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Lognormal Distribution Function for Describing Anelastic and Other Relaxation Processes

I. Theory and Numerical Computations

Abstract: Such phenomena as dielectric, magnetic, and anelastic relaxation are often described in terms of a distribution of relaxation times. It is shown that a relaxation process which exhibits a Gaussian distribution in the logarithm of the relaxation times (a "lognormal" distribution) can be specified completely by three parameters. These are: the mean relaxation time (τ_m), the width of the distribution (β), and the magnitude of the relaxation (δJ). The relationships of these parameters to experimentally measurable functions are usually complicated. These relationships were obtained in numerical form by machine computation. Finally, a simple formula is derived which expresses the parameter β in terms of the widths of the distribution of the activation energies and that of the attempt frequencies.

Introduction

The term "relaxation" is used to denote the time-dependent self-adjustment of a system to a new equilibrium condition when an external variable is changed. In the *static manifestation* of relaxation an appropriate "force" is usually changed abruptly and held at a new value while the conjugate "displacement" is observed as a function of time. Thus, in the case of dielectric relaxation, the polarization (and therefore the electric displacement) is observed to change with time in response to an instantaneous change in the applied electric field; in magnetic relaxation the "force" is the magnetic field, while the "displacement" is the magnetic induction; in anelastic relaxation a time-dependent strain may be observed when the applied stress is changed.

In the present paper we will deal with equations that relate to all of these relaxation phenomena in linear systems. In order to express the results compactly, however, only the notation of anelastic relaxation will be used. In this notation the static behavior may be expressed as the time dependence of the strain, $\varepsilon(t)$, when a stress, σ, is abruptly imposed at $t=0$ and held constant for $t>0$. Under the assumption that the response of the system is linear, its static behavior may be expressed as:

$$\varepsilon(t)/\sigma=J_U+(J_R-J_U)\psi(t)\equiv J_U+\delta J\cdot\psi(t)\,, \tag{1}$$

where $\psi(t)$ is a dimensionless function which goes from zero at $t=0$ to unity as $t\rightarrow\infty$ (and which will here be called the *static response function*)[1]; J_U is the compliance of the sample (strain/stress) measured at $t=0$, and is, therefore, called the *unrelaxed compliance;* and J_R is the compliance of the sample under equilibrium conditions (at $t=\infty$), called the *relaxed compliance.*[2] The quantity $\delta J=J_R-J_U$ is then a measure of the magnitude of the relaxation effect. For conversion of this notation into the terminology of dielectric and magnetic relaxation, use may be made of Table 1, which serves as a dictionary. Henceforth, we will continue to use the terminology of anelasticity, recognizing that translation is easily possible.

The *dynamic manifestations* of a relaxation may be studied by the application of a periodic force. In complex notation, the stress may be given by

$$\sigma=\sigma_0e^{i\omega t}\,, \tag{2}$$

where ω is the circular frequency of the applied stress. The strain will, in general, not be in phase with the stress if relaxation effects are present but rather will lag behind the stress by an angle ϕ, so that

$$\varepsilon=\varepsilon_0e^{i(\omega t-\phi)}=(\varepsilon_1-i\varepsilon_2)e^{i\omega t}\,, \tag{3}$$

Table 1 **Relationship between quantities which appear in various relaxation phenomena.**

Type of quantity	*Symbol used here*	*Name of quantity*		
		Dielectric relaxation	*Magnetic relaxation*	*Anelastic relaxation*
"Force"	σ	Electric field	Magnetic field	Stress
"Displacement"	ε	Electric displacement	Magnetic induction	Strain
Ratio: displacement/force	J	Permittivity or dielectric constant	Permeability	Compliance

where ε_1 and ε_2 are, respectively, the components of strain in phase with, and in quadrature with, the stress. The relationship between stress and strain may, therefore, be expressed as

$$\varepsilon = J^* \sigma \,, \tag{4}$$

where J^*, called the *complex compliance,* is given by

$$J^* = J_1(\omega) - iJ_2(\omega) \,. \tag{5}$$

Note that the real part of J^*, $J_1 = \varepsilon_1/\sigma_0$, and the imaginary part, $J_2 = \varepsilon_2/\sigma_0$, are in general both functions of the applied frequency.[4] The relationship of the phase angle ϕ to these quantities is, from Eq. (3),

$$\tan \phi = J_2/J_1 \,. \tag{6}$$

The quantities $J_1(\omega)$, $J_2(\omega)$ and $\tan \phi(\omega)$ constitute the "dynamic response functions."

In the description of relaxation phenomena, it is customary to start with the simple case in which the function $\psi(t)$ is an exponential, of the form $1 - e^{-t/\tau}$. Here τ is called the *relaxation time*[5] and the relaxation process is described as showing a "single time of relaxation." Correspondingly, the dynamic properties $J_1(\omega)$ and $J_2(\omega)$ are described by the well-known Debye equations:

$$J_1(\omega) = J_U + \frac{\delta J}{1+\omega^2\tau^2} \tag{7}$$

$$J_2(\omega) = \delta J \cdot \frac{\omega\tau}{1+\omega^2\tau^2} \,. \tag{8}$$

The function J_2, when plotted against $\log \omega\tau$, gives a symmetrical peak centered about $\log \omega\tau = 0$ (called the Debye peak), while on the same plot, the function J_1 is antisymmetrical about the point $J_1 = (J_U + J_R)/2$, $\log \omega\tau = 0$. Many phenomena in crystals are describable in these terms, in the sense that J_2 vs $\ln \omega\tau$ gives a well-defined peak, but it is often, if not generally, found that the J_2 peak is somewhat broader than that predicted by the simple theory. Correspondingly, the static response function $\psi(t)$ changes in time more gradually than a simple exponential. Such behavior is undoubtedly due to the existence of a distribution of relaxation times rather than a single relaxation time. Nevertheless, many authors have tended to fit the observed behavior to the theory of a single relaxation time as well as possible. For example, the quantity $\tan \phi$ (which is often measured directly[6]) shows a peak which is commonly broader than that given by inserting the Debye equations into Eq. (6). In spite of this fact, it is often customary to use the Debye equations to obtain an effective relaxation time from the position of the peak, and the strength or magnitude of the relaxation from the height of the peak. The difference between the actual behavior and that of a single relaxation is then quoted as the percentage by which the width of the actual peak exceeds that predicted from the Debye equations. Such a procedure has two weaknesses: 1) it essentially ignores valuable additional information contained in the experimental data, and 2) it leads to errors in the value of the mean relaxation time obtained from the data.

The purpose of the present pair of papers is to study the manner in which the introduction of a distribution in the relaxation times produces a broadening in the J_2 and $\tan \phi$ peaks, as well as to obtain the equivalent changes in the other properties, $J_1(\omega)$ and $\psi(t)$. It is hoped in this way to lead to a formal description of solids which will in turn be valuable in the analysis of the detailed atomistic mechanisms in crystals. Attention will be focused on the information to be gathered by adopting the assumption that the relaxation times are distributed according to a Gaussian (or normal) distribution in $\ln \tau$. This distribution, which is also known as the *lognormal distribution,* was originally suggested by Wiechert[7] as early as 1893 to explain the elastic after-effect of solids. It was later applied in more detail to dielectric relaxation by Wagner[8] and further developed for dielectrics by Yager,[9] who also carried out some numerical computations for the functions $J_1(\omega)$ and $J_2(\omega)$. Although the formal theory has been in the literature for quite some time, methods for utilizing it fully have not been recognized. In order to achieve these ends, the present papers will have as their principal objectives: a) to give methods for obtaining the lognormal distribution parameter from experimental data, including the case where the dynamic data are measured as a function of temperature rather than the frequency; b) to obtain the theoretical significance of the temperature dependence of the Gaussian distribution parameter; and c) to show how to obtain precise values of the mean relaxation time from static and dynamic data. Item (c) is particularly important when the activation energy for the relaxation process is to be determined precisely, so that, for example, a comparison may be made with high-temperature diffusion data.

Mathematical formalism and numerical computations

As already mentioned in the Introduction, the existence of a distribution of relaxation times leads to a broadening of the J_2 peak. Such a distribution may consist of a discrete set of τ-values or of a continuous distribution. In general, unless a detailed knowledge of the relaxation mechanism gives one reason to believe that the possible τ-values form a discrete set, it seems most sensible to assume a continuous distribution. For example, in a crystal, internal strains or statistical variations in local composition can produce a continuous distribution of relaxation times.

General formalism

It is most convenient to define the distribution in terms of the variable $\ln \tau$, rather than in τ itself.[10] Accordingly, we obtain for the dynamic response functions[3]

$$J_1(\omega)=J_U+\delta J\int_{-\infty}^{\infty}\frac{\Psi(\ln\tau)}{1+\omega^2\tau^2}\,d\ln\tau \tag{9}$$

$$J_2(\omega)=\delta J\int_{-\infty}^{\infty}\Psi(\ln\tau)\,\frac{\omega\tau}{1+\omega^2\tau^2}\,d\ln\tau \tag{10}$$

and for the static function

$$\begin{aligned}\psi(t)&=\int_{-\infty}^{\infty}\Psi(\ln\tau)(1-e^{-t/\tau})\,d\ln\tau\\ &=1-\int_{-\infty}^{\infty}\Psi(\ln\tau)e^{-t/\tau}\,d\ln\tau\,.\end{aligned} \tag{11}$$

In these equations, $\Psi(\ln\tau)d\ln\tau$ is the relative number of contributing processes for which the logarithm of the relaxation time falls in the range between $\ln\tau$ and $\ln\tau+d\ln\tau$. It should be noted that the function $\Psi(\ln\tau)$ is defined in such a way that

$$\int_{-\infty}^{\infty}\Psi(\ln\tau)\,d\ln\tau=1\,. \tag{12}$$

The function $\Psi(\ln\tau)$ therefore represents the normalized distribution function.

The function $\tan\phi=J_2/J_1$ is now a ratio of two integrals, given by Eqs. (9) and (10), and is, therefore, not expressible in a simple form unless $\delta J\ll J_U$; when this latter approximation is valid, $\tan\phi\equiv J_2/J_U$, so that $\tan\phi$ and J_2 then differ only by a constant factor. In general, however, this approximation is not valid. Methods for dealing with data for $\tan\phi$ will be discussed in Part II. For the present, we will confine ourselves to the dynamical functions J_1 and J_2 only.

