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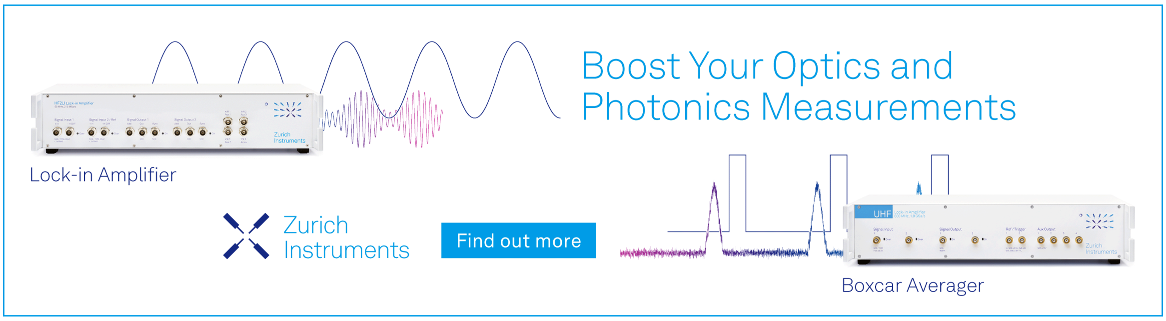

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## I. INTRODUCTION

In the course of a study aimed at obtaining analytic frequency-dependent hypermagnetizabilities $\eta_{\alpha\beta,\gamma\delta}(-\omega;\omega,0,0)$ and Cotton–Mouton constants ${}_mC(\omega,T)$ of molecules using London atomic orbitals (LAO's),[1] we came to discover the occurrence of an unfortunate mistake in the determination of the sign of the paramagnetic contribution $\Delta\eta^{\mathrm{para}}(\omega)$ to the anisotropy of the hypermagnetizability $\Delta\eta(\omega)$ published in the studies of Refs. 2–4. In the latter, moreover, the sign of the hyperpolarizability contribution to Buckingham birefringence $b(\omega)$ was wrong. Here we correct these mistakes and elaborate on the consequences for the discussions and comparisons made in the original papers.

## II. $C_6H_6$

Table I replaces the original Table IV of Ref. 2. Since the paramagnetic $\Delta\eta^{\mathrm{para}}$ and diamagnetic $\Delta\eta^{\mathrm{dia}}$ contributions are roughly of the same magnitudes and of opposite signs, the effect of the correction of the sign of $\Delta\eta^{\mathrm{para}}$ is rather dramatic, with $\Delta\eta(\omega)$ greatly reduced with respect to the erroneous estimates of Ref. 2. The effect on the Cotton-Mouton constant ${}_mC$ and on the anisotropy of the refractive index $\Delta n_u$ is marginal. With respect to the discussion of Cotton-Mouton effect (CME) in Sec. V D in Ref. 2, the need for significantly large basis sets, such as enhancement with double augmentation of the correlation consistent basis sets, is strengthened. Likewise, the originally reported agreement with the Hartree–Fock estimate in Ref. 5, where a lower quality basis was used, is no longer found. Indeed now the Hartree–Fock estimate for $\Delta\eta$ given by Augspurger and Dykstra in Ref. 5 is about five times larger than the value we compute. The effects induced by the change of sign of $\Delta\eta^{\mathrm{para}}$ on the temperature dependence of the Cotton–Mouton constant are such that they do not affect the appearance of Fig. 2 in Ref. 2. In the temperature range of that figure, the contribution of $\Delta\eta(\omega)$ is at most 0.4% of the total effect, instead of 2.2% as stated in Ref. 2. We add that the wide range of variations noted in the original paper for $\Delta\eta(\omega)$ with the choice of the functional appears to be magnified.

