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Revised values for the nuclear quadrupole moments of ^{33}S and ^{35}S

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High-level quantum-chemical calculations are reported for the sulfur electric-field gradients of the CS and SiS molecules. Highly accurate values are obtained in these calculations by using coupled-cluster methods for the treatment of electron correlation together with large atomic-orbital basis sets and by taking into account relativistic effects. The computational results for the sulfur electric-field gradient are used to determine revised values for the ^{33}S and ^{35}S quadrupole moments, thereby taking advantage of available accurate values for the sulfur quadrupole couplings of CS and SiS from the analysis of rotational spectra. The derived values of −69.4(4) and 48.3(3) mb for ^{33}S and ^{35}S, respectively, are slightly larger in absolute values than the currently accepted values of −67.8(13) and 47.1(9) mb and, most importantly, have significantly reduced uncertainties.

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I. INTRODUCTION

The electric quadrupole moment of a nucleus provides information about its charge distribution, i.e., its shape [1]. Thus, along with the mass and the charge number, the quadrupole moment is a parameter often used for the characterization of a nuclide. Nuclear quadrupole moments also play an important role for a variety of spectroscopic techniques, as interactions with a corresponding electric-field gradient can lead to splittings in the spectra. These so-called nuclear quadrupole couplings are, for example, seen in rotational, NMR, and Mössbauer spectra [2–6]. As the values of the nuclear quadrupole moments thus are of general importance, there exist tabulations compiling the best values available. The most recent one has been published in 2008 by Pyykkö [7].

Experimental values for the nuclear quadrupole moments are probably best determined by converting measured values for the nuclear quadrupole couplings into a value for the quadrupole moment, thereby using accurately computed nuclear electric-field gradients. This combined experimental and theoretical approach is the basis for most of the values reported in Ref. [7].

Among the stable isotopes of sulfur, only ^{33}S and ^{35}S have a nonvanishing quadrupole moment. The currently accepted values of −67.8(13) and 47.1(9) mb stem from the work of Sundholm and Olsen [8]. The values are based on an analysis of the measured quadrupole coupling of ^{33}S$^-$ in its $^2P_{3/2}$ state [9] and extensive multiconfigurational self-consistent-field (MCSCF) calculations for the corresponding electric-field gradient. The accuracy of the reported values is clearly limited by the error bars attached to the experimental quadrupole coupling for ^{33}S$^-$ [9] which are with 2% rather large. The quadrupole moment value for ^{35}S was subsequently obtained via the known ratio of the ^{33}S and ^{35}S quadrupole moments [10].

While Sundholm and Olsen carried out a "state-of-the-art" analysis of the sulfur quadrupole moment, one should note that new, more refined sulfur quadrupole-coupling constants have been reported in the literature afterwards. Noteworthy are in this context the experimental studies of CS and SiS by Müller and co-workers [11–13]. For both diatomic systems, the ^{33}S quadrupole-coupling constants could be determined with a rather small error bar (i.e., less than 0.1%). In addition, it was possible to obtain equilibrium values for the quadrupole coupling via a Dunham analysis [14]. The availability of these highly accurate values for the sulfur quadrupole-coupling constants of CS and SiS clearly suggests a redetermination of the sulfur quadrupole moment with the aim of significantly reducing the uncertainties in the available values.

We will report in the following such a reanalysis based on highly accurate quantum-chemical calculations for the sulfur electric-field gradient of CS and SiS. However, unlike Sundholm and Olsen, we will use coupled-cluster theory [15] in our calculations for the treatment of electron correlation in combination with a systematic enlargement of the atomic-orbital basis set towards the basis-set limit. In addition, we will also account for relativistic effects which, however, are expected to be small.

II. COMPUTATIONAL DETAILS

A value for the sulfur quadrupole moment eQ can be determined from the experimental equilibrium ^{33}S quadrupole-coupling constants χ of CS and SiS via

$$eQ = \hbar \frac{\chi}{q} \tag{1}$$

and computed values for the electric-field gradient q at the sulfur nucleus. Note that in the case of diatomic molecules, χ and q refer to the zz component of both the quadrupole-coupling and electric-field gradient tensors. The experimental values for χ are taken from Refs. [11] and [12] and are 13.0265(68) MHz for CS and 11.07684(148) MHz for SiS, respectively. These are equilibrium values and thus no vibrational effects need to be accounted for in the following. However, the zero-point vibrational corrections to χ were also determined in Refs. [11] and [12]. The corresponding values are −199.73(1.95) kHz for CS [12] and −124.64(0.31) kHz for SiS [11] and their analysis can provide further support for the accuracy of the ^{33}S quadrupole moment values to be derived in the following.

