Benchmarking magnetizabilities with recent density functionals

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Abstract

We have assessed the accuracy for magnetic properties of a set of 51 density functional approximations, including both recently published as well as already established functionals. The accuracy assessment considers a series of 27 small molecules and is based on comparing the predicted magnetizabilities to literature reference values calculated using coupled cluster theory with full singles and doubles and perturbative triples [CCSD(T)] employing large basis sets. The most accurate magnetizabilities, defined as the smallest mean absolute error, were obtained with the BHandHLYP functional. Three of the six studied Berkeley functionals and the three range-separated Florida functionals also yield accurate magnetizabilities. Also some older functionals like CAM-B3LYP, KT1, BHLYP (BHandH), B3LYP and PBE0 perform rather well. In contrast, unsatisfactory performance was generally obtained with Minnesota functionals, which are therefore not recommended for calculations of magnetically induced current density susceptibilities within this approach even when analytical approaches for magnetizabilities as the second derivative of the energy have not been implemented. The magnetizability density can also be visualized, providing additional information that is not otherwise easily accessible on the spatial origin of the magnetizabilities.

1 Introduction

Computational methods based on density-functional theory (DFT) are commonly used in quantum chemistry, because DFT calculations are rather accurate despite their relatively modest computational costs. Older functionals such as the Becke’88–Perdew’86 (BP86), Becke’88–Lee–Yang–Parr (BLYP) and Perdew–Burke–Ernzerhof (PBE) functionals at the generalized gradient approximation (GGA) as well as the B3LYP and PBE0 hybrid functionals are still often employed, even though newer functionals with improved accuracy for energies and electronic properties have been developed.

The accuracy and reliability of various den-
density functional approximations (DFAs) has been assessed in a huge number of applications and benchmark studies. It is important to note that functionals that are accurate for energetics may be less suited for calculations of other molecular properties. In specific, the accuracy of magnetic properties calculated within DFAs has been benchmarked by comparing magnetizabilities and nuclear magnetic shieldings to those obtained from coupled-cluster calculations using large basis sets, although modern DFAs have been less systematically investigated.

The same also holds for nuclear independent chemical shifts and magnetically induced current density susceptibilities, which have been studied for a large number of molecules, but whose accuracy has never been benchmarked properly. Magnetizabilities are usually calculated as the second derivative of the electronic energy with respect to the external magnetic perturbation:

$$\xi_{\alpha\beta} = -\frac{\partial^2 E}{\partial B_\alpha \partial B_\beta} \bigg|_{B=0}. \quad (1)$$

Such analytic implementations for magnetizabilities exist in several quantum chemistry programs. However, since the magnetic interaction energy in equation (2) can also be written as an integral over the magnetic interaction energy density $\rho^B(r)$ that is given by the scalar product of the magnetically induced current density $J^B(r)$ with the vector potential $A^B(r)$ of the external magnetic field $B$:

$$E = \int \rho^B(r) \, d^3r = -\frac{1}{2} \int A^B(r) \cdot J^B(r) \, d^3r, \quad (2)$$

an approach based on quadrature is also possible. As will be seen in section 2, the numerical integration approach for the magnetizability provides additional information about its spatial origin that is not available with the analytic approach based on second derivatives: the tensor components of the magnetizability density defined in section 2 are scalar functions that can be visualized, and the integration approach can be used to provide detailed information about the origin of the corresponding components of the magnetizability tensor. Similar approaches have been used in the literature for studying spatial contributions to nuclear magnetic shielding constants.

We will describe our methods for numerical integration of magnetizabilities using the current density susceptibility in sections 2 and 3. Then, in section 4, we will list the studied set of density functionals, and present the results in section 5: the functional benchmark is discussed in section 5.1, and magnetizability densities and spatial contributions to magnetizabilities are analyzed in section 5.2. The conclusions of the study are summarized in section 6.

### 2 Theory

The current density $J^B(r)$ in equation (2) is formally defined as the real part ($R$) of the mechanical momentum density,

$$J^B(r) = -R[\Psi^*(r)(p - A^B(r))\Psi(r)], \quad (3)$$

where $p = -i\nabla$ is the momentum operator. Substituting equation (2) into equation (1) straightforwardly leads to

$$\xi_{\alpha\beta} = \frac{\partial^2}{\partial B_\alpha \partial B_\beta} \frac{1}{2} \int A^B(r) \cdot J^B(r) \, d^3r \bigg|_{B=0}. \quad (4)$$

The current density susceptibility tensor (CDT) is defined as the first derivative of the magnetically induced current density with respect to the components of the external magnetic field in the limit of a vanishing magnetic field:

$$J^B_\gamma = \frac{\partial J^B}{\partial B_\beta} \bigg|_{B=0}. \quad (5)$$

The vector potential $A^B(r)$ of an external static homogeneous magnetic field is expressed as

$$A^B(r) = \frac{1}{2}B \times (r - R_O), \quad (6)$$
where $R_O$ is the chosen gauge origin. The $\alpha \beta$ component of the magnetizability tensor can then be obtained from equations (4), (5) and (6) as

$$\xi_{\alpha \beta} = \int \rho^\xi_{\alpha \beta}(r) d^3r, \quad (7)$$

where the magnetizability density is defined as

$$\rho^\xi_{\alpha \beta}(r) = \frac{1}{2} \sum_{\delta \gamma} \epsilon_{\alpha \delta \gamma} r_\delta J^B_{\gamma i}(r), \quad (8)$$

where $\epsilon_{\alpha \delta \gamma}$ is the Levi–Civita symbol, $\alpha$, $\beta$, $\gamma$, and $\delta$ are one of the Cartesian directions ($x$, $y$, $z$), and $r_\delta$ also denotes one of ($x$, $y$, $z$). The components of the magnetizability density tensor $\rho^\xi_{\alpha \beta}(r)$ are scalar functions that can be visualized to obtain information about the spatial contributions to the corresponding element of the magnetizability tensor $\xi_{\alpha \beta}$.

As the isotropic magnetizability ($\bar{\xi}$) is obtained as the average of the diagonal elements of the magnetizability tensor

$$\bar{\xi} = \frac{1}{3} \text{Tr} \, \xi = \int \bar{\rho}^\xi(r) d^3r, \quad (9)$$

we introduce the isotropic magnetizability density $\bar{\rho}^\xi(r)$ defined as

$$\bar{\rho}^\xi(r) = \frac{1}{3} \text{Tr} \, \rho^\xi(r), \quad (10)$$

which yields information about the spatial origin of the isotropic magnetizability, as we will demonstrate in section 5.2.

Although there is freedom with regard to the choice of the gauge origin of $A^B(r)$, the magnetic flux density $B$ is uniquely defined via equation (6), because $B = \nabla \times (A(r) + \nabla f(r))$ holds for any differentiable scalar function $f(r)$. The exact solution of the Schrödinger equation should also be gauge invariant. However, the use of finite one-particle basis sets introduces gauge dependence in quantum chemical calculations of magnetic properties. The CDT can be made gauge-origin independent by using gauge-including atomic orbitals (GIAOs), also known as London atomic orbitals (LAOs),32,54,55

$$\chi_{\mu}(r) = e^{-i(B \times [R_\mu - R_O]; r)/2} \chi^{(0)}_{\mu}(r), \quad (11)$$

where $i$ is the imaginary unit and $\chi^{(0)}_{\mu}(r)$ is a standard atomic-orbital basis function centered at $R_\mu$. GIAOs eliminate the gauge origin from the expression used for calculating the CDT; the expression we use is given in the supporting information (SI). Since the expression for the magnetizability density in equations (7) and (8) can be computed by quadrature, magnetizabilities can be obtained from the CDT even if the corresponding analytical calculation of magnetizabilities as the second derivative of the energy has not been implemented.

### 3 Implementation

The present implementation is based on the GIMIC program,56 and the NUMGRID library, which are both freely available open-source software. Gauge-independent CDTs can be calculated with GIMIC32–35 using the density matrix, the magnetically perturbed density matrices and information about the basis set.

In order to evaluate equation (7), a molecular integration grid is first generated from atom-centered grids with the NUMGRID library, as described by Becke58. In NUMGRID, the grid weights are scaled according to the Becke partitioning scheme using a Becke hardness of 3;58 the atom-centered grids are determined by a radial grid generated as suggested by Lindh et al., and angular grids due to Lebedev are used.

Given the quadrature grid, the diagonal elements of the magnetizability tensor are calculated in GIMIC from the Cartesian coordinates of the $n$ grid points multiplied with the CDT calculated in the grid points. For example, the $\xi_{xx}$ element of the magnetizability tensor is obtained from equation (7) as

$$\xi_{xx} = \sum_{i=1}^{n} \rho^\xi_{i,xx} \quad (12)$$

where the $xx$ component of the magnetizability density tensor at grid point $i$ is

$$\rho^\xi_{i,xx} = \frac{1}{2} \left[ (y J^B_x)_i - (z J^B_y)_i \right] \quad (13)$$
where \((yJ_x^{Bz})\) and \((zJ_y^{Bz})\) are the product of the \(z\) and \(y\) components of the CDT calculated in grid point \(i\) with the Cartesian coordinates \(y\) and \(z\) of the grid point, respectively, and the external magnetic field perturbation is along the \(x\) axis, \(B_x\). The \(\xi_{yy}\) and \(\xi_{zz}\) elements are obtained analogously.

4 Computational Methods

Calculations are performed for the set of 28 molecules studied in ref. 18 that also provides our molecular structures and the CCSD(T) reference values: AlF, C\(_2\)H\(_4\), C\(_3\)H\(_4\), CH\(_2\)O, CH\(_3\)F, CH\(_4\), CO, FCCH, FCN, H\(_2\)C\(_2\)O, H\(_2\)O, H\(_2\)S, H\(_2\)C\(_2\)O, HCN, HCP, HF, HFCO, HOF, LiF, LiH, N\(_2\), N\(_2\)O, NH\(_3\), O\(_3\), OCS, OF\(_2\), PN, and SO\(_2\). However, as in ref. 18, O\(_3\) was omitted from the analysis, since it is an outlier, and due to the fact that the reliability of the CCSD(T) level of theory is not guaranteed for this system: the perturbative triples correction to the magnetizability of O\(_3\) is \(-46.2 \times 10^{-30}\) J/T\(^2\), indicating that the CCSD(T) result might still have large error bars.\(^{18}\) The results of this work thus only pertain to the 27 other molecules, as in ref. 18.

Electronic structure calculations were performed with Hartree–Fock (HF) and the functionals listed in tables 1 and 2 using TURBOMOLE 7.5.\(^{110}\) Several rungs of Jacob’s ladder were considered when choosing the functionals listed in tables 1 and 2: local density approximations (LDA), generalized gradient approximations (GGAs), and meta-GGAs (mGGAs). Several kinds of functionals are also included: (pure) density functional approximations, global hybrid (GH) functionals with a constant amount of HF exchange, as well as range-separated (RS) hybrids with a given amount of HF exchange in the short range (SR) and the long range (LR). As can be seen in tables 1 and 2, the evaluated functionals consist of one pure LDA, 8 pure GGAs, 8 global hybrid GGAs, 10 range-separated hybrid GGAs, 12 mGGAs, 8 global hybrid mGGAs, and 4 range-separated mGGAs, in addition to HF.

The Dunning aug-cc-pCVQZ basis set\(^{111–115}\) (with aug-cc-pVQZ on the hydrogen atoms) and benchmark quality integration grids were employed in all calculations. Universal auxiliary basis sets\(^{116}\) were used with the resolution-of-the-identity approximation for the Coulomb interaction in all TURBOMOLE calculations. All density functionals were evaluated in TURBOMOLE with LIBXC,\(^{117}\) except the calculations with the recently published CAMh-B3LYP functional for which XCFUN was used.\(^{118}\) Magnetizabilities were subsequently evaluated with GIMIC by numerical integration of equation (7). The data necessary for evaluating the CDT in GIMIC were obtained from TURBOMOLE calculations of nuclear magnetic resonance (NMR) shielding constants employing GIAOs.\(^{54,55,110,119,120}\)

Although response calculations are not possible at the moment in the presence of the non-local correlation kernel used in \(\omega\)B97X-V, B97M-V, and \(\omega\)B97M-V, we have estimated the importance of the van der Waals (vdW) effects on the magnetic properties by comparing magnetizabilities obtained with orbitals optimized with and without the vdW term in the case of SO\(_2\). The magnetizability obtained with the vdW optimized orbitals differed by only \(0.4 \times 10^{-30}\) J/T\(^2\) (0.14\%) from that obtained from a calculation where the vdW term was omitted in the orbital optimization. Thus, the vdW term appears to have very little influence on magnetizabilities, as is already well-known in the literature for other properties.\(^{121}\) The vdW term was therefore not included in the calculations using the \(\omega\)B97X-V, B97M-V, and \(\omega\)B97M-V functionals in this study.

The accuracy of the numerical integration in GIMIC was assessed by comparing the TURBOMOLE/GIMIC magnetizability data to analytical values from PySCF,\(^{122}\) in which LIBXC\(^{117}\) was also used to evaluate the density functionals. Since PySCF does not currently support magnetizability calculations with mGGA functionals or range-separated functionals, further calculations were undertaken with Gaussian 16.\(^{123}\) The analytical magnetizabilities from PySCF and Gaussian were found to be in perfect agreement for the studied LDA and GGA functionals available in both codes (LDA,
Table 1: Functionals at the local density approximation (LDA) and the generalized gradient approximation (GGA) considered in this work. GH stands for global hybrid and RS for range separated hybrid. The amount of Hartree–Fock (HF) exchange, or exact exchange in the short range (SR) and long range (LR) are also given.

| Functional  | Hybrid | Type | Notes                  | Libxc ID$^a$   | References |
|-------------|--------|------|------------------------|----------------|------------|
| LDA         | LDA    |      |                        | 1+7            | 61–63      |
| BLYP        | GGA    |      |                        | 106+131        | 1,3,64     |
| BP86        | GGA    |      |                        | 106+132        | 1,2        |
| CHACHIYO    | GGA    |      |                        | 298+309        | 65,66      |
| KT1         | GGA    |      |                        | 167            | 67         |
| KT2         | GGA    |      |                        | 146            | 67         |
| KT3         | GGA    |      | PySCF data used        | 587            | 68         |
| N12         | GGA    |      |                        | 82+80          | 69         |
| PBE         | GGA    |      |                        | 101+130        | 4,5        |
| B3LYP       | GH     | GGA  | 20% HF                 | 402            | 6          |
| revB3LYP$^b$| GH     | GGA  | 20% HF                 | 454            | 70         |
| B97-2       | GH     | GGA  | 21% HF                 | 410            | 71         |
| B97-3       | GH     | GGA  | 26.9% HF               | 414            | 72         |
| BHLYP$^c$   | GH     | GGA  | 50% HF                 | 435            | 61,62,73   |
| BHandHLYP$^d$| GH   | GGA  | 50% HF                 | 436            | 1,73       |
| PBE0        | GH     | GGA  | 25% HF                 | 406            | 7,8        |
| QTP17       | GH     | GGA  | 62% HF                 | 416            | 74         |
| N12-SX      | RS     | GGA  | 25% SR, 0% LR          | 81+79          | 75         |
| CAM-B3LYP   | RS     | GGA  | 19% SR, 65% LR         | 433            | 76         |
| CAMh-B3LYP$^e$| RS | GGA  | 19% SR, 50% LR         | –              | 77         |
| CAM-QTP-00  | RS     | GGA  | 54% SR, 91% LR         | 490            | 78         |
| CAM-QTP-01  | RS     | GGA  | 23% SR, 100% LR        | 482            | 79         |
| CAM-QTP-02  | RS     | GGA  | 28% SR, 100% LR        | 491            | 80         |
| ωB97        | RS     | GGA  | 0% SR, 100% LR         | 463            | 81         |
| ωB97X       | RS     | GGA  | 15.8% SR, 100% LR      | 464            | 81         |
| ωB97X-D     | RS     | GGA  | 22.2% SR, 100% LR      | 471            | 82         |
| ωB97X-V     | RS     | GGA  | 16.7% SR, 100% LR      | 531            | 83         |

$^a$ Two numbers indicate the exchange and the correlation functional respectively. A single number indicates an exchange-correlation functional.

