Relative Permittivity of Dipolar Model Fluids from Molecular Simulation and Co-Oriented Fluid Functional Equation for Electrostatic Interactions

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Relative permittivity, dielectric constant, molecular simulation, orientation, perturbation theory

ABSTRACT

The relative permittivity of dipolar fluids is important in many industrial and scientific applications, e.g. whenever electrolytes or electromagnetic fields are present. For non-polarizable model molecules, it is directly linked to the mutual molecular orientation and thereby usually not accessible by equations of state. However, the recently developed Co-Oriented Fluid Functional Equation for Electrostatic interactions (COFFEE) allows for calculating the orientation

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distribution function of simple polar molecules and thereby establishes a connection between the thermodynamic behavior and the relative permittivity. In this article, we develop an expression to calculate the relative permittivity from the orientation distribution known from COFFEE. Furthermore, we calculate the relative permittivity of simple polar fluids using molecular simulations. We study the original Stockmayer fluid and the shifted Stockmayer fluid, in which the dipole is shifted away from the Lennard-Jones center along the dipole axis. For both fluids, different dipole strengths are investigated. The results from the theoretical expression from COFFEE are compared to the simulation data. Thereby, a possible link between polar equations of state and electric fields or electrolytes is developed.

1. Introduction

The relative permittivity of solvents is important in many applications where either electromagnetic fields or electrolytes are present. In electrolyte solutions, by virtue of the underlying Coulomb potential, all expressions for thermodynamic properties depend on the relative permittivity, which is a strong function of the polar properties of the solvent and its thermodynamic state. However, engineering type equations of state (EOS) for electrolytes (e.g. ePC-SAFT¹), which are used for the description and prediction of thermodynamic properties, typically contain the relative permittivity of the pure solvent only as a constant or as an expression parameterized to experimental data, while different mixing rules are used for mixtures^{2–10}. Some exceptions are based on the non-primitive mean spherical approximation due Blum and Wei¹¹. This overview of literature sources is not comprehensive by any means, but is meant to highlight the necessity to link the equation of state describing e.g. dipolar contributions to the free energy and the relative permittivity. A comprehensive review of the different possible expressions is given by Loehe and Donohue¹², as well as in the introduction of Zhao et al.¹³.

There is an abundant body of literature on electrolyte EOS, most of which use the pure component relative permittivity. For relatively recent reviews on different approaches to the thermodynamics of electrolyte solutions the interested reader is referred to Kontogeorgis and Folas⁸, Kontogeorgis¹⁰, and Tan et al.⁹. A notable exception is the EOS developed in McCabe's group. In a series of articles^{14–18} from that group, an equation of state is combined with results from integral equation theory to couple dipole interactions and electric permittivity. The integral equation theory results are based on an empirical modification of the dipolar mean spherical approximation (dMSA) by Wertheim¹⁹. However, the relative permittivity is not directly linked to the EOS.

The main difficulty in modeling the relative permittivity of pure compounds is that the short ranged angular correlation of dipoles needs to be known. If the short-range angular correlation between dipoles is known, relative permittivity can be determined using methods developed by Onsager²⁰, Kirkwood²¹ and Fröhlich²². Integral equation theory is one route to this, which, however, is not directly linked to the EOS. Typical EOS are perturbation theories around the reference structure and, as such, contain no information on the orientational structure of the dipolar fluid. It is possible to derive a separate perturbation for the pair distribution function or to use integral equation theory beyond the dMSA (e.g. cluster expansions). Stell et al.²³ give a comprehensive account of such theories in their seminal paper. Again, these approaches would not be linked to the EOS, but rather be independent of it. Therefore, they are not discussed further here. The only EOS which directly accounts for the orientation distribution function (ODF) and thereby for the angular correlation of dipoles is the Co-Oriented Fluid Functional Equation for Electrostatic interactions (COFFEE) by Langenbach²⁴.

