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Abstract:

From the principle of virtual work it is shown that, if equilibrium stress fluctuations are described by a Langevin equation, there is only one possible extension of this equation to describe stress fluctuations in a shear flow. Also, it is shown that the resulting equation is consistent with linear response theory. The formalism developed may be looked upon as a method for extending differential constitutive relations to incorporate thermal fluctuations. A few simple models are discussed as illustrations. These include a model where the stress fluctuates freely below a certain limit, and a model constructed to mimic the Eyring-Tobolsky phenomenological theory of viscoelastic liquids from the 1940's. It is concluded that these and similar models, however, do not realisticly describe real polymeric liquids. To reach this goal, models involving several stress degrees of freedom will have to be considered.

1. INTRODUCTION

The presently most popular models of the viscoelasticity of polymeric liquids are based on a microscopic description, where the bead coordinates are the degrees of freedom [1,2]. These models are rather successful but, unfortunately, also quite complicated to solve. The present paper investigates the more phenomenological approach to take as basic degree of freedom the quantity of main interest, the shear stress. In part, this represents a return to ideas proposed by Eyring, Tobolsky and coworkers many years ago [3-5]. Here, however, a formalism is developed which is consistent with statistical mechanics. This is done by assuming a Langevin equation for the shear stress dynamics in equilibrium. From this the stress fluctuations may be calculated and thereby, via the fluctuation-dissipation theorem, the linear frequency-dependent viscosity. The main result of the paper is a proof that the non-linear response to any shear displacement is uniquely determined by the requirement that the principle of virtual work is obeyed. This result is derived in Sec. 2. In Sec. 3 a few simple models are worked out. Finally, in Sec. 4 a discussion is given. It is concluded that, in order to realisticly model reality, the proposed formalism must be generalised to consider spatially varying stresses.

2. THE UNIQUE COUPLING OF DEFORMATION TO STRESS FLUCTUATIONS

Here and henceforth the term stress refers to the shear stress σ_{xy}. The corresponding x-y-shear rate is denoted by $\dot{\gamma}(t)$. The fluctuation-dissipation theorem [2] allows a calculation of the frequency-dependent viscosity, $\eta(\omega)$, in terms of equilibrium stress fluctuations. The theorem states that the stress relaxation modulus, $G(t)$, which is characterized by

$$\eta(\omega) = \int_0^\infty G(t)\, e^{-i\omega t} dt \quad , \tag{1}$$

is given by

$$G(t) = \beta \frac{\langle s(0)\, s(t) \rangle_0}{V} \quad . \tag{2}$$

Here, a sample of volume V is considered, s is the "total" stress (i. e. $\sigma_{xy} = s/V$), and $\beta = 1/k_B T$. The subscript zero on the right hand side of Eq. (2) is introduced to remind that the autocorrelation function refers to fluctuations in thermal equilibrium. If $\mathbf{R}_i$ is the coordinate vector of the i'th bead and $\mathbf{F}_{ij}$ is the force from the i'th to the j'th bead, the quantity s is given [2] by

$$s = -\sum_i \boldsymbol{R}_{i,x}\, \boldsymbol{F}_{i,y} \quad , \tag{3}$$

(Note that the relaxation modulus of Eq. (2) has a well-defined limit for $V \to \infty$: this is the usual bulk shear stress relaxation modulus.)

From now on the following simple model is adopted. The liquid is regarded as divided into regions whose stresses fluctuate independently. A discussion of this rather severe approximation is postponed to Sec. 4. There is just one relevant degree of freedom in the model, the quantity s of Eq. (3) where the sum is now restricted to one region. The quantity s has dimension energy, but will still be referred to as the "stress". It is convenient also to redefine "viscosity" by absorbing the region volume, so that viscosity is from now on simply $\langle s\rangle/\dot{\gamma}$. With these definitions Eqs. (1) and (2) become

$$\eta(\omega) = \beta\int_0^\infty \langle s(0)\,s(t)\rangle_0 e^{-i\omega t}dt \quad . \tag{4}$$

We remind that Eq. (4) is equivalent to saying that the average stress for small shear rate $\dot{\gamma}(t)$ is given by

$$\langle s(t)\rangle_{\dot{\gamma}} = \beta\int_0^\infty \langle s(0)\,s(\tau)\rangle_0\dot{\gamma}(t-\tau)\,d\tau \quad . \tag{5}$$

According to statistical mechanics, the probability of an s-fluctuation in equilibrium is given by the free energy as function of s , $F(s)$. If Γ denotes the complete set of microscopic coordinates referring to one region, and $s(\Gamma)$ and $E(\Gamma)$ denote respectively the value of s and of energy in state Γ , $F(s)$ is given by

$$e^{-\beta F(s)} = \int e^{-\beta E(\Gamma)}\,\delta[s-s(\Gamma)]\,d\Gamma \quad . \tag{6}$$

It is assumed that $F(s)\to\infty$ as $|s|\to\infty$. Note that the free energy of one region is given by

$$e^{-\beta F} = \int_{-\infty}^{\infty} e^{-\beta F(s)} ds \quad . \tag{7}$$

Now the further assumption is made that the s-fluctuations in equilibrium are described by a Langevin equation,

$$\dot{s} = -\mu \frac{dF}{ds} + \xi(t) \quad . \tag{8}$$

Here, μ is the "mobility" ("velocity"/"force") which determines the time scale, and $\xi(t)$ is a Gaussian white noise term:

$$\langle \xi(t) \xi(t') \rangle = 2\, \mu\, k_B T\, \delta(t-t') \quad . \tag{9}$$

While Eq. (8) is a completely phenomenological postulate, it has the crucial property [2,6,7] that the stationary s-probability distribution, $P_0(s)$, is that required by statistical mechanics:

$$P_0(s) = N^{-1}\, e^{-\beta F(s)} \quad . \tag{10}$$

Any initial probability distribution converges to $P_0(s)$ as $t \to \infty$; the equation governing this is the well-known Smoluchowski equation [2,6,7] (sometimes referred to as the Fokker-Planck equation or just the diffusion equation)

$$\frac{\partial P}{\partial t} = \frac{\partial}{\partial s}\left(\mu \frac{dF}{ds} P + \mu k_B T \frac{\partial P}{\partial s}\right) \quad . \tag{11}$$

A substitution of Eq. (10) into Eq. (11) confirms that $P_0(s)$ is the stationary solution.

How is the s-dynamics changed when the liquid flows? The simplest way to modify Eq. (8) is to add an extra term coupling to $\dot{\gamma}(t)$, writing

$$\dot{s} = -\mu\frac{dF}{ds}+\dot{\gamma}(t)J(s)+\xi(t) \quad . \tag{12}$$

In Eq. (12) the shear rate plays the role of an external field. Obviously, $J(s)$ is a kind of s-dependent infinite frequency shear modulus. The Smoluchowski equation corresponding to Eq. (12) is

$$\frac{\partial P}{\partial t} = \frac{\partial}{\partial s}\left(\left[\mu\frac{dF}{ds}-\dot{\gamma}(t)J\right]P\right) + \mu k_B T\frac{\partial^2 P}{\partial s^2} \quad . \tag{13}$$

We now proceed to show that $J(s)$ is uniquely determined by the principle of virtual work. Then it is shown that, with this choice of $J(s)$, linear response theory is reproduced, as is neccesary to have a consistent theory.

For a given probability distribution $P(s)$ the dynamical free energy, A , is defined [2] by

$$A = \int_{-\infty}^{\infty} ds\ P(s)\left[F(s)+k_B T\ \ln P(s)\right] \quad , \tag{14}$$

The principle of virtual work says that, after the virtual displacement

$$\dot{\gamma}(t) = \delta\gamma\ \delta(t) \quad , \tag{15}$$

A is changed by

$$\delta A = \langle s\rangle\ \delta\gamma \quad . \tag{16}$$

From Eq. (14) the variation in A is given by

$$\delta A = \int_{-\infty}^{\infty} ds\ \delta P(s)\ \left[F(s) + k_B T[\ln(P(s)) + 1]\right] \quad . \tag{17}$$

Substituting Eq. (15) into Eq. (13) and integrating over a small interval around 0 gives, to lowest order in $\delta\gamma$:

$$\delta P = -\delta\gamma \frac{\partial}{\partial s}(JP) \quad . \tag{18}$$

By combining Eqs. (17) and (18) one finds by partial integrations

$$\begin{aligned} \delta A &= -\delta\gamma \int_{-\infty}^{\infty} ds \left[F \frac{\partial}{\partial s}(JP) + k_B T(\ln(P) + 1) \frac{\partial}{\partial s}(JP)\right] \\ &= \delta\gamma \int_{-\infty}^{\infty} ds \left[\frac{dF}{ds} JP + k_B T \frac{\partial P}{\partial s} J\right] \\ &= \delta\gamma \int_{-\infty}^{\infty} ds \left[\frac{dF}{ds} J - k_B T \frac{dJ}{ds}\right] P \end{aligned} \tag{19}$$

If this is to be consistent with Eq. (16) for all P(s) , J(s) must obey

$$\frac{dJ}{ds} = \beta \frac{dF}{ds} J - \beta s \quad . \tag{20}$$

The solution of this equation is

$$J(s) = e^{\beta F(s)} \int_{s}^{\infty} ds' \beta s' e^{-\beta F(s')} \quad . \tag{21}$$

All other solutions lead to an exponentially increasing J(s) and thereby an inconsistent model where s runs off to infinity in any shear flow.