$^b$ Revised version

$^c$ Following King el al. in refs. 84–86, BHLYP is defined as 50% LDA exchange, 50% of HF exchange, and 100% LYP correlation. It is sometimes also known as BHandH, which is its keyword in GAUSSIAN.

$^d$ BHandHLYP is 50% Becke’88 exchange, 50% HF exchange, and 100% LYP correlation.

$^e$ CAMh-B3LYP is defined using the XCFUN library with $\alpha = 0.19; \beta = 0.31; \mu = 0.33$. 


Table 2: Meta-GGA functionals (mGGA) considered in this work. The notation is the same as in table 1.

| Functional     | Hybrid | Type     | Notes     | LIBXC ID* | References |
|----------------|--------|----------|-----------|-----------|------------|
| B97M-V         | mGGA   |          |           | 254       | 87         |
| M06-L          | mGGA   |          |           | 449+235   | 88         |
| revM06-Lb      | mGGA   |          |           | 293+294   | 89         |
| M11-L          | mGGA   |          |           | 226+75    | 90         |
| MN12-L         | mGGA   |          |           | 227+74    | 91         |
| MN15-L         | mGGA   |          |           | 268+269   | 92         |
| TASK           | mGGA   |          |           | 707+13    | 93,94      |
| MVS            | mGGA   |          |           | 257+83    | 95,96      |
| SCAN           | mGGA   |          |           | 263+267   | 97         |
| rSCANc         | mGGA   |          |           | 493+494   | 98         |
| TPSS           | mGGA   |          |           | 457       | 99,100     |
| revTPSSb       | mGGA   |          |           | 212+241   | 96,101     |
| TPSSh          | GH     | mGGA     | 10% HF    | 457       | 102        |
| revTPSShb      | GH     | mGGA     | 10% HF    | 458       | 96,101,102 |
| M06            | GH     | mGGA     | 27% HF    | 449+235   | 103        |
| revM06b        | GH     | mGGA     | 40.4% HF  | 305+306   | 104        |
| M06-2X         | GH     | mGGA     | 54% HF    | 450+236   | 103        |
| M08-HX         | GH     | mGGA     | 52.2% HF  | 295+78    | 105        |
| M08-SO         | GH     | mGGA     | 56.8% HF  | 296+77    | 105        |
| MN15           | GH     | mGGA     | 44% HF    | 268+269   | 106        |
| M11            | RS     | mGGA     | 42.8% SR, 100% LR | 297+76 | 107        |
| revM11b        | RS     | mGGA     | 22.5% SR, 100% LR | 304+172 | 108        |
| MN12-SX        | RS     | mGGA     | 25% SR, 0% LR | 248+73  | 75         |
| ωB97M-V        | RS     | mGGA     | 15% SR, 100% LR | 531     | 109        |

* Two numbers indicate the exchange and the correlation functional respectively.
  
  A single number indicates an exchange-correlation functional.
  
  b Revised version
  
  c Regularized version
BP86, PBE, PBE0, BLYP, B3LYP and BH-LYP). Comparison of the data from PySCF to the GIMIC data revealed the numerically integrated magnetizabilities to be accurate, as the magnetizabilities agreed within $0.5 \times 10^{-30}$ J/T$^2$ for all molecules using the B3LYP, B97-2, B97-3, BLYP, BP86, KT1, KT2, LDA, PBE, and PBE0 functionals; the small discrepancy may arise from use of the resolution-of-identity approximation in TURBOMOLE or from the numerical integration of the magnetizability density. A comparison of the raw data for BP86 and B3LYP is given in the SI.

The magnetizabilities calculated with Gaussian and TURBOMOLE using the meta-GGA functionals were found to differ. The discrepancies between the magnetizabilities obtained with the two programs are due to the use of different approaches to handle the gauge invariance of the kinetic energy density in meta-GGAs, which are described in refs. 125 and 126 for GAUSSIAN and TURBOMOLE, respectively. We found the TURBOMOLE data to be significantly closer to the CCSD(T) reference values.

Finally, since we found the implementation of the KT3 functional in Libxc version 5.0.0 used by TURBOMOLE to be flawed, the KT3 results in this study are based on calculations with PySCF with a corrected version of Libxc.

5 Results

5.1 Functional benchmark

The deviations of the DFT magnetizabilities from the CCSD(T) reference values of ref. 18 are visualized as ideal normal distributions (NDs) in figure 1. The visualization shows the idealized distribution of the error in the magnetizability for each functional, based on the computed mean errors (ME) and standard deviation of the error (STD) given in table 3. The raw data on the magnetizabilities and the differences from the CCSD(T) reference are available in the SI. Although the error distributions in figure 1 are instructive, we will employ mean absolute errors (MAEs) in order to rank the functionals studied in this work in a simple, unambiguous fashion. The MAEs are also given in table 3.

Examination of the data in table 3 shows that range-separated (RS) functionals generally yield accurate magnetizabilities. Judged by the mean absolute error, the best performance is obtained with the BHandHLYP GH functional. BHandHLYP is followed by 10 RS functionals, which have much sharper distributions than the rest of the studied functionals. The best performing RS functionals are three of the six Berkeley RS functionals ($\omega$B97X-V, $\omega$B97, $\omega$B97M-V) and the three RS functionals from the University of Florida’s Quantum Theory Project (QTP) CAM-QTP-00, CAM-QTP-01, and CAM-QTP-02. Five of these functionals have 100% long-range (LR) HF exchange, while the CAM-QTP-00 functional has 91% LR HF exchange. The two other RS Berkeley functionals with 100% LR exchange are ranked 11th ($\omega$B97X) and 21st ($\omega$B97X-D) among the studied functionals. The NDs of the studied RS GGA functionals are shown in figures 1(a) and 1(b), whereas the NDs of the studied RS mGGA functionals are shown in figure 1(c).

The CAM-B3LYP (65% LR HF exchange) and CAMh-B3LYP (50% LR HF exchange) functionals are among the top ten functionals (ranked 8th and 10th, respectively). CAM-B3LYP was designed for the accurate description of charge transfer excitations in a dipeptide model, while CAMh-B3LYP functional is aimed at excitation energies of biochromophores.

The best Minnesota functional, MN12-SX, is ranked 9th. MN12-SX is a highly parameterized functional with 58 parameters that is known to require the use of extremely accurate integration grids. Furthermore, since MN12-SX is a RS functional with HF exchange only in the short range (SR), it may have problems modeling magnetic properties of antiaromatic molecules sustaining strong ring currents in the paratropic (nonclassical) direction. We illustrate this with calculations on the strongly antiaromatic tetraoxa-isophlorin molecule in the Supporting Information: MN12-SX yields a magnetizability that is four times larger than the LMP2 [local second-order Møller–Plesset
Table 3: The mean absolute errors (MAEs), mean errors (MEs), and standard deviations (STDs) for the magnetizabilities of the 27 studied molecules in units of $10^{-30} \text{J/T}^2$ from the CCSD(T) reference with the studied functionals. The functionals are ordered in increasing MAE.

| Rank | Functional | MAE  | ME   | STD  | Rank | Functional | MAE  | ME   | STD  |
|------|------------|------|------|------|------|------------|------|------|------|
| 1    | BHandHLYP  | 3.11 | 2.15 | 4.65 | 27   | revTPSSh   | 7.14 | 7.05 | 5.94 |
| 2    | CAM-QTP-00 | 3.22 | 0.88 | 4.67 | 28   | TPSSh      | 7.20 | 7.07 | 6.02 |
| 3    | $\omega$B97X-V | 3.22 | 2.51 | 4.36 | 29   | B97-2      | 7.24 | 7.07 | 6.40 |
| 4    | CAM-QTP-01 | 3.23 | 0.59 | 4.49 | 30   | M08-HX     | 7.34 | 5.17 | 10.27|
| 5    | CAM-QTP-02 | 3.28 | -0.23| 4.36 | 31   | BLYP       | 7.91 | 5.69 | 8.75 |
| 6    | $\omega$B97 | 3.54 | 2.44 | 4.75 | 32   | N12-SX     | 8.04 | 7.89 | 7.48 |
| 7    | $\omega$B97M-V | 3.61 | 0.41 | 4.75 | 33   | revTPSS    | 8.20 | 7.86 | 6.68 |
| 8    | CAM-B3LYP  | 3.73 | 2.38 | 4.86 | 34   | TPSS       | 8.22 | 7.85 | 6.85 |
| 9    | MN12-SX    | 3.80 | 0.22 | 5.34 | 35   | revM11     | 8.23 | 6.83 | 10.03|
| 10   | CAMh-B3LYP | 4.23 | 3.22 | 5.17 | 36   | TASK       | 8.27 | 7.31 | 7.43 |
| 11   | $\omega$B97X | 4.25 | 3.71 | 5.22 | 37   | BP86       | 8.59 | 7.30 | 8.75 |
| 12   | QTP-17     | 4.58 | 3.77 | 5.45 | 38   | M11-L      | 8.92 | 5.20 | 9.26 |
| 13   | BHLYP      | 4.73 | 0.10 | 6.47 | 39   | revM06     | 8.94 | 8.67 | 10.27|
| 14   | B97M-V     | 5.19 | 4.13 | 5.58 | 40   | PBE        | 9.13 | 7.07 | 9.42 |
| 15   | revB3LYP   | 5.45 | 4.34 | 6.13 | 41   | KT3        | 9.19 | 8.38 | 8.08 |
| 16   | B3LYP      | 5.47 | 4.72 | 5.97 | 42   | LDA        | 9.55 | 5.37 | 11.36|
| 17   | MN12-L     | 5.79 | -2.03| 8.02 | 43   | CHACHIYO   | 9.76 | 9.17 | 8.88 |
| 18   | KT1        | 5.87 | 1.15 | 7.11 | 44   | M11        | 9.93 | 7.61 | 13.77|
| 19   | rSCAN      | 5.91 | 5.00 | 6.06 | 45   | M06-2X     | 10.15| 9.01 | 13.12|
| 20   | PBE0       | 5.96 | 5.56 | 6.81 | 46   | MVS        | 10.35| 9.92 | 9.20 |
| 21   | $\omega$B97X-D | 6.22 | 5.89 | 6.35 | 47   | M08-SO     | 10.40| 8.09 | 14.34|
| 22   | SCAN       | 6.30 | 5.89 | 5.96 | 48   | N12        | 10.89| 10.01| 9.58 |
| 23   | KT2        | 6.42 | 5.58 | 7.21 | 49   | MN15       | 11.45| 10.45| 12.82|
| 24   | MN15-L     | 6.57 | -5.27| 6.94 | 50   | M06-L      | 12.49| 12.45| 9.42 |
| 25   | B97-3      | 6.61 | 6.61 | 6.26 | 51   | M06        | 13.34| 13.11| 13.16|
| 26   | revM06-L   | 7.00 | 6.23 | 5.98 | 52   | HF         | 18.40| 7.48 | 61.81|
Figure 1: Normal distributions (ND) representing the errors in the magnetizabilities for the 27 benchmark reproduced by the studied functionals, obtained by plotting the data presented in table 3. The curves are ordered in each figure by increasing standard deviation. The NDs of RS functionals are shown in figures 1(a), 1(b) and 1(c). The NDs of the GH functionals are shown in figures 1(d), 1(e), 1(f) and 1(g). The NDs of the mGGA functionals are shown in figures 1(h), 1(i) and 1(j). The NDs of the LDA and GGA functionals are shown in figures 1(k) and 1(l).
perturbation theory] reference value, while the magnetizabilities from BHandHLYP and CAM-B3LYP are in good agreement with LMP2. The N12-SX functional ranked 32nd is also a RS functional with 0% LR exchange. The RS Minnesota functionals with 100% LR HF exchange (M11 and revM11) have large MAEs of \(9.93 \times 10^{-30} \text{ J}/\text{T}^2\) and \(8.87 \times 10^{-30} \text{ J}/\text{T}^2\) and are ranked 44th and 35th, respectively.

The best global hybrid (GH) functional is BHandHLYP, which is ranked 1st among all functionals of this study, as was already mentioned above. Among GHs, BHandHLYP is followed by QTP-17, which is ranked 12th. Old and established GH functionals like BHLYP a.k.a. BHandH, B3LYP, and PBE0 perform almost as well as QTP-17 and are ranked 13th, 16th, and 20th, respectively. The performance of revB3LYP is practically the same as for B3LYP; the same holds for revTPSSh and TPSSh. The other established GH functionals like B97-2, B97-3, TPSSh and newer ones like revTPSSh and M08-HX are found in the beginning of the second half of the ranking list, whereas M08-SO, M06, revM06, M06-2X, MN15, and M06 are ranked between 30th and 51st. The NDs of the GH functionals are compared in figures 1(d), 1(e), 1(f) and 1(g).

B97M-V, at the 14th place, is the best pure mGGA functional. The rSCAN and SCAN functionals are ranked 19th and 22nd, respectively, whereas revTPSS and TPSS appear at positions 33 and 34, respectively. The pure mGGA functionals of the Minnesota series are ranked 17th (MN12-L), 24th (MN15-L), 26th (revM06-L), and 50th (M06-L). The performance of the Minnesota pure mGGA functionals, excluding M06-L, is about the same as that of TASK and the other mGGA functionals. The magnetizabilities calculated with the revised M06-L (revM06-L) functional are more accurate than those with M06-L. The MVS mGGA functional is ranked 46th. The NDs for the mGGA functionals are shown in figures 1(h), 1(i) and 1(j).