Equations for the lognormal distribution

In principle, it is possible to invert Eqs. (9) to (11) to obtain the distribution function from an experimental response function.[11] In practice, however, this inversion is not easily accomplished. There is, on the other hand, a considerable advantage to working with a definite form for the distribution function, $\Psi(\ln\tau)$, namely, that it makes possible the description of the specific distribution function by a single parameter. Various choices have been made in the past. The "box distribution," which is constant and finite over a limited range of $\ln\tau$ and zero elsewhere, has been a popular choice[12] because it makes possible evaluation of the integrals in Eqs. (9) to (11) in terms of known functions; nevertheless, it has the distinct disadvantage that it cannot be expected to represent a physical situation realistically. The use of a Gaussian distribution in $\ln\tau$ (the lognormal distribution), is more likely to represent realistically a case in which a distribution of relaxation times arises as the result of the distribution of atomic environments about some mean value. Numerous other distribution functions have been suggested in the literature, usually on empirical rather than theoretical grounds. Many of these have been compiled by Gross.[11]

The authors have made a comparison of widely different distribution functions and have found that the shapes of the J_2 peaks are not very sensitive to the choice of the distribution function, up to values of the peak width about twice that for a single relaxation. Accordingly, the present approach is to use the Gaussian as a reasonable distribution function and to explore fully the consequences of the existence of such a distribution. In the notation of Wagner, we introduce a variable

$$z=\ln(\tau/\tau_m)\,, \tag{13}$$

where τ_m is the most probable value of τ. The normalized distribution function $\Psi(\ln\tau)$ may then be written:

$$\Psi(z)=\frac{b}{\sqrt{\pi}}\exp(-b^2z^2)=\frac{1}{\beta\sqrt{\pi}}\exp[-(z/\beta)^2], \tag{14}$$

where b is the Gaussian distribution parameter. The quantity $\beta\equiv b^{-1}$ measures the half-width of the distribution at the point where $\Psi(z)$ falls to $1/e$ of its maximum value, $\Psi(0)$. Substituting (14) into the general equations (9) to (11) and introducing the variables

$$\left.\begin{aligned}x&=\ln\omega\tau_m\\ y&=\ln(t/\tau_m)\end{aligned}\right\}, \tag{15}$$

we obtain for the dynamic response functions $J_1(x)$ and $J_2(x)$,

$$\frac{J_1(x)-J_U}{\delta J}=\frac{1}{\sqrt{\pi}}\int_{-\infty}^{\infty}e^{-u^2}\frac{du}{1+\exp[2(x+\beta u)]}\equiv f_1(x,\beta) \tag{16}$$

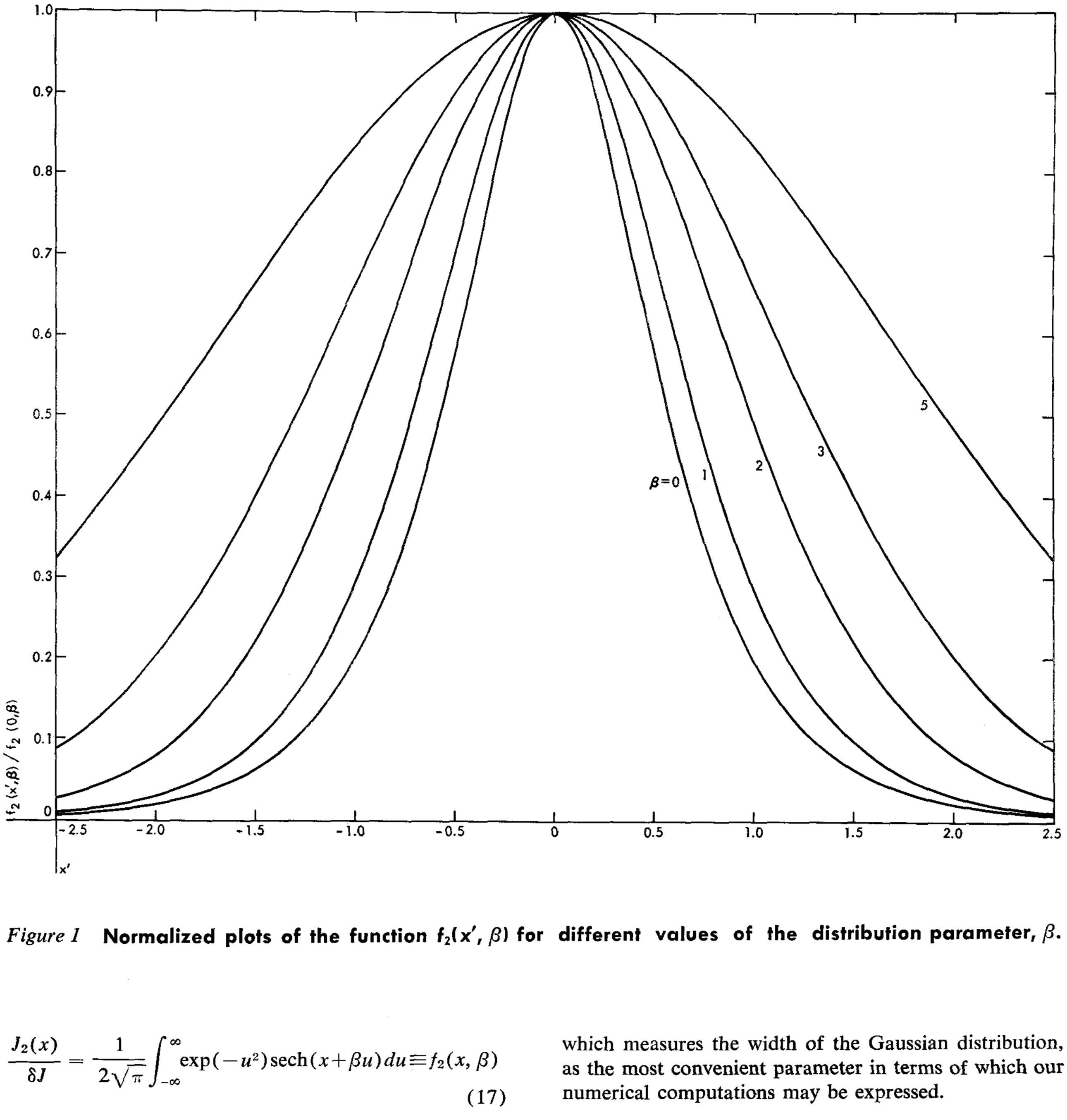


Figure 1 **Normalized plots of the function $f_2(x',\beta)$ for different values of the distribution parameter, β.**

$$\frac{J_2(x)}{\delta J}=\frac{1}{2\sqrt{\pi}}\int_{-\infty}^{\infty}\exp(-u^2)\operatorname{sech}(x+\beta u)\,du\equiv f_2(x,\beta) \tag{17}$$

and for the static response function $\psi(y)$,

$$1-\psi(y)=\frac{1}{\sqrt{\pi}}\int_{-\infty}^{\infty}\exp(-u^2)\exp[-e^{(y-\beta u)}]\,du\equiv g(y,\beta). \tag{18}$$

Since these integrals cannot be evaluated in terms of well-tabulated functions, numerical computations are required. In each of these equations, the quantity β appears as a parameter. For $\beta=0$, the Gaussian function goes into a Dirac δ-function, and Eqs. (16) to (18) degenerate into the corresponding equations for the case of a single relaxation time. It is, therefore, appropriate to regard β, which measures the width of the Gaussian distribution, as the most convenient parameter in terms of which our numerical computations may be expressed.

Results of computations for the lognormal distribution[13]

The above description gives the functions $J_1(\omega)$ and $\varepsilon(t)/\sigma$ in terms of four parameters J_U, δJ, τ_m, and β, and the function $J_2(\omega)$ in terms of the last three of these parameters. This amounts to the addition of just one parameter, β, above and beyond those required to describe the behavior of a material for the case of a single relaxation time. We will now be concerned with methods for obtaining the three parameters of the relaxation (δJ, τ_m, and β) from experimental data, as well as for checking that the lognormal distribution fits the data in a satisfactory way. The fourth parameter, J_U, is not a parameter of the relaxation; it may be obtained directly from the value of J_1 at high frequencies.

For actual applications, it is useful to obtain J_1 and J_2 not as functions of x, but of the variable

$$x' = \log_{10} \omega\tau_m = x/2.303 \tag{19}$$

and ψ as a function of

$$y' = \log_{10}(t/\tau_m) = y/2.303\ . \tag{20}$$

The functions $f_1(x', \beta)$ and $f_2(x', \beta)$, given by Eqs. (16) and (17), (with the change of variables given by (19) and (20)) have been computed by Yager[9] for the range $\beta = 0$ to 6.7, but the intervals selected for both β and x' were rather large, thus limiting the precision of the information that may be derived from his computations.[14] We have, therefore, recomputed the function $f_2(x'\ \beta)$ and at the same time calculated $g(y', \beta)$ over the range $\beta = 0$ to 7 in close intervals, using an IBM 704. We found that the infinite integrals could be replaced by integrals from -4 to $+4$ with no errors up to the sixth decimal place. The finite integrals were then evaluated by the trapezoidal rule. Typical results for the normalized peak function, $f_2(x', \beta)/f_2(0, \beta)$, and for the function $g(y', \beta)$ are plotted in Figs. 1 and 2, respectively. At the same time, the numerical results for these functions are listed in Tables 2 and 3, respectively. The peak height $f_2(0, \beta)$ is tabulated as a function of β in Table 4.

