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Comparison of theories for the translational and rotational diffusion coefficients of rod-like macromolecules. Application to short DNA fragments

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Two theories relating the translational and rotational diffusion coefficients D_t and D_r of a rod-like macromolecule to its length and diameter, proposed by Broersma [J. Chem. Phys. 74, 6989 (1981)], and Tirado and García de la Torre [J. Chem. Phys. 71, 2581 (1979); 73, 1986 (1980)] are shown to predict different values of the coefficients for a particle of given dimensions. Next, we use the two theories to analyze existing experimental data of sedimentation coefficients s and translational and rotational diffusion coefficients of short DNA fragments, and obtain values of the hydrated diameter of DNA d which is treated as an adjustable parameter. The results are compared with the expected value, $d \simeq 26\text{Å}$. This comparison favors clearly the Tirado–Garcia de la Torre theory in the case of D_t and s. For D_r, and using a rise per base pair $r = 3.4$ Å, this theory gives best agreement for all the data examined, while when $r = 3.3$ Å, the agreement depends on the source of data.

INTRODUCTION

The rigid rod, or cylinder, is a macromolecular model of great relevance. There is a large number of biopolymers including some polypeptides, proteins, nucleic acids, and viruses which under certain conditions exhibit the typical rod-like conformation, and their hydrodynamic properties can therefore be analyzed in terms of cylindrical models.

In 1960, Broersma[1,2] presented a theory for the translational and rotational motions of cylinders, and his results have been in widespread use over the past 20 years. The limiting form of his equations for very long cylinders (axial ratio $L/d \rightarrow \infty$) has been confirmed by other workers.[3,4] However, such limiting form is not valid for finite cylinders (say $L/d < 50$) owing to end effects, and it is in fact this range of small or moderate axial ratios where most biologically interesting applications fall. In his papers Broersma reported also results for finite cylinders, but it was later realized that there was a poor agreement between his results and those from other experimental or theoretical works.

Then, our group[5–7] carried out a numerical evaluation of the translational and rotational coefficients of cylinders based on a rigorous version of the Kirkwood-Riseman theory with extrapolation to the shell model limit.[8,9] Our results agreed very well with experimental data for short macroscopic cylinders and deviated remarkably from the results obtained by Broersma.

Now, we know that Broersma himself found some defects in his old theory, obtaining improved results which were first communicated privately[10] and later in a fuller form.[11] It is therefore interesting to compare the two currently available theories; this is one of the purposes of this paper.

Our second purpose is to analyze existing data of translational (sedimentation) and rotational coefficients of short DNA fragments. With the recent advent of endonucleases, DNA fragments with a perfectly known number of base pairs (n) have been prepared and characterized using sedimentation,[12] electric birefringence,[13–16] linear dichroism,[17,18] and dynamic light scattering.[17,19] The analysis of the hydrodynamic properties so obtained can be made without interference from polydispersity. As the molecular weight increases, the flexibility of the double helix gains importance, and the persistence length can be estimated from the equations of Hagerman and Zimm.[20] However, for restriction fragments as short as 40–50 base pairs, the hydrodynamic behavior must strictly coincide with that of straight cylinders, which is determined by just the particle's length and diameter. As n is well defined for a given fragment, its length is in turn determined by an unique constant, the rise per base pair (r). Here we will compare properties of DNA restriction fragments with predictions of the two theories, regarding d as an adjustable parameter.

THEORIES

For a cylinder of length L and diameter d, the translational diffusion coefficients for lengthwise and sidewise motion $D_t^{\parallel}$ and $D_r^{\perp}$ can be expressed as

$$2\pi\eta_0 L D_t^{\parallel}/k_B T = \ln p + \nu_{\parallel} \qquad (1)$$

and

$$4\pi\eta_0 L D_r^{\perp}/k_B T = \ln p + \nu_{\perp}, \qquad (2)$$

where $p = L/d$, $k_B T$ is Boltzmann factor, and η_0 is the viscosity of the solvent. The macroscopic diffusion coefficient D_t corresponds to motion in a random direction and is given by

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$$3\pi\eta_0 LD_t/k_B T = \ln p + \nu. \tag{3}$$

The sedimentation coefficient s can be obtained from D_t using the Svedberg formula

$$s = LM_L(1 - \bar{v}\rho)D_t/RT. \tag{4}$$

M_L is the molecular weight per unit length, $\bar{v}$ is the partial specific volume of the macromolecule, and ρ is the solution density.

