

Near degeneracies between opposite parity states in the heavy DyAg molecule

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We theoretically investigate the rare-earth $^{163}\text{Dy}^{107}\text{Ag}$ molecule as a quantum sensor to amplify weak symmetry-breaking interactions not included in the standard model of physics. We focus on parity-breaking interactions from the octupole-deformed ^{163}Dy nucleus. To characterize key properties for this molecule, we have performed relativistic electronic configuration-interaction and rovibrational coupled-channels calculations. We show that near the equilibrium geometry, the bond is ionic with the Dy atom losing an electron from its open, submerged 4f electron shell to Ag. In fact, the $^{163}\text{Dy}^{107}\text{Ag}$ ground-state configuration corresponds to a Dy ion with a large electronic angular momentum of $j_{\text{Dy}^+} = 15\hbar/2$ and a spinless silver ion. Here, $\hbar$ is the reduced Planck constant. We also observe that the crystal-field splittings among states with projections Ω of j_{Dy^+} along the internuclear axis are on the order of the vibrational spacings in $^{163}\text{Dy}^{107}\text{Ag}$ and orders of magnitude larger than the rotational spacings, Coriolis-induced Ω -type doubling, the hyperfine interactions between the spins of the electrons and the ^{163}Dy nucleus, and Zeeman interactions when an external magnetic field is applied. The spin-stretched state with $|\Omega| = j_{\text{Dy}^+}$ is the ground state. The splittings between the Ω -doublets, a pair of states with opposite parity, dramatically depend on Ω . We predict values on the order of $h \times 10\text{ GHz}$, $h \times 10\text{ MHz}$, and $h \times 1\text{ kHz}$ for $\Omega = \hbar/2$, $3\hbar/2$, and $5\hbar/2$, respectively, with Planck constant h . For larger values of Ω , the splittings decrease by additional orders of magnitude. Parity violations and thus the mixing of the Ω -doublets can be achieved by simply applying external electric fields or by interactions induced by the octupole-deformed ^{163}Dy nucleus through coupling of its nuclear spin to molecular rotation. The actual energy shifts from the latter contribution are not known as they originate from nuclear forces.

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I. INTRODUCTION

Experimental studies of lanthanide atoms in the gas phase are used to search for symmetry-breaking forces in quantum electrodynamics and chromodynamics. These interests are not only generated by their nuclear structure but also by their intriguing electronic structure. Most attention is devoted to the stable dysprosium isotope ^{163}Dy with its two nearly degenerate nuclear states of opposite parity [1–4]. In fact, experiments using radiofrequency electronic-electric-dipole transitions in Dy to search for time variations in the fine-structure constant have been performed [5–7] while Zeeman-tuned level crossings between two electronic hyperfine states of opposite parity have been suggested as a means to detect parity violations [8]. In addition, dysprosium can be sensitive to violations of special and general relativity and used to test Lorentz invariance [9–11].

Investigations into symmetry-breaking forces in the ^{163}Dy isotope could be improved by bonding it to atoms with a large

electron affinity. In addition, ultracold and trapped molecules at temperatures below 1 mK can provide long spin-coherence times and thus excellent energy resolution. In recent theoretical proposals [12,13], DyO and DyF molecules were suggested as candidates with high sensitivity to parity violating forces. This proposal was followed by hyperfine-resolved spectroscopy of two isotopologues of DyO [14]. Reference [15] considered states of DyOH and ErOH as relevant for fundamental symmetry tests. In their absolute ground state, DyOH and ErOH are linear molecules with a nonzero projection Ω of the total electronic angular momentum along the molecular axis as a good quantum number. This leads to Ω -type doubling or simply Ω -doubling between states of opposite parity, once small rotation-induced level interactions are included [16–18]. This in turn implies that these triatomic molecules can be polarized with static electric fields by the linear Stark effect. In heavy molecules, the Ω -doubling states have the potential to greatly increase the sensitivity of experiments searching for electric dipole moments (EDMs) [19].

In this paper, we focus on a promising alternative, the $^{163}\text{Dy}^{107}\text{Ag}$ molecule formed in overlapping samples of laser-cooled dysprosium and silver atoms. Silver atoms were cooled to temperatures of hundreds of microkelvin in the year 2000 [20] and, more recently, in Ref. [21], while samples of bosonic dysprosium isotopes were brought to quantum degeneracy a little over a decade ago [22,23]. Extremely weakly bound ultracold $^{163}\text{Dy}_2$ dimers have been formed

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using magnetic Fano-Feshbach resonances [24,25]. Stable bosonic dysprosium isotopes have no nuclear spin. Even though the ^{163}Dy isotope is a fermion and has a half-integer nuclear spin, no roadblocks are expected in laser cooling this isotope as those methods do not (significantly) rely on the statistics of the particles.

Co-trapped, ultracold samples, i.e., trapping of both atomic clouds in the same spatial region, are a starting point from which to assemble DyAg molecules without unwanted heating. Developed over the past 20 years, we can rely on methods by the atomic physics community for forming bi-alkali-metal molecules [26,27] as well as on protocols for state preparation of rotational and hyperfine levels. See review articles in Ref. [28]. Based on the existing experience, 10^4 optically trapped ^{163}Dy ^{107}Ag molecules can be expected. The samples do not need to be near quantum degeneracy for these purposes.

Following the proposal in Ref. [29] using the radioactive FrAg molecule, the silver atom is chosen as it has a large electron affinity of $hc \times 10\,521\text{ cm}^{-1}$ [30], where h is the Planck constant and c is the speed of light in vacuum. In the DyAg molecular bond, as we will show, the dysprosium atom then loses an electron to Ag. The bond is thus ionic. This leads to a gradient in the electron charge density at the position of the Dy nucleus, which increases the sensitivity to parity-breaking interactions. In fact, weak symmetry-breaking electron-nucleon interactions and small nuclear anapole moments manifest themselves as parity-breaking coupling $W_{\text{br}}(R)\hat{\mathbf{R}} \cdot \mathbf{i}_{\text{Dy}}/\hbar$ between the orientation of the internuclear axis, described by unit vector $\hat{\mathbf{R}}$, and the nuclear spin $\mathbf{i}_{\text{Dy}}$ of ^{163}Dy [31–33]. Its strength $W_{\text{br}}(R)$ depends on atom-atom separation R , and $\hbar$ is the reduced Planck constant.

