the accuracy of the theory could be improved by using this equation, in a self-consistent fashion, instead of (3.5.17), and this improvement would maintain most of the simplicity and general utility of the RPA theory. Unfortunately, such an improvement seems to occur only for the Heisenberg ferromagnet discussed previously, and the nearly-saturated anisotropic ferromagnet, which we will consider later. Equation (3.5.23) allows different choices of the Green functions $G_{\nu\mu,rs}(\mathbf{q},\omega)$ for calculating $\langle a_{\nu\nu}\rangle$, and the results in general depend on this choice. Furthermore, (3.5.23) may lead to non-zero values for $\langle a_{\nu\mu}(i)\, a_{rs}(i)\rangle$, when $\mu \neq r$, despite the fact that $< \mu_i\,|r_i > = 0$ by definition. The two-ion correlation effects which are neglected by the RPA decoupling in (3.5.18) might be as important, when using eqn (3.5.23) with $i = j$, as those effects which are accounted for by the RPA. Nevertheless, it might be possible that certain choices of the Green functions, or a linear combination of them, would lead to an accurate determination of $\langle a_{\nu\nu}\rangle$ (the most natural choice would be to use $G''_{\nu 0,0\nu}(\mathbf{q},\omega)$). However, a stringent justification of a specific choice would require an analysis of the errors introduced by the RPA decoupling. We conclude that a reliable improvement of the theory can only be obtained by a more accurate treatment of the higher-order Green functions than that provided by the RPA. General programs for accomplishing this have been developed, but they have only been carried through in the simplest cases, and we reserve the discussion of these analyses to subsequent sections, where a number of specific systems are considered.

3.5.2 MF-RPA theory of the Heisenberg ferromagnet

We conclude this chapter by applying the RPA to the Heisenberg model, thereby demonstrating the relation between (3.5.8) and the results presented in the previous section. In order to do this, we must calculate $\overline{\overline{\chi}}^{\,o}(\omega)$. The eigenstates of the MF Hamiltonian (3.4.4*b*) are $|S^z = M>$, with $M = -S, -S+1, \cdots, S$, and we neglect the constant contribution to the eigenvalues

$$E_M = -M\mathcal{J}(\mathbf{0})\langle S^z\rangle_0 = -M\Delta \qquad \text{with} \quad \Delta = \mathcal{J}(\mathbf{0})\langle S^z\rangle_0,$$

denoting the MF expectation-value (3.4.5*a*) of S^z by $\langle S^z\rangle_0$. According to (3.3.4*a*), we then have (only terms with $\alpha = M+1$ and $\alpha' = M$ contribute):

$$\begin{aligned}
\chi^o_{+-}(\omega) &= \sum_{M=-S}^{S-1} \frac{<M+1|S^+|M><M|S^-|M+1>}{E_M - E_{M+1} - \hbar\omega}\,(n_{M+1} - n_M) \\
&= \frac{1}{Z}\sum_{-S}^{S-1} \frac{S(S+1) - M(M+1)}{\Delta - \hbar\omega}\left(e^{\beta(M+1)\Delta} - e^{\beta M\Delta}\right) \\
&= \frac{1}{\Delta - \hbar\omega}\,\frac{1}{Z}\Big(\sum_{-S+1}^{S}\big\{S(S+1) - (M-1)M\big\}e^{\beta M\Delta} \\
&\qquad\qquad - \sum_{-S}^{S-1}\big\{S(S+1) - M(M+1)\big\}e^{\beta M\Delta}\Big) \\
&= \frac{1}{\Delta - \hbar\omega}\,\frac{1}{Z}\sum_{-S}^{S} 2Me^{\beta M\Delta} \;=\; \frac{2\langle S^z\rangle_0}{\Delta - \hbar\omega}\,,
\end{aligned}$$

as all the sums may be taken as extending from $-S$ to S. Similarly $\chi^o_{-+}(\omega) = \chi^o_{+-}(-\omega)$, whereas $\chi^o_{++}(\omega) = \chi^o_{--}(\omega) = 0$, from which we obtain

$$\chi^o_{xx}(\omega) = \chi^o_{yy}(\omega) = \frac{1}{4}\left\{\chi^o_{+-}(\omega) + \chi^o_{-+}(\omega)\right\} = \frac{\Delta\langle S^z\rangle_0}{\Delta^2 - (\hbar\omega)^2}, \quad (3.5.24a)$$

and

$$\chi^o_{xy}(\omega) = -\chi^o_{yx}(\omega) = \frac{i}{4}\left\{\chi^o_{+-}(\omega) - \chi^o_{-+}(\omega)\right\} = \frac{i\hbar\omega\langle S^z\rangle_0}{\Delta^2 - (\hbar\omega)^2}. \quad (3.5.24b)$$

