Scattering Experiments on Condensed Matter

The theoretical principles for the experimental methods

W.E. Fischer

Paul Scherrer Institute, CH-5232 Villigen PSI

1 Introduction to Scattering Theory (Various Approximations)

Let us consider the scattering of a particle (probe) on a target sample consisting of many bound scattering centers. This problem is described by a manybody hamiltonian

$$H = H_o + V \tag{1}$$

where H_o represents the hamiltonian for the situation where the sample and the probe are separated by a large distance. V describes the interaction between them as they are brought closer together. Note that the probing particle can be an electron, a photon or a neutron. V depends of course on the nature of coupling of this particle with the sample. This determines the kind of information and its complementarity which can be extracted from the data of these scattering experiments. The basic theoretical framework for the interpretation of scattering experiments which we are going to present in this chapter is essentially independent of the type of the incident probing particle.

The initial state before the scattering process — sample and probe being far apart — is described by the free channel state Φ to an energy E. Hence,

$$H_o\Phi = E\Phi \tag{2}$$

The scattering state is an eigenstate of H for the same energy E,

$$\boxed{H\Psi = E\Psi}. \tag{3}$$

If we introduce the Green's function

$$G = (E + i\epsilon - H_o)^{-1} \tag{4}$$

we can replace (3) by a Lippman-Schwinger equation

$$\Psi = \Phi + GV\Psi. \tag{5}$$

This guarantees that the state Ψ satisfies naturally the correct boundary conditions for the scattering problem. The $(i\epsilon)$ in the free Green's function (4) takes care of the proper

analyticity properties in the upper E-plane. In (5) we can identify

$$\begin{aligned} \Phi &: \text{with the incident wave} \\ GV\Psi &: \text{with the scattered wave} \end{aligned}$$

It is useful to define the T-matrix by [1)]

$$V\Psi = T\Phi \tag{6}$$

and obtain with

$$T = V(1 - GV)^{-1} \tag{7}$$

a formal solution of (5)

$$\boxed{\Psi = (1 + GT)\Phi} \tag{8}$$

For the T-matrix we have

$$\boxed{T = V + TGV} \tag{9}$$

Of course, the equation (9) stands in any specific representation for a system of integral equations. Formally, at least, we can expand (9) into a so-called Born series

$$T = V + VGV + VGVGV + \ldots \tag{10}$$

The first-Born approximation is given by

$$T \cong V. \tag{11}$$

Since the sample is a bulk system composed of a very large number of scattering centers labelled by an index i, we write

$$V = \sum_i V_i \tag{12}$$

We then get for (10)

$$T = \sum_i V_i + \sum_{i,j} V_i G V_j + \sum_{ijn} V_i G V_j G V_n + \ldots \tag{13}$$

$$\left.\begin{array}{rrcl} \text{with} & i \neq j & : & \text{interparticle} \\ & i = j & : & \text{intraparticle} \end{array}\right\} \text{multiple scattering}$$

Let us now write for the interparticle multiple scattering at the i^{th} constituent

$$T_i = V_i + T_i G V_i \tag{14}$$

T_i defines then the scattering amplitude on the constituents of the sample, e.g.

- atoms, for electron - or light scattering magnetic neutron scattering
- nuclei, for neutron scattering

V_i defines then a phenomenological potential leading to an "elementary" cross section or form factor,

$$V_i = T_i - T_i G V_i \tag{15}$$

$$V_i = T_i - T_i G T_i + T_i G T_i G T_i - \ldots \tag{16}$$

We then obtain

$$T = \sum_i T_i + \sum_{i \neq j} T_i G T_j + \sum_{i \neq j \neq n} T_i G T_j G T_n + \ldots \tag{17}$$

for the scattering amplitude, where the interparticle multiple scattering is now implicitly included up to infinite orders.

We interrupt here in order to give two examples from neutron- and electron scattering.

From (14) we have

$$T_i = V_i (1 - G V_i)^{-1} \simeq V_i$$

in first-Born approximation hereafter referred to as the Born approximation. $V_i(\vec{x})$ will be a phenomenological optical potential.

i) Neutron as a probe

In this case V_i is a generally complex potential with a range of $\sim 10^{-12}$ cm and a depth of ~ 40 MeV. The interatomic potential has a range of 10^{-8} cm and a depth of the order of 1 eV. Due to the short interaction time of the neutron with a nucleus we can neglect interatomic forces and an impulse approximation is justified. We can then define a pseudo-potential, which — for low energy neutrons – contains one parameter; the scattering length b.

$$T_i = \frac{2\pi\hbar^2}{m_o} a_i \delta(\vec{x}) = \frac{2\pi\hbar^2}{m} b_i \delta(\vec{x}) \tag{18}$$

Instead of solving (14), we assume (18) and determine b_i from empirical data. Hence the Born approximation is valid by the definiton of this procedure. (Note that the Fourier transform of the δ-function is a constant. Therefore the elementary scattering is isotropic and does not depend on the momentum transfer.)

ii) Electron as a probe

Electrons interact with the atomic charge distribution $\rho(\vec{x})$

$$V_i(\vec{x}) = \int d^3x \frac{\rho_i(\vec{x})}{|\vec{x} - \vec{x}'|} \tag{19}$$

In Born approximation we then obtain

$$\begin{aligned} T_i(\vec{q}) &= \int d^3x e^{i\vec{q}\vec{x}} V(\vec{x}) \\ &= \frac{2\pi}{q^2} F(\vec{q}) \end{aligned} \tag{20}$$

with the form factor

$$\begin{aligned} F(\vec{q}) &= \int d^3x e^{i\vec{q}\vec{x}} \rho(\vec{x}) \\ F(o) &= Q \end{aligned} \tag{21}$$

where Z is the total charge of the atomic electrons. The square of the amplitude therefore is given by the Rutherford cross section. A similar treatment for photons leads to the Thomson cross section for light scattering.

We now go back to equation (17) and write it in the following form (Ewald equations).

$$\begin{aligned} T &= \sum T_i(1+GZ_i) \\ Z_i &= \sum_{j\neq i} T_j(1+GZ_j) \end{aligned} \tag{22}$$

These equations become somewhat more transparent if we write them for the states. With the definition

$$\chi_i = (1+GZ_i)\Phi \tag{23}$$

$$\boxed{\begin{aligned} \Psi &= \Phi + \textstyle\sum_i GT_i\chi_i \qquad (24a) \\ \chi_i &= \Phi + \textstyle\sum_{j\neq i} GT_j\chi_j. \qquad (24b) \end{aligned}}$$

Equation (24a) shows that the scattered wave Ψ consists of

Φ	:	the incident wave
$GT_i\chi_i$	:	the sum of scattered waves from each scattering center in the sample

χ_i, according to (24b) is the local field at the i^{th} particle and contains

Φ	:	the incident wave
$GT_j\chi_j$	:	the scattered contribution from every **other** center in the sample

GZ_i is called the local field correction to the transition operator T_i.

1.1 Kinematical Approximation

This approximation is defined by neglection of the intraparticle multiple-scattering or equivalently the local field corrections. That is in (22),

$$T = \sum_i T_i \tag{25}$$

Obviously, this approximation can only be valid for samples, which — related to the strength of the interaction — are "small". The smallness depends on the size and the texture of the sample. Only if these kind of conditions are fulfilled, does it make sense to interprete the scattering data within this approximation and usually called the kinematically approx.

For the investigations of optical phenomena like dispersion, reflection and refraction (especially of perfect crystals) the kinematical approximation is usually not sufficient. Phenomena like these, and like extinction effects, are explained by multiple scattering processes and hence call for a **dynamical treatment**.

However, if the experimental set up permits an interpretation of the data within the kinematical approximation, its linearity facilitates the procedure. For experiments for structural research this is almost always the case for neutrons as probing particles, often for X-rays and almost never for charged particles.

Let us now formulate the scattering problem in a somewhat less formal style. To be more explicit we write the free hamiltonian

$$H_o = h + H_o^{(s)} \tag{26}$$

where

$$h = \frac{1}{2m}\vec{p}^{\,2} \tag{27}$$

describes the incident particle and $H_o^{(s)}$ the unperturbed target sample. The corresponding states belong to (and form) two different Hilbert spaces $\mathcal{H}$ in the following sense

$$\text{i)} \quad \mathcal{H}_n: \quad h|\vec{k}>= E_{\vec{k}}|\vec{k}> \tag{28}$$

$$\text{ii)} \quad \mathcal{H}_s: \quad H_o^{(s)}|\alpha>= E_a|\alpha> \tag{29}$$

$$<\alpha|\alpha'>= \delta_{\alpha\alpha'}$$

We assume $\mathcal{H}_s$ to be complete, that is

$$\sum_\alpha |\alpha><\alpha| = 1 \tag{30}$$

We then define the channel state as an element of the product space

$$\mathcal{H} = \mathcal{H}_n \otimes \mathcal{H}_s \tag{31}$$

and ϕ as a product wave function

$$\Phi = |\vec{k}> |\alpha >= |\vec{k}, \alpha > \tag{32}$$

to the energy

$$E = E_{\vec{k}} + E_\alpha \tag{33}$$

The channel state Φ develops into the scattering state Ψ by switching on the interaction potential,

$$\Psi = \sum_{\alpha'} \psi_{\alpha\alpha'} |\alpha' > \tag{34}$$

with

$$\psi_{\alpha\alpha'} =< \alpha'|\Psi > \epsilon \mathcal{H}_n$$

being the (one body) scattering state of the incident particles, corresponding to a transition in the sample $\alpha \longrightarrow \alpha'$.

According to (8) we have,

$$\begin{aligned} \psi_{\alpha\alpha'} &= < \alpha'|(1 + GT)|\vec{k}, \alpha > \\ &= \left\{ < \alpha'|\alpha > + \sum_{\alpha''} < \alpha'|G|\alpha'' >< \alpha''|T|\alpha > \right\} |\vec{k} > \end{aligned} \tag{35}$$

and

$$G = \left(E_{\vec{k}} + E_\alpha + i\epsilon - h - H_o^{(s)} \right)^{-1}. \tag{36}$$

For the matrix element of G we obtain

$$\begin{aligned} < \alpha'|G|\alpha'' > &= g' \delta_{\alpha'\alpha''} \\ & \qquad g' = \left(E_{\vec{k}'} + i\epsilon - h \right)^{-1} \end{aligned} \tag{37}$$

with

$$E_{\vec{k}'} = E_{\vec{k}} + E_\alpha - E_{\alpha'}.$$

Here, $\vec{k}'$ is the wave vector of the scattered neutron.

