Search for parity and time reversal violating effects in HgH: Relativistic coupled-cluster study

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The high effective electric field ($E_{\text{eff}}$) experienced by the unpaired electron in an atom or a molecule is one of the key ingredients in the success of electron electric dipole moment (eEDM) experiment and its precise calculation require a very accurate theory. We, therefore, employed the Z-vector method in the relativistic coupled-cluster framework and found that HgH has a very large $E_{\text{eff}}$ value (123.2 GV/cm) which makes it a potential candidate for the next generation eEDM experiment. Our study also reveals that it has a large scalar-pseudoscalar $P, T$-violating interaction constant, $W_s = 284.2$ kHz. To judge the accuracy of the obtained results we have calculated parallel and perpendicular magnetic HFS constants and compared with the available experimental values. The results of our calculation are found to be in nice agreement with the experimental values. Therefore, by looking at the HFS results we can say that both $E_{\text{eff}}$ and $W_s$ values are also very accurate. Further, We have derived the relationship between these quantities and the ratio which will help to get model independent value of eEDM and S-PS interaction constant.

I. INTRODUCTION

In the quest of new physics, there have been an extensive search in order to observe the violation of parity ($P$) and time reversal ($T$) symmetries. An accurate measurement of electric dipole moment of an electron (eEDM)2,3, which arises due to the violation of both $P$ and $T$ is the most promising way to explore in this direction. Although, the intensive search over the past half a century did not conclude in any final value of eEDM, however, it leads to achieve a tremendous increase in the experimental sensitivity, and an upper bound limit of eEDM4. The enhancement of eEDM effects in heavy polar diatomic molecules is the main reason for the higher sensitivity of modern eEDM experiment5,6. The sensitivity of the eEDM experiment using a heavy polar diatomic molecule depends on the molecule’s permanent molecular EDM7,8. There are two main sources of permanent molecular EDM of a paramagnetic molecule; the eEDM and the coupling interaction between the scalar-hadronic current and the pseudoscalar electronic current. However, in most of the calculation either eEDM or the scalar-pseudoscalar (S-PS) interaction is considered as the only possible source of permanent molecular EDM. For example, the best upper bound limit of eEDM ($d_e$) and S-PS interaction constant ($k_s$) is obtained from the ThO experiment by ACME collaboration4 where they have used the theoretically calculated value of effective electric field ($E_{\text{eff}}$) and S-PS $P, T$-odd interaction constant ($W_s$)8,10. In the calculation of $d_e$, the S-PS coupling constant $k_s$, is assumed to be zero and vice versa, although both of these contribute to the $P, T$-odd frequency shift in the experiment. However, it is possible to get independent limit of $d_e$ and $k_s$ by using the results from two different experiments11 and for this the accurate value of $E_{\text{eff}}$, $W_s$ and their ratio are very important. Since, $E_{\text{eff}}$ and $W_s$ cannot be measured by any experimental technique, therefore, these quantities have to be calculated by means of an accurate theoretical method, which can incorporate both the effects of relativity and electron correlation in an intertwined manner.

In this article, we focus on HgH molecule as it offers very high value of $E_{\text{eff}}$ and $W_s$ in its ground electronic state ($^2\Sigma$), which makes it a potential candidate for the future eEDM experiments. The Z-vector method in the relativistic coupled-cluster formalism is used to calculate $E_{\text{eff}}$ and $W_s$ as it is the most reliable method for the calculation of ground state properties. However, high values of $E_{\text{eff}}$ and $W_s$ are not the only requirement for the precise measurement of eEDM, but the molecule must be fully polarized with a low external electric field to fully utilize the $E_{\text{eff}}$. The large rotational constant and small dipole moment of HgH suggest that one needs to apply a much higher electric field to polarize HgH in a spectroscopic experiment. However, Kozlov and Derevianko have suggested that it can be polarized easily in the matrix isolated solid state non-spectroscopic experiment15, which also offers 2-3 orders of higher sensitivity than the current limit. Therefore, the detailed investigation of $E_{\text{eff}}$ and $W_s$ of HgH and their inter-relation is very important for the eEDM experiment based on HgH molecule.

The manuscript is organized as follows. Concise details of the calculated properties including a brief overview of the Z-vector method are described in Sec. II. Computational details are given in Sec. III. We present our calculated results and discuss about those in Sec. IV be-

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fore making final remark in Sec. V. Atomic unit is used consistently unless stated.