## III. $C_6F_6$

Tables II and III replace Tables IV and V, respectively, in Ref. 3. The whole paragraph at the end of the left column on p. 234314–7 in Ref. 3 should be replaced by the following:

"The value of $\Delta\eta(\lambda=632.8\ \mathrm{nm})$ is negative and very close to the center of the distribution of the experimental data $(\Delta\eta(\omega)=-100\pm880\ \mathrm{a.u.}$, as measured at $\lambda=441.6\ \mathrm{nm})$. The effect of electron correlation depends strongly on the functional, leading to a reduction of as much as ≈50% for LB94/LDA, and to increases of up to 25% for the other functionals. With the aug-cc-pVTZ basis set, the increase is ≈8% for B3LYP."

No changes are needed for the rest of the Discussion in Ref. 3, and Fig. 3 thereof also remains unchanged.

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TABLE I. $C_6H_6$. CME at $\lambda = 632.8$ nm. Anisotropy at $T = 273.15$ K. All data in a.u., with ${}_mC(\lambda, T)$ in cm$^3$ G$^{-2}$ mol$^{-1}$ $(4\pi\epsilon_0)$. $\Delta n_u(\lambda, T)$ is defined as $\Delta n$ observed for an induction field $B$ of 1 T and a pressure $P$ of 1 atm.

| Wf/Kernel | Basis | $\alpha_{\text{ani}}(\omega)$ | $\xi_{\text{ani}}$ | $\Delta\eta^{\text{dia}}$ | $\Delta\eta^{\text{para}}$ | $\Delta\eta$ | ${}_mC \times 10^{16}$ (cgs) | $\Delta n_u \times 10^{11}$ |
|---|---|---|---|---|---|---|---|---|
| HF-SCF | aug-cc-pVDZ | −35.7550 | −13.6111 | 483.9 | −323.3 | 160.5 | 2.83 | 1.70 |
| HF-SCF | aug-cc-pVTZ | −36.1277 | −14.3324 | 519.1 | −359.6 | 159.5 | 3.01 | 1.81 |
| HF-SCF | d-aug-cc-pVDZ | −36.2245 | −14.1156 | 539.2 | −389.1 | 150.1 | 2.97 | 1.79 |
| LDA | aug-cc-pVDZ | −40.5267 | −13.4103 | 619.5 | −518.6 | 100.9 | 3.15 | 1.90 |
| LB94/LDA | | −44.6525 | −14.7917 | 519.0 | −535.7 | −16.7 | 3.83 | 2.30 |
| BLYP | | −40.7098 | −13.1723 | 655.3 | −500.8 | 154.5 | 3.11 | 1.87 |
| B3LYP/LDA | | −33.4137 | −13.3613 | 491.1 | −317.6 | 173.5 | 2.59 | 1.56 |
| B3LYP/BLYP | | −33.8619 | −13.3613 | 462.6 | −305.8 | 156.8 | 2.63 | 1.58 |
| B3LYP | | −39.5071 | −13.3613 | 584.1 | −435.2 | 148.9 | 3.06 | 1.85 |
| PBE | | −40.5302 | −13.4096 | 620.4 | −519.3 | 101.1 | 3.15 | 1.90 |
| LDA | aug-cc-pVTZ | −40.4563 | −13.5377 | 622.9 | −516.4 | 106.5 | 3.18 | 1.91 |
| LB94/LDA | | −43.8415 | −14.0426 | 516.0 | −512.3 | 3.6 | 3.57 | 2.15 |
| BLYP | | −40.5590 | −13.1263 | 658.7 | −500.1 | 158.5 | 3.09 | 1.86 |
| B3LYP/LDA | | −33.2932 | −13.4845 | 458.1 | −294.0 | 164.1 | 2.61 | 1.57 |
| B3LYP/BLYP | | −33.3223 | −13.4845 | 456.3 | −293.4 | 162.8 | 2.61 | 1.57 |
| B3LYP | | −39.4865 | −13.4845 | 591.6 | −434.4 | 157.2 | 3.09 | 1.86 |
| PBE | | −40.4589 | −13.5368 | 623.8 | −517.2 | 106.6 | 3.18 | 1.91 |
| CCSD | aug-cc-pVDZ | −37.7312 | −12.5906 | 527.2 | −362.2 | 165.1 | 2.76 | 1.66 |
| | d-aug-cc-pVDZ | −38.2624 | −12.961 | 542.4 | −389.3 | 153.1 | 2.88 | 1.73 |
| Expt. | | $-37.79 \pm 1.15$ [b] | $-13.13 \pm 0.51$ [b] | | | $-2700 \pm 2000$ [d] | | $1.29 \pm 0.06$ [a]<br>$1.51 \pm 0.04$ [c]<br>$1.50 \pm 0.04$ [d] |

