

10.E.2. Causality

We now consider the effects of causality on the response matrix $\mathbb{K}(\tau)$, or equivalently but more conveniently, its Fourier transform $\chi(\omega)$.

To begin, consider a time-independent unit force

$$\mathbf{F}(t) = \hat{\mathbf{r}}$$

so that (10.113c) becomes

$$\alpha_{\mathbf{F}}(t) = \int_0^{\infty} d\tau \mathbb{K}(\tau) \cdot \hat{\mathbf{r}} \quad \forall t \quad (10.118a)$$

Thus, a time-independent force \mathbf{F} induces a time-independent deviation $\alpha_{\mathbf{F}}(t)$, which is expected to be finite from physical experience. Since $\hat{\mathbf{r}}$ can point in any direction, we conclude that

$$\int_0^{\infty} d\tau \mathbb{K}(\tau) \text{ must be finite.} \quad (10.118)$$

By definition [see (10.115b)],

$$\begin{aligned} \mathbb{K}(\tau) &= \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{-i\omega\tau} \chi(\omega) \\ &= \oint_C \frac{dz}{2\pi} e^{-iz\tau} \chi(z) \end{aligned} \quad (10.118b)$$

where $z = \omega + i\xi$ is the complex plane with ω & ξ as its real and imaginary axes, respectively. C is a contour consisting of closing the ω axis with a semi-circle at infinity that gives no contribution to the integration. Since

$$e^{-iz\tau} = e^{-i\omega\tau} e^{-\xi\tau}$$

this means C must close in the

$$\begin{aligned} &\text{lower-half } z\text{-plane, where } \xi \rightarrow -\infty, && \text{for } \tau > 0 \\ &\text{upper-half } z\text{-plane, where } \xi \rightarrow \infty, && \text{for } \tau < 0 \end{aligned}$$

The **causality condition** [see (10.114)]

$$\mathbb{K}(\tau) = 0 \quad \text{for } \tau < 0$$

can thus be achieved if $\chi(z)$ is analytic (has no poles) in the upper-half z -plane.

Note that $\chi(z)$ can also be considered as the extension of the Fourier transform of $\mathbb{K}(\tau)$ into the complex frequency domain:

$$\begin{aligned} \chi(z) &= \int_{-\infty}^{\infty} d\tau e^{iz\tau} \mathbb{K}(\tau) \\ &= \int_0^{\infty} d\tau e^{iz\tau} \mathbb{K}(\tau) \end{aligned} \quad (10.119)$$

As usual, the reality of $\mathbb{K}(\tau)$ means that its Fourier transform satisfies

$$\chi^*(\omega) = \chi(-\omega) \quad (10.118c)$$

$$\rightarrow \chi'(\omega) - i\chi''(\omega) = \chi'(-\omega) + i\chi''(-\omega)$$

$$\therefore \chi'(-\omega) = \chi'(\omega) \quad \chi''(-\omega) = -\chi''(\omega) \quad (10.118d)$$

Furthermore, (10.116) requires $\chi(\omega)$ to be finite since both $\langle \tilde{\alpha}(\omega) \rangle_{\mathbf{F}}$ & $\mathbf{F}(\omega)$ are.

Thus, for u real, the matrix

$$f(z) = \frac{\chi(z)}{z - u} \tag{10.120}$$

is analytic in the upper z -plane except for a simple pole on the real axis at $z = u$.

Let C_{\pm} be a contour obtained from C by replacing the infinitesimal segment through u with a semi-circle centered at u and lies ^{above}/_{below} the ω -axis. Since C_{\pm} is a counterclockwise contour that ^{excludes}/_{includes} the pole

at $z = u$, we have

$$\begin{aligned} \mathbb{I}_{\pm} &= \oint_{C_{\pm}} dz f(z) \\ &= \mathcal{P} \int_{-\infty}^{\infty} d\omega f(\omega) + \mathbb{J}_{\pm} = 2\pi i \left(\underset{z=u}{\overset{0}{\text{Res } f(z)}} \right) \end{aligned} \tag{10.120a}$$

where $\mathcal{P}f$ is the **Cauchy principal part** of the function f defined by

$$\mathcal{P}f(x) = \begin{cases} f(x) & x \neq 0 \\ \text{not defined} & x = 0 \end{cases} \tag{10.123a}$$

In other word, $\mathcal{P}f(x)$ is $f(x)$ with the point $x = 0$ removed. Thus,

$$\mathcal{P} \int_{-\infty}^{\infty} d\omega f(\omega) = \lim_{r \rightarrow 0} \left(\int_{-\infty}^{u-r} d\omega + \int_{u+r}^{\infty} d\omega \right) f(\omega) \tag{10.123}$$