Results for the J_2 peak will first be examined. For the case of a single relaxation time, the peak occurs at $\omega\tau = 1$, which makes possible the determination of τ from the value of ω at the peak. In the present more general case, we have the peak at $x = 0$, or $\omega\tau_m = 1$. The value of τ_m is, therefore, obtainable from the peak position. For a single relaxation time, $\beta = 0$, the height of the J_2 peak is (from Eq. (8)) $\delta J/2$, while the peak width is given by

$$\Delta_2 x'(0) = 1.144\ . \tag{21}$$

Figure 2 **Plots of the function $g(y', \beta)$ for different values of the distribution parameter, β.**

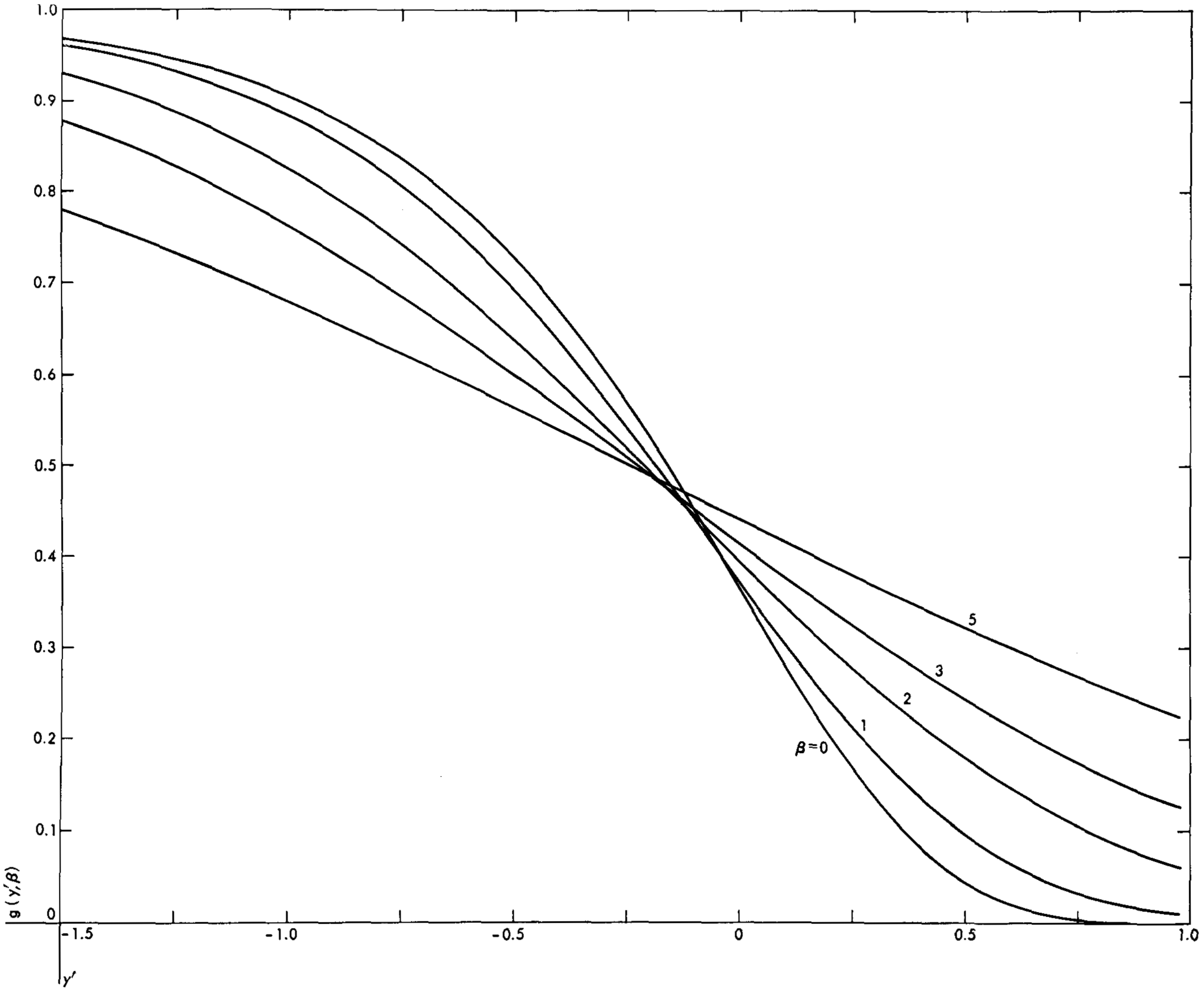


Table 2 **Results of numerical computations for the normalized peak functions $f_2(x', \beta)/f_2(0, \beta)$.**

x'	$\beta = 0$	0.2	0.4	0.6	0.8	1.00	1.25	1.50	1.75	2.00	2.25
0.00	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
0.05	.9934	.9936	.9942	.9949	.9956	.9963	.9969	.9974	.9978	.9981	.9984
0.10	.9740	.9750	.9773	.9801	.9829	.9853	.9878	.9898	.9914	.9927	.9937
0.15	.9431	.9451	.9501	.9561	.9621	.9674	.9729	.9774	.9809	.9837	.9860
0.20	.9025	.9057	.9138	.9239	.9339	.9430	.9525	.9602	.9663	.9713	.9753
0.25	.8544	.8588	.8702	.8847	.8994	.9127	.9270	.9386	.9480	.9555	.9617
0.30	.8011	.8066	.8210	.8399	.8593	.8774	.8970	.9130	.9261	.9367	.9454
0.35	.7447	.7511	.7682	.7910	.8151	.8379	.8629	.8838	.9009	.9150	.9264
0.40	.6872	.6943	.7133	.7394	.7677	.7950	.8256	.8514	.8729	.8905	.9051
0.45	.6302	.6377	.6580	.6865	.7184	.7498	.7856	.8164	.8422	.8637	.8816
0.50	.5749	.5825	.6034	.6336	.6681	.7030	.7436	.7792	.8094	.8348	.8561
0.55	.5221	.5296	.5507	.5816	.6180	.6556	.7004	.7404	.7748	.8041	.8288
0.60	.4725	.4798	.5004	.5314	.5687	.6082	.6565	.7004	.7389	.7719	.8000
0.65	.4263	.4332	.4531	.4835	.5209	.5616	.6125	.6598	.7019	.7385	.7699
0.70	.3837	.3902	.4090	.4383	.4752	.5164	.5689	.6189	.6643	.7042	.7388
0.75	.3447	.3507	.3684	.3962	.4320	.4728	.5263	.5784	.6264	.6693	.7069
0.80	.3092	.3147	.3311	.3572	.3915	.4314	.4850	.5384	.5886	.6341	.6744
0.85	.2769	.2820	.2971	.3214	.3538	.3923	.4453	.4994	.5512	.5988	.6417
0.90	.2478	.2524	.2662	.2886	.3189	.3557	.4075	.4616	.5144	.5638	.6088
0.95	.2216	.2258	.2383	.2588	.2870	.3217	.3717	.4252	.4785	.5292	.5760
1.00	.1980	.2018	.2131	.2318	.2577	.2903	.3381	.3905	.4438	.4953	.5435
1.05	.1768	.1802	.1904	.2074	.2312	.2614	.3068	.3576	.4104	.4623	.5115
1.10	.1578	.1609	.1701	.1855	.2071	.2350	.2777	.3266	.3784	.4302	.4801
1.15	.1408	.1436	.1519	.1657	.1854	.2110	.2508	.2974	.3479	.3993	.4494
1.20	.1256	.1281	.1355	.1480	.1658	.1892	.2261	.2703	.3191	.3697	.4197
1.25	.1121	.1143	.1209	.1321	.1481	.1694	.2035	.2451	.2919	.3413	.3910
1.30	.0999	.1019	.1079	.1179	.1323	.1516	.1829	.2218	.2664	.3144	.3634
1.35	.0891	.0909	.0962	.1052	.1181	.1355	.1642	.2003	.2426	.2889	.3370
1.40	.0794	.0810	.0858	.0938	.1054	.1211	.1472	.1806	.2205	.2649	.3118
1.45	.0708	.0722	.0765	.0837	.0941	.1082	.1318	.1626	.2000	.2424	.2878
1.50	.0631	.0644	.0682	.0746	.0839	.0966	.1180	.1462	.1811	.2213	.2651
1.55	.0563	.0574	.0608	.0665	.0748	.0862	.1055	.1313	.1637	.2017	.2437
1.60	.0502	.0512	.0542	.0593	.0667	.0769	.0943	.1178	.1477	.1834	.2236
1.65	.0447	.0456	.0483	.0528	.0595	.0686	.0842	.1056	.1331	.1666	.2047
1.70	.0398	.0406	.0430	.0471	.0530	.0612	.0752	.0945	.1198	.1510	.1871
1.75	.0355	.0362	.0384	.0420	.0473	.0546	.0671	.0846	.1077	.1366	.1707
1.80	.0316	.0323	.0342	.0374	.0421	.0486	.0599	.0756	.0967	.1235	.1555
1.85	.0282	.0288	.0305	.0333	.0376	.0434	.0534	.0676	.0868	.1114	.1414
1.90	.0251	.0256	.0271	.0297	.0335	.0386	.0477	.0604	.0778	.1005	.1283
1.95	.0224	.0228	.0242	.0265	.0298	.0344	.0425	.0539	.0697	.0904	.1164
2.00	.0199	.0203	.0216	.0236	.0266	.0307	.0379	.0481	.0624	.0813	.1053
2.05	.0178	.0181	.0192	.0210	.0237	.0274	.0338	.0430	.0558	.0731	.0952
2.10	.0158	.0162	.0171	.0187	.0211	.0244	.0301	.0383	.0499	.0656	.0860
2.15	.0141	.0144	.0152	.0167	.0188	.0217	.0268	.0342	.0446	.0588	.0776
2.20	.0126	.0128	.0136	.0149	.0168	.0194	.0239	.0305	.0398	.0527	.0699
2.25	.0112	.0114	.0121	.0133	.0149	.0173	.0213	.0272	.0355	.0472	.0629
2.30	.0100	.0102	.0108	.0118	.0133	.0154	.0190	.0242	.0317	.0422	.0566
2.35	.0089	.0091	.0096	.0105	.0119	.0137	.0169	.0216	.0283	.0378	.0508
2.40	.0079	.0081	.0086	.0094	.0106	.0122	.0151	.0192	.0252	.0338	.0456
2.45	.0070	.0072	.0076	.0083	.0094	.0109	.0134	.0172	.0225	.0302	.0409
2.50	.0063	.0064	.0068	.0074	.0084	.0097	.0120	.0153	.0201	.0270	.0367
2.55	.0056	.0057	.0060	.0066	.0075	.0086	.0107	.0136	.0179	.0241	.0328
2.60	.0050	.0051	.0054	.0059	.0066	.0077	.0095	.0121	.0160	.0215	.0294
2.65	.0044	.0045	.0048	.0052	.0059	.0068	.0085	.0108	.0142	.0192	.0263
2.70	.0039	.0040	.0043	.0047	.0053	.0061	.0075	.0096	.0127	.0171	.0235
2.75	.0035	.0036	.0038	.0042	.0047	.0054	.0067	.0086	.0113	.0153	.0210
2.80	.0031	.0032	.0034	.0037	.0042	.0048	.0060	.0076	.0101	.0136	.0188
2.85	.0028	.0028	.0030	.0033	.0037	.0043	.0053	.0068	.0090	.0121	.0168
2.90	.0025	.0025	.0027	.0029	.0033	.0038	.0047	.0061	.0080	.0108	.0150
2.95	.0022	.0022	.0024	.0026	.0029	.0034	.0042	.0054	.0071	.0096	.0134
3.00	.0020	.0020	.0021	.0023	.0026	.0030	.0038	.0048	.0063	.0086	.0119

Table 2 (Contd.)

