# On the Continuum Between the Solid and the Liquid State

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With a view on nuclear matter, the continuity between the solid and the liquid states of a material is discussed. For such a viscoelastic medium an acoustical analogue of the Kramers-Kronig relation of linear optics is derived.

According to our usual experience there is a rather clear cut distinction between *solids* and *liquids.* The vast majority of condensed materials is, under normal conditions, either shape preserving and thus in a solid state, or quickly fluid and thus in a liquid state. (This fact is stressed by the remarkable discontinuity known as *melting* which seems to extend to very high pressures and temperatures.) Of course, we know of counter examples as tar or (silly) putty, but they are rare and show a rather complicated behavior. There are two phenomena that bridge the gap between the solids and the fluids: *viscosity* tends to make a fluid somewhat like a solid, whereas *relaxation* tends to make a solid somewhat like a fluid. In the following we will give a short account of these concepts which interpolate between *elastodynamics* and *hydrodynamics* in the much broader frame of *plastodynamics.*

In order to simplify the discussion the idealization of *complete incompressibility* will be made, thus *excluding* all phenomena which are peculiar to *aerodynamics.* Let us remember: a *gas* (and similarly a *plasma*) is a material which assumes infinite volume when the outer pressure is made to vanish; whereas a *condensate*, whether solid or liquid, stays within a finite volume at zero and even at some negative pressure. (Thus nuclear matter in or close to its ground state is undoubtedly in a condensed state.) For a material body under pressure the distinction between the gaseous and the condensed states may become meaningless. (This is especially true just above the critical temperature.) One essential step toward inclusion of aerodynamical possibilities would be to allow for a finite though density independent compressibility. But we will keep to the very extreme of an *ideal condensate* by assuming that all material constants including density be pressure independent. Thermodynamic, relativistic, atomistic (or quantal) intricacies will also be disregarded; and we will avoid all complications due to chemical inhomogeneity or crystallographic anisotropy [1].

Thus we have to deal with an internally homogeneous and isotropic substance whose *mass density* $\mu$ is absolutely constant,

$$\partial\mu/\partial t = 0, \qquad \partial\mu/\partial r = \boldsymbol{0} \tag{1}$$

and which moves with a smooth *velocity field* $u(t, r)$. A necessary (though not sufficient) condition of the time space independence (1) is the *incompressibility*

$$d\mu/dt = 0, \tag{2}$$

formulated by means of the *material time derivation*

$$\frac{d}{dt} = \frac{\partial}{\partial t} + \boldsymbol{u} \cdot \frac{\partial}{\partial r}. \tag{3}$$

This operator forms the time rate along the moving mass particles in contrast to the *local time derivative* $\partial/\partial t$ which forms the time rate at a fixed location $r$. From the *differential mass conservation*

$$\partial\mu/\partial t + \partial \cdot \mu\,\boldsymbol{u}/\partial r = 0 \quad \text{or} \quad d\mu/dt + \mu\,\partial\boldsymbol{u}/\partial r = 0$$

the *nondivergence*

$$\partial \cdot \boldsymbol{u}/\partial r = 0 \tag{4}$$

is obtained as a necessary and sufficient condition for (2). This is a purely kinematical equation, which will be used tacitly.

The dynamical law of motion is given by the *differential momentum balance*

$$\mu\,\frac{du}{dt} + \frac{\partial \cdot \mathbf{p}}{\partial r} = \mathbf{k}, \tag{5}$$

which contains the (*momentum flow density* or) *pressure tensor* **p** and the macroscopic *force density* **k**.

This vector comprises the external or long range properties of the mass element relative to its remote surroundings. The *stress tensor* **p**, on the other hand, summarizes the internal or short range properties of the material, which depend only on the kinematical state of affairs in an infinitesimal neighborhood of the mass element. It is this dependence that will tell us with which kind of plasticity we have to count.