\mathbb{J}_{\pm} is the line integral along the ^{upper}/_{lower} semi-circle centered at u :

$$\begin{aligned} \mathbb{J}_{+} &= \lim_{r \rightarrow 0} \int_{\pi}^0 d\phi i r e^{i\phi} f(u + r e^{i\phi}) && [z - u = r e^{i\phi} \rightarrow dz = i r e^{i\phi} d\phi] \\ &= - \lim_{r \rightarrow 0} \int_0^{\pi} d\phi i r e^{i\phi} f(u + r e^{i\phi}) \end{aligned} \tag{10.120b}$$

$$\mathbb{J}_{-} = \lim_{r \rightarrow 0} \int_{\pi}^{2\pi} d\phi i r e^{i\phi} f(u + r e^{i\phi}) \tag{10.120c}$$

Putting (10.120) into (10.120b-c) gives

$$\begin{aligned} \mathbb{J}_{+} &= - \lim_{r \rightarrow 0} \int_0^{\pi} d\phi i r e^{i\phi} \frac{\chi(u + r e^{i\phi})}{r e^{i\phi}} \\ &= -i \int_0^{\pi} d\phi \chi(u) = -i\pi \chi(u) \end{aligned}$$

and

$$\mathbb{J}_{-} = \lim_{r \rightarrow 0} \int_{\pi}^{2\pi} d\phi i r e^{i\phi} \frac{\chi(u + r e^{i\phi})}{r e^{i\phi}} = i\pi \chi(u)$$

Together with

$$\text{Res } f(z) \underset{z=u}{=} \text{Res } \frac{\chi(z)}{z - u} \underset{z=u}{=} \chi(u)$$

(10.120a) becomes

$$\begin{aligned} \mathcal{P} \int_{-\infty}^{\infty} d\omega \frac{\chi(\omega)}{\omega - u} + \begin{pmatrix} -i\pi \chi(u) \\ i\pi \chi(u) \end{pmatrix} &= \begin{cases} 0 \\ 2\pi i \text{Res } f(z) \end{cases} \\ \rightarrow \mathcal{P} \int_{-\infty}^{\infty} d\omega \frac{\chi(\omega)}{\omega - u} &= i\pi \chi(u) \end{aligned} \tag{10.124}$$

or

$$\chi(u) = \mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega}{i\pi} \frac{\chi(\omega)}{\omega - u} \quad (10.125)$$

Let $\chi'(\omega)$ & $\chi''(\omega)$ be the real & imaginary part of $\chi(\omega)$, respectively. Then

$$\chi(\omega) = \chi'(\omega) + i\chi''(\omega) \quad (10.126)$$

and (10.125) becomes

$$\chi'(u) + i\chi''(u) = \mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega}{i\pi} \frac{\chi'(\omega) + i\chi''(\omega)}{\omega - u}$$

Equating the real & imaginary parts separately gives

$$\chi'(u) = \mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \frac{\chi''(\omega)}{\omega - u} \quad (10.127)$$

and

$$\chi''(u) = -\mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \frac{\chi'(\omega)}{\omega - u} \quad (10.128)$$

which are known as the **Kramers-Kronig relations (KKR)**.

Since $\chi'(\omega)$ & $\chi''(\omega)$ are related by the KKR, it is necessary to have full knowledge of only one of them.

Before going further, we need to prove the identity

$$\lim_{\eta \rightarrow 0} \frac{1}{x \pm i\eta} = \mathcal{P} \frac{1}{x} \mp i\pi \delta(x) \quad (10.130)$$

as follows.

$$\begin{aligned} L.H.S. &= \lim_{\eta \rightarrow 0} \frac{x \mp i\eta}{x^2 + \eta^2} \\ &= \begin{cases} \frac{1}{x} & x \neq 0 \\ \lim_{\eta \rightarrow 0} \frac{\mp i\eta}{x^2 + \eta^2} = \mp i\pi \delta(x) & x = 0 \end{cases} \end{aligned}$$

where we have used the Lorentzian representation of the delta function

$$\delta(x) = \lim_{\eta \rightarrow 0} \frac{\eta}{\pi(x^2 + \eta^2)} \quad \text{with} \quad \int_{-\infty}^{\infty} dx \frac{\eta}{\pi(x^2 + \eta^2)} = 1 \quad (10.137)$$

$$\begin{aligned} R.H.S. &= \begin{cases} \frac{1}{x} & x \neq 0 \\ \mp i\pi \delta(x) & x = 0 \end{cases} \\ &= L.H.S. \end{aligned}$$

QED.

