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 $\chi(\omega)$.