Next it is shown that the J(s) of Eq. (21) ensures that linear response theory is reproduced. First Eq. (12) is rewritten as

$$\dot{s} = -\mu \frac{\partial}{\partial s} [F - \dot{\gamma}(t)\Phi] + \xi(t) \quad , \tag{22}$$

where

$$\frac{d\Phi}{ds} = \frac{J}{\mu} \quad . \tag{23}$$

Equation (22) shows that the coupling to the shear displacement field appears as an extra term, $-\dot{\gamma}(t)\Phi(s)$, in the Hamiltonian. In the small shear rate limit, linear response theory **applied to Eq. (22)** [2] leads to

$$\langle s(t) \rangle_{\dot{\gamma}} = \beta \int_0^\infty \dot{\gamma}(t-\tau) \frac{-d}{d\tau} \langle \Phi(0) s(\tau) \rangle_0 \, d\tau \quad . \tag{24}$$

Equation (24) is consistent with Eq. (5) if

$$-\frac{d}{d\tau} \langle \Phi(0) s(\tau) \rangle_0 = \langle s(0) s(\tau) \rangle_0 \quad . \tag{25}$$

To prove Eq. (25) note that the Φ-s correlation function is given by

$$\langle \Phi(0) s(\tau) \rangle_0 = \int_{-\infty}^{\infty} ds' s' \int_{-\infty}^{\infty} ds \; \Phi(s) P_0(s) G_0(s, s'; \tau) \quad . \tag{26}$$

where $G_0(s,s';\tau)$ is the equilibrium Green's function. By substituting into this expression the time integrated version of detailed balance, $P_0(s)G_0(s,s';\tau) = P_0(s')G_0(s',s;\tau)$, one finds

$$-\frac{d}{d\tau}\langle\Phi(0)\,s(\tau)\rangle_0 = -\int_{-\infty}^{\infty}ds's'P_0(s')\int_{-\infty}^{\infty}ds\;\Phi(s)\,\frac{\partial G_0}{\partial\tau}(s',s;\tau) \tag{27}$$

The Green's function considered as function of the second variable satisfies Eq. (11), of course, and therefore one gets

$$\begin{aligned}-\frac{d}{d\tau}\langle\Phi(0)\,s(\tau)\rangle_0 &= -\int_{-\infty}^{\infty}ds's'P_0(s')\int_{-\infty}^{\infty}ds\;\Phi(s)\left[\mu\frac{\partial}{\partial s}\left(\frac{dF}{ds}G_0\right)+\frac{\mu}{\beta}\frac{\partial^2 G_0}{\partial s^2}\right]\\ &= \int_{-\infty}^{\infty}ds's'P_0(s')\int_{-\infty}^{\infty}ds\left(J(s)\,\frac{dF}{ds}-\frac{1}{\beta}\frac{dJ}{ds}\right)G_0(s',s;\tau)\end{aligned} \tag{28}$$

Because $J(s)$ satisfies Eq. (20) it is now clear that Eq. (25) is obeyed.

3. SOME SIMPLE MODELS

A. The Gaussian Model

In this model the free energy is assumed to be quadratic in s :

$$F(s) = \tfrac{1}{2}\alpha s^2 \quad . \tag{29}$$

The equilibrium probability distribution is a Gaussian

$$P_0(s) = \sqrt{\frac{\alpha\beta}{2\pi}}\, e^{-\frac{1}{2}\alpha\beta s^2} \quad , \tag{30}$$

which implies

$$\langle s^2\rangle_0 = \frac{1}{\alpha\beta} \quad . \tag{31}$$

The "equation of motion" for s is

$$\dot{s}(t) = -\mu\alpha s(t) + \xi(t) \quad . \tag{32}$$

If Eq. (32) is multiplied by s(0) and averaged, one finds

$$\frac{d}{dt}\langle s(0)\, s(t)\rangle_0 = -\mu\alpha\langle s(0)\, s(t)\rangle_0 \;, \quad (t>0) \quad . \tag{33}$$

The solution of Eq. (33) which satisfies Eq. (31) is

$$\langle s(0)\, s(t)\rangle_0 = \frac{1}{\alpha\beta} e^{-\mu\alpha t} \quad , \quad (t>0) \quad . \tag{34}$$

The calculation of J(s) is straightforward; from Eq. (21) one finds

$$J(s) = e^{\frac{1}{2}\alpha\beta s^2}\int_s^\infty \beta s' e^{-\frac{1}{2}\alpha\beta s'^2} ds' = \frac{1}{\alpha} \quad . \tag{42}$$

The Smoluchowski equation (13) thus is

$$\frac{\partial P}{\partial t} = \frac{\partial}{\partial s}\left[\left(\mu\alpha s - \frac{\dot\gamma(t)}{\alpha}\right)P\right] + \mu k_B T\frac{\partial^2 P}{\partial s^2} \quad . \tag{36}$$

From Eq. (36) a simple equation for the average $\langle s(t)\rangle_{\dot\gamma}$ may be derived by application of the obvious identity

$$\frac{d}{dt}\langle s(t)\rangle_{\dot\gamma} = \int_{-\infty}^{\infty} s\frac{\partial P}{\partial t}(s,t)\,ds \quad . \tag{37}$$

Substituting Eq. (36) into Eq. (37) one finds after partial integrations

$$\frac{d}{dt}\langle s(t)\rangle_{\dot\gamma} = -\mu\alpha\langle s(t)\rangle_{\dot\gamma} + \frac{\dot\gamma(t)}{\alpha} \quad . \tag{38}$$

As usual it is assumed that the shear rate "field" $\dot\gamma(t)$ is introduced gradually in the distant past. The solution of Eq. (38) which vanishes as $t\to-\infty$, is

$$\langle s(t)\rangle_{\dot\gamma} = \int_{-\infty}^{t}\frac{\dot\gamma(t')}{\alpha}e^{-\mu\alpha(t-t')}dt' \quad . \tag{39}$$

By means of Eq. (34) this may be rewritten as

$$\langle s(t)\rangle_{\dot\gamma} = \beta\int_0^\infty \langle s(0)\,s(\tau)\rangle_0\,\dot\gamma(t-\tau)\,d\tau \quad . \tag{40}$$

Equation (40) is nothing but the prediction of linear response theory (Eq. (5)). Thus, the Gaussian model is linear for all displacements.

B. The Boxmodel

This model is defined by

$$F(s) = \begin{cases} 0, & |s| < s_0 \\ \infty, & |s| > s_0 \end{cases} \qquad (41)$$

The model is named after the boxmodel in elementary quantum mechanics; it should not be mixed up with the boxmodel distribution of relaxation times sometimes used in rheology [9]. Since there is a maximum value of the stress, the boxmodel must exhibit shear thinning at large shear rates. To find the nonlinear viscosity, $\eta(\dot{\gamma})$, we need to determine first $J(s)$. In the present case Eq. (20) reduces to $dJ/ds = -\beta s$. The solution of this equation, which satisfies the boundary conditions $J(-s_0) = J(s_0) = 0$, is

$$J(s) = \frac{\beta}{2}(s_0^2 - s^2) \quad . \qquad (42)$$

The Φ-function of Eq. (23) thus becomes

$$\Phi(s) = \frac{\beta}{2\mu}(s_0^2 s - \tfrac{1}{3}s^3) \quad . \qquad (43)$$

For a given shear rate $\dot{\gamma}$, the stationary solution of Eq. (13) is clearly

$$P(s) = N^{-1} e^{\beta\dot{\gamma}\Phi(s)} \;, \quad (|s| < s_0) \quad , \qquad (44)$$

where N is a normalization constant, and $P(s)=0$ for $|s|>s_0$

. If the dimensionless shear rate

$$\dot{\gamma}^* = \frac{\beta^2 s_0^3}{\mu}\dot{\gamma} \tag{45}$$

is introduced, one finds from Eq. (44)

$$\langle s\rangle_{\dot{\gamma}} = s_0 \frac{\int_{-1}^{1} t e^{\dot{\gamma}^*(\frac{t}{2}-\frac{t^3}{6})} dt}{\int_{-1}^{1} e^{\dot{\gamma}^*(\frac{t}{2}-\frac{t^3}{6})} dt} \quad . \tag{46}$$

From this the nonlinear viscosity, $\eta = \langle s\rangle/\dot{\gamma}$, is readily evaluated. The result is shown in Fig. 1a and is not unlike that seen in polymeric liquids. In the linear limit one finds, by expanding Eq. (46) to first order in γ^*, the shear rate independent viscosity η_0 given by

$$\eta_0 = \frac{2\beta^2 s_0^4}{15\mu} \quad . \tag{47}$$

The equilibrium dynamics is governed by the simple diffusion equation (Eq. (11))

$$\frac{\partial P}{\partial t} = \frac{\mu}{\beta}\frac{\partial^2 P}{\partial s^2} \quad , \tag{48}$$

subject to the boundary condition

$$\frac{\partial P}{\partial s}(s=-s_0) = \frac{\partial P}{\partial s}(s=s_0) = 0 \quad . \tag{49}$$

The eigenfunctions of this problem are $\cos[n\pi(s_0-s)/(2s_0)]$ $(n=0,1,2,...)$. From this one finds by standard methods [10] that the equilibrium Green's function is given by

$$G_0(s,s';t) = \frac{1}{2s_0} + \sum_{n=1}^{\infty} \frac{1}{s_0} e^{-\omega_n t} \cos[\lambda_n(s_0-s)]\cos[\lambda_n(s_0-s')] \tag{50}$$

where

$$\lambda_n = \frac{\pi}{2s_0} n \quad , \quad \omega_n = \frac{\mu}{\beta}\lambda_n^2 \quad (n=1,2,\ldots) \quad . \tag{51}$$

It is now easy to derive an analytical expression for the frequency-dependent viscosity , $\eta(\omega)$. Since the equilibrium probability distribution is $P_0(s)=1/(2s_0)$, the autocorrelation function becomes

$$\langle s(0)\, s(t)\rangle_0 = \int_{-s_0}^{s_0} \frac{ds}{2s_0} \int_{-s_0}^{s_0} ds'\; s\; s'\; G_0(s,s';t) \quad . \tag{52}$$

From Eqs. (50) and (52) and the fluctuation-dissipation theorem (Eq. (4)), one finds

$$\eta(\omega) = \frac{32\beta s_0^2}{\pi^4} \sum_{odd\, n} \frac{1}{n^4 (i\omega + \omega_n)} \quad . \tag{53}$$

In the zero-frequency limit Eq. (53) reduces to Eq. (47) via the identity [11]

$$\frac{1}{1^6} + \frac{1}{3^6} + \frac{1}{5^6} + \cdots = \frac{\pi^6}{960} \quad . \tag{54}$$

Figure 1b shows the real part of $\eta(\omega)$. The spectrum contains infinitely many relaxation times but these are hardly visible, being completely dominated by the fundamental frequency ω_1 . In effect, $\eta(\omega)$ is almost indistinguishable from the prediction of a simple Maxwell model where Re $\eta(\omega)$ is proportional to ω^{-2} for $\omega \to \infty$.

