

# Lorentz and *CPT* violation and the hydrogen and antihydrogen molecular ions. III. Rovibrational spectrum and the nonminimal standard model extension

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Rovibrational transitions in the hydrogen and antihydrogen molecular ions  $H_2^+$  and  $\bar{H}_2^-$  offer the possibility of testing Lorentz and *CPT* symmetry to extremely high precision, in principle attaining  $O(10^{-17})$ . In this paper, the third in a series, we give a comprehensive derivation of the rovibrational spectrum of  $H_2^+$  and  $\bar{H}_2^-$  in the standard model extension (SME), an effective quantum field theory incorporating Lorentz and *CPT* violation. New developments described here include a complete analysis of the molecular dynamics from first principles in terms of the spherical tensor representation of the SME couplings, the systematic extension of our previous results to the nonminimal SME, a full description of the quantum number dependence of the rovibrational energy levels in the spherical tensor formalism with both high and low background magnetic fields, and an extended discussion of sidereal and annual variations of transition frequencies arising from both rotations and Lorentz boosts. The resulting sensitivity of the rovibrational spectrum to an extended range of SME couplings, together with the ability to isolate their individual effects using the quantum number dependence of the transition frequencies, enhances the opportunities to detect Lorentz and *CPT* symmetry breaking through rovibrational spectroscopy of  $H_2^+$  and  $\bar{H}_2^-$ .

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## I. INTRODUCTION

Lorentz invariance and *CPT* symmetry are fundamental elements of the local relativistic quantum field theories that provide our modern understanding of elementary particle physics. It is therefore important to find ways to test them experimentally at the highest levels of precision [1]. Rovibrational spectroscopy of the hydrogen and antihydrogen molecular ions  $H_2^+$  and  $\bar{H}_2^-$  is an especially promising direction, offering the possibility of direct tests of Lorentz and *CPT* symmetry at precisions potentially of  $O(10^{-17})$  [2].

This paper is the third in a series [3,4] (referred to from now on as Papers 1 and 2) in which we study the rovibrational spectrum of  $H_2^+$  and  $\bar{H}_2^-$  in an effective theory of Lorentz and *CPT* violation, the standard model extension (SME) [5]. New developments here include a full derivation of the molecular dynamics from first principles in terms of

the spherical tensor representation of the SME couplings, the systematic extension of our previous results to the non-minimal SME [6], a full description of the quantum number dependence of the rovibrational energy levels in the spherical tensor formalism at both high and low background magnetic fields, and an extended discussion of sidereal and annual variations of transition frequencies arising from both rotations and Lorentz boosts.

Significant advances in experimental techniques have allowed rapid progress in the rovibrational spectroscopy of  $H_2^+$  in RF traps, with current measurements already achieving  $O(10^{-12})$  precision [7]. In parallel, recent developments toward transporting antiprotons away from the CERN AD/ELENA facility to a dedicated off-site spectroscopy laboratory, together with progress in understanding routes to synthesize  $\bar{H}_2^-$  [8–10] and the increasing precision achieved by the ALPHA collaboration in 1S-2S spectroscopy of atomic antihydrogen [11,12], are opening a path toward future rovibrational spectroscopy of  $\bar{H}_2^-$ . This would enable direct high-precision tests of *CPT* by comparison of the spectra of  $H_2^+$  and  $\bar{H}_2^-$  in a common experiment.

The SME Lagrangian [5] is parametrized by a set of Lorentz tensor couplings to operators formed from the standard model fields, in contrast to the scalar couplings of conventional Lorentz-invariant QFTs, *viz.*

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$$\begin{aligned} \mathcal{L}_{\text{SME}} = & \frac{1}{2} \sum_w \int d^4x \left[ \bar{\psi} (i\gamma^\mu \partial_\mu - m_w) \psi - a_\mu^w \bar{\psi} \gamma^\mu \psi + i c_{\mu\nu}^w \bar{\psi} \gamma^\mu \partial^\nu \psi + a_{\mu\nu\lambda}^w \bar{\psi} \gamma^\mu \partial^\nu \partial^\lambda \psi \right. \\ & \left. - b_\mu^w \bar{\psi} \gamma^5 \gamma^\mu \psi + i d_{\mu\nu}^w \bar{\psi} \gamma^5 \gamma^\mu \partial^\nu \psi - \frac{1}{2} H_{\mu\nu}^w \bar{\psi} \sigma^{\mu\nu} \psi + \frac{1}{2} i g_{\mu\nu\lambda}^w \bar{\psi} \sigma^{\mu\nu} \partial^\lambda \psi + \dots \right] + \text{H.c.} \end{aligned} \quad (1.1)$$

where  $w$  denotes the particle type, so here the sum is over the electron and the two protons in the molecular ion. The dots represent further couplings to higher-dimensional, nonrenormalizable operators, of which  $a_{\mu\nu\lambda}^w$  is the first example. The  $a$ ,  $b$  and  $g$  couplings also violate  $CPT$  symmetry, while  $c$ ,  $d$  and  $H$  are Lorentz violating but preserve  $CPT$ . It is often useful to think of these couplings as VEVs of vector and tensor fields, perhaps of some more fundamental theory.

To analyze atomic and molecular systems, we need the nonrelativistic Hamiltonian derived from this, and in this paper we use from the outset the very convenient formalism (see [6] for an extensive description) in which the couplings are represented as spherical tensors. This is especially well-suited to the symmetries of atoms and molecules and makes the extension to the nonminimal SME, including nonrenormalizable operators, particularly natural. This Hamiltonian, for para- $\text{H}_2^+$ , may be written as

$$\begin{aligned} H_{\text{SME}} = & - \sum_{njm} p_e^n Y_{jm}(\hat{\mathbf{p}}_e) \mathcal{V}_{njm}^{e\text{NR}} - \sum_{njm} p_1^n Y_{jm}(\hat{\mathbf{p}}_1) \mathcal{V}_{njm}^{p\text{NR}} - \sum_{njm} p_2^n Y_{jm}(\hat{\mathbf{p}}_2) \mathcal{V}_{njm}^{p\text{NR}} - \sigma_{\text{HEL}}^r \sum_{njm} p_e^n Y_{jm}(\hat{\mathbf{p}}_e) \mathcal{T}_{njm}^{e\text{NR}(0B)} \\ & + \sigma_{\text{HEL}}^\pm \sum_{njm} p_{e\pm 1}^n Y_{jm}(\hat{\mathbf{p}}) \left( \pm \mathcal{T}_{njm}^{e\text{NR}(1B)} + i \mathcal{T}_{njm}^{e\text{NR}(1E)} \right), \end{aligned} \quad (1.2)$$

where  $\sigma_{\text{HEL}}^{r,\pm}$  denote the components of the electron spin in the helicity frame (HEL) associated with its momentum. The notation follows that of [6]— $\sigma_{\text{HEL}}^r$  denotes the spin component along the momentum direction, while  $\sigma_{\text{HEL}}^\pm$  are the spin components in the transverse directions. See Ref. [6] for further details. Here,  ${}_s Y_{jm}$  are the spin-weighted spherical harmonics, defined in Sec. III. The dictionary relating the spherical and Lorentz tensor couplings is given in [6] and Paper 2, with further identities shown here where necessary.

The breaking of Lorentz invariance means that frames of reference play an important rôle in our analysis. First of these is the reference frame (MOL) aligned with the molecular axis. This is used to analyse the electron dynamics, which in the standard Born-Oppenheimer framework determines the inter-nucleon potential. A notable point here is that even in the electron  $1s\sigma_g$  ground state, the molecule only has cylindrical and not full spherical symmetry, which results in sensitivity to the electron spherical tensor couplings  $\mathcal{V}_{njm}^e$  and  $\mathcal{T}_{njm}^e$  with  $j \neq 0$ . Our results for the rovibrational energies themselves are then calculated in the EXP frame, with quantization axis aligned with the background magnetic field. An important rôle is also played by a solar-centred frame (SUN), which has become the standard in which to compare constraints on the SME couplings arising from different experiments [13]. This frame is also the basis for our discussion of transition frequency variations from rotations and Lorentz boosts.

In Papers 1 and 2 we have given an extensive description in the Born-Oppenheimer framework of how the

rovibrational energies are determined from the SME-modified internucleon potential  $V_M(R) + V_{\text{SME}}^e(R, \mathbf{S})$ , described in Sec. II, together with the direct nucleon (proton) contribution  $\Delta E_{\text{SME}}^n$ . The rovibrational energy spectrum is expressed as an expansion in powers of  $(v + \frac{1}{2})$  and  $N(N+1)$  as follows:

$$\begin{aligned} E_{vNM_J} = & V_{\text{SME}}^e + (1 + \delta_{\text{SME}}^e + \delta_{\text{SME}}^n) \left( v + \frac{1}{2} \right) \omega_0 \\ & - (x_0 + x_{\text{SME}}^e + x_{\text{SME}}^n) \left( v + \frac{1}{2} \right)^2 \omega_0 \\ & + (B_0 + B_{\text{SME}}^e + B_{\text{SME}}^n) N(N+1) \omega_0 \\ & - (\alpha_0 + \alpha_{\text{SME}}^e + \alpha_{\text{SME}}^n) \left( v + \frac{1}{2} \right) N(N+1) \omega_0 + \dots \end{aligned} \quad (1.3)$$

where  $\omega_0$  is the fundamental vibration frequency. Each coefficient of  $\omega_0$  is an expansion in powers of a small parameter  $\lambda$ , parametrically of  $O(\sqrt{m_e/m_p})$ , with the leading terms in the coefficients  $x_0$ ,  $B_0$ ,  $\alpha_0$  being of order  $\lambda$ ,  $\lambda$ ,  $\lambda^2$ , respectively. In the rest of this paper, we calculate these coefficients in terms of the spherical tensor couplings as perturbations on the hyperfine-Zeeman energy levels for para- $\text{H}_2^+$ , determining their dependence on the rovibrational and spin quantum numbers and their magnetic field dependence for both high and low fields. This means our results may be applied directly both to current measurements with  $\text{H}_2^+$  in low-field RF traps [7] and future

experiments with  $\bar{\text{H}}_2^-$ , and  $\text{H}_2^+$ , confined in Penning traps [14].

A new feature in this work is the extended discussion of daily (sidereal) variations due to the Earth's rotation, for which spherical tensor formalism is especially well suited, and sidereal and annual variations due to Lorentz boosts with both the Earth's rotational and orbital velocities. Here, the spherical tensor formalism is not in itself sufficient and we have to calculate directly with the Lorentz tensor couplings, but here we exhibit the dictionary to translate these variations back in terms of the spherical tensor couplings. This reveals a sensitivity to further SME couplings with different values of both  $n$  and  $j$  quantum numbers, emphasizing the need to exploit these variations and not rely purely on time-averaged measurements.

One of the initial objectives of this paper was to bring together the approach of Papers 1 and 2 with the work [15] (see also [16]). This reference addresses the same topics, though in less generality and keeping only some of the leading terms in the rovibrational energies. This means that the sensitivity to several SME couplings is not present in the results quoted there. On the other hand, [15] provides detailed explicit formulas for the frequency and rotational variations of a specific experimentally important rovibrational transition in  $\text{H}_2^+$ , which we have checked are in agreement with the general results given here. Other earlier work on rovibrational spectroscopy and the SME includes [17] and [18], the latter having particularly influenced Paper 1.

The paper is organized as follows. Section II gives a brief review of the modifications to the Born-Oppenheimer approximation applied to the SME. Our main analysis of the internucleon potential and rovibrational energy levels is given in Secs. III A, III B and IV, for the electron spin-independent and spin-dependent couplings and the proton couplings respectively. These results are quoted in the EXP frame, and translated to the SUN frame in Sec. V, where transition frequency variations due to rotations and Lorentz boosts are described. Section VI presents a brief summary and outlook on future work, while three Appendixes provide further technical details of our calculations.

## II. BORN-OPPENHEIMER ANALYSIS IN THE SME

We begin with a brief recap of the Born-Oppenheimer analysis incorporating Lorentz and CPT violation described in Papers 1 and 2. In this approximation, the full Schrödinger equation is factorized into an “electron Schrödinger equation,” which determines the inter-nucleon potential  $V_M(R)$ , and a “nucleon Schrödinger equation” from which we find the rovibrational energy eigenstates. We restrict here to the experimentally relevant para- $\text{H}_2^+$ , where the total nucleon spin is 0 and the rotational angular

momentum quantum number  $N$  is even. As always the electron is taken in the  $1s\sigma_g$  ground state.

Factorizing the full wave function into electron and nucleon parts as<sup>1</sup>

$$\Psi(\mathbf{R}, \mathbf{r}; M_S) = \Phi(\mathbf{R})\psi(\mathbf{r}; R)|M_S\rangle, \quad (2.1)$$

the electron Schrödinger equation including the SME Hamiltonian is

$$\left[ -\frac{1}{2\hat{\mu}}\nabla_{\mathbf{r}}^2 + V_{\text{mol}}(R, r_{1e}, r_{2e}) + \tilde{H}_{\text{SME}}^e(\mathbf{p}; \mathbf{S}) \right] \psi(\mathbf{r}; R)|M_S\rangle = E_e(\mathbf{R}, \mathbf{S})\psi(\mathbf{r}; R)|M_S\rangle, \quad (2.2)$$

where we understand  $\mathbf{p} \rightarrow -i\nabla_{\mathbf{r}}$ , and  $V_{\text{mol}}(R, r_{1e}, r_{2e})$  is the electrostatic potential binding the molecule. Here, the electron SME Hamiltonian  $\tilde{H}_{\text{SME}}^e(\mathbf{p}; \mathbf{S})$  also contains a contribution from the spin-independent proton SME couplings, as explained in Papers 1 and 2 and here in Sec. IV. This effect is included in the full internucleon potential by the substitution given in (4.10).

The eigenvalues define the internucleon potential,

$$E_e(\mathbf{R}, \mathbf{S}) = V_M(R) + V_{\text{SME}}^e(\mathbf{R}, \mathbf{S}), \quad (2.3)$$

where

$$V_{\text{SME}}^e(\mathbf{R}, \mathbf{S}) = \int d^3\mathbf{r} \psi^*(\mathbf{r}; R) \tilde{H}_{\text{SME}}^e(\mathbf{p}; \mathbf{S}) \psi(\mathbf{r}; R), \quad (2.4)$$

is the expectation value of  $\tilde{H}_{\text{SME}}^e(\mathbf{p}; \mathbf{S})$  in the electron  $1s\sigma_g$  state. It depends on the expectation values  $\langle p_a p_b \rangle$  of the electron evaluated in the MOL frame.

Inserting into the nucleon Schrödinger equation this gives

$$\begin{aligned} & \left[ -\frac{1}{2\mu}\nabla_{\mathbf{R}}^2 + V_M(R) + V_{\text{SME}}^e(\mathbf{R}; \mathbf{S}) + H_{\text{SME}}^p(\mathbf{P}) \right] \\ & \times \Phi_{vNM_N}(\mathbf{R})|M_S\rangle \\ & = E_{vNM_N M_S} \Phi_{vNM_N}(\mathbf{R})|M_S\rangle, \end{aligned} \quad (2.5)$$

with  $\mathbf{P} = -i\nabla_{\mathbf{R}}$ . Here,

$$E_{vNM_N M_S} = \tilde{E}_{vNM_N M_S} + \Delta E_{\text{SME}}^n, \quad (2.6)$$

with  $\tilde{E}_{vNM_N M_S}$  found by setting  $\Phi_{vNM_N}(\mathbf{R}) = \frac{1}{R} \phi_{vNM_N}(R) Y_{NM_N}(\theta, \phi)$  and solving the 1-dim rovibrational equation

<sup>1</sup>For kinematics and notation, see Paper 1 and Sec. IV here.

$$\left[ -\frac{1}{2\mu} \frac{d^2}{dR^2} + \frac{1}{2\mu R^2} N(N+1) + V_M(R) + V_{\text{SME}}^e(\mathbf{R}; \mathbf{S}) \right] \times \phi_{vNM_N}(R) |M_S\rangle = \tilde{E}_{vNM_N M_S} \phi_{vNM_N}(R) |M_S\rangle, \quad (2.7)$$

where,

$$V_{\text{SME}}^e(\mathbf{R}; \mathbf{S}) = \langle NM_N | V_{\text{SME}}^e(\mathbf{R}; \mathbf{S}) | NM_N \rangle, \quad (2.8)$$

together with,

$$\Delta E_{\text{SME}}^n = \langle v N M_N | H_{\text{SME}}^p | v N M_N \rangle. \quad (2.9)$$

In the following two Sections, we evaluate the SME contribution  $V_{\text{SME}}^e(R)$  to the internucleon potential in the hyperfine-Zeeman states and find the corresponding shifts in the rovibrational energy levels, then construct the direct proton contribution  $\Delta E_{\text{SME}}^n$ .

### III. ELECTRON SME COUPLINGS AND THE INTERNUCLEON POTENTIAL

We now consider the construction of  $V_{\text{SME}}^e(\mathbf{R})$ , the electron contribution to the inter-nucleon potential. (We suppress the explicit dependence on the spin label from now on for convenience and to match the notation in Paper 2.) We start here with the spin-independent couplings  $\mathcal{V}_{njm}$ , leaving the spin-dependent  $T_{njm}^{(0B)}$ ,  $T_{njm}^{(1B)}$  and  $T_{njm}^{(1E)}$  to the following Subsection.

#### A. Electron spin-independent SME couplings

The results at  $O(p^2)$  have already been derived using the couplings  $c_{\mu\nu}$  and  $a_{\mu\nu\lambda}$  appearing in the original SME Lagrangian. Here, we instead start from the relativistic expansion of the Hamiltonian expressed in terms of spherical tensor couplings and extend the discussion to include the leading terms arising at  $O(p^4)$  in the non-minimal SME. In this expansion, the SME Hamiltonian for the electron is written in terms of nonrelativistic, spin-independent couplings  $\mathcal{V}_{njm}^{\text{NR}}$  as

$$H_{\text{SME}}^{\mathcal{V}} = - \sum_{njm} p^n Y_{jm}(\hat{\mathbf{p}}) \mathcal{V}_{njm}^{\text{NR}}, \quad (3.1)$$

with  $n$  summed over  $0, 2, 4, \dots$ . The couplings  $\mathcal{V}_{njm}^{\text{NR}}$  are related in a nontrivial way to the original  $\mathcal{V}_{njm}^e$ , the relations being given explicitly in [6]. These are especially important below when we consider various critical identities amongst the nonrelativistic couplings. Also recall that the couplings may be separated into *CPT* even ( $c_{njm}^{\text{NR}}$ ) and odd ( $a_{njm}^{\text{NR}}$ ) components as follows:

$$\mathcal{V}_{njm}^{\text{NR}} = c_{njm}^{\text{NR}} - a_{njm}^{\text{NR}}. \quad (3.2)$$

In this form, the Hamiltonian  $H_{\text{SME}}^e$  is independent of the frame, so we may consider it first in the MOL frame, denoting the relevant couplings by  $\hat{\mathcal{V}}_{njm}^{\text{NR}}$ . The momentum expectation values are then evaluated as in Paper 1 with the familiar  $1s\sigma_g$  electron wave functions.