*Conservation of angular momentum* can be shown to demand that the pressure tensor be *symmetric:* $\mathbf{p} = \mathbf{p}^T$. An important invariant is its *trace* $\langle \mathbf{p} \rangle = p_{xx} + p_{yy} + p_{zz}$, which accounts for the *isotropic part* $p \cdot \mathbf{1}$ of **p**, where

$$p = \tfrac{1}{3} \langle \mathbf{p} \rangle \tag{6}$$

is the *pressure scalar* and **1** the unit tensor ($1_{ab} = \delta_{ab}$, so that $\mathbf{1} = \mathbf{1}^T$ and $\langle \mathbf{1} \rangle = 3$). Thus we obtain the decomposition

$$\mathbf{p} = p \cdot \mathbf{1} + \boldsymbol{\pi} \,, \tag{7}$$

the remainder $\boldsymbol{\pi}$ being the traceless *shear pressure*

$$\boldsymbol{\pi} = \boldsymbol{\pi}^T, \quad \langle \boldsymbol{\pi} \rangle = 0 \,. \tag{8}$$

The *shear stress* $- \boldsymbol{\pi}$ is therefore a symmetric tensor with five independent components (which behave under coordinate transformations as some five real-valued spherical harmonics $Y^2$ of the second order).

It follows from isotropy and incompressibility that the *stress scalar* $-p$ cannot be determined from the kinematical state of the material. The quantity $p$ enters the Navier-Stokes equation (5) like a Legendre parameter: its value can be found only *a posteriori* by solving this equation of motion together with the subsidiary condition (4).

The purely anisotropic part $\boldsymbol{\pi}$, on the other hand, is determined *a priori*, by the *kinematical state* of the material continuum in the infinitesimal neighborhood of the mass element under consideration. This state concept is mathematically expressed by two tensors: the *inverse distortion* $\partial s / \partial r$, describing the differential deviations of the mass elements from static equilibrium, and the *velocity gradient* $\partial u / \partial r$. In these derivatives, $\partial s = u \, dt$ is an anholonomic infinitesimal which in general *cannot* be integrated uniquely to deliver an "equilibrium field $s$"; and $u = dr/dt$. It is thus supposed that $\boldsymbol{\pi}$ is a unique function of the *distortion* $\partial r / \partial s = (\partial s / \partial r)^{-1}$ and the flow gradient $\partial u / \partial r$. We may assume that

this function is well approximated by a quickly converging Taylor series.

The most simple hypothesis with regard to $\partial r / \partial s$ is *Hook's elasticity law* which states proportionality between stress and strain. To neglect nonlinearities and, consequently, all kinds of elastic failure, is an assumption akin to our disregard of all aerodynamic, thermodynamic and atomistic complications mentioned before. Similarly we may neglect nonlinearities in $\partial u / \partial r$ (assuming that its components stay much smaller than the relevant molecular frequencies), which leads to *Newton's viscosity law*. Cross terms bilinear in $\partial r / \partial s$ and $\partial u / \partial r$ will also be discarded.

This negligibility of nonlinearities in the velocity gradient implies that the antimetric part of $\partial u / \partial r$, which is geometrically equivalent to the *vorticity*

$$\omega = \frac{\partial}{\partial r} \times u \,, \tag{9}$$

has no appreciable influence on the pressure tensor, because Coriolis and centrifugal forces are bilinear in the velocity components. As the trace of $\partial u / \partial r$ vanishes according to (4), we are left with the traceless *gliding* tensor

$$\boldsymbol{\psi} = \frac{\partial u}{\partial r} + \frac{\partial u^T}{\partial r} - \frac{2}{3} \frac{\partial \cdot u}{\partial r} \mathbf{1} \,. \tag{10}$$

The last term may be omitted because of (1), but for the sake of kinematical lucidity we have kept it in the definition, so that the *symmetries*

$$\boldsymbol{\psi} = \boldsymbol{\psi}^T, \quad \langle \boldsymbol{\psi} \rangle = 0 \tag{11}$$

hold in complete analogy with (8). As already mentioned, we take the *bounding property*

$$\tfrac{1}{2} | \boldsymbol{\psi} | \ll \Omega \tag{12}$$

with some high molecular or lattice frequency $\Omega$ for granted.