(* Normalization of Lorentzian *)

$$\text{Assuming } [\eta > 0, \int_{-\infty}^{\infty} \frac{\eta}{\pi(x^2 + \eta^2)} dx]$$

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We can re-write (10.127) as

$$\chi'(u) = \mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega'}{\pi} \frac{\chi''(\omega')}{\omega' - u} \quad [(u, \omega) \rightarrow (\omega, \omega')]]$$

$$\begin{aligned}
 &= \lim_{\eta \rightarrow 0} \int_{-\infty}^{\infty} \frac{d\omega'}{\pi} \left[\frac{1}{\omega' - \omega - i\eta} - i\pi \delta(\omega' - \omega) \right] \chi''(\omega') \quad [(10.130) \text{ used.}] \\
 &= \lim_{\eta \rightarrow 0} \int_{-\infty}^{\infty} \frac{d\omega'}{\pi} \frac{\chi''(\omega')}{\omega' - \omega - i\eta} - i\pi \chi''(\omega) \\
 \rightarrow \quad \chi(\omega) &= \lim_{\eta \rightarrow 0} \int_{-\infty}^{\infty} \frac{d\omega'}{\pi} \frac{\chi''(\omega')}{\omega' - \omega - i\eta} \quad (10.129)
 \end{aligned}$$

(10.115b) then gives

$$\langle \alpha(t) \rangle_F = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{-i\omega t} \langle \tilde{\alpha}(\omega) \rangle_F \quad (10.115)$$

$$= \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{-i\omega t} \chi(\omega) \cdot \tilde{F}(\omega) \quad (10.139)$$

$$= \lim_{\eta \rightarrow 0} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{-i\omega t} \int_{-\infty}^{\infty} \frac{d\omega'}{\pi} \frac{\chi''(\omega')}{\omega' - \omega - i\eta} \cdot \tilde{F}(\omega) \quad [(10.129) \text{ used.}]$$

$$= \lim_{\eta \rightarrow 0} \int_{-\infty}^{\infty} dt' \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{-i\omega t} \int_{-\infty}^{\infty} \frac{d\omega'}{\pi} \frac{\chi''(\omega')}{\omega' - \omega - i\eta} \cdot F(t') e^{i\omega t'} \quad (10.131)$$

The integral

$$I = \lim_{\eta \rightarrow 0} \int_{-\infty}^{\infty} d\omega \frac{e^{-i\omega t}}{\omega + i\eta}$$

can be evaluated as a $\left(\begin{array}{c} \text{clockwise} \\ \text{counter-clockwise} \end{array} \right)$ contour that closes in the $\left(\begin{array}{c} \text{lower} \\ \text{upper} \end{array} \right)$ complex plane with ω

as the real axis for $\left(\begin{array}{c} t > 0 \\ t < 0 \end{array} \right)$. Hence,

$$I = \begin{cases} -2\pi i & t > 0 \\ 0 & t < 0 \end{cases}$$

$$\rightarrow \lim_{\eta \rightarrow 0} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{ie^{-i\omega t}}{\omega + i\eta} = \theta(t) \quad (10.132)$$

(10.131) thus becomes

$$\begin{aligned}
 \alpha_F(t) &= - \lim_{\eta \rightarrow 0} \int_{-\infty}^{\infty} dt' \int_{-\infty}^{\infty} \frac{d\omega'}{\pi} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{e^{-i\omega(t-t')}}{\omega - \omega' + i\eta} \chi''(\omega') \cdot F(t') \\
 &= i \int_{-\infty}^{\infty} dt' \theta(t-t') \int_{-\infty}^{\infty} \frac{d\omega'}{\pi} e^{-i\omega'(t-t')} \chi''(\omega') \cdot F(t') \\
 &= i \int_{-\infty}^t dt' \int_{-\infty}^{\infty} \frac{d\omega}{\pi} e^{-i\omega(t-t')} \chi''(\omega) \cdot F(t') \quad [\omega' \rightarrow \omega] \quad (10.133a)
 \end{aligned}$$

Setting

$$\mathbb{K}''(\tau) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{-i\omega\tau} \chi''(\omega) \quad (10.133b)$$

(10.133a) becomes

$$\alpha_F(t) = 2i \int_{-\infty}^t dt' \mathbb{K}''(t-t') \cdot F(t') \quad (10.133)$$

