LETTER TO THE EDITOR

A Note on T/η Scaling of Dynamic Light-Scattering Spectrum

Dear Sir:

Newman and Carlson (1980) have mentioned, in a discussion based on Pecora's formula (1968), that the field correlation functions $g^{(1)}(\tau)$ of light scattered from dilute solutions of monodisperse rigid rods at different temperatures can scale as $(T/\eta)\tau$, where T/η is the ratio of the absolute temperature to the solvent viscosity (theorem 1). Our discussion in this letter concerns $g^{(1)}(\tau)$ for very large KL (K and L are the length of the scattering vector and the contour length of the scatterer, respectively), where contributions from internal micro-Brownian motions are significant. To verify this theory of T/η scaling, they studied a sample of fd virus as a prototype of a rigid-rod system. A previous study (Newman et al., 1977) concluded that fd virus (~0.9 μm × 9 nm) behaves hydrodynamically as a rigid rod in solution. Their results clearly showed that over a range of 4°–45°C, $g^{(1)}(\tau)$ for fd virus do indeed scale as $(T/\eta)\tau$.

Generally speaking, the converse of theorem 1 is not always true; that is, the presence of T/η scaling does not necessarily mean that the scatterer is a rigid rod. Based on a computer simulation of $g^{(1)}(\tau)$ for dilute solution of monodisperse filamentous particles, Maeda and Fujime (1981) showed that the light-scattering data on fd virus by Newman and Carlson could reasonably be interpreted if fd virus was assumed to be a semiflexible filament having a flexibility parameter γL of ~0.1. Here we would like to propose a more general theorem on T/η-scaling of $g^{(1)}(\tau)$: if γL does not depend on temperature and/or solvent conditions, $g^{(1)}(\tau)$ at different temperatures and/or solvent conditions can scale as $(T/\eta)\tau$ independently of the γL value (theorem 2). The converse of this theorem is true. Therefore we have a corollary; the absence (presence) of T/η-scaling of $g^{(1)}(\tau)$ at different temperatures means that the γL of the scatterer does (does not) depend on temperature. Theorem 2 includes theorem 1 at $\gamma L \ll 1$, and the theorem for the Rouse-Zimm chain at $\gamma L \gg 1$. To sum up, when the γL value, or the conformation, of the filament depends on the environmental condition, $g^{(1)}(\tau)$ cannot scale with T/η but with a combination of T/η and γL. When $g^{(1)}(\tau)$ obeys T/η scaling within experimental errors, we have to examine carefully a possibility that the scatterer is a flexible filament whose flexibility parameter is independent of, or weakly dependent on, temperature.

Derivation of theorem 2 is given below. In what follows, we refer to equations from Maeda and Fujime (1981) as MF and the equation number, e.g., MF05. We neglect a hydrodynamic interaction. From Eq. MF20, we have $g^{(1)}(\tau) = \exp(-D_0K^2\tau)\,S(\tau)/S(0)$ and

$$S(\tau) = (1/L)^2 \int\int_{-L/2}^{L/2} \exp\left\{-(K^2/6) \sum_n{}' \langle q_n^2\rangle [Q(n,s)^2 + Q(n,s')^2 - 2Q(n,s)Q(n,s') \exp(-\tau/\tau_n)]\right\} ds\,ds', \quad (1)$$

where we have from Eqs. MF03, 09, 11, and 16

$$\langle q_n^2\rangle = (3k_BT/\lambda_n) = (4\gamma L)L^3/[(\beta_nL)^4 + (4\gamma L)(L^2/\langle R^2\rangle)(\beta_nL)^2] \quad (2)$$

$$\tau_n^{-1} = \lambda_n/\zeta = (3k_BT/\zeta)/\langle q_n^2\rangle. \quad (3)$$

Because the friction constant ζ is proportional to η, the relaxation rate τ_n^{-1} of the nth internal mode is also proportional to T/η. As eigenvalues β_nL are determined only by the γL value, both eigenfunctions $Q(n,s)$ and $\langle q_n^2\rangle$ depend only on γL. Thus we have theorem 2. This theorem has been derived from a particular model of polymer dynamics (Harris and Hearst, 1966; Fujime and Maruyama, 1973). However, the theorem is true insofar as (*a*) the Langevin equation for polymer dynamics is linear, where complete separation of normal modes will result with a separation constant λ_n and Eqs. 1 and 3 hold as they are, and (*b*) the force constant(s) is proportional to k_BT, where λ_n is proportional to k_BT and $\langle q_n^2\rangle$ does not depend explicitly on temperature as in Eq. 3. Even for polydisperse samples, the above result is valid provided that the size distribution of particles is independent of temperature.

The concept of scaling is very useful in various branches of physics. The proposed theorem is believed to be useful in dynamic light scattering, although its application may sometimes not be straightforward because of possible complexities associated with changes in environmental conditions.

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