The rotational diffusion coefficient for reorientation of the cylinder axis D_r can be written as

$$\pi\eta_0 L^3 D_r/3k_B T = \ln p + \delta_\perp. \tag{5}$$

In Eqs. (1)–(3) and (5), the quantities on the right-hand side adding to $\ln p$ are the so-called end-effect corrections, which are functions of p and converge to asymptotic values when $p \to \infty$. There is a relationship between $\nu_\parallel$, $\nu_\perp$ and ν:

$$\nu = (\nu_\perp + \nu_\parallel)/2. \tag{6}$$

All the theories agree in the functional forms of Eqs. (1)–(3), (5), and (6), and differ in the dependence of the end-effect corrections on p.

Tirado and García de la Torre[5,6] obtained numerical values of $\nu_\parallel$, $\nu_\perp$, ν and $\delta_\perp$ as functions of p. Simple quadratic fits to $1/p$ can be used for interpolation[7]:

$$\nu_\perp = 0.839 + 0.185/p + 0.233/p^2, \tag{7}$$

$$\nu_\parallel = -0.207 + 0.980/p - 0.133/p^2, \tag{8}$$

$$\nu = 0.312 + 0.565/p - 0.100/p^2, \tag{9}$$

and

$$\delta_\perp = -0.662 + 0.917/p - 0.050/p^2. \tag{10}$$

Here we would like to make some comments on the validity of Eqs. (7)–(10). The data from which these equations have been calculated were obtained for cylinders of finite length modeled as stacks of rings, and extrapolated to zero bead size.[5,6] The models had values of p in the range of about 2–20. The extrapolation to infinite p failed to reproduce the correct limiting values previously obtained by other workers.[1–4] This was already pointed out in the original references,[5,6] and the origin of such failure could be a strong curvature in the vicinity of $p^{-1} = 0$. However, we believe that the results in Refs. 5 and 6 for short cylinders are correct. In fact, our results were in excellent agreement with experimental data for $p < 30$ (see Fig. 3 in Ref. 5 and Fig. 3 in Ref. 6). Therefore, we think that the interpolation equations (7)–(10) are correct for, approximately, $2 < p < 30$ (the DNA fragments which we shall study later fall within this range). Conversely, these equations should not be used for very long rods like some filamentous viruses, although it is doubtful that such long rods are strictly rigid and straight in solution.

On the other hand, the new Broersma's equations reported by Newman et al.[10] are

$$\nu_\perp = 0.50 + 4.2 (1/\ln 2p - 0.39)^2, \tag{11}$$

$$\nu_\parallel = -0.58 + 7.4(1/\ln 2p - 0.34)^2, \tag{12}$$

and

$$\delta_\perp = -0.76 + 7.5(1/\ln 2p - 0.27)^2 \tag{13}$$

valid[11] for $0.15 < 1/\ln 2p < 0.35$, or $400 \gtrsim p \gtrsim 9$.