x′	β = 2.50	2.75	3.00	3.50	4.00	4.50	5.00	5.50	6.00	6.50	7.00
0.00	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
0.05	.9986	.9988	.9989	.9991	.9993	.9994	.9995	.9996	.9996	.9997	.9997
0.10	.9946	.9952	.9958	.9967	.9973	.9978	.9981	.9984	.9986	.9988	.9990
0.15	.9878	.9894	.9906	.9926	.9940	.9951	.9959	.9965	.9970	.9974	.9977
0.20	.9785	.9812	.9835	.9869	.9894	.9913	.9927	.9938	.9947	.9954	.9960
0.25	.9667	.9709	.9743	.9797	.9836	.9865	.9887	.9904	.9918	.9929	.9938
0.30	.9525	.9584	.9633	.9709	.9765	.9806	.9838	.9863	.9882	.9898	.9911
0.35	.9359	.9438	.9504	.9606	.9681	.9737	.9780	.9814	.9840	.9862	.9879
0.40	.9172	.9273	.9357	.9489	.9586	.9659	.9714	.9758	.9792	.9820	.9842
0.45	.8965	.9089	.9194	.9358	.9479	.9570	.9640	.9694	.9738	.9772	.9801
0.50	.8739	.8889	.9015	.9214	.9361	.9472	.9557	.9624	.9677	.9720	.9755
0.55	.8496	.8672	.8821	.9057	.9232	.9365	.9467	.9547	.9611	.9662	.9704
0.60	.8239	.8442	.8614	.8889	.9094	.9249	.9369	.9463	.9539	.9599	.9649
0.65	.7968	.8198	.8395	.8710	.8945	.9125	.9264	.9373	.9461	.9532	.9590
0.70	.7687	.7944	.8165	.8520	.8787	.8992	.9151	.9277	.9377	.9459	.9526
0.75	.7396	.7680	.7925	.8321	.8621	.8852	.9032	.9175	.9289	.9382	.9458
0.80	.7099	.7408	.7676	.8114	.8447	.8705	.8906	.9066	.9195	.9300	.9386
0.85	.6796	.7129	.7421	.7899	.8266	.8551	.8775	.8953	.9096	.9213	.9309
0.90	.6490	.6846	.7159	.7677	.8078	.8391	.8637	.8834	.8992	.9122	.9229
0.95	.6182	.6559	.6894	.7450	.7884	.8224	.8494	.8710	.8884	.9027	.9145
1.00	.5875	.6271	.6625	.7218	.7685	.8053	.8346	.8581	.8771	.8928	.9057
1.05	.5569	.5982	.6354	.6982	.7481	.7876	.8192	.8447	.8654	.8824	.8966
1.10	.5266	.5694	.6082	.6743	.7273	.7696	.8035	.8309	.8533	.8718	.8871
1.15	.4969	.5408	.5810	.6502	.7061	.7511	.7873	.8168	.8409	.8607	.8772
1.20	.4677	.5126	.5540	.6260	.6847	.7323	.7708	.8022	.8280	.8493	.8671
1.25	.4393	.4849	.5273	.6018	.6631	.7132	.7540	.7874	.8148	.8376	.8567
1.30	.4116	.4577	.5009	.5776	.6414	.6938	.7368	.7722	.8014	.8256	.8459
1.35	.3848	.4311	.4749	.5535	.6195	.6743	.7194	.7567	.7876	.8133	.8349
1.40	.3590	.4053	.4494	.5295	.5977	.6546	.7018	.7410	.7735	.8007	.8236
1.45	.3342	.3802	.4245	.5059	.5759	.6348	.6840	.7250	.7592	.7879	.8121
1.50	.3105	.3560	.4003	.4825	.5542	.6150	.6661	.7089	.7447	.7749	.8003
1.55	.2879	.3327	.3768	.4596	.5326	.5951	.6481	.6926	.7300	.7616	.7883
1.60	.2664	.3103	.3540	.4370	.5112	.5753	.6299	.6761	.7151	.7481	.7762
1.65	.2460	.2889	.3320	.4150	.4901	.5556	.6118	.6596	.7001	.7345	.7638
1.70	.2268	.2685	.3108	.3935	.4692	.5360	.5936	.6429	.6849	.7207	.7512
1.75	.2086	.2490	.2905	.3725	.4487	.5165	.5755	.6262	.6696	.7068	.7385
1.80	.1916	.2306	.2711	.3521	.4285	.4972	.5574	.6095	.6543	.6927	.7257
1.85	.1757	.2131	.2525	.3324	.4087	.4781	.5394	.5928	.6389	.6785	.7127
1.90	.1608	.1967	.2348	.3133	.3894	.4593	.5216	.5761	.6234	.6643	.6996
1.95	.1469	.1812	.2180	.2949	.3705	.4408	.5039	.5594	.6079	.6500	.6864
2.00	.1340	.1666	.2021	.2772	.3521	.4225	.4863	.5429	.5924	.6356	.6731
2.05	.1221	.1530	.1870	.2601	.3342	.4046	.4690	.5264	.5770	.6212	.6598
2.10	.1111	.1403	.1728	.2438	.3169	.3871	.4518	.5100	.5615	.6068	.6464
2.15	.1009	.1284	.1594	.2282	.3000	.3699	.4350	.4938	.5462	.5924	.6330
2.20	.0915	.1174	.1469	.2132	.2838	.3532	.4183	.4777	.5309	.5780	.6195
2.25	.0829	.1072	.1351	.1990	.2681	.3368	.4020	.4618	.5157	.5636	.6060
2.30	.0750	.0977	.1241	.1855	.2529	.3209	.3860	.4461	.5006	.5493	.5926
2.35	.0678	.0889	.1139	.1727	.2383	.3054	.3703	.4306	.4857	.5351	.5791
2.40	.0612	.0809	.1043	.1605	.2243	.2904	.3549	.4154	.4708	.5209	.5657
2.45	.0552	.0734	.0954	.1490	.2109	.2759	.3398	.4004	.4562	.5068	.5523
2.50	.0498	.0666	.0872	.1382	.1981	.2618	.3251	.3856	.4417	.4928	.5390
2.55	.0448	.0603	.0796	.1279	.1858	.2481	.3108	.3711	.4274	.4790	.5257
2.60	.0403	.0546	.0725	.1183	.1741	.2350	.2969	.3569	.4133	.4653	.5125
2.65	.0362	.0494	.0660	.1093	.1629	.2223	.2833	.3430	.3994	.4517	.4994
2.70	.0325	.0446	.0600	.1008	.1523	.2101	.2701	.3293	.3857	.4382	.4864
2.75	.0292	.0402	.0545	.0929	.1422	.1984	.2573	.3160	.3723	.4249	.4735
2.80	.0262	.0363	.0495	.0855	.1326	.1871	.2450	.3030	.3591	.4118	.4607
2.85	.0234	.0327	.0448	.0786	.1236	.1763	.2330	.2903	.3461	.3989	.4480
2.90	.0210	.0294	.0406	.0722	.1150	.1660	.2214	.2780	.3334	.3862	.4355
2.95	.0188	.0264	.0367	.0662	.1069	.1561	.2102	.2659	.3210	.3736	.4231
3.00	.0168	.0237	.0332	.0606	.0993	.1467	.1994	.2542	.3088	.3613	.4108

Table 3 **Results of numerical computations for the function $g(y', \beta)$.**

y'	$\beta = 0$	0.25	0.50	0.75	1.00	1.25	1.50	1.75
-1.50	0.9689	0.9684	0.9670	0.9645	0.9607	0.9555	0.9496	0.9400
-1.46	0.9659	0.9654	0.9638	0.9611	0.9570	0.9514	0.9440	0.9348
-1.42	0.9627	0.9621	0.9604	0.9575	0.9531	0.9470	0.9391	0.9293
-1.38	0.9592	0.9586	0.9567	0.9535	0.9487	0.9422	0.9337	0.9234
-1.34	0.9553	0.9547	0.9527	0.9492	0.9440	0.9370	0.9279	0.9170
-1.30	0.9511	0.9504	0.9482	0.9444	0.9389	0.9313	0.9217	0.9102
-1.26	0.9465	0.9458	0.9434	0.9393	0.9333	0.9252	0.9150	0.9028
-1.22	0.9415	0.9407	0.9381	0.9337	0.9272	0.9186	0.9078	0.8950
-1.18	0.9361	0.9352	0.9324	0.9276	0.9207	0.9114	0.9000	0.8867
-1.14	0.9301	0.9291	0.9262	0.9210	0.9136	0.9038	0.8917	0.8778
-1.10	0.9236	0.9226	0.9194	0.9138	0.9059	0.8954	0.8828	0.8683
-1.06	0.9166	0.9154	0.9120	0.9060	0.8976	0.8865	0.8732	0.8583
-1.02	0.9089	0.9077	0.9040	0.8976	0.8886	0.8769	0.8631	0.8476
-0.98	0.9006	0.8993	0.8952	0.8885	0.8789	0.8666	0.8522	0.8364
-0.94	0.8915	0.8901	0.8858	0.8786	0.8684	0.8556	0.8407	0.8245
-0.90	0.8817	0.8802	0.8756	0.8679	0.8572	0.8438	0.8284	0.8119
-0.86	0.8711	0.8694	0.8646	0.8564	0.8452	0.8312	0.8155	0.7988
-0.82	0.8595	0.8578	0.8526	0.8440	0.8322	0.8179	0.8018	0.7849
-0.78	0.8471	0.8452	0.8398	0.8307	0.8184	0.8036	0.7873	0.7704
-0.74	0.8336	0.8317	0.8259	0.8164	0.8037	0.7886	0.7721	0.7552
-0.70	0.8191	0.8171	0.8110	0.8011	0.7880	0.7726	0.7561	0.7394
-0.66	0.8035	0.8014	0.7950	0.7848	0.7714	0.7558	0.7394	0.7230
-0.62	0.7867	0.7845	0.7779	0.7674	0.7537	0.7381	0.7219	0.7059
-0.58	0.7687	0.7664	0.7596	0.7488	0.7350	0.7195	0.7036	0.6883
-0.54	0.7495	0.7471	0.7401	0.7291	0.7153	0.7001	0.6847	0.6700
-0.50	0.7289	0.7264	0.7194	0.7083	0.6947	0.6798	0.6651	0.6512
-0.46	0.7070	0.7045	0.6974	0.6864	0.6730	0.6587	0.6448	0.6320
-0.42	0.6837	0.6812	0.6741	0.6633	0.6504	0.6369	0.6239	0.6122
-0.38	0.6591	0.6566	0.6496	0.6391	0.6268	0.6143	0.6025	0.5920
-0.34	0.6331	0.6307	0.6239	0.6139	0.6025	0.5910	0.5806	0.5714
-0.30	0.6058	0.6035	0.5970	0.5877	0.5773	0.5672	0.5582	0.5505
-0.26	0.5772	0.5750	0.5690	0.5606	0.5514	0.5428	0.5354	0.5293
-0.22	0.5474	0.5454	0.5400	0.5326	0.5249	0.5180	0.5123	0.5079
-0.18	0.5165	0.5147	0.5101	0.5039	0.4979	0.4928	0.4890	0.4864
-0.14	0.4846	0.4831	0.4794	0.4747	0.4705	0.4674	0.4656	0.4647
-0.10	0.4519	0.4508	0.4481	0.4451	0.4429	0.4419	0.4421	0.4431
-0.06	0.4186	0.4179	0.4164	0.4152	0.4152	0.4164	0.4186	0.4215
-0.02	0.3848	0.3846	0.3844	0.3852	0.3874	0.3909	0.3953	0.4000
0.02	0.3509	0.3512	0.3526	0.3554	0.3600	0.3657	0.3721	0.3787
0.06	0.3172	0.3181	0.3209	0.3260	0.3328	0.3408	0.3493	0.3577
0.10	0.2840	0.2854	0.2898	0.2970	0.3062	0.3164	0.3268	0.3370
0.14	0.2515	0.2535	0.2596	0.2688	0.2802	0.2925	0.3048	0.3166
0.18	0.2201	0.2228	0.2303	0.2416	0.2551	0.2693	0.2834	0.2968
0.22	0.1902	0.1934	0.2024	0.2156	0.2309	0.2468	0.2625	0.2774
0.26	0.1621	0.1658	0.1760	0.1908	0.2078	0.2253	0.2424	0.2586
0.30	0.1360	0.1401	0.1514	0.1675	0.1858	0.2047	0.2230	0.2403
0.34	0.1122	0.1166	0.1287	0.1458	0.1651	0.1850	0.2044	0.2228
0.38	0.0908	0.0955	0.1080	0.1257	0.1458	0.1665	0.1867	0.2059
0.42	0.0721	0.0768	0.0895	0.1074	0.1279	0.1491	0.1699	0.1897
0.46	0.0559	0.0605	0.0731	0.0909	0.1114	0.1328	0.1540	0.1743
0.50	0.0423	0.0467	0.0588	0.0761	0.0964	0.1177	0.1390	0.1597
0.54	0.0312	0.0353	0.0466	0.0631	0.0827	0.1038	0.1250	0.1458
0.58	0.0223	0.0260	0.0363	0.0517	0.0705	0.0910	0.1120	0.1327
0.62	0.0155	0.0186	0.0278	0.0419	0.0596	0.0793	0.0999	0.1204
0.66	0.0104	0.0130	0.0209	0.0336	0.0500	0.0688	0.0887	0.1089
0.70	0.0067	0.0088	0.0154	0.0266	0.0416	0.0593	0.0785	0.0982
0.74	0.0041	0.0058	0.0111	0.0208	0.0343	0.0508	0.0691	0.0882
0.78	0.0024	0.0036	0.0079	0.0160	0.0281	0.0433	0.0606	0.0790
0.82	0.0014	0.0022	0.0054	0.0122	0.0228	0.0366	0.0529	0.0705
0.86	0.0007	0.0013	0.0037	0.0092	0.0183	0.0308	0.0459	0.0627
0.90	0.0004	0.0007	0.0024	0.0068	0.0146	0.0258	0.0397	0.0555
0.94	0.0002	0.0004	0.0016	0.0050	0.0115	0.0214	0.0342	0.0490
0.98	0.0001	0.0002	0.0010	0.0036	0.0090	0.0177	0.0293	0.0432