Even though the value for $W_{\text{br}}(R)$ is currently unknown and its evaluation is not the purpose of this article, we can derive an estimate based on literature values for related molecules. We observe that $W_{\text{br}}(R) = W_{\text{S}}(R)S_{\text{lab}}$, where the nuclear Schiff moment S_{lab} in the laboratory frame is $0.36\bar{\theta}\text{ e}(\text{fm})^3$ for ^{161}Dy [34], similar in size to many other heavy nuclei. Here, e is the elementary charge, and the degree of nuclear parity and time-reversal symmetry breaking is encapsulated in the dimensionless quantum chromodynamics (QCD) coupling constant $\bar{\theta}$. The upper bound of the absolute value of $\bar{\theta}$ is 2.4×10^{-10} from Refs. [35,36]. Electronic molecular effects are isolated in $W_{\text{S}}(R)$ with $|W_{\text{S}}(R)|$ at most a few times $10^4 E_{\text{h}}/(ea_0^3)$ at the equilibrium separation of various heavy diatomic molecules [13]. Finally, E_{h} is the Hartree energy and a_0 is the Bohr radius. Combining these data, we expect that $|W_{\text{br}}(R)|/\hbar < 100\text{ }\mu\text{Hz}$ at the equilibrium separation of DyAg.

The electronic eigenstates of DyAg in a body-fixed coordinate system that follows along with the rotating linear molecule have quantum number Ω of the projection of the total electronic angular momentum along the internuclear axis as a good quantum number, just like the linear triatomic hydroxide molecules introduced above. Hence, the DyAg electronic potentials are anisotropic and thus depend on vector $\hat{\mathbf{R}}$. For $\Omega > 0$, the electronic potentials for $-\Omega$ and Ω are degenerate. Coriolis forces lift this degeneracy leading to Ω doubling with two states of opposite parity. In this article, we quantify the size of Ω doubling for $^{163}\text{DyAg}$ in its energetically lowest vibrational levels for various Ω states.

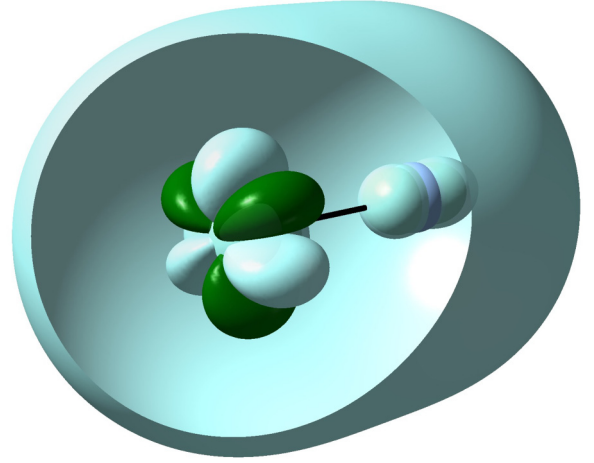


FIG. 1. Isosurfaces of electron orbitals of DyAg at its equilibrium separation obtained within density-functional theory. The alternating cyan and green lobes represents one of the dysprosium 4f orbitals, while the small dumbbell-shaped object to its right represents a 4d orbital of Ag. The 4f and 4d orbitals lie well inside the large hybridized 5s silver and 6s dysprosium orbital, shown with a cutout in order to see the compact internal dysprosium and silver orbitals.

This paper has two distinct components. After a simplified introduction of the molecular electronic wave function in Sec. II, the determination of the relevant adiabatic relativistic electronic potentials of the dimer is described in detail in Sec. II A. We focus on the energetically lowest electronic potentials dissociating to Dy and Ag atoms. In their electronic ground state, these atoms are described by configurations $4f^{10}(^5\text{I}_8)6s^2$ and $4d^{10}5s(^2\text{S})$, respectively. Secondly, we perform quantum-mechanical coupled-channels calculations to determine the energetically lowest rovibrational levels of ^{163}Dy ^{107}Ag , focusing on Ω -doubling in Sec. II B, initially ignoring hyperfine interactions. Section II C describes the effects of hyperfine and Zeeman interactions on Ω -type doubling observing that ^{163}Dy has a large electronic angular momentum from its aligned 4f electrons and a nuclear spin of $5\hbar/2$. The ^{107}Ag isotope has nuclear spin $\hbar/2$. We conclude in Sec. III. Values of fundamental constants in SI units have been taken from Refs. [37,38]. Atomic masses in atomic mass units m_{u} are found in Ref. [39].

II. RESULTS

We start by describing the electronic ground state of DyAg at its equilibrium geometry based on scalar-relativistic spin-unrestricted density-functional theory within GAUSSIAN [40,41]. We have used the B3PW91 functional, def2-QZVP electronic basis sets for the Dy and Ag atoms, and corresponding effective core potentials describing the 28 core electrons of each atom. Figure 1 shows isosurfaces of electronic molecular orbitals in DyAg at its equilibrium separation of $5.46a_0$ with the silver core and dysprosium $4f^9$ orbitals completely embedded inside the hybridized $5s^2-6s^2$ orbitals. This unusual bond, where the inner 4f electron orbital rather than the outer 6s orbital loses an electron to Ag, has also been observed for DyF and DyO [12–14] as well as DyOH [15].

The density-functional simulation with the B3PW91 functional also shows that the permanent molecular electronic dipole moment $\mathbf{d}$ is large at $1.54ea_0$ along the internuclear axis. In fact, we have evaluated the permanent dipole moment within density functional theory using various functionals and basis sets and recommend the value

$$d_e = 1.5(3)ea_0, \quad (1)$$

where the number in parentheses represents the standard-deviation uncertainty. Our value is in line with that for YbAg in Ref. [42].

Our value for the permanent dipole moment can be partially explained by transfer of Dy electrons towards the Ag atom. In fact, a natural-bond-order calculation within density-functional theory shows that the Dy atom loses approximately 0.68 electrons from its open 4f shell, rather than its closed 6s² shell, to the Ag atom. The “partial” electron is accepted into the 5s shell of Ag. In any case, the bond is strongly ionic and, consequently, there exists a nonzero gradient in the electron charge density at the nuclear positions, which enhances the effects of parity-breaking molecular interactions.

A. Configuration-interaction calculations of the electronic structure of DyAg

The density-functional calculations are not sufficiently accurate for our purposes. Hence, we perform “nonrelativistic” multireference configuration-interaction (CI) calculations implemented in MOLPRO [41,43] with single and double excitations, including the Davidson correction, coupled with calculations of spin-orbit matrix elements using the Breit-Pauli Hamiltonian between the ground and several excited electronic eigenstates. We use the cc-pvtz-DK3 and Sapporo-DKH3-TZVP Douglass-Kroll scalar-relativistic atomic orbitals for the valence electrons of Dy and Ag, respectively. The many-electron basis functions for the “nonrelativistic” CI calculations are obtained from multireference self-consistent-field calculations with active 6s and 4f orbitals for Dy and the 5s orbital for Ag.

The term “nonrelativistic” is in quotes to indicate that, although the CI calculations include scalar relativistic effects, they do not couple an electron’s spin to its momentum or to other electron orbital angular momenta. We will drop the quotes from hereon.