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The calculations reported in the present work are all concerned with the electric-field gradient at the sulfur nucleus in CS and SiS which is computed as a first-order property with

$$\hat{q}^K = \frac{\mathbf{1}(\mathbf{r}-\mathbf{R}_K)^2 - 3(\mathbf{r}-\mathbf{R}_K)(\mathbf{r}-\mathbf{R}_K)^T}{|\mathbf{r}-\mathbf{R}_K|^5} \tag{2}$$

as the corresponding property operator [16]. In Eq. (2), $\mathbf{r}$ and $\mathbf{R}_K$ are the position of the electron and the considered nucleus, respectively. The first-order property is then computed using analytic-derivative theory [17] as a first derivative of the energy.

To ensure an adequate treatment of electron-correlation effects, the calculations presented in the following are all based on coupled-cluster (CC) theory [15,18,19]. The standard choice is here the CC singles, doubles (CCSD) scheme [20] augmented by a perturbative treatment of triple excitations, i.e., CCSD(T) [21]. This scheme is applicable together with rather large basis sets and has been shown to provide high-accuracy results. However, in the present work it is necessary to account also for higher-order effects not covered by CCSD(T), i.e., the difference between a rigorous treatment of triple excitations at the CC singles, doubles, triples (CCSDT) level [22,23] and CCSD(T) and in particular for contributions due to quadruple excitations. For this reason, additional computations were performed at the CCSDT as well as at the CC singles, doubles, triples, quadruples (CCSDTQ) level [24]. As the CCSDTQ treatment of quadruple excitations is rather expensive, we also carried out CCSDT(Q) and CCSDT(Q)$_\Lambda$ calculations [25,26] in which the quadruple excitations are treated perturbatively on top of CCSDT. These schemes have been formulated in analogy to CCSD(T) and its CCSD(T)$_\Lambda$ variant [27,28] which both provide a perturbative treatment of triple excitations.

As the calculations beyond the CCSD(T) level are rather expensive and cannot be carried out with large basis sets, the CCSDT, CCSDTQ, CCSDT(Q), and CCSDT(Q)$_\Lambda$ calculations with smaller basis sets were used to obtain the corresponding differences to be added to the results of the large-basis CCSD(T) computations. In contrast to the all-electron CCSD(T) calculations, these corrections were obtained within the frozen-core (fc) approximation with the $1s$ carbon and the $1s$, $2s$, and $2p$ sulfur and silicon shells excluded in the correlation treatment. We consider in the following corrections (a) due to full treatment of triple excitations [difference between CCSDT and CCSD(T)], (b) due to quadruple excitations based on a perturbative treatment [difference between CCSDT(Q) and CCSDT], and (c) due to a full treatment of quadruples [difference between CCSDTQ and CCSDT(Q)].

To ensure that the results for the sulfur electric-field gradients are close to the basis-set limit, the calculations were carried out with a series of basis sets obtained by augmenting uncontracted versions of standard basis sets with additional steep core functions. We followed here a strategy which was already used with success in our previous computational studies of copper and bromine electric-field gradients [29,30]. Our starting points were the uncontracted versions of Dunning's core-polarized cc-pCVXZ sets [31,32]. However, to eliminate linear dependencies, the core-correlating s and p functions need to be skipped. We refer to these basis sets as BAS-3 to BAS-5 depending on X. X equal T thus corresponds to BAS-3, X equal 4 to BAS-4, and X equal 5 to BAS-5. The sets BAS-6 to BAS-12 are then obtained by appending additional steep functions to BAS-5. For BAS-6 to BAS-9 this was done by adding in each step one set of p, d, and f functions with the exponents obtained by multiplying the largest exponent among the basis functions with same angular momentum by three. The sets BAS-10 to BAS-12 were obtained in the same way by augmenting the BAS-9 set with additional steep s, p, d, f, g, and h functions. The basis sets BAS-5+ to BAS-12+ also contain additional diffuse functions taken from Refs. [33] and [34]. The calculations at levels higher than CCSD(T) were carried out with the standard cc-pCVXZ basis sets (X=T and Q) [31,32].