Very recently, Broersma[11] has published more com-

plete results, applicable over a wider range of p, in the form of polynomials in $(\ln 2p)^{-1}$:

$$\nu_\perp = 0.866 - 0.15/\ln 2p - 8.1/(\ln 2p)^2$$
$$+ 18/(\ln 2p)^3 - 9/(\ln 2p)^4, \tag{14}$$

$$\nu_\parallel = -0.114 - 0.15/\ln 2p - 13.5/(\ln 2p)^2$$
$$+ 37/(\ln 2p)^3 - 22/(\ln 2p)^4, \tag{15}$$

$$\delta_\perp = -0.446 - 0.2/\ln 2p - 16/(\ln 2p)^2$$
$$+ 63/(\ln 2p)^3 - 62/(\ln 2p)^4. \tag{16}$$

These equations are supposed to be valid for $1/\ln 2p < 0.45$, or $p > 4.6$. Note that the first constants in the right-hand side of Eqs. (11)–(16) differ from those in the original references[10,11] because we prefer to add the end-effect correction to $\ln p$ instead of substracting it from $\ln 2p$.

Although the detection of $D_t^\parallel$ and $D_t^\perp$ separately is becoming possible,[21] here we analyze only D_t. Figure 1 displays the values of the normalized translational and rotational diffusion coefficients according to our calculations and the two versions of the Broersma theory. The two sets of Broersma's formulas give practically the same results for $p > 9$, and therefore they will not be distinguished hereafter.

Figures 1(a) or 1(b) can be used to obtain by interpolation the diameter if the length and one of the diffusion coefficients of the macromolecule are known. Of course, it is also possible to obtain the length from one of the diffusion coeffi-
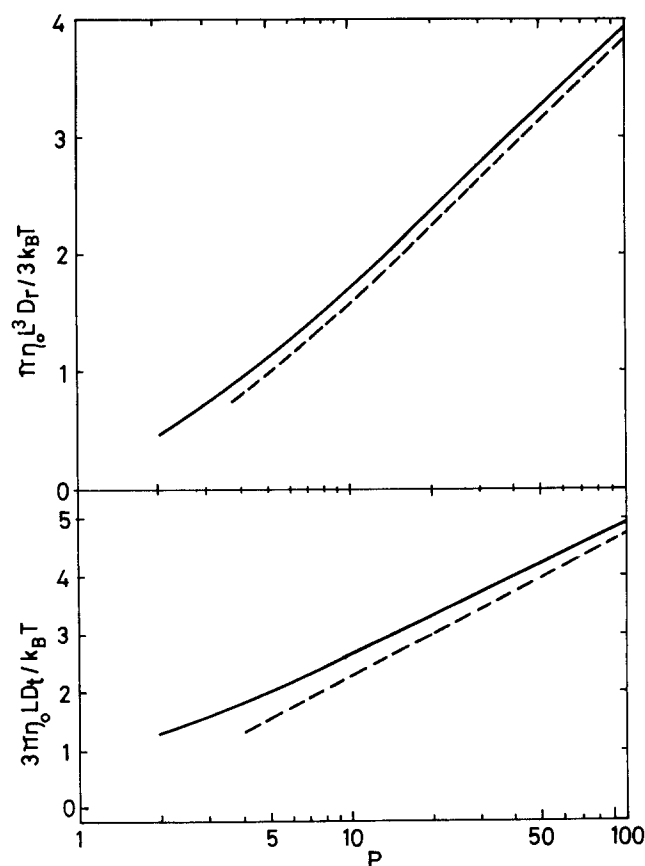

FIG. 1. Bottom: plot of $3\pi\eta_0 L\, D_t/k_B T$ vs p. (---), Broersma (Refs. 10 and 11). Eqs. (11)–(13) or (14)–(16); the two theories give nearly identical results. (——) This work. Top: Similar plot of $\pi\eta_0 L^3 D_r/k_B T$ vs p.
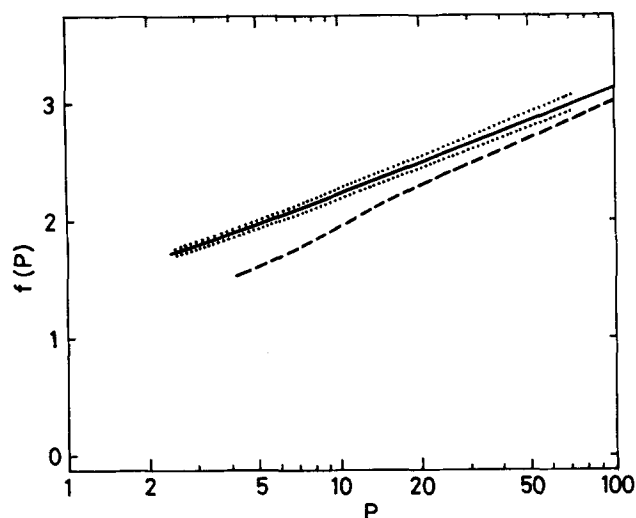
FIG. 2. Plot of $f(p)$ vs p. (---) Broersma; (—) this work. (···), limits for the $f(p)$ function with relative errors of $\pm 1\%$ in D_t and $\pm 3\%$ in D_r.