[a] $\lambda = 632.8$ nm, $T = 293.15$ K, Ref. 13.

[b] Reference 14.

[c] $\lambda = 632.8$ nm, $T = 293$ K, Ref. 15.

[d] $\lambda = 441.6$ nm, $T = 300.1$ K, measurements in the temperature range of 300.1– 455.5 K, Ref. 12.

## IV. $BF_3$

Table IV replaces Table IV of Ref. 4. As a first consequence of the changes, in particular due to the reversal of the sign of $b(\omega)$, the revised value of the traceless quadrupole moment of $BF_3$ originally measured in Ref. 6 is $\Theta^{\text{rev}} = +2.90 \pm 0.15$ (instead of the previous incorrect value of $+2.72 \pm 0.15$). The entry in the next to the last row in Table III of Ref. 4 must therefore be updated. Note then that, contrary to what we stated originally, the revision of the "apparent" quadrupole moment given in Ref. 6, made taking into account the nonvanishing $b(\omega)$ contribution, brings the Buckingham-birefringence-derived experimental value *closer* to our *ab initio* theoretical best estimate $(\Theta = +3.00 \pm 0.01)$. The inclusion of zero-point vibrational average further improves the comparison.

The temperature-independent contribution to the CME $\Delta\eta(\omega)$ is still significant, but yet on the average, when

TABLE II. Dynamic second electric-dipole hyperpolarizability and mixed electric-dipole hypersusceptibilities (see text for definitions). Atomic units, $\lambda = 632.8$ nm.

| Wave function | Basis | $\gamma_K(\times 10^3)$ | $b(\omega)$ | $\Delta\eta(\omega)$ | $\Lambda(\times 10^3)$ |
|---|---|---|---|---|---|
| HF-SCF | Sadlej | 3.24 | −322 | −151 | 4.57 |
| LDA | | 6.07 | −443 | −165 | 8.27 |
| LB94/LDA | | 4.19 | −367 | −121 | 5.59 |
| BLYP | | 6.61 | −440 | −183 | 8.99 |
| B3LYP | | 5.04 | −397 | −171 | 7.13 |
| HF-SCF | aug-cc-pVDZ | 2.87 | −293 | −62 | 2.52 |
| LDA | | 5.39 | −415 | −60 | 6.04 |
| LB94/LDA | | 3.82 | −344 | −33 | 3.31 |
| BLYP | | 5.76 | −411 | −78 | 6.93 |
| B3LYP | | 4.64 | −368 | −71 | 5.05 |
| HF-SCF | aug-cc-pVTZ | 3.25 | −312 | −98 | 4.71 |
| B3LYP | | 5.09 | −395 | −106 | 7.05 |
| Expt. | | $(11 \pm 11) \times 10^3$ [a] | $-2900 \pm 2000$ [b] | $-100 \pm 880$ [c] | |

[a] Reference 16.

[b] Reference 14.

[c] $\lambda = 441.6$ nm, $T = 304.1$ K, with measurements in the temperature range of 304.1–453.5 K, Ref. 12.