Table 3 (Contd.)

y'	β = 2.00	2.50	3.00	3.50	4.00	5.00	6.00	7.00
-1.50	0.9297	0.9051	0.8779	0.8505	0.8246	0.7794	0.7430	0.7140
-1.46	0.9240	0.8985	0.8706	0.8430	0.8170	0.7721	0.7363	0.7079
-1.42	0.9179	0.8914	0.8630	0.8351	0.8092	0.7647	0.7295	0.7016
-1.38	0.9114	0.8840	0.8550	0.8270	0.8011	0.7571	0.7226	0.6954
-1.34	0.9044	0.8762	0.8468	0.8186	0.7929	0.7495	0.7156	0.6891
-1.30	0.8970	0.8680	0.8382	0.8100	0.7844	0.7417	0.7085	0.6827
-1.26	0.8892	0.8594	0.8293	0.8011	0.7759	0.7337	0.7014	0.6762
-1.22	0.8808	0.8504	0.8200	0.7920	0.7669	0.7257	0.6942	0.6698
-1.18	0.8720	0.8409	0.8105	0.7826	0.7579	0.7175	0.6868	0.6632
-1.14	0.8627	0.8310	0.8006	0.7729	0.7486	0.7092	0.6795	0.6566
-1.10	0.8528	0.8208	0.7903	0.7630	0.7392	0.7008	0.6720	0.6500
-1.06	0.8424	0.8100	0.7797	0.7528	0.7295	0.6923	0.6645	0.6433
-1.02	0.8314	0.7989	0.7688	0.7424	0.7197	0.6836	0.6569	0.6366
-0.98	0.8199	0.7873	0.7576	0.7318	0.7097	0.6749	0.6492	0.6298
-0.94	0.8078	0.7753	0.7461	0.7209	0.6996	0.6661	0.6415	0.6230
-0.90	0.7951	0.7629	0.7343	0.7098	0.6892	0.6571	0.6337	0.6161
-0.86	0.7819	0.7500	0.7221	0.6985	0.6788	0.6481	0.6259	0.6092
-0.82	0.7681	0.7368	0.7097	0.6870	0.6681	0.6390	0.6180	0.6023
-0.78	0.7538	0.7231	0.6970	0.6753	0.6573	0.6298	0.6101	0.5953
-0.74	0.7389	0.7091	0.6840	0.6634	0.6464	0.6205	0.6021	0.5884
-0.70	0.7234	0.6946	0.6708	0.6513	0.6353	0.6112	0.5940	0.5813
-0.66	0.7074	0.6799	0.6573	0.6390	0.6241	0.6018	0.5860	0.5743
-0.62	0.6910	0.6648	0.6436	0.6266	0.6128	0.5923	0.5778	0.5672
-0.58	0.6740	0.6494	0.6297	0.6140	0.6014	0.5828	0.5697	0.5601
-0.54	0.6566	0.6336	0.6155	0.6013	0.5899	0.5732	0.5615	0.5530
-0.50	0.6387	0.6176	0.6012	0.5885	0.5784	0.5635	0.5533	0.5459
-0.46	0.6204	0.6014	0.5868	0.5755	0.5667	0.5538	0.5451	0.5387
-0.42	0.6018	0.5849	0.5722	0.5625	0.5550	0.5441	0.5368	0.5316
-0.38	0.5828	0.5682	0.5574	0.5494	0.5432	0.5344	0.5285	0.5244
-0.34	0.5636	0.5514	0.5426	0.5362	0.5313	0.5246	0.5203	0.5172
-0.30	0.5441	0.5344	0.5277	0.5229	0.5195	0.5148	0.5120	0.5100
-0.26	0.5244	0.5173	0.5127	0.5097	0.5076	0.5051	0.5037	0.5028
-0.22	0.5045	0.5001	0.4977	0.4964	0.4957	0.4953	0.4954	0.4956
-0.18	0.4846	0.4829	0.4826	0.4830	0.4838	0.4854	0.4870	0.4884
-0.14	0.4646	0.4657	0.4676	0.4697	0.4719	0.4757	0.4788	0.4812
-0.10	0.4446	0.4485	0.4526	0.4564	0.4600	0.4659	0.4705	0.4740
-0.06	0.4247	0.4313	0.4376	0.4432	0.4481	0.4561	0.4622	0.4669
-0.02	0.4049	0.4143	0.4227	0.4300	0.4363	0.4464	0.4539	0.4597
0.02	0.3852	0.3973	0.4079	0.4169	0.4245	0.4366	0.4457	0.4526
0.06	0.3658	0.3805	0.3932	0.4038	0.4128	0.4270	0.4374	0.4454
0.10	0.3466	0.3639	0.3786	0.3909	0.4012	0.4173	0.4292	0.4383
0.14	0.3278	0.3476	0.3642	0.3780	0.3896	0.4077	0.4211	0.4312
0.18	0.3093	0.3314	0.3499	0.3653	0.3782	0.3982	0.4129	0.4241
0.22	0.2912	0.3156	0.3358	0.3527	0.3668	0.3887	0.4048	0.4170
0.26	0.2736	0.3000	0.3220	0.3403	0.3555	0.3793	0.3968	0.4100
0.30	0.2564	0.2848	0.3084	0.3280	0.3444	0.3700	0.3888	0.4030
0.34	0.2398	0.2699	0.2950	0.3159	0.3334	0.3607	0.3808	0.3960
0.38	0.2238	0.2554	0.2818	0.3040	0.3225	0.3515	0.3728	0.3891
0.42	0.2083	0.2412	0.2690	0.2922	0.3118	0.3424	0.3650	0.3822
0.46	0.1934	0.2276	0.2564	0.2807	0.3012	0.3334	0.3572	0.3753
0.50	0.1792	0.2143	0.2442	0.2694	0.2908	0.3244	0.3494	0.3685
0.54	0.1656	0.2014	0.2322	0.2583	0.2805	0.3156	0.3417	0.3617
0.58	0.1526	0.1890	0.2206	0.2475	0.2704	0.3068	0.3341	0.3550
0.62	0.1403	0.1771	0.2092	0.2369	0.2605	0.2982	0.3265	0.3482
0.66	0.1287	0.1656	0.1983	0.2265	0.2508	0.2897	0.3190	0.3416
0.70	0.1177	0.1547	0.1876	0.2164	0.2413	0.2813	0.3116	0.3350
0.74	0.1074	0.1442	0.1774	0.2066	0.2319	0.2730	0.3042	0.3284
0.78	0.0977	0.1341	0.1674	0.1970	0.2228	0.2648	0.2970	0.3219
0.82	0.0886	0.1246	0.1579	0.1877	0.2139	0.2568	0.2898	0.3155
0.86	0.0802	0.1155	0.1487	0.1786	0.2052	0.2489	0.2827	0.3091
0.90	0.0724	0.1069	0.1398	0.1698	0.1966	0.2411	0.2756	0.3027
0.94	0.0652	0.0987	0.1313	0.1614	0.1884	0.2335	0.2687	0.2964
0.98	0.0585	0.0910	0.1232	0.1531	0.1803	0.2260	0.2618	0.2902