Only in the relaxation towards equilibrium from a strongly non-equilibrium state do the higher harmonics give significant contributions. If $\langle s(t)\rangle_s$ donotes the average stress, given the value s at t=0 , one has obviously

$$\langle s(t)\rangle_s = \int_{-s_0}^{s_0} s' \, G_0(s,s';t)\, ds' \quad . \tag{55}$$

Relaxation from a state with probability distribution P(s) at t=0 is given by

$$\langle s(t)\rangle_P = \int_{-s_0}^{s_0} \langle s(t)\rangle_{s'} P(s')\, ds' \quad . \tag{56}$$

Two well-known examples are stress relaxation after cessation of a steady flow, and stress relaxation after a sudden shear displacement starting from equilibrium. In the first case, the probability distribution P(s) is given by Eq. (44) at t=0 . In the latter case, after the sudden shear displacement given by $\dot{\gamma}(t)=\gamma_0\delta(t)$, it is possible to show that P(s) is given by

$$P(s) = \frac{2\lambda s_0}{[(1+\lambda)s_0-(1-\lambda)s]^2} \quad , \; \lambda = e^{-\beta\gamma_0 s_0} \quad . \tag{57}$$

In both cases P(s) is strongly peaked around $s=s_0$. From Eq. (50) we find

$$\langle s(t)\rangle_s = \frac{2}{s_0} \sum_{odd\; n} \frac{1}{\lambda_n^2} e^{-\omega_n t} \cos[\lambda_n(s_0-s)] \quad . \tag{58}$$

This implies

$$s-\langle s(t)\rangle_s = \frac{2}{s_0}\sum_{odd\,n}\frac{1}{\lambda_n^2}\left(1-e^{-\omega_n t}\right)\cos[\lambda_n(s_0-s)] \quad . \tag{59}$$

The interpretation of Eq. (59) is as follows. Whenever t is so large that $\lambda_n(s_0-s)<<1$ for all n with $\omega_n t<1$, the cosine factor may be ignored all together, and below a limiting $n=p$ given by $\omega_p t=1$, the exponential may be expanded to first order. Writing $p=at^{-1/2}$, one has

$$s-\langle s(t)\rangle_s \simeq Const.\int_1^p t\,dn + Const.\int_p^\infty \frac{dn}{n^2} \propto t^{\frac{1}{2}} \quad . \tag{60}$$

At **very** short times the quantity $s-\langle s(t)\rangle_s$ is exponentially close to zero. Then comes according to Eq. (60) a range of times where this quantity increases like $t^{1/2}$, and finally it converges to s. A similar result applies to relaxation from a state with probability distribution $P(s,t=0)$, since

$$\langle s(0)\rangle_P-\langle s(t)\rangle_P = \int_{-s_0}^{s_0}[s-\langle s(t)\rangle_s]\,P(s,t=0)\,ds \quad . \tag{61}$$

If the width of $P(s,t=0)$, Δs, is defined by the integral of P from $s_0-\Delta s$ to s_0 being $1/2$, it is not hard to show that

$$\langle s(0)\rangle_P-\langle s(t)\rangle_P = \begin{cases} \simeq 0 & , \ t\ll t_1 \\ \propto t^{1/2} & , \ t_1\ll t\ll t_2 \\ \simeq \langle s(0)\rangle & , \ t_2\ll t \end{cases} \tag{62}$$

where

$$t_1 = \frac{\beta}{\mu}(\Delta s)^2 \quad , \quad t_2 = \frac{\beta}{\mu} s_0{}^2 \quad . \tag{63}$$

In the case of relaxation after a sudden cessation of a steady shear flow, Δs is given by (compare Eq. (44))

$$(\Delta s)^2 \simeq \frac{\mu}{s_0 \beta^2} \frac{1}{\dot{\gamma}} \quad . \tag{64}$$

In the case of a sudden large shear displacement, γ_0, Δs is given by (compare Eq. (57))

$$\Delta s \simeq 2 s_0 e^{-\beta \gamma_0 s_0} \quad . \tag{65}$$

Note that, in both cases, the non-Debye character of the relaxation is apparent only because we have considered the quantity $<s(0)>_p - <s(t)>_p$. If one looks at just $<s(t)>_p$, this quantity would be hard to distinguish from a simple exponential decay in time.

C. The Cosine Hyperbolic Model

A phenomenological model for stress relaxation was proposed by Tobolsky and coworkers in the 1940´s [3-5]. The model is a Maxwell element consisting of a Hooke´s law spring and a non-Newtonian dashpot in series, the viscosity of the dashpot obeying the Eyring viscosity equation. The model leads to

$$\dot{\gamma} = A\,\dot{s} + B\sinh\left(\frac{s}{s_0}\right) \quad , \qquad (66)$$

where A , B , and s_0 are constants. Equation (66) reproduces Eyring's viscosity equation and predicts a logarithmic stress relaxation at constant extension: At large s(t=0) one has approximately $\dot{s}$=-Const. exp(s/s_0) which implies at intermediate times [4]

$$s(t) \cong \alpha - \beta \ln(t) \quad . \qquad (67)$$

Both predictions of Eq. (66) mimic experiment on typical polymeric liquids. The model, however, does not take into account thermal fluctuations. The formalism developed in Sec. 2 allows one to construct a model based on Eq. (66) which is consistent with statistical mechanics. Since relaxation towards equilibrium is governed by $\dot{s}$=-Const. sinh(s/s_0) , the obvious choice for F(s) is

$$F(s) = f_0 \cosh\left(\frac{s}{s_0}\right) \quad . \qquad (68)$$

The Langevin equation corresponding to Eq. (68) is then

$$\dot{s} = -\mu \frac{f_0}{s_0} \sinh\left(\frac{s}{s_0}\right) + \xi(t) \quad . \qquad (69)$$

We now proceed to investigate this model, being particularly interested in to which extent it reproduces the predictions of Eq. (66).

First the nonlinear steady state viscosity is considered. At low temperatures it is possible to derive an analytical expression for the viscosity. The derivation is given here for a general F(s) . If <s> denotes the average of s during a steady shear flow, one has from Eq. (12) at low temperatures where fluctuations are small

$$\dot{\gamma} J(\langle s \rangle) = \mu \frac{dF}{ds}(\langle s \rangle) \; . \tag{70}$$

The quantity <s> is a function of $\dot{\gamma}$. If the derivative of this function is denoted by <s>', Eq.(70) implies by differentiation

$$J + \dot{\gamma} \frac{dJ}{ds} \langle s \rangle' = \mu \frac{d^2F}{ds^2} \langle s \rangle' \; . \tag{71}$$

Combining this with Eq. (20) leads to

$$J = \langle s \rangle' \left[\mu \frac{d^2F}{ds^2} - \beta \dot{\gamma} \left(\frac{dF}{ds} J - \langle s \rangle \right) \right] \; . \tag{72}$$

For $\beta \to \infty$ the term in the inner paranthesis must vanish. In conjuction with Eq. (70) one finds

$$\langle s \rangle = \frac{\mu}{\dot{\gamma}} \left(\frac{dF}{ds} (\langle s \rangle) \right)^2 \; , \tag{73}$$

or for the viscosity

$$\eta = \frac{1}{\mu} \left(\frac{\langle s \rangle}{\frac{dF}{ds}(\langle s \rangle)} \right)^2 \; . \tag{74}$$

For the cosh-model we thus have

$$\eta = \eta_0 \left(\frac{\langle s \rangle / s_o}{\sinh(\langle s \rangle / s_0)} \right)^2 \qquad (75)$$

where

$$\eta_0 = \frac{1}{\mu} \frac{s_0^4}{f_0^2} \quad . \qquad (76)$$

is the linear viscosity. Perhaps surprisingly, one does **not** recover the Eyring viscosity expression

$$\eta = \eta_0 \frac{\langle s \rangle / s_0}{\sinh(\langle s \rangle / s_0)} \quad . \qquad (77)$$

But, as is clear from Fig. 2, the cosh model viscosity may be fitted reasonably well by Eq. (77).

Consider now relaxation towards equilibrium from a non-equilibrium state s . At low temperatures $J(s)$ may be found explicitly from Eq. (21) which for $\beta \to \infty$ reduces to

$$J(s) = \frac{s_0^2}{f_0} \frac{s/s_0}{\sinh(s/s_0)} \quad . \qquad (78)$$

When substituted into Eq. (12) this leads to, in the zero noise limit,

$$\dot{\gamma} = \frac{f_0}{s_0^2} \frac{\sinh(s/s_0)}{s/s_0} \dot{s} + \mu \frac{f_0^2}{s_0^3} \frac{\sinh^2(s/s_0)}{s/s_0} \quad . \qquad (79)$$

This looks nothing like Eq. (66). But again we find the nonlinear viscosity given by Eq. (75). Furthermore, for constant elongation, s relaxes according to

$$\dot{s} = -\mu \frac{f_0}{s_0} \sinh\left(\frac{s}{s_0}\right) \quad , \tag{80}$$

as expected from Eq. (69).