TABLE III. Linear birefringences of $C_6F_6$. $\lambda=632.8$ nm. Atomic units except where noted.

| | | Kerr effect[a] | | CME[b] | | Buckingham[c] | | Jones[d] | |
|---|---|---|---|---|---|---|---|---|---|
| Wave function | Basis | ${}_mK\times10^{26}$ | $\Delta n\times10^{11}$ | ${}_mC\times10^{16}$ | $\Delta n_u\times10^{12}$ | ${}_mQ(\omega,T)\times10^{-28}$ | $\Delta n\times10^{14}$ | ${}_mJ\times10^{-26}$ | $\Delta n\times10^{15}$ |
| HF-SCF | Sadlej | 1.91 | 7.66 | 0.44 | 2.46 | −11.23 | 11.31 | 11.52 | 3.54 |
| LDA | | 2.98 | 11.98 | 0.23 | 1.31 | −10.36 | 10.44 | 20.85 | 6.41 |
| LB94/LDA | | 2.89 | 11.62 | 0.53 | 2.88 | −18.84 | 18.97 | 14.10 | 4.33 |
| BLYP | | 3.02 | 12.14 | 0.51 | 2.84 | −12.17 | 12.26 | 22.67 | 6.97 |
| B3LYP | | 2.69 | 10.81 | 0.61 | 3.42 | −11.96 | 12.05 | 17.99 | 5.53 |
| HF-SCF | aug-cc-pVDZ | 1.95 | 7.85 | 0.23 | 1.29 | −11.48 | 11.57 | 6.36 | 1.96 |
| LDA | | 3.01 | 12.08 | 1.73 | 9.73 | −10.26 | 10.34 | 15.23 | 4.68 |
| LB94/LDA | | 2.87 | 11.51 | 1.99 | 11.17 | −18.49 | 18.62 | 8.35 | 2.56 |
| BLYP | | 3.05 | 12.26 | 1.55 | 8.72 | −12.13 | 12.21 | 17.49 | 5.37 |
| B3LYP | | 2.72 | 10.95 | 1.34 | 7.54 | −11.98 | 12.06 | 12.73 | 3.91 |
| HF-SCF | aug-cc-pVTZ | 1.93 | 7.76 | 1.91 | 10.72 | −11.60 | 11.68 | 11.88 | 3.65 |
| B3LYP | | 2.69 | 10.79 | 2.09 | 11.72 | −11.90 | 11.99 | 17.78 | 5.46 |
| Derived from Experiment | | 2.61 ± 0.17 [e] | | 1.46 ± 0.03 [f]<br>1.68 ± 0.03 [h] | 8.81 ± 0.18 [f]<br>10.11 ± 0.17 [h] | −11.2 ± 1.2 [g] | | | |

[a] ${}_mK$ given in SI units of $V^{-2}$ $m^5$ $mol^{-1}$. Pressure of 1 bar, $T=273.15$ K and electric field strength $E$ of $2.6\times10^6$ V $m^{-1}$;

[b] ${}_mC$ given in cgs units; $cm^3$ $G^{-2}$ $mol^{-1}$ $(4\pi\epsilon_0)$. $\Delta n_u$ defined for an induction field $B$ of 1 T, pressure $P$ of 1 atm, and $T=293.15$ K;

[c] $T=273.15$ K, $P=1$ bar, and $\nabla E=-1\times10^9$ V $m^{-2}$;

[d] $T=273.15$ K, $P=1$ bar, $B=3$ T, and $E=2.6\times10^6$ V $m^{-1}$;

[e] $\lambda=632.8$ nm from Ref. 16. We have extrapolated their fitted temperature-dependence linear equation to $T=273.15$ K to calculate ${}_mK$;

[f] $\Delta n$ from Ref. 13 with $\lambda=632.8$ nm, $T=293.15$ K. We have extrapolated their data to calculate ${}_mC$ to $T=293.15$ K;

[g] $\lambda=632.8$ nm from Ref. 14. We have extrapolated their fitted temperature-dependence linear equation to $T=273.15$ K to calculate ${}_mQ$;

[h] $\Delta n$ measured at $\lambda=441.6$ nm and $T=304.1$ K across the temperature range of 304.1–453.5 K [Ref. 12]. We have extrapolated their data to 273.15 K to calculate ${}_mC$.

browsing through the results in Table IV, is only about 3% to 4% of ${}_mC(\lambda,T)$ (instead of the 20% given in Ref. 4).