We determine nonrelativistic eigenpairs with a total electron spin quantum number $S = 5/2$ for each of the four irreducible representations of the C_{2v} point group [44] on a grid of internuclear separations R between $3.25a_0$ and $20a_0$. These states are spin-stretched states as the atomic electron spins of the nonrelativistic ground states of neutral Dy and Ag are 2 and 1/2, respectively. In fact, we compute two eigenstates for the A_2 representation of C_{2v} and three for the remaining A_1 , B_1 , and B_2 representations. The reasons for these choices are discussed in Appendix.

Inspection of the 11 nonrelativistic eigenstates near the equilibrium separation shows that they predominantly contain the ionic configuration $\text{Dy}^+[4f^9(^6H)6s^2] - \text{Ag}^-[4d^{10}5s^2(^1S)]$, where the Dy atom has lost a 4f electron to Ag, just as in the density-functional calculation.

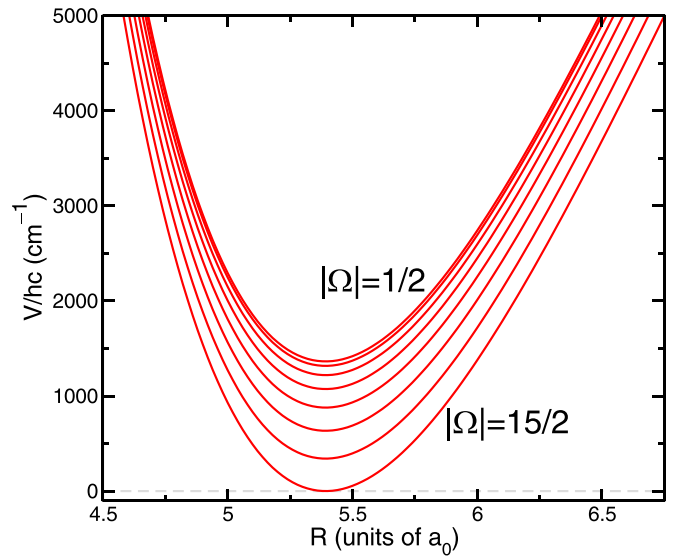


FIG. 2. Relativistic DyAg potentials $V(\Omega; R)$ as functions of separation R based on configuration-interaction calculations belonging to the ground electronic state manifold. Shown are eight closely related potentials corresponding to the ground state of each $|\Omega| = 15/2, 13/2, 11/2, 9/2, 7/2, 5/2, 3/2$, and $1/2$ with increasing potential energy. The zero of energy is located at the equilibrium separation of the $|\Omega| = 15/2$ state.

Relativistic electronic eigenenergies at each R are found by computing all spin-orbit matrix elements among the nonrelativistic eigenstates and diagonalizing a small matrix eigenvalue system for each value of projection Ω of the combined spin and orbital electronic angular momentum along the internuclear axis. The matrix has nonrelativistic eigenenergies as diagonal matrix elements, coupled and shifted by spin-orbit matrix elements. In fact, given the ionic configuration $\text{Dy}^+[4f^9(^6H)6s^2] - \text{Ag}^-[4d^{10}5s^2(^1S)]$ we have a six-dimensional matrix for $|\Omega| = 1/2$ decreasing to a one-dimensional matrix for the largest $|\Omega| = 15/2$.

Figure 2 shows potential energies $V(\Omega; R)$ of our relativistic calculations as functions of internuclear separation for the lowest-energy eigenstate for each absolute value of the projection quantum number $|\Omega|$ between 1/2 and 15/2. Excited electronic states for these Ω values as well as ground and excited states with $|\Omega| > 15/2$ have energies that fall well outside the energy region shown in the figure. We observe that the absolute ground state has quantum number $|\Omega| = 15/2$ and the system has inverted fine structure. Within the accuracy of our simulations, the eight potentials have the same equilibrium separation of $R_e = 5.39a_0$, slightly smaller but more accurate than that obtained with the density-functional calculations, and a spring constant of $hc \times 9500(100) \text{ cm}^{-1}a_0^{-2}$ with the standard uncertainty derived from the spread of spring constants from the adiabatic Ω states. Not shown in Fig. 2 is that these eight potentials dissociate to neutral ground-state Dy and Ag atoms. At about $hc \times 30\,000 \text{ cm}^{-1}$, the DyAg potentials are significantly deeper than those for, say, KRb, RbCs, and Cs₂ [26,45–47].

A consequence of the observations regarding Fig. 2 is that the lowest-energy, relativistic electronic eigenfunctions can be

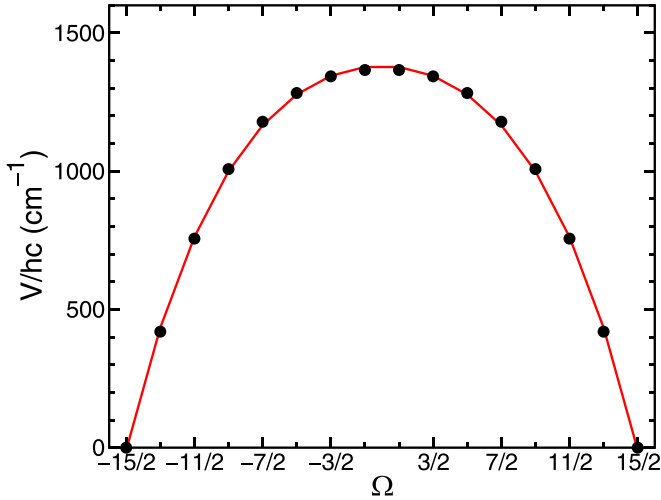


FIG. 3. The relativistic electronic potentials $V(\Omega; R)$ of DyAg at $R = 5.5a_0$, near the equilibrium separation, as a function of projection quantum number Ω . The red curve is a fit to the crystal-field or spin-tensor operator discussed in the text. The energies are relative to the deepest potential. For diatomic molecules, electronic states $-\Omega$ and Ω are degenerate.

described by a positively charged $4f^9 6s^2$ dysprosium ion with electronic angular momentum quantum number $j_{\text{Dy}^+} = 15/2$ and a negatively charged $4d^{10} 5s^2$ silver ion with $j_{\text{Ag}^-} = 0$.