Since the accurate determination of electric-field gradients requires consideration of relativistic effects, [35,36] two different scalar-relativistic treatments were used. The first one was direct perturbation theory (DPT) [37] and its choice was motivated by the fact that we are dealing in the case of CS and SiS with systems containing rather light elements only. We employed the lowest-order scheme, i.e., second-order DPT (DPT2), and evaluated the scalar-relativistic corrections to the sulfur electric-field gradients analytically as described in Ref. [38]. The second scheme used in the present work was the spin-free exact two-component approach in its one-electron variant (SFX2C-1e) [39–43] and we used our recent implementation of analytic derivatives [43] for the evaluation of the electric-field gradients. The scalar-relativistic treatments otherwise employed the same methods and basis sets as the nonrelativistic calculations; i.e., they were performed at the CCSD(T) level using the uncontracted BAS-n and BAS-n+ basis sets.

In the case of CS and SiS, spin-orbit (SO) effects on the electric-field gradients can be assumed to be small. Nevertheless, we estimated their magnitude using fourth-order DPT [44]. The SO corrections to the electric-field gradients were here obtained in a numerical manner at the Hartree-Fock (HF) level as described in Ref. [45].

As there have been indications [35] that Gaunt corrections provide a nonnegligible contribution to computed electric-field gradients, additional calculations were performed at the Dirac-Hartree-Fock (DHF) level with and without the Gaunt term [46] included in the Hamiltonian. These calculations were performed with the BAS-5 set using the kinetic-balance condition for obtaining the corresponding basis for the small component.

All quantum-chemical calculations for the equilibrium value of the sulfur electric-field gradients were carried out at the theoretical best-estimate equilibrium geometries of CS and SiS, obtained as described in Ref. [47]. The distances used are 1.53484 Å for CS and 1.92957 Å for SiS, respectively.

Finally, zero-point vibrational corrections were computed for the sulfur electric-field gradients of CS and SiS. The corrections were not needed for the analysis of the experimental equilibrium quadrupole couplings, but they enabled an analysis of the experimentally determined vibrational corrections to the quadrupole couplings and thus provide a consistency check for the derived value of the sulfur quadrupole moment. Two schemes were used to obtain the vibrational corrections. The first one is second-order vibrational perturbation theory

(VPT2) [48,49]. The vibrational corrections for the properties are here determined in a perturbative manner based on the quadratic and cubic force fields as well as the corresponding property derivatives. The second scheme employed allowed a more rigorous treatment and comprises a variational solution of the vibrational problem using a discrete variable representation (DVR) [50–53]. We used 13 quadrature points (within a Gauss-Hermite quadrature) for the representation of the potential curves of CS and SiS and solved the vibrational problem in a basis consisting of 13 harmonic-oscillator functions. The zero-point vibrational corrections were determined at the CCSD(T) levels of theory with the potential and property curves as well as the corresponding force fields and property derivatives taken from calculations using the BAS-5 basis set.

All quantum-chemical calculations were performed with a local version of the CFOUR program package [54] except for the CCSDTQ, CCSDT(Q), and CCSDT(Q)$_\Lambda$ calculations which were carried out with the MRCC program [55] interfaced to CFOUR and the DHF calculations which were performed using the DIRAC program suite [56].

III. RESULTS AND DISCUSSION

An accurate value can only be derived for the ^{33}S quadrupole moment if the error in the used experimental quadrupole coupling is sufficiently small and if the required electric-field gradient is computed with sufficient accuracy. In the present case, the errors in the experimental quadrupole couplings [11,12] are so small that they will only lead to uncertainties of about 0.04 (CS) and 0.01 mb (SiS) in the quadrupole moment. More critical, however, are remaining errors in the computations of the electric-field gradient: for an accuracy of about 1 mb in the quadrupole moment, the accuracy in the electric-field gradient calculations should be around 0.01 a.u.; for an accuracy of about 0.1 mb, the electric-field gradient should be accurate to about 0.001 a.u. As it is a challenge to reach such an accuracy in the computations, we start in the following with a detailed analysis of our results from the electric-field gradient calculations, thereby focusing on basis-set convergence, higher-order correlation effects, as well as the importance of relativistic contributions. After this, we will report our analysis concerning a revision of the value for the sulfur quadrupole moment.