A similar consideration holds for the distortion $\partial r / \partial s$. Its antimetric part, being equivalent to the *turning* $\omega = \partial \times r / \partial s$, is completely irrelevant, since an equilibrium does not loose its static quality when submitted to a rotation by a fixed angle $\omega$. Regarding the antimetric part we assume that it is small relative to unity,

$$\frac{1}{2} \left| \frac{\partial r}{\partial s} + \frac{\partial r^T}{\partial s} \right| \ll 1 \,, \tag{13}$$

because otherwise Hook's law would be rather unplausible. Incompressibility tells us the determinant equation $[\partial r/\partial s] = 1$, which by (13) simplifies to the approximate trace equation $\partial \cdot r/\partial s + \ldots = 0$. Thus we see that the trace $\partial \cdot r/\partial s = [\partial r/\partial s] - 1 + \ldots$ is almost inoperative, and we are left with the shear strain tensor

$$\chi = \frac{\partial r}{\partial s} + \frac{\partial r^{\mathsf T}}{\partial s} - \frac{2}{3} \frac{\partial \cdot r}{\partial s}\, \mathbf 1\,, \tag{14}$$

which has the *symmetry*

$$\chi = \chi^{\mathsf T}\,, \quad \langle \chi \rangle = 0\,, \tag{15}$$

and the *bounding property*

$$\tfrac{1}{2}\,|\chi| \ll 1\,. \tag{16}$$

The anholonomic infinitesimal $\partial s$ is fictitious and to some extent ambiguous and has to be well distinguished from the differential $dq$ of the particle parameter $q$ that is fundamental for the Lagrangean form of continuum kinematics. This quantity $q$ may, or may not, equal the initial position $r_0$ (or $r_{-\infty}$) of the particles. All the dictionary translating between this and the Eulerian form, which is based on the space-fixed position coordinate $r$, is contained in the transformation equation

$$\left(\frac{\partial}{\partial t}\right)_q = \frac{d}{dt} \tag{17}$$

where the abbreviation (3) has been used again. A special case of (17) is the definition $u = (\partial r/\partial t)_q$. Contrary to $q$ and $dq$, we are not going to make any real use of $\partial s$.

We are now in the position to write down the linear *stress law of Hook and Newton*:

$$-\pi = \varepsilon\,\chi + \zeta\,\psi\,, \tag{18}$$

where the *rigidity* $\varepsilon$ and the *obstinacy* $\zeta$ are assumed to be independent of the pressure components $\pi$ and $p$ as well as of the strain $\chi$ and the velocity gradient $\partial u/\partial r$. Their metrological dimensions are $[\varepsilon] = ML^{-1}T^{-2}$ and $[\zeta] = ML^{-1}T^{-1}$. The semiqualities *

$$0 \leqq \varepsilon\,, \quad 0 \leqq \zeta \tag{19}$$

are known from hydrostatic and thermodynamic stability considerations.

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* We do not call them "inequalities", because the equalities $0 = \varepsilon$ or $0 = \zeta$ are not excluded.

In order to have a complete system of equations not outnumbered by the unknown functions $p$, $u$, $\pi$, and $\chi$, we need an equation of state for the strain $\chi$ or the gliding $\psi$ or both, which would supplement (4), (5), (18). For a *perfectly solid* body we may put $\partial s = ds = dq$ and therefore

$$\frac{d}{dt}\chi = \psi\,. \tag{20}_S$$

In a *perfect liquid*, on the other hand, all mass elements should always be in equilibrium with respect to shearing, so that $\partial s/\partial r = \mathbf 1$ (besides some undetermined $\omega$) and thus

$$\chi = \mathbf 0\,. \tag{20}_L$$

For a material not perfectly liquid, the most simple hypothesis is to assume that, apart from the feeding input $\psi$ acting via $(20)_S$, the strain $\chi$ should *relax* toward the equilibrium $(20)_L$ with a time rate proportional to its instantaneous value. We are then led to *Maxwell's relaxation law*

$$\left(\frac{d}{dt} + \frac{1}{\tau}\right)\chi = \psi\,, \tag{20}$$

where the *relaxation time* $\tau$ is taken to be independent of $\chi$, $\psi$ and $p$. Of course, $[\tau] = T$, and the obvious inequality

$$0 < \tau \tag{21}$$

conforms with the irreversible levelings of thermodynamics.