Finally,

$$\boxed{\psi_{\alpha\alpha'} = \{\delta_{\alpha\alpha'} + g' < \alpha'|T|\alpha >\} |\vec{k} >}. \tag{38}$$

We now write T as

$$T = t + \delta T \tag{39}$$

where $t =< T >$ is the average of the scatt. matrix e.g. for neutrons given by

$$V(\vec{x}) =< \sum_j \frac{2\pi\hbar^2}{m} b_j \delta\left(\vec{x} - \vec{X}_j\right) > \tag{40}$$

The variation δT describes a range of processes, including:

i) b_j may fluctuate about an average due to random distributions of isotopes in the sample (remember that different isotopes have different scattering cross sections). A similar argument is valid for nuclei with spins (spin exchange scattering).

ii) The positions of the nuclei fluctuate, due to thermal motion in the sample.

For the T-matrix element we then write

$$< \alpha'|T|\alpha >= t\delta_{\alpha\alpha'} + < \alpha'|\delta T|\alpha > \tag{41}$$

and obtain for (38)

$$\boxed{\psi_{\alpha\alpha'} = \psi\delta_{\alpha'\alpha} + \delta\psi_{\alpha\alpha'}} \tag{42}$$

with

$$\psi = (1 + gt)|\vec{k} >; \quad g = \left(E_{\vec{k}} + i\epsilon - h\right)^{-1} \tag{43}$$

and

$$\delta\psi_{\alpha'\alpha} = g' < \alpha'|\delta T|\alpha > |\vec{k} > \tag{44}$$

where g' is given by (37).

The first term of (42) describes the **strictly elastically** scattered wave. It is **coherent in the absolute sense**, that is there is interference between the incident wave and the scattered wave. $\delta\psi_{\alpha\alpha'}$ (sometimes called diffuse scattering) is **incoherent in the absolute sense**. It does not interfere with the incident wave. It might be **coherent in the relative sense**, that is there may by interference among the scattered contributions from the different scattering centers (coherent inelastic). Usually $\delta\psi_{\alpha\alpha'}$ has also a contribution, which is incoherent.

2 Cross Section and Scattering Function

2.1 Coherence and Incoherence

The measured cross section of the inelastic scattering process is defined as the ratio of the number of scattered particles with momentum $\vec{k}_1$ within the solid angle $d\Omega$ and an energy resolution ΔE, to the incident flux with momentum $\vec{k}_o$. Note that $\vec{k}_1$ determines the direction Ω and the energy E_1 of the scattered particles.

$$\vec{\kappa} = \vec{k}_o - \vec{k}_1, \qquad\qquad \hbar\omega = E_o - E_1 \tag{45}$$

are the momentum — and the energy transfer in the scattering process. The partial differential cross section is then

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k_1}{k_o} x_1^2 d\Omega \sum_{\Delta E} |\psi_s|^2 \tag{46}$$

x_1 is the distance from the sample to the detector; it has to be large compared to the dimensions of the sample. It then finally drops out of the final expression since $|\psi_s|^2$ turns out to be proportional to x_1^{-2}. The sum runs over the number of terms in the resolution range ΔE.

Hence, all we have to do is to calculate $|\psi_s|^2$ — the scattered wave — in the appropriate approximations. We have

i) Born approximation $T \simeq V$

ii) kinematical approximation with

$$V(t) = \sum_j \frac{2\pi\hbar^2}{m} b_j \delta\left(\vec{x} - \vec{X}_j(t)\right) \tag{47}$$

for neutron scattering.

The $\vec{X}_j(t)$ are the positions of the scattering centers in the sample.

For the incident wave (channel state) we take

$$< \vec{x}|\vec{k} >= e^{i\left(\vec{k}_o\vec{x} - \omega_o t\right)} \tag{48}$$

The time-dependent version of the scattered wave written out explicitly is,

$$\psi_s(\vec{x}', t) = \int d^3x \int dt' G(\vec{x} - \vec{x}'; t - t') \cdot \sum_j b_j \delta\left(\vec{x} - \vec{X}_j(t')\right) \cdot e^{i\left(\vec{k}_o\vec{x} - \omega_o t'\right)}. \tag{49}$$

where the Green's function G satisfies

$$\left(\Delta + \frac{2mi}{\hbar}\partial_t\right) G\left(\vec{x} - \vec{x}'_j; t - t'\right) = 4\pi\delta\left(\vec{x} - \vec{x}', t - t'\right). \tag{50}$$

We do not have the time here to go through the full procedure to obtain $|\psi_s|^2$. Although the job is somewhat tedious, every student should however have seen it once. I shall therefore distribute a hand-written version of this calculation – including (hopefully) every i and π. The final result is

$$\boxed{\frac{d^2\sigma}{d\Omega dE} = \frac{k_1}{k_o}\frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty} d\tau e^{-i\omega\tau} \sum_{ij} b_i^* b_j \overline{e^{i\vec{\kappa}\vec{X}_i(\tau)} e^{-i\vec{\kappa}\vec{X}_j(0)}}}. \tag{51}$$

The bar over the exponentials indicates a time average of the type

$$\lim_{T\to\infty}\frac{1}{T}\int_o^T dt' \tag{52}$$

The scattering lengths b_i are assumed to be (exclude compound unclear states) constant in time; we may take them therefore in front of the integral. On the other hand they may be different for different indices (e.g. different isotopes as scattering centers). For the average over the sample we write

$$\overline{b_i^* b_j} = \bar{b}_i^* \bar{b}_j = |\bar{b}|^2 \text{ for } i \neq j \tag{53}$$

assuming random distribution of isotopes (hence no correlation between $i \neq j$). Furthermore

$$\overline{b_i^* b_j} = \overline{|b_j|^2} = \overline{|b|^2} \text{ for } i = j. \tag{54}$$

For the double sum we get then

$$\sum_{i\neq j}\overline{b_i^* b_j} = N(N-1)|\bar{b}|^2 \tag{55}$$

$$\sum_{i=j}\overline{b_i^* b_j} = N\overline{|b|}^2 \tag{56}$$

All together,

$$\begin{aligned}\sum_{ij}\overline{b_i^* b_j} &= N^2|\bar{b}|^2 + N\left(\overline{|b|^2} - |\bar{b}|^2\right)\\ &= N^2|\bar{b}|^2 + N\overline{(\delta b)^2}\end{aligned}$$

The second term is the average of the squared fluctuation. It is proportional to the number of scattering centers ($\sim N$) and describes therefore incoherent scattering

$$\overline{(\delta b)^2} = b^2{}_{\text{inc}} \tag{57}$$

The first term — proportional to N^2 — describes the coherent scattering; in the absolute sense for elastic ($\tau \to \infty$), in the relative sense for inelastic scattering.

$$|\bar{b}|^2 = b_c^2 \tag{58}$$

The exponentials in (51) form a time correlation function, whose variables are the position operators of the scattering centers. Assuming the ergodic principle (in the sense of Birkhoff) for the sample, we may replace the time average by an average over a canonical statistical ensemble.

$$< e^{i\vec{\kappa}\vec{X}_i(\tau)} e^{-i\vec{\kappa}\vec{X}_j(o)} >_T = \mathrm{Sp}\left(\rho e^{i\vec{\kappa}\vec{X}_i(\tau)} e^{-i\vec{\kappa}\vec{X}_j(0)}\right) \tag{59}$$

where ρ is the density operator of the canonical ensemble

$$\rho = \frac{e^{-\beta H_o^{(s)}}}{\mathrm{Sp}\left(e^{-\beta H_o^{(s)}}\right)} \tag{60}$$

for the temperature $T(\beta = \frac{1}{kT})$ of the sample. Note that $H_o^{(s)}$ is the hamiltonian of the sample to be investigated.

The partition function

$$Z = \mathrm{Sp}\left(e^{-\beta H}\right) = e^{-\beta F} \tag{61}$$

where

$$F = -kTlnZ \tag{62}$$

is the free energy of the sample. (We write from now on H instead of $H_o^{(s)}$ for its hamiltonian). The expectation value of an operator $\hat{O}$ which represents an observable of the sample in thermal equilibrium at temperature T is

$$< \hat{O} >_T = \mathrm{Sp}\ (\rho \hat{O}) = \frac{\mathrm{Sp}\left(e^{-\beta H}\hat{O}\right)}{\mathrm{Sp}\,(e^{-\beta H})} = \frac{\mathrm{Sp}\left(e^{-\beta H}\hat{O}\right)}{Z} \tag{63}$$

and explicitly in terms of energy eigenvalues and eigenvectors,

$$< \hat{O} >_T = \sum_i \frac{e^{-\beta E_i}}{Z} < i|\hat{O}|i > . \tag{64}$$

We now use this development to define the coherent and incoherent differential cross section and write

$$\left.\frac{d^2\sigma}{d\Omega dE}\right|_{\mathrm{coh}} = \frac{k_1}{k_o}\frac{\sigma_c}{4\pi}\frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty} d\tau e^{-i\omega\tau} \sum_{ij} < e^{i\vec{\kappa}\vec{X}_i(\tau)} e^{-i\vec{\kappa}\vec{X}_j(0)} >_T \tag{65}$$

$$\left.\frac{d^2\sigma}{d\Omega dE}\right|_{\text{incoh}} = \frac{k_1}{k_o}\frac{\sigma_i}{4\pi}\frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty} d\tau e^{-i\omega\tau}\sum_j < e^{i\vec{\kappa}\vec{X}_j(\tau)}e^{-i\vec{\kappa}\vec{X}_j(0)} >_T \tag{66}$$

where we have introduced,

$$\sigma_c = 4\pi|\bar{b}|^2, \quad \sigma_i = 4\pi\overline{|\delta b|^2}. \tag{67}$$

Coherent and incoherent cross sections are then time-Fourier transforms of a correlation function

$$Y_{ij}(\kappa,\tau) =< e^{+i\vec{\kappa}\vec{X}_i(\tau)}e^{-i\vec{\kappa}\vec{X}_j(o)} >_T \tag{68}$$

We now show that this corresponds to a density correlation function in space and time

$$C_{ij}(\vec{x},\tau) = \left(\frac{1}{2\pi}\right)^3 \int d^3\kappa e^{-i\vec{\kappa}\vec{x}} Y_{ij}(\kappa\tau). \tag{69}$$

With the exponential representation of the δ-function e.g.

$$\left(\frac{1}{2\pi}\right)^3 \int d^3\kappa e^{-i\vec{\kappa}\left(\vec{x}-\vec{x}'+\vec{X}_j(o)\right)} = \delta^{(3)}\left(\vec{x}-\vec{x}'+\vec{X}_j(0)\right) \tag{70}$$

we obtain

$$C_{ij}(\vec{x},\tau) = \int d^3x' < \delta\left(\vec{x}'-\vec{X}_i(\tau)\right)\delta\left(\vec{x}-\vec{x}'+\vec{X}_j(0)\right) >_T . \tag{71}$$

For the double sum

$$\boxed{C(\vec{x},\tau) = \sum_{ij} C_{ij}(\vec{x},\tau) = \frac{1}{N}\int d^3x' < \rho\left(\vec{x}',\tau\right)\cdot\rho\left(\vec{x}-\vec{x}',0\right) >_T} \tag{72}$$

with

$$\boxed{\rho\left(\vec{x},\tau\right) = \sum_j \delta\left(\vec{x}-\vec{X}_j(\tau)\right)} \tag{73}$$

According to (65) and (66) we define two different correlation functions, depending on the summation procedure over the scattering centers, namely the

i) sum over all i and j gives the general coherent correlation function $C(\vec{x}, \tau)$. It defines pair correlation in space and time.

ii) the sum over $i = j$ only, gives the correlation function which determines the incoherent scattering $C_S(\vec{x}, \tau)$. It defines a one particle (self)-correlation in space and time.