It is very convenient in working with the spherical tensors to introduce the Wigner matrices  $d_{m'm}^j(\theta)$ , in terms of which the spin-weighted spherical harmonics are written as

$${}_s Y_{jm}(\theta, \phi) = \sqrt{\frac{2j+1}{4\pi}} d_{ms}^j(\theta) e^{im\phi}, \quad (3.3)$$

with the usual spherical harmonics corresponding to  $s = 0$ . The necessary properties of the  $d_{m'm}^j(\theta)$  in our conventions are given in Sec. VI and Appendix A of Paper 2.

Using the explicit expressions for  $d_{m0}^2$  and  $d_{m0}^4$ , and imposing cylindrical symmetry on the momentum expectation values, we find in the MOL frame,

$$\begin{aligned} V_{\text{SME}}^{\mathcal{V}}(\mathbf{R}) = & -\frac{1}{\sqrt{4\pi}} \left[ \hat{\mathcal{V}}_{000}^{\text{NR}} + \langle p^2 \rangle \hat{\mathcal{V}}_{200}^{\text{NR}} \right. \\ & - \frac{1}{2} \sqrt{5} \langle (p^2 - 3p_z^2) \rangle \hat{\mathcal{V}}_{220}^{\text{NR}} \\ & + \langle p^4 \rangle \hat{\mathcal{V}}_{400}^{\text{NR}} - \frac{1}{2} \sqrt{5} \langle (p^4 - 3p^2 p_z^2) \rangle \hat{\mathcal{V}}_{420}^{\text{NR}} \\ & \left. + \frac{3}{8} \langle (3p^4 - 30p^2 p_z^2 + 35p_z^4) \rangle \hat{\mathcal{V}}_{440}^{\text{NR}} + \dots \right]. \end{aligned} \quad (3.4)$$

Note that in our previous work, we generally used the notation  $\langle p^2 \rangle = \text{tr} \langle p_a p_b \rangle$  and  $\langle (p^2 - 3p_z^2) \rangle = \text{tr}_Y \langle p_a p_b \rangle$ .

The cylindrical symmetry of the molecule enforces  $m = 0$  here, since the momentum expectation values corresponding to  $m \neq 0$  necessarily vanish. However, the absence of spherical symmetry does allow  $V_{\text{SME}}^{\mathcal{V}}(\mathbf{R})$  to have contributions from the couplings with  $j \neq 0$ , to this order  $\hat{\mathcal{V}}_{220}^{\text{NR}}$ ,  $\hat{\mathcal{V}}_{420}^{\text{NR}}$  and  $\hat{\mathcal{V}}_{440}^{\text{NR}}$ .

In our previous work (Paper 1), we have evaluated  $\text{tr} \langle p_a p_b \rangle$  and  $\text{tr}_Y \langle p_a p_b \rangle$  as functions of  $R$  and found their derivatives at the minimum  $R_0$  of the internucleon potential, as required to evaluate the rovibrational energies in the form (1.3). The nonzero value of  $\text{tr}_Y \langle p_a p_b \rangle$  arises because the molecule only has cylindrical and not full spherical symmetry. It is smaller, but of the same magnitude as  $\text{tr} \langle p_a p_b \rangle$  and cannot be neglected. For example, evaluated at  $R_0$ ,  $\text{tr} \langle p_a p_b \rangle = 1.173$  while  $\text{tr}_Y \langle p_a p_b \rangle = 0.360$  in units of the Bohr radius squared.

In the next step we rewrite the SME couplings in the EXP frame, with the quantization ( $z$ ) axis aligned with the

background magnetic field  $\mathbf{B}$ . The transformation is given in terms of the Wigner matrices by

$$\hat{V}_{njm'} = \hat{V}_{njm} d_{mm'}^j(\theta) e^{im\phi}, \quad (3.5)$$

where  $(\theta, \phi)$  are the standard spherical polar angles specifying the orientation of the MOL axes in the EXP frame.

It follows immediately that

$$\begin{aligned} V_{\text{SME}}^e \mathcal{V}(\mathbf{R}) = & - \left[ \frac{1}{\sqrt{4\pi}} \mathcal{V}_{000}^{\text{eNR}} + \frac{1}{\sqrt{4\pi}} \text{tr}\langle p_a p_b \rangle \mathcal{V}_{200}^{\text{eNR}} \right. \\ & - \frac{1}{2} \text{tr}_Y \langle p_a p_b \rangle \mathcal{V}_{22m}^{\text{eNR}} Y_{2m}(\theta, \phi) + \frac{1}{\sqrt{4\pi}} \langle p^4 \rangle \mathcal{V}_{400}^{\text{eNR}} \\ & - \frac{1}{2} \langle (p^4 - 3p^2 p_z^2) \rangle \mathcal{V}_{42m}^{\text{eNR}} Y_{2m}(\theta, \phi) \\ & \left. + \frac{1}{8} \langle (3p^4 - 30p^2 p_z^2 + 35p_z^4) \rangle \mathcal{V}_{44m}^{\text{eNR}} Y_{4m}(\theta, \phi) \right]. \end{aligned} \quad (3.6)$$

Note that  $\mathcal{V}_{000}^{\text{eNR}}$  is usually neglected on the grounds that it is unobservable.

Following the discussion above, the next step is to take the expectation values of the spherical harmonics in the angular momentum states  $|NM_N\rangle$  to find  $V_{\text{SME}}^e \mathcal{V}(R)$  from (2.8). In terms of Clebsch-Gordan coefficients,

$$\begin{aligned} \langle NM_N | Y_{2m}(\theta, \phi) | NM_N \rangle &= \sqrt{\frac{5}{4\pi}} C_{NM_N, 20}^{NM_N} C_{N0, 20}^{N0} \delta_{m0} \\ &\equiv \sqrt{\frac{5}{4\pi}} c_{NM_N}, \end{aligned} \quad (3.7)$$

where,

$$c_{NM_N} = \frac{N(N+1) - 3M_N^2}{(2N-1)(2N+3)}. \quad (3.8)$$

This notation was used in our previous work. Here, we also need the extension to  $j = 4$ ,

$$\begin{aligned} \langle NM_N | Y_{4m}(\theta, \phi) | NM_N \rangle &= \frac{3}{\sqrt{4\pi}} C_{NM_N, 40}^{NM_N} C_{N0, 40}^{N0} \delta_{m0} \\ &\equiv \frac{3}{\sqrt{4\pi}} c_{NM_N}^{(4)}, \end{aligned} \quad (3.9)$$

with

$$c_{NM_N}^{(4)} = \frac{3}{4} \frac{[3N(N+2)(N+1)(N-1) - 5(6N^2 + 6N - 5)M_N^2 + 35M_N^4]}{[(2N+5)(2N+3)(2N-1)(2N-3)]}. \quad (3.10)$$

The special case  $N = 2$  is of current experimental interest, so we note here the special cases<sup>2</sup>

$$c_{2M_N} = \frac{1}{7} (2 - M_N^2), \quad (3.11)$$

and

$$c_{2M_N}^{(4)} = \frac{1}{252} (72 - 155M_N^2 + 35M_N^4). \quad (3.12)$$

These results are directly applicable in the strong magnetic field limit, where  $|v N M_J M_S\rangle$  are the appropriate ‘‘Zeeman’’ eigenstates. Recall that  $M_J = M_N + M_S$  is a good quantum number for all magnetic fields as the operator  $J_z$  commutes with the full hyperfine-Zeeman Hamiltonian. This is the case of most interest for  $\bar{\text{H}}_2^-$  where it is envisaged spectroscopy will be carried out in Penning traps [14]. In this limit, we find  $V_{\text{SME}}^e \mathcal{V}(R)$  evaluated in the states  $M_J, M_S = \pm \frac{1}{2}$  is

$$\begin{aligned} V_{\text{SME}\pm}^e \mathcal{V}(R) = & - \frac{1}{\sqrt{4\pi}} \left[ \mathcal{V}_{000}^{\text{eNR}} + \text{tr}\langle p_a p_b \rangle \mathcal{V}_{200}^{\text{eNR}} - \frac{\sqrt{5}}{2} \text{tr}_Y \langle p_a p_b \rangle c_{NM_J \mp \frac{1}{2}} \mathcal{V}_{220}^{\text{eNR}} \right. \\ & + \langle p^4 \rangle \mathcal{V}_{400}^{\text{eNR}} - \frac{\sqrt{5}}{2} \langle (p^4 - 3p^2 p_z^2) \rangle c_{NM_J \mp \frac{1}{2}} \mathcal{V}_{420}^{\text{eNR}} + \frac{3}{8} \langle (3p^4 - 30p^2 p_z^2 + 35p_z^4) \rangle c_{NM_J \mp \frac{1}{2}}^{(4)} \mathcal{V}_{440}^{\text{eNR}} \left. \right]. \end{aligned} \quad (3.13)$$

For large, but finite,  $B$  fields, the coefficients acquire corrections, but only of  $O(1/B)^2$ ; these are calculated in Appendix A.

<sup>2</sup>These expressions appear in [15], Eq. (27), in the discussion of proton couplings.

On the other hand, precision spectroscopy of  $\text{H}_2^+$  is already being carried out in RF traps at extremely small magnetic fields [7] where, as discussed in Paper 2, the relevant “hyperfine” eigenstates are  $|vNJM_J\rangle$  where  $\mathbf{J} = \mathbf{N} + \mathbf{S}$  is the total molecular angular momentum. Here,

$$|vNJM_J\rangle = \sum_{M_S} C_{NM_N, \frac{1}{2}M_S}^{JM_J} |vNM_N M_S\rangle, \quad (3.14)$$

with  $J = N \pm \frac{1}{2}$  and  $M_J = -J, \dots, J-1, J$  as usual. We therefore need,

$$V_{\text{SME}}^e \mathcal{V}(R) = \langle vNJ'M_J | V_{\text{SME}}^e(\mathbf{R}) | vNJM_J \rangle, \quad (3.15)$$

where, for each  $M_J$ , this is a  $2 \times 2$  matrix with rows/columns signifying  $J', J = N \pm \frac{1}{2}$ .

At this point, we refer to Paper 2 and Appendix A, which describe the hyperfine-Zeeman Hamiltonian and its diagonalization for nonvanishing magnetic fields, in order to find the SME perturbed energy eigenvalues. The eigenvalues may be written as

$$V_{\text{SME}\pm}^e \mathcal{V}(R) = -\frac{1}{\sqrt{4\pi}} \left[ \mathcal{V}_{000}^{\text{eNR}} + \text{tr}\langle p_a p_b \rangle \mathcal{V}_{200}^{\text{eNR}} - \frac{\sqrt{5}}{2} \text{tr}_Y \langle p_a p_b \rangle \hat{c}_{NM_J}^{\pm}(B) \mathcal{V}_{220}^{\text{eNR}} \right. \\ \left. + \langle p^4 \rangle \mathcal{V}_{400}^{\text{eNR}} - \frac{\sqrt{5}}{2} \langle (p^4 - 3p^2 p_z^2) \rangle \hat{c}_{NM_J}^{\pm}(B) \mathcal{V}_{420}^{\text{eNR}} + \frac{3}{8} \langle (3p^4 - 30p^2 p_z^2 + 35p_z^4) \rangle \hat{c}_{NM_J}^{(4)\pm}(B) \mathcal{V}_{440}^{\text{eNR}} \right] \quad (3.16)$$

with the coefficients,

$$\hat{c}_{NM_J}^+(B) = \frac{1}{(2N+1)(2N+3)} \left[ \left( N + \frac{1}{2} \right) \left( N + \frac{3}{2} \right) - 3M_J^2 \right] + 24 \frac{[(N + \frac{1}{2})^2 - M_J^2] M_J}{(2N+1)^3 (2N-1)(2N+3)} \mathcal{B}, \quad (3.17)$$

and

$$\hat{c}_{NM_J}^-(B) = \frac{1}{(2N-1)(2N+1)} \left[ \left( N - \frac{1}{2} \right) \left( N + \frac{1}{2} \right) - 3M_J^2 \right] - 24 \frac{[(N + \frac{1}{2})^2 - M_J^2] M_J}{(2N+1)^3 (2N-1)(2N+3)} \mathcal{B}. \quad (3.18)$$

Here, we have introduced the notation

$$\mathcal{B} = \frac{1}{c_e(v, N)} (g_e + g_m(v, N)) \mu_B B \quad (3.19)$$

for the expansion parameter, where  $c_e(v, N), g_e, g_m(v, N)$  are coefficients in the hyperfine-Zeeman Hamiltonian.

In  $O(p^4)$ , citing only the terms independent of  $B$  for simplicity, we find

$$\hat{c}_{NM_J}^{(4)+} = \frac{3}{64} \frac{3(2N+5)(2N+3)(2N+1)(2N-1) - 40(12N^2 + 24N - 1)M_J^2 + 560M_J^4}{(2N+5)(2N+3)(2N-1)(2N-3)} \quad (3.20)$$

and

$$\hat{c}_{NM_J}^{(4)-} = \frac{3}{64} \frac{3(2N+3)(2N+1)(2N-1)(2N-3) - 40(12N^2 - 13)M_J^2 + 560M_J^4}{(2N+3)(2N+1)(2N-1)(2N-3)}, \quad (3.21)$$

using results from Appendix B.

In the presently relevant experimental case with  $N = 2$ , these expressions simplify at zero magnetic field to<sup>3</sup>

<sup>3</sup>For  $N = 2$ , these factors have been given in [15], Eq. (11). To compare, the dictionary for our notations is,

$$\alpha_{\frac{3}{2}M_F}^{(2)} = \sqrt{\frac{5}{4\pi}} \hat{c}_{2M_J}^-, \quad \alpha_{\frac{5}{2}M_F}^{(2)} = \sqrt{\frac{5}{4\pi}} \hat{c}_{2M_J}^+, \quad \alpha_{\frac{3}{2}M_F}^{(4)} = \frac{3}{\sqrt{4\pi}} \hat{c}_{2M_J}^{(4)-}, \quad \alpha_{\frac{5}{2}M_F}^{(4)} = \frac{3}{\sqrt{4\pi}} \hat{c}_{2M_J}^{(4)+}.$$

Note that  $\hat{c}_{2M_J}^{(4)-}$  vanishes for both allowed values  $M_J = 1/2$  or  $3/2$ .

$$\begin{aligned}
\hat{c}_{2M_J}^+ &= \frac{1}{140}(35 - 12M_J^2), \\
\hat{c}_{2M_J}^- &= \frac{1}{20}(5 - 4M_J^2), \\
\delta_{\text{SME}}^e &= \frac{1}{2} \frac{1}{V_M''} \left[ V_{\text{SME}}^{e''} - \frac{V_M'''}{V_M''} V_{\text{SME}}^{e'} \right], \\
B_{\text{SME}}^e &= \lambda \frac{1}{V_M''} \left[ \frac{1}{R_0} V_{\text{SME}}^{e'} \right],
\end{aligned} \tag{3.22}$$

and

$$\begin{aligned}
\hat{c}_{2M_J}^{(4)+} &= \frac{1}{432}(567 - 760M_J^2 + 112M_J^4), \\
\hat{c}_{2M_J}^{(4)-} &= \frac{1}{64}(9 - 40M_J^2 + 16M_J^4).
\end{aligned} \tag{3.23}$$

The formulas (3.13) and (3.16) for  $V_{\text{SME}\pm}^{e\mathcal{V}}(R)$  form the starting point to express the rovibrational energies as a function of its derivatives with respect to  $R$ , as shown in Paper 1. Keeping terms up to  $O(\lambda^2)$ , we have the SME contributions,

$$\begin{aligned}
E_{vNM_J\pm}^{e\mathcal{V}} &= V_{\text{SME}\pm}^{e\mathcal{V}} + \delta_{\text{SME}\pm}^{e\mathcal{V}} \left( v + \frac{1}{2} \right) \omega_0 - x_{\text{SME}\pm}^{e\mathcal{V}} \left( v + \frac{1}{2} \right)^2 \omega_0 \\
&\quad + B_{\text{SME}\pm}^{e\mathcal{V}} N(N+1) \omega_0 - \alpha_{\text{SME}\pm}^{e\mathcal{V}} \left( v + \frac{1}{2} \right) \\
&\quad \times N(N+1) \omega_0 + \dots
\end{aligned} \tag{3.24}$$

where [with all derivatives evaluated at the minimum  $R_0$  of the original internucleon potential  $V_M(R)$ ],

and similar, much lengthier, expressions for  $\alpha_{\text{SME}}^e$  and  $x_{\text{SME}}^e$  (see Paper 1).

To complete the determination of the contributions to the rovibrational energies from the SME, we need the expectation values of the momenta in (3.13), (3.16) and their  $R$  derivatives. This calculation was carried out in detail for the terms of  $O(p^2)$  in Paper 1, Appendix A, and numerical results were presented there. This can be repeated in exactly the same way for the  $O(p^4)$  expectation values. However, at this point in the experimental program it is probably excessive to present these here, so we content ourselves with noting that compared to the  $O(p^2)$  expectation values, they will be parametrically suppressed by  $O(1/a_0^2) = O(\alpha^2 m_e^2)$ , where  $a_0$  is the Bohr radius. Their importance therefore depends on the relative magnitude of the  $n = 4$  SME couplings  $\mathcal{V}_{4j0}^{e\text{NR}}$  compared to the  $n = 2$  couplings  $\mathcal{V}_{2j0}^{e\text{NR}}$ , which in this context should be viewed as *a priori* unknown.