cients and an estimate of the diameter. However, if one knows both D_t and D_r for a given macromolecule, its dimensions, d and L, can be obtained simultaneously using the $f(p)$ function,[22]

$$f(p) \equiv (9\pi\eta_0/k_B T)^{2/3} D_t / D_r^{1/3} \qquad (17a)$$

$$= (\ln p + \nu)/(\ln p + \delta_\perp)^{1/3}. \qquad (17b)$$

This function is plotted in Fig. 2 for both the Broersma and the Tirado–Garcia de la Torre theories.

To ascertain the accuracy of the theories, their results should be compared with experimental data. For macroscopic cylinders of precisely known length, the comparison with data reported by Happel and Brenner[23] and Broersma[1] has been already presented,[6,7] and favors clearly our equations. In the next section we shall again compare the performance of the two theories in the analysis of experimental data for short fragments of DNA.

APPLICATION TO DNA FRAGMENTS

A rather definitive test of the performance of the two theories can be made by comparing their predictions with literature data on sedimentation and translational and rotational diffusion coefficients of rigid, rod-like DNA fragments. The dimension to which both D_t (or s) and D_r are least sensitive is d, the hydrated diameter of the double helix. This implies that if d is considered as an adjustable parameter (the length can be obtained from n and the rise per base pair) the value obtained by fitting experimental data can be greatly influenced by possible imperfections of the theories. In fact, the right-hand side of, for instance, Eq. (3) can be written as $\ln L$-$\ln d + \nu$, so that small errors in ν could cause small errors in $\ln d$ but large errors in d.

It is now well accepted that the hydrated diameter of DNA is nearly 26 Å. This figure has been confirmed by recent simulations of DNA hydration by Corongiu and Clementi,[24] who showed that nearly all the water molecules are within 13 Å from the helical axis. Thus, the results of fitting experimental data with the two theories can be checked against this expectation. Furthermore, as the theories give

both D_t and D_r, the agreement between the values of d for each property can be taken as a self-consistency test.

RESULTS

Kovacic and Van Holde[12] have measured the sedimentation coefficients $s_{20,w}$ of DNA fragments as short as 50 base pairs. Their experimental data are plotted in Fig. 3. On the other hand, theoretical results for the variation of $s_{20,w}$ with n can be obtained from Eqs. (3) and (4), since their combination with $L = nr$ gives

$$s = \frac{M_L(1 - \bar{v}\rho)}{3\pi\eta_0 N_A} (\ln n + \ln r - \ln d + \nu), \qquad (18)$$

where N_A is Avogadro's number. For the other quantities in Eq. (18) we use the following values[25]: $M_L = 195$ dalton/Å, $1 - \bar{v}\rho = 0.457$, and $\eta_0 = 0.0100$ P. In Fig. 3 we have also plotted the theoretical curves for several values of d obtained using the ν of Tirado and Garía de la Torre. It is seen that the experimental data begin to deviate from the cylindrical behavior at $n \simeq 200$, which is due to the onset of flexibility. Taking only the values for $n = 50, 94, 117, 145,$ and 160, by numerical fitting to the theoretical curves we find a best value of $d = 25$ Å. If curves from Broersma's theory are drawn in Fig. 3, they show a similar aspect, but for a given d they fall appreciably lower that ours. The best fit for Broersma's theory is found for $d = 18$ Å.