The effect of electron correlation on $\Delta\eta(\omega)$ is far more dramatic than seen in the original paper mostly due to the fact that the near cancellation of the paramagnetic and diamagnetic contribution yields an anisotropy of the hypermagnetizability that is far smaller than originally computed, and therefore far more sensitive to changes in the electron correlation treatment. Note that basis sets of double zeta quality yield the sign of $\Delta\eta(\omega)$ opposite to that obtained with more extended (triple and quadruple zeta) basis sets. As a consequence of the reduced importance of the contribution of $\Delta\eta(\omega)$ to ${}_mC(\lambda,T)$ and $\Delta n_u(\lambda,T)$, we reduce our estimates given in Ref. 4 to ${}_mC(\lambda,T)\approx(10\pm1)\times10^{-19}$ $cm^3$ $G^{-2}$ $mol^{-1}$ $(4\pi\epsilon_0)$, and $\Delta n_u(\lambda,T)\approx(6\pm1)\times10^{-14}$, respectively. With the change of sign in $b(\omega)$ also, the prediction of the Buckingham Effect (BE) constant and BE birefringence of $BF_3$ changes, although only slightly: ${}_mQ(\lambda,T)\approx(-4.4\pm0.3)\times10^{27}$ a.u. and $\Delta n_u(\lambda,T)\approx(4.5\pm0.3)\times10^{-15}$, respectively. While the change in sign of $b(\omega)$ modifies remarkably our revised value of the quadrupole moment of $BF_3$ (see above), the revised value of ${}_mQ(\lambda,T)$ at $T=293.15$ K, given in the original paper as $(-3.9\pm0.2)\times10^{27}$ a.u., does not change.

## V. $BCl_3$

Table V replaces Table V of Ref. 4, whereas Fig. 1 replaces Fig. 1 thereof.

The temperature-independent CME contribution of $BCl_3$ is indeed similar to that of $BF_3$, as stated in Ref. 4, meaning that on the average, it is ≈5% of the ${}_mC(\lambda,T)$, never exceeding 10%. The value of 20% given originally is therefore overestimated.

The revision of the magnetizability anisotropy value of $BCl_3$ given by Lamb and Ritchie,[7,8] revision made by employing the new best estimate for the hypermagnetizability anisotropy, the B3LYP-DFT/d-aug-cc-pVTZ value in Table V ($\Delta\eta=+11.5$ a.u.), now *confirms* not surprisingly the validity of the assumptions made by Lamb and Ritchie[7,8] when they neglected the temperature-independent contribution to the CME. The first two sentences of the second paragraph on p. 114307–10 of Ref. 4 now read:

"Ritchie and Lamb in Refs. 7 and 8 neglected the temperature-independent contribution. We have rederived the value for $\xi_{\mathrm{ani}}$ by fitting their experimental data such that the line in Eq. (2) passes through our estimated intercept (B3LYP/daug-cc-pVTZ: $\Delta\eta=+11.5$ a.u.), assuming as Lamb and Ritchie a value for the anisotropy of the electric dipole polarizability of $\alpha_{\mathrm{ani}}=-21.5\pm0.7$.[9] The experimental estimate of $\xi_{\mathrm{ani}}$ of $-0.71\pm0.09$ a.u. was confirmed."

The last row of Table II of Ref. 4, with its old erroneously revised value of $(-0.45\pm0.09)$ a.u. for $\xi_{\mathrm{ani}}$ of $BCl_3$, should therefore be taken away together with the associated footnote.