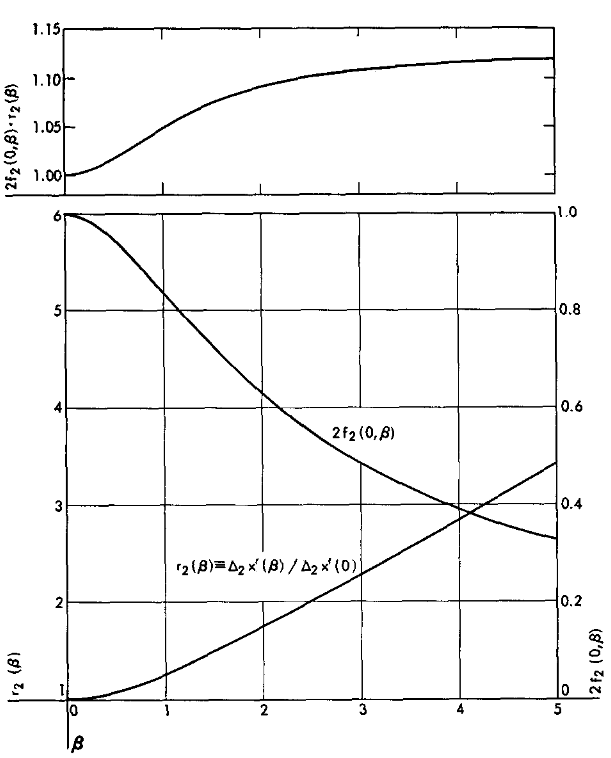


Figure 3 **Dependence of relative peak height, $2f_2(0,\beta)$, relative peak width, $r_2(\beta)$, and the product of these quantities, on the distribution parameter, β.**

For a distribution of relaxation times, the height is smaller than the corresponding value for a single relaxation. This is apparent from the tabulated dependence of the peak height on the distribution width, β, in Table 4. It is also shown in Fig. 3, where we have plotted $2f_2(0,\beta)=f_2(0,\beta)/f_2(0,0)$, which is the height of the J_2 peak relative to the height for the case of a single relaxation time of the same δJ. It is easy to show[15] from Eq. (17) that for large β the peak height asymptotically approaches the value

$$f_2(0,\beta)\equiv J_2(0)/\delta J\sim\sqrt{\pi}/2\beta\,. \tag{22}$$

Further, this asymptotic expression must always be greater than the exact expression. It is noteworthy that for $\beta=7$ the exactly computed value (Table 4) is 4% below that obtained from Eq. (22). The width of the J_2 peak may also be obtained from the numerical values in Table 2. The ratio of the width of the J_2 peak at half maximum, $\Delta_2x'(\beta)$, relative to the width for a single relaxation, $\Delta_2x'(0)$, is

$$r_2(\beta)\equiv\Delta_2x'(\beta)/\Delta_2x'(0)\,. \tag{23}$$

This quantity is plotted vs β in Fig. 3 and the computed values are also listed in Table 4. The quantity $\Delta_2x'(0)$ is given by Eq. (21). The purpose in plotting and tabulating the ratio r_2 rather than the width Δ_2x' itself is in recognition of the fact (see Part II), that the dynamic functions are often measured as a function of T^{-1} rather than of x' (where T is the absolute temperature). When this is the case, it is convenient to obtain an experimental value for r_2 by dividing the observed width $\Delta_2(T^{-1})$ by the corresponding width for the case of a single relaxation (see Eq. (8) in Part II).

An asymptotic form for the peak width Δ_2x' which is valid for large β, may be derived from Eq. (17). The result (which is always less than the exact expression) is

$$\Delta_2x'(\beta)\sim 2\beta\sqrt{\ln 2}/2.303=0.723\beta\,. \tag{24}$$

For $\beta=7$, the exact value is 4.5% greater than that obtained from Eq. (24).

The way in which the three relaxation parameters may be obtained from experimental data for the J_2 function is summarized in the first row of Table 5. First, the width, relative to that of a single time of relaxation, r_2, is determined from the data and used to obtain β from the $r_2(\beta)$ plot of Fig. 3. Then the known value of β is used to read off the value $2f_2(0,\beta)$ from this same Figure. Comparison of this last result with the measured peak height, $J_2(0)=f_2(0,\beta)\cdot\delta J$, gives the value of δJ. Finally, as already mentioned, τ_m is obtained from the condition that $\omega\tau_m=1$ at the peak. Thus, it is possible to obtain the three parameters τ_m, β, and δJ from the peak location, width, and height, respectively. If one now wishes to check how well the lognormal distribution actually fits the entire peak, the normalized experimental curve may be plotted together with the calculated curves for the nearest β values from Table 2. If the lognormal distribution actually applies, the experimental curve will agree with the curve obtained by interpolation from the two closest theoretical curves.

It is interesting to note that the magnitude of the relaxation effect, δJ, can always be obtained, without a knowledge of β, from the area under the J_2 peak. It is readily shown, from Eqs. (10) and (12), that $\int_{-\infty}^{\infty}J_2dx=(\pi/2)\delta J$, i.e., the same result as in the case of a single relaxation time. Since area under an experimental peak is not conveniently obtained, it is interesting to see how accurately the product of height times width may be regarded as an indication of the area. For a single relaxation this product is $J_2(0)\cdot\Delta_2x'=(0.5720)\delta J$. The results of Fig. 3 make it possible to investigate how this product varies with β for the case of a lognormal distribution. The top curve of Fig. 3 shows this product, relative to that for a single relaxation of the same δJ, plotted as a function of β. It is clear that, unlike the area, the product of height and width does vary with β, but this variation is slow, going from a relative value of unity for $\beta=0$ to an asymptotic value of 1.120 for $\beta\rightarrow\infty$. (The latter value is obtained from Eqs. (22) and (24).)

The parameters of the relaxation may also be obtained from a knowledge of the function $J_1(x)$. Yager[9] has calculated the function $f_1(x',\beta)$ of Eq. (16) for several values of β, but unfortunately the increments both in x'

and in β are relatively large. Since the J_1 function is not usually studied experimentally as often, and with as great a precision, as the J_2 peak, we have not recomputed the function $f_1(x', \beta)$. Nevertheless, it is possible to make use of Yager's calculations to obtain a relevant parameter. In this connection, it is noteworthy that the absolute magnitude of the slope $\partial f_1(x', \beta)/\partial x'$ at $x'=0$ decreases steadily with increasing β. A knowledge of this slope could then be used to obtain β from experimental data. However, at small β-values the slope varies rapidly near the inflection point $(x'=0)$, so that an experimentally measured slope will always be somewhat lower in magnitude than the theoretical slope. It therefore appeared to the authors to be more useful to define a related quantity which measures the range of rapid decrease of the function f_1. This quantity is $\Delta_1 x'$, defined as the difference in the variable x' between the point at which $f_1=0.25$ and that at which $f_1=0.75$. For the case of a single relaxation time, the function J_1 gives the value $\Delta_1 x'=0.4722$. For the case of a Gaussian distribution, the dependence of $\Delta_1 x'$ on β is obtained from Yager's calculations and plotted in Fig. 4 relative to the value for $\beta=0$. This ratio $\Delta_1 x'(\beta)/\Delta_1 x'(0)$ is defined as $r_1(\beta)$ by analogy to Eq. (23). The plot of $r_1(\beta)$ vs β given in Fig. 4 may then be used to obtain β from the experimentally measured value of r_1. The asymptotic value of $\Delta_1 x'$ at large β-values is readily found, with the help of a table of the error integral, to be

$$\Delta_1 x' \sim 0.414\beta\,. \qquad (25a)$$

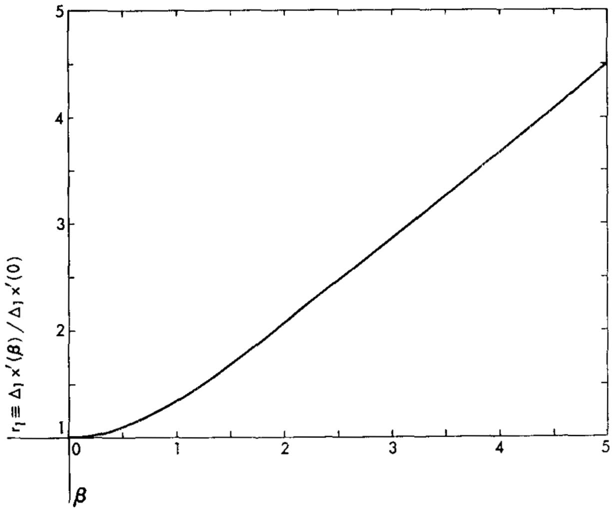
As for the additional parameters to be determined, it should be realized that $J_1(\omega=0)=J_R$ and $J_1(\omega=\infty)=J_U$, so that J_U and δJ are obtained directly. The final parameter τ_m is obtained from the symmetry of the function $f_1(x, \beta)$, since it is readily shown that, regardless of the value of β, $f_1=\frac{1}{2}$ when $x=0$ (or $\omega\tau_m=1$). The second row in Table 5 summarizes the methods for obtaining the parameters β, δJ, and τ_m from the J_1 function.

Table 4 **Results of numerical computations for the dependence of relative peak height, $2f_2(0, \beta)$, and relative peak width, $r_2(\beta)$ on the distribution parameter, β.**

β	$2f_2(0, \beta)$	$r_2(\beta)$
0.00	1.00000	1.00000
0.10	0.99752	1.00327
0.20	0.99024	1.01296
0.30	0.97866	1.02866
0.40	0.96346	1.04981
0.50	0.94540	1.07576
0.60	0.92520	1.10585
0.70	0.90350	1.13946
0.80	0.88088	1.17605
0.90	0.85774	1.21515
1.00	0.83448	1.25637
1.25	0.77722	1.36670
1.50	0.72316	1.48480
1.75	0.67332	1.60849
2.00	0.62798	1.73640
2.25	0.58698	1.86763
2.50	0.54998	2.00159
2.75	0.51664	2.13783
3.00	0.48654	2.27602
3.25	0.45934	2.41590
3.50	0.43466	2.55727
3.75	0.41224	2.69996
4.00	0.39182	2.84382
4.25	0.37314	2.98873
4.50	0.35604	3.13459
4.75	0.34032	3.28130
5.00	0.32584	3.42879
5.25	0.31248	3.57698
5.50	0.30010	3.72580
5.75	0.28860	3.87522
6.00	0.27792	4.02517
6.25	0.26796	4.17559
6.50	0.25866	4.32647
6.75	0.24996	4.47776
7.00	0.24180	4.62943

It should be noted that both J_1 and J_2 are often obtained in the same experiment. Provided that δJ is not too small, this quantity can then be obtained from the difference between J_U and J_R measured as the limiting values of J_1. The quantity β is usually best obtained, utilizing Fig. 3, from the width of the J_2 peak. Thus the height of the J_2 peak represents additional information which can be used to determine δJ, as described above. Agreement between the two values of δJ, obtained on the one hand from the height of the J_2 peak, and on the other from the limiting values of J_1, serves as a check on the validity of the lognormal distribution.