A final correspondance of the cosh-model to Tobolsky's phenomenological model is the frequency-dependent viscosity. As is the case for any differential constitutive relation [1], Eq. (66) reduces to the Maxwell model in the linear limit. In the cosh-model one might expect a more interesting frequency-dependence, at least at high temperatures where the sinh-factor of Eq. (69) cannot be replaced by a s^2-factor. But it turns out that, even at high temperatures, the autocorrelation function $\langle s(0)s(t)\rangle_0$ is actually very close to an exponential (which corresponds to the Maxwell model). What happens is that the logarithmic s-relaxation of Eq. (80), even at quite short times, is killed by the s-diffusion due to the noise term.

D. The Power-law Model

As a final example we briefly discuss the power-law model where

$$F(s) = f_0 \left|\frac{s}{s_0}\right|^n , \quad (n>0) \quad . \tag{81}$$

The fact that $F(s)$ is non-analytical at $s=0$ is insignificant. The case $n=2$ is the Gaussian model and the $n\to\infty$

limit is the box model.

The power-law model makes sense only for $n > 3/2$. To prove this, the low temperature limit is considered. For $\beta \to \infty$ $J(s)$ may be evaluated asymptotically from Eq. (21). Writing $s'^n - s^n = ns^{n-1}(s'-s)$, one gets

$$J(s) = \beta \int_s^\infty ds' s' e^{-\beta \frac{f_0}{s_0^n} n s^{n-1}(s'-s)} \propto s\,(s^{n-1})^{-1} = s^{2-n} \quad . \tag{82}$$

Thus, whenever $\dot{\gamma}$ is positive there is a "force" proportional to s^{2-n} working to increase s. The "restoring force" from the potential varies as s^{n-1}. This force must dominate at large s in order to avoid s running off to infinity, thus $n-1 > 2-n$ or $n > 3/2$. Mathematically, there is no normalizable stationary state whenever $n < 3/2$. The border case $n = 3/2$ leads to a model which is well-defined up to a certain shear rate, above which s runs off to infinity. This corresponds to there being a phase transition at a definite shear rate to a state of infinitely high viscosity.

To estimate the viscosity we use Eq. (73) which implies

$$\dot{\gamma} \propto \frac{1}{s}\left(\frac{dF}{ds}\right)^2 \propto s^{2n-3} \quad . \tag{83}$$

This implies

$$\eta = s/\dot{\gamma} \propto \dot{\gamma}^{\frac{4-2n}{2n-3}} \quad . \tag{84}$$

For $3/2 < n < 2$ the model exhibits **shear thickening** , whereas for $2 < n$ there is **shear thinning.** The case $n = 2$ gives a shear rate independent viscosity, as shown already.

4. DISCUSSION

The model of stress fluctuations considered in the present paper involves several non-trivial assumptions. The liquid is regarded as divided into "regions", and correlations between stress fluctuations of different regions are ignored completely. This is an assumption which is made for simplicity, but which may very well be unrealistic since elastic forces are long ranged. The region picture becomes even harder to justify when a shear flow is considered. Such a flow deforms the regions and the picture can only be maintained whenever the longest correlation time is smaller than the inverse shear rate.

The main result of the paper is the proof that, if equilibrium stress fluctuations follow a Langevin equation, there is only one possible stress dynamics in non-equilibrium which is consistent with the principle of virtual work. Not only is the non-linear response uniquely determined, but this is true also for the stress fluctuations in non-equilibrium. Crucial to this theorem is the assumption of a Langevin dynamics for the stress. This is a phenomenological postulate, but it should be emphasized that Langevin dynamics is the "canonical" guess if one is to discuss dynamics purely from a knowledge of equilibrium statistical mechanics. There simply is no other way of estimating the dynamics from a knowledge of only the equilibrium free energy $F(s)$. But, of course, this does not garanty that the Langevin equation leads to correct results.

The assumption of a linear coupling of the shear rate "field" in Eq. (12) is not essential. In fact, it is easy to show that the principle of virtual work implies the coupling **must** be linear. This is because the shear rate does not occur in Eq. (16).

The formalism developed may be generalized by replacing the term "stress" (which is the transverse momentum current) by any other current. One thus has a method for predicting the nonlinear response from a knowledge of equilibrium current fluctuations. The considering of currents as independent degrees of freedom has become popular in recent years, being the basis of extended irreversible thermodynamics [12]. The approach of Sec. 2 may be regarded as a statistical mechanical counterpart to extended irreversible thermodynamics.

To illustrate the formalism a few simple models were studied in Sec. 3. The Gaussian model leads to an exactly linear response even at large shear rates, reducing simply to the standard Maxwell model. The fact that the Gaussian model is linear is quite satisfactory, since a similar result is valid in ordinary statistical mechanics. Here, strictly Gaussian equilibrium fluctuations of, e. g., the magnetization, implies a field independent susceptibility.

A more interesting model is the boxmodel. It predicts a nonlinear viscosity (simply because there is a maximum possible stress), and a spectrum of relaxation times. In equilibrium this spectrum is not really visible, however; the autocorrelation function $\langle s(0)s(t)\rangle_0$ is approximately an exponential, leading to almost a Maxwell type frequency-dependence of the viscosity. Only in the relaxation from a strongly non-equilibrium state does the spectrum play any significant role, and even here the lowest eigenfrequency dominates the overall picture.

The cosh-model was constructed to mimic Tobolsky's phenomenological model for stress relaxation. But although the zero noise relaxation equation is equal to Tobolsky's (Eq. (66)), the predictions of the model are not identical to those of Eq. (66). This is an excellent illustration of a point made by van Kampen [7] that, by adding a noise term to a phenomenological model of the form $\dot{s}=f(s)$, some of the properties of the equation are lost because of the noise. In the case of the cosh-model, the properties are retained in a qualitative sense, though. Thus, there is an Eyring-like viscosity in the model (Fig. 2), and the frequency-dependence of the phenomenological model and the model of Eq. (69) are almost equal. The latter point may seem surprising, given the fact that the cosh-model leads to a logarithmic relaxation towards zero in the zero noise limit (which, as is well-known, corresponds to a spectrum of relaxation times varying like τ^{-1}). However, just as in the boxmodel, the spectrum is not significant in equilibrium where the noise term completely dominates the autocorrelation function, resulting in an almost exponential decay.

The cosh-model corresponds to an exponentially increasing free energy $F(s)$. The case of $F(s)$ increasing following a power-law was also considered in Sec. 3. (The case of a logarithmically increasing $F(s)$ leads to an inconsistent model where s runs off to infinity whenever $\dot{\gamma}=0$.) The power-law model is consistent only for $n > 3/2$, $n = 3/2$ being a border case where the model makes sense for not too large shear rates. Whenever $3/2 < n < 2$ the model exhibits shear thickening, whereas $2 < n$ corresponds to the experimentally more common case of shear thinning. The case $n = 2$ is the Gaussian model, and the limit $n \to \infty$ is the boxmodel. A closer analysis than given in Sec. 3 reveals that the shear thickening in the case $3/2 < n < 2$ is a consequence of one **not** having $J(s) \to 0$ as $|s| \to \infty$ (while for $2 < n$ $J(s) \to 0$ as $|s| \to \infty$). Since $J(s)$ may be interpreted as an s-dependent infinite frequency shear modulus, the study of the power-law model leads to a perhaps new view on the origin of nonlinearity: Nonlinearity may be viewed as a consequence of a stress dependent G_∞ . The case when G_∞ increases with shear rate corresponds to shear thickening while a decreasing G_∞ (i. e., softening) corresponds to shear thinning. The Gaussian linear case $n = 2$ corresponds to a shear rate independent G_∞ . -- A final thing to be noted about the power-law model is that, in the zero noise limit, this model has a power-law time dependence of the stress relaxation, as is easy to show.

The predictions of the models are approximately on the level of differential constitutive relations: these have realistic nonlinear steady state viscosities but only a simple Maxwell frequency-dependence of the viscosity [1]. Compared to differential constitutive relations, the presently considered models have the advantage of being consistent with statistical mechanics. Thus, the method presented may be regarded as a means of modifying differential constitutive relations to include thermal fluctuations. The modification, however, is nontrivial in the sense that the differential consitutive relation is **not** recovered exactly in the low temperature limit, as shown in detail for the cosh model.

Several important features of polymeric liquids are not mimiced by the models of Sec. 3. The predicted almost single relaxation time frequency-dependence of the viscosity is very far from that observed in polymeric liquids. Another important point which is not captured by the models is the fact that, experimentally, non-linearity sets in at a shear rate about equal to the frequency marking the onset of frequency-dependence of the viscosity [1,13]. Finally, the temperature-dependence of the viscosity is weak and there is a well-defined viscosity in the zero temperature limit. While this last point could be handled by assuming the mobility μ is temperature-dependent, the two other points are quite serious, indeed. A further, perhaps more esoteric objection, is the fact that in the present model there is time-reversibility in a steady shear flow: The steady flow Langevin equation obeys detailed balance for a suitably chosen energy function (compare Eq. (26)). In a real flow one expects a genuine violation of time-reversibility.

In conclusion, the types of models studied in Sec. 3 are not satisfactory as models of reality. To arrive at more realistic models one has to consider several stress coordinates interacting with each others, for instance by taking into account the spatial variation of the stress. This leads to a field theory in which the free energy is a functional of the stress field. If this function has several minima, the Langevin dynamics gives thermally activated relxation times (just like in the description of chemical reactions), and thereby more realistic temperature- and frequency-dependences. Also, it may be shown that in a model with more than one degree of freedom there is genuine time-irreversibility in any shear flow. Work along these lines is in progress.