The magnetizabilities calculated with several of the Minnesota functionals are inaccurate. Seven of the eight worst performing functionals (M11, M06-2X, MVS, M08-SO, N12, MN15, M06-L, M06) in table 3 are Minnesota functionals. Five other Minnesota functionals are also ranked in the lower half, placing 30th (M08-HX), 32nd (N12-SX), 35th (revM11), 38th (M11-L), and 39th (revM06).

The KT1 and KT2 functionals are the best GGA functionals, ranking 18th and 23rd, respectively; both KT1 and KT2 have been optimized for NMR shieldings.\(^{67}\) The older commonly-used GGAs i.e., BLYP, BP86, and PBE are ranked 31st, 37th, and 40th, respectively, which is only slightly better than KT3 ranked 41st and LDA ranked 42nd. The CHACHIYO and N12 functionals, which are newer GGAs, are ranked 43rd and 48th, respectively. The NDs of the GGA functionals and the LDA are shown in figures 1(k) and 1(l).

The magnetizabilities calculated at the HF level are significantly less accurate and have a much larger MAE-STD than those obtained at the DFT levels, and we cannot recommend the use of HF for magnetic properties.

### 5.2 Magnetizability densities

Spatial contributions to the magnetizability densities, i.e., the integrand in equation (7), are illustrated for \(\text{H}_2\text{O}, \text{NH}_3\) and \(\text{SO}_2\) in figure 2, with figure 3 showing the corresponding CDTs. The magnetizability densities are calculated with the gauge origin of the external magnetic field \((\mathbf{R}_O)\) at \((x, y, z) = (0, 0, 0)\). In the calculations on \(\text{H}_2\text{O}\) and \(\text{SO}_2\), the magnetic field perturbation is perpendicular to the molecular plane, while for \(\text{NH}_3\) the perturbation is parallel to the \(C_3\) symmetry axis. In the case of \(\text{H}_2\text{O}\), the current-density flux around the whole molecule (figure 3(a)) leads to the ring-shaped contribution shown in figure 2(a). The magnetic field along the symmetry axis of \(\text{NH}_3\) also results in a current-density flux around the molecule at the hydrogen atoms (figure 3(c)), giving rise to a similar ring-shaped contribution shown in figure 2(c).

The isotropic magnetizability density of \(\text{SO}_2\) shown in figure 2(b) has positive (green) and negative (pink) values. Calculations of the CDT show that the oxygens sustain a strong diatropic atomic CDT that flows around the
Figure 2: Visualization of the isotropic magnetizability density $\rho^e(r)$ (equation (10)) shown in the molecular plane of H$_2$O 2(a) and SO$_2$ 2(b) as well as in the plane formed by the hydrogen atoms of NH$_3$ 2(c), positioned 0.06 $a_0$ away from the N atom towards the hydrogen atoms. Negative contributions are shown in pink, and positive ones in green. The gauge origin $R_O$ is $(0, 0, 0) a_0$.

Figure 3: Streamline representation of the CDT (equation (5)) of H$_2$O (3(a)), SO$_2$ (3(b)) and NH$_3$ (3(c)). The CDT is calculated with the magnetic field perpendicular to the molecular plane of H$_2$O and SO$_2$ as well as with it along the symmetry axis of NH$_3$. The color scale represents the strength of the CDT in $nAT^{-1}a_0^{-2}$. 
atom, whereas the atomic CDT of the sulfur atom is much weaker (figure 3(b)). The $p$-orbital shaped contributions to the magnetizability density of SO$_2$ around the oxygens in figure 2(b) originate from the atomic CDTs. The patterns of the CDT of H$_2$O and SO$_2$ lead to the different magnetizability densities seen in figures 2(a) and 2(b), respectively. The positive magnetizability densities in H$_2$O and NH$_3$ are extremely localized close to the atomic nuclei, also because of vortices of the atomic CDT.

The magnetizability density depends on the gauge origin of the vector potential of the external magnetic field, even though the magnetizability is independent of the gauge origin.

The magnetizability densities for H$_2$O, NH$_3$ and SO$_2$ calculated with the gauge origin at $R_O = (1, 1, 1) \ a_0$ are shown in the SI. The contribution of the choice of the gauge origin to the magnetizability computed from equation (7) vanishes when the CDT fulfills the charge conservation condition

$$\int J_0^{B_\beta}(r)d^3r = 0.$$ (14)

Calculating the magnetizability for NH$_3$ with a gauge origin set to $R_O = (100, 100, 100) \ a_0$ yielded a value that differs by 0.32% from the one computed for $R_O = (0, 0, 0)$. When the gauge origin is set to $R_O = (1, 1, 1) \ a_0$, the deviation is two orders of magnitude smaller, because the change in the magnetizability depends linearly on the relative position of the gauge origin. The magnetizabilities of H$_2$O and SO$_2$ also change by only 0.46% and 0.03% when moving the gauge origin from $(0, 0, 0) \ a_0$ to $(100, 100, 100) \ a_0$, respectively, showing that that charge conservation is practically fulfilled in our calculations. All other positions than $(0, 0, 0)$ for the gauge origin lead to a spurious CDT contribution to the magnetizability density.

The GIAO ansatz modifies the atomic orbitals leading to a magnetic response of an external magnetic field that is correct to the first order for the one-center problem. Even though they do not guarantee that the integral condition for the charge conservation of the CDT is fulfilled, the basis set convergence is faster and the leakage of the CDT is much smaller when GIAOs are used.

### 6 Conclusions

We have calculated magnetizabilities for a series of small molecules using both recently published density functionals, as well as older, established density functionals. The accuracy of the magnetizabilities predicted by the various density functional approximations has been assessed by comparison to coupled-cluster calculations with singles and doubles and perturbative triples [CCSD(T)] reported by Lutnaes et al. Our results are summarized graphically in figure 4: the top functionals afford both small mean absolute errors and standard deviations, but the same is not true for all recently suggested functionals.

Numerical methods for calculating magnetizabilities based on quadrature of the magnetizability density have been implemented. We have shown that this method allows studies of spatial contributions to the magnetizabilities by visualization of the magnetizability density. The method has been employed to calculate magnetizabilities from magnetically induced current density susceptibilities, which were obtained from TURBOMOLE calculations of nuclear magnetic shielding constants. Thus, magnetizabilities can be calculated in this way with TURBOMOLE even though analytical methods to calculate magnetizabilities as the second derivative of the energy are not yet available in this program. Further information about spatial contributions to the magnetizability could be obtained in the present approach by studying atomic contributions and investigating the positive and negative parts of the integrands separately in analogy to our recent work on nuclear magnetic shieldings in ref. 53, which may be studied in future work.

Our calculations show that the most accurate magnetizabilities (judged by the smallest MAE) for the studied database are obtained with BHandHLYP, which is an old global hybrid with 50% HF exchange and 50% B88 exchange. The calculations also show that...
Figure 4: The mean absolute errors (blue solid line) as well as the errors’ standard deviations (red crosses) of the magnetizabilities in $10^{-30}$ J/T$^2$ of the 27 studied molecules obtained with the 51 functionals compared to the CCSD(T) reference.

the modern range-separated functionals with 100% long-range HF exchange developed by Head-Gordon and co-workers and by Bartlett and co-workers yield accurate magnetizabilities for the database. Calculations with other range-separated functionals like CAM-B3LYP and CAMh-B3LYP as well as with global hybrid functionals like QTP-17, BHLYP a.k.a. BHandH, B3LYP and PBE0 yield relatively accurate magnetizabilities for the studied molecules. Meta-GGA functionals are found to yield somewhat better magnetizabilities than GGA and LDA functionals.

However, functionals developed by Truhlar and co-workers do not appear to be well-aimed for calculations of magnetizabilities and other magnetic properties that involve magnetically induced current densities. Magnetizabilities calculated using the popular M06-2X functional are found to be unreliable, and we do not recommend the use of the M06-2X functional in calculations of nuclear magnetic shieldings, magnetizabilities, ring-current strengths and other magnetic properties that depend on magnetically induced current density susceptibilities. Previous studies have also suggested that the M06-2X functional sometimes underestimates magnetizabilities and ring-current strengths.$^{128,129,132}$ Revised versions of Minnesota functionals have been studied in this work, and found to yield somewhat more accurate magnetizabilities than the original parameterizations. However, the revised versions also still appear on the second half of the ranking list.

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Supporting Information Available

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Magnetically induced current-density susceptibilities

The use of GIAOs eliminates the gauge origin ($\mathbf{R}_O$) from the expression we use for calculating the CDT, which is given in equation (15) in table 4. In the expression, $\mathbf{p}$ is the momentum operator, $m_\alpha$ are the Cartesian components ($\alpha$) of the magnetic moment of nucleus $I$, $B_\beta$ are the Cartesian components ($\beta$) of the external magnetic field, $\mathbf{D}$ is the density matrix in the atomic-orbital basis, $[\partial \mathbf{D} / \partial B]_{B=0}$ are the magnetically perturbed density matrices, $\epsilon_{\alpha\beta\gamma}$ is the Levi-Civita symbol, $\mathbf{h}(\mathbf{r})$ denotes the magnetic...
interaction operator without the $| \mathbf{r} - \mathbf{R}_I |^{-3}$ denominator with

$$
\frac{\partial \tilde{h}(\mathbf{r})}{\partial \mathbf{m}_I} = (\mathbf{r} - \mathbf{R}_I) \times \mathbf{p}
$$

(16)

and

$$
\frac{\partial^2 \tilde{h}(\mathbf{r})}{\partial \mathbf{m}_I \partial \mathbf{B}} = \frac{1}{2} [(\mathbf{r} - \mathbf{R}_O) \cdot (\mathbf{r} - \mathbf{R}_I) 1 - (\mathbf{r} - \mathbf{R}_O) (\mathbf{r} - \mathbf{R}_I)],
$$

(17)

and $\mathbf{R}_I$ is the position of nucleus $I$. All terms that contain the gauge origin $\mathbf{R}_O$ cancel in equation (15), making the CDT calculation independent of the gauge origin; this is demonstrated in figure 5 for a different choice of the gauge origin. All terms containing the nuclear position $\mathbf{R}_I$ also cancel, eliminating explicit references to the nuclear coordinates.

Calculations on tetraoxa-isophlorin

Valiev et al.\textsuperscript{127} found the isotropic magnetizability of tetraoxa-isophlorin (molecule V in their work) to be 15.8 a.u. at the LMP2/cc-pVDZ level of theory [local second-order Møller–Plesset perturbation theory], while B3LYP/def2-TZVP calculations yielded a value of 65.9 a.u., which is over four times the LMP2 value. Repeating the calculations of Valiev et al.\textsuperscript{127} with the present approach using the def2-TZVP basis set, we obtained a magnetizability of 65.2 a.u. at the B3LYP level, which agrees within 1% with the value of Valiev et al.\textsuperscript{127}; this difference can be tentatively attributed to the use of density fitting in the present work. MN12-SX, which has no exact exchange in the long range, predicts a susceptibility of 63.4 a.u., which is four times larger than the LMP2/cc-pVDZ value and close to the B3LYP value. Functionals with no exact exchange like PBE yield even larger values, > 100 a.u. In contrast, calculations at the CAM-B3LYP level with 65% LR HF exchange yield a magnetizability of 20.7 a.u., which agrees well with the LMP2 reference value. BHHandHLYP contains 50% LR HF exchange, and yields a magnetizability of 23.7 a.u., which is also in qualitative agreement with LMP2. Range-separated functionals with 100% LR HF exchange like $\omega B97X$ and $\omega B97$ yield a magnetizability for tetraoxa-isophlorin that is close to zero or even negative like the HF value, which is $-11.6$ a.u.\textsuperscript{127} The B3LYP/def2-TZVP optimized geometry of ref. 127, attached here in xyz format, was used in the calculations on tetraoxa-isophlorin.
Figure 5: Visualization of the isotropic magnetizability density $\tilde{\rho}^c(\mathbf{r})$ shown in the molecular plane of H$_2$O 5(a) and SO$_2$ 5(b) as well as in the plane formed by the hydrogen atoms of NH$_3$ 5(c), positioned 0.06 $a_0$ away from the N atom towards the hydrogen atoms. Negative contributions are shown in pink, and positive ones in green. The gauge origin $\mathbf{R}_O$ is $(1,1,1)$ $a_0$.

Table 5: Magnetizabilities in units of $10^{-30}$J/T$^2$ for the B3LYP, B97-2, B97-3, B97M-V, and BHandHLYP functionals in the aug-cc-pCVQZ basis set from calculations with TURBOMOLE and GIMIC compared to CCSD(T) data from ref. 18.

| Molecule | B3LYP | B97-2 | B97-3 | B97M-V | BHandHLYP | CCSD(T) |
|----------|-------|-------|-------|--------|------------|---------|
| AlF      | -396.5 | -393.4 | -392.6 | -396.1 | -395.6 | -394.5 |
| C$_2$H$_4$ | -336.7 | -334.3 | -334.6 | -334.8 | -343.0 | -345.6 |
| C$_3$H$_4$ | -463.1 | -462.6 | -464.1 | -460.6 | -468.8 | -478.9 |
| CH$_2$O  | -114.9 | -116.6 | -115.4 | -132.8 | -123.8 | -127.4 |
| CH$_3$F  | -312.4 | -312.2 | -313.3 | -309.4 | -314.9 | -315.7 |
| CH$_4$   | -317.0 | -314.6 | -314.6 | -313.2 | -315.7 | -316.9 |
| CO       | -206.6 | -202.5 | -201.2 | -208.5 | -205.0 | -209.5 |
| FCCH     | -440.1 | -438.3 | -439.2 | -440.5 | -443.6 | -441.6 |
| FCN      | -367.4 | -365.4 | -365.9 | -368.3 | -370.5 | -370.0 |
| H$_2$C$_2$O | -422.1 | -421.2 | -421.0 | -425.9 | -425.0 | -423.9 |
| H$_2$O   | -236.7 | -233.5 | -233.9 | -233.4 | -234.0 | -235.1 |
| H$_2$S   | -455.1 | -452.0 | -452.4 | -452.3 | -453.5 | -455.1 |
| H$_4$C$_2$O | -526.9 | -527.3 | -529.0 | -519.7 | -534.5 | -535.2 |
| HCN      | -269.4 | -265.0 | -265.4 | -268.9 | -272.7 | -271.8 |
| HCP      | -487.4 | -481.9 | -482.9 | -485.9 | -494.3 | -492.8 |
| HF       | -178.4 | -175.9 | -176.2 | -176.1 | -175.8 | -176.4 |
| HFCO     | -300.5 | -298.0 | -298.0 | -302.5 | -304.0 | -307.2 |
| HOF      | -231.1 | -230.7 | -232.2 | -231.5 | -236.7 | -235.4 |
| LiF      | -194.7 | -193.4 | -194.5 | -195.6 | -192.6 | -195.5 |
| LiH      | -130.8 | -129.6 | -127.0 | -132.5 | -126.4 | -127.2 |
| N$_2$    | -202.0 | -197.2 | -197.3 | -203.2 | -201.6 | -205.2 |
| N$_2$O   | -333.8 | -332.6 | -334.1 | -333.0 | -336.7 | -339.1 |
| NH$_3$   | -291.2 | -287.9 | -288.4 | -287.0 | -289.3 | -290.3 |
| O$_3$    | 238.7  | 239.4  | 264.0  | 99.3   | 336.6   | 121.5   |
| OCS      | -579.6 | -577.2 | -578.8 | -577.6 | -585.6 | -584.1 |
| OF$_2$   | -234.1 | -235.2 | -238.2 | -240.0 | -250.2 | -247.1 |
| PN       | -292.2 | -285.4 | -284.3 | -302.0 | -295.5 | -308.2 |
| SO$_2$   | -296.1 | -289.5 | -290.9 | -301.3 | -296.6 | -314.3 |
Table 6: Magnetizabilities in units of $10^{-30}$ J/T$^2$ for the BHLYP, BLYP, BP86, and CAM-B3LYP functionals in the aug-cc-pCVQZ basis set from calculations with TURBOMOLE and GIMIC compared to CCSD(T) data from ref. 18.