This point of view to interpret such scattering experiment has been introduced by Leon van Hove [2]). In a somewhat popular language these experiments answer the question:

"Where are the positions of the atoms and how do they move."

It is evident that the correlation function $C(\vec{x}, \tau)$ contains information about collective excitations in dependence of the temperature of the sample. The incoherent correlation function $C_s(\vec{x}, \tau)$ (among others) describes diffusion processes in the sample.

Note that in principle a similar approach is also possible for magnetic neutron scattering and also for X-ray and electron-scattering experiments. Due to the different coupling of these probes to the scattering centers, the information obtained is complementary. The various probes are sensitive to different kind of correlations (fluctuations) in the sample; namely:

Neutrons	nuclear	mass-density
	magnetic	density of magnetization
X-rays		electric charge density and to order $\left(\frac{v}{c}\right)$ also density of spin and orbital magnetizations
Electrons		(like x-rays)

We shall discuss this point further, later on. Following van Hove, let us define here the

scattering function:

$$S(\vec{\kappa},\omega) = \frac{1}{N}\frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty} d\tau e^{-i\omega\tau}\sum_{ij} Y_{ij}(\vec{\kappa},\tau) \tag{74}$$

which has the dimension of (y energy). For the cross sections we then have

$$\boxed{\left.\frac{d^2\sigma}{d\Omega dE}\right|_{\text{coh}} = N\frac{k_1}{k_o}\frac{\sigma_c}{4\pi}S(\vec{\kappa},\omega)} \tag{75}$$

$$\boxed{\left.\frac{d^2\sigma}{d\Omega dE}\right|_{\text{incoh}} = N\frac{k_1}{k_o}\frac{\sigma_{in}}{4\pi}S_i(\vec{\kappa},\omega)} \tag{76}$$

where S_i contains only the sum of the diagonal terms of Y_{ij}. The structure function is the spatial and temporal Fourier transform of the correlation function $C(\vec{x},\tau)$.

2.2 Elastic- and Inelastic Scattering [3)]

From (73) we write for the microscopic particle density, which has dimension (y volume),

$$\rho(\vec{x},\tau) = \sum_j \delta\left(\vec{x} - \vec{X}_j(\tau)\right) = \frac{1}{V}\sum_{\vec{\kappa}} \rho_{\vec{\kappa}} e^{i\vec{\kappa}\vec{x}}, \tag{77}$$

with the Fourier components,

$$\rho_{\vec{\kappa}} = \sum_j e^{-i\vec{\kappa}\vec{X}_j(\tau)}, \tag{78}$$

and then for the scattering function according to (68) and (74)

$$S\left(\vec{\kappa},\omega\right) = \frac{1}{2\pi\hbar}\frac{1}{N}\int_{-\infty}^{+\infty} d\tau e^{-i\omega\tau} < \rho_{\vec{\kappa}}(\tau)\rho^+_{\vec{\kappa}}(0) >_T . \tag{79}$$

For very large times $\rho_{\vec{\kappa}}(\tau)$ and $\rho^+_{\vec{\kappa}}(0)$ can be considered as uncorrelated, hence

$$\lim_{\tau\to\infty} < \rho_{\vec{\kappa}}(\tau)\rho^+_{\vec{\kappa}}(0) >_T = < \rho_{\kappa}(\tau) >< \rho^+_{\kappa}(0) > \tag{80}$$

Accordingly we split the correlation function (the coherent as well as the incoherent) into two parts.

$$C\left(\vec{x},\tau\right) = C(\vec{x},\infty) + C'(\vec{x},\tau) \tag{81}$$

with

$$\lim_{\tau\to\infty} C'(\vec{x},\tau) = 0. \tag{82}$$

The first term describes the sustainable part of the structure (e.g. crystal structure), the second part is given by the actual fluctuations. We now show, that the first term is described by elastic scattering and the second by inelastic scattering phenomena. For the coherent correlation function this is exactly in the spirit of the interpretation of equations (41 – 44).

The cross section related to $C(x,\infty)$ is

$$\begin{aligned}\frac{d^2\sigma}{d\Omega dE} &= N\frac{\sigma_c}{4\pi}\delta(\hbar\omega)\int d^3x e^{i\vec{\kappa}\vec{x}} C(\vec{x},\infty) \\ &= \frac{\sigma_c}{4\pi}\delta(\hbar\omega)\left|\int d^3x e^{i\vec{\kappa}\vec{x}} < \rho(\vec{x}) >\right|^2 .\end{aligned} \tag{83}$$

For a static (rigid) Bravais Lattice defined by vectors $\{1\}$,

$$\rho(\vec{x}) = \sum_l \delta(\vec{x} - \vec{l}) \tag{84}$$

we have

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{coh}} = \frac{\sigma_c}{4\pi}\left|\sum_{\vec{l}} e^{i\vec{\kappa}\vec{l}}\right|^2 \tag{85}$$

There is no inelastic scattering from, a static lattice. Similarly, for the self-correlation $C_S(\vec{x},\infty)$ we obtain

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{inc}} = \frac{\sigma_i}{4\pi}\sum_j \left|e^{i\vec{\kappa}\vec{X}_j}\right|^2 = N\frac{\sigma_i}{4\pi}, \tag{86}$$

an isotropic diffuse scattering.

Note in (85) we can use the identify

$$\left|\sum_{\vec{l}} e^{i\vec{\kappa}\vec{l}}\right|^2 = N\frac{(2\pi)^3}{V_o}\sum_{\tau}\delta\left(\vec{\kappa}-\vec{\tau}\right), \tag{87}$$

where V_o is the volume of the unit cell, and $\{\vec{\tau}\}$ the lattice vectors of the reciprocal lattice defined from $\{1\}$. (87) shows the Bragg peaks i.e. the elastic scattering is zero at all wave vectors except $k = \tau$..

For a **non rigid** lattice we assume the positions of the scattering centers to be given by

$$\vec{X}_j = \vec{l}_j + \vec{u}_j\left(\vec{l}_j\right) \tag{88}$$

This represents a Bravais lattice with average positions $\vec{l}_j$ and (time dependent) deviations from these positions $\vec{u}_j(\vec{l}_j)$ (e.g. due to thermal motion).

$$< u_j\left(\vec{l}_j\right) >_T = 0 \tag{89}$$

With (88) we can write for the average particle density

$$\begin{aligned} < \rho(\vec{x}) > &= \sum_j < \delta\left(\vec{x}-\vec{X}_j\right) >_T = \\ &= \left(\frac{1}{2\pi}\right)^3 \int d^3\kappa \sum_j e^{i\vec{\kappa}(\vec{x}-\vec{l})} < e^{-i\vec{\kappa}\vec{u}(\vec{l})} >_T \end{aligned} \tag{90}$$

By means of the Bloch relation, valid for harmonic vibrations,

$$< e^{\hat{Q}} > = e^{\frac{1}{2}<\hat{Q}^2>} \tag{91}$$

we rewrite the last term under the integral (90) as

$$< e^{-i\vec{\kappa}\vec{u}(\vec{l})} > = e^{-\frac{1}{2}<(\vec{\kappa}\vec{u}(l))^2>_T} = e^{-W(\vec{\kappa})} \tag{92}$$

in (92),

$$\boxed{2W(\vec{\kappa}) =< (\vec{\kappa}\vec{u}(\vec{l}))^2 >_T} \tag{93}$$

is the famous Debye-Waller factor. Note that (91) is an identity for a quantum mechanical observable which is a linear combination of annihilation and creation phonon operators. We have to deal with $\vec{u}(\vec{l})$ accordingly.

We do not give here the prove of Bloch's relation; it can be found in the literature [3]. It contains basically the quantum mechanical treatment of the degrees of freedom of the sample. Accordingly, due to zero point motions the Debye-Waller factor does not vanish at $T = 0$.

For ℓ the special case of the three cubic Bravais lattices we can write

$$W(\vec{\kappa}) = \frac{1}{2} < (\vec{\kappa}\vec{u})^2 >= \frac{1}{6}\kappa^2 < \vec{u}^2 > \tag{94}$$

and according to (90)

$$< \rho(\vec{\kappa}) >= \left(\frac{3}{2\pi < u^2 >}\right)^{\frac{3}{2}} \sum_l e^{-\frac{|\vec{x}-\vec{l}|^2}{2<u^2>}} \tag{95}$$

is a Gaussian form-factor centered at the equilibrium position of the scattering centers. The elastic cross sections are now

$$\left(\frac{d\sigma}{d\Omega}\right)^{\text{el}}_{\text{coh}} = \frac{N\sigma_c}{4\pi}\frac{(2\pi)^3}{V_o}\sum_{\vec{\tau}} \delta\left(\vec{\kappa} - \vec{\tau}\right) e^{-2W(\vec{\kappa})} \tag{96}$$

and,

$$\left(\frac{d\sigma}{d\Omega}\right)^{\text{el}}_{\text{incoh}} = \frac{N\sigma_i}{4\pi} e^{-2W(\vec{\kappa})}. \tag{97}$$

We recognize that the Debye-Waller factor reduces the elastic cross sections. Indeed, it is a measure of the probability for the reaction being elastic. Furthermore, the incoherent scattering — due to the $\vec{\kappa}$-dependence of the Debye-Waller factor — in general, is not isotropic anymore.