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FIGURE CAPTIONS

Fig. 1: Boxmodel predictions for the viscosity. Fig. 1a shows a log-log plot of the viscosity relative to the linear viscosity, as function of the dimensionless shear rate given by Eq. (45). At large shear rates the viscosity varies as one over the shear rate; this is a consequence of there being a maximum possible stress in the model. Fig. 1b shows a log-log plot of the real part of the frequency-dependent viscosity relative to the zero frequency viscostity, as function of the dimensionless frequency ω/ω_1 . Despite the fact that the model has infinitely many relaxation times, the longest relaxation time dominates the frequency-dependence completely. In effect, the frequency-dependence is almost indistinguishable from that of a standard Maxwell model where the real part of the viscosity varies as ω^{-2} at large frequencies.

Fig. 2: Log-log plot of the steady state nonlinear viscosity of the cosh-model. The nonlinear viscosity is shown relative to the linear viscosity, as function of the dimensionless shear rate $\dot{\gamma}/(s_0/\eta_0)$. The nonlinear viscosity of the cosh-model is not identical to the Eyring viscosity of Eq. (77) (marked by dots).

Fig 1a

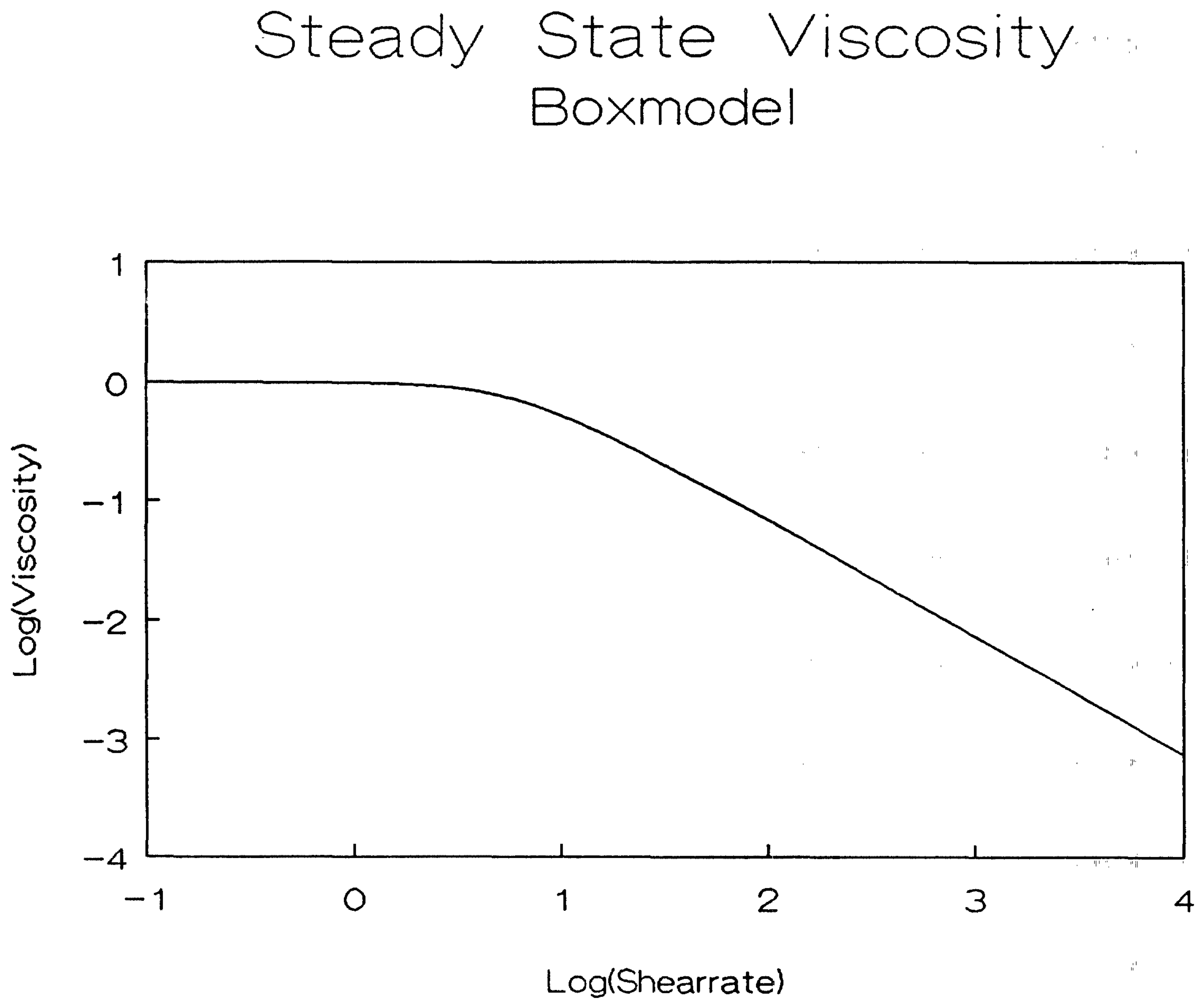

Steady State Viscosity
Boxmodel
Log(Viscosity)
1
0
−1
−2
−3
−4
−1
0
1
2
3
4
Log(Shearrate)

Fig 1k

Frequency–dependent Viscosity

Boxmodel

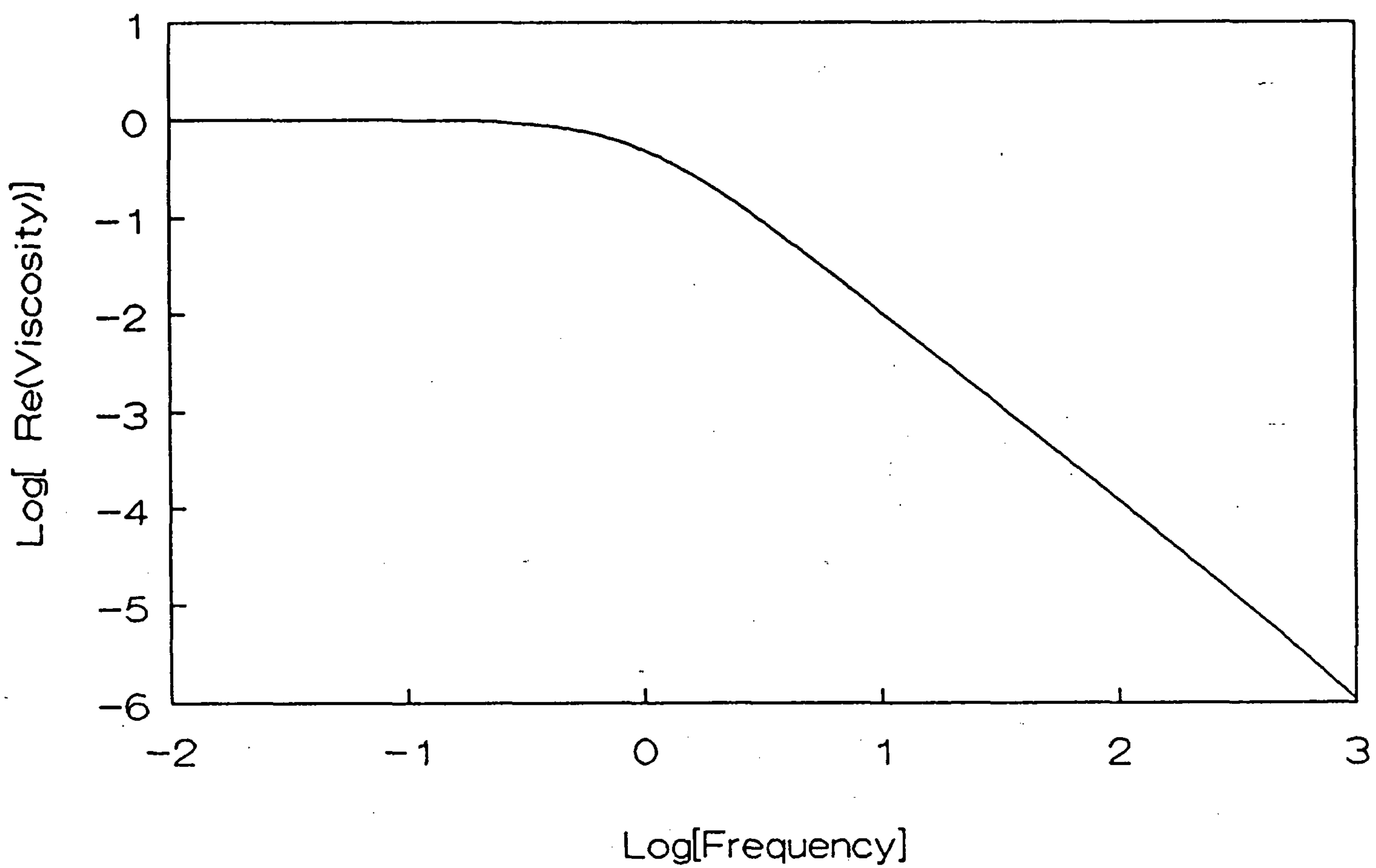
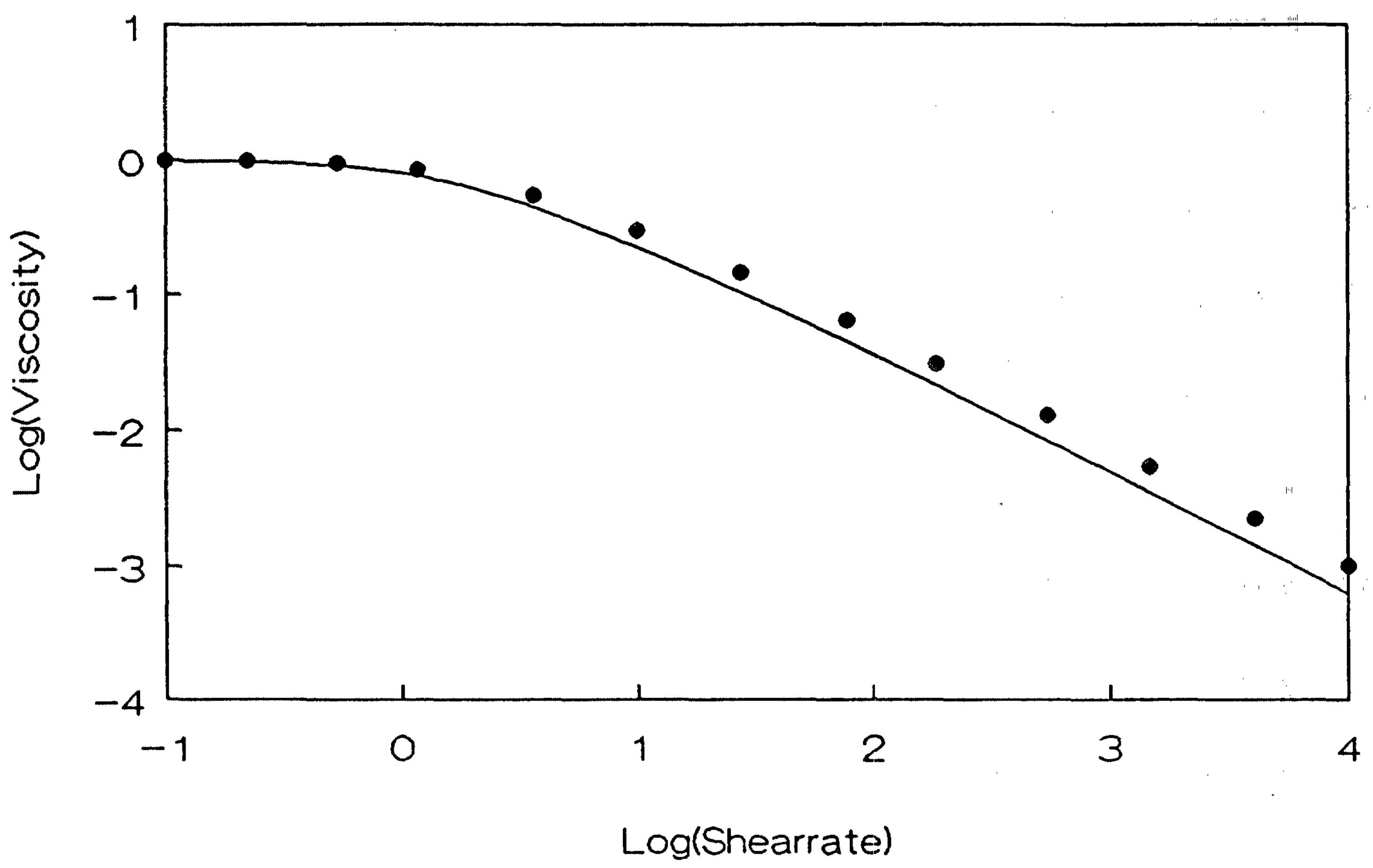