Note that $\mathbb{K}''(\tau) \neq \text{Im } \mathbb{K}(\tau)$ partly because $\text{Im } \mathbb{K}(\tau) \equiv 0$. Since $\chi''(\omega)$ is odd in ω [see (10.118d)], we have

$$\begin{aligned}\mathbb{K}''(\tau) &= \int_0^\infty \frac{d\omega}{2\pi} e^{-i\omega\tau} \chi''(\omega) - \int_0^\infty \frac{d\omega}{2\pi} e^{i\omega\tau} \chi''(\omega) \\ &= \int_0^\infty \frac{d\omega}{\pi i} \chi''(\omega) \sin \omega\tau\end{aligned}\quad (10.133c)$$

(10.133a) can therefore be written as

$$\alpha_F(t) = 2 \int_{-\infty}^t dt' \int_0^\infty \frac{d\omega}{\pi} \sin \omega(t-t') \chi''(\omega) \cdot F(t') \quad (10.133d)$$

Step Force

Consider the step force

$$F(t) = F\theta(-t) = \begin{cases} F & t < 0 \\ 0 & t \geq 0 \end{cases} \quad (10.134)$$

with Fourier transform

$$\begin{aligned}\tilde{F}(\omega) &= \int_{-\infty}^\infty dt e^{i\omega t} F(t) \\ &= F \int_{-\infty}^0 dt e^{i\omega t} = \lim_{t \rightarrow -\infty} F \frac{1 - e^{i\omega t}}{i\omega}\end{aligned}$$

In order to evaluate

$$\lim_{t \rightarrow -\infty} e^{i\omega t}$$

we set $\omega \rightarrow \omega - i\epsilon$ with $\epsilon \rightarrow 0^+$ so that

$$\lim_{t \rightarrow -\infty} e^{i\omega t} \rightarrow \lim_{t \rightarrow -\infty} e^{i\omega t + \epsilon t} = 0$$

and

$$\tilde{F}(\omega) = \lim_{\epsilon \rightarrow 0} F \frac{1}{i\omega + \epsilon} = -iF \lim_{\epsilon \rightarrow 0} \frac{1}{\omega - i\epsilon} \quad (10.138a)$$

$$= -i \left[\mathcal{P} \frac{1}{\omega} + i\pi\delta(\omega) \right] F \quad [(10.130) \text{ used. }]$$

$$= \left[\mathcal{P} \frac{1}{i\omega} + \pi\delta(\omega) \right] F \quad (10.138)$$

Putting (10.134) into (10.113c) gives

$$\begin{aligned}\alpha_F(t) &= \int_0^\infty d\tau \mathbb{K}(\tau) \cdot F\theta(-t+\tau) \\ &= \begin{cases} \int_0^\infty d\tau \mathbb{K}(\tau) \cdot F & t \leq 0 \\ \int_t^\infty d\tau \mathbb{K}(\tau) \cdot F & t \geq 0 \end{cases}\end{aligned}\quad (10.146a)$$

Using (10.117a), we have

$$\alpha_F(t) = \chi(0) \cdot F \quad \text{for } t \leq 0 \quad (10.146)$$

Also,

$$\alpha_F(\infty) = \int_\infty^\infty d\tau \mathbb{K}(\tau) \cdot F = 0 \quad (10.146b)$$

As expected from physical experiences, the deviation $\alpha_F(t)$ is a constant as long as F is kept on, but

decays eventually to the equilibrium value 0 after F is turned off.

Note that (10.146) is the generalization of the familiar electrostatic relation

$$\mathbf{P} = \chi \cdot \mathbf{E}$$

between the polarization \mathbf{P} and the electric field \mathbf{E} .

Since $\alpha_F(t)$ & F are real, so should $\chi(0)$. This is ensured as follows.

$$\mathbb{K}(\tau) \text{ is real} \quad \rightarrow \quad \chi^*(\omega) = \chi(-\omega) \quad \rightarrow \quad \chi^*(0) = \chi(0) \text{ is real} \quad (10.146c)$$

Setting $u = 0$ in (10.125) gives

$$\chi(0) = \mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega}{i\pi} \frac{\chi(\omega)}{\omega} \quad (10.144)$$

Alternatively, setting $u = 0$ in (10.129) gives

$$\begin{aligned} \chi(0) &= \lim_{\eta \rightarrow 0} \int_{-\infty}^{\infty} \frac{d\omega'}{\pi} \frac{\chi''(\omega')}{\omega' - i\eta} \\ &= \mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \frac{\chi''(\omega)}{\omega} + i\chi''(0) \quad [(10.130) \text{ used. }] \\ &= \mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \frac{\chi''(\omega)}{\omega} \quad [(10.146c) \text{ used. }] \end{aligned} \quad (10.144a)$$

$$= 2 \mathcal{P} \int_0^{\infty} \frac{d\omega}{\pi} \frac{\chi''(\omega)}{\omega} \quad (10.144b)$$

which may be taken as a **sum rule** for $\chi''(\omega)$.