| Molecule | BHLYP   | BLYP | BP86   | CAM-B3LYP | CCSD(T) |
|----------|---------|------|--------|-----------|---------|
| AlF      | -397.1  | -399.2 | -394.3 | -397.0    | -394.5  |
| C$_2$H$_4$ | -343.5  | -333.4 | -331.0 | -339.4    | -345.6  |
| C$_3$H$_4$ | -473.1  | -458.4 | -460.1 | -468.1    | -478.9  |
| CH$_2$O  | -114.1  | -109.3 | -108.1 | -115.3    | -127.4  |
| CH$_3$F  | -319.0  | -309.5 | -311.4 | -314.6    | -315.7  |
| CH$_4$   | -325.2  | -318.1 | -318.5 | -320.0    | -316.9  |
| CO       | -205.9  | -209.1 | -205.2 | -208.4    | -209.5  |
| FCCCH    | -445.4  | -438.5 | -437.9 | -441.8    | -441.6  |
| FCN      | -371.6  | -366.4 | -365.0 | -369.5    | -370.0  |
| H$_2$C$_2$O | -431.0  | -420.7 | -422.0 | -424.8    | -423.9  |
| H$_2$O   | -236.6  | -239.4 | -237.5 | -237.5    | -235.1  |
| H$_2$S   | -462.6  | -457.0 | -456.6 | -456.7    | -455.1  |
| H$_4$C$_2$O | -542.1  | -520.4 | -523.5 | -531.3    | -535.2  |
| HCN      | -272.9  | -268.7 | -264.4 | -272.0    | -271.8  |
| HCP      | -492.9  | -485.6 | -479.2 | -488.6    | -492.8  |
| HF       | -177.0  | -181.0 | -179.3 | -179.0    | -176.4  |
| HFCO     | -303.8  | -299.4 | -296.5 | -302.9    | -307.2  |
| HOF      | -238.8  | -226.7 | -227.8 | -233.4    | -235.4  |
| LiF      | -193.1  | -196.3 | -197.0 | -195.6    | -195.5  |
| LiH      | -129.4  | -136.5 | -133.2 | -129.3    | -127.2  |
| N$_2$    | -202.4  | -203.7 | -199.6 | -204.3    | -205.2  |
| N$_2$O   | -338.8  | -332.0 | -332.6 | -336.0    | -339.1  |
| NH$_3$   | -294.3  | -293.4 | -291.9 | -292.7    | -290.3  |
| O$_3$    | 356.9   | 180.1  | 180.9  | 258.1     | 121.5   |
| OCS      | -588.0  | -576.1 | -575.0 | -583.4    | -584.1  |
| OF$_2$   | -251.5  | -220.6 | -222.1 | -239.8    | -247.1  |
| PN       | -293.9  | -292.4 | -284.7 | -297.4    | -308.2  |
| SO$_2$   | -297.0  | -298.4 | -292.7 | -300.7    | -314.3  |
Table 7: Magnetizabilities in units of $10^{-30} \text{J/T}^2$ for the CAMh-B3LYP, CAM-QTP-00, and CAM-QTP-01 functionals in the aug-cc-pCVQZ basis set from calculations with TURBOMOLE and GIMIC compared to CCSD(T) data from ref. 18.

| Molecule | CAMh-B3LYP | CAM-QTP-00 | CAM-QTP-01 | CCSD(T) |
|----------|------------|------------|------------|---------|
| AlF      | -396.9     | -394.5     | -397.2     | -394.5  |
| C$_2$H$_4$ | -338.5    | -344.7     | -341.6     | -345.6  |
| C$_3$H$_4$ | -466.2     | -472.4     | -471.9     | -478.9  |
| CH$_2$O  | -115.6     | -124.5     | -115.3     | -127.4  |
| CH$_3$F  | -313.6     | -316.7     | -316.6     | -315.7  |
| CH$_4$   | -318.6     | -317.5     | -322.7     | -316.9  |
| CO       | -207.9     | -205.2     | -209.4     | -209.5  |
| FCCH     | -441.2     | -444.8     | -443.5     | -441.6  |
| FCN      | -368.8     | -371.8     | -371.2     | -370.0  |
| H$_2$C$_2$O | -423.6   | -427.0     | -427.4     | -423.9  |
| H$_2$O   | -237.2     | -234.0     | -238.0     | -235.1  |
| H$_2$S   | -455.8     | -454.0     | -458.5     | -455.1  |
| H$_4$C$_2$O | -529.5   | -538.2     | -535.6     | -535.2  |
| HCN      | -271.2     | -273.9     | -273.9     | -271.8  |
| HCP      | -488.4     | -494.5     | -489.8     | -492.8  |
| HF       | -178.8     | -175.5     | -179.1     | -176.4  |
| HFCO     | -302.2     | -305.3     | -304.5     | -307.2  |
| HOF      | -232.5     | -238.7     | -235.6     | -235.4  |
| LiF      | -195.4     | -192.6     | -195.8     | -195.5  |
| LiH      | -129.8     | -124.9     | -128.6     | -127.2  |
| N$_2$    | -203.6     | -202.3     | -205.6     | -205.2  |
| N$_2$O   | -335.2     | -338.2     | -337.7     | -339.1  |
| NH$_3$   | -292.0     | -289.8     | -293.9     | -290.3  |
| O$_3$    | 250.1      | 373.1      | 283.2      | 121.5   |
| OCS      | -582.1     | -588.0     | -586.4     | -584.1  |
| OF$_2$   | -237.7     | -255.0     | -244.9     | -247.1  |
| PN       | -295.9     | -297.6     | -300.3     | -308.2  |
| SO$_2$   | -298.8     | -298.6     | -303.1     | -314.3  |
Table 8: Magnetizabilities in units of $10^{-30}$ J/T$^2$ for the CAM-QTP-02, CHACHIYO, HF, KT1, KT2, and KT3 functionals in the aug-cc-pCVQZ basis set from calculations with TURBOMOLE and GIMIC compared to CCSD(T) data from ref. 18.

| Molecule     | CAM-QTP-02 | CHACHIYO | HF   | KT1  | KT2  | KT3  | CCSD(T) |
|--------------|------------|----------|------|------|------|------|---------|
| AlF          | -397.4     | -392.2   | -399.2 | -398.4 | -392.4 | -394.2 | -394.5 |
| C$_2$H$_4$   | -343.0     | -329.0   | -354.8 | -338.6 | -335.2 | -332.4 | -345.6 |
| C$_3$H$_4$   | -473.3     | -458.7   | -478.1 | -461.5 | -457.3 | -453.2 | -478.9 |
| CH$_2$O      | -116.5     | -109.3   | -139.4 | -116.8 | -118.0 | -117.9 | -127.4 |
| CH$_3$F      | -317.4     | -310.8   | -317.9 | -309.8 | -307.4 | -305.3 | -315.7 |
| CH$_4$       | -323.3     | -315.5   | -313.6 | -320.7 | -316.0 | -311.8 | -316.9 |
| CO           | -209.3     | -202.6   | -204.5 | -214.0 | -209.1 | -206.1 | -209.5 |
| FCCH         | -444.5     | -436.3   | -452.2 | -445.0 | -440.2 | -437.0 | -441.6 |
| FCN          | -372.1     | -363.4   | -378.0 | -372.4 | -367.6 | -365.1 | -370.0 |
| H$_2$C$_2$O  | -428.6     | -419.6   | -432.6 | -428.1 | -422.1 | -416.9 | -423.9 |
| H$_2$O       | -237.8     | -235.9   | -231.2 | -238.8 | -235.0 | -233.8 | -235.1 |
| H$_2$S       | -459.1     | -453.4   | -452.6 | -462.1 | -455.7 | -450.8 | -455.1 |
| H$_4$C$_2$O  | -537.8     | -522.2   | -544.9 | -527.0 | -521.3 | -516.5 | -535.2 |
| HCN          | -274.8     | -261.7   | -280.1 | -274.8 | -270.5 | -267.1 | -271.8 |
| HCP          | -491.2     | -475.5   | -511.6 | -493.6 | -487.9 | -483.9 | -492.8 |
| HF           | -178.8     | -178.4   | -172.7 | -179.8 | -176.8 | -176.5 | -176.4 |
| HFCO         | -305.5     | -294.7   | -311.5 | -303.3 | -299.0 | -297.5 | -307.2 |
| HOF          | -237.1     | -227.2   | -244.6 | -231.4 | -227.6 | -224.9 | -235.4 |
| LiF          | -195.5     | -196.1   | -190.7 | -199.1 | -196.1 | -193.8 | -195.5 |
| LiH          | -128.0     | -131.7   | -125.3 | -139.1 | -137.1 | -138.0 | -127.2 |
| N$_2$        | -205.9     | -197.0   | -202.9 | -209.8 | -205.0 | -201.2 | -205.2 |
| N$_2$O       | -338.6     | -331.6   | -342.8 | -334.6 | -330.5 | -328.0 | -339.1 |
| NH$_3$       | -294.1     | -289.6   | -287.4 | -293.9 | -289.6 | -287.0 | -290.3 |
| O$_3$        | 303.8      | 183.6    | 578.9  | 131.9  | 138.6  | 149.2  | 121.5  |
| OCS          | -587.9     | -573.1   | -597.5 | -582.1 | -575.6 | -571.6 | -584.1 |
| OF$_2$       | -248.4     | -222.1   | -271.8 | -231.7 | -226.4 | -223.3 | -247.1 |
| PN           | -300.9     | -279.9   | -304.2 | -302.1 | -297.0 | -291.1 | -308.2 |
| SO$_2$       | -303.7     | -288.9   | 0.0    | -304.6 | -297.1 | -292.6 | -314.3 |
Table 9: Magnetizabilities in units of $10^{-30} \text{J/T}^2$ for the LDA, M06, M06-2X, M06-L, M08-HX, and M08-SO functionals in the aug-cc-pCVQZ basis set from calculations with Turbomole and Gimic compared to CCSD(T) data from ref. 18.

| Molecule | LDA    | M06   | M06-2X | M06-L  | M08-HX | M08-SO | CCSD(T) |
|----------|--------|-------|--------|--------|--------|--------|---------|
| AlF      | -395.8 | -387.1| -392.9 | -382.7 | -397.2 | -395.8 | -394.5  |
| C$_2$H$_4$ | -331.1 | -332.4| -329.9 | -327.1 | -331.2 | -334.3 | -345.6  |
| C$_3$H$_4$ | -464.4 | -465.3| -461.4 | -461.5 | -462.7 | -463.7 | -478.9  |
| CH$_2$O  | -95.9  | 104.5 | -94.1  | -123.6 | -94.1  | -89.4  | -127.4  |
| CH$_3$F  | -315.4 | -313.4| -317.2 | -311.1 | -319.5 | -319.0 | -315.7  |
| CH$_4$   | -329.4 | -316.3| -319.9 | -309.4 | -322.6 | -323.0 | -316.9  |
| CO       | -206.6 | -192.3| -193.9 | -195.3 | -201.7 | -195.2 | -209.5  |
| FCCH     | -438.6 | -434.5| -439.7 | -434.0 | -442.4 | -441.0 | -441.6  |
| FCN      | -365.3 | -358.7| -364.6 | -359.9 | -368.3 | -365.2 | -370.0  |
| H$_2$C$_2$O | -427.7 | -417.3| -418.1 | -418.8 | -423.1 | -419.3 | -423.9  |
| H$_2$O   | -240.9 | -233.1| -235.6 | -230.2 | -235.8 | -236.7 | -235.1  |
| H$_2$S   | -466.0 | -451.7| -457.7 | -447.1 | -457.1 | -459.1 | -455.1  |
| H$_3$C$_2$O | -529.8 | -529.4| -540.3 | -521.1 | -543.4 | -543.5 | -535.2  |
| HCN      | -265.1 | -252.4| -260.4 | -252.6 | -265.2 | -262.4 | -271.8  |
| HCP      | -477.5 | -465.0| -476.4 | -462.9 | -483.3 | -480.5 | -492.8  |
| HF       | -181.1 | -175.1| -176.6 | -173.8 | -177.0 | -177.5 | -176.4  |
| HFCO     | -296.9 | -292.1| -292.9 | -292.4 | -295.7 | -293.5 | -307.2  |
| HOF      | -229.0 | -228.3| -235.1 | -230.0 | -236.4 | -235.3 | -235.4  |
| LiF      | -196.3 | -190.4| -193.4 | -191.8 | -193.2 | -193.0 | -195.5  |
| LiH      | -136.0 | -130.3| -128.1 | -127.7 | -129.1 | -129.6 | -127.2  |
| N$_2$    | -201.1 | -181.2| -189.1 | -186.5 | -195.7 | -189.7 | -205.2  |
| N$_2$O   | -334.3 | -326.3| -332.8 | -329.0 | -335.7 | -331.7 | -339.1  |
| NH$_3$   | -298.1 | -288.2| -291.9 | -283.0 | -292.0 | -293.4 | -290.3  |
| O$_3$    | -195.2 | -163.4| 492.9  | 156.2  | 348.2  | 647.4  | 121.5   |
| OCS      | -576.6 | -570.7| -578.3 | -569.4 | -583.5 | -580.2 | -584.1  |
| OF$_2$   | -220.3 | -228.5| -242.9 | -234.3 | -247.0 | -241.6 | -247.1  |
| PN       | -284.6 | -249.4| -259.9 | -267.0 | -283.7 | -259.9 | -308.2  |
| SO$_2$   | -295.1 | -276.3| -277.4 | -285.4 | -287.7 | -271.8 | -314.3  |
Table 10: Magnetizabilities in units of $10^{-30} \text{J}/\text{T}^2$ for the M11, M11-L, MN12-L, MN12-SX, and MN15 functionals in the aug-cc-pCVQZ basis set from calculations with Turbomole and Gimic compared to CCSD(T) data from ref. 18.