The correlations in the sample described by $C'(\vec{x}, \tau)$ in a nonrigid lattice are responsible for inelastic scattering processes. Energy carried by the internal degrees of freedom can be transferred to the probe. The reversed process is also possible. The probability of these two processes are intimately related with each other, as we shall see.

We do not intend in this lecture to discuss the inelastic cross sections in detail. I just would like to emphasize a few points, which will turn out to be of importance for the

discussion of general properties of the correlation function and the approach of "Linear Response".

We start according to (81) with

$$C'(\vec{x},\tau) = C(\vec{x},\tau) - C(\vec{x},\infty). \tag{98}$$

With (88) and a similar procedure as above we obtain

$$\begin{aligned} C'(\vec{x},\tau) &= \left(\frac{1}{2\pi}\right)^3 \sum_{ik} \int d^3\kappa e^{-i\vec{\kappa}\vec{x}} e^{-i\vec{\kappa}\left(\vec{l}_j - \vec{l}_k\right)} e^{-2W(\vec{\kappa})} \\ &\quad \cdot \left\{ \exp\left[< \vec{\kappa}\vec{u}\left(\vec{l}_j\right) \cdot \vec{\kappa}\vec{u}\left(\vec{l}_k,\tau\right) >_T \right] - 1 \right\}. \end{aligned} \tag{99}$$

The deviation of the position of an atom from its equilibrium position can be represented as a linear combination of the eigenmodes of the crystal oscillations,

$$\vec{u}(\vec{l}) = \sum_{j,\vec{q}} \left(\frac{\hbar}{3NM\omega_j(\vec{q})}\right)^{1/2} \left[\vec{\alpha}^{(j)}(\vec{q}) e^{i\vec{q}\vec{l}} a_j(\vec{q},t) + \vec{\alpha}^{(j)*}(\vec{q}) e^{-i\vec{q}\vec{l}} a_j^+\right]. \tag{100}$$

The dynamics of the oscillations is governed by the equation

$$i\hbar\partial_t a_j = [a_j, \mathcal{H}] \tag{101}$$

where $\mathcal{H}$ is the hamiltonian of the lattice oscillations. In the harmonic approximation (101) reduces to $i\hbar\partial_t a_j = \hbar\omega_i a_j$ and describes $3N$ independent harmonic oscillations

$$a_j(\vec{q},t) = a_j(\vec{q}) e^{-i\omega_j(\vec{q})t} \tag{102a}$$

$$a_j^+(\vec{q},t) = a_j^+(\vec{q}) e^{i\omega_j(\vec{q})t} \tag{102b}$$

The a_j and a_j^+ are the usual annihilation and creation operators, fulfilling the well-known commutation relations of the linear oscillator. Furthermore the following averages over the canonical partition are obtained

$$< a_j^+(\vec{q}) a_k(\vec{q}\,') >_T = \delta_{jk}\delta_{qq'} n_j(\vec{q}) \tag{103a}$$

$$< a_j(\vec{q}) a_k^+(\vec{q}\,') >_T = \delta_{jk}\delta_{qq'}(n_j(\vec{q}) + 1), \tag{103b}$$

in which $n_j(\vec{q})$ describes the population of the mode j

$$n_j(\vec{q}) = (e^{\beta\hbar\omega_j(\vec{q})} - 1)^{-1} \tag{104}$$

The exponent in the Debye-Waller factor then becomes

$$< \left[\vec{\kappa}\vec{u}(\vec{l})\right]^2 >_T \sim \sum_{j,\vec{q}} \frac{\left|\vec{\kappa}\vec{\alpha}^{(j)}(\vec{q})\right|^2}{\omega_j(\vec{q})} (2n_j(\vec{q}) + 1) \tag{105}$$

which demonstrates, how this factor depends on the population of the lattice excitations.

For the time-dependent exponent in (99) we get

$$\begin{aligned} < \quad & \vec{\kappa}\vec{u}(\vec{l_i}) \cdot \vec{\kappa}\vec{u}(\vec{l_k}, \tau) >_T = \\ & \sum_{j,\vec{q}} \frac{|\vec{\kappa}\vec{\alpha}^j(\vec{q})|^2}{\omega_j(\vec{q})} \left[e^{i\vec{q}(\vec{l}_k - \vec{l}_j)} e^{i\omega_j(\vec{q})\tau} (1 + n_j(\vec{q})) + e^{-i\vec{q}(\vec{l}_k - \vec{l}_j)} e^{-i\omega_j(\vec{q})\tau} \cdot n_j(\vec{q})\right] \end{aligned} \tag{106}$$

Expansion of exp[< $>_T$] in (99) and keeping the linear term only, leads — using (106) — to a scattering function and hence an inelastic coherent cross-section which contains typical terms proportional to

$$1) \qquad n_j(\vec{q})\delta(\omega + \omega_j(\vec{q}))\delta(\vec{\kappa} + \vec{q} - \vec{K}) \tag{107}$$

$$2) \qquad (n_j(\vec{q}) + 1)\delta(\omega - \omega_j(\vec{q}))\delta(\vec{\kappa} - \vec{q} - \vec{K}). \tag{108}$$

The first term represents to a process of phonon annihilation (absorption) $(\omega_j(\vec{q}), \vec{q})$ by the probing incident particle. The second term is the corresponding phonon creation term. Energy- and momentum conservation for these inelastic processes are,

$$E' = E \pm \hbar\omega_j(\vec{q}) \tag{109}$$

$$\vec{k}' = \vec{k} \pm \vec{q} - \vec{K} \tag{110}$$

where $\vec{K}$ is a reciprocal lattice vector.

3 Properties of the Scattering - and the Correlation-Function[3)]

With the help of (77) and (78) we write the scattering function in the following form

$$S(\vec{\kappa},\omega) = \frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty} dt e^{-i\omega t} < \rho_{\vec{\kappa}}(0)\rho_{-\vec{\kappa}}(t) >_T . \tag{111}$$

Since the position- and momentum operators are self-adjoint (observables), the Fourier-component of the density operator has the property

$$\rho_{-\vec{\kappa}} = \rho_{\vec{\kappa}}^{+} \tag{112}$$

where + stands for hermitian conjugation.

$\rho_{-\vec{\kappa}}(t)$ with $t \neq 0$ and $\rho_{\vec{\kappa}}(0)$ do not usually commute - therefore, instead of the relation

$$e^A \cdot e^B = e^{A+B} \tag{113}$$

which is for classical number, A and B, when A and B are quantum operators we have to use,

$$e^A \cdot e^B = e^{A+B+C} \tag{114}$$

when the operator C can be expressed as an expansion of nested commutators,

$$C = \frac{1}{2}[A,B] + \frac{1}{12}[[A,B],B] + \frac{1}{12}[[B,A],A] + ... \tag{115}$$

This is easily verified by expanding the exponents for

$$A = -i\kappa\vec{X}_i(0), \qquad B = i\vec{\kappa}\vec{X}_j(t). \tag{116}$$

Returning to (112) we can show that

$$\boxed{< \rho_{\vec{\kappa}}(0)\rho_{\vec{\kappa}}^{+}(t) >_T^{*} = < \rho_{\vec{\kappa}}(t)\rho_{\kappa}^{+}(0) >_T} \tag{117}$$

where * stands for conjugate complex.

The average for $<>_T$ describes the many-body system in thermal equilibrium. In this stationary state, it is therefore invariant under a translation operation in time, and

$$\boxed{< \rho_{\vec{\kappa}}(-t)\rho^+_{\vec{\kappa}}(0) >_T = < \rho_{\vec{\kappa}}(0)\rho^+_{\vec{\kappa}}(t) >_T} \tag{118}$$

Note that these relations are generally only true for the thermal average and not as proper operator relations.

These two symmetry relations guarantee that the Fourier transform (111) is a purely real function. The scattering function $S(\vec{\kappa}, \omega)$ is therefore real - as it has to be the case for an experimentally accessible cross section.

The correlation function $C(\vec{x}, t)$ is generally not purely real. Under the conditions mentioned above, the following relationship is obtained

$$\boxed{C(\vec{x}, t) = C^*(-\vec{x}, -t)} \tag{119}$$

Let us express ReC and ImC in the form of anticommutator and commutator

$$ReC(\vec{x}, t) = \frac{1}{2} \int d^3x' < \{\rho(\vec{x}\,', t), \rho(\vec{x}\,' - \vec{x}, 0)\} >_T \tag{120}$$

and

$$ImC(\vec{x}, t) = \frac{i}{2} \int d^3x' < [\rho(\vec{x}\,', t), \rho(\vec{x}\,' - \vec{x}, 0)] >_T \tag{121}$$

Since the density operators do not commute for $t \neq 0$ the imaginary part of C (121) does not vanish. This condition - not valid generally - would be equivalent to

$$< \rho_{\vec{\kappa}}(t)\rho^+_{\vec{\kappa}}(0) >_T = < \rho^+_{\vec{\kappa}}(0)\rho_{\vec{\kappa}}(t) >_T \tag{122}$$

That (122) is not valid can be demonstrated by an analytical extension to imaginary time.

$$\begin{aligned} < \rho_{\vec{\kappa}}(t)\rho^+_{\vec{\kappa}}(0) >_T &= \frac{1}{Z} Sp\left[e^{-\beta H} e^{i/\hbar H t} \rho_{\vec{\kappa}} e^{-i/\hbar H t} \rho^+_{\vec{\kappa}}\right] \\ &= \frac{1}{Z} Sp\left[e^{-\beta H} \rho^+_{\kappa} e^{-\beta H + i/\hbar H t} \rho_{\vec{\kappa}} e^{\beta H - i/\hbar H t}\right] \\ &= \frac{1}{Z} Sp\left[e^{-\beta H} \rho^+_{\vec{\kappa}} \rho_{\kappa}(t + i\hbar\beta)\right] \\ &= < \rho^+_{\vec{\kappa}}(0)\rho_{\vec{\kappa}}(t + i\hbar\beta) >_T \,. \end{aligned} \tag{123}$$

As a reminder - we have made use of cyclic exchange of noncommuting operators, which is allowed under the trace operation. We recognize from (123) that (122) becomes valid in the limit $\hbar \to 0$. Hence, the classical correlation function is real, and $ImC \neq 0$ is a quantum mechanical effect. Particularly for fluids (however not "quantum" fluids like liquid He) the classical, real correlation function is normally sufficient for the description of its structure. The same is obviously true for gaseous systems.