Static relative permittivity can also be calculated using standard computational chemistry techniques such as molecular dynamics and Monte Carlo simulations. It is usually obtained by sampling the formula derived by Kirkwood²¹. There is a large body of literature work on molecular simulations of permittivities. Due to their importance for the present work, we limit the present discussion to Stockmayer and Stockmayer-like fluids. Pioneering works on the relative permittivity of Stockmayer fluids were carried out by e.g. Neumann and co-workers^{25–28} in the framework of developing long-range corrections for the electrostatic interactions such as the reaction field method. Later, simulation data for the permittivity of Stockmayer fluids were employed to assess perturbation theories^{29–31} and to identify the cross-over between isotropic and ferroelectric behavior^{32–35}. However, this transition occurs at very high dipole strengths and is thus irrelevant for the present work, i.e. all fluids investigated here are isotropic.

In this article, the Kirkwood²¹ equation is used together with COFFEE²⁴ to yield a model for the relative permittivity which is directly linked with an EOS. To test our theory, we perform molecular simulations. We calculate the relative permittivity by sampling Kirkwood’s formula²¹ for the Stockmayer fluid and the closely related shifted Stockmayer fluid. In the shifted Stockmayer fluid, the dipole is not centered on the Lennard-Jones sphere, but shifted away from it along its own axis by a distance d (cf. Figure 1). We perform a systematic study of the relative permittivity on the saturated liquid and vapor lines of the vapor-liquid equilibrium for several temperatures, two dipole moments, and the shift distances $d = 0$ and $d = 0.1 \sigma$. The saturated densities for these models at several state points are taken from previous work.²⁴ For the unshifted Stockmayer fluid with a reduced dipole moment of $\mu^* = 1.0$, we compare our simulation results to literature data²⁹. To the best of our knowledge, the relative permittivity of

the shifted Stockmayer fluid has never been reported before, while the amount of data for the Stockmayer fluid on the saturation line is very limited.

While this work is limited to model fluids, application to real fluids is straightforward. Work in this direction is currently undertaken and generalizations of COFFEE to more complex fluids are planned.

Figure 1. Depiction of the two different model fluids investigated in this article. The blue (online) large spheres depict the reference potential centered at the black small spheres. Red (online) arrows show dipole moments with center in the small red (online) spheres. From left to right the depictions are reference potentials with a) central dipole, b) dipole shifted by d . Reprinted from Chemical Engineering Science, 174, K. Langenbach, Co-Oriented Fluid Functional Equation for Electrostatic Interactions (COFFEE), 40-55, Copyright (2017), with permission from Elsevier.

2. Molecular Simulation

We study fluids interacting with the Lennard-Jones potential and a superimposed point dipole, so that the total energy in a system of N particles is given by

$$U = \sum_{i=1}^N \sum_{j=1}^N \left(4\epsilon_{LJ} \left[\left(\frac{\sigma}{r_{ij}} \right)^{12} - \left(\frac{\sigma}{r_{ij}} \right)^6 \right] - \frac{1}{4\pi\epsilon_0} \frac{\mu^2}{r_{D_i D_j}^3} (2\cos\varphi'_i \cos\varphi'_j - \sin\varphi'_i \sin\varphi'_j \cos\gamma'_{ij}) \right) \quad (1)$$

where σ and ε_{LJ} are the Lennard-Jones size and energy parameters, respectively, r_{ij} is the distance between particles i and j , $r_{D_i D_j}$ is the distance between the dipoles on particles i and j , ε_0 is the vacuum permittivity, μ is the magnitude of the dipole moment, and the φ'_i and γ'_{ij} are angles describing the mutual orientation of the dipoles as depicted in Figure 2 a). The prime indicates that the angles are related to the dipoles and their mutual connecting line. Figure 2 b) shows the angles of the molecule with respect to the Lennard-Jones center as defined in this article.

Figure 2. a) Angles used in the dipole-dipole potential. b) Distinguishable angles and vectors in coordinate system for ODF calculation, reprinted from Chemical Engineering Science, 174, K. Langenbach, Co-Oriented Fluid Functional Equation for Electrostatic Interactions (COFFEE), 40-55, Copyright (2017), with permission from Elsevier.

