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Addition/Correction

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Correction to "Analytic Density-Functional Theory Calculations of Pure Vibrational Hyperpolarizabilities: The First Dipole Hyperpolarizability of Retinal and Related Molecules"

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We recently presented a study of the second-harmonic generation (SHG) of retinal and five related molecules using a new, general approach for the analytic calculation of pure vibrational contributions to the molecular (hyper)-polarizabilities at the density-functional (B3LYP) level of theory. The molecules were selected by considering the availability of experimental data.[1,2] In addition to retinal, SHG hyperpolarizabilities have been measured for retinol, retinoic acid, vitamin A acetate, retinal Schiff base, and protonated retinal Schiff base.[1,2] Unfortunately, we used in our calculations the chemical structures depicted in Figure 1 of Hendrickx et al.[1] (in particular, molecule no. 4) and Figure 1 of Ghosh et al.[2] (molecule d); thus our results were obtained for five molecules corresponding to those studied in experiments, but not in the case of vitamin A acetate. Because it is clear from the text of the experimental works that the measured values were in fact determined for vitamin A acetate, not for the molecule depicted in these figures, we report here for completeness the properties of vitamin A acetate. The results are shown in Tables 1−3, and these tables should replace the corresponding data in the original Tables 1−3.

The computed properties of vitamin A acetate are similar to the properties of the other five molecules. In particular, this applies to the Hartree−Fock and B3LYP frequency-dependent first hyperpolarizability (in contrast to the previously studied molecule). The ordering of the β (electronic) magnitudes is now

PRSB > retinal > retinoic acid > RSB > vitamin A acetate > retinol

both at the B3LYP and Hartree−Fock level of theory, whereas in experiment[1,2] it was

PRSB > retinal > RSB > retinoic acid > vitamin A acetate > retinol

REFERENCES

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(2) Ghosh, S.; Ranjini, A. S.; Pandey, R.; Das, P. K. First Hyperpolarizability of Bacteriorhodopsin, Retinal and Related Molecules Revisited. *Chem. Phys. Lett.* **2009**, *474*, 307−310.

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Table 1. Electric Dipole Polarizabilities (10^{-25} esu)

wavelength, nm	retinal	retinol	retinoic acid	vitamin A acetate	retinal Schiff base	protonated RSB
			Electronic, HF			
532	558.90	507.79	547.52	553.23	686.64	1420.28
771	497.37	466.93	492.35	509.79	612.81	865.69
953	483.15	456.85	479.36	499.08	595.71	794.54
1064	478.27	453.33	474.89	495.35	589.84	772.79
1543	468.53	446.22	465.93	487.80	578.12	732.68
1907	465.65	444.10	463.28	485.54	574.66	721.63
∞	460.40	440.19	458.42	481.41	568.33	702.31
			Vibrational, HF			
532	−0.17	−0.08	−0.20	−0.14	−0.12	−1.76
771	−0.36	−0.18	−0.43	−0.30	−0.25	−3.73
953	−0.56	−0.28	−0.67	−0.46	−0.39	−5.73
1064	−0.71	−0.35	−0.84	−0.58	−0.49	−7.18
1543	−1.57	−0.80	−1.88	−1.30	−1.13	−15.55
1907	−2.57	−1.35	−3.09	−2.13	−1.92	−24.55
∞	156.57	355.29	132.06	502.32	80.83	492.34
			Electronic, DFT			
532	809.45	634.85	763.57	694.03	964.09	−8552.58
771	603.95	540.48	593.33	592.39	741.08	1151.73
953	571.16	521.23	564.29	571.73	703.24	975.43
1064	560.72	514.81	554.93	564.85	691.08	929.39
1543	540.92	502.23	537.04	551.36	667.86	852.06
1907	535.33	498.58	531.96	547.44	661.27	832.25
∞	525.41	492.00	522.90	540.38	649.54	798.99
			Vibrational, DFT			
532	−0.15	−0.07	−0.17	−0.12	−0.10	−0.69
771	−0.31	−0.15	−0.36	−0.25	−0.21	−1.45
953	−0.48	−0.24	−0.56	−0.39	−0.32	−2.24
1064	−0.61	−0.30	−0.70	−0.50	−0.41	−2.80
1543	−1.35	−0.68	−1.55	−1.10	−0.93	−6.09
1907	−2.20	−1.15	−2.54	−1.80	−1.58	−9.66
∞	132.73	117.71	111.13	421.96	67.75	256.99

Table 2. SHG Hyperpolarizabilities (10^{-30} esu)

wavelength, nm	retinal	retinol	retinoic acid	vitamin A acetate	retinal Schiff base	protonated RSB
			Electronic, HF			
1064	40.99	−2.20	32.76	6.54	14.17	−601.59
1543	27.07	−1.46	21.99	4.26	8.67	−223.57
1907	24.10	−1.30	19.65	3.75	7.53	−179.04
∞	19.58	−1.05	16.07	2.97	5.81	−124.46
			Vibrational, HF			
1064	−1.05	−0.00	−0.86	−0.10	−0.47	9.72
1543	−2.25	−0.01	−1.83	−0.21	−1.01	20.89
1907	−3.51	−0.01	−2.83	−0.31	−1.59	32.65
∞	168.37	88.45	136.01	73.86	61.16	−805.60
			Electronic, DFT			
1064	250.17	−18.97	190.99	48.90	116.65	6542.78
1543	101.32	−9.34	83.44	23.26	45.52	−305.43
1907	81.71	−7.74	68.17	19.06	35.94	−204.25
∞	57.15	−5.60	48.58	13.46	24.03	−113.36
			Vibrational, DFT			
1064	−1.07	0.03	−0.91	−0.19	−0.59	2.30
1543	−2.30	0.06	−1.95	−0.40	−1.27	4.93
1907	−3.59	0.09	−3.01	−0.60	−1.99	7.68
∞	187.76	−26.34	142.64	52.39	87.30	−213.31

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Table 3. Rescaled SHG Electronic Hyperpolarizabilities[a]

wavelength, nm	retinal	retinol	retinoic acid	vitamin A acetate	retinal Schiff base	protonated RSB
			This Work, HF			
∞	1.00	−0.05	0.82	0.15	0.30	−6.36
			This Work, DFT			
∞	1.00	−0.10	0.85	0.24	0.42	−1.98
			Frequency Dispersion			
			This Work, HF			
1064	2.093	2.089	2.039	2.198	2.437	4.834
			This Work, DFT			
1064	4.377	3.389	3.931	3.633	4.855	−57.716

[a]All the static hyperpolarizabilities are defined with respect to the analogous static retinal values, and all the dynamic hyperpolarizabilities are defined with respect to the corresponding static values of the same molecule.

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