Fig 2

Steady State Viscosity
Cosh Model



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Vejleder: Bent Sørensen.

74/84 "MINIATURISERING AF MIKROELEKTRONIK" - om videnskabeliggjort teknologi og nytten af at lære fysik.
Projektrapport af: Bodil Harder og Linda Szkotak Jensen.
Vejledere: Jens Højgaard Jensen og Bent C. Jørgensen.

75/84 "MATEMATIKUNDERVISNINGEN I FREMTIDENS GYMNASIUM"
- Case: Lineær programmering.
Projektrapport af: Morten Blomhøj, Klavs Frisdahl og Frank Mølgaard Olsen.
Vejledere: Mogens Brun Heefelt og Jens Bjørneboe.

76/84 "KERNEKRAFT I DANMARK?" - Et høringssvar indkaldt af miljøministeriet, med kritik af miljøstyrelsens rapporter af 15. marts 1984.
ENERGY SERIES No. 1o
Af: Niels Boye Olsen og Bent Sørensen.

77/84 "POLITISKE INDEKS - FUP ELLER FAKTA?"
Opinionsundersøgelser belyst ved statistiske modeller.
Projektrapport af: Svend Åge Houmann, Keld Nielsen og Susanne Stender.
Vejledere: Jørgen Larsen og Jens Bjørneboe.

78/84 "JÆVNSTRØMSLEDNINGSEVNE OG GITTERSTRUKTUT I AMORFT GERMANIUM".
Specialrapport af: Hans Hedal, Frank C. Ludvigsen og Finn C. Physant.
Vejleder: Niels Boye Olsen.

79/84 "MATEMATIK OG ALMENDANNELSE".
Projektrapport af: Henrik Coster, Mikael Wennerberg Johansen, Povl Kattler, Birgitte Lydholm og Morten Overgaard Nielsen.
Vejleder: Bernhelm Booss.

80/84 "KURSUSMATERIALE TIL MATEMATIK B".
Af: Mogens Brun Heefelt.

81/84 "FREKVENSAFHÆNGIG LEDNINGSEVNE I AMORFT GERMANIUM".
Specialerapport af: Jørgen Wind Petersen og Jan Christensen.
Vejleder: Niels Boye Olsen.

82/84 "MATEMATIK - OG FYSIKUNDERVISNINGEN I DET AUTOMATISEREDE SAMFUND".
Rapport fra et seminar afholdt i Hvidovre 25-27 april 1983.
Red.: Jens Højgaard Jensen, Bent C. Jørgensen og Mogens Niss.

83/84 "ON THE QUANTIFICATION OF SECURITY":
PEACE RESEARCH SERIES NO. 1
Af: Bent Sørensen
nr. 83 er p.t. udgået

84/84 "NOGLE ARTIKLER OM MATEMATIK, FYSIK OG ALMENDANNELSE".
Af: Jens Højgaard Jensen, Mogens Niss m. fl.

85/84 "CENTRIFUGALREGULATORER OG MATEMATIK".
Specialerapport af: Per Hedegård Andersen, Carsten Holst-Jensen, Else Marie Pedersen og Erling Møller Pedersen.
Vejleder: Stig Andur Pedersen.

86/84 "SECURITY IMPLICATIONS OF ALTERNATIVE DEFENSE OPTIONS FOR WESTERN EUROPE".
PEACE RESEARCH SERIES NO. 2
Af: Bent Sørensen.

87/84 "A SIMPLE MODEL OF AC HOPPING CONDUCTIVITY IN DISORDERED SOLIDS".
Af: Jeppe C. Dyre.

88/84 "RISE, FALL AND RESURRECTION OF INFINITESIMALS".
Af: Detlef Laugwitz.

89/84 "FJERNVARMEOPTIMERING".
Af: Bjarne Lillethorup og Jacob Mørch Pedersen.

90/84 "ENERGI I 1.G - EN TEORI FOR TILRETTELÆGGELSE".
Af: Albert Chr. Paulsen.

91/85 "KVANTETEORI FOR GYMNASIET".
1. Lærervejledning
Projektrapport af: Biger Lundgren, Henning Sten Hansen og John Johansson.
Vejleder: Torsten Meyer.

92/85 "KVANTETEORI FOR GYMNASIET".
2. Materiale
Projektrapport af: Biger Lundgren, Henning Sten Hansen og John Johansson.
Vejleder: Torsten Meyer.

93/85 "THE SEMIOTICS OF QUANTUM - NON - LOCALITY".
Af: Peder Voetmann Christiansen.

94/85 "TREENIGHEDEN BOURBAKI - generalen, matematikeren og ånden".
Projektrapport af: Morten Blomhøj, Klavs Frisdahl og Frank M. Olsen.
Vejleder: Mogens Niss.

95/85 "AN ALTERNATIV DEFENSE PLAN FOR WESTERN EUROPE".
PEACE RESEARCH SERIES NO. 3
Af: Bent Sørensen

96/85 "ASPEKTER VED KRAFTVARMEFORSYNING".
Af: Bjarne Lilletorup.
Vejleder: Bent Sørensen.

97/85 "ON THE PHYSICS OF A.C. HOPPING CONDUCTIVITY".
Af: Jeppe C. Dyre.

98/85 "VALGMULIGHEDER I INFORMATIONSALDEREN".
Af: Bent Sørensen.

99/85 "Der er langt fra Q til R".
Projektrapport af: Niels Jørgensen og Mikael Klintorp.
Vejleder: Stig Andur Pedersen.

100/85 "TALSYSTEMETS OPBYGNING".
Af: Mogens Niss.

101/85 "EXTENDED MOMENTUM THEORY FOR WINDMILLS IN PERTURBATIVE FORM".
Af: Ganesh Sengupta.

102/85 OPSTILLING OG ANALYSE AF MATEMATISKE MODELLER, BELYST VED MODELLER OVER KØERS FODEROPTAGELSE OG - OMSÆTNING".
Projektrapport af: Lis Eilertzen, Kirsten Habekost, Lill Røn og Susanne Stender.
Vejleder: Klaus Grünbaum.

103/85 "ØDSLE KOLDKRIGERE OG VIDENSKABENS LYSE IDEER".
Projektrapport af: Niels Ole Dam og Kurt Jensen.
Vejleder: Bent Sørensen.

104/85 "ANALOGREGNEMASKINEN OG LORENZLIGNINGER".
Af: Jens Jæger.

105/85 "THE FREQUENCY DEPENDENCE OF THE SPECIFIC HEAT AF THE GLASS REANSITION".
Af: Tage Christensen.