II. THEORY

A. Properties

The matrix element of $E_{\text{eff}}$ is given by the following expression,

$$E_{\text{eff}} = |W_d\Omega| = |\langle \Psi_\Omega | \sum_j^n \frac{H_d(j)}{d_e} | \Psi_\Omega \rangle|,$$

where $\Omega$ is the projection of total angular momentum along the molecular axis and $\Psi_\Omega$ is the electronic wavefunction corresponding to $\Omega$ state. $n$ is the total number of electrons and $H_d$ is the interaction Hamiltonian of $d_e$ with internal electric field and is given by

$$H_d = 2icd_e \gamma^0 \gamma^5 p^2,$$

where $\gamma$ are the usual Dirac matrices and $p$ is the momentum operator. Another $P, T$-odd interaction constant, $W_s$, can be obtained by evaluating the following matrix element

$$W_s = \frac{1}{\Omega k_s} \langle \Psi_\Omega | \sum_j^n H_{SP}(j) | \Psi_\Omega \rangle,$$

where $k_s$ is the dimensionless electron-nucleus scalar-pseudoscalar coupling constant which is defined as $Z_{k_s} = (Z_{k_s,p} + N k_{s,n})$, where $k_{s,p}$ and $k_{s,n}$ are electron-proton and electron-neutron coupling constant, respectively. The interaction Hamiltonian is defined as

$$H_{SP} = \frac{G_F}{\sqrt{2}} Z k_s \gamma^0 \gamma^5 \rho_N(r),$$

where $\gamma$ are the usual Dirac matrices, $\rho_N(r)$ is the nuclear charge density normalized to unity and $G_F$ is the Fermi constant. The calculation of the above matrix elements depends on the accurate wavefunction in the near nuclear region and the standard way to determine the accuracy of the electronic wavefunction in that region is to compare the theoretically calculated hyperfine structure (HFS) constant with the experimental value. The parallel ($A_{||}$) and perpendicular ($A_{\perp}$) magnetic HFS constants of a diatomic molecule can be written as

$$A_{||,\perp} = \frac{\mu_k}{\Omega} \cdot \langle \Psi_\Omega | \sum_i^n \left( \frac{\vec{a}_i \times \vec{r}_i}{r_i^3} \right) z(x/y) | \Psi_\Omega(-\Omega) \rangle,$$

where $\Omega$ represents the $z$-component of the total angular momentum of the diatomic molecule and it is 1/2 for the ground ($^2\Sigma$) state of HgH.

B. Relation between independent limit of $d_e$ and $k_s$, with experimentally determined $d_{e,\text{exp}}$

Since, the dominant contribution of $P, T$-odd frequency shift comes from the eEDM and S-PS interaction, considering only those two effects we can get the following relation

$$d_e E_{\text{eff}} + \frac{W_s k_s}{2} = \hbar \omega_{p, T},$$

where $\hbar$ is the Planck’s constant and $\omega_{p, T}$ is the experimentally measured $P, T$-odd frequency shift.

$$\Rightarrow d_e + \frac{W_s k_s}{2E_{\text{eff}}} = \frac{\hbar \omega_{p, T}}{E_{\text{eff}}},$$

or,

$$d_e + \frac{k_s}{2R} = d_{e,\text{exp}}|_{k_s=0}.$$  

Here, $d_{e,\text{exp}}|_{k_s=0}$ is the eEDM limit derived from the experimentally measured $P, T$-odd frequency shift where $k_s$ is assumed to be 0 and $R$ is defined as

$$R = \frac{E_{\text{eff}}}{W_s}.$$

Eq.8 defines the interrelation of the independent limit of $d_e$, $k_s$ and experimentally determined $d_{e,\text{exp}}$.

C. Z-vector method

The relativistic coupled-cluster (CC) method is employed to calculate the molecular wavefunction. The CC energy is a function of both the molecular orbital coefficients and determinantal coefficients in the expansion of a many electron correlated wavefunction for a fixed nuclear geometry. Thus, the CC energy derivative calculation needs to incorporate the derivative of energy with respect to these two parameters in addition to the derivative of these two terms with respect to the external field of perturbation. However, the derivative of energy with respect to determinantal coefficients and the derivative of determinant coefficients with respect to external perturbation field can be included with the introduction of a perturbation independent linear equation whose solution yields the $Z$-vector.