Mandelkern et al.[19] have reported light scattering decay times $\tau^{(t)}$ from which D_t can be obtained as

$$D_t = 1/2q^2\tau^{(t)}. \qquad (19)$$
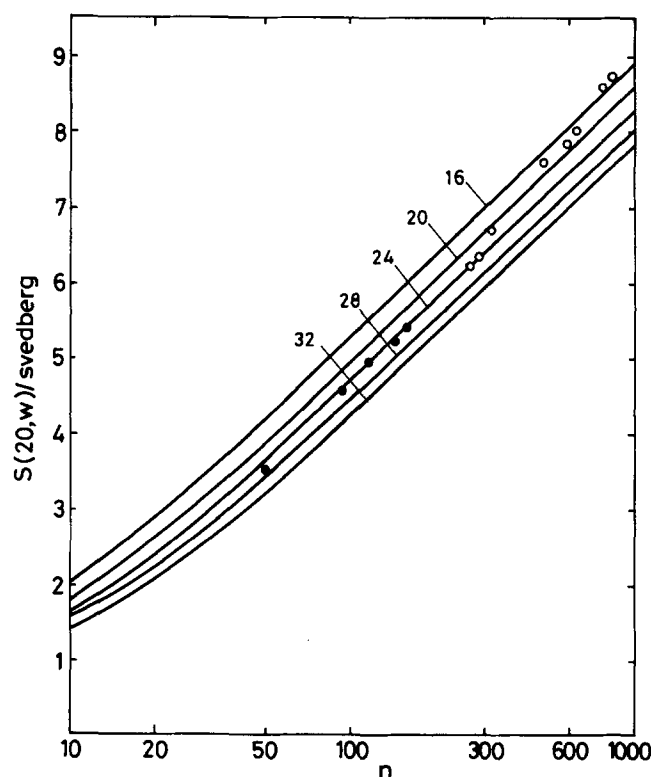

FIG. 3. Variation of $s_{20,w}$ with n. (●) Experimental data (Ref. 23) with $n < 200$. (○) Experimental data with $n > 200$. The curves are calculated from the Tirado–García de la Torre formulas for the indicated values of d in Å.
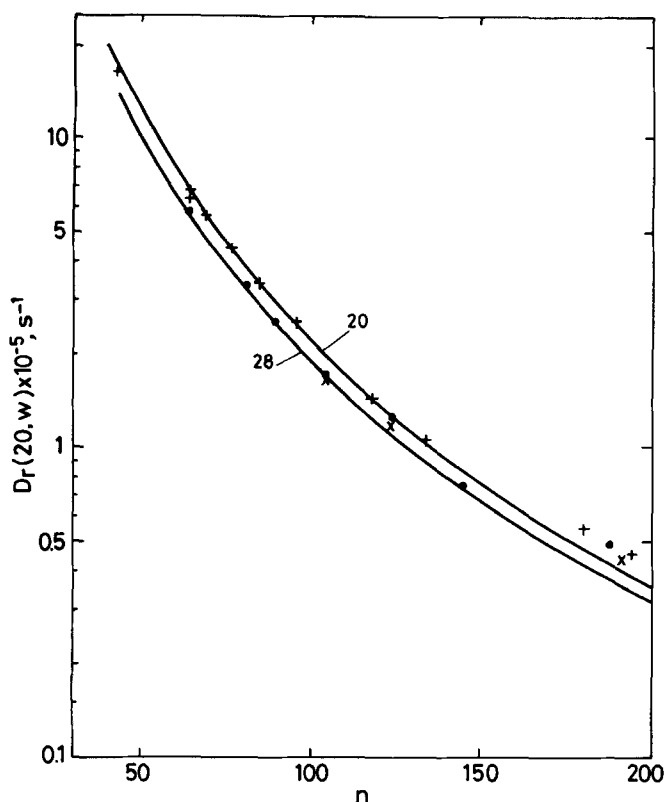
FIG. 4. Variation of $D_r(20,w)$ with n. Experimental data: (●) Elías and Eden (Ref. 13); (×) Hagerman (Ref. 16); (+) Dieckmann et al. (Ref. 18). The curves are calculated from the Tirado–García de la Torre formulas for the indicated values of d in Å.

