Linear response, generalized subsctibility and dispersion theory

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LINEAR RESPONSE, GENERALIZED SUSCEPTIBILITY AND DISPERSION THEORY

D. DES CLOIZEAUX
SERVICE DE PHYSIQUE THEORIQUE,
CENTRE D'ETUDES NUCLEAIRES,
SACLAY, FRANCE

Abstract

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1. LINEAR RESPONSE TO A DYNAMICAL DISTURBANCE. THEORY

1.1. Response function and generalized susceptibility

The system to which we apply an external force at a given time is assumed to be initially in a state of thermodynamic equilibrium. The unperturbed system is characterized by a density operator D which is non-negative definite with:

$$\mathrm{Tr}\, D = 1 \qquad (1.1)$$

The mean value of any operator B related to the system is given by:

$$\langle B \rangle = \mathrm{Tr}\,(DB) \qquad (1.2)$$

and therefore D determines the state of the system completely.

For a classical system, D would be a probability distribution in a phase space, rather than an operator, and each observable B would be a function defined in this space. In the classical limit, the formalism is not fundamentally different; consequently, we shall deal mainly with quantum systems, but the results will often be extended to classical systems by passing to the limit $\hbar = 0$.

When the system is at thermal equilibrium, the density operator must be:

$$D = Z^{-1} e^{-\beta H}$$

where H is the effective Hamiltonian and Z the partition function. Actually, if, for example, the system consists of a set of identical particles and if the number of particles is not fixed we must put:

$$H = \mathscr{H} - \mu N \tag{1.4}$$

where $\mathscr{H}$ is the true Hamiltonian, μ the chemical potential and N the number of particles.

Let us now apply a time-dependent external disturbance to the system. In this case, the Hamiltonian H is replaced by the time-dependent Hamiltonian H(t):

$$H(t) = H + v(t) \tag{1.5}$$

where v (t) is supposed to be a small perturbation. Since we are only interested in linear responses, we may assume without loss of generality that v (t) has the simple form:

$$v(t) = -a(t)\,A \tag{1.6}$$

where A is a constant operator, and a(t) a function of time representing a generalized external force and vanishing for remote times. Actually, it is convenient to assume that a (t) vanishes exponentially when t goes to infinity:

$$\exists\epsilon \text{ with } \epsilon > 0 \rightarrow \lim_{t \to -\infty} e^{-\epsilon t}\, a(t) = 0 \tag{1.7}$$

As a consequence of the perturbation, the average of the operator B becomes time-dependent and its mean value, at time t, will be denoted by $\langle B(t)\rangle_v$. The linear relation between this quantity and the small perturbing potential can be written in the form:

$$\langle B(t)\rangle_v - \langle B\rangle = \int_{-\infty}^{+\infty} X_{BA}(t-t')\, a(t')\, dt' \tag{1.8}$$

where $X_{BA}(t)$ is assumed to be a bounded function of t:

$$|X_{BA}(t)| < C \tag{1.9}$$

This mathematical assumption expresses the fact that the system reacts in a rather smooth way to any percussion, i.e. any strong instantaneous perturbation.

Thus, $X_{BA}(t)$ defines a linear response. Owing to causality requirements, we have, however,

$$X_{BA}(t) = 0 \qquad t < 0 \tag{1.10}$$

Therefore, the preceding equation must be written:

$$\langle B(t)\rangle_v - \langle B\rangle = \int_{-\infty}^{t} X_{BA}(t-t')\, a(t')\, dt' \tag{1.11}$$

This relation takes a very simple form if we use Fourier transforms. For this purpose, it is useful to associate a function $X_{BA}(z)$ of the complex variable $z = z' + iz''$ with $X_{BA}(t)$: for $z'' > 0$ this function is defined by the following (Lebesgue) integral:

$$X_{BA}(z) = \int_{-\infty}^{\infty} X_{BA}(t)\, e^{izt}\, dt \equiv \int_{0}^{\infty} X_{BA}(t)\, e^{izt}\, dt \tag{1.12}$$

As $X_{BA}(t)$ is assumed to be bounded, the integral converges uniformly in any domain $z'' \geq \epsilon > 0$ and therefore defines an analytic function $X_{BA}(z)$ of z in the upper part of the complex plane of z (i.e. $z'' > 0$).

Actually, by putting $z = \omega + i\epsilon$, we can write:

Fourier

$$X_{BA}(\omega + i\epsilon) = \int_{-\infty}^{+\infty} X_{BA}(t)\, e^{-\epsilon t}\, e^{i\omega t}\, dt \tag{1.13}$$

which shows that for a given value of ϵ, $X_{BA}(\omega + i\epsilon)$ can be considered as the Fourier transform in ω of the function $X_{BA}(t)\exp(-\epsilon t)$. By passing to the limit $\epsilon \to 0$, we can define a function (or, in special cases, a distribution) $X_{BA}(\omega)$:

$$X_{BA}(\omega) \equiv X_{BA}(\omega + i0) \equiv \lim_{\epsilon \to 0} X_{BA}(\omega + i\epsilon) \tag{1.14}$$

which is the boundary value of the analytic function $X_{BA}(z)$ on the real axis.

Conversely, we have:

$$X_{BA}(t) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} X_{BA}(\omega)\, e^{-i\omega t}\, d\omega \tag{1.15}$$

With the same kind of notation, the Fourier transform $\alpha(\omega)$ of a(t) can be defined by:

$$\alpha(\omega) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} e^{i\omega t}\, e^{-0t}\, a(t)\, dt \equiv \lim_{\epsilon \to 0} \int_{-\infty}^{+\infty} dt\, e^{i\omega t}\, e^{-\epsilon t}\, a(t) \tag{1.16}$$

Thus, a(t) is equal to:

$$a(t) = \int_{-\infty}^{+\infty} \alpha(\omega)\, e^{-i\omega t}\, e^{+0t}\, d\omega \tag{1.17}$$

In the same way, the function $\langle B(t)\rangle - \langle B\rangle$ can be expressed in terms of its spectral distribution:

$$\langle B(t)\rangle_v - \langle B\rangle = \int_{-\infty}^{+\infty} \beta(\omega)\, e^{-i\omega t + 0t}\, dt \tag{1.18}$$

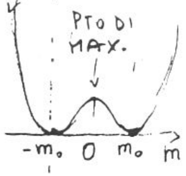
where $\beta(\omega)$ is given by:

$$\beta(\omega) = \frac{1}{2\pi}\int_{-\infty}^{+\infty}\Big[\langle B(t)\rangle_{\varphi} - \langle B\rangle\Big]e^{i\omega t + 0t}dt \qquad (1.19)$$

Then Eqs (1.8) and (1.9) can be written in the simple form:

energia libera per $T<T_c$

$$\beta(\omega) = \chi_{BA}(\omega)\,\alpha(\omega) \qquad (1.20)$$



$m=0$ è un minimo per $T>T_c$ mentre è un max. per $T<T_c$ ⇒ è instabile

Thus, $\chi_{BA}(\omega)$ can be regarded as a generalized susceptibility.

At this point, we must remark that the appearance of broken symmetries may sometimes obscure our simple picture. Let us consider, for example, a ferromagnetic system at a temperature below the Curie point. In the absence of any magnetic field, the average magnetic moment is zero, but if we apply to the system a very small magnetic field $\vec{b}_0$, a finite magnetic moment appears. Thus, the influence of this infinitesimal field changes, in a drastic way, the nature of the density operator. However, for a given value of $\vec{b}_0$, we can define a magnetic susceptibility $\chi(\omega, \vec{b}_0)$ which describes the variations of the magnetic moment produced by adding (for instance in the same direction) a small field $\vec{b}(t)$ to $\vec{b}_0$. Thus a magnetic susceptibility for a zero field can be defined as the limit of $\chi(\omega, \vec{b}_0)$ when $\vec{b}_0 \to 0$. The same kind of behaviour is to be expected when strong modifications of the state of a system can result from its interaction with infinitesimal symmetry-breaking external sources.

1.2. Reactive and absorptive part of a susceptibility. Definition

The response function $X_{BA}(t)$ can always be written in the form:

considerare caso $B = A$

$$\mathbb{R} \ni X_{BA}(t) = X'_{BA}(t) + iX''_{BA}(t) \qquad (1.21)$$

where by definition we have:

$$\begin{cases} X'_{BA}(-t) = X'_{AB}(t) & (1.22)\\ X''_{BA}(-t) = -X''_{AB}(t) & (1.23)\end{cases}$$

In fact, $X'_{BA}(t)$ and $X''_{BA}(t)$ are also defined by:

$$X'_{BA}(t) = \frac{1}{2}\Big[X_{BA}(t) + X_{AB}(-t)\Big] \qquad (1.24)$$

$$X''_{BA}(t) = -\frac{i}{2}\Big[X_{BA}(t) - X_{AB}(-t)\Big] \qquad (1.25)$$

For reasons which will be given later on, $X'_{BA}(t)$ and $X''_{BA}(t)$ will be called the reactive and the absorptive part of the response function, respectively. (Note that $X'_{BA}(t)$ is real and $X''_{BA}(t)$ is purely imaginary.)