Sometimes it is nice to replace the material constant $\tau$ by its inverse, the *relaxivity*

$$\varrho = 1/\tau\,. \tag{22}$$

A material behaves almost like a solid if the *frequencies* $\omega$ of the occurring disturbances stay much higher than $\varrho$; and it behaves almost like a liquid if the relevant $\omega$-values remain much lower than $\varrho$. In short, we have

$$\left.\begin{array}{l} \omega \ll \varrho \\ \varrho \ll \omega \end{array}\right\} \quad \text{in the} \quad \left\{\begin{array}{l} \text{liquid case (L)} \\ \text{solid case (S)} \end{array}\right\}\,. \tag{23}$$

The ratio $\varrho/\omega$ plays the role of a dimensionless *damping constant* for the transverse sound waves of frequency $\omega$. In the liquid case this damping is of the strongly aperiodic type, whereas the solid case is characterized by weakness of relaxation damping. (Damping by viscosity is a different phenomenon.)

What is to say about the integration constant $\mathbf{c}$ left open by the differential equation (20)? The formal solution of this equation is

$$\boldsymbol{\chi}(t) = \left(\varrho + \frac{\mathrm{d}}{\mathrm{d}t}\right)^{-1} \boldsymbol{\psi}(t) + \mathbf{c} \cdot 2\pi\,\delta\left(\varrho + \frac{\mathrm{d}}{\mathrm{d}t}\right). \quad (24)$$

The second term, being a solution of the homogeneous equation $\left(\varrho + \frac{\mathrm{d}}{\mathrm{d}t}\right)\boldsymbol{\chi}_0 = 0$, is proportional to $e^{-\varrho t}$. Therefore

$$\mathbf{c} = 0 \qquad (25)$$

as a consequence of (12) and (16) in the limit $t \to -\infty$, because the first term of $\boldsymbol{\chi}$ in (24) can never exceed the bounded quantity $(\varrho + \bar{\omega})\,\boldsymbol{\psi}$ by orders of magnitude. Inserting (24) with (25) into (18) one obtains the *linear stress law*

$$-\boldsymbol{\pi} = \eta\,\boldsymbol{\psi}\,, \qquad (26)$$

where the operator

$$\eta = \zeta + \varepsilon\left(\varrho + \frac{\mathrm{d}}{\mathrm{d}t}\right)^{-1} \qquad (27)$$

of complex *viscosity* has been used.

The case $\omega \gg \varrho$ of a flow containing only "solid" frequencies is covered by *elastodynamics*. This is propitiously treated by the Lagrangean kinematics where the material time derivative occurring in (20) or (27) becomes a simple partial derivative, as stated in (17). This advantage does not persist in the opposite domain $\omega \ll \varrho$ of flows that carry exclusively "liquid" frequencies. Here, in *hydrodynamics*, the particle positions $\mathbf{r}$ may before long deviate from the particle parameters $\mathbf{q}$ vastly and in an approximately ergodic manner which makes $\partial\mathbf{r}/\partial\mathbf{q}$ very different from $\mathbf{1}$. But fortunately, the operator $\mathrm{d}/\mathrm{d}t$ is now effectively much smaller than the factor $\varrho$, and so (27) may be well evaluated by a geometrical series:

$$\eta = \zeta + \varepsilon\,\tau \sum_{n=0}^{\infty} (-1)^n\,\tau^n\,\frac{\mathrm{d}^n}{\mathrm{d}t^n}\,. \qquad (28)$$

In the low frequency limit, $\omega = 0$, this becomes simply $\eta = \zeta + \tau \cdot \varepsilon$.

A *perfect solid* may now be defined as the limiting case $\tau = +\infty$, which simplifies (20) to $(20)_S$ and (18) to

$$-\boldsymbol{\pi} = \left(\varepsilon + \zeta\,\frac{\mathrm{d}}{\mathrm{d}t}\right)\boldsymbol{\chi}\,. \qquad (18)_S$$

Here, (17) and (14) with $\partial\mathbf{s} = \partial\mathbf{q}$ may be conveniently used. In order to obtain an *ideal solid*, we have to add the condition $\zeta = 0$ of *inobstinacy*. Alternatively, an absolutely *rigid body* comes about in the limit $\varepsilon = +\infty$ and $\tau > 0$, which implies $\boldsymbol{\chi} = \mathbf{0}$ and $\boldsymbol{\psi} = \mathbf{0}$, regardless of the $\zeta$ value.