Table I summarizes the results concerning the basis-set convergence in our nonrelativistic (nrl) and relativistic CCSD(T) electric-field gradient calculations. Convergence to about 0.01 a.u. is reached with the BAS-5 set; convergence to about 0.001, however, is only achieved with the BAS-9 basis. Further extension of the basis up to BAS-12 essentially leaves the computed results unchanged. The effect of additional diffuse functions monitored through the use of the BAS-n+ sets is also negligible. The best nonrelativistic CCSD(T) value for the sulfur electric-field gradients thus is −0.7969 a.u. in the case of CS and −0.6733 a.u. in the case of SiS. The remaining basis-set error is estimated to about 0.0001 a.u.

These best CCSD(T) values need to be adjusted in a second step for higher-order correlation effects not covered by the CCSD(T) treatment. Table II gathers the corresponding results which have been obtained at the CCSDT, CCSDT(Q), CCSDT(Q)$_\Lambda$, and CCSDTQ levels of theory. Due to the high cost of these additional calculations, they were performed with much smaller basis sets than the CCSD(T) calculations in Table I and thus we use the results in Table II just for obtaining corrections that can be addded to our best CCSD(T) values. Turning first to the CCSDT results, we note that CCSDT/cc-pCVQZ calculations with only the 1s orbitals frozen indicate that the effect of a rigorous instead of a perturbative treatment of triple excitations amounts to +0.0147 a.u. in the case of CS and to +0.0056 a.u. in the case of SiS, respectively. The variation in the different CCSDT results in Table II [in comparison to CCSD(T)] indicates that the remaining error is probably of the order 0.001 to 0.002 a.u. for both molecules. The contributions due to quadruple excitations amount to about −0.0065 a.u. for CS and −0.0084 a.u. for SiS, respectively, when computed at the CCSDT(Q)/cc-pCVTZ level with the 1s shells kept frozen. CCSDT(Q) calculations with all inner-shell orbitals frozen indicate that the use of a quadruple-zeta instead of a triple-zeta basis slightly increases the quadruple corrections. This effect

TABLE I. Computed sulfur electric-field gradients (in a.u.) of CS and SiS as obtained in nonrelativistic and relativistic CCSD(T) calculations using the BAS-n and BAS-n+ basis sets.

	CS			SiS		
	nrl	SFX2C-1e	DPT2	nrl	SFX2C-1e	DPT2
BAS-3	−0.8098	−0.8167	−0.8167	−0.6832	−0.6861	−0.6860
BAS-4	−0.8106	−0.8177	−0.8177	−0.6816	−0.6848	−0.6847
BAS-5	−0.7981	−0.8053	−0.8053	−0.6745	−0.6778	−0.6778
BAS-9	−0.7971	−0.8044	−0.8044	−0.6733	−0.6767	−0.6767
BAS-10	−0.7967	−0.8041	−0.8042	−0.6732	−0.6767	−0.6767
BAS-11	−0.7968	−0.8041	−0.8041	−0.6732	−0.6767	−0.6766
BAS-12	−0.7968	−0.8042	−0.8042	−0.6732	−0.6767	−0.6767
BAS-5+	−0.7983	−0.8055	−0.8055	−0.6746	−0.6779	−0.6778
BAS-9+	−0.7973	−0.8046	−0.8046	−0.6734	−0.6768	−0.6768
BAS-10+	−0.7969	−0.8043	−0.8043	−0.6733	−0.6768	−0.6767
BAS-11+	−0.7970	−0.8043	−0.8043	−0.6733	−0.6768	−0.6767
BAS-12+	−0.7969	−0.8044	−0.8044	−0.6733	−0.6768	−0.6768

TABLE II. Contributions of higher excitations to the sulfur electric-field gradients (in a.u.) of CS and SiS as obtained in CCSD(T), CCSDT, CCSDT(Q), CCSDT(Q)$_\Lambda$, and CCSDTQ calculations.