Figure 3 shows the potential energies of the relativistic ground states at separation $5.5a_0$, close to the equilibrium geometry, as a function of Ω . The energies form a “flattened” parabola. More precisely, the Ω and R dependence of the potential energies near the equilibrium separation is well described by the crystal-field operators [18]

$$\begin{aligned} \hat{V}(\mathbf{R}) = & V_0(R) + V_2 T_2(\mathbf{j}_{\text{Dy}^+}, \mathbf{j}_{\text{Dy}^+}) \cdot C_2(\hat{\mathbf{R}}) / \hbar^2 \\ & + V_4 T_4(T_2(\mathbf{j}_{\text{Dy}^+}, \mathbf{j}_{\text{Dy}^+}), T_2(\mathbf{j}_{\text{Dy}^+}, \mathbf{j}_{\text{Dy}^+})) \cdot C_4(\hat{\mathbf{R}}) / \hbar^4, \end{aligned} \quad (2)$$

where $T_{kq}(R_{k_1}, S_{k_2})$ are rank- k spherical tensor operators formed from a rank k_1 and a rank k_2 spherical tensor operator following the notation of Ref. [48]. Moreover, the $C_{kq}(\hat{\mathbf{R}})$ are rank- k spherical harmonic functions with $C_{kq}(\hat{\mathbf{0}}) = \delta_{q0}$, recalling that unit vector $\hat{\mathbf{R}} = (\theta, \varphi)$, specified by polar and azimuthal angles θ and φ , respectively, describes the orientation of the internuclear axis. Function δ_{ij} is the Kronecker delta.

The eigenvalues of operator $\hat{V}(\mathbf{R})$ when choosing a body-fixed coordinate system with the z axis along the internuclear axis, i.e., $\hat{\mathbf{R}} = \hat{\mathbf{0}}$, correspond to the curves in Fig. 2. For $R \in [R_e - 0.5a_0, R_e + 0.5a_0]$, coefficient $V_0(R)$ is a parabola as a function of R , while the R dependence of coefficients V_2 and V_4 can be neglected. The fitted values for V_i are $V_0(5.5a_0)/\hbar c = 920 \text{ cm}^{-1}$, $V_2/\hbar c = -20 \text{ cm}^{-1}$, and $V_4/\hbar c = -0.078 \text{ cm}^{-1}$, respectively.

B. Omega-doubling in $^{163}\text{Dy}^{107}\text{Ag}$

Ω -type doubling, as stated in the Introduction, is the process whereby the degeneracy of electronic states with $-\Omega$ and

Ω with $\Omega > 0$ is lifted by the rotation of the molecule. The resulting nondegenerate pair of states will have opposite parity and, thus, can be relevant for precision measurements searching for small parity violations. The heteronuclear $^{163}\text{Dy}^{107}\text{Ag}$ molecule has multiple states with nonzero projections of the total electron angular momentum along the internuclear axis.

To describe the rotating and vibrating $^{163}\text{Dy}^{107}\text{Ag}$ dimer, we assume a positively charged $^{163}\text{Dy}^+(4f^9 6s^2)$ ion with electron angular momentum $j_{\text{Dy}^+} = 15/2$, following our observations in the previous section, and nuclear spin $i_{\text{Dy}^+} = 5/2$. Similarly, we assume a negatively charged $\text{Ag}^-(5s^2(^1\text{S}))$ with electron angular momentum $j_{\text{Ag}^-} = 0$ and nuclear spin $i_{\text{Ag}^-} = 1/2$. Absent hyperfine interactions, the Hamiltonian of the dimer then contains the sum of a radial kinetic energy operator $-\hbar^2/(2\mu)d^2/dR^2$ with reduced mass μ derived from the masses of the neutral atoms [49], the rotational interaction $V_{\text{rot}} = \ell^2/(2\mu R^2)$, where ℓ is the relative mechanical angular momentum, and the potential operator in Eq. (2) with the R -dependence of $V_0(R)$ derived from fitting to the (anharmonic) data in Fig. 2 and $V_4 = 0$.

We discretize the radial kinetic energy on a uniform spatial grid R_i with $i = 1, \dots, N_r - 1$ around the equilibrium separation using the discrete variable representation with zero-valued wave functions at separations R_0 and R_{N_r} [50]. The total angular momentum $\mathbf{J}_{\text{tot}} = \mathbf{j}_{\text{Dy}^+} + \mathbf{j}_{\text{Ag}^-} + \ell$ and parity $p = p_{\text{Dy}^+} p_{\text{Ag}^-} (-1)^\ell$ are conserved quantities. Here, parity $p_{\text{Dy}^+} = -1$ for our excited ionic Dy^+ state and $p_{\text{Ag}^-} = +1$ for the closed-shell Ag^- . That is, the matrix elements of the discretized Hamiltonian are determined in the coupled basis $|R_i, (j_{\text{Dy}^+} \ell) J_{\text{tot}} M_{\text{tot}}\rangle$, and the eigenpairs of the matrix are numerically calculated with a linear algebra solver on matrices of dimension $(N_r - 1) \times N_s$, where N_s is the finite number of $|(j_{\text{Dy}^+} \ell) J_{\text{tot}} M_{\text{tot}}\rangle$ spin states for a given J_{tot} and parity. We have omitted $j_{\text{Ag}^-} = 0$ in the ket for simplicity and note that the molecular Hamiltonian is independent of the space-fixed projection quantum number M_{tot} of $\mathbf{J}_{\text{tot}}$. For DyAg omitting the nuclear spins, we have $N_s \leq 8$ and use $N_r \approx 70$ to converge the energies of the lowest 20 vibrational levels.

The energetically lowest vibrational eigenenergies for total molecular angular momentum quantum number $J_{\text{tot}} = 15/2$ and odd parity $p = -1$ are shown in Fig. 4. Each level is labeled by approximate quantum number $|\Omega| = 1/2$ to $15/2$ and vibrational level $v = 0, 1, 2, \dots$, where the $v = 0$ level with projection $\Omega = 15/2$ is the absolute ground state of the molecule. We observe that the spacing between vibrational levels is on the order of the spacing between the different Ω potentials, making a complex, interleaved level spectrum. In fact, the harmonic vibrational energy of $^{163}\text{Dy}^{107}\text{Ag}$ is about $\hbar c \times 130 \text{ cm}^{-1}$ with equilibrium rotational constant $B_e \equiv \hbar^2/(2\mu R_e^2) = \hbar c \times 0.032 \text{ cm}^{-1} = h \times 0.96 \text{ GHz}$.