In the same way, we define $\chi'_{BA}(\omega)$ and $\chi''_{BA}(\omega)$ by (see Eq. (1.13))

Se uso direttamente la trasf. di Fourier ($\epsilon = 0$) non devo mettere questo INCOMPRENSIBILE modulo di t

$$\chi'_{BA}(\omega) = \int_{-\infty}^{+\infty} X'_{BA}(t)\, e^{-0|t|}\, e^{i\omega t}\,dt \qquad (1.26)$$

$$\chi''_{BA}(\omega) = \int_{-\infty}^{+\infty} X''_{BA}(t)\, e^{-0|t|}\, e^{i\omega t}\,dt \qquad (1.27)$$

Thus, we have: $\chi'_{AB}(\omega) = \frac{1}{2}\int_{-\infty}^{+\infty}(X_{BA}(t) + X_{AB}(-t))e^{-0t}e^{i\omega t}dt$ → vedi appunti pag 114

$$\chi'_{BA}(\omega) = \frac{1}{2}\Big[\chi_{BA}(\omega) + \chi_{AB}(-\omega)\Big] \qquad (1.28)$$

$$\chi''_{BA}(\omega) = -\frac{i}{2}\Big[\chi_{BA}(\omega) - \chi_{AB}(-\omega)\Big] \qquad (1.29)$$

These quantities satisfy symmetry conditions:

$$\chi'_{BA}(\omega) = \chi'_{AB}(-\omega) = \Big[\chi'_{AB}(\omega)\Big]^* \qquad (1.30)$$

$$\chi''_{BA}(\omega) = -\chi''_{AB}(-\omega) = \Big[\chi''_{AB}(\omega)\Big]^* \qquad (1.31)$$

Thus, in the space of the operators A or B, $\chi'(\omega)$ and $\chi''(\omega)$ may be considered Hermitian.

Incidentally, we remark that $\chi'_{AA}(\omega)$ and $\chi''_{AA}(\omega)$ are both real. In this case, we have:

$$\chi'_{AA}(\omega) = \int_0^{\infty} \cos\omega t\; X_{AA}(t)\, e^{-0t}\,dt \qquad (1.32)$$

$$\chi''_{AA}(\omega) = \int_0^{\infty} \sin\omega t\; X_{AA}(t)\, e^{-0t}\,dt \qquad (1.33)$$

and, conversely:

$$X'_{AA}(t) = \frac{1}{\pi}\int_0^{\infty} \cos\omega t\; \chi'_{AA}(\omega)\,d\omega \qquad (1.34)$$

$$X''_{AA}(t) = -\frac{i}{\pi}\int_0^{\infty} \sin\omega t\; \chi''_{AA}(\omega)\,d\omega \qquad (1.35)$$

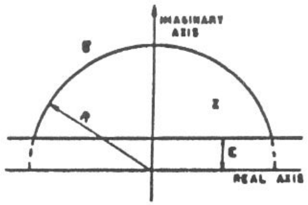


FIG. 1. Integration contour $\mathscr{C}$.

1.3. Kramers and Kronig dispersion relations

The causal nature of the response function implies relations between $\chi'_{BA}(\omega)$ and $\chi''_{BA}(\omega)$. These dispersion relations are derived by expressing the analytic function $\chi_{BA}(z)$ in terms of its boundary value $\chi_{BA}(\omega)$.

This result is usually obtained by writing $\chi_{BA}(z)$ as a Cauchy integral on the contour $\mathscr{C}$ of Fig. 1. This contour consists of a fraction of a line parallel to the real axis (at a distance ϵ above it) and of a fraction of a circle centered at the origin (radius R).

$$\chi_{BA}(z) = \frac{1}{2\pi i} \int_{\mathscr{C}} \frac{\chi_{AB}(x)}{x-z}\, dx \tag{1.36}$$

Now the Lebesgue lemma [1] says that $\chi(\omega + i\epsilon)$ which is the Fourier transform of the "good" function $X_{BA}(t)\exp(-\epsilon t)$ goes to zero when $|\omega| \to \infty$. On the other hand, since $X_{BA}(t)$ is bounded in the domain $z'' > \epsilon$, Phragmén-Lindelöf theorem [2] can be applied and it shows that in the domain $z'' > \epsilon$, the preceding conditions imply the uniform convergence of $\chi_{BA}(z)$ to zero when $|z| \to \infty$.

Let us then keep ϵ fixed and let R increase. We see that, as a consequence of the preceding remark, the integral on the circle must vanish in the limit $R \to \infty$. Therefore, in this limit, Eq. (1.35) becomes:

$$\chi_{BA}(z) = \frac{1}{2\pi i} \int_{-\infty}^{+\infty} \frac{\chi_{BA}(\omega + i\epsilon)}{\omega + i\epsilon - z}\, d\omega \tag{1.37}$$

The same result can be obtained by direct application of the following theorem given by Titchmarsh [3].

<u>Theorem</u>

Let $\Phi(z)$ be an analytic function regular for $y > 0$ and let

$$\int_{-\infty}^{+\infty} |\Phi(x+iy)^2|\, dx$$

exist and be bounded. Then, as $y \to 0$, $\Phi(x+iy)$ converges in mean toward a function $\Phi(x)$ and also $\Phi(x+iy) \to \Phi(x)$ for almost all x. For $y > 0$:

$$\Phi(z) = \frac{1}{2\pi i} \int_{-\infty}^{+\infty} \frac{\Phi(u)}{u-z}\, du \tag{1.38}$$

In order to apply this theorem, we put:

$$z = x + iy + i\epsilon \qquad y \geq 0 \tag{1.39}$$

$$\chi_{BA}(z) \equiv \chi_{BA}(x + iy + i\epsilon) = \Phi(x + iy) \tag{1.40}$$

On the other hand, as $\chi_{BA}(z' + iz'')$ is the Fourier transform in z' of $X_{BA}(t)\exp(-z''t)$, Parseval's theorem leads to the following condition for $z'' \geq \epsilon$:

$$\int_{-\infty}^{+\infty} |\chi_{BA}(z' + iz'')|^2\, dz' = 2\pi \int_{-\infty}^{+\infty} |X_{BA}(t)|^2 e^{-2z''t}\, dt \leq \frac{\pi \Psi^2}{z''} \leq \frac{\pi\Psi^2}{\epsilon} \tag{1.41}$$

Thus, Titchmarsh's theorem can be directly applied and we obtain Eq. (1.37) again.

In order to express $\chi_{BA}(z)$ in terms of $\chi_{BA}(\omega)$ which is a quantity of physical interest, we consider now the limit $\epsilon \to 0$. We note that if the function $X_{BA}(t)$ is square integrable, Titchmarsh's theorem can be applied for $\epsilon = 0$. Then, $\chi_{BA}(\omega)$ is also a function of square integrable modulus and we have:

$$\chi_{BA}(z) = \frac{1}{2\pi i} \int_{-\infty}^{+\infty} \frac{\chi_{BA}(\omega + i0)}{\omega + i0 - z}\, d\omega \equiv \frac{1}{2\pi i} \int_{-\infty}^{+\infty} \frac{\chi_{BA}(\omega)}{\omega - z}\, d\omega \tag{1.42}$$

Incidentally, we verify that the integral on the right hand side of this equation is convergent. However, if the square of $X_{BA}(t)$ is not integrable, the preceding equation can be given a meaning if we consider $\chi_{BA}(\omega)$ to be a distribution.

Finally, dispersion relations are obtained when z becomes real ($z'' \to +0$). We get:

$$\chi_{BA}(\omega) \equiv \chi_{BA}(\omega + i0) = \frac{1}{2\pi i} \int_{-\infty}^{+\infty} \frac{\chi_{BA}(\omega')}{\omega' - \omega - i0}\, d\omega' \tag{1.43}$$

Making use of the relation

$$\frac{1}{\omega' - \omega - i0} = \frac{\mathscr{P}}{\omega' - \omega} + i\pi\delta(\omega' - \omega) \tag{1.44}$$

Eq. (1.43) can be written in the simple form:

$$\chi_{BA}(\omega) = \frac{1}{i\pi} \mathscr{P}\!\!\int_{-\infty}^{+\infty} \frac{\chi_{BA}(\omega')}{\omega' - \omega}\, d\omega' \tag{1.45}$$

This equation can be written more explicitly by separating the Hermitian and the anti-Hermitian part of $\chi_{BA}(\omega)$ (see Eqs (1.28) and (1.29)).

This operation leads to the Kramers-Kronig relations:

$$\chi'_{BA}(\omega) = \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{\chi''_{BA}(\omega')}{\omega' - \omega} \, d\omega' \tag{1.46}$$

$$\chi''_{BA}(\omega) = -\frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{\chi'_{BA}(\omega')}{\omega' - \omega} \, d\omega' \tag{1.47}$$

which are very useful for the interpretation of many experiments.

Thus, we see that the total response $\chi_{BA}(\omega)$ can be expressed in terms of $\chi'_{BA}(\omega)$ or $\chi''_{BA}(\omega)$, only. However, in general, the function $\chi''_{BA}(\omega)$ (which corresponds to the absorptive part of the response function) is more localized than $\chi'_{BA}(\omega)$. For this reason, it is interesting to express $\chi_{BA}(\omega)$ in terms of $\chi''_{BA}(\omega)$ only. Actually, from the Kramers-Kronig relations, we deduce:

$$\chi_{BA}(\omega) = \frac{1}{i\pi} \int_{-\infty}^{+\infty} \frac{\chi''_{BA}(\omega')}{\omega' - \omega - i0} \, d\omega' \tag{1.48}$$

We may remark also that (for $z'' > 0$):

$$\chi_{BA}(z) = \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{\chi''_{BA}(\omega)}{\omega - z} \, d\omega \tag{1.49}$$

a relation which can be derived easily from Eq. (1.42) by using relation (1.46).