On the other side, we may characterize a *perfect liquid* as belonging to the limit $\tau = +0$ and $\varepsilon < \infty$, which simplifies (20) to $(20)_L$ and (18) to

$$-\boldsymbol{\pi} = \zeta \cdot \boldsymbol{\psi}\,, \qquad (18)_L$$

making $\zeta = \eta$. An *ideal liquid* is obtained by adding the condition $\eta = 0$ of *inviscidity* and by supplementing it with the condition $\boldsymbol{\omega} = \mathbf{0}$ of *irrotationality*. This supplementation stays valid all times if it is assumed valid for some time, as long as the long range forces are conservative ($\mathbf{k} = -\partial\Phi/\partial\mathbf{r}$). This follows from the *vorticity law*

$$\frac{\mathrm{d}\boldsymbol{\omega}}{\mathrm{d}t} - \boldsymbol{\omega} \cdot \frac{\partial\mathbf{u}}{\partial\mathbf{r}} = \frac{\partial \times \mathbf{k}}{\partial\mathbf{r}} + \eta \cdot \frac{\partial^2\boldsymbol{\omega}}{\partial\mathbf{r}^2} \qquad (29)$$

of Helmholtz, which may be easily derived from (4), (5), (7), and $(18)_L$. An *ideal fluid* is not necessarily incompressible.

If the series in (26) converges quickly, it may well be terminated and is thus effectively a differential operator. Otherwise it is really an *integral operator*. This is most easily seen by an exact integration of the differential equation (20), which may be done either by variation of constants or by a Fourier analysis of (24). The solution

$$\boldsymbol{\chi}(t) = e^{-\varrho t} \int_{-\infty}^{t} \mathrm{d}t'\, e^{\varrho t'}\,\boldsymbol{\psi}(t') \qquad (30)$$

can be easily checked; and the absence of a homogeneous contribution $\mathbf{c}\,e^{-\varrho t}$ has already been stated in (24). In the solid limit $\varrho = +0$ this becomes the obvious solution

$$\boldsymbol{\chi}(t) = \int_{-\infty}^{t} \mathrm{d}t'\,\boldsymbol{\psi}(t') \qquad (30)_S$$

of $(20)_S$; and in the liquid limit $\varrho = +\infty$ we obtain just

$$\boldsymbol{\chi}(t) = 0\,, \qquad (30)_L$$

which is identical with $(20)_L$.

An important generalization of the material relation (20) allows for *dispersion*, a phenomenon much more familiar in electrodynamics or optics than in

elastodynamics or acoustics. In the latter case it means a dependence of rigidity and viscosity on the *frequency* $\omega$ of the (shearing) matter disturbances known as (transversal) sound waves. (According to our incompressibility assumption the longitudinal sound has infinite velocity and vanishing amplitude.) Thus we keep $\tau$ or $\varrho$ unchanged but understand $\varepsilon$, $\zeta$, and $\eta$ as $\varepsilon_\omega$, $\zeta_\omega$, and $\eta_\omega$, respectively. All equations and inequalities stay formally unchanged but have to be slightly reinterpreted: numerical factors become linear operators. Let us shortly recollect how this works.

The precise meaning of the $\omega$-dependence is, as usual, realized by a *Fourier analysis*. If $\Phi$ is any time dependent quantity as, for example, $p$ or some component of $\boldsymbol{u}$, $\boldsymbol{k}$, $\mathbf{p}$, $\boldsymbol{\pi}$, $\chi$ or $\boldsymbol{\psi}$, we shall write

$$\Phi(t) = \frac{1}{2\pi} \cdot \int_{-\infty}^{\infty} d\omega\, e^{-i\omega t} \Phi_\omega \Leftrightarrow$$
$$\Phi_\omega = \int_{-\infty}^{\infty} dt\, e^{i\omega t} \Phi(t). \tag{31}$$

This $\Phi_\omega$ is a complex quantity, and as for *reality* we have

$$\Phi(t) = \Phi(t)^* \Leftrightarrow \Phi_\omega = \Phi_{-\omega}^*. \tag{32}$$

In connection with (20) we need the operator relation

$$d/dt \Leftrightarrow -i\omega, \tag{33}$$

which, of course, means that $(\dot\Phi)_\omega = -i\omega \cdot \Phi_\omega$. Then (25) and (26) are taken to mean

$$-\boldsymbol{\pi}_\omega = \eta_\omega \boldsymbol{\psi}_\omega \tag{26}_\omega$$

and

$$\eta_\omega = \zeta_\omega + \frac{i\varepsilon_\omega}{\omega + i\varrho}, \tag{27}_\omega$$

respectively. Transforming back, we find the $t$-representation

$$-\boldsymbol{\pi}(t) = \int_{-\infty}^{\infty} dt'\, \eta(t-t')\, \boldsymbol{\psi}(t') \tag{26}_t$$

of (26) with the viscosity kernel

$$\eta(t-t') = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega\, \eta_\omega e^{-i\omega(t-t')}. \tag{34}_t$$