The experimental value of q was not given explicitly, but can be calculated from the theoretical decay times presented in their paper. Thus, we find $q = 2.42 \times 10^5$ cm^{-1}. The D_t's of Mandelkern et al. for $n = 89,104,124$, and 188 follow well within experimental error the theoretical predictions from the Tirado–García de la Torre and Broersma theories for $d = 21$ and 15 Å, respectively.

Now we turn to the analysis of the rotational diffusion data. Eden and co-workers[13,14,19] have reported several sets of experimental D_r's obtained from electric birefringence decays. The same technique has been used by Hagerman and

Zimm,[20] and data from the electric dichroism study of Diekcmann et al.[18] are also available. In Fig. 4 we have plotted the experimental data for $n < 200$ along with theoretical curves for $d = 20$ and 28 Å. The start of chain flexibility, manifested by a clear separation between the trends of the theoretical and experimental values, takes place at a lower n than in the case of sedimentation (this is a consequence of the greater sensitivity of D_r to chain conformation). To make sure that we are considering strictly rod-like pieces of DNA, in the estimation of d we have considered only fragments with $n < 125$ base pairs. It is noteworthy that the electric dichroism decay data of Dieckmann et al.[18] are substantially higher (about 15%) than the birefringence decay data.[13,14,19,20]

In Table I we list the values of the hydrodynamic diameter d obtained by fitting each data set to the two theories. The commonly accepted value of $r = 3.4$ Å for the B form of Na$^+$-DNA has been used for D_t and D_r. We shall later discuss the choice of r.

As stated earlier in this paper, both d and r could be obtained from experimental data on D_t and D_r for the same samples by the use of the $f(p)$ function. We have analyzed in this way the data of Mandelkern et al.[19] with $n = 89$, 104, and 124 base pairs, and the results are listed in Table II. A similar method is to plot D_t vs D_r curves for different values of d, and inserting in the diagram the experimental values. This procedure is illustrated in Fig. 5.

DISCUSSION

It is clear from Tables I and II that for nearly all the experimental data examined the hydrodynamic diameter of DNA obtained using the Tirado–García de la Torre equations with $r = 3.4$ Å is closer to the expected value (26 Å) than that obtained from Broersma's equations. We think that this is a rather definitive test of the performance of the two theories. Table I also shows that in the case of Broersma's equations, the translational results are more discrepant than the rotational ones.

Our analysis of d could be affected by the choice of r and M_L. Some recent works have raised the possibility that in solution r could be somewhat shorter than 3.4 Å. In fact, by

TABLE I. Values of the hydrodynamic diameter d of DNA obtained by fitting several sets of experimental data to the two theories.

Property	Reference	n	$r =$	Broersma (Ref. 11) 3.4 Å	3.3 Å	Tirado–García de la Torre (Refs. 5 and 6) 3.4 Å	3.3 Å
s	Kovacic and Van Holde (Ref. 12)	50, 94, 117, 145, 160		18[a]	19[b]	25[a]	27[b]
D_t	Mandelkern et al. (Ref. 19)	89, 104, 124, 188		15	16	21	23
D_r^c	Elías and Eden (Ref. 14)	64, 80, 89, 104		24	27	28	32
		64, 80, 89, 104, 124		23	26	26	31
D_r	Hagerman (Ref. 16)	104, 124		23	26	26	30
D_r	Dieckmann et al. (Ref. 18)	43, 64, 69, 76, 84, 95, 118		17	20	20	24

[a] $M_L = 195$ dalton/Å.
[b] $M_L = 200$ dalton/Å.
[c] The experimental D_r data of Elías and Eden (Ref. 13) and Mandelkern et al. (Ref. 19) lead to values of d which differ by less than 1 Å.