Showing just the terms up to  $O(p^2)$ , we find from (3.16) for weak magnetic fields,<sup>4</sup>

$$\begin{aligned}
V_{\text{SME}\pm}^{e\mathcal{V}} &= \frac{1}{\sqrt{4\pi}} \left[ -\mathcal{V}_{000}^{e\text{NR}} - 2.346m_e \mathcal{V}_{200}^{e\text{NR}} + 0.360\sqrt{5}\hat{c}_{NM_J}^\pm(B)m_e \mathcal{V}_{220}^{e\text{NR}} + \dots \right], \\
\delta_{\text{SME}\pm}^{e\mathcal{V}} &= \frac{1}{\sqrt{4\pi}} \left[ 3.009m_e \mathcal{V}_{200}^{e\text{NR}} - 0.817\sqrt{5}\hat{c}_{NM_J}^\pm(B)m_e \mathcal{V}_{220}^{e\text{NR}} + \dots \right], \\
B_{\text{SME}\pm}^{e\mathcal{V}} &= \lambda \frac{1}{\sqrt{4\pi}} \left[ 2.002m_e \mathcal{V}_{200}^{e\text{NR}} - 0.336\sqrt{5}\hat{c}_{NM_J}^\pm(B)m_e \mathcal{V}_{220}^{e\text{NR}} + \dots \right], \\
x_{\text{SME}\pm}^{e\mathcal{V}} &= \lambda \frac{1}{\sqrt{4\pi}} \left[ 4.887m_e \mathcal{V}_{200}^{e\text{NR}} - 0.195\sqrt{5}\hat{c}_{NM_J}^\pm(B)m_e \mathcal{V}_{220}^{e\text{NR}} + \dots \right], \\
\alpha_{\text{SME}\pm}^{e\mathcal{V}} &= \lambda^2 \frac{1}{\sqrt{4\pi}} \left[ 6.456m_e \mathcal{V}_{200}^{e\text{NR}} - 0.285\sqrt{5}\hat{c}_{NM_J}^\pm(B)m_e \mathcal{V}_{220}^{e\text{NR}} + \dots \right],
\end{aligned} \tag{3.26}$$

with analogous results for strong fields from (3.13) with  $\hat{c}_{NM_J}^\pm \rightarrow c_{NM_J\mp\frac{1}{2}}^\pm$ . Note that  $m_e \mathcal{V}_{2jm}^{e\text{NR}}$  and the coefficients  $\delta_{\text{SME}\pm}^{e\mathcal{V}}, \dots$  are dimensionless. We are using atomic units here (see Paper 1) in which all energies are given in units of the Rydberg energy  $R_H$ .

<sup>4</sup>There is a significant sign error in the analogous expressions in Paper 1. This originates in Eq. (4.23) where the sign of the coefficient of the  $\text{tr}_Y \tilde{E}_{ij}^e$  term should be reversed. This is propagated through Sec. VI, especially (6.8), which should be compared with the corresponding expressions here, and Sec. 7, especially (7.1), where the same sign error affects the coefficient of all terms with  $(c_{220}^{\text{NR}e} - a_{220}^{\text{NR}e})$ . This sign error is repeated in Paper 2 in the numerical results in Eqs. (7.6) and (7.10), although all algebraic expressions in this paper are correct. These are now corrected in the arXiv versions of the papers.

### B. Electron spin-dependent couplings

We now come to the electron spin-dependent couplings. The nonrelativistic SME Hamiltonian is in this case [6],

$$H_{\text{SME}}^e = -\sigma_{\text{HEL}}^r \sum_{njm} p^n Y_{jm}(\hat{\mathbf{p}}) \mathcal{T}_{njm}^{e\text{NR}(0B)} + \sigma_{\text{HEL}}^{\pm} \sum_{njm} p_{\pm 1}^n Y_{jm}(\hat{\mathbf{p}}) \left( \pm \mathcal{T}_{njm}^{e\text{NR}(1B)} + i \mathcal{T}_{njm}^{e\text{NR}(1E)} \right), \quad (3.27)$$

where the spins are given in the helicity frame (HEL) associated with the electron momentum. The  $\mathcal{T}_{njm}^{\text{NR}}$  are combinations of the equivalent *CPT* violating and conserving couplings,

$$\mathcal{T}_{njm}^{\text{NR}} = g_{njm}^{\text{NR}} - H_{njm}^{\text{NR}}, \quad (3.28)$$

respectively, for each of the (0B), (1B) and (1E) type couplings. We only require [6] those with  $n = 0, 2, 4, \dots$  and  $j = (n+1), (n-1), \dots \geq 0$  for the (0B) couplings [ $\geq 1$  for (1B)] and  $j = n, (n-2), \dots \geq 1$  for the (1E) type.

Now, while the couplings in the full relativistic theory are independent, those in the nonrelativistic approximation (3.27) are not, but rather satisfy some identities that will be important in our analysis. Specifically, we can show<sup>5</sup>

$$\mathcal{T}_{01m}^{\text{NR}(1B)} = \mathcal{T}_{01m}^{\text{NR}(0B)}, \quad (3.29)$$

and

$$\mathcal{T}_{23m}^{\text{NR}(1B)} = \sqrt{\frac{2}{3}} \mathcal{T}_{23m}^{\text{NR}(0B)}. \quad (3.30)$$

Note that these identities are not restricted to the minimal SME.

The first step is to rewrite the Hamiltonian (3.27) entirely in the MOL frame. This means transforming the spin

<sup>5</sup>From the relations (112) in [6], we can reexpress the following special cases of the nonrelativistic couplings  $g_{njm}^{\text{NR}}$  in terms of the fully relativistic  $g_{njm}^{(d)}$  as follows ( $d$  is the dimension of the operator),

$$g_{01m}^{\text{NR}(0B)} = \sum_{d, \text{even}} m_w^{d-3} g_{01m}^{(d)(0B)} = g_{01m}^{\text{NR}(1B)},$$

$$g_{23m}^{\text{NR}(0B)} = 3 \sum_{d, \text{even}} m_w^{d-5} g_{23m}^{(d)(0B)} = \sqrt{\frac{3}{2}} g_{23m}^{\text{NR}(1B)},$$

after a short calculation, reflecting the fact that the putative couplings  $g_{01m}^{(d)(1B)}$  and  $g_{23m}^{(d)(1B)}$  are not permitted by the restrictions  $n \geq 2$ ,  $j = (n-1), (n-3), \dots \geq 1$  on the relativistic couplings  $g_{njm}^{(d)(1B)}$ . See [6], Table III. Similar results hold for the  $H_{njm}^{\text{NR}}$ . The particle-type label  $w$  on the couplings is omitted here for clarity.

variables from the original helicity frame as presented in [6]. The transformations are given in terms of the Wigner matrices by

$$\sigma_{s'}^{\text{HEL}} = \hat{\sigma}_s d_{ss'}^1(\theta) e^{-is\phi}, \quad (3.31)$$

where  $\hat{\sigma}$  denotes the MOL frame variables and  $(\theta, \phi)$  the direction of the electron momentum in the MOL frame. Together with the definition (3.3) of the spin-weighted spherical harmonics, we can rewrite (3.27) in the MOL frame as

$$H_{\text{SME}}^e = -\sum_{njm} p^n \sqrt{\frac{2j+1}{4\pi}} e^{i(m-s)\phi} \left[ \hat{\sigma}_s d_{s0}^1 d_{m0}^j \hat{\mathcal{T}}_{njm}^{e\text{NR}(0B)} + \hat{\sigma}_s (d_{s,-1}^1 d_{m,-1}^j + d_{s1}^1 d_{m1}^j) \hat{\mathcal{T}}_{njm}^{e\text{NR}(1B)} + \hat{\sigma}_s (d_{s,-1}^1 d_{m,-1}^j - d_{s1}^1 d_{m1}^j) i \hat{\mathcal{T}}_{njm}^{e\text{NR}(1E)} \right]. \quad (3.32)$$

This expression is simplified using special cases of the following relations for products of Wigner matrices,

$$d_{s's'm'}^j(\theta) d_{sm}^j(\theta) = \sum_{J=|j-1|}^{j+1} C_{j's',js}^{J's'+s} C_{j'm',jm}^{Jm'+m} d_{s'+s,m'+m}^J(\theta), \quad (3.33)$$

and exploiting the useful relations

$$d_{m'm}^j = (-1)^{m'-m} d_{mm'}^j = d_{-m-m'}^j. \quad (3.34)$$

The calculation is described in detail for the minimal SME in Appendix A of Paper 2 and will not be repeated here.<sup>6</sup> Also, to keep the presentation relatively simple, we only quote results here up to  $O(p^2)$  for the spin-dependent contributions. We find, including the nonminimal  $\hat{\mathcal{T}}_{23m}^{e\text{NR}}$  couplings but *without* at this stage applying the identities (3.29) and (3.30),

<sup>6</sup>The calculation of the  $\hat{\mathcal{T}}_{22m}^{e\text{NR}(1E)}$  coupling presented in Paper 2 is unnecessarily complicated and is greatly simplified by using the identity  $d_{s,1}^1 = (-1)^{s-1} d_{-s,-1}^1$  before applying (3.33).

$$\begin{aligned}
H_{\text{SME}}^{e\mathcal{T}} = & Y_{00}(\theta, \phi) \hat{\sigma}_s (-1)^s C_{1-s,1m}^{00} \left( \hat{\mathcal{T}}_{01m}^{e\text{NR}(0B)} + 2\hat{\mathcal{T}}_{01m}^{e\text{NR}(1B)} \right) - \sqrt{\frac{2}{5}} Y_{20}(\theta, \phi) \hat{\sigma}_s (-1)^s C_{1-s,1m}^{20} \left( \hat{\mathcal{T}}_{01m}^{e\text{NR}(0B)} - \hat{\mathcal{T}}_{01m}^{e\text{NR}(1B)} \right) \\
& + p^2 Y_{00}(\theta, \phi) \hat{\sigma}_s (-1)^s C_{1-s,1m}^{00} \left( \hat{\mathcal{T}}_{21m}^{e\text{NR}(0B)} + 2\hat{\mathcal{T}}_{21m}^{e\text{NR}(1B)} \right) - \sqrt{\frac{2}{5}} p^2 Y_{20}(\theta, \phi) \hat{\sigma}_s (-1)^s C_{1-s,1m}^{20} \left( \hat{\mathcal{T}}_{21m}^{e\text{NR}(0B)} - \hat{\mathcal{T}}_{21m}^{e\text{NR}(1B)} \right) \\
& - \sqrt{2} p^2 Y_{20}(\theta, \phi) \hat{\sigma}_s (-1)^s C_{1-s,2m}^{20} i \hat{\mathcal{T}}_{22m}^{e\text{NR}(1E)} + \sqrt{\frac{3}{5}} p^2 Y_{20}(\theta, \phi) \hat{\sigma}_s (-1)^s C_{1-s,3m}^{20} \\
& \times \left( \hat{\mathcal{T}}_{23m}^{e\text{NR}(0B)} + 2\sqrt{\frac{2}{3}} \hat{\mathcal{T}}_{23m}^{e\text{NR}(1B)} \right) - \frac{2}{3} p^2 Y_{40}(\theta, \phi) \hat{\sigma}_s (-1)^s C_{1-s,3m}^{40} \times \left( \hat{\mathcal{T}}_{23m}^{e\text{NR}(0B)} - \sqrt{\frac{3}{2}} \hat{\mathcal{T}}_{23m}^{e\text{NR}(1B)} \right). \quad (3.35)
\end{aligned}$$

Notice the important simplification at this point that the “unnatural” terms proportional just to  $Y_{20}$  and  $p^2 Y_{40}$  vanish by virtue of the identities (3.29) and (3.30). This just leaves the familiar combinations  $p^2 Y_{00} = \frac{1}{\sqrt{4\pi}} \text{tr} p_a p_b$  and  $p^2 Y_{20} = -\frac{\sqrt{5}}{2} \frac{1}{\sqrt{4\pi}} \text{tr}_Y p_a p_b$ .

The next step is to write the remaining Clebsch-Gordan coefficients in more manageable form. We need,<sup>7</sup>

$$\begin{aligned}
(-1)^s C_{1-s,1m}^{00} &= -\frac{1}{\sqrt{3}} \delta_{sm}, & (-1)^s C_{1-s,1m}^{20} &= -\frac{1}{\sqrt{6}} Y_{sm}, \\
(-1)^s C_{1-s,2m}^{20} &= -\frac{1}{\sqrt{2}} T_{sm}, & (-1)^s C_{1-s,3m}^{20} &= -\sqrt{\frac{2}{7}} X_{sm}. \quad (3.36)
\end{aligned}$$

So finally, now applying the identities (3.29) and (3.30), the spin-dependent nonminimal SME Hamiltonian can be written in the MOL frame up to  $O(p^2)$  as<sup>8</sup>

$$\begin{aligned}
H_{\text{SME}}^{e\mathcal{T}} = & -\frac{1}{\sqrt{4\pi}} \left[ \sqrt{3} \hat{\sigma}_s \delta_{sm} \hat{\mathcal{T}}_{01m}^{e\text{NR}(0B)} + \text{tr} p_a p_b \left( \frac{1}{\sqrt{3}} \hat{\sigma}_s \delta_{sm} (\hat{\mathcal{T}}_{21m}^{e\text{NR}(0B)} + 2\hat{\mathcal{T}}_{21m}^{e\text{NR}(1B)}) \right) \right. \\
& \left. + \text{tr}_Y p_a p_b \left( \frac{1}{2\sqrt{3}} \hat{\sigma}_s Y_{sm} (\hat{\mathcal{T}}_{21m}^{e\text{NR}(0B)} - \hat{\mathcal{T}}_{21m}^{e\text{NR}(1B)}) - \sqrt{\frac{7}{6}} \hat{\sigma}_s X_{sm} \hat{\mathcal{T}}_{23m}^{e\text{NR}(0B)} + \frac{\sqrt{5}}{2} \hat{\sigma}_s T_{sm} i \hat{\mathcal{T}}_{22m}^{e\text{NR}(1E)} \right) \right]. \quad (3.37)
\end{aligned}$$

With the Hamiltonian specified in the MOL frame, we can use the expectation values  $\text{tr} \langle p_a p_b \rangle$  and  $\text{tr}_Y \langle p_a p_b \rangle$  of the electron momenta evaluated in Paper 1, exploiting the cylindrical symmetry of the  $\text{H}_2^+$  and  $\bar{\text{H}}_2^-$  molecular ions. Then, we need to express  $H_{\text{SME}}^{e\mathcal{T}}$  in terms of the EXP frame couplings.

The spins and couplings are transformed from the MOL to EXP frames by

$$\begin{aligned}
\hat{\sigma}_{s'} &= \sigma_s d_{ss'}^1(\theta) e^{-is\phi}, \\
\hat{\mathcal{T}}_{n j m'} &= T_{n j m} d_{m m'}^j(\theta) e^{-im\phi}, \quad (3.38)
\end{aligned}$$

where now the angles  $(\theta, \phi)$  describe the orientation of the molecular axis relative to the EXP frame, whose  $z$  axis is aligned with the background magnetic field. Notice especially the importance of including all the  $\hat{\mathcal{T}}_{n j m}$  couplings here, not simply those with  $m = 0$ .

The techniques required for the conversion are similar to those described above for the spin-independent couplings and the derivation has been carried through in the minimal SME sector in Paper 2. Here, we simply quote the result, as always interpreting the expectation value of  $H_{\text{SME}}^{e\mathcal{T}}$  in the electron  $1s_{\sigma_g}$  state as a contribution  $V_{\text{SME}}^{e\mathcal{T}}(\mathbf{R})$  to the internucleon potential,

<sup>7</sup>We use the notation from Paper 2 for  $T_{sm}$  and  $Y_{sm}$  and also define the new matrix  $X_{sm}$  arising in the nonminimal sector as,

$$T_{sm} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad Y_{sm} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{pmatrix}, \quad X_{sm} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \sqrt{\frac{3}{2}} \end{pmatrix}.$$

<sup>8</sup>For  $s = 0$ , the coefficient of  $\hat{\mathcal{T}}_{230}^{e\text{NR}(0B)}$  has previously appeared in Eq. (132) of [17].

$$\begin{aligned}
V_{\text{SME}}^e \mathcal{T}(\mathbf{R}) = & -\sqrt{\frac{3}{4\pi}} \left[ \sigma_m \mathcal{T}_{01m}^{e\text{NR}(0B)} + \frac{1}{3} \text{tr} \langle p_a p_b \rangle \sigma_m \left( \mathcal{T}_{21m}^{e\text{NR}(0B)} + 2\mathcal{T}_{21m}^{e\text{NR}(1B)} \right) \right] + \sqrt{\frac{1}{10}} \text{tr}_Y \langle p_a p_b \rangle \sigma_s (-1)^s \\
& \times \sum_M C_{1-s,1m}^{2M} Y_{2M}(\theta, \phi) \left( \mathcal{T}_{21m}^{e\text{NR}(0B)} - \mathcal{T}_{21m}^{e\text{NR}(1B)} \right) - \sqrt{\frac{1}{2}} \text{tr}_Y \langle p_a p_b \rangle \sigma_s (-1)^s \sum_M C_{1-s,2m}^{2M} Y_{2M}(\theta, \phi) i \mathcal{T}_{22m}^{e\text{NR}(1E)} \\
& - \frac{1}{2\sqrt{5}} \frac{7}{\sqrt{6}} \text{tr}_Y \langle p_a p_b \rangle \sigma_s (-1)^s \sum_M C_{1-s,3m}^{2M} Y_{2M}(\theta, \phi) \mathcal{T}_{23m}^{e\text{NR}(0B)}. \quad (3.39)
\end{aligned}$$

We emphasize here how the sensitivity of the rovibrational energies to the nonminimal couplings  $\mathcal{T}_{230}^{e\text{NR}(0B)}$  arises only with  $\text{tr}_Y \langle p_a p_b \rangle$ , again reflecting the fact that the molecular ion only has cylindrical symmetry. This explains their absence in the corresponding energy formulas quoted in [15].

We can now impose the simplification discussed in Paper 2 that allows us to restrict to the EXP frame couplings with  $m = 0$  only. The argument is that the hyperfine-Zeeman interactions do not mix states with different values of  $M_J$ . Any matrix element of  $V_{\text{SME}}^e \mathcal{T}(\mathbf{R})$  which is off-diagonal in  $M_J$  is therefore of  $O(\text{SME})$  and therefore contributes to energy levels only at  $O(\text{SME})^2$ , which we neglect. Imposing  $\Delta M_J = 0$  on the matrix elements of  $V_{\text{SME}}^e \mathcal{T}(\mathbf{R})$  implies the constraint  $M + s = 0$  in (3.39), and since  $M = -s + m$  this in turn requires  $m = 0$ .