1.4. Formal expression of the response function. Kubo formula

For a quantum system in a state of equilibrium the density operator D is a constant in any representation (Heisenberg, Schrödinger, or interaction representation). On the other hand, the formal expressions giving the response function $\chi_{BA}(\omega)$ in terms of H, A and B may be derived by using any representation. However, it may be simpler to use the interaction representation as will be done here.

First, we shall define the time-dependent operators A(t) and B(t) by putting:

$$A(t) = e^{i\frac{Ht}{\hbar}} \, A \, e^{-i\frac{Ht}{\hbar}} \tag{1.50}$$

$$B(t) = e^{i\frac{Ht}{\hbar}} \, B \, e^{-i\frac{Ht}{\hbar}} \tag{1.51}$$

In the interaction representation, the density operator D(t) is given by its initial value and the equation:

$$i\hbar \dot{D}(t) = \left[e^{i\frac{Ht}{\hbar}} v(t) \, e^{-i\frac{Ht}{\hbar}}, D(t) \right] = -[A(t), D(t)] \, a(t) \tag{1.52}$$

On the other hand, we have:

$$\langle B(t)\rangle_v - \langle B\rangle = \mathrm{Tr}\left([D(t) - D] \, B(t) \right) \tag{1.53}$$

Now, we may put:

$$D(t) = D + \delta D(t) \tag{1.54}$$

where $\delta D(t)$ is given in the linear approximation by:

$$\delta D(t) = i\hbar^{-1} \int_{-\infty}^{t} [A(t'), D] \, a(t') \, dt' \tag{1.55}$$

This expression is rather formal, but a really meaningful result is obtained by using this expression in Eq. (1.53):

$$\langle B(t)\rangle_v - \langle B\rangle = i\hbar^{-1} \int_{-\infty}^{t} dt' \, a(t') \, \mathrm{Tr}\left([A(t'), D] \, B(t) \right) \tag{1.56}$$

ADB − DAB = DBA − DAB

By using the cyclic invariance of the trace, we obtain:

$$\langle B(t)\rangle_v - \langle B\rangle = i\hbar^{-1} \int_{-\infty}^{t} dt' \, a(t') \, \mathrm{Tr}\left(D \, [B(t), A(t')] \right) \tag{1.57}$$

$$= i\hbar^{-1} \int_{-\infty}^{t} \langle [B(t), A(t')] \rangle \, a(t') \, dt' \tag{1.58}$$

By comparing this expression with the definition (1.11), we get finally:

$$\chi_{BA}(t-t') = i\hbar^{-1} \langle [B(t), A(t')] \rangle \, \Theta(t-t') \tag{1.59}$$

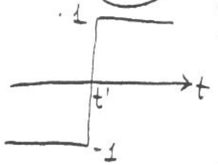
where $\Theta(t)$ is the step function ($\Theta(0) = \frac{1}{2}$, $\Theta(x) = +1$ for $x > 0$, $\Theta(x) = 0$ for $x < 0$). Incidentally, we see immediately that this function satisfies the causality requirements (Eq. (1.10)). On the other hand, the response function must be real since B(t) and A(t') are Hermitian, which implies that the mean value of their commutator is purely imaginary. Finally, the fact that the response function depends only on the difference (t−t') is immediately evident since we have:

$$\chi_{BA}(t-t') = -\frac{\Theta(t-t')}{i\hbar} \langle [B(t), A(t')] \rangle = -\frac{\Theta(t-t')}{i\hbar Z} \mathrm{Tr}\left(e^{-\beta H} [B(t), A(t')] \right) \tag{1.60}$$

$$= -\frac{\Theta(t-t')}{i\hbar Z} \mathrm{Tr} \, e^{-\beta H} [e^{iH(t-t')} B e^{-iH(t-t')}, A] = -\frac{\Theta(t-t')}{i\hbar} \langle [B(t-t'), A] \rangle$$

$\epsilon(t) = \theta(t-t') - \theta(t'-t)$

$\epsilon(t)$



The Hermitian and anti-Hermitian parts of the response function have very simple expressions; according to Eq.(1.59) and the definitions (1.24) and (1.25) we have ($\epsilon(t) = -1 + 2\Theta(t)$):

$$\chi'_{BA}(t-t') = i(2\hbar)^{-1}\,\epsilon(t-t')\,\langle[B(t),\ A(t')]\rangle \qquad (1.61)$$

$$\chi''_{BA}(t-t') = (2\hbar)^{-1}\langle[B(t),\ A(t')]\rangle \qquad (1.62)$$

A PROPOSITO

Incidentally, we remark that $\chi_{BA}(t-t')$ can be written in terms of $\chi''_{BA}(t-t')$ or $\chi'_{BA}(t-t')$ alone:

$$\boxed{\chi_{BA}(t-t') = 2i\Theta(t-t')\,\chi''_{BA}(t-t') = 2\Theta(t-t')\,\chi'_{BA}(t-t')} \qquad (1.63)$$

Actually, the first relation coincides with the dispersion relation (1.48).

1.5. Symmetries

The reality of the response function implies relations between the real and the imaginary parts of $\chi_{BA}(\omega)$:

$$\mathrm{Re}\,\chi_{BA}(\omega) = \mathrm{Re}\,\chi_{BA}(-\omega) \qquad (1.64)$$

$$\mathrm{Im}\,\chi_{BA}(\omega) = -\,\mathrm{Im}\,\chi_{BA}(-\omega) \qquad (1.65)$$

and also, as we have seen, between the Hermitian and the anti-Hermitian parts of $\chi_{BA}(\omega)$ (Eqs(1.30) and (1.31)):

$$\chi'_{BA}(\omega) = \chi'_{AB}(-\omega) = [\chi'_{AB}(\omega)]^* \qquad (1.66)$$

$$\chi''_{BA}(\omega) = -\,\chi''_{AB}(-\omega) = [\chi''_{AB}(\omega)]^* \qquad (1.67)$$

Additional symmetry properties can be found by time reversal. Let θ be this transformation. The Hamiltonian H is, in general, invariant wit[h] respect to time reversal; however, if a magnetic field $\vec{b}$ is applied to the system, then, the time reversal operation changes $\vec{b}$ into $-\vec{b}$. On the othe[r] hand, the operators A and B have often simple symmetry properties under time reversal such as:

$$\theta A\theta^{-1} = \epsilon_A A \qquad \theta B\theta^{-1} = \epsilon_B B. \qquad (1.68)$$

and for any operator $\mathcal{O}$, the time reversal operator θ satisfies also the relation

$$\mathrm{Tr}\,\theta\mathcal{O}\theta^{-1} = \mathrm{Tr}\,\mathcal{O}^{\dagger} \qquad (1.69)$$

$i\hbar \frac{d}{dt}\langle \rho g H\rangle = $ [illegible] $(t), \rho g]$ A

Consequently, the time reversal invariance of the system can be expressed as follows:

$$\chi_{BA}(t-t',\vec{b}) \equiv -\,\Theta(t-t')Z^{-1}\,\mathrm{Tr}\left(\frac{e^{-\beta H(\vec{b})}}{i\hbar}\,[B(t),\ A(t')]\right) \qquad (1.70)$$

$$= -\,\Theta(t-t')Z^{-1}\,\mathrm{Tr}\left(\theta[A(-t'),\ B(-t)]\,\frac{e^{-\beta H(\vec{b})}}{-i\hbar}\,\theta^{-1}\right) \qquad (1.71)$$

where $H(\vec{b})$ is the Hamiltonian considered to be a function of $\vec{b}$ (Z is invariant under time reversal). The last equation can be written ($\theta i\theta^{-1} = -i$)

$$\chi_{BA}(t-t',\ \vec{b}) = -\,\epsilon_A\epsilon_B\,\Theta(t-t')\,Z^{-1}\,\mathrm{Tr}\left([A(t'),\ B(t)]\,\frac{e^{-\beta H(-\vec{b})}}{i\hbar}\right) \qquad (1.72)$$

$$= -\,\epsilon_A\epsilon_B\,\Theta(t-t')\,Z^{-1}\,\mathrm{Tr}\left(\frac{e^{-\beta H(-\vec{b})}}{-i\hbar}\,[B(t),\ A(t')]\right) \qquad (1.73)$$

Finally, we get:

$$\chi_{BA}(t-t',\ \vec{b}) = \epsilon_A\epsilon_B\,\chi_{AB}(t-t',\ -\vec{b}) \qquad (1.74)$$

or, by using Fourier transforms:

$$\chi_{BA}(\omega,\vec{b}) = \epsilon_A\epsilon_B\,\chi_{AB}(\omega,\ -\vec{b}) \qquad (1.75)$$

1.6. Absorption and interpretation of $\chi''(\omega)$

Our aim is to calculate the energy produced in a system by the external perturbation $v(t)$. This energy is of course always positive and due to absorption in the system. But dissipative effects are non-linear. For this reason and in order to preserve interference terms, it will be assumed in this section that the perturbing potential is a sum of terms:

$$v(t) = -\sum_j a_j(t)\,A_j \qquad (1.76)$$

For the sake of simplicity the response function corresponding to A_i and A_j will be simply written $\chi_{ji}(t)$. In Heisenberg representation, we have:

$$i\hbar\frac{dH}{dt} = [H,\ H(t)] = [H,\ v(t)] = -\sum_j a_j(t)\,[H,\ A_j] \qquad (1.77)$$

All the operators in this equation are assumed to be time-dependent. This means that we have:

$$i\hbar\frac{d}{dt}\langle H\rangle_v = -\sum_j a_j(t)\,\langle[H,\ A_j]\rangle_v \qquad (1.78)$$