The last two paragraphs of Sec. IV C 2 should be replaced by the following:

"We predict a Cotton–Mouton constant of ${}_mC(\lambda,T)=(9\pm1)\times10^{-18}$ $cm^3$ $G^{-2}$ $mol^{-1}$ $(4\pi\epsilon_0)$ with an associated birefringence of $\Delta n_u(\lambda,T)=(5\pm1)\times10^{-13}$, under the conditions in Table V. With the exception of LDA-DFT, which

TABLE IV. CME and BE for $BF_3$ at $\lambda=632.8$ nm and $T=273.15$ K. Atomic units, with ${}_mC(\lambda,T)$ in cgs units of $cm^3\ G^{-2}\ mol^{-1}$ $(4\pi\epsilon_0)$. $\Delta n_u$ is the birefringence defined for an induction field $B$ of 1 T and a pressure $P$ of 1 atm according to Ref. 17. $\Delta n$ for Buckingham birefringence is given for a pressure of $P=1$ bar and EFG of $\nabla E=-1\times10^9$ V $m^{-2}$.

| | | CME | | | Buckingham | | |
|---|---|---|---|---|---|---|---|
| Wave function | Basis | $\Delta\eta(\omega)$ | ${}_mC(\lambda,T)\times10^{19}$ | $\Delta n_u\times10^{14}$ | $b(\omega)$ | ${}_mQ(\lambda,T)\times10^{-28}$ | $\Delta n\times10^{14}$ |
| HF-SCF | aug-cc-pVDZ | −6.4 | 6.17 | 3.72 | 26.3 | −3.71 | 3.74 |
| | d-aug-cc-pVDZ | −9.2 | 3.46 | 2.09 | 33.6 | −3.78 | 3.81 |
| | aug-cc-pVTZ | 0.8 | 5.55 | 3.34 | 30.7 | −3.74 | 3.77 |
| | d-aug-cc-pVTZ | 3.9 | 5.35 | 3.22 | 34.0 | −3.61 | 3.64 |
| | aug-cc-pVQZ | 2.5 | 5.38 | 3.24 | 32.9 | −3.62 | 3.65 |
| LDA | aug-cc-pVDZ | −1.8 | 13.70 | 8.25 | 45.7 | −4.28 | 4.31 |
| | d-aug-cc-pVDZ | −7.0 | 11.86 | 7.15 | 58.2 | −4.35 | 4.38 |
| | aug-cc-pVTZ | 5.3 | 12.82 | 7.72 | 53.3 | −4.44 | 4.47 |
| | d-aug-cc-pVTZ | 9.1 | 12.37 | 7.45 | 59.8 | −4.26 | 4.29 |
| | aug-cc-pVQZ | 7.0 | 12.46 | 7.50 | 57.1 | −4.29 | 4.32 |
| B3LYP | aug-cc-pVDZ | −3.0 | 10.79 | 6.50 | 40.6 | −4.32 | 4.35 |
| | d-aug-cc-pVDZ | −7.7 | 8.78 | 5.29 | 51.8 | −4.36 | 4.39 |
| | aug-cc-pVTZ | 4.2 | 9.86 | 5.94 | 47.3 | −4.41 | 4.44 |
| | d-aug-cc-pVTZ | 7.6 | 9.44 | 5.69 | 53.2 | −4.23 | 4.26 |
| | aug-cc-pVQZ | 5.8 | 9.65 | 5.81 | 50.5 | −4.27 | 4.30 |
| KT1 | aug-cc-pVDZ | −0.4 | 11.72 | 7.06 | 51.4 | −4.07 | 4.10 |
| | d-aug-cc-pVDZ | −5.6 | 9.15 | 5.51 | 64.5 | −4.12 | 4.14 |
| | aug-cc-pVTZ | 7.2 | 10.40 | 6.26 | 60.0 | −4.19 | 4.22 |
| | d-aug-cc-pVTZ | 11.0 | 10.13 | 6.10 | 66.9 | −4.03 | 4.06 |
| | aug-cc-pVQZ | 8.8 | 10.33 | 6.22 | 64.0 | −4.07 | 4.10 |
| CCSD | aug-cc-pVDZ | −5.9 | 10.40 | 6.26 | 30.0 | −4.43 | 4.46 |
| | d-aug-cc-pVDZ | −10.5 | 8.49 | 5.12 | 47.6 | −4.44 | 4.47 |
| | aug-cc-pVTZ | 1.9 | 9.35 | 5.63 | 41.9 | −4.34 | 4.38 |

[a] B3LYP GIAO magnetizability anisotropies are employed in place of the nonGIAO CCSD results.