Computationally, Z-vector calculation in the CC framework is a three step process - (i) calculation of normal coupled-cluster excitation operator $(T)$, (ii) calculation of perturbation independent deexcitation operator $(\Lambda)$, and (iii) calculation of energy derivative using $T$ and $\Lambda$. The form of the $T$ and $\Lambda$ operators are given by

$$X = X_1 + X_2 + \ldots + X_N = \sum_n^n X_n$$

with

$$X_n = \sum_{q_1 \ldots q_n} a_{p_1}^{q_1} \ldots a_{p_n}^{q_n} a_{p_1} \ldots a_{p_n}.$$
where $x^{q_1,q_2,\ldots}$ are the cluster amplitudes corresponding to the cluster operator $X_m$, $X$ is $T$ when $p(q)$ are hole/(particle) index and $X$ is $A$ when $p(q)$ are particle/(hole) index. In the coupled-cluster single- and double-(CCSD) model, $T$ is $T_1 + T_2$ and $A$ is $A_1 + A_2$. The coupled-cluster $T_1$ and $T_2$ amplitudes can be calculated by solving the following equations,

$$
\langle \Phi_0 | (H_N e^T) \langle \rangle | \Phi_0 \rangle = 0, \quad \langle \Phi_0 | (H_N e^T) \langle \rangle | \Phi_0 \rangle = 0, \quad (12)
$$

where $H_N$ is the normal-ordered Dirac-Coulomb Hamiltonian and subscript $c$ means only the connected terms exist in the contraction between $H_N$ and $T$. Size-extensivity is ensured by this connectedness. The explicit equations for the amplitudes of $A_1$ and $A_2$ operators are given by

$$
\langle \Phi_0 | [A(H_N e^T)]_{ij} | \Phi_0 \rangle = 0, \quad (13)
$$

Finally, the equation for energy derivative can be written as

$$
\Delta E' = \langle \Phi_0 | (O_N e^T)_{ij} | \Phi_0 \rangle + \langle \Phi_0 | [\Delta(O_N e^T)]_{ij} | \Phi_0 \rangle, \quad (15)
$$

where $O_N$ is the normal-ordered property operator.

### III. COMPUTATIONAL DETAILS

The locally modified version of DIRAC10 program package\textsuperscript{21} is used to solve the Dirac-Hartree-Fock equation where the Dirac-Coulomb Hamiltonian is used. On the other hand, the $Z$-vector method in the CCSD framework is used for the correlation treatment. The wavefunction is four-component in nature and the small-component functions are linked to the large-components by the restricted kinetic balance (RKB) condition.\textsuperscript{22} Finite nucleus size is considered and the nuclear potential is calculated considering Gaussian charge distribution.\textsuperscript{23} We have done two calculations - one with triple zeta (TZ) basis (dyall.cv3z for Hg\textsuperscript{24} and cc-pCVTZ for H\textsuperscript{25}) and the other with quadruple zeta (QZ) basis (dyall.cv4z for Hg\textsuperscript{24} and cc-pCVQZ for H\textsuperscript{25}). As the higher energy virtual orbitals contribute very less in the correlation calculations, the virtual orbitals whose energy exceeds 500 a.u. are removed from our calculations. None of the occupied orbitals are frozen in our correlation calculation as the core polarization effect plays a vital role for the type of properties of interest.\textsuperscript{14} The experimental bond length of HgH (1.766 Å)\textsuperscript{26} is used to calculate the properties in its ground state ($^2\Sigma_{1/2}$).

### IV. RESULTS AND DISCUSSION

The accuracy of the property values given in Eq.1 and Eq.3 can be determined by comparing the theoretically obtained magnetic HFS constants with the experimental values since all these matrix elements require an accurate wavefunction in the near nuclear region of the heavy nucleus. The parallel ($A_{ll}$) and perpendicular ($A_{\perp}$) magnetic HFS constant values of $^{199}$Hg and $^{201}$Hg in HgH are presented in Table I. The experimental values are taken from Ref.\textsuperscript{27}, where Stowe et al measured the magnetic HFS constant of HgH trapped in neon and argon matrices at 4K by electron spin resonance study. The agreement of our calculated $A_{ll}$ and $A_{\perp}$ results with the experimental values shows that the wavefunction evaluated in $Z$-vector method is very accurate in the near nuclear region and thus it also shows the reliability of our calculated $E_{\text{eff}}$ and $W_s$ values. Further, we have calculated molecular-frame dipole moment ($\mu$) of the HgH molecule using same $Z$-vector method. The obtained ($\mu$) values are 0.25 D and 0.27 D in TZ and QZ basis, respectively. These results are also compiled in the same table and compared with the available experimental value.\textsuperscript{28} However, the experimental $\mu$ value of HgH reported in Ref.\textsuperscript{28} was measured with an unusual and indirect way and the value is also given without any experimental uncertainty.