The static part $\langle [H, A_j] \rangle$ vanishes for a system in thermal equilibrium.
Therefore from (1.57), we get:

$$i\hbar \frac{d}{dt}\langle H \rangle_v = -i\hbar^{-1} \sum_{j\ell} a_j(t) \int_{-\infty}^{t} \left\langle \left[[H, A_j(t)],\ A_\ell(t') \right] \right\rangle a_\ell(t')\, dt'$$

$$= -\sum_{j\ell} a_j(t) \int_{-\infty}^{t} \frac{\partial}{\partial t} \langle [A_j(t), A_\ell(t')] \rangle a_\ell(t')\, dt' \qquad (1.79)$$

Therefore by using definition (1.59), we obtain:

le somme, di commut...

$$\frac{d}{dt}\langle H \rangle_v = \sum_{j\ell} \int_{-\infty}^{+\infty} a_j(t) \frac{\partial}{\partial t} X_{j\ell}(t-t')\, a_\ell(t')\, dt' \qquad (1.80)$$

$(X_{ij} + X_{ji})\, a_i a_j$

For reasons of simplicity, we assume that the perturbation is monochromat (but real); therefore we put:

$$a_j(t) = \frac{1}{2}\left[a_j e^{-i\omega t} + a_j^* e^{i\omega t} \right] \qquad (1.81)$$

Then, the preceding expression becomes:

$$\frac{d}{dt}\langle H \rangle_v = \frac{-i\omega}{4} \sum_{j\ell} \left(a_j e^{-i\omega t} + a_j^* e^{i\omega t} \right) \left[a_\ell e^{-i\omega t} X_{j\ell}(\omega) - a_\ell^* e^{i\omega t} X_{j\ell}(-\omega) \right] \qquad (1.82)$$

Now, we can drop the periodic terms which are irrelevant here and calculat the average flow of energy:

$$\overline{\frac{d}{dt}\langle H \rangle_v} = -\frac{i\omega}{4} \sum_{j\ell} \left[a_j^* a_\ell X_{j\ell}(\omega) - a_\ell^* a_j X_{j\ell}(-\omega) \right] \qquad (1.83)$$

Then, by using definition (1.29), we obtain finally:

$$\frac{d}{dt}\langle H \rangle_v = \frac{1}{2} \sum_{j\ell} a_j^* a_\ell\, \omega X''_{j\ell}(\omega) \qquad (1.84)$$

This result shows that $X''_{j\ell}(\omega)$ can really be identified with the dissipative part of $X_{j\ell}(\omega)$.

1.7. A Kubo formula

Another formal expression of the response function has been given by Kubo [4]. In order to derive it, we write $X_{BA}(t)$ in the following form which

results from the cyclical invariance of the trace of a product of operators; in doing so we start from Eq. (1.60):

$$X_{BA}(t) = \frac{\Theta(t)}{i\hbar Z} \operatorname{Tr}\left([e^{-\beta H}, A]\, B(t) \right) \qquad (1.85)$$

Now, we use the following identity:

$$[e^{-\beta H}, A] = \int_0^\beta e^{-\beta H} e^{\lambda H} [A, H]\, e^{-\lambda H}\, d\lambda \qquad (1.86)$$

which can be easily proved by multiplying both sides by $\exp(\beta H)$ and differentiating. Thus,

$$[e^{-\beta H}, A] = i\hbar \int_0^\beta e^{-\beta H} \dot{A}(-i\hbar\lambda)\, d\lambda \qquad (1.87)$$

Finally, by introducing this expression in Eq. (1.85) we obtain Kubo's formula:

$$X_{BA}(t) = \Theta(t) \int_0^\beta \langle \dot{A}(-i\hbar\lambda)\, B(t) \rangle\, d\lambda$$

$$= -\Theta(t) \int_0^\beta \langle A(-i\hbar\lambda)\, \dot{B}(t) \rangle\, d\lambda \qquad (1.88)$$

1.8. Fluctuation dissipation theorems

The natural fluctuations occurring in a system at equilibrium are related to the dissipation effects resulting from an interaction of the system with external forces. Of course, this connection proves to be very important and, though it has been clarified only rather recently [5], it has been recognized and used a long time ago (Einstein relation, Nyquist theorem [6]) in special cases.

The time-dependent fluctuation function $F_{BA}(t)$ is defined by the anti-commutator:

$$F_{BA}(t-t') = \frac{1}{2} \langle \{ (B(t) - \langle B \rangle),\ (A(t') - \langle A \rangle) \} \rangle \qquad (1.89)$$

Our aim is to establish a relation between the Fourier transform $\Phi_{BA}(\omega)$ of $F_{BA}(t)$ and the dissipative part of the susceptibility $X''_{BA}(\omega)$.

$$\Phi_{BA}(\omega) = \int_{-\infty}^{+\infty} F_{BA}(t)\, e^{i\omega t - 0|t|}\, dt \qquad (1.90)$$

For this purpose, we introduce the function $S_{BA}(t)$ and its Fourier transform $\Sigma_{BA}(\omega)$:

$$S_{BA}(t) = \langle (B(t) - \langle B \rangle)(A - \langle A \rangle) \rangle = \langle (B - \langle B \rangle)(A(-t) - \langle A \rangle) \rangle \tag{1.91}$$

$$\Sigma_{BA}(\omega) = \int_{-\infty}^{+\infty} S_{BA}(t)\, e^{i\omega t - 0|t|}\, dt \tag{1.92}$$

By using these definitions, we see immediately that:

$$\chi''_{BA}(t) = (2\hbar)^{-1}\,[S_{BA}(t) - S_{AB}(-t)] \tag{1.93}$$

$$F_{BA}(t) = \frac{1}{2}\,[S_{BA}(t) + S_{AB}(-t)] \tag{1.94}$$

But on the other hand:

$$\begin{aligned} \langle A(-t)B \rangle &= Z^{-1}\,\mathrm{Tr}\left(e^{-\beta H} e^{-i\frac{Ht}{\hbar}} A\, e^{i\frac{Ht}{\hbar}} B\right) \\ &= Z^{-1}\,\mathrm{Tr}\left(e^{-\beta H} e^{\frac{i}{\hbar}H(t - i\hbar\beta)} B\, e^{-\frac{i}{\hbar}H(t - i\hbar\beta)} A\right) \\ &= \langle B(t - i\hbar\beta) A \rangle \end{aligned} \tag{1.95}$$

Consequently we obtain

$$S_{AB}(-t) = S_{BA}(t - i\hbar\beta) \tag{1.96}$$

and, therefore, we have:

$$\chi''_{BA}(t) = (2\hbar)^{-1}\,[S_{BA}(t) - S_{BA}(t - i\hbar\beta)] \tag{1.97}$$

$$F_{BA}(t) = \frac{1}{2}\,[S_{BA}(t) + S_{BA}(t - i\hbar\beta)] \tag{1.98}$$

Let us now take the Fourier transforms of these expressions. If we assume that

1) $\left\langle \left(A(t) - \langle A \rangle\right)\left(B - \langle B \rangle\right) \right\rangle \to 0$ for $t \to \infty$

2) $\left\langle \left(A(t) - \langle A \rangle\right)\left(B - \langle B \rangle\right) \right\rangle$ is analytic in the domain $0 < \mathrm{Im}\, t < \beta h$

we can write:

$$\begin{aligned} \int_{-\infty}^{+\infty} e^{i\omega t} S_{AB}(-t)\, dt &\equiv \int_{-\infty}^{+\infty} e^{i\omega t} S_{BA}(t - i\hbar\beta)\, dt \\ &= e^{-\beta\hbar\omega} \int_{-\infty}^{+\infty} e^{i\omega t} S_{BA}(t)\, dt = e^{-\beta\hbar\omega}\, \Sigma_{BA}(\omega) \end{aligned} \tag{1.99}$$

Therefore, we obtain in this case:

$$\chi''_{BA}(\omega) = (2\hbar)^{-1}\,(1 - e^{-\beta\hbar\omega})\, \Sigma_{BA}(\omega) \tag{1.100}$$

$$\Phi_{BA}(\omega) = \frac{1}{2}\,(1 + e^{-\beta\hbar\omega})\, \Sigma_{BA}(\omega) \tag{1.101}$$

Thus, we are led to the fluctuation dissipation theorem:

$$\chi''_{BA}(\omega) = \hbar^{-1}\, \mathrm{th}\,(\beta\hbar\omega/2)\, \Phi_{BA}(\omega) \tag{1.102}$$

1.9. Moments and sum rules

The corresponding classical formula is, of course, obtained by passing to the limit $\hbar \to 0$; in this way we obtain:

$$\chi''_{BA}(\omega) = \frac{1}{2}\,\beta\omega\, \Phi_{BA}(\omega) \tag{1.103}$$

Note that, if $A = B$, $\chi''_{BA}(\omega)$ and $\Phi_{BA}(\omega)$ are real.