In Fig. 5(a), we focus on the lowest vibrational level for $|\Omega| = 1/2$ and show both even- and odd-parity states as a function of J_{tot} . For each J_{tot} , we have one even- and one odd-parity state, where the energy ordering alternates with increasing J_{tot} . Moreover, state $J_{\text{tot}} = 7/2$, $p = -1$ has the lowest energy of all $|\Omega| = 1/2$ levels. The alternating even-odd parity splitting is due to the Coriolis coupling $2(\mathbf{J}_{\text{tot}} \cdot \mathbf{j}_{\text{Dy}^+})/(2\mu R^2)$ originating from the rotational interaction V_{rot} after inserting $\ell = \mathbf{J}_{\text{tot}} - \mathbf{j}_{\text{Dy}^+}$ and expanding term by

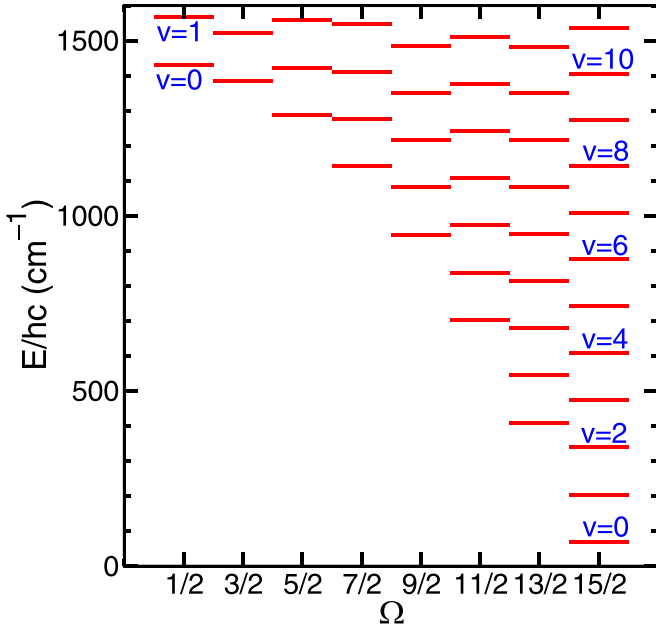


FIG. 4. Vibrational levels of $^{163}\text{Dy } ^{107}\text{Ag}$ near the minima of the electronic potentials with $J_{\text{tot}} = 15/2$ and odd parity as functions of approximate projection quantum number Ω . For each Ω , we count vibrational levels $v = 0, 1, \dots$ with increasing energy. The zero of energy is at the minimum of the $|\Omega| = 15/2$ potential energy curve.

term. See, for example, Refs. [16–18,51]. In fact, degenerate first-order perturbation theory in V_{rot} for $|\Omega| = 1/2$ leads to rotational energies

$$B_e \left[J_{\text{tot}}(J_{\text{tot}} + 1) + \epsilon \left(J_{\text{tot}} + \frac{1}{2} \right) \left(j_{\text{Dy}^+} + \frac{1}{2} \right) \right] \quad (3)$$

with $\epsilon = p(-1)^{J_{\text{tot}} - j_{\text{Dy}^+}}$, a generalization of spin-doubling in $^2\Sigma$ molecules [51]. Hence, the even-odd parity level splittings are almost $h \times 10$ GHz. Curves from Eq. (3) are also shown in Fig. 5(a).

Figures 5(b) and 5(c) show the differences or splittings between the energies of the even- and odd-parity states of the $v = 0$ level of $|\Omega| = 3/2$ and $5/2$ states, respectively, as a function of J_{tot} . The sign of the splittings again alternates, but their absolute values are now very small compared to the rotational constant B_e . We have splittings on the order of $h \times 10$ MHz and $h \times 1$ kHz for $|\Omega| = 3/2$ and $5/2$, respectively. For $|\Omega| > 1/2$, the Coriolis operator no longer couples states $-\Omega$ and Ω in first-order perturbation theory. Higher-order perturbation theory results in splittings between even- and odd-parity states of order

$$\left(\frac{B_e J_{\text{tot}}}{V_2} \right)^{|\Omega| - 1/2} B_e J_{\text{tot}} \equiv \Delta_e \quad (4)$$

for angular momentum $J_{\text{tot}} \gg |\Omega|$, where the negative crystal-field coefficient V_2 is defined in Eq. (2) and thus $B_e/V_2 \approx -2 \times 10^{-3}$. Clearly, for $|\Omega| > 5/2$, the splittings will be of order or less than $h \times 1$ Hz, while simultaneously the J_{tot} dependence is more rapid. It is worth noting that Eq. (4) only describes the dominant exponential dependence on Ω . It misses a nontrivial polynomial-dependent prefactor.

We end this subsection with estimates of the size of the Stark effect for vibrational levels of the DyAg molecule when

a constant electric field $\mathbf{E}$ is applied. The Stark Hamiltonian $-\mathbf{d} \cdot \mathbf{E}$ mixes the Ω -doubled states. Then the electric field strength at which the two interactions are comparable, Δ_e/d_e , is several kV/cm for $|\Omega| = 1/2$ and a few V/cm for $|\Omega| = 3/2$ when $J_{\text{tot}} = |\Omega|$. See also the derivation of Lambda-doubling for OH in Ref. [52] and observing that the atomic unit of electric field is $E_h/(ea_0) \approx 5.14 \times 10^{11}$ V/m. We have omitted the small effects of the R dependence of the permanent dipole moment.

C. Hyperfine and Zeeman interactions

We now turn our attention to hyperfine and Zeeman interactions due to external magnetic fields, with the goal to bring states with different molecular angular momentum and parity to exact degeneracy. The hyperfine and Zeeman interactions between the nuclear spins and electronic angular momenta of the ionically bonded $^{163}\text{Dy } ^{107}\text{Ag}$ near their equilibrium separation are unknown. We add the hyperfine interactions [53,54]

$$\begin{aligned} H_{\text{hfs}} = & a_{\text{Dy}} (\mathbf{j}_{\text{Dy}^+} \cdot \mathbf{i}_{\text{Dy}}) / \hbar^2 \\ & + \frac{6}{4} b_{\text{Dy}} \frac{T_2(\mathbf{j}_{\text{Dy}^+}, \mathbf{j}_{\text{Dy}^+}) \cdot T_2(\mathbf{i}_{\text{Dy}}, \mathbf{i}_{\text{Dy}}) / \hbar^4}{j_{\text{Dy}^+}(2j_{\text{Dy}^+} - 1) i_{\text{Dy}}(2i_{\text{Dy}} - 1)} \\ & + g_{\text{Dy}} \mu_B (\mathbf{j}_{\text{Dy}^+} \cdot \mathbf{B}) / \hbar \end{aligned} \quad (5)$$

to the Hamiltonian described in the previous section, where energies a_{Dy} and b_{Dy} are the molecular magnetic dipole and electric quadrupole constants, respectively, g_{Dy} is a molecular electronic g -factor, μ_B is the Bohr magneton, and $\mathbf{B}$ is a homogeneous, constant external magnetic field pointing along the z axis of our space-fixed coordinate system. The constants a_{Dy} , b_{Dy} , and g_{Dy} are assumed to be independent of R near the equilibrium separation. We do not include hyperfine terms that depend on orientation $\hat{\mathbf{R}}$ and neglect couplings between the nuclear spin of Ag and other angular momenta. Specifically, we can neglect atomic silver hyperfine interactions because the Ag ion in the molecule has closed electron shells. Finally, the weak nuclear Zeeman interactions for the nuclei have been omitted.