In molecular simulations, the relative permittivity can be sampled according to Kirkwood's formula²¹

$$\varepsilon_r = 1 + \frac{4\pi}{3kTV} \left(\langle \mathbf{M}^2 \rangle - \langle \mathbf{M} \rangle^2 \right) \quad (2)$$

where

$$\mathbf{M} = \sum_{i=1}^N \boldsymbol{\mu}_i \quad (3)$$

is the total dipole moment of the simulation box and $\boldsymbol{\mu}_i$ is the dipole moment vector of molecule i , k is Boltzmann's constant, T is the temperature and V is the volume. For isotropic fluids as the ones studied here, the term $\langle \mathbf{M} \rangle^2$ vanishes.

All simulations of this work were carried out with an extended version of the molecular simulation program *ms2*³⁶. Kirkwood's formula was sampled in Molecular Dynamics (MD) simulations in the NVT ensemble. The total number of particles was 1024 throughout, and the saturated states of selected dipolar fluids presented by Langenbach²⁴ were studied, cf. also Table 2 of the appendix. The MD simulations were conducted with the reduced time step $\Delta t^* = \Delta t / \sigma \sqrt{\varepsilon_{LJ} / m} = 0.0025$ with m being the Lennard-Jones mass, and the cutoff radius was equal to half the box length. The equilibration and production runs took 50 000 and 10 000 000 time steps, respectively. The electrostatic long-range corrections were treated with the reaction field method, assuming an infinite relative permittivity of the continuum outside the reaction field sphere (tin foil boundary conditions). Statistical uncertainties were estimated with the block average method by Flyvbjerg and Petersen³⁷.

The parameters of the investigated models are listed in Table 1.

Table 1. Reduced dipole moment and dipole shift of the investigated models.

μ^*	1.0	1.0	1.5	1.5	2.0	2.0
d^*	0	0.1	0	0.1	0	0.1

3. Theory

For non-polarizable dipolar molecules, the Kirkwood-equation²¹ is a good starting point for a theory of the dielectric

$$\frac{(\varepsilon_r - 1)(2\varepsilon_r + 1)}{\varepsilon_r} = 4\pi\rho^* \varepsilon_{LJ}^* \mu^{*2} (1 + z \langle \cos(\Theta) \rangle) \quad (4)$$

where $\varepsilon_r = \varepsilon/\varepsilon_0^{-1}$ is the electric permittivity relative to the electric permittivity of vacuum ε_0 , $\rho^* = \rho\sigma^3$ is the number density reduced with the Lennard-Jones diameter σ , $\mu^{*2} = \mu^2 (4\pi\varepsilon_0\varepsilon_{LJ}\sigma^3)^{-1}$ is the dipole strength divided by the permittivity of vacuum ε_0 times 4π and reduced with the Lennard-Jones diameter and interaction energy ε_{LJ} , $\varepsilon_{LJ}^* = \varepsilon_{LJ} (kT)^{-1}$ is the reduced dispersion interaction energy with k being Boltzmann's constant and T being the temperature, z is the number of nearest neighbors, i.e. the average coordination number, and $\langle \cos(\Theta) \rangle$ is the ensemble average angle between two dipoles of these neighbors. Temperatures are reported in the reduced form $T^* = kT\varepsilon_{LJ}^{-1}$.

Generally, both the number of nearest neighbors and the average angle are needed for predicting the relative permittivity. While these properties can be obtained from molecular simulation, equations of state based on perturbation theory usually assume the reference fluid's structure

and, thereby, contain no information on the angular correlation of dipoles and only limited information on the number of nearest neighbors. In COFFEE it is assumed that the radial packing is unaltered by orientation. Hence, we used the expression for the radial distribution function by Bamdad et al.³⁸ to calculate the number of nearest neighbors as a function of temperature and density. The temperature dependence was found to be negligible and therefore we arrive at the simple expression

$$z = \rho^* (a + b\rho^*) \quad (5)$$

where the values for the constants are $a = 0.0675$ and $b = 14.164$.