As an interesting application of the preceding theorem, we have also:

$$\hbar \int_{-\infty}^{+\infty} \coth\left(\frac{\beta\hbar\omega}{2}\right) e^{-i\omega(t-t')}\, \chi''_{AA}(\omega)\, d\omega = \int_{-\infty}^{+\infty} e^{-i\omega(t-t')}\, \Phi_{AA}(\omega)\, d\omega = 2\pi F_{AA}(t-t') \tag{1.104}$$

which leads to the sum rule (for $t = t'$):

$$\int_{-\infty}^{+\infty} \coth\left(\frac{\beta\hbar\omega}{2}\right) \chi''_{AA}(\omega)\, d\omega = 2\pi\hbar^{-1} \left\langle \left(A - \langle A \rangle\right)^2 \right\rangle \tag{1.105}$$

or for a classical system

$$\int_{-\infty}^{+\infty} \chi''_{AA}(\omega)\, \frac{d\omega}{\omega} = \pi\beta \left\langle \left(A - \langle A \rangle\right)^2 \right\rangle \tag{1.106}$$

This equation can be generalized by taking derivatives of Eq. (1.104) with respect to t and t'. Thus, we get (when the equation has a meaning):

$$\int \omega^{2n} \coth\left(\frac{\beta\hbar\omega}{2}\right)\chi''_{AA}(\omega)\, d\omega = 2\pi\hbar^{-1}\left\langle\left(A^{(n)}(t)\right)^2\right\rangle \tag{1.107}$$

where $A^{(n)}(t)$ is the n^{th} order time derivative of $A(t)$ (t is arbitrary). In the classical limit, we have also:

$$\int_{-\infty}^{+\infty} \omega^{2n-1}\chi''_{AA}(\omega)\, d\omega = \pi\beta\left\langle\left(A^{(n)}(t)\right)^2\right\rangle \tag{1.108}$$

We note that if the potentials acting on the particles of the system are regular, all the moments of $\chi''_{AA}(\omega)$ exist since the mean values appearing on the right-hand side of (1.107) are finite. But, of course, this is not true in the case of Coulomb interactions.

1.10. Classical case. Additional sum rules

In the classical case, useful relations are obtained also between the reactive part of the susceptibility and the fluctuations by using the Kramers-Kronig relation (1.46):

$$\chi'_{AA}(\omega) = \frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{\chi''_{AA}(\omega')}{\omega'-\omega}\, d\omega' \tag{1.109}$$

Thus, by putting $\omega = 0$ in this equation and by taking into account the symmetry properties of $\chi'_{AA}(\omega)$ and $\chi''_{AA}(-\omega)$ we deduce from Eq. (1.106):

$$\chi_{AA}(0) = \chi'_{AA}(0) = \frac{1}{\pi}\int_{-\infty}^{+\infty}\omega^{-1}\chi''_{AA}(\omega)\, d\omega = \beta\langle(A(t) - \langle A\rangle)^2\rangle \tag{1.110}$$

On the other hand, by examining the behaviour of $\chi'_{AA}(\omega)$ for large values of ω, we obtain also an interesting relation (see Eq. (1.108) for $n = 1$):

guardando 1.109
l'integr. è grosso solo se integrale a ω anche ω'→∞
⇒ $\int \omega\, \chi''_{AA}(\omega')\,d\omega' \simeq \int \omega'\, \chi''_{AA}(\omega')\,d\omega'$

$$\lim_{\omega\to\infty}\omega^2\chi'_{AA}(\omega) = -\frac{1}{\pi}\int_{-\infty}^{+\infty}\omega\chi''_{AA}(\omega)\, d\omega = -\beta\langle(\dot A(t))^2\rangle \tag{1.111}$$

If this equation has a meaning, we can infer from the convergence of the integral that $\lim_{\omega\to\infty} \omega^2\chi''_{AA}(\omega) = 0$. Thus, we get finally:

$$\lim_{\omega\to\infty}\omega^2\chi_{AA}(\omega) = \lim_{\omega\to\infty}\omega^2\chi'_{AA}(\omega) = -\beta\langle(\dot A(t))^2\rangle \tag{1.112}$$

1.11. Spectral representations and energy levels of the unperturbed system

The meaning of all the preceding relations may become more evident by using explicit representations in terms of the eigenstates and eigenenergies of the system. Thus we have immediately (see Eq. (1.91)):

$$S_{BA}(t) = Z^{-1}\Big(\sum_{mn} e^{-\beta E_n} e^{i(E_n - E_m)t/\hbar}\langle n|A|m\rangle\langle m|B|n\rangle - \sum_n e^{-\beta E_n}\langle n|A|n\rangle\sum_m e^{-\beta E_m}\langle m|B|m\rangle\Big) \tag{1.113}$$

Hence, we get the Fourier transform:

$$\Sigma_{BA}(\omega) = \hbar Z^{-1}\sum_{mn} e^{-\beta E_n}\delta(\hbar\omega - (E_m - E_n))\langle n|A|m\rangle\langle m|B|n\rangle \tag{1.114}$$

× 2π

and by using Eqs (1.100) and (1.101) we obtain:

$$\chi''_{BA}(\omega) = \frac{1}{2}Z^{-1}\sum_{mn}(e^{-\beta E_n} - e^{-\beta E_m})\,\delta(\hbar\omega - (E_m - E_n))\langle n|A|m\rangle\langle m|B|n\rangle \tag{1.115}$$

× 2π

$$\Phi_{BA}(\omega) = \frac{\hbar}{2}Z^{-1}\sum_{mn}(e^{-\beta E_n} + e^{-\beta E_m})\,\delta(\hbar\omega - (E_m - E_n))\langle n|A|m\rangle\langle m|B|n\rangle \tag{1.116}$$

In the same way, it is easy to find an explicit representation for $\chi_{BA}(\omega)$, since we have according to Eqs (1.63) and (1.97):

$$\chi_{BA}(t) = 2i\Theta(t)\chi''_{BA}(t)$$

$$= i\hbar^{-1}\Theta(t)\sum_{mn}(e^{-\beta E_n} - e^{-\beta E_m})e^{i(E_n - E_m)t/\hbar}\langle n|A|m\rangle\langle m|B|n\rangle \tag{1.117}$$

Therefore we obtain the following expression:

$$\chi_{BA}(\omega) = -Z^{-1}\sum_{mn}(e^{-\beta E_n} - e^{-\beta E_m})\frac{\langle n|A|m\rangle\langle m|B|n\rangle}{\hbar\omega - (E_m - E_n) + i0} \tag{1.118}$$

check sign $E_n - E_m$

which shows the relationship between the excitations of a system and the response function.

1.12. Density fluctuations, f sum rule and longitudinal sum rule

The general theory is frequently applied to the study of large systems consisting of a set of identical particles of mass m interacting between each other and with the medium in which they move. In this case, the Hamiltonian can be written in the form:

$$H = \frac{1}{2m}\sum_{i=1}^{N}\vec P_i^2 + U(r_1 \ldots r_N) \tag{1.119}$$

where $U(r_1 \ldots r_N)$ is an operator which depends on the medium and on the positions $r_1 \ldots r_N$ of the particles under consideration; on the other hand, $\vec{P}_s$ is the momentum of the s^{th} particle. The form of this Hamiltonian implies an interesting relation, the f sum-rule, which plays an important role in the theory of conductivity and will be derived now.

First, we introduce the density operator $n(\vec{r})$ and the current operator $\vec{j}(\vec{r})$:

$$n(\vec{r}) = \sum_s \delta(\vec{r} - \vec{r}_s) \tag{1.120}$$

$$\vec{j}(\vec{r}) = \frac{1}{2m} \sum_s (\vec{P}_s\, \delta(\vec{r} - \vec{r}_s) + \delta(\vec{r} - \vec{r}_s)\, \vec{P}_s) \tag{1.121}$$

and their Fourier transforms:

$$n(\vec{k}) = \sum_s e^{i\vec{k}\cdot\vec{r}_s} \tag{1.122}$$

$$\vec{j}(\vec{k}) = \frac{1}{2m} \sum_s (\vec{P}_s\, e^{i\vec{k}\cdot\vec{r}_s} + e^{i\vec{k}\cdot\vec{r}_s}\, \vec{P}_s) \tag{1.123}$$

which are related by:

$$[H, n(\vec{k})] = \hbar\, \vec{k}\cdot\vec{j}(\vec{k}) \tag{1.124}$$

a relation which implies the conservation of particles; in fact, if we define:

$$n(\vec{k}, t) = e^{i\frac{Ht}{\hbar}}\, n(\vec{k})\, e^{-i\frac{Ht}{\hbar}} \tag{1.125}$$

$$\vec{j}(\vec{k}, t) = e^{i\frac{Ht}{\hbar}}\, \vec{j}(\vec{k})\, e^{-i\frac{Ht}{\hbar}} \tag{1.126}$$

these time-dependent operators satisfy the conservation equation:

$$\frac{d}{dt} n(\vec{k}, t) = \frac{i}{\hbar} [H, n(\vec{k}, t)] = i\,\vec{k}\cdot\vec{j}(\vec{k}, t) \tag{1.127}$$

Now, we note that:

$$[\vec{j}(\vec{k}),\, n(-\vec{k})] = -\frac{N}{m}\hbar\vec{k} \tag{1.128}$$

With the help of Eq. (1.124), we deduce:

$$[[H, n(\vec{k})],\, n(-\vec{k})] = -\frac{N}{m}\hbar^2 k^2 \tag{1.129}$$

For a system at temperature T the mean value of the left-hand side can be expressed in terms of the eigenstates $|n\rangle$ of H:

$$\langle [[H, n(\vec{k})],\, n(-\vec{k})] \rangle = Z^{-1} \sum_{\ell m} e^{-\beta E_\ell} (E_\ell - E_m)\, (\langle \ell | n(\vec{k}) | m \rangle \langle m | n(-\vec{k}) | \ell \rangle + \langle \ell | n(-\vec{k}) | m \rangle \langle m | n(\vec{k}) | \ell \rangle) \tag{1.130}$$