TABLE II. Analysis of the experimental data of Mandelkern *et al.* (Ref. 19) for DNA.

n	Experimental (20, w) $D_t \times 10^7$, cm^2 s^{-1}	$D_r \times 10^{-5}$, s^{-1}	$f(p)$	Broersma p	L, Å	d, Å	r, Å	Tirado–García de la Torre p	L, Å	d, Å	r, Å
89	4.27	2.36	2.52	33	355	11	4.0	21	340	16	3.8
104	3.88	1.73	2.54	34	397	12	3.8	22.5	382	17	3.7
124	3.41	1.24	2.49	31	438	14	3.5	20	419	21	3.4

comparison of experimental data on D_r with theoretical predictions, Elías and Eden[14] and Mandelkern *et al.*[19] have obtained $r = 3.3$ Å. In regard to M_L, the 195 dalton/Å figure has been confirmed experimentally.[26] Anyhow, a 3% decrease in r would bring about an identical increase in M_L, so that a reasonable value in such case would be $M_L = 200$ Å. This value is in turn closer to those obtained from scattering measurements.[27,28]

We have included in Table I the results for $r = 3.3$ Å. For the rotational data of Eden and co-workers[13,14,19] and Hagerman[16] the Broersma equations give a better value of d, but for the data of Dieckman *et al.*[18] the Tirado–García de la Torre equations still seem best. In regard to the results from the translational properties (s, D_t) the comparison again favors clearly the latter theory.

We can see from Table II and Fig. 5 that the values of the molecular dimensions, r and d, obtained by means of the

FIG. 5. Plot of D_r vs D_t for cylinders with the indicated values of d in Å. The points correspond to experimental data of Mandelkern *et al.* (Ref. 19) for $n = 89$, 104, and 124.

$f(p)$ function or D_r vs D_t plot, are physically unreasonable: $d = 16$–21 Å with $r = 3.8$–3.4 Å from the Tirado–García de la Torre equations and $d = 11$–14 Å with $r = 4.0$–3.5 Å from Broersma equations. (Anyhow, the values from the Tirado–García de la Torre theory are better again.) In a similar analysis, Mandelkern *et al.*[19] obtained $d = 22$–26 Å, which seems more reasonable. However, it should be pointed out that his analysis was not consistent since it mixed the Tirado–García de la Torre equations for D_t[5] with the *old* Broersma equations[1] for D_r. It is also important to note that these methods are quite sensitive to errors in the experimental data, because the errors of D_t and D_r are accumulated and magnified in d owing to the logarithmic dependence.

It is not the purpose of this paper to obtain the solution value of the rise per base pair, r. Indeed, we believe that refinements in r of the order of 0.1 Å can not be made by analyzing the presently available hydrodynamic measurements. It is evident from this work, however, that there are appreciable differences between the results that one obtains from different properties, techniques, and laboratories. Unfortunately, the methods for determining simultaneously r and d [the $f(p)$ function or D_r vs D_t plot] are strongly affected by errors or uncertainties in the experimental data.

Finally, we wish to discuss some problematic aspects of our analysis. One of them could be the use of a number of sets of experimental data which, as pointed out previously, show in some cases remarkable and systematic differences. It is not the purpose of this paper to pinpoint the origin of the discrepancies between experimental data from different laboratories. We think, however, that the simultaneous use of *all* the data published so far makes our results and conclusions more reliable and significant (from the statistical point of view) than those of other works in which a single set of data was analyzed.[14,19]

A second point is that we have chosen $d = 26$ Å as the reference value in the comparison of the predictions of the two theories. We should recognize that this value, although confirmed by a structural study of hydration,[23] has been mainly obtained from hydrodynamic studies. However, such studies were about DNA of high molecular weight treated as a worm-like coil.[3,25,26] Thus both the samples and the models considered in those studies are clearly different from the ones used in our analysis. Appreciably smaller values of d are very unlikely, and if we took higher values (say $d = 30$ Å), our conclusion about the performance of the two theories would remain unchanged since, as seen in Tables I and II, Broersma's theory underestimates systematically the cylinder diameter.