| Molecule | M11  | M11-L | MN12-L | MN12-SX | MN15  | CCSD(T) |
|----------|------|-------|--------|---------|-------|---------|
| AlF      | -391.5 | -403.1 | -407.1 | -403.4 | -400.2 | -394.5  |
| C$_2$H$_4$ | -331.3 | -334.1 | -340.4 | -338.4 | -330.4 | -346.6  |
| C$_3$H$_4$ | -466.6 | -461.3 | -471.1 | -467.1 | -460.0 | -478.9  |
| CH$_2$O  | -89.0  | -135.4 | -145.0 | -128.4 | -91.0  | -127.4  |
| CH$_3$F  | -320.7 | -307.8 | -313.8 | -315.5 | -314.9 | -315.7  |
| CH$_4$   | -323.7 | -312.0 | -315.6 | -317.7 | -319.4 | -316.9  |
| CO       | -199.2 | -203.4 | -211.4 | -207.8 | -194.3 | -209.5  |
| FCCH     | -440.0 | -444.9 | -446.6 | -445.2 | -437.2 | -441.6  |
| FCN      | -365.7 | -368.1 | -371.7 | -370.9 | -361.1 | -370.0  |
| H$_2$C$_2$O | -424.3 | -431.4 | -434.6 | -428.8 | -416.3 | -423.9  |
| H$_2$O   | -236.4 | -227.8 | -230.5 | -233.4 | -235.1 | -235.1  |
| H$_2$S   | -459.1 | -450.6 | -454.1 | -454.8 | -454.8 | -455.1  |
| H$_4$C$_2$O | -545.2 | -518.5 | -525.5 | -532.5 | -531.5 | -535.2  |
| HCN      | -262.7 | -263.9 | -273.5 | -271.6 | -259.8 | -271.8  |
| HCP      | -471.9 | -489.0 | -501.5 | -494.9 | -481.5 | -492.8  |
| HF       | -177.9 | -169.8 | -173.5 | -175.2 | -176.8 | -176.4  |
| HFCO     | -293.7 | -300.3 | -305.4 | -303.3 | -289.1 | -307.2  |
| HOF      | -234.8 | -228.6 | -234.8 | -237.4 | -230.1 | -235.4  |
| LiF      | -194.0 | -188.3 | -191.3 | -190.8 | -195.5 | -195.5  |
| LiH      | -126.4 | -150.0 | -145.0 | -140.5 | -131.5 | -127.2  |
| N$_2$    | -193.7 | -193.7 | -204.3 | -201.3 | -186.9 | -205.2  |
| N$_2$O   | -334.5 | -330.7 | -336.1 | -336.1 | -328.4 | -339.1  |
| NH$_3$   | -292.7 | -284.3 | -286.3 | -289.5 | -291.0 | -290.3  |
| O$_3$    | 484.4  | 84.9  | 45.6   | 131.1  | 571.0  | 121.5   |
| OCS      | -582.9 | -579.9 | -587.3 | -586.0 | -574.3 | -584.1  |
| OF$_2$   | -241.3 | -242.3 | -249.8 | -253.4 | -230.3 | -247.1  |
| PN       | -266.2 | -293.5 | -330.2 | -311.0 | -271.6 | -308.2  |
| SO$_2$   | -273.4 | -290.9 | -312.3 | -303.3 | -268.9 | -314.3  |
Table 11: Magnetizabilities in units of $10^{-30} \text{J/T}^2$ for the MN15-L, MVS, N12, N12-SX, PBE, and PBE0 functionals in the aug-cc-pCVQZ basis set from calculations with Turbomole and Gimic compared to CCSD(T) data from ref. 18.

| Molecule | MN15-L | MVS   | N12   | N12-SX | PBE   | PBE0  | CCSD(T) |
|----------|--------|-------|-------|--------|-------|-------|---------|
| AlF      | -410.2 | -384.3| -394.1| -394.7 | -396.9| -394.3| -394.5  |
| C$_2$H$_4$ | -343.2 | -319.8| -331.1| -333.8 | -330.8| -335.3| -345.6  |
| C$_3$H$_4$ | -470.4 | -459.6| -456.3| -463.8 | -459.5| -465.1| -478.9  |
| CH$_2$O  | -142.4 | -123.5| -112.9| -113.3 | -104.9| -112.8| -127.4  |
| CH$_3$F  | -314.1 | -309.1| -307.8| -313.8 | -311.2| -314.4| -315.7  |
| CH$_4$   | -318.8 | -315.1| -315.6| -316.5 | -320.3| -318.4| -316.9  |
| CO       | -215.3 | -198.0| -205.2| -201.1 | -205.6| -202.9| -209.5  |
| FCCH     | -450.2 | -436.9| -433.4| -436.2 | -437.6| -439.9| -441.6  |
| FCN      | -376.7 | -365.7| -360.8| -362.6 | -365.0| -366.6| -370.0  |
| H$_2$C$_2$O | -438.1 | -428.8| -416.6| -419.6 | -421.8| -423.6| -429.3  |
| H$_2$O   | -235.8 | -232.3| -234.3| -233.7 | -238.5| -235.2| -235.1  |
| H$_2$S   | -459.0 | -456.0| -451.4| -453.9 | -458.6| -456.2| -455.1  |
| H$_4$C$_2$O | -530.1 | -519.8| -516.6| -528.2 | -523.9| -531.5| -535.2  |
| HCN      | -277.0 | -256.3| -265.3| -264.0 | -264.4| -266.0| -271.8  |
| HCP      | -508.5 | -467.7| -477.2| -480.1 | -479.3| -482.7| -492.8  |
| HF       | -176.9 | -174.8| -176.2| -176.0 | -180.1| -177.1| -176.4  |
| HFCO     | -309.6 | -297.4| -294.8| -295.6 | -296.8| -298.7| -307.2  |
| HOF      | -239.5 | -226.7| -222.6| -230.0 | -227.4| -232.8| -235.4  |
| LiF      | -197.3 | -194.5| -191.7| -193.3 | -196.2| -194.1| -195.5  |
| LiH      | -137.9 | -125.5| -139.1| -128.9 | -135.2| -129.1| -127.2  |
| N$_2$    | -208.7 | -191.9| -199.8| -195.5 | -199.8| -198.2| -205.2  |
| N$_2$O   | -340.1 | -332.9| -326.5| -329.9 | -331.8| -334.2| -339.1  |
| NH$_3$   | -291.9 | -287.4| -288.9| -288.6 | -293.1| -290.4| -290.3  |
| O$_3$    | 63.6   | 136.6 | 185.3 | 292.9  | 183.4 | 257.6 | 121.5   |
| OCS      | -590.7 | -576.0| -568.4| -576.7 | -574.8| -579.6| -584.1  |
| OF$_2$   | -255.0 | -228.6| -217.6| -233.7 | -221.6| -238.3| -247.1  |
| PN       | -330.5 | -277.7| -285.9| -283.2 | -284.3| -285.0| -308.2  |
| SO$_2$   | -318.4 | -289.8| -283.4| -284.2 | -293.8| -291.5| -314.3  |
Table 12: Magnetizabilities in units of $10^{-30}$ J/T$^2$ for the QTP-17, revB3LYP, revM06, and revM06-L functionals in the aug-cc-pCVQZ basis set from calculations with TURBOMOLE and GIMIC compared to CCSD(T) data from ref. 18.

| Molecule    | QTP-17 | revB3LYP | revM06 | revM06-L | CCSD(T) |
|-------------|--------|----------|--------|----------|---------|
| AlF         | -397.4 | -396.9   | -389.7 | -385.9   | -394.5  |
| C$_2$H$_4$  | -338.5 | -337.0   | -329.6 | -326.6   | -345.6  |
| C$_3$H$_4$  | -464.1 | -463.6   | -462.6 | -461.2   | -478.9  |
| CH$_2$O     | -116.8 | -113.8   | -104.4 | -134.0   | -127.4  |
| CH$_3$F     | -312.7 | -312.8   | -315.2 | -310.5   | -315.7  |
| CH$_4$      | -316.9 | -318.3   | -318.4 | -312.0   | -316.9  |
| CO          | -207.1 | -206.8   | -197.2 | -206.4   | -209.5  |
| FCCH        | -441.1 | -440.4   | -438.2 | -437.8   | -441.6  |
| FCN         | -368.4 | -367.7   | -364.2 | -366.5   | -370.0  |
| H$_2$C$_2$O | -422.8 | -422.9   | -421.1 | -427.7   | -423.9  |
| H$_2$O      | -236.6 | -237.1   | -234.7 | -230.8   | -235.1  |
| H$_2$S      | -455.3 | -456.3   | -456.3 | -446.9   | -455.1  |
| H$_4$C$_2$O | -528.2 | -527.8   | -533.4 | -517.3   | -535.2  |
| HCN         | -270.8 | -269.6   | -261.3 | -264.7   | -271.8  |
| HCP         | -490.0 | -487.5   | -476.1 | -478.4   | -492.8  |
| HF          | -178.2 | -178.6   | -176.2 | -174.4   | -176.4  |
| HFCO        | -301.7 | -300.7   | -294.4 | -298.9   | -307.2  |
| HOF         | -232.2 | -231.4   | -232.7 | -231.8   | -235.4  |
| LiF         | -194.6 | -194.8   | -194.0 | -195.1   | -195.5  |
| LiH         | -130.9 | -131.3   | -127.8 | -123.9   | -127.2  |
| N$_2$       | -202.7 | -202.2   | -191.7 | -200.5   | -205.2  |
| N$_2$O      | -334.4 | -334.1   | -333.2 | -334.6   | -339.1  |
| NH$_3$      | -291.2 | -292.0   | -290.6 | -284.5   | -290.3  |
| O$_3$       | 251.4  | 239.5    | 395.7  | 93.9     | 121.5   |
| OCS         | -581.2 | -580.0   | -578.1 | -577.1   | -584.1  |
| OF$_2$      | -237.2 | -234.2   | -238.1 | -241.0   | -247.1  |
| PN          | -294.2 | -292.3   | -269.8 | -305.2   | -308.2  |
| SO$_2$      | -297.1 | -296.4   | -280.9 | -301.8   | -314.3  |
Table 13: Magnetizabilities in units of $10^{-30}$J/T$^2$ for the revM11, revTPSS, revTPSSh, rSCAN, and SCAN functionals in the aug-cc-pCVQZ basis set from calculations with TURBOMOLE and GIMIC compared to CCSD(T) data from ref. 18.

| Molecule | revM11 | revTPSS | revTPSSh | rSCAN | SCAN | CCSD(T) |
|----------|--------|---------|----------|-------|------|---------|
| AlF      | -393.5 | -393.2  | -392.7   | -395.0| -392.0| -394.5  |
| C$_2$H$_4$ | -333.0 | -332.0  | -333.7   | -332.3| -333.0| -345.6  |
| C$_3$H$_4$ | -467.4 | -457.7  | -460.2   | -463.4| -462.4| -478.9  |
| CH$_2$O  | -99.7  | -123.2  | -124.6   | -121.9| -126.4| -127.4  |
| CH$_3$F  | -316.8 | -308.9  | -310.3   | -311.8| -310.4| -315.7  |
| CH$_4$   | -320.1 | -308.8  | -309.3   | -318.5| -314.4| -316.9  |
| CO       | -201.9 | -204.9  | -204.1   | -206.7| -206.9| -209.5  |
| FCCH     | -437.8 | -436.7  | -437.8   | -439.2| -438.3| -441.6  |
| FCN      | -365.4 | -365.2  | -365.9   | -366.6| -365.8| -370.0  |
| H$_2$C$_2$O | -418.7 | -416.3  | -417.7   | -427.3| -426.0| -423.9  |
| H$_2$O   | -238.1 | -235.6  | -234.6   | -235.0| -234.2| -235.1  |
| H$_2$S   | -456.2 | -448.5  | -448.7   | -457.5| -453.9| -455.1  |
| H$_4$C$_2$O | -538.2 | -520.7  | -523.9   | -524.6| -519.8| -535.2  |
| HCN      | -264.1 | -264.8  | -265.5   | -266.4| -265.3| -271.8  |
| HCP      | -473.1 | -481.1  | -482.5   | -482.5| -481.7| -492.8  |
| HF       | -179.9 | -178.5  | -177.5   | -176.8| -176.5| -176.4  |
| HFCO     | -298.1 | -298.0  | -298.9   | -298.8| -299.3| -307.2  |
| HOF      | -233.8 | -230.3  | -232.2   | -230.1| -230.9| -235.4  |
| LiF      | -196.9 | -196.2  | -195.3   | -195.9| -195.6| -195.5  |
| LiH      | -123.4 | -128.5  | -127.2   | -130.9| -130.4| -127.2  |
| N$_2$    | -195.5 | -199.0  | -198.7   | -201.4| -199.8| -205.2  |
| N$_2$O   | -334.0 | -331.3  | -332.3   | -333.6| -332.6| -339.1  |
| NH$_3$   | -293.0 | -287.5  | -287.1   | -290.3| -288.7| -290.3  |
| O$_3$    | 445.5  | 142.5   | 167.9    | 138.7 | 143.4 | 121.5   |
| OCS      | -580.8 | -573.6  | -575.8   | -578.0| -577.7| -584.1  |
| OF$_2$   | -239.0 | -231.2  | -236.8   | -232.4| -234.3| -247.1  |
| PN       | -274.0 | -288.1  | -288.6   | -294.8| -292.3| -308.2  |
| SO$_2$   | -287.3 | -291.7  | -291.5   | -297.5| -296.4| -314.3  |
Table 14: Magnetizabilities in units of $10^{-30}$J/T$^2$ for the TASK, TPSS, TPSSh, $\omega$B97, $\omega$B97M-V, and $\omega$B97X functionals in the aug-cc-pCVQZ basis set from calculations with TURBOMOLE and GIMIC compared to CCSD(T) data from ref. 18.