Inserting the required Clebsch-Gordan coefficients<sup>9</sup> we find, effectively,

$$\begin{aligned}
V_{\text{SME}}^e \mathcal{T}(\mathbf{R}) = & -\sqrt{\frac{3}{4\pi}} \left[ \sigma_0 \mathcal{T}_{010}^{e\text{NR}(0B)} + \frac{1}{3} \text{tr} \langle p_a p_b \rangle \sigma_0 \left( \mathcal{T}_{210}^{e\text{NR}(0B)} + 2\mathcal{T}_{210}^{e\text{NR}(1B)} \right) \right] + \sqrt{\frac{1}{15}} \text{tr}_Y \langle p_a p_b \rangle \sigma_0 Y_{20}(\theta, \phi) \\
& \times \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} + \frac{3}{2} \sqrt{\frac{7}{6}} \mathcal{T}_{230}^{e\text{NR}(0B)} \right) - \frac{1}{2\sqrt{5}} \text{tr}_Y \langle p_a p_b \rangle \left[ \sigma_1 Y_{2-1}(\theta, \phi) \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} \right) \right. \\
& \left. - \sqrt{\frac{7}{6}} \mathcal{T}_{230}^{e\text{NR}(0B)} - \sqrt{5} i \mathcal{T}_{220}^{e\text{NR}(1E)} \right] + \sigma_{-1} Y_{21}(\theta, \phi) \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} - \sqrt{\frac{7}{6}} \mathcal{T}_{230}^{e\text{NR}(0B)} + \sqrt{5} i \mathcal{T}_{220}^{e\text{NR}(1E)} \right) \Big]. \quad (3.40)
\end{aligned}$$

The final step in calculating the required rovibrational energies in the weak magnetic field regime is to find  $V_{\text{SME}}^e \mathcal{T}(\mathbf{R})$  by taking the matrix elements of (3.40) in the hyperfine states  $|vNJM_J\rangle$  as in (3.15). Since this now also involves the spin operators  $\sigma_s$ , the calculation is much more intricate, but all the required steps have already been set out in Paper 2 (Eqs. (5.12)–(5.17)) and we will only quote the result here. For a given value of  $M_J$ , the states with  $J = N \pm \frac{1}{2}$  are mixed and  $V_{\text{SME}}^e \mathcal{T}(\mathbf{R})$  is best presented as a  $2 \times 2$  matrix with rows/columns representing  $J', J = N \pm \frac{1}{2}$ ,

$$V_{\text{SME}}^e \mathcal{T}(\mathbf{R}) = \sqrt{\frac{3}{4\pi}} \begin{pmatrix} \alpha & \beta \\ \beta^* & \delta \end{pmatrix}. \quad (3.41)$$

After some calculation, we find

$$\begin{aligned}
\alpha = & -2 \frac{1}{2N+1} M_J \mathcal{T}_{010}^{e\text{NR}(0B)} - \text{tr} \langle p_a p_b \rangle \frac{2}{3} \frac{1}{2N+1} M_J \left( \mathcal{T}_{210}^{e\text{NR}(0B)} + 2\mathcal{T}_{210}^{e\text{NR}(1B)} \right) + \text{tr}_Y \langle p_a p_b \rangle \frac{1}{(2N+1)(2N-1)(2N+3)} \\
& \times \left[ -\frac{2}{3} N(2N-1) M_J \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} \right) + \left( 3N^2 + 6N + \frac{5}{4} - 5M_J^2 \right) M_J \sqrt{\frac{7}{6}} \mathcal{T}_{230}^{e\text{NR}(0B)} \right] \quad (3.42)
\end{aligned}$$

and

<sup>9</sup>For  $M = -s$ , the Clebsch-Gordan coefficients appearing in (3.39) are,

$$\begin{aligned}
(-1)^s C_{1-s,10}^{2-s} &= -\frac{1}{\sqrt{2}}, -\frac{1}{\sqrt{2}}, \sqrt{\frac{2}{3}}, & (-1)^s C_{1-s,20}^{2-s} &= -\frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}}, 0, \\
(-1)^s C_{1-s,30}^{2-s} &= -\frac{1}{\sqrt{7}}, -\frac{1}{\sqrt{7}}, \sqrt{\frac{3}{7}}, & \text{for } s &= 1, -1, 0 \text{ respectively.}
\end{aligned}$$

$$\delta = 2 \frac{1}{2N+1} M_J \mathcal{T}_{010}^{e\text{NR}(0B)} + \text{tr}\langle p_a p_b \rangle \frac{2}{3} \frac{1}{2N+1} M_J \left( \mathcal{T}_{210}^{e\text{NR}(0B)} + 2\mathcal{T}_{210}^{e\text{NR}(1B)} \right) - \text{tr}_Y \langle p_a p_b \rangle \frac{1}{(2N+1)(2N-1)(2N+3)} \\ \times \left[ -\frac{2}{3} (N+1)(2N+3) M_J \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} \right) + \left( 3N^2 - \frac{7}{4} - 5M_J^2 \right) M_J \sqrt{\frac{7}{6}} \mathcal{T}_{230}^{e\text{NR}(0B)} \right] \quad (3.43)$$

while,

$$\text{Re}\beta = \sqrt{\left(N + \frac{1}{2}\right)^2 - M_J^2} \left( 2 \frac{1}{2N+1} \mathcal{T}_{010}^{e\text{NR}(0B)} + \text{tr}\langle p_a p_b \rangle \frac{2}{3} \frac{1}{2N+1} \left( \mathcal{T}_{210}^{e\text{NR}(0B)} + 2\mathcal{T}_{210}^{e\text{NR}(1B)} \right) + \text{tr}_Y \langle p_a p_b \rangle \right. \\ \left. \times \frac{1}{(2N+1)(2N-1)(2N+3)} \left[ -\frac{1}{6} (2N-1)(2N+3) \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} \right) - \left( N^2 + N - \frac{3}{4} - 5M_J^2 \right) \sqrt{\frac{7}{6}} \mathcal{T}_{230}^{e\text{NR}(0B)} \right] \right). \quad (3.44)$$

The coupling  $\mathcal{T}_{230}^{e\text{NR}(1E)}$  drops out from  $\text{Re}\beta$ .<sup>10</sup> This leaves just three independent SME couplings contributing to  $V_{\text{SME}}^e(R)$  at  $O(p^2)$ , including the non-minimal coupling  $\mathcal{T}_{230}^{e\text{NR}(0B)}$ , in line with the analysis in Paper 2 using the Lorentz tensor couplings of (1.1).

We can now summarize our results for the eigenvalues  $V_{\text{SME}\pm}^e(R)$  in a concise form suitable for reading off the contributions of the spin-dependent SME couplings to the rovibrational energies. For weak magnetic fields, applying the methods of Appendix A, and using the notation (3.19) for the expansion parameter, we find<sup>11</sup>:

$$V_{\text{SME}\pm}^e(R) = \sqrt{\frac{3}{4\pi}} \left( \hat{e}_{NM_J}^{\pm}(B) \mathcal{T}_{010}^{e\text{NR}(0B)} - \frac{1}{3} \text{tr}\langle p_a p_b \rangle \hat{f}_{NM_J}^{\pm}(B) \left( \mathcal{T}_{210}^{e\text{NR}(0B)} + 2\mathcal{T}_{210}^{e\text{NR}(1B)} \right) \right. \\ \left. - \frac{1}{3} \text{tr}_Y \langle p_a p_b \rangle \hat{f}_{NM_J}^{Y\pm}(B) \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} \right) + \sqrt{\frac{7}{6}} \text{tr}_Y \langle p_a p_b \rangle \hat{f}_{NM_J}^{X\pm}(B) \mathcal{T}_{230}^{e\text{NR}(0B)} \right), \quad (3.45)$$

where

$$\hat{e}_{NM_J}^{\pm}(B) = \mp \frac{2}{2N+1} M_J \mp \frac{8}{(2N+1)^3} \left[ \left(N + \frac{1}{2}\right)^2 - M_J^2 \right] \mathcal{B}, \quad (3.46)$$

$$\hat{f}_{NM_J}^{\pm}(B) = \pm \frac{2}{2N+1} M_J \pm \frac{8}{(2N+1)^3} \left[ \left(N + \frac{1}{2}\right)^2 - M_J^2 \right] \mathcal{B}, \quad (3.47)$$

while

$$\hat{f}_{NM_J}^{Y+}(B) = \frac{2N}{(2N+1)(2N+3)} M_J - \frac{2}{(2N+1)^3} \left[ \left(N + \frac{1}{2}\right)^2 - M_J^2 \right] \mathcal{B}, \quad (3.48)$$

$$\hat{f}_{NM_J}^{Y-}(B) = -\frac{2(N+1)}{(2N-1)(2N+1)} M_J + \frac{2}{(2N+1)^3} \left[ \left(N + \frac{1}{2}\right)^2 - M_J^2 \right] \mathcal{B}, \quad (3.49)$$

and

$$\hat{f}_{NM_J}^{X+}(B) = \frac{1}{4} \frac{12N^2 + 24N + 5 - 20M_J^2}{(2N+1)(2N-1)(2N+3)} M_J + \frac{(2N-1)(2N+3) - 20M_J^2}{(2N+1)^3(2N-1)(2N+3)} \left[ \left(N + \frac{1}{2}\right)^2 - M_J^2 \right] \mathcal{B}, \quad (3.50)$$

<sup>10</sup>Recall from Paper 2 that in calculating the eigenvalues of the combined hyperfine-Zeeman and SME perturbations, only the combination  $\text{Re}\beta = \frac{1}{2}(\beta + \beta^*)$  occurs. The full expression is quoted in Appendix A, (A5) and the simplification for weak magnetic fields in (A6).

<sup>11</sup>In this paper, we are using a self-consistent notation where hatted coefficients refer to the weak magnetic field regime with states  $|vNJM_J\rangle$ , while unhatted coefficients refer to the strong field regime with states  $|vNM_JM_S\rangle$ . This means that the hatted  $\hat{f}_{NM_J}$  coefficients in (3.47), (3.48) and (3.49) here are identical to the corresponding unhatted  $f_{NM_J}$  in Paper 2.

$$\hat{f}_{NM_J}^{X\pm}(B) = -\frac{1}{4} \frac{12N^2 - 7 - 20M_J^2}{(2N+1)(2N-1)(2N+3)} M_J - \frac{(2N-1)(2N+3) - 20M_J^2}{(2N+1)^3(2N-1)(2N+3)} \left[ \left(N + \frac{1}{2}\right)^2 - M_J^2 \right] \mathcal{B}. \quad (3.51)$$

Notice that in all cases, the  $O(B)$  correction vanishes for the unmixed, or stretched, states with  $M_J = \pm(N + \frac{1}{2})$ .

For large magnetic fields, we repeat this analysis taking the matrix elements of  $V_{\text{SME}}^e(\mathbf{R})$  in (3.40) in the basis states  $|vNM_JM_S\rangle$ . Some intermediate details are given in Appendix A. Here, we simply present the result for the eigenvalues  $V_{\text{SME}\pm}^e(\mathbf{R})$ . This takes the same form as above,

$$V_{\text{SME}\pm}^e(\mathbf{R}) = \sqrt{\frac{3}{4\pi}} \left( e_{NM_J}^{\pm} \mathcal{T}_{010}^{e\text{NR}(0B)} - \frac{1}{3} \text{tr}\langle p_a p_b \rangle f_{NM_J}^{\pm} \left( \mathcal{T}_{210}^{e\text{NR}(0B)} + 2\mathcal{T}_{210}^{e\text{NR}(1B)} \right) \right. \\ \left. - \frac{1}{3} \text{tr}_Y \langle p_a p_b \rangle f_{NM_J}^{Y\pm}(B) \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} \right) + \sqrt{\frac{7}{6}} \text{tr}_Y \langle p_a p_b \rangle f_{NM_J}^{X\pm}(B) \mathcal{T}_{230}^{e\text{NR}(0B)} \right), \quad (3.52)$$

with coefficients,

$$e_{NM_J}^{\pm} = \mp 1, \quad (3.53)$$

$$f_{NM_J}^{\pm} = \pm 1, \quad (3.54)$$

$$f_{NM_J}^{Y\pm}(B) = \mp c_{NM_J \mp \frac{1}{2}} \pm 3 \frac{[(N + \frac{1}{2})^2 - M_J^2]}{(2N-1)(2N+3)} M_J \frac{1}{\mathcal{B}}, \quad (3.55)$$

$$f_{NM_J}^{X\pm}(B) = \pm \frac{1}{2} c_{NM_J \mp \frac{1}{2}} \pm \frac{[(N + \frac{1}{2})^2 - M_J^2]}{(2N-1)(2N+3)} M_J \frac{1}{\mathcal{B}}. \quad (3.56)$$

These coefficients, together with their weak-field counterparts, in principle allow constraints on the individual components of the electron SME couplings  $\mathcal{T}_{njm}^{e\text{NR}}$  to be distinguished by measuring suitably chosen combinations of transition frequencies.

In the same way as for the spin-independent couplings, we can calculate the contribution of the internucleon potentials  $V_{\text{SME}\pm}^e(\mathbf{R})$  in (3.45) and (3.52) to the rovibrational energies in the form,

$$E_{vNM_J\pm}^e = V_{\text{SME}\pm}^e + \delta_{\text{SME}\pm}^e \left( v + \frac{1}{2} \right) \omega_0 - x_{\text{SME}\pm}^e \left( v + \frac{1}{2} \right)^2 \omega_0 \\ + B_{\text{SME}\pm}^e N(N+1) \omega_0 - \alpha_{\text{SME}\pm}^e \left( v + \frac{1}{2} \right) N(N+1) \omega_0 + \dots \quad (3.57)$$

Here, for weak magnetic fields, we find from (3.45),

$$V_{\text{SME}\pm}^e = \sqrt{\frac{3}{4\pi}} \left( \frac{2}{\alpha^2} \hat{e}_{NM_J}^{\pm} \frac{1}{m_e} \mathcal{T}_{010}^{e\text{NR}(0B)} - 0.782 \hat{f}_{NM_J}^{\pm} m_e \left( \mathcal{T}_{210}^{e\text{NR}(0B)} + 2\mathcal{T}_{210}^{e\text{NR}(1B)} \right) \right. \\ \left. - 0.240 \hat{f}_{NM_J}^{Y\pm} m_e \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} \right) + 0.294 \sqrt{7} \hat{f}_{NM_J}^{X\pm} m_e \mathcal{T}_{230}^{e\text{NR}(0B)} \right), \quad (3.58)$$

while

$$\begin{aligned}
\delta_{\text{SME}\pm}^e \mathcal{T} &= \sqrt{\frac{3}{4\pi}} \left( 1.003 \hat{f}_{NM_J}^{\pm} m_e \left( \mathcal{T}_{210}^{e\text{NR}(0B)} + 2\mathcal{T}_{210}^{e\text{NR}(1B)} \right) \right. \\
&\quad \left. + 0.545 \hat{f}_{NM_J}^{Y\pm} m_e \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} \right) - 0.667 \sqrt{7} \hat{f}_{NM_J}^{X\pm} m_e \mathcal{T}_{230}^{e\text{NR}(0B)} \right), \\
B_{\text{SME}\pm}^e \mathcal{T} &= \lambda \sqrt{\frac{3}{4\pi}} \left( 0.667 \hat{f}_{NM_J}^{\pm} m_e \left( \mathcal{T}_{210}^{e\text{NR}(0B)} + 2\mathcal{T}_{210}^{e\text{NR}(1B)} \right) \right. \\
&\quad \left. + 0.224 \hat{f}_{NM_J}^{Y\pm} m_e \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} \right) - 0.275 \sqrt{7} \hat{f}_{NM_J}^{X\pm} m_e \mathcal{T}_{230}^{e\text{NR}(0B)} \right), \\
x_{\text{SME}\pm}^e \mathcal{T} &= \lambda \sqrt{\frac{3}{4\pi}} \left( 1.629 \hat{f}_{NM_J}^{\pm} m_e \left( \mathcal{T}_{210}^{e\text{NR}(0B)} + 2\mathcal{T}_{210}^{e\text{NR}(1B)} \right) \right. \\
&\quad \left. + 1.130 \hat{f}_{NM_J}^{Y\pm} m_e \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} \right) - 1.384 \sqrt{7} \hat{f}_{NM_J}^{X\pm} m_e \mathcal{T}_{230}^{e\text{NR}(0B)} \right), \\
\alpha_{\text{SME}\pm}^e \mathcal{T} &= \lambda^2 \sqrt{\frac{3}{4\pi}} \left( 2.152 \hat{f}_{NM_J}^{\pm} m_e \left( \mathcal{T}_{210}^{e\text{NR}(0B)} + 2\mathcal{T}_{210}^{e\text{NR}(1B)} \right) \right. \\
&\quad \left. + 0.195 \hat{f}_{NM_J}^{Y\pm} m_e \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} \right) - 0.239 \sqrt{7} \hat{f}_{NM_J}^{X\pm} m_e \mathcal{T}_{230}^{e\text{NR}(0B)} \right), \tag{3.59}
\end{aligned}$$

all in atomic units, with  $m_e \mathcal{T}_{2jm}^{e\text{NR}}$  dimensionless and all energies in units of  $R_H$ . For clarity, we have omitted the explicit  $B$  dependence in the notation for the coefficients here.

The analogous results for strong magnetic fields follow in the same way from (3.52), with the substitution from hatted to unhatted coefficients,  $\hat{f}_{NM_J}^{\pm} \rightarrow f_{NM_J}^{\pm}$  etc.

#### IV. PROTON SME COUPLINGS AND ROVIBRATIONAL ENERGIES

We now come to the proton sector of the SME and calculate the dependence of the rovibrational energy levels on the couplings  $\mathcal{V}_{njm}^p$ . Since we only consider para- $\text{H}_2^+$  where the total proton spin  $\mathbf{I} = 0$ , we may restrict here to the spin-independent proton SME couplings alone.

In the nonrelativistic approximation, the SME Hamiltonian for the two protons, with momenta  $\mathbf{p}_1$  and  $\mathbf{p}_2$ , is

$$H_{\text{SME}}^p = -\sum_{n,jm} p_1^n Y_{jm}(\hat{\mathbf{p}}_1) \mathcal{V}_{njm}^{p\text{NR}} - \sum_{n,jm} p_2^n Y_{jm}(\hat{\mathbf{p}}_2) \mathcal{V}_{njm}^{p\text{NR}} \tag{4.1}$$

with the sum running over  $n = 0, 2, 4, \dots$ . This expression is frame independent, and we immediately interpret it in the EXP frame.