Using COFFEE²⁴, in contrast to other equations of state, information on the orientation distribution function (ODF) $O(\omega_1, \omega_2)$ describing the probability of finding two molecules in the general orientations ω_1 and ω_2 is accessible for the first coordination shell. COFFEE is a perturbation theory for molecules with the intermolecular potential²⁴

$$u = u_{ref} + \lambda o(\mathbf{x}_1, \mathbf{x}_2) \quad (6)$$

with the spherically symmetric reference potential u_{ref} and the general dependence $o(\mathbf{x}_1, \mathbf{x}_2)$ of the perturbing potential (in this case dipolar) on the generalized coordinates $\mathbf{x}_i = \{\mathbf{r}_i, \omega_i\}$ where $\mathbf{r}_i$ is the position of molecule i . The free energy of the reference fluid is taken to be given by the SAFT-VR Mie EOS³⁹ with 12-6 potential. The perturbation parameter λ above can be identified with the squared dipole moment $\mu'^2 = \mu^2 (4\pi\epsilon_0)^{-1}$ in the full perturbation limit.

Using COFFEE, it is assumed that only in the first coordination shell, the molecules may orient preferentially²⁴. Therefore, the residual free energy due to dipolar interactions is split into the near-field (NF) and the far-field (FF) part. In the FF, the usual expansion of the pair distribution function around $\lambda = 0$ is used, while in the NF, the pair distribution is expanded around the fully perturbed state. For simplification, it is assumed that the radial distribution function is that of the reference fluid g_{ref} and can be factored out. In addition, it is assumed that the ODF in the first coordination shell does not depend on distance. This leads to the expression

$$g(r_{12}, \omega_1, \omega_2, \lambda = \mu^2) = g_{ref}(r_{12}) O(\omega_1, \omega_2) \quad (7)$$

for the first term in the series expansion, which is truncated at this point. Furthermore it is assumed that the contact value of $g_{ref}(\sigma)$ can be used as a simple expression for the pair distribution in the first coordination shell. In the free energy expression, the contact value for the hard sphere distribution is used as a further simplification. For the contact value, the expression of Boublík⁴⁰

$$g_{hs}(\sigma, \eta) = \frac{1}{1-\eta} + \frac{3\eta}{(1-\eta)^2} + \frac{2\eta}{(1-\eta)^3} \quad (8)$$

for the hard sphere with the packing fraction $\eta = \frac{\pi}{6} \rho^* d_{LJ}^3$ is used, where d_{LJ} is the temperature dependent Lennard-Jones diameter according to the SAFT-VR Mie EOS³⁹. The NF part of the free energy is a functional of the ODF

$$\begin{aligned}
\frac{F_{\mu\mu}^{NF}}{NkT} = & \frac{19\pi}{12} \rho^* g_{hs}(\sigma, \eta) \int d\mathbf{\omega}_1 \int d\mathbf{\omega}_2 O(\mathbf{\omega}_1, \mathbf{\omega}_2) \ln(\Omega O(\mathbf{\omega}_1, \mathbf{\omega}_2)) \\
& + 2\pi\rho^* \varepsilon_{LJ}^* \mu^{*2} g_{hs}(\sigma, \eta) I_{\mu\mu}^{NF} \int_1^{3/2} dr_{12}^* \int d\mathbf{\omega}_1 \int d\mathbf{\omega}_2 O(\mathbf{\omega}_1, \mathbf{\omega}_2) o(r_{D_1 D_2}^*, \mathbf{\omega}_1, \mathbf{\omega}_2)
\end{aligned} \tag{9}$$

where r_{12}^* is the distance between two neighboring molecules reduced with σ . The ODF follows from minimization of the NF free energy under the constraint that the integral of O over all orientations is 1. Dropping the dependence of o on the distance and orientations for ease of notation, the ODF is then given by

$$O(\xi_1, \gamma_{12}, \xi_2) = \frac{\exp\left(-\frac{24}{19} \mu^{*2} \varepsilon_{LJ}^* I_{\mu\mu}^{NF} \int_{r_{12}=0}^{3/2} dr_{12}^* o\right)}{\int d\xi_1 \int d\gamma_{12} \int d\xi_2 \exp\left(-\frac{24}{19} \mu^{*2} \varepsilon_{LJ}^* I_{\mu\mu}^{NF} \int_{r_{12}=0}^{3/2} dr_{12}^* o\right)} \tag{10}$$