In the right-hand side of this expression, both terms are even in $\vec{k}$; therefore, by comparing this expression with Eq. (1.129), we obtain:

$$2 Z^{-1} \sum_{\ell m} e^{-\beta E_\ell} (E_m - E_\ell) |\langle \ell | n(k) | m \rangle|^2 = \frac{N}{m}\hbar^2 k^2 \tag{1.131}$$

It is customary to define the oscillator strength of the level $|n\rangle$ by:

$$f_\ell(\vec{k}, T) = \frac{2m}{N Z \hbar^2 k^2} \sum_m e^{-\beta E_\ell} (E_m - E_\ell) |\langle \ell | n(k) | m \rangle|^2 \tag{1.132}$$

and thus, we get the f sum rule:

$$\sum_\ell f_\ell(\vec{k}, T) = 1 \tag{1.133}$$

This relation can be expressed in a slightly different form which is also very useful. In agreement with definition (1.62), we put:

$$\chi''_{j_\alpha j_\beta}(\vec{r} - \vec{r}', t - t') = (2\hbar)^{-1} \langle [j_\alpha(\vec{r}, t),\, j_\beta(\vec{r}', t')] \rangle \tag{1.134}$$

where

$$\vec{j}(\vec{r}, t) = e^{i\frac{Ht}{\hbar}}\, \vec{j}(\vec{r})\, e^{-i\frac{Ht}{\hbar}} \tag{1.135}$$

and its Fourier transform:

$$\chi''_{j_\alpha j_\beta}(\vec{k}, \omega) = \int d^3r\, e^{i\vec{k}\cdot(\vec{r}-\vec{r}')} \int dt\, e^{i\omega(t-t')}\, \chi''_{j_\alpha j_\beta}(\vec{r} - \vec{r}', t - t')$$

$$= (2\hbar\Omega)^{-1} \int dt\, e^{i\omega(t-t')} \langle [j_\alpha(\vec{k}, t),\, j_\beta(-\vec{k}, t')] \rangle \tag{1.136}$$

(Ω = volume of the sample).

Our aim is to find a sum rule which is just another version of the f sum rule for the susceptibility $\chi''_{j_\alpha j_\beta}(\vec{k}, \omega)$.

In a homogeneous medium, we can separate $\chi^{"}_{j_\alpha j_\beta}(\vec{k},\omega)$ into a longitudinal and a transversal part; by definition we have

$$\chi^{"}_{j_\alpha j_\beta}(\vec{k},\omega) = \frac{k_\alpha k_\beta}{k^2}\chi^{"L}_{jj} + \left(\delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{k^2}\right)\chi^{"T}_{jj} \qquad (1.137)$$

Therefore:

$$\chi^{"L}_{jj}(\vec{k},\omega) = (2\hbar k^2 \Omega)^{-1}\int dt\, e^{i\omega(t-t')}\langle[\vec{k}\cdot\vec{j}(\vec{k},t),\ \vec{k}\cdot\vec{j}(-\vec{k},t')]\rangle \qquad (1.138)$$

This expression can be transformed by using the continuity equation (1.127):

$$\chi^{"L}_{jj}(\vec{k},\omega) = i(2\hbar k^2\Omega)^{-1}\int dt\, e^{i\omega(t-t')}\langle[\vec{k}\cdot\vec{j}(\vec{k},t),\ \dot{n}(-\vec{k},t')]\rangle \qquad (1.139)$$

$$= -\omega(2\hbar k^2\Omega)^{-1}\int dt\,\langle[\vec{k}\cdot\vec{j}(\vec{k},t),\ n(-\vec{k},t')]\rangle \qquad (1.140)$$

Thus, we derive the longitudinal sum rule:

$$\frac{1}{2\pi}\int_{-\infty}^{+\infty} d\omega\, \omega^{-1}\chi^{"L}_{jj}(\vec{k},\omega) = -(2\hbar k^2\Omega)^{-1}\langle[\vec{k}\cdot\vec{j}(\vec{k}),\ n(-\vec{k})]\rangle \qquad (1.141)$$

which by using Eq.(1.128) becomes:

$$\frac{1}{2\pi}\int_{-\infty}^{+\infty} d\omega\,\omega^{-1}\chi^{"L}_{jj}(\vec{k},\omega) = \frac{n}{2m} \qquad (1.142)$$

(where $n = N/\Omega$ is the density of particles).

Or by application of the Kramers-Kronig relation (1.46) and of the symmetry conditions (1.66) and (1.67):

$$\chi^{L}_{jj}(\vec{k},0) = \chi'^{L}_{jj}(\vec{k},0) = \frac{n}{m} \qquad (1.143)$$

2. LINEAR RESPONSE TO A DYNAMICAL DISTURBANCE. EXAMPLES AND APPLICATIONS

2.1. Classical oscillator

As a simple illustration of the theory, we consider now the case of a simple classical oscillator driven by an external force. The coordinate $x(t)$ of the oscillating mass m satisfies the equation:

$$m\ddot{x}(t) + \gamma\dot{x}(t) + m\omega_0^2 x(t) = f(t) \qquad (2.1)$$

where m is the mass, $m\omega_0^2$ the spring constant, γ a friction coefficient and $f(t)$ an external force which can be regarded as resulting from a perturbing potential:

$$v(t) = -x f(t) \qquad (2.2)$$

The preceding equation and the usual assumption $x(-\infty) = 0$ determines the variations of $x(t)$ completely:

$$x(t) = \int_{-\infty}^{t} \chi_{xx}(t-t')\, f(t')\, dt' \qquad (2.3)$$

Now, we get:

$$f(t) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} e^{-i\omega t}\varphi(\omega)\, d\omega \qquad (2.4)$$

$$x(t) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} e^{-i\omega t}\xi(\omega)\, d\omega \qquad (2.5)$$

and thus the preceding equation can be transformed into:

$$\xi(\omega) = \chi_{xx}(\omega)\varphi(\omega) \qquad (2.6)$$

But Eq.(2.1) gives:

$$[m(\omega_0^2 - \omega^2) - i\gamma\omega]\,\xi(\omega) = \varphi(\omega) \qquad (2.7)$$

and therefore, we have:

$$\chi_{xx}(\omega) = \frac{1}{m(\omega_0^2-\omega^2) - i\gamma\omega} \qquad (2.8)$$

This function has two poles; two regimes are possible:

a) $\gamma < 2m\,\omega_0$ (weakly damped oscillator).

then the roots are given by:

$$\omega = \pm\omega_1 - i\omega_2 \qquad \omega_1 > 0 \qquad \omega_2 > 0 \qquad (2.9)$$

b) $\gamma > 2m\,\omega_0$ (strongly damped oscillator);

Si $\gamma < 0$
$|x(t)|$ [illegible]

then

$$\omega = -i(\omega_1 \pm \omega_2) \qquad \omega_1 > \omega_2 > 0 \qquad (2.10)$$

In any case, the roots have negative imaginary parts and therefore, in agreement with the causality requirements $\chi_{xx}(\omega)$ is analytic in the half plane $\mathrm{Im}\,\omega > 0$.

The Krönig-Kramers relation (see Eq. (1.45)):

$$\chi_{xx}(\omega) = \frac{1}{i\pi} \mathcal{P}\!\!\int_{-\infty}^{+\infty} \frac{\chi_{xx}(\omega')}{\omega' - \omega}\, d\omega' \tag{2.11}$$

turns out to be obvious because we can write:

$$\frac{\mathcal{P}}{\omega - \omega'} = \frac{1}{2}\left(\frac{1}{\omega' - \omega + i0} + \frac{1}{\omega' - \omega - i0}\right) \tag{2.12}$$

perché chiudo sopra? e poi che significa il fatto che $\chi(\omega)$ è il residuo?

why?

on the right-hand side of Eq. (2.11), close the contour upwards and calculate the residue at the pole $\omega' = \omega + i0$; this residue is of course $\chi_{xx}(\omega)$. embè?

The real and imaginary parts of $\chi_{xx}(\omega)$ are given by:

$$\chi'_{xx}(\omega) = \frac{m(\omega_0^2 - \omega^2)}{m^2(\omega_0^2 - \omega^2)^2 + \gamma^2\omega^2} \tag{2.13}$$

$$\chi''_{xx}(\omega) = \frac{\gamma\omega}{m^2(\omega_0^2 - \omega^2)^2 + \gamma^2\omega^2} \tag{2.14}$$

and we verify immediately the symmetry conditions of section 1.4. The shapes of the curves corresponding to these functions are very characteristic for small values of γ; they are represented in Fig. 2.

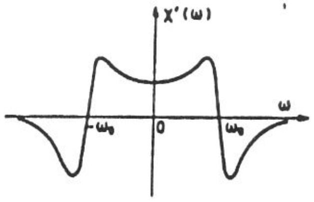


FIG. 2a. $\chi'(\omega)$ for small values of γ.

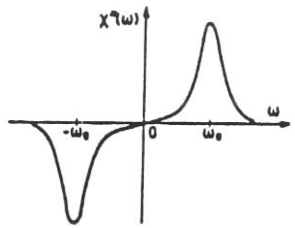


FIG. 2b. $\chi''(\omega)$ for small values of γ.