| Molecule | TASK     | TPSS     | TPSSh    | $\omega$B97 | $\omega$B97M-V | $\omega$B97X | CCSD(T) |
|----------|----------|----------|----------|-------------|----------------|--------------|---------|
| AlF      | -383.3   | -393.9   | -393.3   | -397.0      | -400.1         | -395.5       | -394.5  |
| C$_2$H$_4$ | -325.4   | -332.1   | -333.8   | -338.1      | -340.5         | -337.5       | -345.6  |
| C$_3$H$_4$ | -460.8   | -458.5   | -460.9   | -471.5      | -469.9         | -469.6       | -478.9  |
| CH$_2$O  | -132.9   | -120.0   | -121.7   | -118.9      | -115.8         | -116.4       | -127.4  |
| CH$_3$F  | -309.3   | -309.5   | -310.9   | -315.3      | -315.0         | -314.9       | -315.7  |
| CH$_4$   | -310.7   | -311.6   | -311.8   | -318.7      | -321.0         | -317.9       | -316.9  |
| CO       | -204.5   | -204.7   | -203.9   | -208.2      | -210.5         | -205.8       | -209.5  |
| FCCH     | -439.1   | -436.7   | -437.8   | -442.1      | -444.6         | -441.2       | -441.6  |
| FCN      | -367.8   | -364.8   | -365.5   | -370.3      | -371.8         | -368.5       | -370.0  |
| H$_2$C$_2$O | -431.3   | -417.9   | -419.1   | -427.0      | -427.2         | -424.6       | -423.9  |
| H$_2$O   | -231.2   | -235.7   | -234.7   | -235.9      | -237.3         | -235.1       | -235.1  |
| H$_2$S   | -454.3   | -450.6   | -450.5   | -454.2      | -458.2         | -454.3       | -455.1  |
| H$_4$C$_2$O | -519.1   | -521.3   | -524.5   | -533.9      | -533.9         | -532.5       | -535.2  |
| HCN      | -262.1   | -264.4   | -265.1   | -269.5      | -274.4         | -268.6       | -271.8  |
| HCP      | -478.2   | -480.5   | -481.9   | -481.9      | -491.0         | -483.8       | -492.8  |
| HF       | -173.7   | -178.4   | -177.3   | -178.4      | -178.7         | -177.2       | -176.4  |
| HFCO     | -298.6   | -297.8   | -298.6   | -303.9      | -304.0         | -301.0       | -307.2  |
| HOF      | -231.1   | -229.3   | -231.3   | -234.5      | -235.3         | -233.5       | -235.4  |
| LiF      | -195.1   | -194.6   | -194.6   | -197.9      | -196.3         | -197.2       | -195.5  |
| LiH      | -120.4   | -129.5   | -128.0   | -126.4      | -133.9         | -129.3       | -127.2  |
| N$_2$    | -197.7   | -198.8   | -198.4   | -202.4      | -207.1         | -200.6       | -205.2  |
| N$_2$O   | -335.7   | -330.8   | -331.8   | -337.7      | -337.5         | -336.0       | -339.1  |
| NH$_3$   | -286.1   | -288.4   | -287.8   | -290.3      | -292.8         | -289.9       | -290.3  |
| O$_3$    | 142.9    | 151.0    | 176.9    | 236.9       | 225.4          | 260.5        | 121.5   |
| OCS      | -580.8   | -573.6   | -575.8   | -585.6      | -586.3         | -583.2       | -584.1  |
| OF$_2$   | -236.8   | -228.9   | -234.8   | -243.4      | -245.4         | -241.1       | -247.1  |
| PN       | -289.1   | -287.3   | -287.7   | -294.6      | -300.7         | -292.2       | -308.2  |
| SO$_2$   | -291.5   | -291.9   | -291.6   | -300.8      | -303.8         | -296.5       | -314.3  |
Table 15: Magnetizabilities in units of $10^{-30} \text{J/T}^2$ for the $\omega$B97X-D, and $\omega$B97X-V functionals in the aug-cc-pCVQZ basis set from calculations with TURBOMOLE and GIMIC compared to CCSD(T) data from ref. 18.

| Molecule | $\omega$B97X-D | $\omega$B97X-V | CCSD(T) |
|----------|----------------|----------------|----------|
| AlF      | -393.1         | -396.5         | -394.5   |
| C$_2$H$_4$ | -335.5         | -338.9         | -345.6   |
| C$_3$H$_4$ | -466.9         | -469.9         | -478.9   |
| CH$_2$O  | -114.7         | -118.6         | -127.4   |
| CH$_3$F  | -314.1         | -314.9         | -315.7   |
| CH$_4$   | -316.1         | -317.7         | -316.9   |
| CO       | -202.4         | -207.4         | -209.5   |
| FCCH     | -439.8         | -442.4         | -441.6   |
| FCN      | -366.2         | -370.1         | -370.0   |
| H$_2$C$_2$O | -422.1         | -425.3         | -423.9   |
| H$_2$O    | -233.8         | -236.0         | -235.1   |
| H$_2$S    | -452.9         | -454.2         | -455.1   |
| H$_4$C$_2$O | -530.1         | -532.8         | -535.2   |
| HCN      | -266.2         | -270.7         | -271.8   |
| HCP      | -482.7         | -486.5         | -492.8   |
| HF       | -175.9         | -178.1         | -176.4   |
| HFCO     | -297.9         | -303.1         | -307.2   |
| HOF      | -232.0         | -234.7         | -235.4   |
| LiF      | -195.6         | -196.4         | -195.5   |
| LiH      | -131.5         | -127.4         | -127.2   |
| N$_2$    | -197.7         | -202.9         | -205.2   |
| N$_2$O   | -334.3         | -336.8         | -339.1   |
| NH$_3$   | -288.7         | -290.5         | -290.3   |
| O$_3$    | 267.2          | 251.2          | 121.5    |
| OCS      | -579.6         | -584.7         | -584.1   |
| OF$_2$   | -238.0         | -243.8         | -247.1   |
| PN       | -286.5         | -296.0         | -308.2   |
| SO$_2$   | -290.6         | -299.8         | -314.3   |
Table 16: Deviations of the magnetizabilities computed with TURBOMOLE and GIMIC for the B3LYP, B97-2, B97-3, B97M-V, and BHandHLYP functionals in the aug-cc-pCVQZ basis set from CCSD(T) data from ref. 18 in units of $10^{-30}\text{J}/\text{T}^2$.

| Molecule | B3LYP | B97-2 | B97-3 | B97M-V | BHandHLYP |
|----------|-------|-------|-------|--------|-----------|
| AlF      | -2.0  | 1.1   | 1.9   | -1.6   | -1.1      |
| C$_2$H$_4$ | 8.9   | 11.3  | 11.0  | 10.8   | 2.6       |
| C$_3$H$_4$ | 15.8  | 16.3  | 14.8  | 18.3   | 10.1      |
| CH$_2$O  | 12.5  | 10.8  | 12.0  | -5.4   | 3.6       |
| CH$_3$F  | 3.3   | 3.5   | 2.4   | 6.3    | 0.8       |
| CH$_4$   | -0.1  | 2.3   | 2.3   | 3.7    | 1.2       |
| CO       | 2.9   | 7.0   | 8.3   | 1.0    | 4.5       |
| FCCH     | 1.5   | 3.3   | 2.4   | 1.1    | -2.0      |
| FCN      | 2.6   | 4.6   | 4.1   | 1.7    | -0.5      |
| H$_2$C$_2$O | 1.8 | 2.7   | 2.9   | -2.0   | -1.1      |
| H$_2$O   | -1.6  | 1.6   | 1.2   | 1.7    | 1.1       |
| H$_2$S   | -0.0  | 3.1   | 2.7   | 2.8    | 1.6       |
| H$_4$C$_2$O | 8.3  | 7.9   | 6.2   | 15.5   | 0.7       |
| HCN      | 2.4   | 6.8   | 6.4   | 2.9    | -0.9      |
| HCP      | 5.4   | 10.9  | 9.9   | 6.9    | -1.5      |
| HF       | -2.0  | 0.5   | 0.2   | 0.3    | 0.6       |
| HFCO     | 6.7   | 9.2   | 9.2   | 4.7    | 3.2       |
| HOF      | 4.3   | 4.7   | 3.2   | 3.9    | -1.3      |
| LiF      | 0.8   | 2.1   | 1.0   | -0.1   | 2.9       |
| LiH      | -3.6  | -2.4  | 0.2   | -5.3   | 0.8       |
| N$_2$    | 3.2   | 8.0   | 7.9   | 2.0    | 3.6       |
| N$_2$O   | 5.3   | 6.5   | 5.0   | 6.1    | 2.4       |
| NH$_3$   | -0.9  | 2.4   | 1.9   | 3.3    | 1.0       |
| O$_3$    | 117.2 | 117.9 | 142.5 | -22.2  | 215.1     |
| OCS      | 4.5   | 6.9   | 5.3   | 6.5    | -1.5      |
| OF$_2$   | 13.0  | 11.9  | 8.9   | 7.1    | -3.1      |
| PN       | 16.0  | 22.8  | 23.9  | 6.2    | 12.7      |
| SO$_2$   | 18.2  | 24.8  | 23.4  | 13.0   | 17.7      |

* Statistics for the mean absolute error (MAE), mean error (ME) and the standard deviation (STD) exclude O$_3$.
Table 17: Deviations of the magnetizabilities computed with TURBOMOLE and GIMIC for the BHLYP, BLYP, BP86, and CAM-B3LYP functionals in the aug-cc-pCVQZ basis set from CCSD(T) data from ref. 18 in units of $10^{-30}$ J/T$^2$.

| Molecule  | BHLYP  | BLYP  | BP86  | CAM-B3LYP |
|-----------|--------|-------|-------|-----------|
| AlF       | -2.6   | -4.7  | 0.2   | -2.5      |
| C$_2$H$_4$| 2.1    | 12.2  | 14.6  | 6.2       |
| C$_3$H$_4$| 5.8    | 20.5  | 18.8  | 10.8      |
| CH$_2$O   | 13.3   | 18.1  | 19.3  | 12.1      |
| CH$_3$F   | -3.3   | 6.2   | 4.3   | 1.1       |
| CH$_4$    | -8.3   | -1.2  | -1.6  | -3.1      |
| CO        | 3.6    | 0.4   | 4.3   | 1.1       |
| FCCH      | -3.8   | 3.1   | 3.7   | -0.2      |
| FCN       | -1.6   | 3.6   | 5.0   | 0.5       |
| H$_2$C$_2$O| -7.1  | 3.2   | 1.9   | -0.9      |
| H$_2$O    | -1.5   | -4.3  | -2.4  | -2.4      |
| H$_2$S    | -7.5   | -1.9  | -1.5  | -1.6      |
| H$_3$C$_2$O| -6.9  | 14.8  | 11.7  | 3.9       |
| HCN       | -1.1   | 3.1   | 7.4   | -0.2      |
| HCP       | -0.1   | 7.2   | 13.6  | 4.2       |
| HF        | -0.6   | -4.6  | -2.9  | -2.6      |
| HFCO      | 3.4    | 7.8   | 10.7  | 4.3       |
| HOF       | -3.4   | 8.7   | 7.6   | 2.0       |
| LiF       | 2.4    | -0.8  | -1.5  | -0.1      |
| LiH       | -2.2   | -9.3  | -6.0  | -2.1      |
| N$_2$     | 2.8    | 1.5   | 5.6   | 0.9       |
| N$_2$O    | 0.3    | 7.1   | 6.5   | 3.1       |
| NH$_3$    | -4.0   | -3.1  | -1.6  | -2.4      |
| O$_3$     | 235.4  | 58.6  | 59.4  | 136.6     |
| OCS       | -3.9   | 8.0   | 9.1   | 0.7       |
| OF$_2$    | -4.4   | 26.5  | 25.0  | 7.3       |
| PN        | 14.3   | 15.8  | 23.5  | 10.8      |
| SO$_2$    | 17.3   | 15.9  | 21.6  | 13.6      |

* Statistics for the mean absolute error (MAE), mean error (ME) and the standard deviation (STD) exclude O$_3$. 

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Table 18: Deviations of the magnetizabilities computed with TURBOMOLE and GIMIC for the CAMh-B3LYP, CAM-QTP-00, and CAM-QTP-01 functionals in the aug-cc-pCVQZ basis set from CCSD(T) data from ref. 18 in units of $10^{-30} J/T^2$.

| Molecule | CAMh-B3LYP | CAM-QTP-00 | CAM-QTP-01 |
|----------|------------|------------|------------|
| AlF      | -2.4       | -0.0       | -2.7       |
| C₂H₄     | 7.1        | 0.9        | 4.0        |
| C₃H₄     | 12.7       | 6.5        | 7.0        |
| CH₂O     | 11.8       | 2.9        | 12.1       |
| CH₃F     | 2.1        | -1.0       | -0.9       |
| CH₄      | -1.7       | -0.6       | -5.8       |
| CO       | 1.6        | 4.3        | 0.1        |
| FCCH     | 0.4        | -3.2       | -1.9       |
| FCN      | 1.2        | -1.8       | -1.2       |
| H₂C₂O    | 0.3        | -3.1       | -3.5       |
| H₂O      | -2.1       | 1.1        | -2.9       |
| H₂S      | -0.7       | 1.1        | -3.4       |
| H₂C₂O    | 5.7        | -3.0       | -0.4       |
| HCN      | 0.6        | -2.1       | -2.1       |
| HCP      | 4.4        | -1.7       | 3.0        |
| HF       | -2.4       | 0.9        | -2.7       |
| HFCO     | 5.0        | 1.9        | 2.7        |
| HOF      | 2.9        | -3.3       | -0.2       |
| LiF      | 0.1        | 2.9        | -0.3       |
| LiH      | -2.6       | 2.3        | -1.4       |
| N₂       | 1.6        | 2.9        | -0.4       |
| N₂O      | 3.9        | 0.9        | 1.4        |
| NH₃      | -1.7       | 0.5        | -3.6       |
| O₃       | 128.6      | 251.6      | 161.7      |
| OCS      | 2.0        | -3.9       | -2.3       |
| OF₂      | 9.4        | -7.9       | 2.2        |
| PN       | 12.3       | 10.6       | 7.9        |
| SO₂      | 15.5       | 15.7       | 11.2       |
| MAE*     | 4.2        | 3.2        | 3.2        |
| ME*      | 3.2        | 0.9        | 0.6        |
| STD*     | 5.2        | 4.7        | 4.5        |

* Statistics for the mean absolute error (MAE), mean error (ME) and the standard deviation (STD) exclude O₃.
Table 19: Deviations of the magnetizabilities computed with TURBOMOLE and GIMIC for the CAM-QTP-02, CHACHIYO, HF, KT1, KT2, and KT3 functionals in the aug-cc-pCVQZ basis set from CCSD(T) data from ref. 18 in units of $10^{-30}$ J/T$^2$.