Here, the arbitrary orientation variables $\mathbf{\omega}$ are replaced by the angles of Figure 2 b) or their cosines $\xi_i = \cos\varphi_i$, respectively. The cosine is chosen because using the actual angles would cause an artifact in the orientation distribution due to the different size of the angular volume elements for different angles. An example of the ODF calculated with COFFEE of the Stockmayer fluid with dipole moment $\mu^* = 1.5$ for the temperature $T^* = 2$ and the density $\rho^* = 0.4$ is shown in Figure 3.

Figure 3. Isosurfaces of the ODF at $T^* = 2$ and $\rho^* = 0.4$ for a Stockmayer fluid with dipole moment $\mu^* = 1.5$. The surfaces are isosurfaces with values 1.4 (red), 1.0 (green), and 0.6 (blue) times the probability of a random distribution (color shown online). Reprinted from Chemical Engineering Science, 174, K. Langenbach, Co-Oriented Fluid Functional Equation for Electrostatic Interactions (COFFEE), 40-55, Copyright (2017), with permission from Elsevier.

The cosine of the angle between two dipoles expressed in the variables of COFFEE is

$$\cos \Theta_{12} = \xi'_1 \xi'_2 - \sqrt{1 - \xi_1'^2} \sqrt{1 - \xi_2'^2} \cos \gamma'_{12} \quad (11)$$

where the prime indicates that the respective angles are those of the dipoles rather than the molecules (cf. Figure 2). These angles will always depend on the molecular angles. The average cosine of the angle is

$$\langle \cos \Theta_{12} \rangle = \iiint d\xi_1 d\xi_2 d\gamma_{12} O(\xi_1, \gamma_{12}, \xi_2) \cos \Theta_{12} \quad (12)$$

For the Stockmayer fluid,

$$\xi'_i = \xi_i \quad (13)$$

and

$$\gamma'_{12} = \gamma_{12} \quad (14)$$

For the shifted Stockmayer fluid, the corresponding expressions are given by Langenbach²⁴. For the shifted Stockmayer fluid, the angle between dipoles depends on the intermolecular distance. The average value over the first coordination shell is used here. In reduced units, the result for the relative permittivity is

$$\epsilon_r = \frac{y+1}{4} + \sqrt{\left(\frac{y+1}{4}\right)^2 + \frac{1}{2}} \quad (15)$$

with the abbreviation

$$y = 4\pi\mu^{*2}\epsilon_{LJ}^*\rho^*(1 + z \langle \cos \Theta_{12} \rangle) \quad (16)$$

This expression does not contain any adjustable parameters or unknowns aside from the relative permittivity and is thus fully predictive. If the parameters for a fluid model are known for COFFEE, the relative permittivity follows directly.

4. Results & Discussion

In Table 2 of the appendix, the simulation results for the relative permittivity in the saturated liquid and vapor state are given together with their statistical uncertainties for the models and state points investigated here. These results are shown together with the theoretical prediction from COFFEE in Figure 4 for three dipole moments and two dipole shifts. The overall agreement between prediction and computer experiment is good. Also shown are literature values for the Stockmayer fluid with $\mu^* = 1$ from Boda et al.²⁹, which are in acceptable agreement with our

own simulation data. The occurring differences are likely due to small differences in saturated density. The best agreement between theory and computer experiments is found for intermediate dipole moments. For small dipole moments, the relative permittivity is somewhat overpredicted. For large dipole moments, it is underpredicted at low temperatures while still being in good agreement to the computer experiments at higher temperatures. The simulation results show that shifting the dipole results in an increased relative permittivity compared to the corresponding unshifted fluid. COFFEE predicts this behavior correctly for high temperatures, whereas at low temperatures only a very small impact of the shift is predicted. In Figure 5 the vapor phase region is enlarged. Overall, the agreement is better than for the liquid phase. The prediction of the impact of a dipole shift on the relative permittivity from COFFEE is also in good agreement with simulation data.