"A SIMPLE MODEL AF AC HOPPING CONDUCTIVITY".
Af: Jeppe C. Dyre.
Contributions to the Third International Conference on the Structure of Non - Crystalline Materials held in Grenoble July 1985.

106/85 "QUANTUM THEORY OF EXTENDED PARTICLES".
Af: Bent Sørensen.

107/85 "EN MYG GØR INGEN EPIDEMI".
- flodblindhed som eksempel på matematisk modellering af et epidemiologisk problem.
Projektrapport af: Per Hedegård Andersen, Lars Boye, CarstenHolst Jensen, Else Marie Pedersen og Erling Møller Pedersen.
Vejleder: Jesper Larsen.

108/85 "APPLICATIONS AND MODELLING IN THE MATEMATICS CURRICULUM" - state and trends -
Af: Mogens Niss.

109/85 "COX I STUDIETIDEN" - Cox's regressionsmodel anvendt på studenteroplysninger fra RUC.
Projektrapport af: Mikael Wennerberg Johansen, Poul Katler og Torben J. Andreasen.
Vejleder: Jørgen Larsen.

110/85 "PLANNING FOR SECURITY".
Af: Bent Sørensen

111/85 JORDEN RUNDT PÅ FLADE KORT".
Projektrapport af: Birgit Andresen, Beatriz Quinones og Jimmy Staal.
Vejleder: Mogens Niss.

112/85 "VIDENSKABELIGGØRELSE AF DANSK TEKNOLOGISK INNOVATION FREM TIL 1950 - BELYST VED EKSEMPLER".
Projektrapport af: Erik Odgaard Gade, Hans Hedal, Frank C. Ludvigsen, Annette Post Nielsen og Finn Physant.
Vejleder: Claus Bryld og Bent C. Jørgensen.

113/85 "DESUSPENSION OF SPLITTING ELLIPTIC SYMBOLS 11".
Af: Bernhelm Booss og Krzysztof Wojciechowski.

114/85 "ANVENDELSE AF GRAFISKE METODER TIL ANALYSE AF KONTIGENSTABELLER".
Projektrapport af: Lone Biilmann, Ole R. Jensen og Arne-Lise von Moos.
Vejleder: Jørgen Larsen.

115/85 "MATEMATIKKENS UDVIKLING OP TIL RENÆSSANCEN".
Af: Mogens Niss.

116/85 "A PHENOMENOLOGICAL MODEL FOR THE MEYER-NELDEL RULE".
Af: Jeppe C. Dyre.

117/85 "KRAFT & FJERNVARMEOPTIMERING"
Af: Jacob Mørch Pedersen.
Vejleder: Bent Sørensen

118/85 "TILFÆLDIGHEDEN OG NØDVENDIGHEDEN IFØLGE PEIRCE OG FYSIKKEN".
Af: Peder Voetmann Christiansen

119/86 "DET ER GANSKE VIST - - EUKLIDS FEMTE POSTULAT KUNNE NOK SKABE RØRE I ANDEDAMMEN".
Af: Iben Maj Christiansen
Vejleder: Mogens Niss.

120/86 "ET ANTAL STATISTISKE STANDARDMODELLER".
Af: Jørgen Larsen

121/86 "SIMULATION I KONTINUERT TID".
Af: Peder Voetmann Christiansen.

122/86 "ON THE MECHANISM OF GLASS IONIC CONDUCTIVITY".
Af: Jeppe C. Dyre.

123/86 "GYMNASIEFYSIKKEN OG DEN STORE VERDEN".
Fysiklærerforeningen, IMFUFA, RUC.

124/86 "OPGAVESAMLING I MATEMATIK".
Samtlige opgaver stillet i tiden 1974-jan. 1986.

125/86 "UVBY,β - systemet - en effektiv fotometrisk spektralklassifikation af B-,A- og F-stjerner".
Projektrapport af: Birger Lundgren.

126/86 "OM UDVIKLINGEN AF DEN SPECIELLE RELATIVITETSTEORI".
Projektrapport af: Lise Odgaard & Linda Szkotak Jensen
Vejledere: Karin Beyer & Stig Andur Pedersen.

127/86 "GALOIS' BIDRAG TIL UDVIKLINGEN AF DEN ABSTRAKTE ALGEBRA".
Projektrapport af: Pernille Sand, Heine Larsen & Lars Frandsen.
Vejleder: Mogens Niss.

128/86 "SMÅKRYB" - om ikke-standard analyse.
Projektrapport af: Niels Jørgensen & Mikael Klintorp.
Vejleder: Jeppe Dyre.

129/86 "PHYSICS IN SOCIETY"
Lecture Notes 1983 (1986)
Af: Bent Sørensen

130/86 "Studies in Wind Power"
Af: Bent Sørensen

131/86 "FYSIK OG SAMFUND" - Et integreret fysik/historieprojekt om naturanskuelsens historiske udvikling og dens samfundsmæssige betingethed.
Projektrapport af: Jakob Heckscher, Søren Brønd, Andy Wierød.
Vejledere: Jens Høyrup, Jørgen Vogelius, Jens Højgaard Jensen.

132/86 "FYSIK OG DANNELSE"
Projektrapport af: Søren Brønd, Andy Wierød.
Vejledere: Karin Beyer, Jørgen Vogelius.

133/86 "CHERNOBYL ACCIDENT: ASSESSING THE DATA. ENERGY SERIES NO. 15.
AF: Bent Sørensen.

134/87 "THE D.C. AND THE A.C. ELECTRICAL TRANSPORT IN AsSeTe SYSTEM"
Authors: M.B.El-Den, N.B.Olsen, Ib Høst Pedersen, Petr Viščor

135/87 "INTUITIONISTISK MATEMATIKS METODER OG ERKENDELSES-TEORETISKE FORUDSÆTNINGER"
MASTEMATIKSPECIALE: Claus Larsen
Vejledere: Anton Jensen og Stig Andur Pedersen

136/87 "Mystisk og naturlig filosofi: En skitse af kristendommens første og andet møde med græsk filosofi"
Projektrapport af Frank Colding Ludvigsen
Vejledere: Historie: Ib Thiersen
Fysik: Jens Højgaard Jensen

137/87 "HOPMODELLER FOR ELEKTRISK LEDNING I UORDNEDE FASTE STOFFER" - Resume af licentiatafhandling
Af: Jeppe Dyre
Vejledere: Niels Boye Olsen og
Peder Voetmann Christiansen.

138/87 "JOSEPHSON EFFECT AND CIRCLE MAP."
Paper presented at The International Workshop on Teaching Nonlinear Phenomena at Universities and Schools, "Chaos in Education". Balaton, Hungary, 26 April-2 May 1987.

By: Peder Voetmann Christiansen

139/87 "Machbarkeit nichtbeherrschbarer Technik durch Fortschritte in der Erkennbarkeit der Natur"

Af: Bernhelm Booss-Bavnbek
Martin Bohle-Carbonell

140/87 "ON THE TOPOLOGY OF SPACES OF HOLOMORPHIC MAPS"

By: Jens Gravesen

141/87 "RADIOMETERS UDVIKLING AF BLODGASAPPARATUR - ET TEKNOLOGIHISTORISK PROJEKT"

Projektrapport af Finn C. Physant

Vejleder: Ib Thiersen

142/87 "The Calderón Projektor for Operators With Splitting Elliptic Symbols"

by: Bernhelm Booss-Bavnbek og
Krzysztof P. Wojciechowski

143/87 "Kursusmateriale til Matematik på NAT-BAS"

af: Mogens Brun Heefelt

144/87 "Context and Non-Locality - A Peircean Approach
Paper presented at the Symposium on the Foundations of Modern Physics The Copenhagen Interpretation 60 Years after the Como Lecture. Joensuu, Finland, 6 - 8 august 1987.

By: Peder Voetmann Christiansen

145/87 "AIMS AND SCOPE OF APPLICATIONS AND MODELLING IN MATHEMATICS CURRICULA"
Manuscript of a plenary lecture delivered at ICMTA 3, Kassel, FRG 8.-11.9.1987

By: Mogens Niss

146/87 "BESTEMMELSE AF BULKRESISTIVITETEN I SILICIUM"
- en ny frekvensbaseret målemetode.

Fysikspeciale af Jan Vedde

Vejledere: Niels Boye Olsen & Petr Viščor

147/87 "Rapport om BIS på NAT-BAS"
redigeret af: Mogens Brun Heefelt

148/87 "Naturvidenskabsundervisning med Samfundsperspektiv"

af: Peter Colding-Jørgensen DLH
Albert Chr. Paulsen

149/87 "In-Situ Measurements of the density of amorphous germanium prepared in ultra high vacuum"

by: Petr Viščor

150/87 "Structure and the Existence of the first sharp diffraction peak in amorphous germanium prepared in UHV and measured in-situ"

by: Petr Viščor

151/87 "DYNAMISK PROGRAMMERING"
Matematikprojekt af:
Birgit Andresen, Keld Nielsen og Jimmy Staal

Vejleder: Mogens Niss

152/87 "PSEUDO-DIFFERENTIAL PROJECTIONS AND THE TOPOLOGY OF CERTAIN SPACES OF ELLIPTIC BOUNDARY VALUE PROBLEMS"

by: Bernhelm Booss-Bavnbek
Krzysztof P. Wojciechowski

153/88 "HALVLEDERTEKNOLOGIENS UDVIKLING MELLEM MILITÆRE OG CIVILE KRÆFTER"

Et eksempel på humanistisk teknologihistorie
Historiespeciale

Af: Hans Hedal

Vejleder: Ib Thiersen

154/88 "MASTER EQUATION APPROACH TO VISCOUS LIQUIDS AND THE GLASS TRANSITION"