The $t$-representation of (27) reads

$$\eta(t-t') = \zeta(t-t')$$
$$+ \int_{-\infty}^{t} dt''\, e^{-\varrho\cdot(t-t'')} \varepsilon(t''-t') \tag{27}_t$$

as can be seen from (30). Of course, the order of the two factor kernels $e^{-\varrho(t-t'')}$ and $\varepsilon(t''-t')$ may also be reversed which brings the product into the form $\int_{t'}^{\infty} dt''\, \varepsilon(t-t'')\, e^{-\varrho(t''-t')}$. The semiqualities (19) now read

$$0 \leq \varepsilon_\omega = \varepsilon_{-\omega}, \qquad 0 \leq \zeta_\omega \leq \zeta_{-\omega} \tag{19}_\omega$$

in the $\omega$-representation; but the $t$-representation is less explicit:

$$0 \leq \int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} dt'\, \Phi(t)^* \varepsilon(t-t')\, \Phi(t'),$$
$$0 \leq \int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} dt'\, \Phi(t)^* \zeta(t-t')\, \Phi(t'), \tag{19}_t$$

for each complex field $\Phi(t)$.

It is plausible to suppose that $(27)_\omega$ is *continuous* and thus *finite* for all $\omega$ values *within the open interval* $(0 \mid \infty)$. With regard to the limit $\omega \to 0$ we know from experience that the viscosity (28) stays finite,

$$0 \leq \eta_0 = \zeta_0 + \tau \cdot \varepsilon_0 < \infty, \tag{35}$$

which implies by $(19)_\omega$ that $\zeta_0$ and $\varepsilon_0$ are finite, too. The same may be assumed, and empirically tested, for the opposite limit $\omega \to \infty$. With this experimental proviso we may thus assume the *finiteness*

$$\zeta_\omega < \infty, \qquad \varepsilon_\omega < \infty \tag{36}$$

for the whole closed interval $[-\infty \mid \infty]$. Furthermore, it seems to be an empirical fact, supported by molecular considerations, that $\zeta_\omega$ becomes rather small for $\omega$-values well above some high molecular frequency $\Omega$. There the matter distortions do not have enough time to build up much of the collective and irreversible obstinacy effects. In the very limit we arrive at the *yielding property*

$$\zeta_\infty = 0, \tag{37}$$

which assumes the sound waves to become reversible lattice oscillations. The second inequality of (36)

says that their velocity $\sqrt{\varepsilon_\omega/\mu}$ stays finite for $\omega \to \infty$. (The lattice does not show up, of course, in our mathematical scheme, which idealizes the material to be a macroscopic continuum of constant mass density.)

In order to simplify the analysis we shall replace the couple of real, non-negative functions $\zeta_\omega$, $\varepsilon_\omega$ by the real and the imaginary part of $\eta_\omega$, which we call $\gamma_\omega$ and $\omega \delta_\omega$, respectively:

$$\eta_\omega = \gamma_\omega + i \omega \, \vartheta_\omega, \tag{38$_\omega$}$$

where the *implasticity* $\gamma_\omega$ and the *inelasticity* $\vartheta_\omega$ satisfy

$$0 \le \gamma_\omega = \zeta_\omega + \frac{\varrho \, \varepsilon_\omega}{\varrho^2 + \omega^2} = \gamma_{-\omega},$$

$$0 \le \vartheta_\omega = \frac{\varepsilon_\omega}{\varrho^2 + \omega^2} = \vartheta_{-\omega}. \tag{39$_\omega$}$$

The *effective reality*

$$\eta_\omega = \eta_{-\omega}^* \tag{40$_\omega$}$$

in the $\omega$-representation is equivalent to the *simple reality*

$$\eta(t - t') = \eta(t - t')^*, \tag{40$_t$}$$

in the $t$-representation, enduring that any real valued $\boldsymbol{\psi}(t')$ in (26)$_t$ will give rise to a $-\boldsymbol{\pi}(t)$ which is also real valued.