Figure 4. Relative permittivity versus temperature of the Stockmayer (full lines and filled symbols) and the shifted Stockmayer fluid (broken lines and open symbols) from molecular simulations (symbols) and the proposed method (lines). The statistical uncertainties of the simulation results are within the symbol size. From bottom to top the dipole moments are

$\mu^* = 1.0$ (black online), $\mu^* = 1.5$ (red online) and $\mu^* = 2.0$ (blue online). The crosses depict data from Boda et al.²⁹ for $\mu^* = 1.0$.

Figure 5. Same as Figure 4, but enlarged on the vapor side.

The differences between prediction and computer experiment mainly for the liquid phase might have several reasons. Deviations between predictions for the influence of shifting the dipole from the center can possibly be explained as follows: The phase behavior itself is predicted and is not in perfect agreement with simulation data, even though the trends are correctly predicted by COFFEE²⁴. The low temperature behavior might be due to the way COFFEE is constructed. The near field term of COFFEE is a series expansion in the unknown orientational structure and the series is expanded only to first order²⁴. This low order may be one reason for the deviations at low temperatures for high dipole moments, since the convergence radius of the series is anti-proportional to the square dipole moment μ^{*2} divided by kT . The second reason might be that the orientation frustration due to high densities⁴¹ is only taken into account by a correlated

factor²⁴, rather than being included in the functional equation through a repulsive term. A third reason could be that at low temperatures and high dipole moments, the second coordination shell shows some angular correlation of the dipoles³⁵ for Stockmayer fluids, which is ignored by the current level of the theory. All three of these shortcomings of the theory could be lifted in principle, however at the expense of numerical effort. A higher order in the series expansion, a functional treatment of the orientation frustration, and a treatment of the second coordination shell would lead to a nonlinear Euler-Lagrange equation and thereby to not analytically solvable transcendental expressions for the ODF.

5. Conclusions & Outlook

In this paper, new simulation data for the relative permittivity at saturated liquid and vapor density over the entire temperature range of the vapor-liquid equilibrium of the Stockmayer fluid and the related Stockmayer fluid with the dipole shifted from the Lennard-Jones center along its axis are presented. The data are given for three dipole moments and two shifts. A theory for the prediction of the pure component relative permittivity based on the Co-Oriented Fluid Functional Equation for Electrostatic (COFFEE) interactions is presented. Predictions from the theory and simulation data are mostly in good agreement, except for high dipole moments at low temperatures. The reason for the worse agreement at these points can be the low order of the series expansion in the orientation distribution function underlying the equation of state or the account of the orientation frustration by a correlated factor. For future work, it would be interesting to lift both of these simplifications. Nonetheless, the developed theory offers the possibility of linking the dipolar contribution of the equation of state to expressions for electrolytes and thereby provides a possible way to develop a new electrolyte equation of state. This shows the possibility to remove one of the shortcomings of common perturbation theories,

i.e. that their contributions are not connected. Furthermore, using the developed theory, the relative permittivity can be included when parameterizing models for COFFEE and thus provides a way to distinguish between the polar and dispersion contributions.

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Appendix

Simulation results for the relative permittivity of the studied model fluids are given in Table 2.

Table 2. MD results for ϵ_r of Stockmayer and shifted Stockmayer fluids at ρ^* in the saturated liquid (L) and vapor (V) state for several μ^* and T^* . Uncertainties in the last digit are given in brackets.