By: Jeppe Dyre

155/88 "A NOTE ON THE ACTION OF THE POISSON SOLUTION OPERATOR TO THE DIRICHLET PROBLEM FOR A FORMALLY SELFADJOINT DIFFERENTIAL OPERATOR"

by: Michael Pedersen

156/88 "THE RANDOM FREE ENERGY BARRIER MODEL FOR AC CONDUCTION IN DISORDERED SOLIDS"

by: Jeppe C. Dyre

157/88 " STABILIZATION OF PARTIAL DIFFERENTIAL EQUATIONS BY FINITE DIMENSIONAL BOUNDARY FEEDBACK CONTROL: A pseudo-differential approach."

by: Michael Pedersen

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159/88 "STUDIES IN SOLAR ENERGY"

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160/88 "LOOP GROUPS AND INSTANTONS IN DIMENSION TWO"

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Dirichlet feedback control problems"

by: Michael Pedersen

162/88 "PIGER & FYSIK - OG MEGET MERE"

AF: Karin Beyer, Sussanne Blegaa, Birthe Olsen, Jette Reich , Mette Vedelsby

163/88 "EN MATEMATISK MODEL TIL BESTEMMELSE AF PERMEABILITETEN FOR BLOD-NETHINDE-BARRIEREN"

Af: Finn Langberg, Michael Jarden, Lars Frellesen

Vejleder: Jesper Larsen

164/88 "Vurdering af matematisk teknologi
Technology Assessment
Technikfolgenabschätzung"

Af: Bernhelm Booss-Bavnbek, Glen Pate med
Martin Bohle-Carbonell og Jens Højgaard Jensen

165/88 "COMPLEX STRUCTURES IN THE NASH-MOSER CATEGORY"

by: Jens Gravesen

166/88 "Grundbegreber i Sandsynligheds-
regningen"

Af: Jørgen Larsen

167a/88 "BASISSTATISTIK 1. Diskrete modeller"

Af: Jørgen Larsen

167b/88 "BASISSTATISTIK 2. Kontinuerte
modeller"

Af: Jørgen Larsen

168/88 "OVERFLADEN AF PLANETEN MARS"
Laboratorie-simulering og MARS-analoger
undersøgt ved Mössbauerspektroskopi.

Fysikspeciale af:

Birger Lundgren

Vejleder: Jens Martin Knudsen
Fys.Lab./HCØ

169/88 "CHARLES S. PEIRCE: MURSTEN OG MØRTEL
TIL EN METAFYSIK."

Fem artikler fra tidsskriftet "The Monist"
1891-93.

Introduktion og oversættelse:

Peder Voetmann Christeansen

170/88 "OPGAVESAMLING I MATEMATIK"

Samtlige opgaver stillet i tiden
1974 - juni 1988

171/88 "The Dirac Equation with Light-Cone Data"

af: Johnny Tom Ottesen

172/88 "FYSIK OG VIRKELIGHED"

Kvantemekanikkens grundlagsproblem
i gymnasiet.

Fysikprojekt af:

Erik Lund og Kurt Jensen

Vejledere: Albert Chr. Paulsen og
Peder Voetmann Christiansen

173/89 "NUMERISKE ALGORITMER"

af: Mogens Brun Heefelt

174/89 " GRAFISK FREMSTILLING AF
FRAKTALER OG KAOS"

af: Peder Voetmann Christiansen

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DEPENDENT SPECTRUM OF THE NON-STATONARY
SOLUTION TO THE OPERATOR RICCATI EQUATION

af: Michael Pedersen

176/89 " A MAXIUM ENTROPY ANSATZ FOR NONLINEAR
RESPONSE THEORY"

af : Jeppe Dyre

177/89 "HVAD SKAL ADAM STÅ MODEL TIL"

af: Morten Andersen, Ulla Engström,
Thomas Gravesen, Nanna Lund, Pia
Madsen, Dina Rawat, Peter Torstensen

Vejleder: Mogens Brun Heefelt

178/89 "BIOSYNTESEN AF PENICILLIN - en matematisk model"

af: Ulla Eghave Rasmussen, Hans Oxvang Mortensen,
Michael Jarden

vejleder i matematik: Jesper Larsen
biologi: Erling Lauridsen

179a/89 "LÆRERVEJLEDNING M.M. til et eksperimentelt forløb
om kaos"

af: Andy Wierød, Søren Brønd og Jimmy Staal

Vejledere: Peder Voetmann Christiansen
Karin Beyer

179b/89 "ELEVHÆFTE: Noter til et eksperimentelt kursus om
kaos"

af: Andy Wierød, Søren Brønd og Jimmy Staal

Vejledere: Peder Voetmann Christiansen
Karin Beyer

180/89 "KAOS I FYSISKE SYSTEMER eksemplificeret ved
torsions- og dobbeltpendul".

af: Andy Wierød, Søren Brønd og Jimmy Staal
Vejleder: Peder Voetmann Christiansen

181/89 "A ZERO-PARAMETER CONSTITUTIVE RELATION FOR PURE
SHEAR VISCOELASTICITY"

by: Jeppe Dyre

183/89 "MATEMATICAL PROBLEM SOLVING, MODELLING. APPLICATIONS
AND LINKS TO OTHER SUBJECTS - State. trends and
issues in mathematics instruction

by: WERNER BLUM, Kassel (FRG) og
MOGENS NISS, Roskilde (Denmark)

184/89 "En metode til bestemmelse af den frekvensafhængige
varmefylde af en underafkølet væske ved glasovergangen"

af: Tage Emil Christensen

185/90 "EN NÆSTEN PERIODISK HISTORIE"

Et matematisk projekt

af: Steen Grode og Thomas Jessen

Vejleder: Jacob Jacobsen

186/90 "RITUAL OG RATIONALITET i videnskabers udvikling"
redigeret af Arne Jakobsen og Stig Andur Pedersen

187/90 "RSA - et kryptografisk system"

af: Annemette Sofie Olufsen, Lars Frellesen
og Ole Møller Nielsen

Vejledere: Michael Pedersen og Finn Munk

188/90 "FERMICONDENSATION - AN ALMOST IDEAL GLASS TRANSITION"

by: Jeppe Dyre

189/90 "DATAMATER I MATEMATIKUNDERVISNINGEN PÅ
GYMNASIET OG HØJERE LÆREANSTALTER

af: Finn Langberg

190/90 "FIVE REQUIREMENTS FOR AN APPROXIMATE NONLINEAR RESPONSE THEORY"

by: Jeppe Dyre

191/90 "MOORE COHOMOLOGY, PRINCIPAL BUNDLES AND ACTIONS OF GROUPS ON C*-ALGEBRAS"

by: Iain Raeburn and Dana P. Williams

192/90 "Age-dependent host mortality in the dynamics of endemic infectious diseases and SIR-models of the epidemiology and natural selection of co-circulating influenza virus with partial cross-immunity"

by: Viggo Andreasen

193/90 "Causal and Diagnostic Reasoning"

by: Stig Andur Pedersen

194a/90 "DETERMINISTISK KAOS"
Projektrapport af : Frank Olsen

194b/90 "DETERMINISTISK KAOS"
Kørselsrapport

Projektrapport af: Frank Olsen

195/90 "STADIER PÅ PARADIGMETS VEJ"
Et projekt om den videnskabelige udvikling der førte til dannelse af kvantemekanikken.

Projektrapport for 1. modul på fysikuddannelsen, skrevet af:

Anja Boisen, Thomas Hougård, Anders Gorm Larsen, Nicolai Ryge.

Vejleder: Peder Voetmann Christiansen

196/90 "ER KAOS NØDVENDIGT?"

- en projektrapport om kaos' paradigmatiske status i fysikken.

af: Johannes K. Nielsen, Jimmy Staal og Peter Bøggild

Vejleder: Peder Voetmann Christiansen

197/90 "Kontrafaktiske konditionaler i HOL

af: Jesper Voetmann, Hans Oxvang Mortensen og Aleksander Høst-Madsen

Vejleder: Stig Andur Pedersen

198/90 "Metal-Isolator-Metal systemer"
Speciale
af: Frank Olsen

199/90 "SPREDT FÆGTNING" Artikelsamling
af: Jens Højgaard Jensen

200/90 "LINEÆR ALGEBRA OG ANALYSE"
Noter til den naturvidenskabelige basisuddannelse.
af: Mogens Niss

201/90 "Undersøgelse af atomare korrelationer i amorfe stoffer ved røntgendiffraktion"

af: Karen Birkelund og Klaus Dahl Jensen

Vejledere: Petr Viščor, Ole Bakander

202/90 "TEGN OG KVANTER"
Foredrag og artikler, 1971-90.

af: Peder Voetmann Christiansen

203/90 "OPGAVESAMLING I MATEMATIK" 1974-1990

afløser tekst 170/88

204/91 "ERKENDELSE OG KVANTEMEKANIK"
et Breddemodul Fysik Projekt
af: Thomas Jessen
Vejleder: Petr Viščor

205/91 "PEIRCE'S LOGIC OF VAGUENESS"
by: Claudine Engel-Tiercelin
Department of Philosophy
Université de Paris-1
(Panthéon-Sorbonne)

206a+b/91 "GERMANIUMBEAMANALYSE SAMT A - GE TYNDFILMS ELEKTRISKE EGENSKABER"
Eksperimentelt Fysikspeciale
af: Jeanne Linda Mortensen
og Annette Post Nielsen
Vejleder: Petr Viščor

207/91 "SOME REMARKS ON AC CONDUCTION IN DISORDERED SOLIDS"

by: Jeppe C. Dyre

208/91 "LANGEVIN MODELS FOR SHEAR STRESS FLUCTUATIONS IN FLOWS OF VISCO-ELASTIC LIQUIDS"

by: Jeppe C. Dyre