	frozen core		1*s* frozen	
	cc-pCVTZ	cc-pCVQZ	cc-pCVTZ	cc-pCVQZ
(a) CS				
CCSD(T)	−0.8140	−0.8213	−0.8112	−0.8151
CCSDT	−0.7998	−0.8061	−0.7974	−0.8004
CCSDT(Q)	−0.8068	−0.8141	−0.8039	
CCSDT(Q)$_\Lambda$	−0.8088	−0.8160	−0.8060	
CCSDTQ	−0.8064			
(b) SiS				
CCSD(T)	−0.6865	−0.6916	−0.6805	−0.6821
CCSDT	−0.6818	−0.6857	−0.6760	−0.6765
CCSDT(Q)	−0.6898	−0.6945	−0.6844	
CCSDT(Q)$_\Lambda$	−0.6904	−0.6949	−0.6849	
CCSDTQ	−0.6898			

is estimated to about −0.0010 a.u. for CS and −0.0008 a.u. for SiS, respectively, so that our best estimates for the perturbative contributions due to quadruple excitations are −0.0075 and −0.0092 a.u., respectively. Frozen-core CCSDTQ/cc-pCVTZ computations furthermore indicate that a rigorous treatment of quadruples leads to results that are more or less identical to those from CCSDT(Q) computations. Interestingly, the performance of the CCSDT(Q)$_\Lambda$ scheme is somewhat inferior than the one of CCSDT(Q) when compared to CCSDTQ. To give error estimates for the contributions due to quadruple excitations is not straightforward. It seems that remaining basis-set effects are here a significant error source and we estimate the remaining error to be of the order of 0.001 to 0.002 a.u. for both CS and SiS. Contributions due to even higher than quadruple excitations are expected to be negligible and have not been further investigated.

The SFX2C-1e and DPT2 calculations, see Table I, indicate that scalar-relativistic effects are, as expected, not too pronounced in the case of CS and SiS. The corresponding contributions are −0.0075 and −0.0035 for CS and SiS, respectively, when computed at the CCSD(T) level with the large BAS-n basis sets. The results obtained at the SFX2C-1e and DPT2 level agree very well; i.e., the discrepancies are 0.0001 a.u. or less. This agreement is an indication that the chosen schemes provide a nearly quantitative treatment of the scalar-relativistic effects. SO effects, however, are assumed to be negligible and DPT4 computations at the HF/BAS-5 level confirm our expectations. The corresponding corrections are 0.00007 a.u. for CS and 0.00019 a.u. for SiS, respectively.

Table III summarizes all contributions considered in the present work in the computation of the sulfur electric-field gradients of CS and SiS. The final values are −0.7975 a.u. for CS and −0.6809 a.u. for SiS. The corresponding error estimates are of the order of 0.002 to 0.003 a.u. for both molecules, mostly due to the remaining uncertainties in the basis-set convergence of higher excitations beyond CCSD(T). The conversion of the experimental sulfur quadrupole-coupling constants using our best-estimate values for the sulfur electric-field gradients leads to values of −69.52(30) mb and −69.24(30) mb for the ^{33}S quadrupole moment, depending on whether one considers CS or SiS. The given uncertainties are based on assumed error estimates of 0.002 to 0.003 a.u. for the sulfur electrical-field gradients, as already discussed. We note that the sole consideration of the nonrelativistic CCSD(T) contribution lead in particular for SiS to a rather larger error of 0.7 mb in the derived value for the quadrupole moment. Contributions due to a full treatment of triples and due to quadruple excitations are nonnegligible, i.e., up to 1.3 mb, but they have opposite signs and partially cancel. Their combined contributions are about −0.64 mb in the case of CS and 0.36 mb in the case of SiS, indicating that CCSD(T) indeed benefits from some error cancellation. Relativistic effects also amount to corrections of the same order of magnitude and thus are not negligible for an accurate determination of the sulfur quadrupole moment. However, unlike the scalar-relativistic corrections, SO contributions turn out to be more or less negligible. The same is not true for the Gaunt corrections computed at the DHF level which contribute about 0.06 and 0.07 mb for CS and SiS, respectively.

The two values, derived from the experimental quadrupole couplings of CS and SiS, differ slightly, but they agree within 0.3 mb. We thus propose to use in the future the mean of both values, i.e., −69.4 mb with an uncertaintity of 0.4 mb as the currently best available value for the ^{33}S quadrupole moment. For the ^{35}S quadrupole moment, we propose to use a value of

TABLE III. Individual contributions to the sulfur electric-field gradient (efg, in a.u.) of CS and SiS and the derived values for the ^{33}S quadrupole moment (eQ, in mb).