The atomic quadrupole term on the second line of Eq. (5) and thus the definition of constant b_{Dy} are equivalent to the “standard form” given in, for example, Ref. [53]. Our symmetric form in $\mathbf{j}_{\text{Dy}^+}$ and $\mathbf{i}_{\text{Dy}}$ is more practical for numerical implementation using the Wigner-Eckart theorem [48].

We have chosen $a_{\text{Dy}}/\hbar = 162.754$ MHz, $b_{\text{Dy}}/\hbar = 1152.869$ MHz, and $g_{\text{Dy}} = +4/3$ corresponding to the hyperfine constants of neutral ^{163}Dy in its $4f^{10}(^5\text{I}_8)6s^2$ ground state from Ref. [55] and a g -factor based on the Landé expression for term $^6\text{H}_{15/2}$ [56]. As far as we are aware, the hyperfine constants of the excited ion $^{163}\text{Dy}^+[4f^9(^6\text{H}_{15/2})6s^2]$ are not available from the literature. Here, we have followed the observations by Ref. [14] on the hyperfine structure of dimer DyO. We believe that this introduces a factor of 2 uncertainty in our estimates of molecular hyperfine splittings. The effects of molecular bonding on constants a_{Dy} , b_{Dy} , and g_{Dy} are expected to be small compared to our uncertainties in these three quantities.

Figures 6(a)–6(c) show hyperfine and parity eigenenergies of the $v = 0$ level of $\Omega = 1/2, 3/2$, and $5/2$, respectively, absent a magnetic field as functions of quantum number F_{tot}

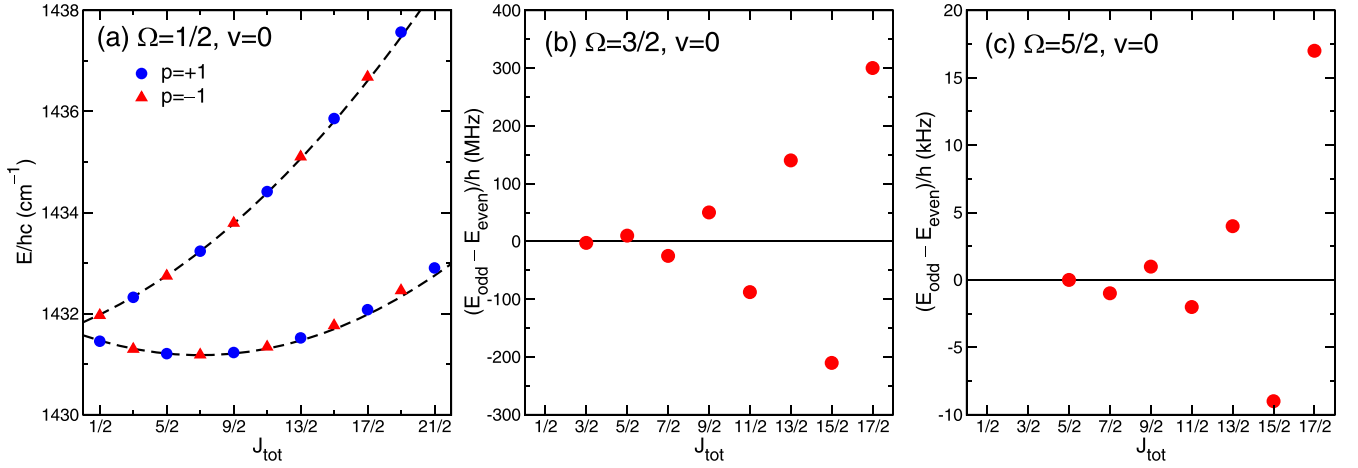


FIG. 5. Panel (a) shows energies of even-parity, $p = 1$ (blue filled circles), and odd-parity, $p = -1$ (red filled circles), states for the $v = 0$ vibrational level of the $\Omega = 1/2$ state of $^{163}\text{Dy}^{107}\text{Ag}$ as functions of angular momentum J_{tot} absent hyperfine interactions. The dashed curves are the predictions of Eq. (3) from degenerate first-order perturbation theory in the rotational Hamiltonian. The zero of energy is at the minimum of the $|\Omega| = 15/2$ potential energy curve. Panels (b) and (c) show energy differences $E_{\text{odd}} - E_{\text{even}}$ between odd- and even-parity states for the $v = 0$ vibrational level of the $\Omega = 3/2$ and $5/2$ states of $^{163}\text{Dy}^{107}\text{Ag}$, respectively, as functions of J_{tot} absent hyperfine interactions. Notice the different energy scales along the y axes of the three panels.

of the conserved molecular angular momentum $\mathbf{F}_{\text{tot}} = \mathbf{j}_{\text{Dy}^+} + \boldsymbol{\ell} + \mathbf{i}_{\text{Dy}} \equiv \mathbf{f}_{\text{Dy}} + \boldsymbol{\ell} \equiv \mathbf{J}_{\text{tot}} + \mathbf{i}_{\text{Dy}}$. We can omit the nuclear spin of the silver nucleus in constructing $\mathbf{F}_{\text{tot}}$ as we do not include hyperfine or Zeeman interactions that depend on $\mathbf{i}_{\text{Ag}}$. The coupled-channels calculations have been performed in the basis $|R_i, (j_{\text{Dy}^+} + i_{\text{Dy}})f_{\text{Dy}}, \ell; F_{\text{tot}}M_{F,\text{tot}}\rangle$. Without a magnetic field, the eigenenergies are independent of projection quantum number $M_{F,\text{tot}}$.

Figure 6(a) shows that the hyperfine splittings for $\Omega = 1/2$ have a similar magnitude to those due to Ω -doubling and rotation, reflecting the fact that $B_e \sim a_{\text{Dy}}, b_{\text{Dy}}$. Nevertheless, the two groups or bands of levels are recognizable in the figure. In fact, we observe that when $F_{\text{tot}} \gg i_{\text{Dy}}$, “rows” of $2i_{\text{Dy}} + 1 = 6$

nearly degenerate levels with the same parity appear. Each row can be assigned an approximate value J_{tot} from a comparison with the data in Fig. 6(a). Not clearly visible in Fig. 6(a) is that the $F_{\text{tot}} = 1$, $p = -1$ state has the lowest energy of all hyperfine states of level $v = 0$, $|\Omega| = 1/2$.