The correlation functions for short time $\tau \longrightarrow 0$ in liquids and gases are

$$C_s(\vec{x}, 0) = \delta(\vec{x}) \tag{124}$$

$$C(\vec{x}, 0) = \delta(\vec{x}) + \sum_{i \neq 0} < \delta(\vec{x} + \vec{X}_0(0) - X_i(0)) >_T \tag{125}$$

whereas for long times $\tau \longrightarrow \infty$

$$C_s(\vec{x}, \infty) = 0 \tag{126}$$

$$C(\vec{x}, \infty) = \rho \tag{127}$$

where ρ is the bulk density of the sample. The long range spatial correlations for short time are

$$C_s(\infty, 0) = 0 \tag{128}$$

$$C(\infty, 0) = \rho \tag{129}$$

(126) and (127) are a consequence of random motion of the scattering centers which smooth out any correlation. Although (128) and (129) is also true for glasses, this is not the case for (126) and (127). For solids short- and long-ranged correlations remain, even for long times. In a crystal they are given by the lattice and somewhat smoothed out at higher temperature by thermal motion (Debye-Waller factor as a form factor).

4 Linear Response

In this chapter we investigate the scattering process of a probe on the target sample from the point of view of a perturbative response. The incident particle acts to the target as a perturbation to which the latter responds. The response function - a generalized susceptibility - is related to the correlation function discussed in the last chapter and contains the information about the scatterer.

4.1 The perturbative interaction

A scattering process corresponds to a change in the condition of the beam radiation incident on the sample to a possibly different condition of the secondary beam, e.g. the incident beam is deflected by the scattering event. The change in condition of the beam is related to changes induced in the sample, which is the information to be extracted.

We summarize the perturbative part of the hamiltonians causing the scattering process. Generally the perturbative hamiltonian has the form

$$H_1(t) = B(t) \cdot h(t) \tag{130}$$

where $h(t)$ describes the perturbing external field induced by the probe. $B(t)$ expresses the coupling of this external field to the dynamical degrees of freedom of the sample.

We assume that for $t \longrightarrow -\infty$ the sample is undisturbed and is described by a hamiltonian H_0. Any time dependent perturbation is in its Fourier transform described by a set of monochromatic components and is hence proportional to $e^{i\omega t}$. In its explicite expression H_1 is switched on adiabatically, that is the sample stays always in thermal equilibrium. This can be accomplished by the choice

$$h(t) \sim e^{i\omega t + \epsilon t}; \qquad \epsilon \longrightarrow 0^+ \tag{131}$$

Normally H_1 contains the sum of couplings to individual particles in the system, usually expressed by a space-integration. Guided by the representation of the various interactions we therefore write for H_1

i) electromagnetic interaction (e.g. X-rays)

$$H_1 = \frac{e}{c} \int d^3x \vec{A}(\vec{x}, t) \vec{j}(\vec{x}) \tag{132}$$

with the current densities of the charge carriers

$$\vec{j}(\vec{x}) = \sum_i \frac{1}{2m} (\vec{p}_j \delta(\vec{x} - \vec{x}_j) + \delta(\vec{x} - \vec{x}_j) \vec{p}_j)$$

and $\vec{A}(\vec{x}, t)$ the electromagnetic potential of the incident field and m is the mass of an electron with change $-e$.

ii) magnetic interaction of neutrons

$$H_1 = \int d^3x \vec{A}(\vec{x}, t) \cdot \vec{m}(\vec{x}) \tag{133}$$

with the magnetization density either given by magnetic moments of the electrons in the sample

$$\vec{m} = 2\mu_B \sum_j \int d^3x \vec{\sigma}_e \delta(\vec{x} - \vec{x}_j) \tag{134}$$

or by their orbital currents

$$\vec{M} = \frac{e}{m} \sum_j \int d^3x (\vec{p}_j \delta(\vec{x} - \vec{x}_j) + \delta(\vec{x} - \vec{x}_j) \vec{p}_j) \tag{135}$$

$\vec{A}(\vec{x}, t)$ is here the electromagnetic potential due to the magnetic moment of the neutron

$$\vec{A}(\vec{x}, t) = \mu_N \vec{\sigma}_N \wedge \vec{\nabla} \frac{1}{\mid \vec{x} \mid} \tag{136}$$

iii) nuclear interaction with neutrons

$$H_1 = \int d^3x e^{i(\omega t - \vec{\kappa}\vec{x})} \rho(\vec{x}) \tag{137}$$

with a weighted nuclear scattering density

$$\rho(\vec{x}) = \sum_j b_j \delta(\vec{x} - \vec{x}_j) \tag{138}$$

4.2 The Dynamics of the Density Operator

We define the hamiltonian, split into two parts

H_0 this part describes the unperturbed sample to be investigated

$H_1(t)$ describes the time-dependent perturbation produced by an external probe interacting with the sample.

Hence the composite system of the radiation and sample is describe on the Hamiltonian,

$$H = H_0 + H_1(t) \tag{139}$$

The expectation value of an operator $\hat{O}$ of the system is

$$< \hat{O} >= Sp(\hat{O}\rho(t)) \tag{140}$$

where $\rho(t)$ is the density operator of the system, governed by the equation of motion

$$i\hbar\partial_t\rho = [H, \rho]. \tag{141}$$

We now work in the interaction-picture (Dirac) where the time development of operators is governed by the unperturbed hamiltonian H_0

$$\hat{O}(t) = e^{(i/\hbar)H_0 t}\hat{O}e^{-(i/\hbar)H_0 t}. \tag{142}$$

The dynamics of the system is then described by

$$i\hbar\partial_t\rho_I = [H_1^I(t), \rho_I(t)] \tag{143}$$

with

$$H_1^I(t) = e^{(i/\hbar)H_0 t}H_1(t)e^{-(i/\hbar)H_0 t} \tag{144}$$

A formal solution of (143) can be represented by the series

$$\rho_I(t) = \rho(0) - \frac{i}{\hbar}\int_{-\infty}^{t} d\tau[H_1^I(\tau), \rho_0] - \frac{1}{\hbar^2}\int_{-\infty}^{t} d\tau \int_{-\infty}^{\tau} d\tau'[H_1^I(\tau'), [H_1^I(\tau), \rho_0]] + ... \tag{145}$$

If we are allowed to neglect the higher order terms in (145) we can express the expectation value of any observable by means of (140) as

$$< \hat{O} >= Sp(O_I(t)\rho_0) - \frac{i}{\hbar}\int\limits_{-\infty}^{t} d\tau Sp(O_I(t)[H_1^I(\tau), \rho_0]) \tag{146}$$

where ρ_0 is the equilibrium density operator. The traces in (146) give the average over the canonical ensemble of the enclosed quantities and we may write for the expected response of the observable

$$\boxed{< \delta\hat{O} >_T =< \hat{O}. - Sp(O_I(t)\rho_0) = -\frac{i}{\hbar}\int\limits_{-\infty}^{t} d\tau < [\hat{O}_I(t), H_1^I(\tau)] >_T} \tag{147}$$

We now introduce the perturbation hamiltonian in its general form. For elegance and convenience - after all, electromagnetic fields are relativistic objects - we chose the relativistic covariant form (see appendix A).

$$H_1^I(\tau) = \int d^3x J_\mu(x)\mathcal{A}^\mu(x) \tag{148}$$

where A^μ is the four-vector potential of the external field. Using (148) we obtain for the response of the current operator according to (147)

$$\begin{aligned} < \delta J_\beta >_T &= -\frac{i}{\hbar}\int\limits_{-\infty}^{t} d\tau < [J_p^+(\vec{x}, t)\int d^3x' J_\mu(x')\mathcal{A}^\mu(x')] >_T \\ &= -\frac{i}{\hbar}\int d^4x' \theta(x_0 - x_0') < [J_\beta(x'), J_\mu(x')] >_T \mathcal{A}^\mu(x') \end{aligned} \tag{149}$$

We write this in the following form

$$\boxed{< \delta J_\beta >_T = \int d^4x' K_{\beta\mu}(x - x')\mathcal{A}^\mu(x')} \tag{150}$$

with a kernel

$$\boxed{K_{\beta\mu}(x - x') = -\frac{i}{\hbar}\theta(x_0 - x_0') < [J_\beta^+(x), J_\mu(x')] >_T} \tag{151}$$

The tensor K admits an interpretation as a generalized susceptibility. In the following it will be one of our tasks to establish the relationship of K with the scattering function or the correlation function.

From now on we call K the response function or the response kernel. (150) and (151) may serve as a starting point for a field theoretical approach for treatment of many-body problems since K is closely related to the polarization tensor and to the field-theoretical four-point functions.

Before proceeding along that line, let us however first discuss the tensor character of K for the electromagnetic case and its relationship to phenomenological electrodynamics.

4.3 The Tensor Character of $K_{\mu\nu}$

We are interested here in the general tensor properties of K in a homogeneous and isotropic medium.

Let $\vec{k}$ be the wave vector of an electromagnetic excitation in this medium. We now choose three basic orthogonal unit vectors $e_\mu^{(\lambda)}$ (λ = 1,2,3) in the following way

$$\lambda = 1, 2 \qquad e_\mu^{(T)}(k, \lambda) \qquad \text{orthogonal to } \vec{k}$$

and

$$\lambda = 3 \qquad e_\mu^{(L)}(k, \lambda) \qquad \text{parallel to } \vec{k}$$

where k is the four-vector $(\omega, \vec{k})$ with the property

$$k^2 = \omega^2 - \vec{k}^{\,2} \tag{152}$$

($k^2 = 0$ in vacuum only).

For the longitudinal vector we write

$$e_\mu^{(L)}(\vec{k}, \omega) = \frac{1}{\sqrt{k^2}} \left(\mid \vec{k} \mid, \frac{\omega \vec{k}}{\mid \vec{k} \mid} \right) \tag{153}$$

which leads to the longitudinal projection operator

$$P^{(L)}_{\mu\nu} = e^{(L)}_{\mu} \cdot e^{(L)}_{\nu} = \begin{cases} \dfrac{\mid \vec{k} \mid^2}{k^2} & \text{for} \quad \mu = \nu = 0 \\ \dfrac{\omega}{k^2} k_j & \text{for} \quad \mu \quad \text{or} \quad \nu = 0 \\ \dfrac{\omega^2}{k^2} \dfrac{k_i k_j}{\mid \vec{k} \mid^2} & \text{otherwise} \end{cases} \tag{154}$$

For the transvers projection operator we obtain

$$P^{(T)}_{\mu\nu} = \sum_{\lambda=1,2} e^{(T)}_{\mu}(k,\lambda) e^{(T)}_{\nu}(k,\lambda) = \begin{cases} 0 \quad \text{for} \quad \mu = \nu = 0 \\ 0 \quad \text{for} \quad \mu \quad \text{or} \quad \nu = 0 \\ \delta_{ij} - \dfrac{k_i k_j}{\mid \vec{k} \mid^2} \quad \text{otherwise} \end{cases} \tag{155}$$

Both operators are obviously projections and fulfill

$$\{P^{(L)}\}^2 = P^{(L)}; \qquad \{P^{(T)}\}^2 = P^{(T)} \tag{156}$$

and furthermore

$$P^{(L)} P^{(T)} = 0. \tag{157}$$

Hence, in a homogeneous and isotropic medium there exist two independent tensor components - a transvers and a longitudinal ones. Note that for real photons ($k^2 = 0$) $P^{(L)}$ is not defined.