We note here that $\chi^o_{xy}{}'(\omega)$ and $\chi^o_{xy}{}''(\omega)$, obtained by replacing ω by $\omega + i\epsilon$ and letting $\epsilon \to 0^+$, are both purely imaginary. Of the remaining components in $\overline{\overline{\chi}}{}^o(\omega)$, only $\chi^o_{zz}(\omega)$ is non-zero, and it comprises only an elastic contribution

$$\chi^o_{zz}(\omega) = \beta\,(\delta S^z)^2\delta_{\omega 0}, \qquad \text{with} \quad (\delta S^z)^2 \equiv \langle (S^z)^2\rangle_0 - \langle S^z\rangle_0^2. \quad (3.5.25)$$

Because $\chi^o_{\pm z}(\omega) = 0$, the RPA equation (3.5.8) factorizes into a 2×2 (xy)-matrix equation and a scalar equation for the zz-component. Inverting the (xy)-part of the matrix $\{1 - \overline{\overline{\chi}}{}^o(\omega)\,\mathcal{J}(\mathbf{q})\}$, we find

$$\chi_{xx}(\mathbf{q},\omega) = \frac{\chi^o_{xx}(\omega) - |\overline{\overline{\chi}}{}^o(\omega)|\,\mathcal{J}(\mathbf{q})}{1 - \{\chi^o_{xx}(\omega) + \chi^o_{yy}(\omega)\}\,\mathcal{J}(\mathbf{q}) + |\overline{\overline{\chi}}{}^o(\omega)|\,\mathcal{J}^2(\mathbf{q})},$$

where the determinant is

$$|\overline{\overline{\chi}}{}^o(\omega)| = \chi^o_{xx}(\omega)\chi^o_{yy}(\omega) - \chi^o_{xy}(\omega)\chi^o_{yx}(\omega) = \frac{\langle S^z\rangle_0^2}{\Delta^2 - (\hbar\omega)^2}.$$

By a straightforward manipulation, this leads to

$$\chi_{xx}(\mathbf{q},\omega) = \frac{E^0_{\mathbf{q}}\langle S^z\rangle_0}{(E^0_{\mathbf{q}})^2 - (\hbar\omega)^2}, \quad (3.5.26a)$$

with

$$E^0_{\mathbf{q}} = \Delta - \langle S^z\rangle_0\mathcal{J}(\mathbf{q}) = \langle S^z\rangle_0\{\mathcal{J}(\mathbf{0}) - \mathcal{J}(\mathbf{q})\}. \quad (3.5.26b)$$

The same result is obtained for $\chi_{yy}(\mathbf{q},\omega)$. We note that (3.5.26$a$) should be interpreted as

$$\chi_{xx}(\mathbf{q},\omega) = \tfrac{1}{2}\langle S^z\rangle_0 \lim_{\epsilon\to 0^+}\left(\frac{1}{E^0_{\mathbf{q}} - \hbar\omega - i\hbar\epsilon} + \frac{1}{E^0_{\mathbf{q}} + \hbar\omega + i\hbar\epsilon}\right).$$

This result is nearly the same as that deduced before, eqns (3.4.10–11), except that the RPA expectation-value $\langle S^z\rangle$ is replaced by its MF value $\langle S^z \rangle_0$, reflecting the lack of self-consistency in this analysis. As a supplement to the previous results, we find that

$$\chi_{zz}(\mathbf{q},\omega) = \frac{\chi^o_{zz}(\omega)}{1 - \chi^o_{zz}(\omega)\,\mathcal{J}(\mathbf{q})} = \frac{\beta(\delta S^z)^2}{1 - \beta(\delta S^z)^2\,\mathcal{J}(\mathbf{q})}\,\delta_{\omega 0}, \qquad (3.5.27a)$$

and the corresponding correlation function is

$$S_{zz}(\mathbf{q},\omega) = 2\pi\hbar \frac{(\delta S^z)^2}{1 - \beta(\delta S^z)^2\,\mathcal{J}(\mathbf{q})}\delta(\hbar\omega). \qquad (3.5.27b)$$

The zz-response vanishes in the zero-temperature limit and, in this approximation, it is completely elastic, since $(\delta S^z)^2$ is assumed independent of time. However, this assumption is violated by the dynamic correlation-effects due to the spin waves. For instance, the $(n = 1)$-sum-rule (3.3.18b) indicates that the second moment $\langle(\hbar\omega)^2\rangle_{zz}$ is non-zero, when $\mathbf{q} \neq \mathbf{0}$ and $T > 0$, which is not consistent with a spectral function proportional to $\delta(\hbar\omega)$.

Although this procedure leads to a less accurate analysis of the Heisenberg ferromagnet than that applied previously, it has the advantage that it is easily generalized, particularly by numerical methods, to models with single-ion anisotropy, i.e. where $\mathcal{H}_{\mathrm{J}}(\mathbf{J}_i)$ in (3.5.1) is non-zero. The simplicity of the RPA result (3.5.8), or of the more general expression (3.5.7), furthermore makes it suitable for application to complex systems. As argued above, its validity is limited to low temperatures in systems with relatively large coordination numbers. However, these limitations are frequently of less importance than the possibility of making quantitative predictions of reasonable accuracy under realistic circumstances. Its utility and effectiveness will be amply demonstrated in subsequent chapters.