For $\Omega > 1/2$ in Figs. 6(b) and 6(c), the splittings from the Ω -doubling process are small compared to those from the hyperfine interactions and are consistent with the splittings shown in Figs. 5(b) and 5(c). The “rows” with nearly degenerate energies on the scale of the panels correspond to states with the same approximate J_{tot} . Equivalently, a rotational progression, with linearly increasing level splittings, can be observed along the vertical direction. Levels with $F_{\text{tot}} = 1$ and

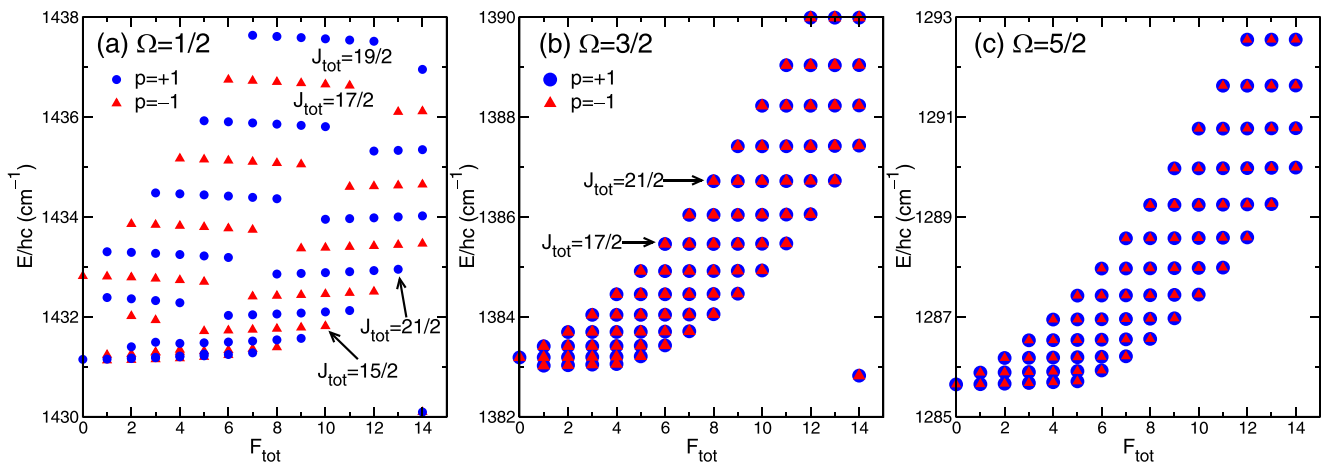


FIG. 6. Panel (a) shows energies of even-parity, $p = 1$ (blue filled circles), and odd-parity, $p = -1$ (red filled circles), states for the $v = 0$ vibrational level of the $\Omega = 1/2$ state of $^{163}\text{Dy}^{107}\text{Ag}$ including hyperfine interactions as function of quantum number F_{tot} . For some “rows” of states with large F_{tot} , the approximate value for J_{tot} is indicated. Panels (b) and (c) show energies of even-parity (blue circles) and odd-parity (smaller red filled circles) levels as functions of F_{tot} for the $v = 0$ vibrational level of the $\Omega = 3/2$ and $5/2$ states, respectively. The splittings between the even- and odd-parity levels are of order $h \times 10$ MHz in panel (b) and on the order of $h \times 1$ kHz in panel (c), invisible on the vertical scale of these panels. The zero of energy in all panels is at the minimum of the $|\Omega| = 15/2$ potential energy curve. In the lower right-hand side of panels (a) and (b), a single marker is visible. They are hyperfine states of a separate v , Ω pair.

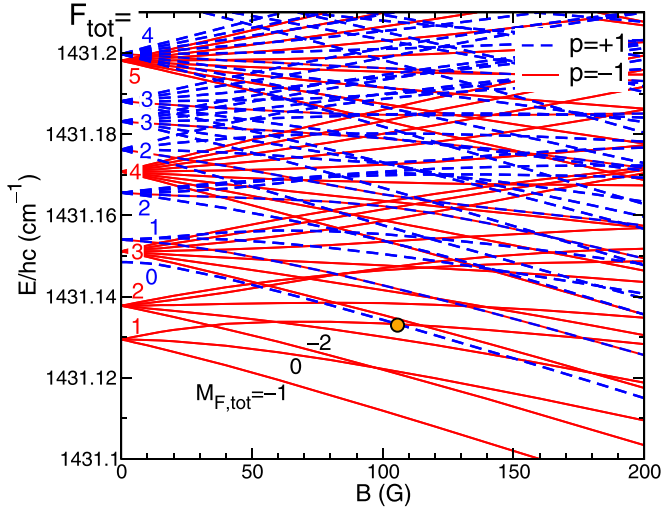


FIG. 7. Zeeman and hyperfine energies of the lowest vibrational level $v = 0$ with $\Omega = 1/2$ of $^{163}\text{Dy } ^{107}\text{Ag}$ as functions of magnetic field strength B . Blue and red curves correspond to even- and odd-parity states, respectively. Levels are labeled by molecular angular momentum quantum numbers F_{tot} and $M_{F,\text{tot}}$. We only draw states with $|M_{F,\text{tot}}| \leq 4$. The orange filled circle near $B = 100$ G corresponds to a crossing of opposite-parity states with $|\Delta M_{F,\text{tot}}| = 1$. The zero of energy is at the minimum of the $|\Omega| = 15/2$ potential energy curve.

0 are the energetically lowest hyperfine levels for state $v = 0$, $|\Omega| = 1/2$ and $5/2$, respectively.

We can now apply a magnetic field to the rotating $^{163}\text{Dy } ^{107}\text{Ag}$ molecule. Figure 7 shows the energetically lowest hyperfine levels of state $v = 0$, $|\Omega| = 1/2$ over an $hc \times 0.1 \text{ cm}^{-1}$ interval as functions of B for easily achievable field strengths up to 200 G. Here, 1 G is 10^{-4} T. For $B > 0$ G, only projection $M_{F,\text{tot}}$ and parity p are good quantum numbers. Both even- and odd-parity states are shown, and data are limited to $|M_{F,\text{tot}}| \leq 4$ to reduce clutter. For small field strengths, states can still be labeled by zero-field quantum number F_{tot} , and their energies split linearly as $g_{\text{mol}}\mu_B M_{F,\text{tot}} B$ with *positive* F_{tot} -dependent molecular g -factor g_{mol} . That is, for each F_{tot} the energetically lowest Zeeman level has $M_{F,\text{tot}} = -F_{\text{tot}}$. Note that for even parity, the energy order with respect to F_{tot} is not monotonically increasing, a consequence of the level order of the lowest branch of states in Fig. 5(a) or in Fig. 6(a).

Precision experiments require states with opposite parity to be degenerate absent parity violating interactions. Many such cases are visible in Fig. 7. One case is shown in the figure satisfying the additional constraint that the difference $\Delta M_{F,\text{tot}}$ in projection quantum number $M_{F,\text{tot}}$ is no more than ± 1 unit, consistent with the selection rules for transitions induced by electromagnetic radiation. Finally, we stress that our choices for the magnetic dipole and electric quadrupole constants are only estimates, and that the actual level ordering and spacings might change.