Our response tensor can now be written in the following form

$$K_{00} = K^{(L)} \cdot \frac{\mid \vec{k} \mid^2}{k^2} \tag{158a}$$

$$K_{0j} = K^{(L)} \cdot \frac{\omega}{k^2} \cdot k_j \tag{158b}$$

$$K_{ij} = K^{(L)} \cdot \frac{k_i k_j}{\mid \vec{k} \mid^2} \frac{\omega^2}{k^2} + K^{(T)} \left(\delta_{ij} - \frac{k_i k_j}{\mid \vec{k} \mid^2} \right) \tag{158c}$$

In order to gain some insight to the significance of (150) we express the zero-zero component for the longitudinal dielectric response

$$\delta < \rho >= -\frac{i}{\hbar} \int d^3x' \int dt' \theta(t-t') < [\rho^+(\vec{x},t), \rho(\vec{x}\,',t')] > \varphi_{ext}(\vec{x}\,',t') \tag{159}$$

where φ_{ext} is the external scalar potential of $\mathcal{A} = (\varphi, \vec{A})$. $\delta < \rho >$ is the source term for the internal induced potential

$$\Delta\varphi_{int} = -4\pi\delta < \rho > \tag{160}$$

or time Fourier-transformed

$$\varphi_{int}(\vec{k},\omega) = \frac{4\pi}{k^2}\delta < \rho(\vec{k},\omega) > . \tag{161}$$

With the phenomenological equations of macroscopic electrodynamics

$$\begin{gathered} \vec{D} = \epsilon\vec{E}, \quad \vec{E} = -\vec{\nabla}\varphi_{int} \\ \vec{D} = -\vec{\nabla}\varphi_{ext} \end{gathered} \tag{162}$$

and considering

$$\varphi_{tot} = \varphi_{ext} + \varphi_{int} \tag{163}$$

we obtain for the longitudinal part of the dielectric function

$$\frac{1}{\epsilon^{(L)}(\vec{k},\omega)} = \frac{\varphi_{tot}}{\varphi_{ext}} = 1 - \frac{4\pi\delta < \rho >}{\varphi_{ext}} = 1 - \frac{4\pi}{k^2}\chi(\vec{k},\omega) \tag{164}$$

where the susceptibility is defined, analogous to (150) and in particular (159) by

$$\delta < \rho(\vec{k},\omega) >= \chi(\vec{k},\omega)\varphi_{ext}(\vec{k},\omega) \tag{165}$$

$\epsilon^{(L)}(\vec{k},\omega)$ describes the response of the charge carriers in the system to an impurity charge. In particular, the screening mechanism to a static test charge is determined by $\epsilon^L(\vec{k},0)$ or $\chi(\vec{k},0)$ for small k.

From the experimental point of view a possible route to $\epsilon^{(L)}(\vec{k},\omega)$ (actually its imaginary part) is the measurement of the stopping power for a charged particle in the system.

For the case of coherent or incoherent nuclear neutron scattering as perturbative action (150) is a scalar equation. The relevant operator is given by the nuclear scattering density

$$B(\vec{x},t) = \sum_j b_j \delta(\vec{x} - \vec{X}_j(t)) \tag{166}$$

or its spatial Fourier transform

$$\rho_{\vec{\kappa}}(t) = \sum_j e^{-i\vec{\kappa}\vec{X}_j(t)} \tag{167}$$

The generalized "susceptibility" is then given by

$$-\frac{i}{\hbar}\theta(t) < [\rho_\kappa(t), \rho_\kappa^+(0)] >_T = -\frac{i}{\hbar}\theta(t) \sum_{i,j} < [e^{i\vec{\kappa}\vec{X}_j(t)}, e^{-i\vec{\kappa}\vec{X}_i(0)}] >_T \tag{168}$$

as a response function for a coherent perturbation. The corresponding expression for incoherent perturbation is

$$-\frac{i}{\hbar}\theta(t) \sum_j < [e^{-i\vec{\kappa}\vec{X}_j(t)}, e^{+i\vec{\kappa}\vec{X}_j(0)}] >_T \tag{169}$$

4.4 Response Kernel and Scattering Function

We shall assume translational invariance in both space and time coordinates and simplify (151)

$$K_{\alpha\beta}(x) = -\frac{i}{\hbar}\theta(x_0) < [J_\alpha^+(x), J_\beta(0)] >_T . \tag{170}$$

We are interested in the Fourier transform of $K_{\alpha\beta}$

$$\tilde{K}_{\alpha\beta}(k) = \tilde{K}_{\alpha\beta}(\omega,\vec{k}) = \left(\frac{\hbar}{2\pi}\right)^2 \int d^4x e^{ikx} K_{\alpha\beta}(x). \tag{171}$$

In order to prepare $K_{\alpha\beta}$ so we can evaluate this transform we first reformulate the second term of the commutator in (170),

$$J_a^+(x)J_\beta(0) - J_\beta(0)J_\alpha^+(x) \tag{172}$$

We use the translation invariance in time and space

$$J_\alpha^+(x) = e^{(i/\hbar)px} J_\alpha^+(0) e^{-(i/\hbar)px} \tag{173}$$

and the fact that cyclic interchange of operators is allowed under the trace operation. We can then write

$$Sp(e^{-\beta H} J_\beta(0) e^{(i/\hbar)Ht} J_\alpha^+(0,\vec{x}) e^{-(i/\hbar)Ht}) = Sp(e^{-\beta H} J_\alpha^+(t - i\hbar\beta, \vec{x}) J_\beta(0)) \tag{174}$$

extending the expression analytically into the domain of complex time coordinate. If we now use the spatial part of translation symmetry of (173), as well and take matrix elements in a complete set of eigenstates of H, we obtain for $K(x)$.

$$\begin{aligned} &K_{\alpha\beta}(x) = \\ &-\tfrac{i}{\hbar}\theta(x_0)\textstyle\sum_{n,m} e^{-\beta(\hbar\omega_n - F)}[< n \mid J_\alpha^+(t,0) \mid m >< m \mid J_\beta(0) \mid n > - \\ &- < n \mid J_\alpha^+(t - i\hbar\beta, 0) \mid m >< m \mid J_\beta(0) \mid n > e^{-i(\vec{k}_n - \vec{k}_m)\vec{x}} \end{aligned} \tag{175}$$

Let us now introduce an integral-representation of the step function $\theta(x_0)$ (B.1)

$$\theta(x_0) = -\frac{1}{2\pi i}\int_{-\infty}^{+\infty} dp_0 \frac{e^{ip_0 x_0}}{p_0 + i\eta} = -\frac{1}{2\pi i}\int_{-\infty}^{+\infty} d\omega' \frac{e^{-i\omega'\cdot t}}{\omega' + i\epsilon} \tag{176}$$

and take the Fourier transform in space and time. The transformed kernel becomes then

$$\begin{aligned} &\tilde{K}_{\alpha\beta}(k) = \tilde{K}_{\alpha\beta}(\omega, \vec{k}) = \\ &\frac{1}{2\pi\hbar}\left(\frac{1}{2\pi}\right)^2 \int d^3x e^{i(\vec{k} + \vec{k}_n - \vec{k}_m)\vec{x}} \int_{-\infty}^{+\infty} d\omega' \int_{-\infty}^{+\infty} dt \frac{e^{i(\omega - \omega')t}}{\omega' + i\epsilon} \sum_{n,m} e^{-\beta(\hbar\omega_n - F)}[\quad] \end{aligned} \tag{177}$$

with

$$[\ ] =< n \mid J_\alpha^+(t,0) \mid m >< m \mid J_\beta(0) \mid n > - < n \mid J_\alpha^+(t - i\hbar\beta, 0) \mid m >< m \mid J_\beta(0) \mid n > \quad (178)$$

By means of (173) we take the t-dependence out of the matrix elements of (178) and obtain

$$[\quad] = e^{i(\omega_n - \omega_m)t}[1 - e^{\hbar\beta(\omega_n - \omega_m)}] < n \mid J_\alpha^+(0) \mid m >< m \mid J_\beta(0) \mid n > \quad (179)$$

With (179) we can now write for $\tilde{K}$ - after a trivial $\vec{x}$-integration

$$\tilde{K}_{\alpha\beta}(\omega, \vec{k}) =$$

$$\frac{1}{\hbar} \sum_{n,m} e^{-\beta(\hbar\omega_n - F)} \delta^{(3)}(\vec{k} + \vec{k}_n - \vec{k}_m) \int_{-\infty}^{+\infty} d\omega' \int_{-\infty}^{+\infty} dt \frac{e^{i(\omega - \omega' + \omega_n - \omega_m)t}}{\omega' + i\epsilon} \quad (180)$$

$$\cdot[1 - e^{\hbar\beta(\omega_n - \omega_m)}] < n \mid J_\alpha^+(0) \mid m >< m \mid J_\beta(0) \mid n >$$

With the substitution $\omega' \longrightarrow \omega - \omega'$ we get for the ω'-integral

$$-\int_{-\infty}^{+\infty} d\omega' \frac{e^{i(\omega' + \omega_n - \omega_m)t}}{(\omega - \omega') + i\epsilon} = -P \int_{-\infty}^{+\infty} d\omega' \frac{e^{i(\omega' + \omega_n - \omega_m)t}}{\omega - \omega'} + i\pi \int_{-\infty}^{+\infty} d\omega' \delta(\omega - \omega') e^{i(\omega' + \omega_n - \omega_m)t} \quad (181)$$

where we have made use of (B.8). P stands for the principal value of the integral. (181) separates $\tilde{K}_{\alpha\beta}$ into its real and imaginary part. We now concentrate on to the imaginary part of $\tilde{K}_{\alpha\beta}$. Using the second integral of (181) we obtain from (180)

$$\mathcal{I}m\tilde{K}_{\alpha\beta} =$$

$$\frac{2\pi^2}{\hbar} \sum_{n,m} \delta^{(3)}(\vec{k} + \vec{k}_n - \vec{k}_m) \delta(\omega + \omega_n - \omega_m) e^{-\beta(\hbar\omega_n - F)} \quad (182)$$

$$\cdot < n \mid J_\alpha^+(0) \mid m >< m \mid J_\beta(0) \mid n > (1 - e^{-\hbar\beta\omega})$$

With the help of the integral representation (B.3) of the δ-functions and the translation relation (173) we transform the kinematical variables back into the current matrix elements. This leads to

$$\mathcal{I}m\tilde{K}_{\alpha\beta} =$$

$$\left(\frac{1}{2\pi}\right)^2 \frac{1}{2\hbar} \cdot \sum_n e^{-\beta(\hbar\omega_n - F)} \cdot < n \mid \tilde{J}_\alpha^+(\omega, \vec{k}) \tilde{J}_\beta(0) \mid n > (1 - e^{-\hbar\beta\omega}) \quad (183)$$

Apart from the factor $(1-e^{-\hbar\beta\omega})$ (183) has the same structure like the scattering function $S(\vec{k},\omega)$.