| Molecule  | CAM-QTP-02 | CHACHIYO | HF   | KT1 | KT2 | KT3 |
|-----------|------------|----------|------|-----|-----|-----|
| AlF       | -2.9       | 2.3      | -4.7 | -3.9| 2.1 | 0.3 |
| C$_2$H$_4$| 2.6        | 16.6     | -9.2 | 7.0 | 10.4| 13.2|
| C$_3$H$_4$| 5.6        | 20.2     | 0.8  | 17.4| 21.6| 25.7|
| CH$_2$O   | 10.9       | 18.1     | -12.0| 10.6| 9.4 | 9.5 |
| CH$_3$F   | -1.7       | 4.9      | -2.2 | 5.9 | 8.3 | 10.4|
| CH$_4$    | -6.4       | 1.4      | 3.3  | -3.8| 0.9 | 5.1 |
| CO        | 0.2        | 6.9      | 5.0  | -4.5| 0.4 | 3.4 |
| FCCH      | -2.9       | 5.3      | -10.6| 1.4 | 4.6 |
| FCN       | -2.1       | 6.6      | -8.0 | -2.4| 4.9 |
| H$_2$C$_2$O| -4.7       | 4.3      | -8.7 | -4.2| 1.8 | 7.0 |
| H$_2$O    | -2.7       | -0.8     | 3.9  | -3.7| 0.1 | 1.3 |
| H$_2$S    | -4.0       | 1.7      | 2.5  | -7.0| -0.6| 4.3 |
| H$_4$C$_2$O| -2.6       | 13.0     | -9.7 | 8.2 | 13.9| 18.7|
| HCN       | -3.0       | 10.1     | -8.3 | -3.0| 1.3 | 4.7 |
| HCP       | 1.6        | 17.3     | -18.8| -0.8| 4.9 | 8.9 |
| HF        | -2.4       | -2.0     | 3.7  | -3.4| -0.4| -0.1|
| HFCO      | 1.7        | 12.5     | -4.3 | 3.9 | 8.2 | 9.7 |
| HOF       | -1.7       | 8.2      | -9.2 | 4.0 | 7.8 | 10.5|
| LiF       | 0.0        | -0.6     | 4.8  | -3.6| -0.6| 1.7 |
| LiH       | -0.8       | -4.5     | 1.9  | -11.9| -9.9|-10.8|
| N$_2$     | -0.7       | 8.2      | 2.3  | -4.6| 0.2 | 4.0 |
| N$_2$O    | 0.5        | 7.5      | -3.7 | 4.5 | 8.6 | 11.1|
| NH$_3$    | -3.8       | 0.7      | 2.9  | -3.6| 0.7 | 3.3 |
| O$_3$     | 182.3      | 62.1     | 457.4| 10.4| 17.1| 27.7|
| OCS       | -3.8       | 11.0     | -13.4| 2.0 | 8.5 | 12.5|
| OF$_2$    | -1.3       | 25.0     | -24.7| 15.4| 20.7| 23.8|
| PN        | 7.3        | 28.3     | 4.0  | 6.1 | 11.2| 17.1|
| SO$_2$    | 10.6       | 25.4     | 314.3| 9.7 | 17.2| 21.7|

| MAE*      | 3.3        | 9.8      | 18.4 | 5.9 | 6.4 | 9.2 |
| ME*       | -0.2       | 9.2      | 7.5  | 1.1 | 5.6 | 8.4 |
| STD*      | 4.4        | 8.9      | 61.8 | 7.1 | 7.2 | 8.1 |

* Statistics for the mean absolute error (MAE), mean error (ME) and the standard deviation (STD) exclude O$_3$. 
Table 20: Deviations of the magnetizabilities computed with TURBOMOLE and GIMIC for the LDA, M06, M06-2X, M06-L, M08-HX, and M08-SO functionals in the aug-cc-pCVQZ basis set from CCSD(T) data from ref. 18 in units of $10^{-30}$J/T$^2$.

| Molecule | LDA | M06 | M06-2X | M06-L | M08-HX | M08-SO |
|----------|-----|-----|--------|-------|--------|--------|
| AlF      | -1.3| 7.4 | 1.6    | 11.8  | -2.7   | -1.3   |
| C$_2$H$_4$ | 14.5| 13.2| 15.7   | 18.5  | 14.4   | 11.3   |
| C$_3$H$_4$ | 14.5| 13.6| 17.5   | 17.4  | 16.2   | 15.2   |
| CH$_2$O  | 31.5| 22.9| 33.3   | 3.8   | 33.3   | 38.0   |
| CH$_3$F  | 0.3 | 2.3 | -1.5   | 4.6   | -3.8   | -3.3   |
| CH$_4$   | -12.5| 0.6| -3.0   | 7.5   | -5.7   | -6.1   |
| CO       | 2.9 | 17.2| 15.6   | 14.2  | 7.8    | 14.3   |
| FCCH     | 3.0 | 7.1 | 1.9    | 7.6   | -0.8   | 0.6    |
| FCN      | 4.7 | 11.3| 5.4    | 10.1  | 1.7    | 4.8    |
| H$_2$C$_2$O | -3.8| 6.6| 5.8    | 5.1   | 0.8    | 4.6    |
| H$_2$O   | -5.8| 2.0 | -0.5   | 4.9   | -0.7   | -1.6   |
| H$_2$S   | -10.9| 3.4| -2.6   | 8.0   | -2.0   | -4.0   |
| H$_4$C$_2$O | 5.4| 5.8| -5.1   | 14.1  | -8.2   | -8.3   |
| HCN      | 6.7 | 19.4| 11.4   | 19.2  | 6.6    | 9.4    |
| HCP      | 15.3| 27.8| 16.4   | 29.9  | 9.5    | 12.3   |
| HF       | -4.7| 1.3 | -0.2   | 2.6   | -0.6   | -1.1   |
| HFCO     | 10.3| 15.1| 14.3   | 14.8  | 11.5   | 13.7   |
| HOF      | 6.4 | 7.1 | 0.3    | 5.4   | -1.0   | 0.1    |
| LiF      | -0.8| 5.1 | 2.1    | 3.7   | 2.3    | 2.5    |
| LiH      | -8.8| -3.1| -0.9   | -0.5  | -1.9   | -2.4   |
| N$_2$    | 4.1 | 24.0| 16.1   | 18.7  | 9.5    | 15.5   |
| N$_2$O   | 4.8 | 12.8| 6.3    | 10.1  | 3.4    | 7.4    |
| NH$_3$   | -7.8| 2.1 | -1.6   | 7.3   | -1.7   | -3.1   |
| O$_3$    | 73.7| 291.9| 371.4  | 34.7  | 226.7  | 525.9  |
| OCS      | 7.5 | 13.4| 5.8    | 14.7  | 0.6    | 3.9    |
| OF$_2$   | 26.8| 18.6| 4.2    | 12.8  | 0.1    | 5.5    |
| PN       | 23.6| 58.8| 48.3   | 41.2  | 24.5   | 48.3   |
| SO$_2$   | 19.2| 38.0| 36.9   | 28.9  | 26.6   | 42.5   |
| MAE$^*$  | 9.6 | 13.3| 10.1   | 12.5  | 7.3    | 10.4   |
| ME$^*$   | 5.4 | 13.1| 9.0    | 12.4  | 5.2    | 8.1    |
| STD$^*$  | 11.4| 13.2| 13.1   | 9.4   | 10.3   | 14.3   |

$^*$ Statistics for the mean absolute error (MAE), mean error (ME) and the standard deviation (STD) exclude O$_3$. 
Table 21: Deviations of the magnetizabilities computed with Turbomole and GIMIC for the M11, M11-L, MN12-L, MN12-SX, and MN15 functionals in the aug-cc-pCVQZ basis set from CCSD(T) data from ref. 18 in units of $10^{-30}$J/T$^2$.

| Molecule | M11  | M11-L | MN12-L | MN12-SX | MN15 |
|----------|------|-------|--------|---------|------|
| AlF      | 3.0  | -8.6  | -12.6  | -8.9    | -5.7 |
| C$_2$H$_4$ | 14.3 | 11.5  | 5.2    | 7.2     | 15.2 |
| C$_3$H$_4$ | 12.3 | 17.6  | 7.8    | 11.8    | 18.9 |
| CH$_2$O  | 38.4 | -8.0  | -17.6  | -1.0    | 36.4 |
| CH$_3$F  | -5.0 | 7.9   | 1.9    | 0.2     | 0.8  |
| CH$_4$   | -6.8 | 4.9   | 1.3    | -0.8    | -2.5 |
| CO       | 10.3 | 6.1   | -1.9   | 1.7     | 15.2 |
| FCCH     | 1.6  | -3.3  | -5.0   | -3.6    | 4.4  |
| FCN      | 4.3  | 1.9   | -1.7   | -0.9    | 8.9  |
| H$_2$C$_2$O | -0.4 | -7.5  | -10.7  | -4.9    | 7.6  |
| H$_2$O   | -1.3 | 7.3   | 4.6    | 1.7     | -0.0 |
| H$_2$S   | -4.0 | 4.5   | 1.0    | 0.3     | 0.3  |
| H$_4$C$_2$O | -10.0 | 16.7  | 9.7    | 2.7     | 3.7  |
| HCN      | 9.1  | 7.9   | -1.7   | 0.2     | 12.0 |
| HCP      | 20.9 | 3.8   | -8.7   | -2.1    | 11.3 |
| HF       | -1.5 | 6.6   | 2.9    | 1.2     | -0.4 |
| HFCO     | 13.5 | 6.9   | 1.8    | 3.9     | 18.1 |
| HOF      | 0.6  | 6.8   | 0.6    | -2.0    | 5.3  |
| LiF      | 1.5  | 7.2   | 4.2    | 4.7     | 0.0  |
| LiH      | 0.8  | -22.8 | -17.8  | -13.3   | -4.3 |
| N$_2$    | 11.5 | 11.5  | 0.9    | 3.9     | 18.3 |
| N$_2$O   | 4.6  | 8.4   | 3.0    | 3.0     | 10.7 |
| NH$_3$   | -2.4 | 6.0   | 4.0    | 0.8     | -0.7 |
| O$_3$    | 362.9| -36.6 | -75.9  | 9.6     | 449.5|
| OCS      | 1.2  | 4.2   | -3.2   | -1.9    | 9.8  |
| OF$_2$   | 5.8  | 4.8   | -2.7   | -6.3    | 16.8 |
| PN       | 42.0 | 14.7  | -22.0  | -2.8    | 36.6 |
| SO$_2$   | 40.9 | 23.4  | 2.0    | 11.0    | 45.4 |

MAE$^*$  | 9.9  | 8.9   | 5.8    | 3.8     | 11.4 |
ME$^*$   | 7.6  | 5.2   | -2.0   | 0.2     | 10.4 |
STD$^*$  | 13.8 | 9.3   | 8.0    | 5.3     | 12.8 |

$^*$ Statistics for the mean absolute error (MAE), mean error (ME) and the standard deviation (STD) exclude O$_3$. 

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Table 22: Deviations of the magnetizabilities computed with TURBOMOLE and GIMIC for the MN15-L, MVS, N12, N12-SX, PBE, and PBE0 functionals in the aug-cc-pCVQZ basis set from CCSD(T) data from ref. 18 in units of $10^{-30} \text{J/T}^2$.

| Molecule | MN15-L | MVS  | N12  | N12-SX | PBE   | PBE0  |
|----------|--------|------|------|--------|-------|-------|
| AlF      | -15.7  | 10.2 | 0.4  | -0.2   | -2.4  | 0.2   |
| C$_2$H$_4$ | 2.4    | 25.8 | 14.5 | 11.8   | 14.8  | 10.3  |
| C$_3$H$_4$ | 8.5    | 19.3 | 22.6 | 15.1   | 19.4  | 13.8  |
| CH$_2$O  | -15.0  | 3.9  | 14.5 | 14.1   | 22.5  | 14.6  |
| CH$_3$F  | 1.6    | 6.6  | 7.9  | 1.9    | 4.5   | 1.3   |
| CH$_4$   | -1.9   | 1.8  | 1.3  | 0.4    | -3.4  | -1.5  |
| CO       | -5.8   | 11.5 | 4.3  | 8.4    | 3.9   | 6.6   |
| FCCH     | -8.6   | 4.7  | 8.2  | 5.4    | 4.0   | 1.7   |
| FCN      | -6.7   | 4.3  | 9.2  | 7.4    | 5.0   | 3.4   |
| H$_2$C$_2$O | -14.2 | -4.9 | 7.3  | 4.3    | 2.1   | 0.3   |
| H$_2$O   | -0.7   | 2.8  | 0.8  | 1.4    | -3.4  | -0.1  |
| H$_2$S   | -3.9   | -0.9 | 3.7  | 1.2    | -3.5  | -1.1  |
| H$_3$C$_2$O | 5.1   | 15.4 | 18.6 | 7.0    | 11.3  | 3.7   |
| HCN      | -5.2   | 15.5 | 6.5  | 7.8    | 7.4   | 5.8   |
| HCP      | -15.7  | 25.1 | 15.6 | 12.7   | 13.5  | 10.1  |
| HF       | -0.5   | 1.6  | 0.2  | 0.4    | -3.7  | -0.7  |
| HFCO     | -2.4   | 9.8  | 12.4 | 11.6   | 10.4  | 8.5   |
| HOF      | -4.1   | 8.7  | 12.8 | 5.4    | 8.0   | 2.6   |
| LiF      | -1.8   | 1.0  | 3.8  | 2.2    | -0.7  | 1.4   |
| LiH      | -10.7  | 1.7  | -11.9| -1.7   | -8.0  | -1.9  |
| N$_2$    | -3.5   | 13.3 | 5.4  | 9.7    | 5.4   | 7.0   |
| N$_2$O   | -0.9   | 6.2  | 12.6 | 9.2    | 7.3   | 4.9   |
| NH$_3$   | -1.6   | 2.9  | 1.4  | 1.7    | -2.8  | -0.1  |
| O$_3$    | -57.9  | 15.1 | 63.8 | 171.4  | 61.9  | 136.1 |
| OCS      | -6.6   | 8.1  | 15.7 | 7.4    | 9.3   | 4.5   |
| OF$_2$   | -7.9   | 18.5 | 29.5 | 13.4   | 25.5  | 8.8   |
| PN       | -22.3  | 30.5 | 22.3 | 25.0   | 23.9  | 23.2  |
| SO$_2$   | -4.1   | 24.5 | 30.9 | 30.1   | 20.5  | 22.8  |

| Statistics | MAE*  | ME*   | STD*  |
|------------|-------|-------|-------|
|            | 6.6   | -5.3  | 6.9   |
|            | 10.4  | 9.9   | 9.2   |
|            | 10.9  | 10.0  | 9.6   |
|            | 8.0   | 7.9   | 7.5   |
|            | 9.1   | 7.1   | 9.4   |
|            | 6.0   | 5.6   | 6.8   |

* Statistics for the mean absolute error (MAE), mean error (ME) and the standard deviation (STD) exclude O$_3$. 
Table 23: Deviations of the magnetizabilities computed with TURBOMOLE and GIMIC for the QTP-17, revB3LYP, revM06, and revM06-L functionals in the aug-cc-pCVQZ basis set from CCSD(T) data from ref. 18 in units of $10^{-30}$J/T².