	CS		SiS	
	efg	eQ	efg	eQ
CCSD(T) basis-set limit	−0.7969	−69.57	−0.6733	−70.02
+ full treatment of triples	0.0147	−1.31	0.0056	−0.59
+ perturbative quadruples	−0.0075	0.67	−0.0092	0.96
+ corrections for full quadruples	0.0004	−0.04	0.0000	0.00
+ scalar-relativistic corrections	−0.0075	0.66	−0.0035	0.36
+ spin-orbit corrections	0.0001	0.01	0.0002	0.02
+ Gaunt corrections	−0.0008	0.06	−0.0007	0.07
final value	−0.7975	−69.52	−0.6809	−69.24

TABLE IV. Calculated vibrational corrections to the sulfur electric-field gradients (efg, in a.u.) and quadrupole-coupling constants (χ, in kHz) of CS and SiS. All calculations were performed at the CCSD(T) level with the BAS-5 basis and a value of –69.4 mb is used for the ^{33}S quadrupole moment in the conversion of the electric-field gradients.

	CS		SiS	
	efg	χ	efg	χ
VPT2[a]	0.01182	−192.7	0.00753	−122.7
DVR[a]	0.01178	−192.1	0.00751	−122.4
DVR[b]	0.01189	−193.9	0.00755	−123.1
exp.		–199.73 (1.95)[c]		–124.64 (0.31)[d]

[a] Potential curve and efg's from nonrelativistic calculations.
[b] efg's from SFX2C-1e calculations.
[c] Reference [12].
[d] Reference [11].

48.3(3) mb based on the experimentally known ratio of −0.695 for the ^{33}S and ^{35}S quadrupole moments [10].

Our revised values for the sulfur quadrupole moments are more or less in line with the earlier results of Sundholm and Olsen [8] who reported values of −67.8(13) and 47.1(9) mb for ^{33}S and ^{35}S, respectively. The present values are slightly larger (i.e., by about 1.6 and 1.2 mb), but the main achievement is the significantly reduced uncertainty. While Sundholm and Olsen estimated the uncertainty of their value to about 1.3 mb in the case of ^{33}S, the present study yields an error estimate of only 0.4 mb. In the same way, we reduce the uncertainty for the ^{35}S value from 0.9 mb in Ref. [8] down to about 0.3 mb.

A consistency check for our revised sulfur quadrupole moment values is provided by a comparison of the computed and experimentally derived vibrational corrections for the quadrupole couplings in CS and SiS (see Table IV). For both molecules, the various computational schemes agree within a few kHz in their values. To be more specific, the DVR scheme provides in absolute terms slightly smaller corrections than VPT2 and consideration of relativistic effects leads to a small increase in the computed values. In comparison to experiment, the computed values are somewhat too small, by about 6 kHz in the case of CS and 1.5 kHz in the case of SiS, respectively. While an uncertainty of about 1 kHz can be attributed to the remaining uncertainty in the revised sulfur quadrupole-moment value, the contributions of other error sources such as inaccuracies in the potential curves, remaining basis-set effects, and neglected higher-order correlation effects are difficult to estimate. Nevertheless, the reached agreement between computed and experimental vibrational corrections can be taken as a further indication of the reliability and accuracy of our revised sulfur quadrupole-moment values.

Finally, we note that our revised ^{33}S quadrupole-moment value was already used in Ref. [57] in the computation of the sulfur quadrupole couplings in H_2S and SO_2.

IV. SUMMARY

Based on our analysis of the experimental sulfur quadrupole-coupling constants for CS and SiS and high-level quantum-chemical calculations of the corresponding sulfur electric-field gradients, we propose a revision of the value for the ^{33}S and ^{35}S quadrupole moments. The revised values are with −69.4(4) and 48.3(3) mb for ^{33}S and ^{35}S slightly larger than the previous values derived by Sundholm and Olsen [8] and most importantly have significantly reduced uncertainties. In comparison with the previous values from Ref. [8], they have been reduced by a factor of about 3.

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more challenging due to the increased significance of relativistic effects and due to the necessity of considering vibrational corrections. A value reported by Le Floch and Masson [A. C. Le Floch and J. Masson, *ibid.* **103**, 408 (1984)] for the sulfur quadrupole coupling of GeS, however, turns out to be not accurate enough for our purpose.

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