With $n(\omega)$ as statistical population of states at temperature $T(\beta = 1/k_B T)$

$$n(\omega) = (e^{\beta\hbar\omega} - 1)^{-1} \tag{184}$$

we can write for the factor in (183)

$$(1 - e^{-\beta\hbar\omega}) = [1 + n(\omega)]^{-1} \tag{185}$$

Since $\tilde{K}_{\alpha\beta}$ has the tensor structure of (158), the imaginary parts of $\tilde{K}^T$ and $\tilde{K}^L$ are according to (183) intrinsically related to the scattering amplitude $S(\vec{k},\omega)$.

$$\boxed{4\pi[1 + n(\omega)]\mathcal{I}m\tilde{K}(\omega,\vec{k}) = S(\vec{k},\omega)}. \tag{186}$$

4.5 Kramers-Kronig relations / Lehmann representation

We have seen that the Fourier transformed expressions of

$$-\frac{i}{\hbar}\Theta(x_0) < [J_\alpha^+(0), J_\rho(x)] >_T \qquad \text{or} \qquad -\frac{i}{\hbar}\Theta(\tau) < [\rho_\kappa(0), \rho_\kappa^+(\tau)] >_T$$

correspond to generalized susceptibilities, or more precisely to their imaginary part. According to (186) these (response) functions are, apart from a thermal detailed-balance factor, proportional to the scattering function $S(\omega,\vec{\kappa})$ and are therefore directly measured by the differential cross section for inelastic scattering. Our aim is now to find the real part of the generalized susceptibility, which is connected to the imaginary part by a causality condition on the scattering process. In order to establish this connection we must first verify a few analytic properties of the response function.

With (183) we can write

$$\phi_\kappa(\tau) = (-i)\int_{-\infty}^{-\infty} d\omega e^{i\omega\tau}[1 - e^{-\hbar\omega\beta}]S(\vec{\kappa},\omega) \tag{187}$$

The response function is then given by

$$\tilde{K}^{(R)}(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} d\tau e^{-i\omega\tau} \Theta(\tau) \phi_\kappa(\tau) \tag{188}$$

$S(\vec{\kappa}, \omega)$ has (as a cross section) the following properties

i) $S(\vec{\kappa}, \omega) \geq 0$ since it is proportional to a cross section

ii) $S(\vec{\kappa}, \omega) = e^{\hbar\omega\beta} S(-\vec{\kappa}, -\omega)$, so-called condition of detailed balance.

iii) $\exists \omega_{max} : S(\omega) = 0$ for $\forall \omega > \omega_{max}$ due to a minimal interatomic distance.

Using these properties and applying them to (187) we obtain the following behaviour for small times

$$\phi_\kappa(\tau) \xrightarrow[\tau \longrightarrow 0]{} > 0 \tag{189}$$

$$\partial_\tau \phi_\kappa(\tau)_{\tau=0} = const \geq 0 \tag{190}$$

$\tilde{K}^{(R)}(\omega)$ is the Laplace transform of $\phi_\kappa(\tau)$ due to the presence of $\Theta(\tau)$ in the integrand of (188), which takes care of the causal property of the response function.

We extend $\tilde{K}^{(R)}(\omega)$ into the complex ω-plane and write[1]

$$\tilde{K}^{(R)}(p) = \int_0^\infty d\tau e^{-p\tau} \phi/(\tau) \tag{191}$$

with

$$p = \sigma + i\omega$$

(188) and (189) determine the behaviour for large ω of $K^{(R)}(p)$ in the half-plane with $\sigma > 0$. Using one of the Abelian theorems [4)] we obtain

$$\tilde{K}^{(R)}(p) \sim O(1/p^2) \tag{192}$$

[1]Since we are in the following interested only in the conjugate variables ω and τ we suppress the variable (index) $\vec{\kappa}$

for large $p's$ in any direction of the above mentioned half-plane.

(117) and (118) imply

$$\phi^*(\tau) = -\phi(-\tau) \tag{193}$$

that is

$$Re\phi(\tau) = -Re\phi(-\tau) \tag{194a}$$

$$\mathcal{I}m\phi(\tau) = \mathcal{I}m\phi(-\tau) \tag{194b}$$

With

$$\tilde{\phi}(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} d\tau e^{-i\omega\tau}\phi(\tau) \tag{195}$$

we get the symmetry relation for $\tilde{\phi}$

$$\tilde{\phi}^*(\omega) = -\tilde{\phi}(\omega^*) \tag{196}$$

$$Re\phi(\omega) = Re\phi(-\omega^*) \tag{197a}$$

$$\mathcal{I}m\phi(\omega) = \mathcal{I}m\phi(-\omega^*) \tag{197b}$$

which means that $\tilde{\phi}(\omega)$ is purely imaginary for real ω.

This can also be put into evidence by the relation

$$\begin{aligned} \tilde{\phi}(\omega) &= \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} d\tau e^{-i\omega\tau}\phi(\tau) = \sqrt{\frac{2}{\pi}} i\mathcal{I}m \int_{0}^{\infty} d\tau e^{-i\omega\tau}\phi(\tau) \\ &= \sqrt{\frac{2}{\pi}} i\mathcal{I}m \int_{-\infty}^{+\infty} d\tau e^{-i\omega\tau}\Theta(\tau)\phi(\tau) = \sqrt{\frac{2}{\pi}} i\mathcal{I}m\tilde{K}^R(\omega) \end{aligned} \tag{198}$$

For the expressions of $\tilde{\phi}$ and $\tilde{K}$ as Laplace transforms we have to rotate the complex plane by the angle $\pi/2$ and the symmetry relation (196) becomes

$$\tilde{\phi}^*(p) = -\tilde{\phi}(p^*) \tag{199}$$

$$Re\tilde{\phi}(p) = Re\tilde{\phi}(p^*) \tag{200}$$

$$\mathcal{I}m\tilde{\phi}(p) = \mathcal{I}m\tilde{\phi}(p^*) \tag{201}$$

hence $\mathcal{I}m\tilde{\phi}(p)$ vanishes on the real p-axis and $\tilde{\phi}(p)$ is then purely real.

We now show that

$$\tilde{K}(p) = \int_0^\infty d\tau e^{-p\tau}\phi(\tau) \tag{202}$$

decreases monotonically on the real p-axis in the inteval $(0,\infty)$[5].

The properties of $\phi(\tau)$ and (202) guarantee the holomorphy (analyticity) of $\tilde{K}(p)$ in the half-plane $\sigma > 0$. Let us now consider the function

$$\tilde{K}(p) - a = f(p) \tag{203}$$

where a is a real constant. $f(p)$ is obviously also holomorphic and we therefore apply the following theorem for analytic functions:

$$\frac{1}{2\pi i}\oint_C \frac{f'(p)}{f(p)}dp = N - P = \frac{1}{2\pi i}\oint_C \frac{d\tilde{K}(p)}{dp}\frac{dp}{\tilde{K}(p) - a} \tag{204}$$

where N is the number of zeros and P the number of poles of $f(p)$ within C, the closed integration circuit along the imaginary axis and closing on a circle C_∞ around the left half-plane. Mapping (204) onto the complex K-plane we can write

$$\frac{1}{2\pi i}\oint_{C'} \frac{d\tilde{K}^R}{\tilde{K}^R - a} = N \tag{205}$$

C' is now a path which cuts the real K-axis exactly twice - namely at

$\tilde{K} = (0,0)$ which is the picture of C_∞ of the p-plane

and

$\tilde{K} = (K_0, 0)$ which is the picture of (0,0) of the p-plane.

In between C' might be arbitrarily complicated, (even curling up) but never cuts the real K-axis again. Hence C' circumvents $a \epsilon Re$ either once or not at all. For the integral in (205) we write

$$\oint_C \frac{d\tilde{K}}{\tilde{K}-a} = ln|\tilde{K}-a| \mid_{C'} = ln|\tilde{K}-a| + i\ arg(\tilde{K}-a) \tag{206}$$

where for a closed path C'

$$\begin{aligned} ln|\tilde{K}-a| &= 0 \\ i\ arg(\tilde{K}-a) &= 2\pi i. \end{aligned}$$

Hence the integral (205) has the values

$$N = \begin{cases} 1 & \text{for } 0 < a < K_0 \\ 0 & \text{otherwise} \end{cases} \tag{207}$$

This shows that $\tilde{K}(p)$ takes on the real axis any value between K_0 (which might be ∞) and zero once and only once; hence $\tilde{K}(p)$ decreases monotonically.

Due to the holomorphy of $\tilde{K}(p)$ in the right half-plane ($\sigma \geq 0$) we can write for a closed path

$$\oint_C \frac{\tilde{K}^R(p)}{p-p_0} dp = 2\pi i \tilde{K}(p_0) \tag{208}$$

where C consists of a part along the complex axis $(-i\infty, +i\infty)$ and closes along a circle at infinity in the positive half-plane. Following a procedure analogous to (B5 - B10) gives

$$P \int \frac{\tilde{K}^R(p)}{p-p_0} dp = i\pi \tilde{K}(p_0) \tag{209}$$

for p_0 on the imaginary axis. P means the principal value at p_0. Transforming back to the coordinate system with the variable ω along the real axis (209) becomes equivalent to

$$\boxed{\begin{aligned} -\frac{1}{\pi}P\int \frac{Re\tilde{K}^R(\omega)}{\omega-\omega_0}d\omega &= \mathcal{I}m\tilde{K}^R(\omega_0) \qquad (210a)\\ \frac{1}{\pi}P\int \frac{\mathcal{I}m\tilde{K}(\omega)}{\omega-\omega_0}d\omega &= Re\tilde{K}^R(\omega_0) \qquad (210b)\end{aligned}}$$

These relations - called Kramers-Kronig - or dispersion relations are a consequence of the causal behaviour of the response functions (no output before input), which leads to the holomorphy of $\tilde{K}^R$ in the upper half-plane. On the other half plane $\tilde{K}^R$ contains usually singularities and its behaviour at infinity is not regular. These relations show that the whole response function $\tilde{K}^R(\omega)$ is determined by the scattering experiment in spite of the relationship (186) which connects only the imaginary part of $\tilde{K}^R$ with the scattering function $S(\vec{\kappa},\omega)$. This is a simple consequence of the causal relation of the applied perturbation on the response observed.