The invariant formulations of (38)$_\omega$ and (39)$_\omega$ are easily written down when (33) is substituted:

$$\eta = \gamma - \vartheta \, (\mathrm{d}/\mathrm{d}t) \tag{38}$$

and

$$0 \le \gamma = \zeta + \varrho \cdot \varepsilon \left( \varrho^2 - \frac{\mathrm{d}^2}{\mathrm{d}t^2} \right)^{-1},$$

$$0 \le \vartheta = \varepsilon \left( \varrho^2 - \frac{\mathrm{d}^2}{\mathrm{d}t^2} \right)^{-1}, \tag{39}$$

respectively. All the operators $\mathrm{d}/\mathrm{d}t$, $\vartheta$, $\varepsilon$, $\zeta$, $\gamma$, $\eta$ and $\varrho$ commute. Therefore the $t$-representation of (38) is simply

$$\eta(t - t') = \gamma(t - t') - \vartheta'(t - t'). \tag{38$_t$}$$

The inverse operators in (39) become integrations:

$$\vartheta(t - t') = \frac{1}{2\varrho} \int_{-\infty}^{\infty} \mathrm{d}t'' [\theta(t - t'') \, e^{-\varrho \cdot (t - t'')} \tag{39$'_t$}$$

$$+ \, \theta(t'' - t) \, e^{-\varrho \cdot (t'' - t)}] \, \varepsilon(t'' - t'),$$

and similarly for $\gamma(t - t')$. The reality statements implied in (39) are $t$-represented by the hermitic equations

$$\gamma(t - t') = \gamma(t' - t)^*, \quad \vartheta(t - t') = \vartheta(t' - t)^*, \tag{39$''_t$}$$

whereas the semiinqualities become statements similar to (19)$_t$.

If the relaxivity constant $\varrho$ is known, it is easy to invert our substitution $(\zeta, \varepsilon) \Rightarrow (\gamma, \vartheta)$ by solving (39)$_\omega$ for $\zeta_\omega$ and $\varepsilon_\omega$ as unknowns:

$$\zeta_\omega = \gamma_\omega - \varrho \cdot \vartheta_\omega, \quad \varepsilon_\omega (\varrho^2 + \omega^2) \, \vartheta_\omega \tag{47$_\omega$}$$

or, invariantly stated

$$\zeta = \gamma - \varrho \cdot \vartheta, \quad \varepsilon = \vartheta \left( \varrho^2 - \frac{\mathrm{d}^2}{\mathrm{d}t^2} \right). \tag{41}$$

This raises the question whether it is possible to define $\varrho$ in terms of $\gamma$ and $\vartheta$, too. From a purely mathematical point of view the answer is ambiguous, because any choice consistent with (21), (22), (19)$_\omega$, (39)$_\omega$ is possible, which means that $\varrho$ must only fulfill the semiqualities

$$0 \le \varrho \le \min_\omega \frac{\gamma_\omega}{\vartheta_\omega}. \tag{42}$$

Is there any reason to select a special value out of this closed interval as an appropriate measure of relaxivity?

In any case we have $\varrho = (\gamma_\omega - \zeta_\omega) : \vartheta_\omega$ independently of $\omega$; yet the occurrence of $\zeta_\omega$ is, of course, circular as long as we have no independent information on this quantity. Fortunately enough, such an *a priori* information is provided by (37), and so we may postulate that

$$\varrho = \lim_{\omega \to \infty} \frac{\gamma_\omega}{\vartheta_\omega}. \tag{43}$$

As this equals $\min_\omega(\gamma_\omega/\vartheta_\omega)$, the condition (42) is not violated.