The momenta  $\mathbf{p}_1, \mathbf{p}_2$  and the electron momentum  $\mathbf{p}_e$  are most conveniently reexpressed in terms of the relative momentum  $\mathbf{P}$  of the two protons, the momentum  $\mathbf{p}$  of the electron relative to the molecule center of mass c.m., and the c.m. momentum  $\mathbf{P}_{\text{c.m.}}$  of the molecule. The momentum

$\mathbf{P}$  is defined with a reduced mass  $\mu = m_p/2$  and  $\mathbf{p}$  with  $\hat{\mu} = 2m_p m_e / (2m_p + m_e)$ . Details are given in Paper 1. Setting  $\mathbf{P}_{\text{c.m.}}$  to zero, the relations we need here are simply,

$$\mathbf{p}_1 = \mathbf{P} - \frac{1}{2}\mathbf{p}, \quad \mathbf{p}_2 = -\mathbf{P} - \frac{1}{2}\mathbf{p}. \tag{4.2}$$

The momentum-independent term in  $H_{\text{SME}}^p$  is evidently just the constant

$$H_{\text{SME}}^p|_{n=0} = -\frac{2}{\sqrt{4\pi}} \mathcal{V}_{000}^{p\text{NR}}, \tag{4.3}$$

and is usually considered unobservable and therefore neglected. At  $O(p^2)$ , recalling the restrictions  $j = n, n-2, \dots \geq 0$  on the spherical tensors [6],

$$\begin{aligned}
H_{\text{SME}}^p|_{n=2} &= -(p_1^2 + p_2^2) \frac{1}{\sqrt{4\pi}} \mathcal{V}_{200}^{p\text{NR}} \\
&\quad - \left( p_1^2 Y_{2m}(\hat{\mathbf{p}}_1) + p_2^2 Y_{2m}(\hat{\mathbf{p}}_2) \right) \mathcal{V}_{22m}^{p\text{NR}}. \tag{4.4}
\end{aligned}$$

Now, from (4.2),

$$p_1^2 + p_2^2 = 2P^2 + \frac{1}{2}p^2, \tag{4.5}$$

where, crucially, the mixed terms involving  $\mathbf{P} \cdot \mathbf{p}$  cancel. In the Born-Oppenheimer analysis, this means that, as already described in Paper 1, the electron Schrödinger equation receives an extra contribution from the proton SME couplings.

To recover the equivalent split for the  $j = 2$  couplings, we need the following key result:

$$p_1^2 Y_{2m}(\hat{\mathbf{p}}_1) + p_2^2 Y_{2m}(\hat{\mathbf{p}}_2) = 2P^2 Y_{2m}(\hat{\mathbf{P}}) + \frac{1}{2} p^2 Y_{2m}(\hat{\mathbf{p}}). \quad (4.6)$$

To establish this, we use the explicit form of the spherical harmonics for each  $m$  to write the lhs in terms of Cartesian components of  $\mathbf{p}_1$  and  $\mathbf{p}_2$ , then reconstruct the rhs following their reexpression in terms of  $\mathbf{P}$  and  $\mathbf{p}$  using (4.2).

At this point, however, the second term is still expressed in the EXP frame. Using the transformation (3.5) of the spherical tensors under rotations together with the expression (3.3) of the spherical harmonics in terms of Wigner matrices, we note that this term is invariant, so we can rewrite it in terms of MOL frame couplings and momenta,

$$p^2 Y_{2m}(\hat{\mathbf{p}}_{\text{EXP}}) \mathcal{V}_{22m}^{p\text{NR}} = p^2 Y_{2m}(\hat{\mathbf{p}}_{\text{MOL}}) \hat{\mathcal{V}}_{22m}^{p\text{NR}}. \quad (4.7)$$

In this form we can take the expectation values of the momentum factors in the  $1s\sigma_g$  state, where cylindrical symmetry restricts to  $m = 0$ , leaving

$$\langle p^2 Y_{20}(\hat{\mathbf{p}}_{\text{MOL}}) \rangle \hat{\mathcal{V}}_{220}^{p\text{NR}} = -\frac{1}{2} \sqrt{\frac{5}{4\pi}} \text{tr}_Y \langle p_a p_b \rangle \hat{\mathcal{V}}_{220}^{p\text{NR}}. \quad (4.8)$$

The total contribution at  $O(P^2)$  to the electron Schrödinger equation is therefore

$$-\frac{1}{2} \frac{1}{\sqrt{4\pi}} \left( \text{tr} \langle p_a p_b \rangle \hat{\mathcal{V}}_{200}^{p\text{NR}} - \frac{1}{2} \sqrt{5} \text{tr}_Y \langle p_a p_b \rangle \hat{\mathcal{V}}_{220}^{p\text{NR}} \right). \quad (4.9)$$

Comparing with (3.4), the effect of these terms on the rovibrational energy levels is therefore included simply by substituting  $\mathcal{V}_{2jm}^{e\text{NR}}$  with

$$\tilde{\mathcal{V}}_{2jm}^{e\text{NR}} \equiv \mathcal{V}_{2jm}^{e\text{NR}} + \frac{1}{2} \mathcal{V}_{2jm}^{p\text{NR}}, \quad (4.10)$$

in (3.13) and (3.16). This is the equivalent of the result initially found in terms of Lorentz tensor SME couplings in Paper 1. [Notice the absence of explicit factors of  $m_e$  and  $m_p$  in (4.10), compatible with the identifications of the CPT even and odd components  $\tilde{c}_{2jm}^e$  and  $\tilde{a}_{2jm}^e$  in Sec. 7 there.]

The remaining terms in the proton SME Hamiltonian, expressed in the EXP frame, are then simply

$$H_{\text{SME}}^p|_{n=0,2} = -\frac{2}{\sqrt{4\pi}} \mathcal{V}_{000}^{p\text{NR}} - 2P^2 \left( \frac{1}{\sqrt{4\pi}} \mathcal{V}_{200}^{p\text{NR}} + Y_{2m}(\hat{\mathbf{P}}) \mathcal{V}_{22m}^{p\text{NR}} \right), \quad (4.11)$$

and we recall from the discussion in the previous Section that only the  $m = 0$  components contribute at  $O(\text{SME})$ . The corresponding rovibrational energies follow by taking the expectation values first in the Zeeman states  $|vNM_J M_S\rangle$  appropriate for high magnetic fields and then in the hyperfine states  $|vNJM_J\rangle$  relevant at weak fields.

The expectation values of the spherical harmonic  $Y_{20}(\hat{\mathbf{P}})$  in these states are the same as already encountered for the electron couplings. In the Zeeman states,

$$\langle vNM_N M'_S | Y_{20}(\hat{\mathbf{P}}) | vNM_N M_S \rangle = \sqrt{\frac{5}{4\pi}} c_{NM_N} \delta_{M_S M'_S}, \quad (4.12)$$

and we find

$$\begin{aligned} \Delta E_{\text{SME}\pm}^n|_{n=0,2} &= -\frac{2}{\sqrt{4\pi}} \mathcal{V}_{000}^{p\text{NR}} - 2\langle vN | P^2 | vN \rangle \\ &\times \left( \frac{1}{\sqrt{4\pi}} \mathcal{V}_{200}^{p\text{NR}} + \sqrt{\frac{5}{4\pi}} c_{NM_N \mp \frac{1}{2}} \mathcal{V}_{220}^{p\text{NR}} \right), \end{aligned} \quad (4.13)$$

up to  $O(P^2)$ , with the  $c_{NM_N}$  coefficients in (3.8).

For the hyperfine states, the expectation values at fixed  $M_J$  are nondiagonal in the  $2 \times 2$  sector with  $J', J = N \pm \frac{1}{2}$  and we need,

$$\begin{aligned} \langle vNJ' M_J | Y_{20}(\hat{\mathbf{P}}) | vNJM_J \rangle \\ = \sqrt{\frac{5}{4\pi}} \sum_{M_S} C_{NM_N \frac{1}{2} M_S}^{J' M_J} C_{NM_N \frac{1}{2} M_S}^{JM_J} c_{NM_N}. \end{aligned} \quad (4.14)$$

Finding the eigenvalues in exactly the same way as described for the electron (see Appendix A) therefore gives

$$\begin{aligned} \Delta E_{\text{SME}\pm}^n|_{n=0,2} &= -\frac{2}{\sqrt{4\pi}} \mathcal{V}_{000}^{p\text{NR}} - 2\langle vN | P^2 | vN \rangle \\ &\times \left( \frac{1}{\sqrt{4\pi}} \mathcal{V}_{200}^{p\text{NR}} + \sqrt{\frac{5}{4\pi}} \hat{c}_{NM_J}^{\pm}(B) \mathcal{V}_{220}^{p\text{NR}} \right), \end{aligned} \quad (4.15)$$

with the same coefficients  $\hat{c}_{NM_J}^{\pm}(B)$  introduced in (3.17) and (3.18).

The next step is to evaluate the expectation value  $\langle vN | P^2 | vN \rangle$  of the proton momentum in the spin-independent rovibrational states  $|vN\rangle$ . These states are constructed in a systematic perturbation expansion about the simple harmonic oscillator (SHO) states found by expanding about the minimum  $R_0$  of the internucleon potential  $V_M(R)$ . The details of this derivation are presented in a self-contained way in Appendix C. The results are expressed in a form directly matching the expression in (1.3) for the full

rovibrational energy spectrum, i.e. an expansion in  $(v + \frac{1}{2})$  and  $N(N+1)$  with coefficients parametrized by  $\lambda \sim \sqrt{m_e/m_p}$ . We find,

$$\begin{aligned} \langle vN|P^2|vN\rangle &= \frac{1}{2}m_p\omega_0 \left[ \left( v + \frac{1}{2} \right) + N(N+1)\lambda \right. \\ &\quad \left. + \frac{3}{2} \left( 3 + \frac{R_0 V_M'''}{V_M''} \right) \left( v + \frac{1}{2} \right) N(N+1)\lambda^2 + \dots \right] \\ &= \frac{1}{2}m_p\omega_0 \left[ \left( v + \frac{1}{2} \right) + N(N+1)\lambda - 3.405 \right. \\ &\quad \left. \times \left( v + \frac{1}{2} \right) N(N+1)\lambda^2 + \dots \right], \end{aligned} \quad (4.16)$$

where  $\omega_0$  is the frequency of the unperturbed SHO oscillations, given in terms of the Rydberg constant by  $\omega_0 \sim R_H \sqrt{m_e/m_p}$ .

Together, (4.16) with (4.14) or (4.15) give the proton contributions at  $O(P^2)$  to the components  $\delta_{\text{SME}}^n$ ,  $B_{\text{SME}}^n$  and  $\alpha_{\text{SME}}^n$  in the rovibrational energy levels.<sup>12</sup>

Now we have seen how to reformulate the derivation of the proton SME couplings to the rovibrational energies at  $O(P^2)$  entirely within the spherical tensor formalism, we are equipped to extend the analysis to the nonminimal SME, incorporating effects at  $O(P^4)$  and the associated nonminimal couplings. At  $O(P^4)$ , the SME Hamiltonian is

$$\begin{aligned} H_{\text{SME}}^P|_{n=4} &= -p_1^4 \left( \frac{1}{\sqrt{4\pi}} \mathcal{V}_{400}^{p\text{NR}} + Y_{2m}(\hat{\mathbf{p}}_1) \mathcal{V}_{42m}^{p\text{NR}} \right. \\ &\quad \left. + Y_{4m}(\hat{\mathbf{p}}_1) \mathcal{V}_{44m}^{p\text{NR}} \right) - (\mathbf{p}_1 \rightarrow \mathbf{p}_2). \end{aligned} \quad (4.17)$$

At this point we would hope to make the change of variables from  $\mathbf{p}_1, \mathbf{p}_2$  to  $\mathbf{P}, \mathbf{p}$  with identities analogous to (4.6) above. However, even for the simplest case, the coefficient of  $\mathcal{V}_{400}^{p\text{NR}}$ , we encounter a difficulty. From the definitions (4.2) we immediately find,

$$p_1^4 + p_2^4 = 2P^4 + P^2 p^2 + (\mathbf{P} \cdot \mathbf{p})^2 + \frac{1}{8} p^4, \quad (4.18)$$

so the mixed terms involving both  $\mathbf{P}$  and  $\mathbf{p}$  do *not* cancel. This spoils the clean separation of the total Schrödinger

<sup>12</sup>To match with the corresponding expression in terms of Lorentz tensor couplings in the minimal SME, the dictionary is,

$$\frac{1}{\sqrt{4\pi}} \mathcal{V}_{200}^{p\text{NR}} = -\frac{1}{3m_p^2} \text{tr} E_{ij}^p, \quad \sqrt{\frac{5}{4\pi}} \mathcal{V}_{220}^{p\text{NR}} = \frac{1}{3m_p^2} \text{tr}_Y E_{ij}^p,$$

with [19],

$$E_{ij}^p = -m_p \left( c_{ij} + \frac{1}{2} c_{00} \delta_{ij} \right) + m_p^2 (3a_{0ij} + a_{000} \delta_{ij}).$$

equation into electron and proton parts, which underpins the simple Born-Oppenheimer split. We can try different strategies to evaluate these mixed terms, but no sufficiently simple results emerge for our present purpose. Systematic improvements on the basic Born-Oppenheimer have been the subject of extensive theoretical work in atomic physics; for a review, see e.g. [20]. These terms nevertheless only give contributions suppressed by powers of  $\lambda$  relative to the leading power in the individual coefficients  $x_{\text{SME}}^n$ ,  $B_{\text{SME}}^n$ ,  $\alpha_{\text{SME}}^n$  in the rovibrational energy expansion (1.3) so may be consistently neglected.<sup>13</sup> In this leading approximation, we therefore simply consider

$$p_1^4 Y_{jm}(\hat{\mathbf{p}}_1) + p_2^4 Y_{jm}(\hat{\mathbf{p}}) = 2P^4 Y_{jm}(\hat{\mathbf{P}}) + \dots \quad (4.19)$$

and take,

$$\begin{aligned} H_{\text{SME}}^P|_{n=4} &= -2P^4 \left( \frac{1}{\sqrt{4\pi}} \mathcal{V}_{400}^{p\text{NR}} + Y_{2m}(\hat{\mathbf{P}}) \mathcal{V}_{42m}^{p\text{NR}} + Y_{4m}(\hat{\mathbf{P}}) \mathcal{V}_{44m}^{p\text{NR}} \right) + \dots \end{aligned} \quad (4.20)$$

The corresponding energies are found as usual by taking expectation values in either the Zeeman or hyperfine states for strong or weak magnetic fields respectively. Again, the expectation values of the spherical harmonics yield coefficients already evaluated for the electron couplings, so we can immediately write the final results. For strong magnetic fields,

$$\begin{aligned} \Delta E_{\text{SME}\pm}^n|_{n=4} &= -2\langle vN|P^4|vN\rangle \left( \frac{1}{\sqrt{4\pi}} \mathcal{V}_{400}^{p\text{NR}} \right. \\ &\quad \left. + \sqrt{\frac{5}{4\pi}} c_{NM_J \mp \frac{1}{2}} \mathcal{V}_{420}^{p\text{NR}} + \frac{3}{\sqrt{4\pi}} c_{NM_J \mp \frac{1}{2}}^{(4)} \mathcal{V}_{440}^{p\text{NR}} \right), \end{aligned} \quad (4.21)$$

while for weak fields,

$$\begin{aligned} \Delta E_{\text{SME}\pm}^n|_{n=4} &= -2\langle vN|P^4|vN\rangle \left( \frac{1}{\sqrt{4\pi}} \mathcal{V}_{400}^{p\text{NR}} \right. \\ &\quad \left. + \sqrt{\frac{5}{4\pi}} \hat{c}_{NM_J}^{\pm} \mathcal{V}_{420}^{p\text{NR}} + \frac{3}{\sqrt{4\pi}} \hat{c}_{NM_J}^{(4)\pm} \mathcal{V}_{440}^{p\text{NR}} \right), \end{aligned} \quad (4.22)$$

with the coefficients quoted in (3.10) and (3.20), (3.21).

The momentum expectation value is evaluated in Appendix C, where we show

<sup>13</sup>To estimate the size of the neglected terms, note that  $\langle p^2 \rangle \sim R_H m_e$  while  $\langle P^2 \rangle \sim R_H \sqrt{m_e m_p}$ , so they will give corrections suppressed by  $O(\sqrt{m_e/m_p}) \sim \lambda$  to the leading terms quoted below separately for each coefficient  $x_{\text{SME}}^n$ ,  $B_{\text{SME}}^n$ ,  $\alpha_{\text{SME}}^n$ , ... in the expansion of the rovibrational energy.

$$\begin{aligned}
\langle vN|P^4|vN\rangle = & \frac{1}{4}m_p^2\omega_0^2\left[\frac{3}{2}\left[\left(v+\frac{1}{2}\right)^2+\frac{1}{4}\right]\right. \\
& -\frac{3}{8}N(N+1)\left(3-\frac{R_0V_M'''}{V_M'''}\right)\lambda^2 \\
& \left.+2\left(v+\frac{1}{2}\right)N(N+1)\lambda+\dots\right], \quad (4.23)
\end{aligned}$$

where the ... denote terms of lower order in  $\lambda$  that should be dropped for consistency given the initial approximation in (4.19). Notice the different orders in  $\lambda$  of these terms compared with their equivalents in the expectation value  $\langle vN|P^2|vN\rangle$  in (4.16).

Together, (4.23) with (4.21) or (4.22) give the proton contributions to the components  $x_{\text{SME}}^n$ ,  $B_{\text{SME}}^n$  and  $\alpha_{\text{SME}}^n$  in the rovibrational energies at  $O(P^4)$ . Notice there is no contribution to  $\delta_{\text{SME}}^n$  in this case.

These  $O(P^4)$  contributions to the rovibrational energy coefficients provide sensitivity to the nonminimal SME couplings  $\mathcal{V}_{400}^{p\text{NR}}$ ,  $\mathcal{V}_{420}^{p\text{NR}}$  and  $\mathcal{V}_{440}^{p\text{NR}}$ . Individual constraints on these, as well as  $\mathcal{V}_{200}^{p\text{NR}}$  and  $\mathcal{V}_{220}^{p\text{NR}}$ , may be achieved by measurements of spectral transitions corresponding to states with different rovibrational quantum numbers  $(v, N)$  through the leading terms in (4.16) and (4.23) and, crucially, through discriminating the different spin components  $\mathcal{V}_{njm}^{p\text{NR}}$  by exploiting the quantum number dependence of the  $c$  and  $\hat{c}$  coefficients.

## V. FRAME TRANSFORMATIONS AND TRANSITION FREQUENCY VARIATIONS

In the SME, Lorentz symmetry is broken because the vector and tensor couplings (which may be usefully thought of as VEVs of fields in some more fundamental theory) take on fixed, nonzero values. Unlike the familiar case of QFTs with scalar couplings, this means that in the SME the results of physical measurements depend on the frame of reference of the experiment.