Figure 4 **Dependence of the relative range of decrease, $r_1(\beta)$, of the f_1-function on the distribution parameter, β.**



Finally, we turn to the problem of obtaining the relaxation parameters from static measurements. Here J_U is obtained from the instantaneous value of ε, and J_R from its final value (see Eq. (1)); therefore, as for the J_1 function, δJ is again measured directly. The problem then centers about means for obtaining τ_m and β from the normalized function, $\psi(y')$. For this purpose, the tabulation of the function $g(y', \beta)$ in Table 3 is most useful.[16] By analogy to the procedure for the f_1 function, it is convenient to define a quantity $\Delta y'$, which measures the range of rapid increase of the ψ function, and which is defined as the difference in the variable y' between the point at which $\psi=0.75$ (or $g=0.25$) and that at which $\psi=0.25$ (or $g=0.75$). For the case of a single relaxation time the value of $\Delta y'$ is 0.6829, while for the case of a Gaussian distribution, values of $\Delta y'$ may be obtained as a function of β from the computed results in Table 3. These values are given in Fig. 5. This plot may then be used to obtain β from the experimental value, $\Delta y'$, for the range of increase of the static function $\psi(y')$. For large β values, the function $g(y, \beta)$ goes asymptotically into the same error integral as the function $f_1(x, \beta)$. Accordingly, $\Delta y'$ asymptotically approaches

$$\Delta y' \sim 0.414\beta\,. \tag{25b}$$

The problem of obtaining the mean relaxation time, τ_m, is not as simple here as it was for the dynamic functions, since $\psi(y)$ does not have the symmetry property about $y=0$ which the functions f_1 and f_2 have about $x=0$. To determine how to proceed, we refer first to the case of a single time of relaxation, $\psi=1-e^{-t/\tau}$. The relaxation time τ is usually obtained from the time for the exponential to fall to a fixed fraction. Thus, for the case of a single relaxation, τ is equal to the time, $t_{1/e}$, for the quantity $1-\psi$ to fall to $1/e$; it is also related to the time, $t_{1/2}$, for ψ to reach the value 1/2, viz., $\tau=t_{1/2}/(\ln 2)=1.44t_{1/2}$. Now, in passing from the case of a single time of relaxation to one in which a lognormal distribution is involved, the same method may be employed to obtain τ_m. Thus, Fig. 6 shows a plot of $t_{1/2}/\tau_m$ as a function of β, obtained from the material in Table 3. This plot can be used to obtain τ_m, once β is obtained from the observed value of $\Delta y'$ and Fig. 5. It is important to note that the ratio $t_{1/2}/\tau_m$ is no longer equal to ln 2 when $\beta \neq 0$, and that correspondingly $t_{1/e}$ is no longer equal to τ_m. These changes with increasing β are not negligible and must be considered if precise values of τ_m are desired (as, for example, in obtaining an accurate value for the activation energy governing the quantity τ_m). The methods for obtaining the relaxation parameters from the static function are reviewed in the third row of Table 5.

Dependence of the distribution parameter on the temperature

In the previous section, methods were given for obtaining the distribution parameter, β, of the lognormal distribution, from the dynamic response functions $J_1(x)$ and $J_2(x)$ and from the static function $\psi(y)$. In Part II, consideration will be given to methods of obtaining the relaxation parameters from dynamic data in which tan ϕ is measured rather than J_2, and in which measurements are made as a function of temperature rather than of frequency. Thus, it can be assumed that we know how to obtain β from experimental data and that, by using a variety of both static and dynamic techniques, it is possible to obtain β over a range of temperatures. In the present section we consider the manner in which the parameter β varies with the temperature. Such considerations will bring out more clearly the theoretical significance of β.

Whenever the relaxation processes are controlled by atom movements, one may expect that each relaxation rate, τ^{-1}, will vary with temperature according to an Arrhenius relation, i.e., $\tau^{-1} \propto e^{-Q/RT}$, where Q is the activation energy for the process. Thus, we may write, for any relaxation time τ in the distribution,

$$\ln \tau = \ln \tau_0 + Q/RT\,. \tag{26}$$

In view of this equation, it becomes clear that a distribu-

Table 5 **Methods for obtaining relaxation parameters from dynamic and static functions.**

Function	*Parameter*		
	β	δJ	τ_m
$J_2(x')$	From peak width relative to that for a single relaxation, using Fig. 3 (or Table 4) and Eq. (21).	From peak height $J_2(0)=\delta J \cdot f_2(0, \beta)$, using $f_2(0, \beta)$ obtained from Fig. 3 or Table 4.	From peak position, using $\omega\tau_m=1$
$J_1(x')$	From "range of decrease," $\Delta_1 x'(\beta)$, relative to that for a single relaxation, using Fig. 4 and $\Delta_1 x'(0)=0.4772$.	$J_1(-\infty)-J_1(\infty)$	From center of symmetry: $J_1=J_U+\frac{1}{2}\delta J$ at $\omega\tau_m=1$
$\psi(y')$	From "range of increase," $\Delta y'$ (Fig. 5)	- - -	From $t_{1/2}(\beta)/\tau_m$ (Fig. 6)

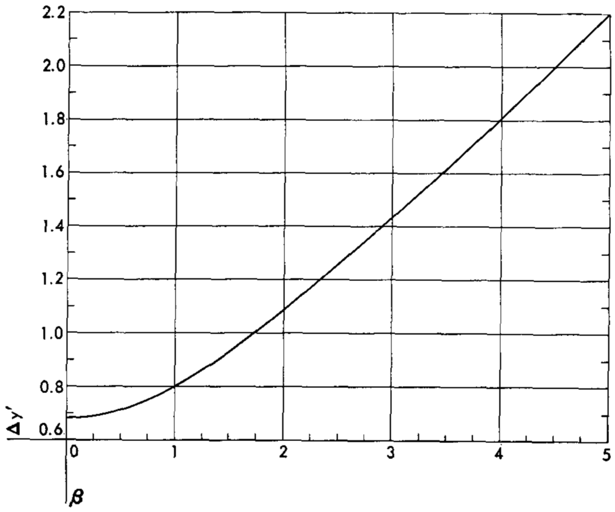


Figure 5 **Dependence of the range of increase, $\Delta y'$, of the g-function on the distribution parameter, β.**

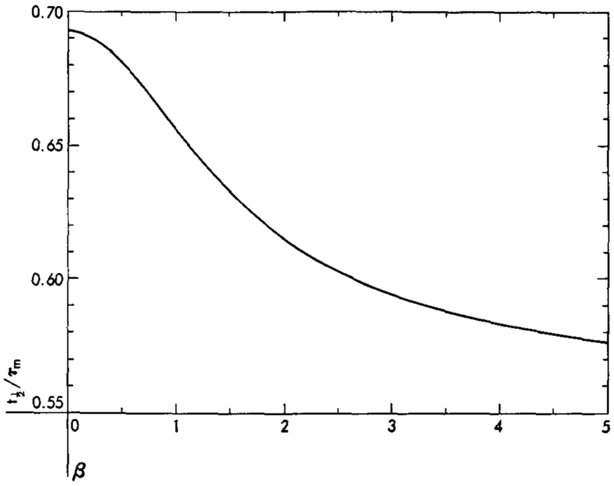


Figure 6 **Dependence of $t_{1/2}/\tau_m$ on the distribution parameter, β. ($t_{1/2}$ is the time for the function $\psi(t)$ to reach the value 1/2, and τ_m is the most probable relaxation time.)**

tion in τ is due either to the existence of a distribution in the temperature independent parameter,[18] τ_0, or in the activation energy, Q, or in both. The quantities Q and $\ln\tau_0$ may be regarded as the fundamental parameters which determine the relaxation time, τ. (The choice of $\ln\tau_0$ rather than τ_0 is based on the fact that $\ln\tau_0$ is linearly related to the entropy of activation.)

In order to obtain the relationship of the parameter β to the distributions in the quantities $\ln\tau_0$ and Q, we make use of the following theorem, which is easily verified: If two variables u and v are linearly related, i.e., $u=cv+d$ (where c and d are constants), and if values of u are distributed about a mean value u_m according to a Gaussian (normal) distribution of the form $\exp[-(u-u_m)/\beta_u]^2$, then the values of v are also distributed in a Gaussian manner, with distribution parameter given by

$$\beta_v=\beta_u/c \tag{27}$$

and mean value $v_m=(u_m-d)/c$.

In applying this theorem, we consider first the case in which the distribution is only in the quantity $\ln\tau_0$, i.e., when the activation energy of all contributing relaxation processes is a constant. From the above theorem and Eq. (26) it is clear that a Gaussian distribution in $\ln\tau_0$ results in a Gaussian distribution in $\ln\tau$, with the same distribution parameter β. Next we consider the other limiting case, where the distribution is only in the activation energy, and τ_0 is a constant for all relaxation processes. Under these circumstances, a Gaussian distribution in Q about a mean value Q_m with a distribution parameter β_Q is equivalent to a Gaussian distribution in $\ln\tau$ with parameter

$$\beta=\beta_Q/RT \qquad (Q\text{-distribution only}). \tag{28}$$

Since β_Q may be considered independent of temperature (insofar as the activation energies themselves are independent of temperature), we are led to the conclusion that Eq. (28) defines the temperature dependence of β for this limiting case. So far, then, we have found that either a Gaussian distribution in $\ln\tau_0$ (with $Q=$ constant) or a Gaussian in Q (with $\tau_0=$constant) gives rise to a Gaussian in $\ln\tau$. In the former case, the parameter β is independent of temperature; in the latter case β varies inversely as T. These conclusions are rigorous, in the sense that they follow directly from Eq. (26) and the concept that the distributions of Q and τ_0 are independent of temperature.