Finally, Fig. 8 shows the energetically lowest hyperfine levels of state $F_{\text{tot}} = 1$, $v = 0$, $|\Omega| = 3/2$, over an $\approx hc \times 0.01 \text{ cm}^{-1}$ interval as functions of B for fields up to 20 G. The splitting at $B = 0$ between the two $F_{\text{tot}} = 1$ parity states is

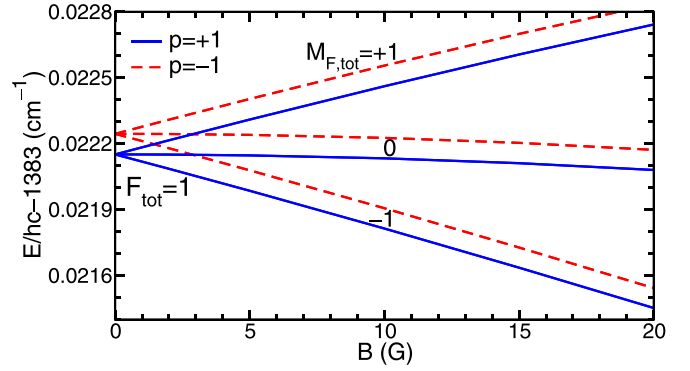


FIG. 8. Zeeman energies of the lowest energy $F_{\text{tot}} = 1$, $v = 0$ level with $\Omega = 3/2$ of $^{163}\text{Dy } ^{107}\text{Ag}$ as functions of magnetic field strength B . Blue and red curves correspond to even- and odd-parity states, respectively. The states are labeled with projection quantum number $M_{F,\text{tot}}$. The zero of energy is at the minimum of the $|\Omega| = 15/2$ potential energy curve.

$\approx h \times 40 \text{ MHz}$, consistent with the small Ω -doubling splitting for $|\Omega| = 3/2$ in Fig. 5(b). A crossing between two opposite-parity states with $|\Delta M_{F,\text{tot}}| = 1$ is visible at 3 G, a much smaller field than for $\Omega = 1/2$ states.

III. CONCLUSION

Advances in building stable lasers in the ultraviolet regime have made it possible to cool atoms from a larger part of the Periodic Table and in some instances put them together to form ultracold molecules. Isotopes of both dysprosium and silver have been cooled by laser light, but have not yet been bound together. In anticipation of the formation of ground-state $^{163}\text{Dy } ^{107}\text{Ag}$ molecules, we have studied the eight relevant lowest-energy electronic states as well as the hyperfine and Zeeman structure of rovibrational levels in these coupled electronic states. In addition, we obtained values for the splittings due to Coriolis-induced Ω -type doubling as a function of the electronic projection quantum number Ω on the internuclear axis.

The study of Ω doubling has been the main driver behind our research. The two “doubled” states have opposite parity and are nearly degenerate for $|\Omega| > 1/2$ and thus can be used to search for parity-breaking interactions in the octupole-deformed ^{163}Dy nucleus. In a molecule and in its simplest form, parity-breaking interactions lead to coupling between the nonzero nuclear spin of ^{163}Dy and the rotation of the molecule. In addition, we showed that magnetic fields can be used to bring states of opposite parity into exact degeneracy with modest magnetic field strengths, making it, in principle, easier to detect weak parity-breaking interactions.

For our proposal to be corroborated, other investigations will be required. This includes finding ways to compute excited electronic states and electric transition dipole moments with electronic structure codes in order to characterize the two-photon Raman transitions that are needed to form molecules from laser-cooled Dy and Ag atoms. In addition, precision experiments rely not only on magnetic fields to shift energy levels but also parity-breaking electric fields. By studying changes in energy shifts when the direction of the

electric field is reversed, unwanted systematic effects in the experiments can be eliminated. Hence, we must be able to add the Stark Hamiltonian to our already complex molecular Hamiltonian. We hope to contribute to some of these aspects in the future.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX: DETAILS ABOUT THE ELECTRONIC CONFIGURATION-INTERACTION CALCULATIONS

This appendix describes the “nonrelativistic” electronic configuration-interaction (CI) calculations in more detail. Specifically, we explain the limits set by our computational resources. We have calculated eigenpairs with a total electron spin quantum number $S = 5/2$ for each of the four irreducible representations of the C_{2v} point group. In fact, we have computed two nonrelativistic eigenstates for the A_2 representation and three for the A_1 , B_1 , and B_2 representations.

For each of the four irreducible representations with spin $S = 5/2$, the dimension of the nonrelativistic CI eigenvalue system using contracted many-electron basis functions is of order 10^7 . The energetically lowest eigenpairs within

MOLPRO are found using iterative algorithms. Attempts to determine more eigenvalues for each representation than stated in the previous paragraph lead to impractical run times. CI calculations with $S < 5/2$ could not be completed as matrix dimensions became prohibitively large. Calculations with $S < 5/2$, however, will only lead to high-energy eigenstates.

The origin of these computational limitations follows from an analysis of the nonrelativistic eigenstates. First, we observe that in diatomic molecules, nonrelativistic eigenstates have projection Λ of the total electron orbital angular momentum along the internuclear axis as a good quantum number, and that states with $-\Lambda$ and Λ when $\Lambda > 0$ are degenerate. Eigenstates in representation A_1 have $|\Lambda|^\sigma = 0^+, 2, 4, \dots$. Representation A_2 contains states with $|\Lambda| = 0^-, 2, 4, \dots$, while representations B_1 and B_2 contain states with $|\Lambda| = 1, 3, 5, \dots$. The $\pm$ superscript for $|\Lambda| = 0$ corresponds to even/odd reflection symmetry of the electronic wave function through a plane containing the internuclear axis.

As stated in the main text, the 11 nonrelativistic eigenstates near the equilibrium separation are well described by the ionic configuration $\text{Dy}^+[4f^9(^6\text{H})6s^2] - \text{Ag}^-[4d^{10}5s^2(^1\text{S})]$, where the Dy atom has lost a 4f electron to Ag. In other words, the negatively charged silver ion only has closed-shell configurations and is spinless, while the positively charged dysprosium configuration has electron spin $5/2$ and electron orbital angular momentum quantum number 5. This molecular configuration corresponds to an excited state of the isolated atomic Dy^+ ion and has $|\Lambda|$ between 0 and 5. State counting and molecular reflection symmetries of this ionic configuration imply three, two, three, and three eigenstates for representations A_1 , A_2 , B_1 , and B_2 , respectively. Crucially, the $|\Lambda| = 0^-$ state does not exist. Hence, the iterative eigenvalue solver used by the CI program is only numerically stable when requesting exactly those numbers of eigenvalues.

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