This has two immediate consequences. First, in comparing the results of experiments at different locations and orientations, we need to make an appropriate Lorentz transformation (rotation or boost) to change the frame of reference of the individual experiments. Second, measurements in the same experiment at a fixed location on Earth will experience daily (sidereal) and annual variations as the frame of reference of the experiment changes as the Earth rotates and orbits the Sun.

To compare the results of different experiments, or the same experiment performed at different times, it is convenient to refer the couplings to a standard frame [13]. It has become established practice to use a solar-centred frame (SUN), defined by an orthonormal set of basis vectors  $\mathbf{e}_I$  with the  $Z$  axis aligned with the Earth's rotation axis and the  $X$  axis with the vernal equinox. This implies that  $\mathbf{e}_X, \mathbf{e}_Y$  lie in

the plane of the Earth's equator, making an angle of  $\eta = 23^\circ$  with the orbital plane. Where necessary, the origin of the time coordinate  $T$  is chosen as the vernal equinox in 2000.

In practice it is convenient to introduce one further frame, a standard laboratory frame (LAB). The LAB frame is defined locally with basis vectors  $\mathbf{e}_i$  with the  $z$  axis pointing vertically to the zenith at the location of the experiment and the  $x$  axis pointing south. The relation of the  $\mathbf{e}_z$  and  $\mathbf{e}_Z$  axes in the LAB and SUN frames is given by the colatitude  $\chi$  of the laboratory.

### A. Rotations and daily (sidereal) variations

Since our results for the energy levels are given in terms of couplings in EXP frame coordinates, we need to convert to the standard SUN frame in two stages, first re-expressing in terms of the LAB frame then a further rotation from LAB to SUN. We illustrate this transformation for a generic spin-independent coupling  $\mathcal{V}_{njm}$ , dropping all other labels since the rotation is entirely determined by the  $SO(3)$  indices  $(j, m)$ . The results for the spin-dependent couplings  $\mathcal{T}_{njm}$ , of all types, are identical.

To transform from EXP to LAB, we need the angle  $\phi$  of the magnetic field measured anticlockwise from due south, and its angle  $\theta$  measured from the local vertical. The couplings transform with the Wigner matrices as (following our notation so far, couplings with no superscript are in the EXP frame),

$$\mathcal{V}_{njm'} = \mathcal{V}_{njm}^{\text{LAB}} e^{im\phi} d_{mm'}^j(\theta). \quad (5.1)$$

Combining with a further rotation to the SUN frame we can show (nontrivially, since in both cases the angular momentum operators defining the Wigner matrices are to be taken in the initial frame [21]), that

$$\mathcal{V}_{njm'} = \mathcal{V}_{njm}^{\text{SUN}} e^{im\omega_\oplus T_\oplus} d_{mm''}^j(\chi) e^{im''\phi} d_{m''m'}^j(\theta), \quad (5.2)$$

where  $\omega_\oplus$  is the Earth's angular rotation frequency corresponding to a sidereal day and with  $T_\oplus$  being local sidereal time in the LAB frame.

Next, we use the Wigner matrix addition theorem<sup>14</sup> to show,

<sup>14</sup>The standard form for the addition theorem for the little Wigner matrices  $d_{mm'}^j$  is [21]

$$d_{mm''}^j(\beta_1) e^{-im''\varphi} d_{m''m'}^j(\beta_2) = e^{-im\alpha} d_{mm'}^j(\beta) e^{-im'\gamma},$$

with

$$\cot \alpha = \cos \beta_1 \cot \varphi + \sin \beta_1 \cot \beta_2 (\sin \varphi)^{-1},$$

$$\cos \beta = \cos \beta_1 \cos \beta_2 - \sin \beta_1 \sin \beta_2 \cos \varphi,$$

$$\cot \gamma = \cos \beta_2 \cot \varphi + \sin \beta_2 \cot \beta_1 (\sin \varphi)^{-1}.$$

Also note the useful relation  $d_{mm'}^j(-\theta) = d_{m'm}^j(\theta)$ .

$$\mathcal{V}_{n j m'} = \mathcal{V}_{n j m}^{\text{SUN}} e^{im(\omega_{\oplus} T_{\oplus} + \alpha)} d_{m m'}^j(\vartheta) e^{im'\gamma}, \quad (5.3)$$

where here,

$$\cos \vartheta = \cos \chi \cos \theta - \sin \chi \sin \theta \cos \phi, \quad (5.4)$$

$$\cot \alpha = \cos \chi \cot \phi + \sin \chi \cot \theta (\sin \phi)^{-1}, \quad (5.5)$$

with an identical formula for  $\cot \gamma$  with  $\chi, \theta$  interchanged.

Next, recalling that only couplings with  $m = 0$  in the EXP frame appear in the energy levels, we only need

$$\begin{aligned} \mathcal{V}_{n j 0}^{w \text{NR}} &= \mathcal{V}_{n j m}^{w \text{NR SUN}} d_{m 0}^j(\vartheta) e^{im(\omega_{\oplus} T_{\oplus} + \alpha)}, \\ \mathcal{T}_{n j 0}^{w \text{NR}} &= \mathcal{T}_{n j m}^{w \text{NR SUN}} d_{m 0}^j(\vartheta) e^{im(\omega_{\oplus} T_{\oplus} + \alpha)}, \end{aligned} \quad (5.6)$$

where we restore the full notation, with  $w = e, p$ . These expressions are to be inserted into the formulas for the internucleon potential and energy levels to give our final results in the standard SUN frame.

We see immediately that rovibrational transition frequencies measured at different times during the Earth's daily rotation will show a time variation. Moreover, this depends

on *all* the azimuthal components of the couplings, not just  $m = 0$ . This is quite natural in a Lorentz violating theory—measurements taken with a different orientation of the experiment will be sensitive to different components of a background Lorentz vector or tensor, such as the effective SME couplings.

In principal, the dependence of these variations on the couplings with different values of  $m$  can be isolated since they vary according to different harmonics of the rotation frequency, arising from the  $\exp(im(\omega_{\oplus} T_{\oplus} + \alpha))$  factor.

Notice also that the orientation of the magnetic field also introduces a constant, experiment-specific, phase into the variation. This will not be important in analyzing time variations in a single experiment, since only time differences will matter, but could be relevant in comparing results from experiments with different magnetic field orientations or in different laboratories, since according to (5.5) the phase  $\alpha$  depends on  $\phi, \theta$  and  $\chi$ .

We can make (5.6) more explicitly tailored to experimental measurements by rewriting entirely in terms of real quantities. Using the relations  $\mathcal{V}_{n j (-m)} = (-1)^m \mathcal{V}_{n j m}^*$  and  $d_{(-m)0}^j = (-1)^m d_{m 0}^j$ , (5.6) becomes,

$$\begin{aligned} \mathcal{V}_{n j 0}^{w \text{NR}} &= d_{00}^j(\vartheta) \mathcal{V}_{n j 0}^{w \text{NR SUN}} \\ &+ \sum_{|m|=1}^j d_{m|0}^j(\vartheta) \left( \cos(m(\omega_{\oplus} T_{\oplus} + \alpha)) \text{Re} \mathcal{V}_{n j |m|}^{w \text{NR SUN}} - \sin(m(\omega_{\oplus} T_{\oplus} + \alpha)) \text{Im} \mathcal{V}_{n j |m|}^{w \text{NR SUN}} \right). \end{aligned} \quad (5.7)$$

Finally, to illustrate how these time variations enter our final results, we quote here the explicit example of the proton contribution to the energy levels, up to  $O(P^2)$  and for weak fields, written out fully in terms of SUN frame couplings [see (4.15)]<sup>15</sup>:

$$\begin{aligned} \Delta E_{\text{SME}\pm}^n|_{n=0,2} &= -\frac{2}{\sqrt{4\pi}} \mathcal{V}_{000}^{p \text{NR SUN}} \\ &- 2 \langle vN | P^2 | vN \rangle \left[ \frac{1}{\sqrt{4\pi}} \mathcal{V}_{200}^{p \text{NR SUN}} + \sqrt{\frac{5}{4\pi}} \hat{c}_{NM_J}^{\pm}(B) \left( -\frac{1}{2} (1 - 3 \cos^2 \vartheta) \mathcal{V}_{220}^{p \text{NR}} \right. \right. \\ &- \frac{1}{2} \sqrt{\frac{3}{2}} \sin 2\vartheta \left( \cos(\omega_{\oplus} T_{\oplus} + \alpha) \text{Re} \mathcal{V}_{221}^{p \text{NR SUN}} - \sin(\omega_{\oplus} T_{\oplus} + \alpha) \text{Im} \mathcal{V}_{221}^{p \text{NR SUN}} \right) \\ &\left. \left. + \frac{1}{2} \sqrt{\frac{3}{2}} \sin^2 \vartheta \left( \cos 2(\omega_{\oplus} T_{\oplus} + \alpha) \text{Re} \mathcal{V}_{222}^{p \text{NR SUN}} - \sin 2(\omega_{\oplus} T_{\oplus} + \alpha) \text{Im} \mathcal{V}_{222}^{p \text{NR SUN}} \right) \right) \right]. \end{aligned} \quad (5.8)$$

All the other expressions for energy levels in the paper can be expanded out in the same way showing the dependence on all the azimuthal components of the spherical tensor couplings at fixed rank  $j$ . As we now show, consideration of the effects of Lorentz boosts also exposes a time-dependence on couplings of different rank.

<sup>15</sup>The corresponding expressions for many of the leading terms for a particular experimentally important transition frequency are tabulated in [15] and may be derived directly from the more general results given here.

### B. Lorentz boosts

The energy levels will also experience variations due to Lorentz boosts arising from the motion of the laboratory in the SUN frame. These are of two types, daily (sidereal) variations determined by the Earth's rotational velocity  $\mathbf{v}_E$  and annual variations governed by the orbital velocity  $\mathbf{v}_\oplus$ . Here [17], taking  $c = 1$ ,

$$\mathbf{v}_E = v_E(-\sin \omega_\oplus T_\oplus \mathbf{e}_X + \cos \omega_\oplus T_\oplus \mathbf{e}_Y), \quad (5.9)$$

while

$$\mathbf{v}_\oplus = v_\oplus(\sin \Omega_\oplus T \mathbf{e}_X - \cos \Omega_\oplus T(\cos \eta \mathbf{e}_Y + \sin \eta \mathbf{e}_Z)). \quad (5.10)$$

These velocities are small, with  $v_E = O(10^{-6})$  and  $v_\oplus = O(10^{-4})$ , so we may consider the corresponding Lorentz boosts to  $O(v)$  only (i.e. set the relativistic  $\gamma$  factors to 1).

Now, while the spherical tensor formalism used here for the SME couplings is extremely convenient in considering

the effects of rotations, it is not well suited to analyse Lorentz boosts. In particular, spherical tensors  $\mathcal{V}_{njm}$  characterized by the same  $(j, m)$  labels may have quite different transformations under Lorentz boosts, since they only distinguish the rotational properties of their constituent 4-vectors and tensors in the fundamental theory. For example, even if we consider the simplest case of the  $\mathcal{V}_{njm}^{\text{NR}} = c_{njm}^{\text{NR}} - a_{njm}^{\text{NR}}$  couplings, we find different transformation properties under boosts for the  $c_{njm}^{\text{NR}}$  and  $a_{njm}^{\text{NR}}$  separately.

We therefore need to re-express the spherical tensor couplings in their original Lorentz tensor form to study the effects of Lorentz boosts. We illustrate this first by evaluating the Lorentz boost transformations of the  $c_{njm}^{\text{NR SUN}}$  couplings, which are related to the original  $c_{\mu\nu}$  tensor couplings. This demonstrates most of the general features in considering boosts with the spherical tensors.

First we need a more complete dictionary of the equivalence of the spherical and Lorentz tensor couplings than given in Paper 2. In the SUN frame, we find [6]

$$\begin{aligned} \frac{1}{\sqrt{4\pi}} c_{000}^{\text{NR SUN}} &= m_w c_{TT}, \\ \sqrt{\frac{3}{4\pi}} c_{11m=0,1,-1}^{\text{NR SUN}} &= 2c_{TZ}, \quad -\sqrt{2}(c_{TX} - ic_{TY}), \quad \sqrt{2}(c_{TX} + ic_{TY}), \\ \frac{1}{\sqrt{4\pi}} c_{200}^{\text{NR SUN}} &= \frac{1}{3m_w} \left( c_{KK} + \frac{3}{2} c_{TT} \right), \\ \sqrt{\frac{5}{4\pi}} c_{220}^{\text{NR SUN}} &= -\frac{1}{3m_w} (c_{XX} + c_{YY} - 2c_{ZZ}), \end{aligned} \quad (5.11)$$

while more generally,

$$c_{22m}^{\text{NR SUN}} = \frac{1}{m_w} C_{JK}^m c_{JK}, \quad (5.12)$$

where  $C_{JK}^m$  are the expansion coefficients for the product of two vectors in spherical harmonics given in [22].<sup>16</sup> Recall

<sup>16</sup>The  $C_{JK}^m$  are defined from the expansion  $p_J p_K = p^2(\frac{1}{3}\delta_{JK} + C_{JK}^m Y_{2m})$ . Explicitly,

$$\begin{aligned} C_{JK}^0 &= \sqrt{\frac{2\pi}{15}} \sqrt{\frac{2}{3}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{pmatrix}, \quad C_{JK}^1 = \sqrt{\frac{2\pi}{15}} \begin{pmatrix} 0 & 0 & -1 \\ 0 & 0 & i \\ -1 & i & 0 \end{pmatrix}, \\ C_{JK}^2 &= \sqrt{\frac{2\pi}{15}} \begin{pmatrix} 1 & -i & 0 \\ -i & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \end{aligned}$$

that here  $m_w$  denotes the mass of the particle of type  $w = e, p$ . We have omitted the matching label  $w$  on the SME couplings to avoid overburdening the notation.

Under a Lorentz boost the transformations of the spherical tensors entering the energy level formulas are,

$$\frac{1}{\sqrt{4\pi}} \delta c_{000}^{\text{NR SUN}} = 2m_w v^J c_{TJ}, \quad (5.13)$$

$$\frac{1}{\sqrt{4\pi}} \delta c_{200}^{\text{NR SUN}} = \frac{5}{3m_w} v^J c_{TJ}, \quad (5.14)$$

and

$$\sqrt{\frac{5}{4\pi}} \delta c_{220}^{\text{NR SUN}} = -\frac{2}{3m_w} (v^J c_{TJ} - 3v^Z c_{TZ}). \quad (5.15)$$

Now, since in this paper we are expressing everything in the spherical tensor formalism, the next step is to rewrite

these in terms of the  $c_{njm}^{\text{NR SUN}}$  couplings. First, writing the velocity in the SUN frame in its spherical vector form with components  $v_0 = v_Z$ ,  $v_{m=\pm 1} = \mp \frac{1}{\sqrt{2}}(v_X \pm i v_Y)$ , we find using (5.11),

$$\delta c_{000}^{\text{NR SUN}} = -\sqrt{3} m_w v_m c_{11m}^{\text{NR SUN}}, \quad (5.16)$$

$$\delta c_{200}^{\text{NR SUN}} = -\frac{5}{2\sqrt{3}} \frac{1}{m_w} v_m c_{11m}^{\text{NR SUN}}, \quad (5.17)$$

and

$$\delta c_{220}^{\text{NR SUN}} = \frac{1}{\sqrt{15}} \frac{1}{m_w} \left( v_m c_{11m}^{\text{NR SUN}} - 3 v_0 c_{110}^{\text{NR SUN}} \right). \quad (5.18)$$

For the velocities in (5.9), (5.10) we have,

$$v_{E0} = 0, \quad v_{Em} = -\frac{i}{\sqrt{2}} v_E e^{im\omega_\oplus T_\oplus}, \quad (m = \pm 1), \quad (5.19)$$

and

$$v_{\oplus 0} = -\sin \eta \cos \Omega_\oplus T, \\ v_{\oplus m} = \frac{i}{2\sqrt{2}} \left[ (1 + \cos \eta) e^{im\Omega_\oplus T} - (1 - \cos \eta) e^{-im\Omega_\oplus T} \right], \\ (m = \pm 1). \quad (5.20)$$

It follows that for sidereal variations, the nonisotropic term in (5.18) does not contribute.

We immediately see from (5.17), (5.18) that the Lorentz boost has introduced a sensitivity to a new set of spherical tensor couplings, in this case  $c_{11m}^{\text{NR SUN}}$ , which did not appear in the original energy level formulas. The numerical factors are largely specific to the  $c_{njm}^{\text{NR SUN}}$  couplings, but we can understand the qualitative features that determine which couplings arise. First, they must satisfy the angular momentum addition rules for the combination of the couplings with the  $(1, m)$  components of the vector  $v_m$ . Then, the  $n$  label must change by  $\pm 1$  to maintain the matching with the power of  $|\mathbf{p}|^n$  in the nonrelativistic Hamiltonian defining the couplings. Finally, the range of allowed  $j$  is further constrained by the restrictions  $j = n, n-2, \dots \geq 0$  on the  $c_{njm}^{\text{NR}}$  couplings [6]. Taken together, these constraints explain the occurrence of the  $c_{11m}^{\text{NR SUN}}$  coupling in the boosts of  $c_{200}^{\text{NR SUN}}$  and  $c_{220}^{\text{NR SUN}}$ . The non-isotropy of  $c_{220}^{\text{NR SUN}}$  is apparent in the independent presence of  $c_{110}^{\text{NR SUN}}$  in (5.18).

Once again, as in (5.8) for  $\Delta E_{\text{SME}\pm}^n$ , these transformations should be inserted into the expressions for the energy levels to see the effect of the boost-induced sidereal or annual variations, which may in principle be used to isolate the dependence on the extra couplings  $c_{11m}^{\text{NR SUN}}$ .

The obvious question now is whether the same results apply also to the corresponding CPT odd couplings  $a_{njm}^{\text{NR SUN}}$  which so far we have treated together in the coupling  $\mathcal{V}_{njm}^{\text{NR SUN}}$ . However, here the answer is no. These couplings arise from a different tensor  $a_{\mu\nu\lambda}$  in the fundamental Lagrangian. This is of rank 3 compared to the rank 2  $c_{\mu\nu}$  and the couplings therefore have quite different Lorentz transformations.