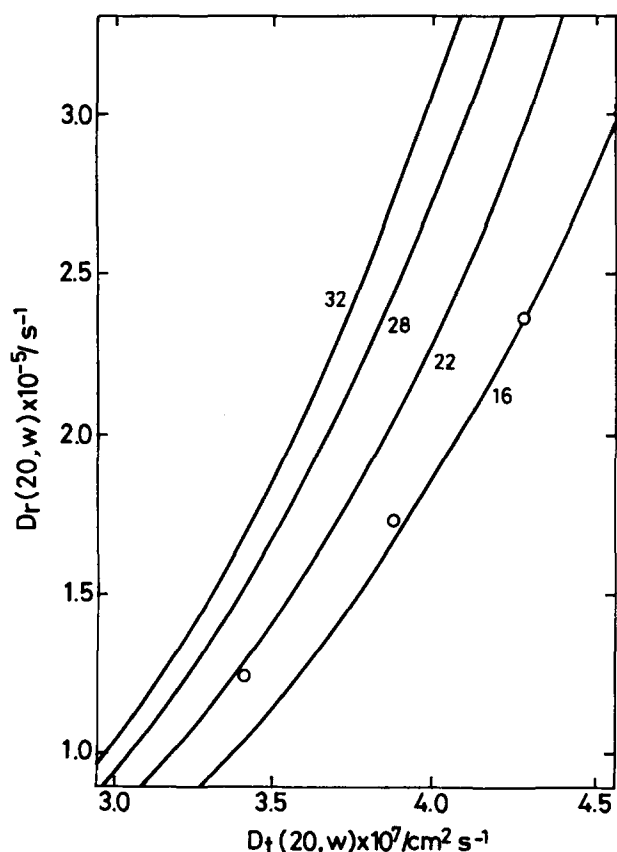Another problematic aspect could arise from some re-

sidual flexibility of the largest DNA fragments considered in our study. Hagerman[16] has combined results for the rotational coefficient of weakly bending rods with the *old* Broersma equation[1] to present in Fig. 13 of Ref. 16 the apparent value of r, r_{app}, that would be obtained for DNA pieces of varying length if they were considered as rigid rods. The deviation of r_{app} from 3.4 Å for DNA fragments with $L / P < 1$ (the persistence length, P, is 640 Å for DNA[16]) is in his opinion significant enough to conclude that these fragments do not behave strictly as rigid rods from the point of view of rotational diffusion.

We think, however that Hagerman's analysis of residual flexibility is not quantitatively correct. With $d \cong 26$ Å, the L/P region in Fig. 13 of Ref. 16 corresponds to $p < 25$, and it is now well known that the *old* Broersma equation is wrong for such sort cylinders. In addition, the results used for weakly bending rods are only valid for $p > 20$ (page 1491 in Ref. 20), or for $L/P > 0.8$. The longest DNA fragment considered in our work has $n = 125$, with $L/P = 0.66$. Disregarding the latter fact and assuming that the only source of error is the use of the old Broersma equation, we have redone Hagerman's calculations for r_{app} employing the newer equations. For $L/P = 0.35$–0.70, which approximately corresponds to $n = 64$–125, the use of the new Broersma equation gives $r_{app} = 3.41$–3.25, and our equation yields $r_{app} = 3.52$–3.35. These values are appreciably closer to 3.4 Å than those in Hagerman's paper.

Without further comments, we recall that for $n < 125$ all the sets of experimental D_r data follow well, within experimental error, the theoretical variation with length for rigid, straight rods. Thus, we beleive that DNA flexibility does not influence the results and conclusions of our work.

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