| Molecule | QTP-17 | revB3LYP | revM06 | revM06-L |
|----------|--------|----------|--------|----------|
| AlF      | -2.9   | -2.4     | 4.8    | 8.6      |
| C₂H₄     | 7.1    | 8.6      | 16.0   | 19.0     |
| C₃H₄     | 14.8   | 15.3     | 16.3   | 17.7     |
| CH₂O     | 10.6   | 13.6     | 23.0   | -6.6     |
| CH₂F     | 3.0    | 2.9      | 0.5    | 5.2      |
| CH₄      | -0.0   | -1.4     | -1.5   | 4.9      |
| CO       | 2.4    | 2.7      | 12.3   | 3.1      |
| FCCH     | 0.5    | 1.2      | 3.4    | 3.8      |
| FCN      | 1.6    | 2.3      | 5.8    | 3.5      |
| H₂C₂O    | 1.1    | 1.0      | 2.8    | -3.8     |
| H₂O      | -1.5   | -2.0     | 0.4    | 4.3      |
| H₂S      | -0.2   | -1.2     | -1.2   | 8.2      |
| H₂C₂O    | 7.0    | 7.4      | 1.8    | 17.9     |
| HCN      | 1.0    | 2.2      | 10.5   | 7.1      |
| HCP      | 2.8    | 5.3      | 16.7   | 14.4     |
| HF       | -1.8   | -2.2     | 0.2    | 2.0      |
| HFCO     | 5.5    | 6.5      | 12.8   | 8.3      |
| HOF      | 3.2    | 4.0      | 2.7    | 3.6      |
| LiF      | 0.9    | 0.7      | 1.5    | 0.4      |
| LiH      | -3.7   | -4.1     | -0.6   | 3.3      |
| N₂       | 2.5    | 3.0      | 13.5   | 4.7      |
| N₂O      | 4.7    | 5.0      | 5.9    | 4.5      |
| NH₃      | -0.9   | -1.7     | -0.3   | 5.8      |
| O₃       | 129.9  | 118.0    | 274.2  | -27.6    |
| OCS      | 2.9    | 4.1      | 6.0    | 7.0      |
| OF₂      | 9.9    | 12.9     | 9.0    | 6.1      |
| PN       | 14.0   | 15.9     | 38.4   | 3.0      |
| SO₂      | 17.2   | 17.9     | 33.4   | 12.5     |

| MAE*     | 4.6    | 5.4    | 8.9    | 7.0      |
| ME*      | 3.8    | 4.3    | 8.7    | 6.2      |
| STD*     | 5.4    | 6.1    | 10.3   | 6.0      |

* Statistics for the mean absolute error (MAE), mean error (ME) and the standard deviation (STD) exclude O₃.
Table 24: Deviations of the magnetizabilities computed with TURBOMOLE and GIMIC for the revM11, revTPSS, revTPSSh, rSCAN, and SCAN functionals in the aug-cc-pCVQZ basis set from CCSD(T) data from ref. 18 in units of $10^{-30} \text{J/T}^2$.

| Molecule | revM11 | revTPSS | revTPSSh | rSCAN | SCAN |
|----------|--------|---------|----------|-------|------|
| AlF      | 1.0    | 1.3     | 1.8      | -0.5  | 2.5  |
| C$_2$H$_4$ | 12.6   | 13.6    | 11.9     | 13.3  | 12.6 |
| C$_3$H$_4$ | 11.5   | 21.2    | 18.7     | 15.5  | 16.5 |
| CH$_2$O  | 27.7   | 4.2     | 2.8      | 5.5   | 1.0  |
| CH$_3$F  | -1.1   | 6.8     | 5.4      | 3.9   | 5.3  |
| CH$_4$   | -3.2   | 8.1     | 7.6      | -1.6  | 2.5  |
| CO       | 7.6    | 4.6     | 5.4      | 2.8   | 2.6  |
| FCCH     | 3.8    | 4.9     | 3.8      | 2.4   | 3.3  |
| FCN      | 4.6    | 4.8     | 4.1      | 3.4   | 4.2  |
| H$_2$C$_2$O | 5.2  | 7.6     | 6.2      | -3.4  | -2.1 |
| H$_2$O   | -3.0   | -0.5    | 0.5      | 0.1   | 0.9  |
| H$_2$S   | -1.1   | 6.6     | 6.4      | -2.4  | 1.2  |
| H$_4$C$_2$O | -3.0 | 14.5    | 11.3     | 10.6  | 15.4 |
| HCN      | 7.7    | 7.0     | 6.3      | 5.4   | 6.5  |
| HCP      | 19.7   | 11.7    | 10.3     | 10.3  | 11.1 |
| HF       | -3.5   | -2.1    | -1.1     | -0.4  | -0.1 |
| HFCO     | 9.1    | 9.2     | 8.3      | 8.4   | 7.9  |
| HOF      | 1.6    | 5.1     | 3.2      | 5.3   | 4.5  |
| LiF      | -1.4   | -0.7    | 0.2      | -0.4  | -0.1 |
| LiH      | 3.8    | -1.3    | -0.0     | -3.7  | -3.2 |
| N$_2$    | 9.7    | 6.2     | 6.5      | 3.8   | 5.4  |
| N$_2$O   | 5.1    | 7.8     | 6.8      | 5.5   | 6.5  |
| NH$_3$   | -2.7   | 2.8     | 3.2      | -0.0  | 1.6  |
| O$_3$    | 324.0  | 21.0    | 46.4     | 17.2  | 21.9 |
| OCS      | 3.3    | 10.5    | 8.3      | 6.1   | 6.4  |
| OF$_2$   | 8.1    | 15.9    | 10.3     | 14.7  | 12.8 |
| PN       | 34.2   | 20.1    | 19.6     | 13.4  | 15.9 |
| SO$_2$   | 27.0   | 22.6    | 22.8     | 16.8  | 17.9 |

MAE* | 8.2 | 8.2 | 7.1 | 5.9 | 6.3 |
ME* | 6.8 | 7.9 | 7.1 | 5.0 | 5.9 |
STD* | 10.0 | 6.7 | 5.9 | 6.1 | 6.0 |

* Statistics for the mean absolute error (MAE), mean error (ME) and the standard deviation (STD) exclude O$_3$. 

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Table 25: Deviations of the magnetizabilities computed with TURBOMOLE and GIMIC for the TASK, TPSS, TPSSh, ωB97, ωB97M-V, and ωB97X functionals in the aug-cc-pCVQZ basis set from CCSD(T) data from ref. 18 in units of $10^{-30}$J/T$^2$.

| Molecule | TASK | TPSS | TPSSh | ωB97 | ωB97M-V | ωB97X |
|----------|------|------|-------|-------|----------|--------|
| AlF      | 11.2 | 0.6  | 1.2   | -2.5  | -5.6     | -1.0   |
| C$_2$H$_4$ | 20.2 | 13.5 | 11.8  | 7.5   | 5.1      | 8.1    |
| C$_3$H$_4$ | 18.1 | 20.4 | 18.0  | 7.4   | 9.0      | 9.3    |
| CH$_2$O  | -5.5 | 7.4  | 5.7   | 8.5   | 11.6     | 11.0   |
| CH$_3$F  | 6.4  | 6.2  | 4.8   | 0.4   | 0.7      | 0.8    |
| CH$_4$   | 6.2  | 5.3  | 5.1   | -1.8  | -4.1     | -1.0   |
| CO       | 5.0  | 4.8  | 5.6   | 1.3   | -1.0     | 3.7    |
| FCCH     | 2.5  | 4.9  | 3.8   | -0.5  | -3.0     | 0.4    |
| FCN      | 2.2  | 5.2  | 4.5   | -0.3  | -1.8     | 1.5    |
| H$_2$C$_2$O | -7.4 | 6.0  | 4.8   | -3.1  | -3.3     | -0.7   |
| H$_2$O   | 3.9  | -0.6 | 0.4   | -0.8  | -2.2     | -0.0   |
| H$_2$S   | 0.8  | 4.5  | 4.6   | 0.9   | -3.1     | 0.8    |
| H$_4$C$_2$O | 16.1 | 13.9 | 10.7  | 1.3   | 1.3      | 2.7    |
| HCN      | 9.7  | 7.4  | 6.7   | 2.3   | -2.6     | 3.2    |
| HCP      | 14.6 | 12.3 | 10.9  | 10.9  | 1.8      | 9.0    |
| HF       | 2.7  | -2.0 | -0.9  | -2.0  | -2.3     | -0.8   |
| HFCO     | 8.6  | 9.4  | 8.6   | 3.3   | 3.2      | 6.2    |
| HOF      | 4.3  | 6.1  | 4.1   | 0.9   | 0.1      | 1.9    |
| LiF      | 0.4  | 0.2  | 0.9   | -2.4  | -0.8     | -1.7   |
| LiH      | 6.8  | -2.3 | -0.8  | 0.8   | -6.7     | -2.1   |
| N$_2$    | 7.5  | 6.4  | 6.8   | 2.8   | -1.9     | 4.6    |
| N$_2$O   | 3.4  | 8.3  | 7.3   | 1.4   | 1.6      | 3.1    |
| NH$_3$   | 4.2  | 1.9  | 2.5   | 0.0   | -2.5     | 0.4    |
| O$_3$    | 21.4 | 29.5 | 55.4  | 115.4 | 103.9    | 139.0  |
| OCS      | 3.3  | 10.5 | 8.3   | -1.5  | -2.2     | 0.9    |
| OF$_2$   | 10.3 | 18.2 | 12.3  | 3.7   | 1.7      | 6.0    |
| PN       | 19.1 | 20.9 | 20.5  | 13.6  | 7.5      | 16.0   |
| SO$_2$   | 22.8 | 22.4 | 22.7  | 13.5  | 10.5     | 17.8   |
| MAE*     | 8.3  | 8.2  | 7.2   | 3.5   | 3.6      | 4.2    |
| ME*      | 7.3  | 7.8  | 7.1   | 2.4   | 0.4      | 3.7    |
| STD*     | 7.4  | 6.8  | 6.0   | 4.8   | 4.8      | 5.2    |

* Statistics for the mean absolute error (MAE), mean error (ME) and the standard deviation (STD) exclude O$_3$. 

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Table 26: Deviations of the magnetizabilities computed with TURBOMOLE and GIMIC for the $\omega$B97X-D, and $\omega$B97X-V functionals in the aug-cc-pCVQZ basis set from CCSD(T) data from ref. 18 in units of $10^{-30}$J/T$^2$.

| Molecule | $\omega$B97X-D | $\omega$B97X-V |
|----------|----------------|---------------|
| AlF      | 1.4            | -2.0          |
| C$_2$H$_4$ | 10.1           | 6.7           |
| C$_3$H$_4$ | 12.0           | 9.0           |
| CH$_2$O  | 12.7           | 8.8           |
| CH$_3$F  | 1.6            | 0.8           |
| CH$_4$   | 0.8            | -0.8          |
| CO       | 7.1            | 2.1           |
| FCCH     | 1.8            | -0.8          |
| FCN      | 3.8            | -0.1          |
| H$_2$C$_2$O | 1.8         | -1.4          |
| H$_2$O   | 1.3            | -0.9          |
| H$_2$S   | 2.2            | 0.9           |
| H$_2$C$_2$O | 5.1          | 2.4           |
| HCN      | 5.6            | 1.1           |
| HCP      | 10.1           | 6.3           |
| HF       | 0.5            | -1.7          |
| HFCO     | 9.3            | 4.1           |
| HOF      | 3.4            | 0.7           |
| LiF      | -0.1           | -0.9          |
| LiH      | -4.3           | -0.2          |
| N$_2$    | 7.5            | 2.3           |
| N$_2$O   | 4.8            | 2.3           |
| NH$_3$   | 1.6            | -0.2          |
| O$_3$    | 145.7          | 129.7         |
| OCS      | 4.5            | -0.6          |
| OF$_2$   | 9.1            | 3.3           |
| PN       | 21.7           | 12.2          |
| SO$_2$   | 23.7           | 14.5          |
| MAE*     | 6.2            | 3.2           |
| ME*      | 5.9            | 2.5           |
| STD*     | 6.3            | 4.4           |

* Statistics for the mean absolute error (MAE), mean error (ME) and the standard deviation (STD) exclude O$_3$. 
Table 27: Comparison of the magnetizabilities in $10^{-30} \text{J/T}^2$ calculated with Turbomole (TM) /GIMIC employing the resolution of the identity approximation, and PySCF employing exact integrals at the BP86/aug-cc-pCVQZ and B3LYP/aug-cc-pCVQZ levels of theory. The PySCF data is in full agreement with that from Gaussian.

| Molecule | BP86 | | B3LYP | |
|----------|------|------|------|------|
|          | TM/GIMIC | PySCF | difference | TM/GIMIC | PySCF | difference |
| AlF      | -394.3 | -394.4 | 0.2 | -396.5 | -396.6 | 0.1 |
| C$_2$H$_4$ | -331.0 | -330.9 | -0.0 | -336.7 | -336.7 | -0.0 |
| C$_3$H$_4$ | -460.1 | -460.1 | -0.0 | -463.1 | -463.0 | -0.0 |
| CH$_2$O  | -108.1 | -108.1 | 0.0 | -114.9 | -114.9 | 0.0 |
| CH$_3$F  | -311.4 | -311.4 | 0.0 | -312.4 | -312.4 | 0.0 |
| CH$_4$   | -318.5 | -318.6 | 0.1 | -317.0 | -317.1 | 0.1 |
| CO       | -205.2 | -205.2 | 0.0 | -206.6 | -206.6 | 0.0 |
| FCCH     | -437.9 | -437.9 | 0.0 | -440.1 | -440.0 | 0.0 |
| FCN      | -365.0 | -365.0 | 0.0 | -367.4 | -367.4 | 0.0 |
| H$_2$C$_2$O | -422.0 | -422.1 | 0.1 | -422.1 | -422.2 | 0.1 |
| H$_2$O   | -237.5 | -237.5 | 0.1 | -236.7 | -236.7 | 0.0 |
| H$_2$S   | -456.6 | -456.7 | 0.2 | -455.1 | -455.3 | 0.2 |
| H$_4$C$_2$O | -523.5 | -523.5 | 0.0 | -526.9 | -526.9 | 0.0 |
| HCN      | -264.4 | -264.4 | 0.0 | -269.4 | -269.4 | 0.0 |
| HCP      | -479.2 | -479.3 | 0.0 | -487.4 | -487.4 | 0.0 |
| HF       | -179.3 | -179.3 | 0.0 | -178.4 | -178.4 | 0.0 |
| HFCO     | -296.5 | -296.5 | 0.0 | -300.5 | -300.5 | 0.0 |
| HOF      | -227.8 | -227.7 | 0.0 | -231.1 | -231.1 | 0.0 |
| LiF      | -197.0 | -197.1 | 0.1 | -194.7 | -194.8 | 0.1 |
| LiH      | -133.2 | -133.2 | 0.0 | -130.8 | -130.7 | 0.0 |
| N$_2$    | -199.6 | -199.5 | -0.1 | -202.0 | -201.9 | -0.1 |
| N$_2$O   | -332.6 | -332.7 | 0.1 | -333.8 | -334.0 | 0.1 |
| NH$_3$   | -291.9 | -292.0 | 0.1 | -291.2 | -291.3 | 0.1 |
| O$_3$    | 180.9  | 181.2  | -0.3 | 238.7  | 239.0  | -0.3 |
| OCS      | -575.0 | -575.1 | 0.1 | -579.6 | -579.7 | 0.1 |
| OF$_2$   | -222.1 | -221.8 | -0.2 | -234.1 | -233.9 | -0.2 |
| PN       | -284.7 | -284.3 | -0.4 | -292.2 | -291.8 | -0.4 |
| SO$_2$   | -292.7 | -292.3 | -0.5 | -296.1 | -295.6 | -0.4 |