TABLE V. CME and BE for $BCl_3$ at $\lambda=632.8$ nm and $T=273.15$ K. Atomic units, with ${}_mC(\lambda,T)$ in cgs units of $cm^3\ G^{-2}\ mol^{-1}$ $(4\pi\epsilon_0)$. $\Delta n_u$ is the birefringence defined for an induction field $B$ of 1 T and a pressure $P$ of 1 atm according to Ref. 17. $\Delta n$ for Buckingham birefringence is given for a pressure of $P=1$ bar and EFG of $\nabla E=-1\times10^9$ V $m^{-2}$.

| | | CME | | | Buckingham | | |
|---|---|---|---|---|---|---|---|
| Wave function | Basis | $\Delta\eta(\omega)$ | ${}_mC(\lambda,T)\times10^{18}$ | $\Delta n_u\times10^{13}$ | $b(\omega)$ | ${}_mQ(\lambda,T)\times10^{-27}$ | $\Delta n\times10^{15}$ |
| HF | aug-cc-pVDZ | −82.4 | 6.93 | 4.17 | 207.4 | −8.46 | 8.52 |
| | d-aug-cc-pVDZ | −138.3 | 5.05 | 3.05 | 278.9 | −7.52 | 7.58 |
| | aug-cc-pVTZ | −54.4 | 7.10 | 4.28 | 260.2 | −7.91 | 7.97 |
| | d-aug-cc-pVTZ | −1.7 | 7.31 | 4.40 | 285.0 | −7.57 | 7.63 |
| LDA-DFT | aug-cc-pVDZ | −30.5 | 13.13 | 7.91 | 247.6 | −4.79 | 4.82 |
| | d-aug-cc-pVDZ | −129.0 | 11.42 | 6.88 | 344.6 | −4.26 | 4.29 |
| | aug-cc-pVTZ | −35.2 | 13.48 | 8.12 | 311.7 | −4.05 | 4.07 |
| | d-aug-cc-pVTZ | 19.9 | 13.70 | 8.25 | 352.7 | −3.74 | 3.77 |
| B3LYP-DFT | aug-cc-pVDZ | −43.7 | 10.64 | 6.41 | 236.3 | −7.81 | 7.87 |
| | d-aug-cc-pVDZ | −133.4 | 8.94 | 5.39 | 328.6 | −7.08 | 7.13 |
| | aug-cc-pVTZ | −40.5 | 10.57 | 6.37 | 296.9 | −7.10 | 7.15 |
| | d-aug-cc-pVTZ | 11.5 | 10.75 | 6.47 | 336.8 | −6.64 | 6.69 |
| KT1-DFT | aug-cc-pVDZ | −36.8 | 8.97 | 5.40 | 261.9 | −4.23 | 4.26 |
| | d-aug-cc-pVDZ | −138.5 | 7.29 | 4.39 | 360.6 | −3.58 | 3.61 |
| | aug-cc-pVTZ | −44.2 | 8.92 | 5.37 | 325.9 | −3.99 | 4.02 |
| | d-aug-cc-pVTZ | 11.3 | 9.47 | 5.70 | 369.7 | −3.67 | 3.69 |
| CCSD[a] | aug-cc-pVDZ | −60.2 | 8.73 | 5.26 | 227.6 | −8.06 | 8.12 |
| | d-aug-cc-pVDZ | −130.2 | 6.45 | 3.89 | 310.1 | −6.97 | 7.02 |
| | aug-cc-pVTZ | −44.2 | 8.78 | 5.29 | 284.7 | −6.37 | 6.42 |
| Extrapolated from expt. | | | 8.8 ± 0.7 [b] | 5.3 ± 0.7 [b] | | | |

[a] B3LYP-DFT GIAO magnetizability anisotropies are employed in place of the nonGIAO CCSD results.