The experimental determination of the limit (43) is facilitated by a *dispersion relation* which is the acoustical variant of a complex equation well known in optics as the Kramers-Kronig relation. It is a consequence of that aspect of "causality" which ensures that no effect can occur earlier in time than its cause. This warrants for the integral operator of (26)$_t$ the *causality condition*

$$\eta(t - t') = 0 \quad \text{whenever} \quad t < t'. \tag{44}$$

From this it may be proved [2] that the Fourier component

$$\eta_\omega = \int_{-\infty}^{\infty} dt\, \eta(t)\, e^{i\omega t} \tag{34}_t$$

is *holomorphic in the upper $\omega$ semiplane*. As a consequence [2] we may formulate a *Hilbert identity* whose exact form depends, to be sure, on the integrability properties of $\eta_\omega$ along the real axis. The asymptotic properties

$$\gamma_\infty = 0, \quad \vartheta_\infty = 0 \quad \text{and thus} \quad \eta_\infty = 0, \tag{45}$$

which follow from (36), (37) when inserted into $(39)_\omega$, $(38)_\omega$, ensure the most simple form, which does not contain any "polynomial substractions". Thus we obtain the acoustical version of the *Kramers-Kronig relation:*

$$\eta_\omega = \frac{\mathscr{P}}{i\pi} \int_{-\infty}^{\infty} d\omega'\, \frac{\eta_{\omega'}}{\omega' - \omega}, \tag{46}$$

where $\omega'$ as well as $\omega$ has to stay on the real axis, and $\mathscr{P}\int$ performs a Cauchy integration at the singularity $\omega' = \omega$.

When the complex equation (46) is decomposed into its real and imaginary parts along (38) and using $(19)_\omega$, $(39)_\omega$, this couple of *mutually inverse integral transformations* issues

$$\gamma_\omega = \frac{2}{\pi} \mathscr{P}\int_0^\infty d\omega'\, \frac{\omega'^2\, \vartheta_{\omega'}}{\omega'^2 - \omega^2},$$

$$\vartheta_\omega = -\frac{2}{\pi} \mathscr{P}\int_0^\infty d\omega'\, \frac{\gamma_{\omega'}}{\omega'^2 - \omega^2}. \tag{47}$$

They interconnect the dispersive functions of implasticity and inelasticity in a way similar to any

relation between an absorption function and its ensuing refraction function. *Without implasticity there can be no inelasticity and vice versa.*

If the Kramers-Kronig relations are considered at $\omega \to 0$, one achieves the *low frequency sum rules:*

$$\gamma_0 = \frac{2}{\pi} \int_0^\infty d\omega\, \vartheta_\omega, \qquad \vartheta_0 = \int_0^\infty d\omega\, \frac{\gamma_0 - \gamma_\omega}{\omega^2}.$$

The first is an immediate consequence of (47), and the other is obtained in the same way, when the analytical function

$$i(\gamma_0 - \eta_\omega)/\omega = \vartheta_\omega + i\,\omega\,(\gamma_0\,\omega^{-2} - \omega^{-2}\gamma_\omega)$$

is analyzed instead of the analytical function

$$\eta_\omega = \gamma_\omega + i\,\omega\,\vartheta_\omega.$$

For $\omega \to \infty$ we find similarly *the high frequency sum rules*

$$v_\infty = \frac{2}{\pi} \int_0^\infty d\omega\, \gamma_\omega, \quad \xi_\infty = \frac{2}{\pi} \int_0^\infty d\omega\, (v_\infty - \omega^2 \vartheta_\omega) \tag{49}$$

for the subsidiary functions

$$v_\omega = \omega^2\, \vartheta_\omega, \qquad \xi_\omega = \omega^2\, \gamma_\omega. \tag{50}$$

Again, the first equation in (49) follows immediately from (47), and the second in the same way by studying $v_\infty + i\,\omega\,\eta_\omega = (v_\infty - v_\omega) + i\,\omega\,\gamma_\omega$ in place of $\eta_\omega = \gamma_\omega + i\,\omega\,\vartheta_\omega$.

Applying these results to our previous definitions (43), (22), we see now that the relaxation time

$$\tau = v_\infty / \xi_\infty \tag{51}$$

can be evaluated by the frequency integrals (49). This may considerably improve the accuracy in cases where the high frequency tale of the viscosity dispersion $\eta_\omega$ is not well known.

[1] E. N. da C. Andrade, Viscosity and Plasticity, Chemical Publ., New York 1951; – C. Torre, Trans. Soc. Rheology **2**, 153 (1958); – A. C. Pipkin, Viscoelasticity Theory, Springer, Berlin 1972; – P. Haupt, Viskoelastizität und Plastizität, Springer, Berlin 1977.
[2] E. C. Titchmarsh, Theory of Fourier Integrals, University press, Oxford 1937, Chapter V.