In Table II, we present the $E_{\text{eff}}$ and $W_s$ values of our calculation. The $E_{\text{eff}}$ value of HgH obtained in QZ basis is 123.2 GV/cm. This result shows that HgH is one of those diatomic molecules which have the largest effective electric field. Previously, Kozlov calculated the $E_{\text{eff}}$ of HgH by using a semiempirical method and the value found to be 79 GV/cm. On the other hand, we have used an $ab$ initio ($Z$-vector method in the relativistic CCSD framework) method with sufficiently large basis sets (TZ and QZ) to calculate the properties of HgH which makes our calculated values more reliable. Our calculated $W_s$ value in QZ basis is 284.2 kHz. This large value of $W_s$ suggests that the S-PS interaction can contribute a significant amount to the permanent molecular EDM. These characteristics of HgH make it an important player in the field of eEDM search. The ratio (R) of $E_{\text{eff}}$ to $W_s$ is also calculated as this is a very important quantity to obtain the model independent limit of $d_e$ and $k_e$ as suggested by Dzuba et al\textsuperscript{11}. They suggest that this ratio would be constant for a particular heavy nucleus for the following reasons: (i) these types of property mainly depend on the core (near nuclear region) electronic wavefunction and in this short distance the one electron Dirac equation becomes identical for all single-electron states with the given angular momenta; (ii) the main contribution of these types of properties comes from the $s_{1/2}$-$p_{1/2}$ matrix elements and thus the many-body effects like core polarization has a very little effect on the ratio R. Our calculated values of R in QZ basis are 106.9 and 104.8 in units of $10^{18}$/e cm in the SCF and in the Z-vector calculations, respectively. These results support the previous argument as the correlation treatment changes the value of R only by 2 units. These values are very close to the value obtained by Dzuba et al (112.5 in the same unit)\textsuperscript{11} where they used an analytic expression to evaluate this ratio. By putting the value of R in Eq.8, we can get the
The difference of calculated values using 500 a.u. cutoff for virtual orbitals and using all virtual orbitals in correlation calculation are 1.7 GV/cm and 3.8 kHz for $E_{\text{eff}}$ and $W_s$, respectively. Therefore, if we use 500 a.u. as cutoff for the virtual orbitals, then the associated errors in both $E_{\text{eff}}$ and $W_s$ values are about 1.4%. The effect of higher order correlation terms can be estimated by comparing our CCSD results with CCSD with partial triples (CCSD(T)) or with full configuration interaction (FCI) calculations. These types of calculations are very expensive and beyond the scope of the present study. However, from our experience we can comment that the error associated with this effect will be within 3.5%. Although these three effects are intertwined in nature, assuming linearity, we estimate our results are correct within 5% uncertainty. It is worth to mention that our results are free from the error associated with the effect of core polarization since all the electrons are correlated in our calculation.

From Table I, II and III, we can see that the HFS constants, $E_{\text{eff}}$ and $W_s$ values are following the same trend and thus, we can comment that the calculated $E_{\text{eff}}$ and $W_s$ values are very accurate. The above results suggest that HgH can be a potential candidate for the future eEDM experiment. However, there are other factors that need to be considered for an eEDM experiment. The ground state of HgH is a $^2\Sigma$ state. It has no orbital angular momentum contribution to the magnetic moment and thus it cannot cancel the unpaired electron’s spin angular momentum contribution to the magnetic moment unlike PbF and ThO. For this reason the $^2\Sigma$ state of HgH has a higher $g$-factor compared to $^2\Pi_{1/2}$ state of PbF$^{39}$ or $^3\Delta_1$ state of ThO$^{30,31}$. Thus, it can give a significant magnetic noise in the spectroscopic eEDM experiment. Being a $^2\Sigma$ state, there are no $\Omega$-doublets$^{32}$ available for the ground state of HgH which can be used as a comagnetometer state$^{31,33,34}$ that can suppress some systematic errors though it is possible to find the sets of “internal comagnetometer” states with the proper combinations of different rotational states$^{35}$. Even though HgH has a high $E_{\text{eff}}$, one needs to fully polarize the molecule in an external laboratory electric field for the maximal utilization of $E_{\text{eff}}$. But the large rotational constant$^{36}$ and small molecular dipole moment$^{28}$ of HgH suggest that enormous amount of external electric field is required to fully polarize the HgH molecule.