[b] Data in Refs. 7 and 8 fitted in this work assuming linear regression with a $T\rightarrow\infty$ value of ${}_mC(\lambda,T)$ equal to our B3LYP-DFT/daug-cc-pVTZ "best value" of $\Delta\eta(\omega)=11.5$ a.u.

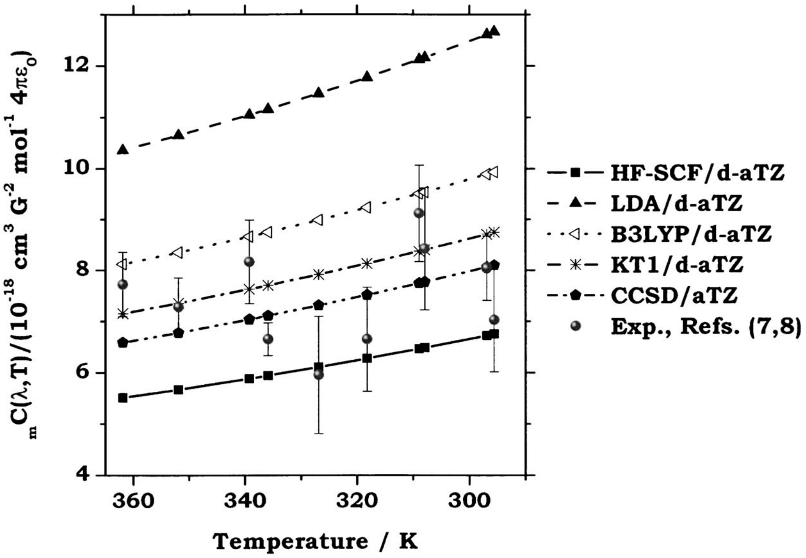

FIG. 1. The temperature dependence of the CME of $BCl_3$ as computed here, compared to the experiment. The labels "aTZ" and "d-aTZ" stand for aug-cc-pVTZ and daug-cc-pVTZ, respectively.

overestimates the effect, we are inside the error bars of experiment and close to the center of the statistical distribution in particular with KT1-DFT and CCSD.

Finally, we predict the BE constant to be $_mQ(\lambda,T) = (-6\pm 1)\times 10^{27}$ a.u. with an associated birefringence of $\Delta n(\lambda,T) = (6\pm 1)\times 10^{-15}$. No experimental measurements are available for these constants."

In the conclusions, Sec. V of Ref. 4, the last three paragraphs should be replaced by the following:

"The agreement with experimental data is satisfactory, particularly in view of the neglect of molecular vibrations. The temperature-independent contribution to the Cotton–Mouton birefringence is about 5% for both molecules, whereas the contributions to the BE are about 5% and 10% for $BF_3$ and $BCl_3$, respectively.

We have carried out a detailed and systematic investigation of the molecular quadrupole moment of both molecules, yielding $(3.00\pm 0.01)$ and $(0.71\pm 0.01)$ a.u. for $BF_3$ and $BCl_3$, respectively. For $BF_3$, this value is within one standard deviation of our revised experimental measurement, while for $BCl_3$, our value supports the claims of Lamb and Ritchie[7,8,10] that the measurement of Gierszal *et al.*[11] is inaccurate.

Our best *ab initio* result for the magnetizability anisotropy of boron trichloride $\xi_{\mathrm{ani}}$ is in good agreement with the results of the measurements performed by Lamb and Ritchie,[7,8] and excellent agreement is also observed between theory and experiment for the Cotton–Mouton constant of $BCl_3$."

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