To consider the more general case in which distributions exist for *both* τ_0 and Q, an assumption is needed concerning the relation between τ_0 and Q. One reasonable assumption that can be made, without further knowledge of the system being studied, is that τ_0 and Q do not vary independently, but that both depend on a single internal parameter p, e.g., local composition or local order. The dependence of $\ln\tau_0$ and Q on p can be described by a Taylor expansion about the mean value p_m in which only the first derivative term is retained (certainly this is approximately correct if the distribution is not a broad one). Thus

$$\left.\begin{aligned}\ln\tau_0-\ln\tau_{0m}&=C(p-p_m)\\ Q-Q_m&=D(p-p_m)\end{aligned}\right\}, \tag{29}$$

where C and D are constants. If we now assume that the parameter p varies according to a Gaussian distribution about a most probable value p_m, then Q must be distributed as a Gaussian about the value Q_m and similarly for $\ln\tau_0$ about $\ln\tau_{0m}$. The variable p may be eliminated from Eqs. (29) to obtain

$$\ln\tau_0-\ln\tau_{0m}=K(Q-Q_m)\,, \tag{30}$$

where $K=C/D$. Finally, Eq. (30) may be combined with (26) and (13) to obtain

$$z\equiv\ln(\tau/\tau_m)=\left(K+\frac{1}{RT}\right)(Q-Q_m)\,, \tag{31}$$

where τ_m is defined by

$$\ln\tau_m=\ln\tau_{0m}+Q_m/RT\,. \tag{32}$$

The theorem which led to Eq. (27) then establishes that the distribution of the quantity z is Gaussian about the value $z=0$, and that its width β is related to the width, β_Q, of the Q-distribution according to

$$\beta=\beta_Q\left(K+\frac{1}{RT}\right)=\beta_0+\frac{\beta_Q}{RT}\,. \tag{33}$$

Note that, from Eq. (30), $\beta_0=K\beta_Q$ is the distribution parameter for the quantity $\ln\tau_0$. Since the parameters β_0 and β_Q are regarded as being independent of temperature, it is concluded that if the assumptions which led to Eq. (33) are correct, the parameter β deduced from measurements at different temperatures should plot as a straight line against T^{-1} with slope equal to β_Q/R and intercept the axis $1/T=0$ at β_0. Such a plot enables us to obtain separately the distribution parameters for the $\ln\tau_0$ and Q distributions. The two limiting cases, of either a distribution in $\ln\tau_0$ or in Q only, can now be regarded as special cases of the present more general result. It should be kept in mind that the results obtained for these two limiting cases were rigorous while Eq. (33) is based on the assumptions involved in Eqs. (29). It may be pointed out, however, that not only are these assumptions reasonable, but that the result obtained, i.e., Eq. (33), is the simplest equation for covering the range between the two limiting cases. The usefulness of Eq. (33) is that it provides a means of separating out the extent to which the distribution in $\ln\tau$ represents a distribution in $\ln\tau_0$ and one in Q.

A somewhat different result is obtained if it is assumed, in place of Eq. (29), that $\ln\tau_0$ and Q are uncorrelated, but that both distributions are Gaussian. Under these circumstances the temperature dependence of β is given as[19]

$$\beta=[\beta_0^2+(\beta_Q/RT)^2]^{1/2}\,. \tag{34}$$

It would be difficult to distinguish Eqs. (33) and (34) experimentally unless measurements were made over a wide range of temperature. In the absence of such data a choice between Eqs. (33) and (34) must be based on what is known about the specific relaxation phenomenon. For processes controlled by elementary atom movements in crystals, a correlation between $\ln\tau_0$ and Q can be expected. Since the applications to be considered in Part II fall into this category, Eq. (33) will be employed in our analysis.

Summary

The content of the present paper may be summarized as follows:

(1) Equations for the static and dynamic response functions are reviewed for the case of a lognormal distribution of relaxation times.

(2) Numerical computations for the integrals involved in the expressions for the static function and the J_2 function are presented.

(3) Methods are presented for obtaining the three relaxation parameters δJ, τ_m, and β from the response functions.

(4) It is shown how the variation of β with temperature can be used to determine separately the parameters for the distribution in $\ln\tau_0$ and that in the activation energy.

Appendix I. Application of the Gaussian distribution to anelastic functions which correspond to a specified strain.

In the case of anelastic relaxation one may study the relaxation of stress under a given (static) strain or, in the dynamic case, the behavior of a system under a specified periodic strain. In the static case, the appropriate function to consider is the *stress relaxation function*, $\phi(t)$, defined by

$$\frac{\sigma(t)}{\varepsilon}=M_R+\delta M\phi(t)\,, \tag{A-1}$$

where $\sigma(t)$ is the dependence of stress on time, ε the constant specified strain, M_R the "relaxed modulus" $(=J_R^{-1})$ and

$$\delta M=M_U-M_R\,, \tag{A-2}$$

where $M_U=J_U^{-1}$ is the "unrelaxed modulus." The function $\phi(t)$ therefore ranges from $\phi(0)=1$ to $\phi(\infty)=0$. Similarly, the dynamic properties are describable in terms of the complex modulus

$$M^*=M_1(\omega)+iM_2(\omega) \tag{A-3}$$

and therefore by the variation of M_1 and M_2 with frequency. The existence of a distribution in relaxation times means here that M_1, M_2, and ϕ are expressible in the forms[3]

$$M_1(\omega)=M_R+\delta M\int_{-\infty}^{\infty}\frac{\Phi(\ln\tau)\omega^2\tau^2}{1+\omega^2\tau^2}\,d\ln\tau \tag{A-4}$$

$$M_2(\omega)=\delta M\int_{-\infty}^{\infty}\Phi(\ln\tau)\frac{\omega\tau}{1+\omega^2\tau^2}\,d\ln\tau \tag{A-5}$$

$$\phi(t)=\ {}_{-\infty}^{\infty}\Phi(\ln\tau)e^{-t/\tau}\,d\ln\tau\,, \tag{A-6}$$

where the distribution function $\Phi(\ln\tau)$ obeys the condition

$$\int_{-\infty}^{\infty}\Phi(\ln\tau)\,d\ln\tau=1\,. \tag{A-7}$$

In general, the Φ distribution is *not* the same as the Ψ distribution which is used to express the functions J_1, J_2, and ψ. If one assumes that the Φ distribution is log-normal (and hence that the Ψ distribution is not) then, using the variables x, y, and β defined in Eqs. (14) and (15), the following relationships are obtained:

$$\frac{M_U - M_1}{\delta M} = f_1(x, \beta) \tag{A-8}$$

$$M_2/\delta M = f_2(x, \beta) \tag{A-9}$$

$$\phi(y) = g(y, \beta)\,. \tag{A-10}$$

In these equations the functions f_1, f_2, and g are precisely those defined in Eqs. (16) to (18) and tabulated in Tables 2 and 3 and in Yager's article.[9]

It is therefore concluded that the integrals $f_1(x, \beta)$, $f_2(x, \beta)$ and $g(y, \beta)$ evaluated numerically by Yager and by the present authors are equally useful in the analysis of the "strain-given" functions M_1, M_2, and ϕ, when the Φ distribution is Gaussian, as it is for the "stress-given" functions J_1, J_2, and ψ, when the Ψ distribution is Gaussian. The methods discussed in Section B (Table 5) and in Part II (Table 1) may then be used in the evaluation of the parameters β, τ_m, and δM which appear in Eqs. (A-8) to (A-10).

References and footnotes

1. We will use the usual terminology that "static" properties are those measured under conditions in which either σ or ε is held constant for some period of time, while "dynamic" properties are those for which σ and ε are periodic.
2. In anelasticity, the meaning of the J's depends on the nature of the stress which is applied. For the purpose of the present paper, the particular type of stress is best left unspecified. However, it is important to note that complications can arise in the interpretation of the response functions in crystals, and even in isotropic materials when the mode of deformation is such that both shear and bulk relaxation occur simultaneously. See Ref. 3.
3. A. J. Staverman and F. Schwartzl, *Die Physik der Hochpolymeren,* Vol. IV, Chap. I. (Ed. H. A. Stuart). J. Springer, Berlin, 1956.
4. In the anelastic case, one sometimes studies the relaxation of stress at constant strain or, in the dynamic case, the behavior of a system under a given sinusoidal strain. It will later be shown in the Appendix that the mathematical functions computed in this paper for the calculation of the functions $\psi(t)$, $J_1(\omega)$, and $J_2(\omega)$, are also applicable to the situation in which the strain is prescribed rather than the stress.
5. In anelasticity, it is customary to distinguish between the "relaxation time at constant stress" which appears above, and the "relaxation time at constant strain" which is obtained from a stress relaxation experiment at constant strain. In the literature on high polymers the former is generally called the *retardation time* and the latter the *relaxation time.* In order to be consistent with the terminology of dielectric relaxation, however, we have chosen the simplified terminology quoted above.
6. In the study of dielectric relaxation ϕ is called the *loss angle*, while in anelasticity tan ϕ is the *internal friction.*
7. E. Wiechert, *Annalen d. Physik,* **50,** 335, 546 (1893).
8. K. W. Wagner, *Annalen d. Physik,* **40,** 817 (1913).
9. W. A. Yager, *Physics,* **7,** 434 (1936).
10. C. Zener, *Elasticity and Anelasticity of Metals,* U. Chicago Press, Chicago, 1948.
11. B. Gross, *Mathematical Structure of the Theories of Viscoelasticity,* Hermann and Co., Paris, 1953, Ch. XIII.
12. A. V. Tobolsky, *Properties and Structure of Polymers,* J. Wiley, New York, 1960.
13. Appendix I shows how the mathematical functions computed here may be applied to the situation in which the strain rather than the stress is the prescribed variable.
14. In addition, recalculation has shown that a number of Yager's figures are slightly in error.
15. By making the change in variable $z = \beta u$ in Eq. (17), it readily follows that the asymptotic form of the function for large β is $f_2(x, \beta) \sim (\sqrt{\pi}/2\beta)\exp(-x^2/\beta^2)$.
16. In the case of dielectric relaxation it is not the electric displacement (or polarization) which is measured directly but rather the current density, which is a time derivative of the polarization. For such measurements, the derivative $\partial g(y', \beta)\partial y'$ is the more useful function. In fact, using the work of Wagner,[8] Jahnke and Emde[17] tabulate a function which is, in our notation, $\sqrt{\pi}\beta e^{-y}\partial g(y, \beta)/\partial y$. These tables cover only cases of $\beta > 1$ and for both β and y' spaced at inconveniently large intervals. The present results for $g(y', \beta)$ in Table 4 may be readily differentiated numerically to yield more complete tables of the type given by Jahnke and Emde.
17. E. Jahnke and F. Emde, *Tables of Functions,* Fourth Ed., Dover, N. Y., 1945, p. 39.
18. The reciprocal of τ_0 is commonly called the *frequency factor* or *attempt frequency*. The fact that τ_0 and Q may be taken as independent of temperature to a high degree of approximation is justified by a combination of the following: (a) the empirical fact that Eq. (26) is very accurately obeyed for atom movements in crystals; (b) the theoretical interpretation of the quantities τ_0 and Q in terms of the thermodynamical theory of activated processes. See, for example, D. Lazarus' article in *Solid-State Physics,* vol. 10, Academic Press, New York, 1960.
19. The authors are grateful to J. R. MacDonald and C. A. Barlow, Jr., of Texas Instruments Inc., for having pointed out this result. MacDonald has also independently derived Eq. (28), in a paper to be published in *J. Chem. Phys.*

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