The work W(t) done by the external force f(t) is given by:

$$dW = \vec{f}\cdot d\vec{s}$$

$$P = \frac{dW}{dt} = \frac{\vec{f}\cdot d\vec{s}}{dt} = \vec{f}\cdot\frac{d\vec{s}}{dt} = \vec{f}\cdot\vec{v}(t)$$

$$\frac{dW(t)}{dt} = \dot{x}(t)\, f(t) = \int_{-\infty}^{\infty} f(t)\, \dot{\chi}_{xx}(t-t')\, f(t')\, dt'$$

$$(-i)$$

$$= \frac{1}{2\pi}\int_{-\infty}^{+\infty} e^{-i\omega' t} f(t)\, e^{i\omega' t'} f(t')\, \omega'\chi_{xx}(\omega')\, d\omega'\, dt' \tag{2.15}$$

Thus, if we put:

$$f(t) = \frac{1}{2}\left(f e^{-i\omega t} + f^* e^{i\omega t}\right) \tag{2.16}$$

we get:

$$\frac{d\overline{W}(t)}{dt} = \frac{\omega}{2}\chi''_{xx}(\omega)\, f f^* \tag{2.17}$$

in agreement with the general results of section 1.5: this result reminds us that $\chi''_{xx}(\omega)$ is the absorptive part and $\chi'_{xx}(\omega)$ the reactive part of the susceptibility.

Let us now examine how the fluctuation dissipation theorem applies to this case. Our system is classical but the friction γ is just a phenomenological coefficient and no real Hamiltonian corresponds to the equation of motion (2.1). The oscillating mass may however be regarded, for example, as a ball moving in a viscous medium. In this case, there exists a Hamiltonian for the whole system consisting of the ball and of the medium in which it oscillates, and γ describes the response of the medium. Then the fluctuation dissipation theorem should be valid for the whole system. Equations (1.110) and (1.112) are written in this case:

$$\chi_{xx}(0) = \beta\langle x^2(t)\rangle \qquad = \frac{1}{m\omega_0^2} \tag{2.18}$$

$$\lim_{\omega\to\infty} \omega^2\chi_{xx}(\omega) = -\beta\langle \dot{x}^2(t)\rangle \qquad = -\frac{1}{m} \tag{2.19}$$

But, on the other hand, from (2.8), we get:

$$\chi_{xx}(0) = \frac{1}{m\omega_0^2}\,; \quad \lim_{\omega\to\infty} \omega^2\chi_{xx}(\omega) = -\frac{1}{m} \tag{2.20}$$

From the comparison of these expressions, we get finally:

$$\frac{1}{2}\, m\,\omega_0^2\langle x^2(t)\rangle = \frac{1}{2}\, m\langle \dot{x}^2(t)\rangle = \frac{1}{2}KT \tag{2.21}$$

This is exactly Boltzmann's equipartition theorem, and this result appears rather remarkable if we consider the phenomenological nature of γ.

We note also that all these results would remain unchanged if the friction forces were frequency-dependent and represented by a coefficient $\gamma(\omega)$ provided that:

$$\gamma(0) = 0 \qquad \lim_{\omega\to\infty} \omega^{-1}\gamma(\omega) = 0 \tag{2.22}$$

2.2. Conductivity tensor

The behaviour of the system consisting of a sample of matter interacting with an electromagnetic field is determined by two kinds of equations, Firstly, the fields satisfy two groups of Maxwell equations which, in the reciprocal space and with proper units, can be written:

($\vec{h}(\vec{k},\omega)$ is the electric field, $\vec{b}(\vec{k},\omega)$ is the magnetic induction)

$$\text{I)}\quad \left[\begin{array}{ll} \vec{k}\times\vec{h}(\vec{k},\omega) - \omega\vec{b}(\vec{k},\omega) = 0 & (2.23) \\ \vec{k}\cdot\vec{b}(\vec{k},\omega) = 0 & (2.24) \end{array}\right.$$

$$\text{II)}\quad \left[\begin{array}{ll} \vec{k}\times\vec{b}(\vec{k},\omega) + \omega\vec{h}(\vec{k},\omega) = -i\vec{J}(\vec{k},\omega) & (2.25) \\ \vec{k}\cdot\vec{h}(\vec{k},\omega) = \rho(\vec{k},\omega) & (2.26) \end{array}\right.$$

where $\rho(\vec{k},\omega) = e\,n(\vec{k},\omega)$.

Secondly, there are the "material equations" which give the current and the density of charge appearing in the material as a result of its interaction with the electromagnetic field. Thus $\vec{J}(\vec{k},\omega)$ and $\rho(\vec{k},\omega)$ are functions of $\vec{h}(\vec{k},\omega)$ and $\vec{b}(\vec{k},\omega)$. However, $\rho(\vec{k},\omega)$ is not independent of $\vec{J}(\vec{k},\omega)$ since we have the continuity relation:

$$\omega\,\rho(\vec{k},\omega) - \vec{k}\cdot\vec{J}(\vec{k},\omega) = 0 \tag{2.27}$$

On the other hand, $\vec{b}(\vec{k},\omega)$ can be expressed in terms of $\vec{h}(\vec{k},\omega)$ by means of Eq. (2.23) which implies also Eq. (2.24). Thus, the response of the material to an electromagnetic stimulation is completely determined by the conductivity which relates $\vec{J}(\vec{k},\omega)$ to $\vec{h}(\vec{k},\omega)$. In an homogeneous medium (and a crystal, for example, can be considered homogeneous for wave lengths which are long compared with the interatomic distances), we can write:

$$J_\alpha(\vec{k},\omega) = \sum_\beta \sigma_{\alpha\beta}(\vec{k},\omega)\,h_\beta(\vec{k},\omega) \tag{2.28}$$

where $\sigma_{\alpha\beta}(\vec{k},\omega)$ is the conductivity tensor.

In order to emphasize this point, we can assume, for example, that the medium is isotropic and expand $\sigma_{\alpha\beta}(\vec{k},\omega)$; for small values of $\vec{k}$ and ω we find in this way:

$$\sigma_{\alpha\beta}(\vec{k},\omega) = \rho^{-1}\,\delta_{\alpha\beta} - i\omega\mathscr{A}\delta_{\alpha\beta} + i\mathscr{B}\omega^{-1}(k_\alpha k_\beta - k^2\delta_{\alpha\beta})$$

$$\mathscr{C}\omega\sum_\gamma \epsilon_{\alpha\beta\gamma}k_\gamma + i(\Lambda\omega)^{-1}\,\delta_{\alpha\beta} \tag{2.29}$$

where ρ is the resistivity, $\mathscr{A}$ the electric polarizability (with $\mathscr{A} = \epsilon - 1$ where ϵ is the dielectric constant), $\mathscr{B}$ a magnetic constant (with $\mathscr{B} = 1 - \mu^{-1}$ where μ is the magnetic permeability), $\mathscr{C}$ a constant of rotatory power (with $\epsilon_{\alpha\beta\gamma}$ completely antisymmetric with respect to the indices and $\epsilon_{xyz} = 1$), and Λ the London constant (if the system is a superconductor).

Thus, we see how all the simple electromagnetic properties of an isotropic medium are related to the form of $\sigma_{\alpha\beta}(\vec{k},\omega)$.

The conductivity tensor $\sigma_{\alpha\beta}(\vec{k},\omega)$ can be expressed in a formal way by means of a Kubo formula which often serves as a starting point for further investigations. The complete Hamiltonian can be written:

$$H(t) = \frac{1}{2m}\sum_{i=1}^{N}[\vec{P}_i - \frac{e}{c}\vec{A}(\vec{r}_i,t)]^2 + e\sum_{i=1}^{N}V(\vec{r}_i,t) + U(r_1\ldots r_N) \tag{2.30}$$

We note now that it is always possible to choose a gauge such that:

$$V(\vec{r},t) = 0 \tag{2.31}$$

On the other hand, since we are interested in the linear response only, we may write:

$$H(t) \simeq H + v(t) \tag{2.32}$$

where H is the unperturbed Hamiltonian and $v(t)$ the perturbation:

$$v(t) = -\frac{e}{2mc}\sum_i[\vec{P}_i\cdot\vec{A}(\vec{r}_i,t) + \vec{A}(\vec{r}_i,t)\cdot\vec{P}_i] = -\frac{e}{c}\int\vec{j}(\vec{r})\cdot\vec{A}(\vec{r},t)\,d^3\vec{r} \tag{2.33}$$

where $j(r)$ is given by (1.121). On the other hand, the total current is given by:

$$\vec{J}(\vec{r}) = e\,j(\vec{r}) - \frac{e^2}{mc}\,n(\vec{r})\,\vec{A}(\vec{r},t) \tag{2.34}$$

By assuming that the system is homogeneous, we may write, in first approximation:

$$J_\alpha(\vec{r},t) = \frac{e^2}{c}\int_{-\infty}^{t}dt'\int d^3\vec{r}'\,X_{j_\alpha j_\beta}(\vec{r}-\vec{r}',t-t')\,A_\beta(\vec{r}',t') - \frac{e^2}{mc}nA\,(\vec{r},t) \tag{2.35}$$

Now, we can use the relation:

$$\vec{h}(\vec{r},t) = -\frac{1}{c}\frac{\partial}{\partial t}\vec{A}(\vec{r},t) \qquad \vec{A}(\vec{r},t) = -c\int_{-\infty}^{t}\vec{h}(\vec{r},t')\,dt' \tag{2.36}$$

or its Fourier transform:

$$\vec{A}(\vec{k},\omega) = \frac{c}{i\omega - 0}\vec{h}(\vec{k},\omega) \tag{2.37}$$

By comparing Eqs (2.28) and (2.33), we get:

$$\sigma_{\alpha\beta}(\vec{k},\omega) = \frac{e^2}{i\omega - 0}[X_{j_\alpha j_\beta}(k,\omega+i0) - \frac{n}{m}\delta_{\alpha\beta}] \tag{2.38}$$

In particular, if we denote the longitudinal part of any vector $\vec{g}(\vec{k})$ by the symbol $\vec{g}^L(\vec{k})$ with:

$$\vec{g}^L(\vec{k}) = \vec{k}(\vec{k}\cdot g(\vec{k}))/k^2 \tag{2.39}$$

we can write:

$$\vec{J}^L(\vec{k}\cdot\omega) = \sigma^L(\vec{k},\omega)\,\vec{h}^L(\vec{k},\omega) \tag{2.40}$$

with

$$\sigma^L(\vec{k},\omega) = \frac{e^2}{i\omega - 0}\left[\chi^L_{jj}(\vec{k},\omega + i0) - \frac{n}{m}\right] \tag{2.41}$$

where $\chi^L_{jj}(\vec{k},\omega)$ is defined as $\chi^{"L}_{jj}(\vec{k},\omega)$ in Eq.(1.137).