μ^*	T^*	ρ_L^*	$\epsilon_{r,L}$	ρ_V^*	$\epsilon_{r,V}$	T^*	ρ_L^*	$\epsilon_{r,L}$	ρ_V^*	$\epsilon_{r,V}$
$d^* = 0$						$d^* = 0.1$				
1.0	0.86	0.820	10.20(7)	0.0047	1.0232(1)	0.95	0.783	8.66(6)	0.0099	1.0438(6)
	0.92	0.792	8.64(5)	0.0081	1.0369(2)	1.00	0.760	7.80(5)	0.0143	1.0603(7)
	0.98	0.766	7.53(4)	0.0131	1.0569(3)	1.05	0.736	6.93(4)	0.0205	1.084(1)
	1.04	0.737	6.62(4)	0.0200	1.0832(5)	1.10	0.708	6.22(4)	0.0277	1.109(1)
	1.10	0.706	5.83(3)	0.0297	1.1165(7)	1.15	0.682	5.64(3)	0.0390	1.148(1)
	1.16	0.670	5.13(3)	0.0413	1.1554(9)	1.20	0.652	5.05(3)	0.0513	1.188(2)
	1.22	0.635	4.56(2)	0.0602	1.217(1)	1.25	0.620	4.52(3)	0.0697	1.249(2)
	1.28	0.589	3.97(2)	0.0836	1.296(2)	1.30	0.580	4.05(2)	0.0934	1.332(3)
1.5	1.34	0.537	3.45(1)	0.1234	1.429(2)	1.35	0.532	3.59(2)	0.1249	1.436(3)
	1.06	0.815	29.2(2)	0.0048	1.0434(2)	1.06	0.820	32.7(3)	0.0044	1.0410(5)
	1.12	0.790	23.6(2)	0.0078	1.0674(4)	1.12	0.798	27.0(3)	0.0073	1.0621(7)
	1.18	0.766	20.3(1)	0.0120	1.1003(5)	1.18	0.774	22.5(2)	0.0111	1.0922(8)
	1.24	0.739	19.1(1)	0.0167	1.1321(7)	1.24	0.747	19.0(2)	0.0156	1.125(1)
	1.30	0.711	14.49(9)	0.0245	1.187(1)	1.30	0.720	16.5(1)	0.0223	1.174(1)
	1.36	0.681	12.65(8)	0.0343	1.253(1)	1.36	0.690	14.0(1)	0.0325	1.247(2)

	1.42	0.649	10.99(6)	0.0476	1.348(2)	1.42	0.658	12.04(9)	0.0438	1.326(2)
	1.48	0.609	9.29(5)	0.0642	1.461(2)	1.48	0.624	10.50(7)	0.0614	1.449(3)
	1.54	0.565	7.96(4)	0.0880	1.626(3)	1.54	0.579	8.81(6)	0.0844	1.607(4)
	1.28	0.836	77.9(8)	0.0026	1.0342(2)	1.38	0.812	58.7(7)	0.0040	1.0509(5)
	1.36	0.809	57.1(5)	0.0044	1.0558(3)	1.46	0.784	46.0(5)	0.0069	1.0861(7)
	1.44	0.781	44.9(4)	0.0064	1.0768(4)	1.54	0.754	36.9(4)	0.0109	1.130(1)
	1.52	0.750	35.5(3)	0.0103	1.1183(6)	1.62	0.724	30.5(3)	0.0150	1.172(1)
2.0	1.60	0.720	29.0(2)	0.0165	1.1849(9)	1.70	0.692	25.5(2)	0.0259	1.287(2)
	1.68	0.684	23.6(2)	0.0248	1.269(1)	1.78	0.653	20.8(2)	0.0351	1.384(3)
	1.76	0.648	19.3(1)	0.0375	1.400(2)	1.86	0.613	17.3(1)	0.0540	1.597(4)
	1.84	0.605	15.8(1)	0.0564	1.610(3)	1.94	0.561	14.1(1)	0.0753	1.838(5)
	1.92	0.554	12.91(8)	0.0815	1.878(4)	2.02	0.495	11.01(8)	0.1217	2.417(8)

TOC Graphic

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