In this case, comparing the nonrelativistic Hamiltonian with the fundamental Lagrangian, and focusing just on the dimension 5  $a_{\mu\nu\lambda}$  couplings (i.e. ignoring the dimension 3  $a_\mu$ ), we can show,

$$\begin{aligned} \frac{1}{\sqrt{4\pi}} a_{000}^{\text{NR SUN}} &= m_\omega^2 a_{TTT}, \\ \sqrt{\frac{3}{4\pi}} a_{11m=0,1,-1}^{\text{NR SUN}} &= -3m_w a_{TTZ}, \quad 3m_w a_{TT-}, \quad -3m_w a_{TT+}, \\ \frac{1}{\sqrt{4\pi}} a_{200}^{\text{NR SUN}} &= a_{TTT} + a_{TKK}, \\ \sqrt{\frac{5}{4\pi}} a_{220}^{\text{NR SUN}} &= -(a_{TXX} + a_{TYY} - 2a_{TZZ}), \end{aligned} \quad (5.21)$$

while more generally,

$$a_{22m}^{\text{NR SUN}} = 3C_{JK}^m a_{TJK}, \quad (5.22)$$

with  $C_{JK}^m$  as above. Here, we define  $a_{TT\pm} = \frac{1}{\sqrt{2}}(a_{TTX} \pm i a_{TTY})$ .

The Lorentz boosts give,

$$\frac{1}{\sqrt{4\pi}} \delta a_{000}^{\text{NR SUN}} = 3m_w^2 v^J a_{TTJ}, \quad (5.23)$$

$$\frac{1}{\sqrt{4\pi}} \delta a_{200}^{\text{NR SUN}} = 5v^J a_{TTJ} + v^J a_{JKK}, \quad (5.24)$$

and

$$\begin{aligned} \sqrt{\frac{5}{4\pi}} \delta a_{220}^{\text{NR SUN}} &= -2v^J a_{TTJ} - v^J a_{JKK} \\ &\quad + 3v^J a_{JZZ} + 6v^Z a_{TTZ}. \end{aligned} \quad (5.25)$$

Reexpressing in spherical tensor form we immediately find

$$\delta a_{000}^{\text{NR SUN}} = \sqrt{3} m_w v_m a_{11m}^{\text{NR SUN}}. \quad (5.26)$$

Compared to  $\delta c_{200}^{\text{NR SUN}}$ , the boost for  $a_{200}^{\text{NR SUN}}$  has an extra type of term proportional to the purely space components  $a_{IJK}$  of the fundamental tensor. In general, writing these in spherical tensor form involves the couplings  $a_{3jm}^{\text{NR SUN}}$  with  $j = 1, 3$ . This case is simplified, however, since we only

require the couplings with contracted indices and we can straightforwardly check,

$$\begin{aligned} & \sqrt{\frac{3}{4\pi}} \left( a_{31m}^{\text{NR SUN}} - \frac{1}{2m_w^2} a_{11m}^{\text{NR SUN}} \right)_{m=0,1,-1} \\ &= -\frac{3}{5m_w} a_{KKZ}, \quad \frac{3}{5m_w} a_{KK-}, \quad -\frac{3}{5m_w} a_{KK+}. \end{aligned} \quad (5.27)$$

Then, collecting terms,

$$\delta a_{200}^{\text{NR SUN}} = \frac{5}{\sqrt{3}} m_w v_m \left( a_{31m}^{\text{NR SUN}} + \frac{1}{2m_w^2} a_{11m}^{\text{NR SUN}} \right). \quad (5.28)$$

We therefore find a contribution in which  $n$  has *increased* by 1. This is due to the rank of the underlying fundamental

Lorentz coupling. The allowed  $(j, m)$  labels on the couplings are constrained by the angular momentum addition rules as before.

Turning finally to  $\delta a_{220}^{\text{NR SUN}}$ , we also encounter a non-isotropic tensor coupling  $a_{JZZ}$ . Inspection of the matching conditions from the Hamiltonians shows that this also involves the  $j = 3$  couplings  $a_{33m}^{\text{NR SUN}}$ . After some further calculation we find

$$\begin{aligned} & \sqrt{\frac{7}{4\pi}} a_{330}^{\text{NR SUN}} + \sqrt{\frac{3}{4\pi}} \left( a_{310}^{\text{NR SUN}} - \frac{1}{2m_w^2} a_{110}^{\text{NR SUN}} \right) \\ &= -\frac{1}{m_w} a_{ZZZ}, \end{aligned} \quad (5.29)$$

while

$$\sqrt{3} \sqrt{\frac{7}{4\pi}} a_{33m}^{\text{NR SUN}} + \sqrt{\frac{3}{4\pi}} \left( a_{31m}^{\text{NR SUN}} - \frac{1}{2m_w^2} a_{11m}^{\text{NR SUN}} \right) = \pm \frac{3}{m_w} a_{ZZ\mp}, \quad (m = \pm 1). \quad (5.30)$$

Then, from (5.25) and collecting terms, we can write the Lorentz boost for the coupling  $a_{220}^{\text{NR SUN}}$  as,

$$\begin{aligned} \delta a_{220}^{\text{NR SUN}} &= m_w v_m \left( \sqrt{\frac{21}{5}} a_{33m}^{\text{NR SUN}} - \frac{2}{3} \sqrt{\frac{3}{5}} \left( a_{31m}^{\text{NR SUN}} + \frac{1}{2m_w^2} a_{11m}^{\text{NR SUN}} \right) \right) \\ &\quad - 3m_w v_0 \left( \left( \frac{1}{\sqrt{3}} - 1 \right) \sqrt{\frac{7}{5}} a_{330}^{\text{NR SUN}} - \frac{2}{3} \sqrt{\frac{3}{5}} \left( a_{310}^{\text{NR SUN}} + \frac{1}{2m_w^2} a_{110}^{\text{NR SUN}} \right) \right). \end{aligned} \quad (5.31)$$

This is to be compared with the corresponding result (5.18) for the Lorentz boost of  $c_{220}^{\text{NR SUN}}$ , the key difference being the appearance of couplings with  $n = 3$ .

Evidently, we can continue to evaluate the Lorentz boosts for all the spherical tensor couplings appearing in the relations for the energy levels. Each must be evaluated separately since the underlying fundamental Lorentz tensors will be different in each case. The general principles and all the rules governing which couplings can appear are the same as demonstrated above for the  $c_{njm}^{\text{NR SUN}}$  and  $a_{njm}^{\text{NR SUN}}$  couplings and we will not quote all of these explicitly here.

The key implication for a future program of high-precision spectroscopy is that the SME predicts sidereal, and annual, variations of transition frequencies which, although suppressed by  $O(v_E)$  or  $O(v_\oplus)$ , depend on *extra* spherical tensor couplings with  $n$  and  $j$  differing by 1 compared to those which would appear in time-averaged measurements.

## VI. OUTLOOK

In this paper, extending the work presented in Papers 1 and 2, we have provided a comprehensive account of the effect of the Lorentz and *CPT* violating interactions in the

SME on the rovibrational spectrum of the molecular ions  $\text{H}_2^+$  and  $\bar{\text{H}}_2^-$ . The description has been entirely in terms of the spherical tensor representation of the couplings, which is the most natural framework to include the nonrenormalizable operators in the nonminimal SME.

We have given our results as perturbations on the full hyperfine-Zeeman spectrum, for both large and small background magnetic fields. This makes them directly applicable both to current high-precision spectroscopy of  $\text{H}_2^+$  using low-field RF traps [7] and to future spectroscopy of its antimatter counterpart  $\bar{\text{H}}_2^-$ , which will be performed in a high magnetic field on molecular ions confined in a Penning trap [14].

The effects of the Lorentz and *CPT* violating interactions involving the proton are calculated directly in perturbation theory. For the electron interactions, their direct effect is to modify the internucleon potential. Following Paper 1, we showed how a detailed analysis of the molecular dynamics, noting particularly the lack of full spherical symmetry of the molecule even in its  $1s\sigma_g$  electron ground state, allows this to be translated into changes to the rovibrational energy levels. We also noted how in the Born-Oppenheimer framework, the proton SME

couplings also feed into the inter-nucleon potential, and subsequently the rovibrational spectrum, via (4.10).

In both cases, we quoted the changes to the rovibrational energy levels in complete generality, giving the dependence of the SME contributions on the quantum numbers  $(v, N)$  and spin  $M_J$  of the hyperfine-Zeeman levels at high and low magnetic field.

We then described the importance of looking for daily (sidereal) and annual variations of the transition frequencies induced by rotations and Lorentz boosts in this Lorentz noninvariant theory. These variations are especially significant as they depend on further components of the SME couplings characterized by different  $(n, j, m)$  labels, which would not be present in single or time-averaged measurements.

All this establishes the theoretical basis to interpret, and guide, experimental searches for Lorentz and *CPT* violation in hydrogen and antihydrogen molecular ion spectroscopy.

Rovibrational spectroscopy of  $\text{H}_2^+$  at  $O(10^{-12})$  has already been carried out [7] with the possibility of eventually attaining precisions as high as  $O(10^{-17})$ . Meanwhile, the success of the ALPHA collaboration in high-precision spectroscopy of atomic antihydrogen, especially the  $1S-2S$  transition [11, 12], together with the recent achievement of BASE-STEP in transporting antiprotons away from the electromagnetically noisy environment of the CERN AD/ELENA facility, has opened an increasingly clear path towards future high-precision spectroscopy of  $\bar{\text{H}}_2^-$ .

An important element in achieving such precisions in molecular ion spectroscopy is to put together combinations of specific rovibrational transitions chosen so as to cancel the systematic shifts due to less precisely controllable effects such as the linear and quadratic Zeeman shifts, the electric quadrupole shift, and dependence on the hyperfine spin structure [23]. Each of these has a distinctive dependence on the  $v, N$  and  $M_J$  quantum numbers [24–26], ultimately determined, like the SME contributions, by combinations of Clebsch-Gordan coefficients.

This raises a serious question for the design of experimental protocols to search for Lorentz and *CPT* violation. Inspection of the quantum number dependence of our results for the SME contributions, encoded in the  $c_{NM_J}$ ,  $e_{NM_J}$ , and  $f_{NM_J}$  coefficients, shows that in selecting

combinations of transition frequencies that cancel the systematic shifts listed above, sensitivity to some or all of the SME couplings could be lost. In the most unfavorable case, an extremely precise transition frequency combination may be entirely insensitive to *CPT* violation.

An important next step is therefore to use our results to analyze in detail possible specific combinations of rovibrational transition frequencies that minimize sensitivity to precision-limiting systematic effects while maintaining maximum sensitivity to potential sources of Lorentz and *CPT* violation. This analysis will be presented in forthcoming work.

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## DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

## APPENDIX A: HYPERFINE-ZEEMAN HAMILTONIAN AND EIGENVALUES OF $V_{\text{SME}}^e(\mathbf{R})$

The hyperfine-Zeeman Hamiltonian may be written as

$$H_{HFS} + H_Z = c_e(v, N) \mathbf{N} \cdot \mathbf{S} + g_e \mu_B \mathbf{B} \cdot \mathbf{S} - g_m(v, N) \mu_B \mathbf{B} \cdot \mathbf{N}, \quad (\text{A1})$$

where the couplings  $c_e(v, N)$  and  $g_m(v, N)$  depend on the rovibrational state. Its expectation value in the states  $|vNJM_J\rangle$  can be written for fixed  $M_J$  (which remains a good quantum number in the presence of the applied magnetic field  $\mathbf{B}$ ) as a  $2 \times 2$  matrix with rows/columns corresponding to  $J, J' = N \pm \frac{1}{2}$  as follows,

$$\langle vNJ'M_J | H_{HFS} + H_Z | vNJM_J \rangle = \begin{pmatrix} A & B \\ B & D \end{pmatrix}, \quad (\text{A2})$$

with

$$\begin{aligned} A &= \frac{1}{2} N c_e(v, N) + \frac{1}{2N+1} (g_e - 2N g_m(v, N)) \mu_B B M_J, \\ D &= -\frac{1}{2} (N+1) c_e(v, N) - \frac{1}{2N+1} (g_e + 2(N+1) g_m(v, N)) \mu_B B M_J, \\ B &= -\frac{1}{2N+1} \sqrt{\left(N + \frac{1}{2}\right)^2 - M_J^2} (g_e + g_m(v, N)) \mu_B B. \end{aligned} \quad (\text{A3})$$

With the matrix elements  $V_{\text{SME}}^e(R)$  in the same states written as

$$V_{\text{SME}}^e(R) = \begin{pmatrix} \alpha & \beta \\ \beta^* & \delta \end{pmatrix}, \quad (\text{A4})$$

the SME contributions to the combined eigenvalues are given by (Paper 2, footnote 9),

$$V_{\text{SME}\pm}^e(R) = \frac{1}{2}(\alpha + \delta) \pm \frac{1}{2} \frac{1}{\sqrt{(A-D)^2 + 4B^2}} \times ((A-D)(\alpha - \delta) + 2B(\beta + \beta^*)). \quad (\text{A5})$$

In the case considered here, the off-diagonal element  $B$  is proportional to the magnetic field, so in the small-field regime where we can neglect terms of  $O(\mu_B B/c_e)^2$ , we have the simple forms,

$$\begin{aligned} V_{\text{SME}+}^e(R) &= \alpha + 2 \frac{B}{A-D} \text{Re}\beta, \\ V_{\text{SME}-}^e(R) &= \delta - 2 \frac{B}{A-D} \text{Re}\beta. \end{aligned} \quad (\text{A6})$$

From above, we find at  $O(B)$ ,

$$\frac{B}{A-D} = -\frac{2}{(2N+1)^2} \sqrt{\left(N + \frac{1}{2}\right)^2 - M_J^2} B, \quad (\text{A7})$$

where we define

$$B = \frac{1}{c_e(v, N)} (g_e + g_m(v, N)) \mu_B B. \quad (\text{A8})$$

This allows the  $O(B)$  corrections to the results quoted in the text for  $V_{\text{SME}\pm}^e(R)$  to be read off directly. For example, to calculate the  $B$  dependence of the coefficients  $\hat{c}_{NM_J}^\pm(B)$  in (3.17), (3.18) in the text, we apply this formula to the coefficient of  $\mathcal{V}_{220}^{e\text{NR}}$  in  $V_{\text{SME}}^{e\mathcal{V}}(R)$  where, taking out the common factor  $\frac{1}{2} \sqrt{\frac{5}{4\pi}} \text{tr}_Y \langle p_a p_b \rangle \mathcal{V}_{220}^{e\text{NR}}$ , we have from Paper 2, Appendix B,

$$\begin{aligned} \alpha &= \frac{1}{(2N+1)(2N+3)} \left[ \left(N + \frac{1}{2}\right) \left(N + \frac{3}{2}\right) - 3M_J^2 \right], \\ \delta &= \frac{1}{(2N-1)(2N+1)} \left[ \left(N - \frac{1}{2}\right) \left(N + \frac{1}{2}\right) - 3M_J^2 \right], \\ \beta &= -6 \frac{1}{(2N-1)(2N+1)(2N+3)} \sqrt{\left(N + \frac{1}{2}\right)^2 - M_J^2} M_J, \end{aligned} \quad (\text{A9})$$

and find

$$\begin{aligned} \hat{c}_{NM_J}^+(B) &= \frac{1}{(2N+1)(2N+3)} \left[ \left(N + \frac{1}{2}\right) \left(N + \frac{3}{2}\right) - 3M_J^2 \right], \\ &\quad + 24 \frac{[(N + \frac{1}{2})^2 - M_J^2] M_J}{(2N+1)^3 (2N-1)(2N+3)} B, \end{aligned} \quad (\text{A10})$$

and

$$\begin{aligned} \hat{c}_{NM_J}^-(B) &= \frac{1}{(2N-1)(2N+1)} \left[ \left(N - \frac{1}{2}\right) \left(N + \frac{1}{2}\right) - 3M_J^2 \right] \\ &\quad - 24 \frac{[(N + \frac{1}{2})^2 - M_J^2] M_J}{(2N+1)^3 (2N-1)(2N+3)} B. \end{aligned} \quad (\text{A11})$$

In the large magnetic field limit, the eigenstates of the hyperfine-Zeeman Hamiltonian are  $|vNM_J M_S\rangle$  (or equivalently,  $|vNM_N M_S\rangle$  since  $M_J = M_N + M_S$ ) so these are the appropriate states to use for large fields. In this case the matrix elements  $\langle H_{\text{HFS}} + H_Z \rangle$  are written for fixed  $M_J$  in  $2 \times 2$  matrix form with  $M'_S, M_S = \pm 1/2$  as,

$$\langle vNM_J M'_S | H_{\text{HFS}} + H_Z | vNM_J M_S \rangle = \begin{pmatrix} A & B \\ B & D \end{pmatrix}, \quad (\text{A12})$$

where now [see Paper 2, Eq. (5.26)],

$$\begin{aligned} A &= \frac{1}{2} c_e(v, N) \left( M_J - \frac{1}{2} \right) + \left( \frac{1}{2} g_e - g_m(v, N) \right) \left( M_J - \frac{1}{2} \right) \mu_B B, \\ D &= -\frac{1}{2} c_e(v, N) \left( M_J + \frac{1}{2} \right) - \left( \frac{1}{2} g_e + g_m(v, N) \right) \left( M_J + \frac{1}{2} \right) \mu_B B, \\ B &= \frac{1}{2} c_e(v, N) \sqrt{\left(N + \frac{1}{2}\right)^2 - M_J^2}. \end{aligned} \quad (\text{A13})$$

The matrix elements  $V_{\text{SME}}^{e\mathcal{V}}(R)$  for the spin-independent couplings are of course diagonal in this basis, so we just have  $V_{\text{SME}}^{e\mathcal{V}}(R) = \begin{pmatrix} \alpha & 0 \\ 0 & \delta \end{pmatrix}$  with the  $O(p^2)$  terms,

$$\begin{aligned} \alpha &= -\frac{1}{\sqrt{4\pi}} \left[ \text{tr} \langle p_a p_b \rangle \mathcal{V}_{200}^{e\text{NR}} - \frac{\sqrt{5}}{2} \text{tr}_Y \langle p_a p_b \rangle c_{NM_J - \frac{1}{2}} \mathcal{V}_{220}^{e\mathcal{V}} \right], \\ \delta &= -\frac{1}{\sqrt{4\pi}} \left[ \text{tr} \langle p_a p_b \rangle \mathcal{V}_{200}^{e\text{NR}} - \frac{\sqrt{5}}{2} \text{tr}_Y \langle p_a p_b \rangle c_{NM_J + \frac{1}{2}} \mathcal{V}_{220}^{e\mathcal{V}} \right]. \end{aligned} \quad (\text{A14})$$