There are three main possible sources of error associated with our calculation - (i) basis set incompleteness, (ii) cutoff used for virtual orbitals in the correlation calculation and (iii) higher order correlation effect. The error associated with the incompleteness of the basis set can be estimated by comparing TZ and QZ basis calculations. The difference between the results calculated in TZ and QZ basis for both $E_{\text{eff}}$ and $W_s$ is less than 0.1%. We have done a series of calculations (compiled in Table III) to estimate the error associated with the restriction of correlation space by neglecting higher energy virtual orbitals. In this calculation, the $E_{\text{eff}}$ and $W_s$ values are calculated by employing double zeta (DZ) basis (dyall.ev2z for Hg$^{24}$ and cc-pCVDZ for H$^{25}$) in the Z-vector method with different cutoff of virtual orbitals.

### Table I. Dipole Moment ($\mu$) (in Debye) and Magnetic HFS constants of HgH (in MHz)

| Basis  | $\mu$ | $A_{\parallel}$ | $A_{\perp}$ |
|--------|-------|----------------|-------------|
| TZ     | 0.25  | 8371           | 6483        |
| QZ     | 0.27  | 8440           | 6575        |
| Expt.  | 0.47  | 7780(5)$^a$    | 6200(3)$^b$ |

| $^{199}\text{Hg}$ | $^{201}\text{Hg}$ |
|-------------------|-------------------|
| $A_{\parallel}$   | $A_{\perp}$       |
| -3090             | -2392             |
| -3116             | -2427             |

$^a$measured in Ne matrices, $^b$measured in Ar matrices.

### Table II. $E_{\text{eff}}$ (in GV/cm), $W_s$ (in kHz) and the ratio of them ($R = E_{\text{eff}}/W_s$ in units of $10^{18}$ e/cm) of HgH.

| Basis  | $E_{\text{eff}}$ | $W_s$ | $R$ |
|--------|------------------|-------|-----|
| SCF    | Z-vector         | SCF   | Z-vector |
| TZ     | 106.8            | 123.3 | 241.2 | 284.3 | 107.1 | 104.9 |
| QZ     | 106.9            | 123.2 | 241.7 | 284.2 | 106.9 | 104.8 |

### Table III. Convergence pattern of $A_{\parallel}$ of $^{199}\text{Hg}$ in HgH, $\mu$, $E_{\text{eff}}$ and $W_s$ of HgH as a function of virtual orbitals

| Cutoff (a.u.) | Virtual spinor | $\mu$ | A$_{\parallel}$ | $E_{\text{eff}}$ | $W_s$ |
|---------------|----------------|------|----------------|-----------------|------|
| 100           | 175            | 0.168| 7911           | 115.6           | 260.5|
| 200           | 199            | 0.169| 7918           | 115.7           | 260.6|
| 400           | 207            | 0.167| 7977           | 116.8           | 263.0|
| 500           | 231            | 0.170| 7980           | 116.8           | 263.1|
| 1000          | 239            | 0.170| 8015           | 117.5           | 264.6|
| no cutoff     | 355            | 0.170| 8069           | 118.5           | 266.9|

Following relation

$$d_e + 4.77 \times 10^{-21} k_s = d_{e\text{exp}}^{\text{virtual orbitals}}$$

where $d_{e\text{exp}}^{\text{virtual orbitals}}$ is the eEDM limit derived from the P,T-odd energy shift of HgH experiment at the limit of $k_s = 0$.
On the contrary, as suggested by Kozlov et al\textsuperscript{15}, HgH can be polarized easily in the matrix isolated solid state non-spectroscopic experiment. They also argued that in this method, it is possible to improve the eEDM limit by 2-3 orders higher than the current limit. Therefore, considering these facts, we can comment that HgH can be a potential candidate for eEDM experiment and the solid state non-spectroscopic experimental technique would be best suitable for it.

V. CONCLUSION

In summary, we have performed the precise calculation of $E_{\text{eff}}$ and $W_z$ of HgH molecule in its open-shell ground state using the most reliable Z-vector method in the relativistic coupled-cluster framework. The outcome of our study reveals that HgH has one of the highest $E_{\text{eff}}$ and $W_z$ known for the polar diatomic molecule. On the other hand, HgH can be polarized easily using a solid state non-spectroscopic technique. Thus, the combination makes HgH a very important candidate for the next generation eEDM experiment.

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