Comparing (10.144a) with (10.144), we get

$$\mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \frac{\chi'(\omega)}{\omega} = 0 \quad (10.144c)$$

which can also be derived from the fact $\chi'(-\omega) = \chi'(\omega)$ [see (10.118d)].

For $t \geq 0$, (10.146a) & (10.115b) give

$$\begin{aligned} \alpha_F(t) &= \int_t^{\infty} d\tau \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{-i\omega\tau} \chi(\omega) \cdot \mathbf{F} \\ &= \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{e^{-i\omega\infty} - e^{-i\omega t}}{-i\omega} \chi(\omega) \cdot \mathbf{F} \\ &= \lim_{\eta \rightarrow 0^+} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi i} \frac{e^{-i(\omega-i\eta)t}}{\omega-i\eta} \chi(\omega) \cdot \mathbf{F} \end{aligned} \quad (10.147a)$$

where, as in (10.138a), we have set $\omega \rightarrow \omega - i\eta$ to get rid of the term $e^{-i\omega\infty}$.

Using (10.130), we have, for $t \geq 0$,

$$\alpha_F(t) = \lim_{\eta \rightarrow 0^+} e^{-\eta t} \left[\mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi i} \frac{e^{-i\omega t}}{\omega} \chi(\omega) \cdot \mathbf{F} + \frac{1}{2} \chi(0) \cdot \mathbf{F} \right] \quad (10.147b)$$

Note that the $e^{-\eta t}$ factor is necessary to keep $\langle \alpha(\infty) \rangle_F = 0$.

To check the result, we set $t = 0$ to get

$$\begin{aligned} \alpha_F(t) &= \mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi i} \frac{1}{\omega} \chi(\omega) \cdot \mathbf{F} + \frac{1}{2} \chi(0) \cdot \mathbf{F} \\ &= \chi(0) \cdot \mathbf{F} \quad [(10.144) \text{ used. }] \end{aligned}$$

in agreement with (10.146).

Alternatively, we can use (10.133d) to get

$$\begin{aligned} \alpha_F(t) &= 2 \int_0^\infty d\tau \int_0^\infty \frac{d\omega}{\pi} \sin \omega \tau \chi''(\omega) \cdot F(t-\tau) \\ &= \begin{cases} 2 \int_0^\infty d\tau \int_0^\infty \frac{d\omega}{\pi} \sin \omega \tau \chi''(\omega) \cdot F & t \leq 0 \\ 2 \int_t^\infty d\tau \int_0^\infty \frac{d\omega}{\pi} \sin \omega \tau \chi''(\omega) \cdot F & t \geq 0 \end{cases} \end{aligned}$$

Using

$$\int_t^\infty d\tau \sin \omega \tau = -\frac{1}{\omega} \cos \omega \tau \Big|_t^\infty = \frac{\cos \omega t}{\omega}$$

we have

$$\alpha_F(t) = \begin{cases} 2 \mathcal{P} \int_0^\infty \frac{d\omega}{\pi} \frac{\chi''(\omega)}{\omega} \cdot F & t \leq 0 \\ 2 \mathcal{P} \int_0^\infty \frac{d\omega}{\pi} \frac{\cos \omega t}{\omega} \chi''(\omega) \cdot F & t \geq 0 \end{cases} \quad (10.147c)$$

Only the principal parts of the integrals are included since the $\omega = 0$ term gives no contribution due to the $\sin \omega \tau$ factor.

Using (10.144b), we have,

$$\alpha_F(t) = \chi(0) \cdot F \quad \text{for} \quad t \leq 0$$

in agreement with (10.146).

For $t \geq 0$,

$$\begin{aligned} \alpha_F(t) &= 2 \mathcal{P} \int_0^\infty \frac{d\omega}{\pi} \frac{\cos \omega t}{\omega} \chi''(\omega) \cdot F \\ &= \mathcal{P} \int_{-\infty}^\infty \frac{d\omega}{\pi} \cos \omega t \frac{\chi''(\omega)}{\omega} \cdot F \end{aligned} \quad (10.147)$$