In this case, we find the magnetic field corrections of  $(c_e/\mu_B B)$  vanish, since  $\beta = 0$ , but we can use the formula (A5) to calculate the next order corrections. We find

$$c_{NM_J \mp \frac{1}{2}} \rightarrow c_{NM_J \mp \frac{1}{2}} \mp \frac{3}{2} \frac{[(N + \frac{1}{2})^2 - M_J^2] M_J}{(2N - 1)(2N + 3)} \left( \frac{c_e(v, N)}{(g_e + g_m(v, N)) \mu_B B} \right)^2. \quad (\text{A15})$$

For the spin-dependent couplings, the matrix elements of  $V_{\text{SME}}^{e\mathcal{T}}(\mathbf{R})$  in the states  $|vNM_J M_S\rangle$  take the form, with matrix rows/columns corresponding to  $M_S = \pm \frac{1}{2}$ ,

$$V_{\text{SME}}^{e\mathcal{T}}(R) = \begin{pmatrix} \alpha & \beta \\ \beta^* & \delta \end{pmatrix}, \quad (\text{A16})$$

with

$$\begin{aligned} \alpha &= -\mathcal{T}_{010}^{e\text{NR}(0B)} - \frac{1}{3} \text{tr} \langle p_a p_b \rangle \left( \mathcal{T}_{210}^{e\text{NR}(0B)} + 2\mathcal{T}_{210}^{e\text{NR}(1B)} \right) \\ &\quad + \frac{1}{3} \text{tr}_Y \langle p_a p_b \rangle c_{NM_J - \frac{1}{2}} \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} + \frac{3}{2} \sqrt{\frac{7}{6}} \mathcal{T}_{230}^{e\text{NR}(0B)} \right), \end{aligned} \quad (\text{A17})$$

$$\begin{aligned} \delta &= \mathcal{T}_{010}^{e\text{NR}(0B)} + \frac{1}{3} \text{tr} \langle p_a p_b \rangle \left( \mathcal{T}_{210}^{e\text{NR}(0B)} + 2\mathcal{T}_{210}^{e\text{NR}(1B)} \right) \\ &\quad - \frac{1}{3} \text{tr}_Y \langle p_a p_b \rangle c_{NM_J + \frac{1}{2}} \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} + \frac{3}{2} \sqrt{\frac{7}{6}} \mathcal{T}_{230}^{e\text{NR}(0B)} \right), \end{aligned} \quad (\text{A18})$$

$$\begin{aligned} \beta &= -\text{tr}_Y \langle p_a p_b \rangle \frac{\sqrt{(N + \frac{1}{2})^2 - M_J^2}}{(2N - 1)(2N + 3)} M_J \\ &\quad \times \left( \mathcal{T}_{210}^{e\text{NR}(0B)} - \mathcal{T}_{210}^{e\text{NR}(1B)} - \sqrt{\frac{7}{6}} \mathcal{T}_{230}^{e\text{NR}(0B)} - \sqrt{5} i \mathcal{T}_{220}^{e\text{NR}(1E)} \right). \end{aligned} \quad (\text{A19})$$

In this case, since  $\beta \neq 0$ , there is a correction of  $O(1/B)$  to the eigenvalues  $V_{\text{SME}\pm}^{e\mathcal{T}}(R)$  and we can again use the formula (A6), but this time for small  $c_e/\mu_B B$  where we have from (A13),

$$\frac{B}{A - D} = \frac{1}{2} \sqrt{\left(N + \frac{1}{2}\right)^2 - M_J^2} \frac{1}{B} + O(1/B)^2. \quad (\text{A20})$$

Together with the coefficients  $\alpha, \delta, \beta$  above, we find  $V_{\text{SME}\pm}^{e\mathcal{T}}(R)$  as given in the main text, (3.52).

## APPENDIX B: CLEBSCH-GORDAN RELATIONS

Extending the set of identities given in Paper 2, Appendix B, to those required for the  $O(p^4, P^4)$  SME contributions considered here, we find

$$\begin{aligned}
\sum_{M_S} (C_{NM_N, \frac{1}{2}M_S}^{N+\frac{1}{2}M_J})^2 c_{NM_N}^{(4)} &\equiv \hat{c}_{NM_J}^{(4)+} \\
&= \frac{3}{64} \frac{3(2N+5)(2N+3)(2N+1)(2N-1) - 40(12N^2 + 24N - 1)M_J^2 + 560M_J^4}{(2N+5)(2N+3)(2N-1)(2N-3)}, \\
\sum_{M_S} (C_{NM_N, \frac{1}{2}M_S}^{N-\frac{1}{2}M_J})^2 c_{NM_N}^{(4)} &\equiv \hat{c}_{NM_J}^{(4)-} \\
&= \frac{3}{64} \frac{3(2N+3)(2N+1)(2N-1)(2N-3) - 40(12N^2 - 13)M_J^2 + 560M_J^4}{(2N+3)(2N+1)(2N-1)(2N-3)}, \\
\sum_{M_S} C_{NM_N, \frac{1}{2}M_S}^{N+\frac{1}{2}M_J} C_{NM_N, \frac{1}{2}M_S}^{N-\frac{1}{2}M_J} c_{NM_N}^{(4)} &= -\frac{15}{4} \frac{(12N(N+1) - 17 - 28M_J^2)[(N+\frac{1}{2})^2 - M_J^2]M_J}{(2N+5)(2N+3)(2N+1)(2N-1)(2N-3)}, \tag{B1}
\end{aligned}$$

where  $c_{NM_N}^{(4)}$ , defined as

$$C_{NM_N, 40}^{NM_N} C_{N0, 40}^{N0} = c_{NM_N}^{(4)}, \tag{B2}$$

is given explicitly by,

$$c_{NM_N}^{(4)} = \frac{3}{4} \frac{[3N(N+2)(N+1)(N-1) - 5(6N^2 + 6N - 5)M_N^2 + 35M_N^4]}{[(2N+5)(2N+3)(2N-1)(2N-3)]}. \tag{B3}$$

### APPENDIX C: PROTON MOMENTUM EXPECTATION VALUES

In the main text, we quote the expectation values  $\langle vN|P^2|vN\rangle$  and  $\langle vN|P^4|vN\rangle$  as expansions in  $(v + \frac{1}{2})$  and  $N(N+1)$ , characterized by the small parameter  $\lambda \sim O(\sqrt{m_e/m_p})$ . Here we briefly illustrate a general method to find the expectation values of any operator in these rovibrational states and apply it to the special case of  $P^2$  and  $P^4$ . The method is essentially described in Appendix B of Paper 2, which we use extensively though many of the results below are new.

The eigenstates  $|vN\rangle$  of the proton Schrödinger equation are found by expanding about the minimum  $R_0$  of the internucleon potential, treating the angular momentum-induced effective potential  $V_N(R)$  as a perturbation. Recall that  $V_M(R)$  is obtained by solving the electron Schrödinger equation, while  $V_N(R) = N(N+1)/2\mu R^2$ . The expansion parameter is  $\lambda = 1/\mu\omega_0 R_0^2$ .

The unperturbed states are therefore those of a SHO with potential  $\frac{1}{2}\mu\omega_0^2 x^2$ , where  $x = R - R_0$  and  $\mu\omega_0^2 = V_M''(R_0)$ . The total perturbation, before including the SME, is  $\delta V = \delta V_M + \delta V_N$  where,

$$\begin{aligned}
\delta V_M &= \frac{1}{6} V_M''' x^3 + \dots \\
\delta V_N &= \lambda\omega_0 N(N+1) \left( -\frac{x}{R_0} + \frac{3}{2} \frac{x^2}{R_0^2} + \dots \right). \tag{C1}
\end{aligned}$$

In what follows we present results up to second order in perturbation theory, truncating the  $\lambda$  expansion at  $O(\lambda^2)$ . Higher order terms are calculated in Paper 1.

First consider the expectation value  $\langle vN|P^2|vN\rangle$ . In the nucleon Schrödinger equation the 3-dim momentum squared  $P^2$  is represented as the Laplacian acting on the appropriate wave function, with

$$\begin{aligned}
P^2 &\rightarrow -\nabla_R^2 = -\frac{d^2}{dR^2} + N(N+1) \frac{1}{R_0^2} \\
&= k^2 + N(N+1) \frac{1}{R_0^2} \left( 1 - 2 \frac{x}{R_0} + 3 \frac{x^2}{R_0^2} + \dots \right), \tag{C2}
\end{aligned}$$

where we denote the 1-dim SHO momentum by  $k = -id/dx$ . We therefore need to find the expectation values of the SHO operators  $k^2$ ,  $x$  and  $x^2$  in the states  $|vN\rangle$  up to second order in the perturbations  $\delta V$ . Taking intermediate results from Paper 1, we find,

$$\langle k^2 \rangle = \mu\omega_0 \left( v + \frac{1}{2} \right) \left[ 1 + \frac{1}{2} N(N+1) \left( 3 + \frac{R_0 V_M'''}{V_M''} \right) \lambda^2 \right], \tag{C3}$$

$$\frac{1}{R_0^3} \langle x \rangle = -\frac{1}{2} \mu\omega_0 \left( v + \frac{1}{2} \right) \left( \frac{R_0 V_M'''}{V_M''} \right) \lambda^2, \tag{C4}$$

$$\frac{1}{R_0^4} \langle x^2 \rangle = \mu\omega_0 \left( v + \frac{1}{2} \right) \lambda^2. \tag{C5}$$

Collecting terms, recalling  $\mu = \frac{1}{2}m_p$ , and truncating at  $O(\lambda^2)$  gives<sup>17</sup>

$$\langle vN|P^2|vN\rangle = \frac{1}{2}m_p\omega_0\left[\left(v+\frac{1}{2}\right) + N(N+1)\lambda + \frac{3}{2}\left(3 + \frac{R_0V_M'''}{V_M''}\right)\left(v+\frac{1}{2}\right)N(N+1)\lambda^2 + \dots\right]. \quad (C6)$$

The analysis of  $\langle vN|P^4|vN\rangle$  follows along the same lines, though is inevitably more complicated. Here, in the nucleon Schrödinger equation,

$$\begin{aligned} P^4 &\rightarrow \nabla_R^2 \nabla_R^2 = \frac{d^4}{dR^4} - N(N+1)\frac{1}{R^2}\left(2\frac{d^2}{dR^2} - \frac{4}{R}\frac{d}{dR} + \frac{6}{R^2}\right) + (N(N+1))^2\frac{1}{R^2} \\ &= k^4 + N(N+1)\frac{1}{R_0^2}\left(2\left(1+3\frac{x^2}{R_0^2}\right)k^2 + 4\frac{1}{R_0}\left(1-3\frac{x}{R_0}\right)ik - \frac{6}{R_0^2}\right) \\ &\quad + (N(N+1))^2\frac{1}{R_0^4} + \dots \end{aligned} \quad (C7)$$

keeping only terms which give relevant contributions at the order considered here. Again working up to second order in perturbation theory making extensive use of results from Paper 1, and truncating at  $O(\lambda^2)$ , we find

$$\langle k^4 \rangle = \frac{3}{2}(\mu\omega_0)^2\left[\left(v+\frac{1}{2}\right)^2 + \frac{1}{4}\right]\left[1 + N(N+1)\left(3 + \frac{R_0V_M'''}{V_M''}\right)\lambda^2\right], \quad (C8)$$

$$\frac{1}{R_0^4}\langle x^2k^2 \rangle = \frac{1}{2}(\mu\omega_0)^2\left[\left(v+\frac{1}{2}\right)^2 - \frac{3}{4}\right]\lambda^2, \quad (C9)$$

$$\frac{1}{R_0^3}\langle ik \rangle = O(\lambda^3), \quad (C10)$$

$$\frac{1}{R_0^4}\langle ixk \rangle = -\frac{1}{2}(\mu\omega_0)^2\lambda^2, \quad (C11)$$

in addition to those quoted above. Collecting all these terms, we obtain

$$\begin{aligned} \langle vN|P^4|vN\rangle &= (\mu\omega_0)^2\left[\frac{3}{2}\left[\left(v+\frac{1}{2}\right)^2 + \frac{1}{4}\right]\left(1 + N(N+1)\left(3 + \frac{R_0V_M'''}{V_M''}\right)\lambda^2\right) \right. \\ &\quad \left. + 2\left(v+\frac{1}{2}\right)N(N+1)\lambda + 3\left[\left(v+\frac{1}{2}\right)^2 - \frac{3}{4}\right]N(N+1)\lambda^2 + (N(N+1))^2\lambda^2 + \dots\right]. \end{aligned} \quad (C12)$$

Omitting the terms of  $O((N(N+1))^2)$  and  $O((v+\frac{1}{2})^2N(N+1))$ , which do not contribute to  $\chi_{\text{SME}}^n$ ,  $B_{\text{SME}}^n$  and  $\alpha_{\text{SME}}^n$ , this is the result quoted in the main text, (4.23). Notice the coefficient of  $N(N+1)$  which appears there. This arises because of contributions from the  $O(1)$  fractions which automatically accompany the terms of  $O(v+\frac{1}{2})^2$ .

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<sup>17</sup>We have displayed here only the terms contributing to the coefficients  $\delta_{\text{SME}}^n$ ,  $B_{\text{SME}}^n$  and  $\alpha_{\text{SME}}^n$  in the rovibrational energy levels (1.3). In particular we have omitted the  $O(\lambda)$  terms proportional to  $(v+\frac{1}{2})^2$  contributing to the  $x_{\text{SME}}^n$  coefficient; these depend on the third and fourth derivatives of  $V_M$  and are quoted in Paper 1.

- [1] M. Charlton, S. Eriksson, and G. M. Shore, *Antihydrogen and Fundamental Physics* (Springer, New York, 2020), 10.1007/978-3-030-51713-7.
- [2] S. Schiller, Precision spectroscopy of molecular hydrogen ions: An introduction, *Contemp. Phys.* **63**, 247 (2022) and Supplementary material.
- [3] G. M. Shore, Lorentz and *CPT* violation and the hydrogen and antihydrogen molecular ions. I. Rovibrational states, *Phys. Rev. D* **112**, 056015 (2025).
- [4] G. M. Shore, Lorentz and *CPT* violation and the hydrogen and antihydrogen molecular ions. II. Hyperfine-Zeeman spectrum, *Phys. Rev. D* **112**, 056016 (2025).
- [5] D. Colladay and V. A. Kostelecky, Lorentz violating extension of the standard model, *Phys. Rev. D* **58**, 116002 (1998).
- [6] A. Kostelecký and M. Mewes, Fermions with Lorentz-violating operators of arbitrary dimension, *Phys. Rev. D* **88**, 096006 (2013).
- [7] M. R. Schenkel, S. Alighanbari, and S. Schiller, Laser spectroscopy of a rovibrational transition in the molecular hydrogen ion  $H_2^+$ , *Nat. Phys.* **20**, 383 (2024).
- [8] M. C. Zammit *et al.*, Laser-driven production of the antihydrogen molecular ion, *Phys. Rev. A* **100**, 042709 (2019).
- [9] E. G. Myers, *CPT* tests with the antihydrogen molecular ion, *Phys. Rev. A* **98**, 010101(R) (2018).
- [10] M. C. Zammit, C. J. Baker, S. Jonsell, S. Eriksson, and M. Charlton, Antihydrogen chemistry, *Phys. Rev. A* **111**, 050101 (2025).
- [11] M. Ahmadi *et al.* (ALPHA Collaboration), Characterization of the  $1S$ - $2S$  transition in antihydrogen, *Nature (London)* **557**, 71 (2018).
- [12] C. J. Baker *et al.*, Precision spectroscopy of the hyperfine components of the  $1S$ - $2S$  transition in antihydrogen, *Nat. Phys.* **21**, 201 (2025).
- [13] V. A. Kostelecky and N. Russell, Data tables for Lorentz and *CPT* violation, *Rev. Mod. Phys.* **83**, 11 (2011).
- [14] S. Schiller *et al.*, Towards a future test of *CPT* invariance by spectroscopy of  $H_2^+$  and anti- $H_2^+$ , in *Talk at Workshop All that Antimatters in the Universe* (CERN, Geneva, 2026).
- [15] A. J. Vargas, Prospects for testing Lorentz and *CPT* symmetry with  $H_2^+$  and  $\bar{H}_2^+$ , [arXiv:2503.06306](https://arxiv.org/abs/2503.06306).
- [16] A. J. Vargas, Nonminimal Lorentz violation in atomic and molecular spectroscopy experiments, [arXiv:2603.08298](https://arxiv.org/abs/2603.08298).
- [17] V. A. Kostelecký and A. J. Vargas, Lorentz and *CPT* tests with hydrogen, antihydrogen, and related systems, *Phys. Rev. D* **92**, 056002 (2015).
- [18] H. Muller, S. Herrmann, A. Saenz, A. Peters, and C. Lammerzahl, Tests of Lorentz invariance using hydrogen molecules, *Phys. Rev. D* **70**, 076004 (2004).
- [19] V. A. Kostelecky and C. D. Lane, Nonrelativistic quantum Hamiltonian for Lorentz violation, *J. Math. Phys. (N.Y.)* **40**, 6245 (1999).
- [20] A. Carrington, I. R. McNab, and C. A. Montgomerie, Spectroscopy of the hydrogen molecular ion, *J. Phys. B* **22**, 3551 (1989).
- [21] D. A. Varshalovich, A. N. Moskalev, and V. K. Khersonskii, *Quantum Theory of Angular Momentum* (World Scientific, Singapore, 1988).
- [22] T. J. Yoder and G. S. Adkins, Higher order corrections to the hydrogen spectrum from the standard-model extension, *Phys. Rev. D* **86**, 116005 (2012).
- [23] S. Schiller and V. I. Korobov, Cancelling spin-dependent contributions and systematic shifts in precision spectroscopy of molecular hydrogen ions, *Phys. Rev. A* **98**, 022511 (2018).
- [24] V. I. Korobov, L. Hilico, and J.-Ph. Karr, Hyperfine structure in the hydrogen molecular ion, *Phys. Rev. A* **74**, 040502(R) (2006).
- [25] J.-P. Karr *et al.*, Vibrational spectroscopy of  $H_2^+$ : Hyperfine structure of two-photon transitions, *Phys. Rev. A* **77**, 06430 (2008).
- [26] J.-Ph. Karr, V. I. Korobov, and L. Hilico, Vibrational spectroscopy of  $H_2^+$ : Precise evaluation of the Zeeman effect, *Phys. Rev. A* **77**, 062507 (2008).