Then the sum rule (1.143) expresses the fact that, for a normal system, the d.c. conductivity ($\omega = 0$, $\vec{k} = 0$) is finite. However, it must be noted that, for free electrons, although the f sum rule remains valid for $\vec{k} \neq 0$, the d.c. conductivity is infinite. This anomaly can be related to the fact that in this case:

$$\lim_{\vec{k}\to 0} \chi^L_{jj}(\vec{k},\omega) \neq \chi_{jj}(0,\omega) = 0 \tag{2.42}$$

Another simple expression of the longitudinal conductivity can be obtained by using the Kubo formula of section 1.7. From Eqs (2.35) and (2.36), we deduce after partial integration with respect to t':

$$\vec{J}^L(\vec{r},t) = e^2\int_{-\infty}^{t} dt' \int d^3r' \int_{-\infty}^{t'} dt''\, \chi^L_{jj}(\vec{r}-\vec{r}', t-t'')\,\vec{h}^L(\vec{r}',t') \tag{2.43}$$

by using Eq.(2.36) and the f sum rule (see Eq.(1.142)):

$$\int_{-\infty}^{t} \chi^L_{jj}(\vec{r}-\vec{r}', t-t')\,dt' = \frac{n}{m}\,\delta(\vec{r}-\vec{r}') \tag{2.44}$$

Or, from Eq.(1.88):

$$\vec{J}^L(\vec{r},t) = e^2\int_{-\infty}^{t} dt' \int_0^{\beta} d\lambda \langle j^L(\vec{r}', -i\hbar\lambda)\, j^L(\vec{r}, t-t')\rangle\, h^L(\vec{r}',t')$$

$$= e^2\int_{-\infty}^{t} dt' \int_0^{\beta} d\lambda \langle j^L(\vec{r}', t'-i\hbar\lambda)\, j^L(\vec{r},t)\rangle\, \vec{h}^L(\vec{r}',t') \tag{2.45}$$

which gives:

$$\sigma^L(k,\omega) = e^2\int e^{ik(r-r')}\,d^3r \int_0^{\infty} dt\, e^{i\omega t}\int_0^{\beta} d\lambda \langle j^L(\vec{r}', -i\hbar\lambda)\, j^L(\vec{r},t)\rangle$$

$$= \Omega^{-1} e^2 \int_0^{\infty} dt\, e^{i\omega t}\int_0^{\beta} d\lambda \langle j^L(-\vec{k}, -i\hbar\lambda)\, j^L(\vec{k},t)\rangle \tag{2.46}$$

2.3. Einstein relation

The well known Einstein relation which connects the mobility of a set of particles with their diffusion constant D:

$$\mu = e\,D/KT \tag{2.47}$$

is a simple consequence of Eq.(2.45), in the classical limit ($\hbar = 0$). In fact, if we denote by v_s the velocity of the s^{th} particle along an arbitrary axis, we may write:

$$\mu = (en)^{-1}\sigma^L(0,0) = N^{-1} e\beta \int_0^{\infty} dt \sum_{ss'} \langle v_s(0)\, v_{s'}(t)\rangle$$

$$= e\beta \int_0^{\infty} dt \,\langle v(0)\, v(t)\rangle \tag{2.48}$$

where v(t) is the velocity of any particle (the mean density is $n = N/\Omega$ and $\beta = 1/KT$).
On the other hand: (for $\mathcal{T} \gg 1$)

$$\int_0^{\infty} dt \,\langle v(0)\, v(t)\rangle = \frac{1}{2\mathcal{T}}\int_0^{\mathcal{T}} dt \int_0^{\mathcal{T}} dt' \,\langle v(t)\, v(t')\rangle$$

$$= \frac{1}{2\mathcal{T}}\langle (x(\mathcal{T}) - x(0))^2\rangle \tag{2.49}$$

But a particle, which is at the origin at t = 0 has a probability $p(\vec{r},t)$ of being at $\vec{r}$ at time t which is given by:

$$p(\vec{r},t) = (4\pi Dt)^{-\frac{3}{2}}\, e^{-\frac{r^2}{4Dt}} \tag{2.50}$$

Therefore:

$$\left\langle \left(x(t) - x(0)\right)^2 \right\rangle = \int x^2 p(\vec{r},t)\, d^3r = 2Dt \tag{2.51}$$

Thus, from Eqs (2.48), (2.49) and (2.51), we can deduce Eq.(2.47).

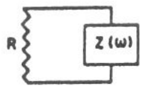


FIG.3. A circuit with a resistance R and a block box of independence $Z(\omega)$.

2.4. Nyquist theorem

The thermal noise of a resistance [7] can be calculated by direct application of the fluctuation dissipation theorem. Thus, let us consider a resistance connected to a black box of impedance $Z(\omega)$ (see Fig. 3) which will be assumed to be purely reactive ($Z(\omega) = iZ''(\omega)$) with $Z''(\omega)$ real. The conductivity of the loop consisting of the resistance and of the black box will be given by:

$$\sigma(\omega) = \frac{1}{i\omega}\,[\chi_{JJ}(\omega) - \chi_{JJ}(0)] \tag{2.52}$$

This formula is completely equivalent to Eq. (2.41) and can be derived in the same way. Here $J(\omega)$ is the Fourier transform of the total current:

$$J(\omega) = \int_{-\infty}^{+\infty} e^{i\omega t} J(t)\, dt$$

and $\chi_{JJ}(\omega)$ is the Fourier transform of:

$$\chi_{JJ}(t-t') = \hbar^{-1}\langle [J(t),\, J(t')]\rangle\, \Theta(t-t') \tag{2.53}$$

On the other hand, by definition, we have:

$$\sigma(\omega) = \frac{1}{R + Z(\omega)} = \frac{R - iZ''(\omega)}{R^2 + [Z''(\omega)]^2} \tag{2.54}$$

Therefore, by comparing Eqs (2.52) and (2.54), we get $(\chi''_{JJ}(0) = 0)$;

$$\omega^{-1}\chi''_{JJ}(\omega) = \frac{R}{R^2 + [Z''(\omega)]^2} \tag{2.55}$$

We can now apply the fluctuation dissipation theorem Eq. (1.102) or, more conveniently, its classical form given by Eq. (1.103) because in all cases of practical interest $\beta\hbar\omega \ll 1$.
Thus, we get:

$$\Phi_{JJ}(\omega) = 2\beta^{-1}\,\frac{R}{R^2 + [Z''(\omega)]^2} \tag{2.56}$$

On the other hand, according to Eqs (1.89) and (1.90), we have:

$$\begin{aligned}\langle\{J(\omega)J(-\omega')\}\rangle &= \int dt\, e^{i\omega t}\int dt'\, e^{-i\omega' t'}\langle\{J(t),\, J(t')\}\rangle \\ &= 2\int dt\int dt'\, e^{i(\omega t - \omega' t')} F_{JJ}(t-t') \qquad (2.57)\\ &= 4\pi\,\delta(\omega-\omega')\,\Phi_{JJ}(\omega)\end{aligned}$$

$J(\omega)$ can be considered here to be classical (i.e., a c-number) and therefore:

$$\langle J(\omega)J(-\omega')\rangle = 2\pi\,\delta(\omega-\omega')\,\Phi_{JJ}(\omega) \tag{2.58}$$

By comparing with Eq. (2.56), we finally obtain the Nyquist theorem:

$$\langle J(\omega)J(-\omega')\rangle = 4\pi\, KT\,\frac{R}{R^2 + [Z''(\omega)]^2}\,\delta(\omega-\omega') \tag{2.59}$$

The meaning of this theorem can be better understood if we simulate the effect of the thermal noise which produces current fluctuations in the black box. For this purpose, we may add:

a) either a current generator (current J_0) of infinite impedance in parallel with the resistance R (Fig. 4a).

b) or a voltage generator (e.m.f. E_0) in series with the resistance (Fig. 4b).

It is trivial to show that J_0 or E_0 must be given by:

$$\langle J_0(\omega)\, J_0(-\omega')\rangle = 4\pi\, KT R^{-1}\,\delta(\omega-\omega') \tag{2.60}$$

$$\langle E_0(\omega)\, E_0(-\omega')\rangle = 4\pi\, KT R\,\delta(\omega-\omega') \tag{2.61}$$

These results indicate clearly that the thermal noise comes only from the resistance and does not depend on the purely reactive black box at all.

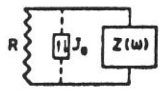


FIG.4a. Current generator in parallel with a resistance.

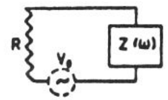


FIG.4b. Voltage generator in series with the resistance.

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