From the dispersion relation and (183) we can find a general representation of the retarded response function $\tilde{K}^R(\omega)$

$$\boxed{\tilde{K}^R(\omega)=\frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{d\omega'\rho(\omega')}{(\omega'-\omega)-i\epsilon}} \qquad (211)$$

with the spectral function $\rho(\omega)$

$$\rho(\omega)=\sum_{n,m}e^{-\beta(\hbar\omega_n-F)}A_{nm}\delta(\omega-\omega_{mn})(1-e^{\beta\hbar\omega_{mn}}) \qquad (212)$$

with

$$\omega_{mn}=\omega_m-\omega_n \qquad (213)$$

and

$$A_{nm}=\begin{cases} <n|\rho_{\vec{\kappa}}|m><m|\rho^+_{\vec{\kappa}}|n> \\ \\ <n|J_\alpha(\vec{\kappa})|m><m|J^+_\beta(\vec{\kappa})|n> \end{cases} \qquad (214)$$

where the first expression is for neutron - the second for light scattering. (211) is called the Lehmann representation.

Let us now look a bit closer to the spectral function $\rho(\omega)$.

The term

$$e^{-\beta(\hbar\omega_n - F)}$$

is the population probability P_n of the state $|n>$.

$$e^{-\beta(\hbar\omega_n - F)} e^{-\beta\hbar\omega_{mn}}$$

with (213) stands for the propability of populating state $|m>$. The product of matrix elements A_{nm} expresses the transition probability $w_{n\to m}$ between the states $|n>$ and $|m>$. Therefore, the sum

$$\sum_m (P_n - P_m) w_{n\to m} \tag{215}$$

admits an interpretation as the transition rate from state $|n>$ into any other (excited) state of the system during the scattering process. The total rate of energy dissipated by the sample is then given by the following master equation

$$\partial_t \dot{E} = \sum_{n,m} \hbar\omega_{nm} (P_n - P_m) w_{n\to m} \tag{216}$$

Considering (186) this relationship is intimitely related to the time deriviative at $\tau = 0$ of (187)

$$\int_{-\infty}^{+\infty} d\omega\omega (1 - e^{-\hbar\omega\beta}) S(\vec{\kappa}, \omega) = \partial_\tau \phi_x(\tau) \mid_{\tau=0} \tag{217}$$

with

$$\phi_\kappa(t) =< [\rho_\kappa(\tau), \rho_\kappa^+(0)] >_T$$

The relationship (216) shows that the spectral function $\rho(\omega)$ aquires its contributions from the excited states (dynamical degrees of freedom) of the system and hence from the variety of possibilities of energy transfer from the neutron to the sample.

Due to the detailed balance factor in (216) the range of integration contributing to the integral of (208) is restricted to the positive ω-axis for $T = 0(\beta \to \infty)$. For higher temperatures this range extends into the negative axis. Using (B8) it can be shown that $\tilde{K}^R(\omega)$ is holomorphic (regular) in the cut complex ω-plane - the cut being along the real axis, starting at some $\omega < 0$ which depends on the temperature of the sample. The discontinuouity between the lower and the upper bord of the cut is

$$\tilde{K}^R(\omega + i\epsilon) - \tilde{K}^R(\omega - i\epsilon) = 2i\rho(\omega) = 2i\mathcal{I}m\tilde{K}^R(\omega) \tag{218}$$

According to (212) the contribution to (218) are the residues of a sequence of poles at ω_{nm}.

A Appendix

For four-dimensioned covariant notation we choose the following convention. The metric tensor is given by the matrix

$$g^{\mu}_{\nu} = \begin{pmatrix} 1 & & & \\ & -1 & & \\ & & -1 & \\ & & & -1 \end{pmatrix} \tag{A.1}$$

and the scalar prodct is defined by

$$x \cdot x = \vec{x}_{\mu} x^{\mu} = g^{\mu}_{\nu} x_{\nu} x_{\mu}$$

in particular for a product of a four-momentum with a space-time vector

$$p_{\mu} x^{\mu} = px = p_0 x_0 - \vec{p} \cdot \vec{x} \tag{A.2}$$

where

$$x = (x_0, \vec{x}) = (ct, \vec{x}) \tag{A.3}$$

and

$$p = (p_0, \quad \vec{p}\,) = (E/c, \vec{p}) \tag{A.4}$$

The energy and the momentum for a mass less particle can be expressed in wave numbers

$$E = \hbar\omega, \quad \vec{p} = \hbar\vec{k} \tag{A.5}$$

such that

$$e^{i/\hbar(px)} = e^{i(\omega t - \vec{k}\vec{x})} \tag{A.6}$$

B Appendix

In this appendix we summarize some expressions on generalized functions, which are useful in the mathematical developments of scattering theory.

With theorem of residues the step function can be shown to have the following integral represenation

$$\theta(\tau) = \frac{-1}{2\pi i} \lim_{\epsilon \to 0} \int_{-\infty}^{+\infty} d\xi \frac{e^{i\xi\tau}}{\xi - i\epsilon} = \frac{1}{2\pi i} \lim_{\epsilon \to 0} \int_{-\infty}^{+\infty} d\xi \frac{e^{-i\xi\tau}}{\xi + i\epsilon} \tag{B.1}$$

The formal derivative of (B.1) leads to the δ-function

$$\theta'(\tau) = \frac{1}{2\pi i} \int_{-\infty}^{+\infty} d\xi \frac{i\xi}{\xi - i\epsilon} e^{i\xi\tau} \quad \xrightarrow{\epsilon \to 0} \quad \frac{1}{2\pi} \int_{-\infty}^{+\infty} e^{i\xi\tau} d\xi = \delta(\tau) \tag{B.2}$$

By means of (B.1) the δ-function can also be represented differently. Consider

$$\begin{aligned} \int_{0}^{+\infty} e^{ikx} dk &= \int_{-\infty}^{+\infty} \theta(k) e^{ikx} dk = \\ &= \frac{1}{2\pi i} \int_{-\infty}^{+\infty} d\xi \int_{-\infty}^{+\infty} dk \frac{e^{ik(\xi + x)}}{\xi - i\epsilon} = -i \int_{-\infty}^{+\infty} d\xi \frac{\delta(x + \xi)}{\xi - i\epsilon} \end{aligned}$$

and similarly

$$\int_{-\infty}^{0} e^{ikx} dk = -i \int_{-\infty}^{+\infty} d\xi \frac{\delta(x - \xi)}{\xi + i\epsilon}$$

Summing up gives

$$\frac{1}{2\pi} \int_{-\infty}^{+\infty} e^{ikx} dk = \delta(x) = \frac{1}{2\pi i} \left[\frac{1}{x - i\epsilon} - \frac{1}{x + i\epsilon} \right] \tag{B.3}$$

and therefore

$$\pi\delta(x) = \lim_{\epsilon \to 0} \frac{\epsilon}{x^2 + \epsilon^2} \tag{B.4}$$

Let $f(\zeta)$ be a regular function in the complex ζ-plane. We assume $f(\zeta)$ to decrease sufficiently fast for large arguments $\mid \zeta \mid \rightarrow \infty$. The value of the function f at a point ω within a certain region R is then given by the values on the boundary δR, that is

$$f(\omega) = \frac{1}{2\pi i} \oint \frac{f(\zeta)}{\zeta - \omega} d\zeta \tag{B.5}$$

If $\omega_1 \epsilon \mathcal{R}e$ and $f(\zeta) \rightarrow 0$ for $\mid \zeta \mid \rightarrow \infty$ we can write for (B.5)

$$\lim_{\omega \rightarrow \omega_1 + i\epsilon} f(\omega_1) = \frac{1}{2\pi i} \int_{-\infty}^{+\infty} \frac{f(\zeta)}{\zeta - \omega_1 - i\epsilon} d\zeta = \frac{1}{2\pi i} \lim_{\epsilon \rightarrow 0} \left[\int_{-\infty}^{\omega_1 - \epsilon} \frac{f(\zeta)}{\zeta - \omega_1} d\zeta + \int_{\omega_1 + \epsilon}^{\infty} \frac{f(\zeta)}{\zeta - \omega_1} d\zeta \right] \tag{B.6}$$

where the third term stems from the integration on the $(\epsilon \rightarrow 0)$ semi-circle round the pole on the real axis. The integral in bracket is by definition the Cauchy-principal value. Hence

$$\lim_{\omega \rightarrow \omega_1} 2\pi i f(\omega) = P \int_{\infty}^{+\infty} \frac{f(\zeta)}{\zeta - \omega_1} d\zeta + \pi f(\omega_1) \tag{B.7}$$

From (B.6) and (B.7) we can read off the formal relationship (Plemelij)

$$\lim_{\epsilon \rightarrow 0} \frac{1}{\zeta - \zeta_0 \pm i\epsilon} = P \frac{1}{\zeta - \zeta_0} \mp i\pi \delta(\zeta - \zeta_0) \tag{B.8}$$

Looking at (B.7) as

$$f(\omega) = \frac{1}{i\pi} P \int_{-\infty}^{+\infty} \frac{f(\zeta)}{\zeta - \omega} d\zeta \tag{B.9}$$

we obtain by separation of real and imaginary parts the relationships

$$\mathcal{R}ef(\omega) = \frac{1}{\pi} P \int \frac{\mathcal{I}mf(\zeta)}{\zeta - \omega} d\xi \tag{B.10a}$$

$$\mathcal{I}mf(\omega) = -\frac{1}{\pi} P \int \frac{Ref(\zeta)}{\zeta - \omega} d\zeta \tag{B.10b}$$

that is, a relation between real and imaginary part of the function f as a consequence of its analyticity properties.

We must emphasize that the derivations in this appendix should be considered as formal heuristic procedures at best - proper mathematical proofs need the concept of distributions.

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