

# Relativistic multireference coupled-cluster theory based on a $B$ -spline basis: Application to atomic francium

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In this paper, we report the relativistic Fock space multireference coupled-cluster method for atomic structure calculations. We use the no-pair Dirac-Coulomb-Breit Hamiltonian, together with a finite  $B$ -spline basis set to expand the large and small components of the Dirac wave function. Our method is applied to calculate ionization energies, reduced matrix elements, lifetimes, and polarizabilities for many states of atomic francium. To evaluate uncertainties of our results and investigate the role of electron correlation effects, we carry out calculations using approximated models at different levels. The quality of our calculations is assessed by comparing with available experimental results, showing a good agreement. In addition, the tune-out wavelengths of the ground state in the range of 340–800 nm, the magic wavelengths for the transition  $7s$ - $8s$  in the range of 800–1500 nm and the transition  $7s$ - $7p$  in the range of 600–1500 nm are determined by evaluating the dynamic polarizabilities of the  $7s$ ,  $8s$ , and  $7p$  states for a linearly polarized light. These tune-out and magic wavelengths may be useful for laser cooling and trapping of the Fr atom, and for related high-precision trapping measurements.

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## I. INTRODUCTION

Accurate knowledge of atomic properties for heavy atomic systems has become increasingly demanding in many research areas, including ultraprecise atomic clocks [1,2], quantum information [3,4], determination of nuclear moments [5–7], search for new physics beyond the standard model [8], and testing of the Lorentz invariance [9]. For example, the uncertainty in blackbody radiation shift, one of the largest contributions to the uncertainty budget of atomic clocks [1], is currently limited by the uncertainty in atomic transition matrix elements. The magnetic dipole moment  $\mu$  and electric quadrupole moment  $Q$  of a nucleus can be derived from the hyperfine constants  $A$  and  $B$  by combining theoretical calculations and experimental measurements of sufficiently high accuracy [5–7]. High-precision theoretical and experimental studies of parity nonconservation can provide an atomic-physics-based test of the standard model of electroweak interaction [8]. However, accurate theoretical predictions of atomic properties for heavy atoms need to include high-order relativistic and electron-electron correlation effects. It is therefore necessary to develop a many-body method that can simultaneously treat relativistic and electron correlation effects on the same footing.

Coupled-cluster (CC) theory is an all-order, size-extensive, systematic, and very accurate many-body approach [10]. In the past several decades, several kinds of coupled-cluster methods have been developed and adapted widely into electronic structure calculations [11]. In the CC domain, the method of Fock-space multireference coupled-cluster (FSMRCC) is one of the most familiar variants. The relativistic FSMRCC in Gaussian basis sets has been developed by Kaldor and coworkers and applied to calculations of energy levels of atomic and molecular systems [12–14], such as the calcula-

tions of ionization potentials, electron affinities, and excitation energies in heavy and superheavy elements [15–19].

In atomic physics, one is not only interested in energy levels, but also interested in transition matrix elements, lifetimes, polarizabilities, hyperfine structure constants, and other atomic properties, involving low-lying and highly excited Rydberg states. For this purpose, we present here a relativistic Fock-space multireference coupled-cluster method for atomic systems. We use the no-pair Dirac-Coulomb-Breit Hamiltonian and expand the large and small components of one-electron Dirac wave function in terms of a finite  $B$ -spline basis set. The advantage of using  $B$  splines as basis functions is that they are not only complete, but also numerically stable; furthermore, they are highly localized, flexible to construct, and capable of describing highly excited Rydberg states [20]. A relativistic version of linear coupled-cluster method in  $B$ -spline basis set was developed by Johnson *et al.* [21,22], and applied successfully to study the atomic properties of monovalent atomic systems [23–25]. Recently, Safronova *et al.* [26] and Dzuba [27] extended this method to multiple-valent atomic systems, in combination with the configuration interaction method. In these approximations, the contributions from the Breit interaction are treated using low-order many-body perturbation theory, and the effects from nonlinear terms and higher-order correlation are considered by a semiempirical scaling process [28]. The present method has important differences from their methods. For example, the Dirac-Fock-Breit equation is solved using a completely self-consistent iteration method based on  $B$ -spline basis set, and all matrix elements are evaluated by using Gaussian-Legendre quadratures. The single and double excitations of cluster operators are considered completely in our approach, which includes all linear and nonlinear terms. The nonlinear terms of cluster operators can be important for heavy atoms [29,30]. We are also aware of other relativistic versions of coupled-cluster methods for atomic systems using Gaussian basis [31–35].

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As a test of our method, we calculate atomic properties of Fr, including ionization energies, reduced matrix elements, lifetimes, and static polarizabilities involving many atomic states. Fr is the heaviest atom in alkali-metal elements, which has been proposed as a candidate for the search of the permanent electric dipole moment of the electron (e-EDM) and of parity-nonconservation (PNC) effects [36–40]. Accurate knowledge of Fr atomic properties are very useful for designing experiments and for analyzing systematic uncertainties. We also present here the tune-out wavelengths of the ground state and the magic wavelengths for the  $7s$ - $7p$  and  $7s$ - $8s$  transitions by evaluating the dynamic polarizabilities of the  $7s$ ,  $7p$ , and  $8s$  states. The tune-out and magic wavelengths may be used for laser cooling and trapping Fr atom, as well as for related precision trapping measurements.

This paper is organized as follows. An introduction to the Dirac-Fock-Breit calculation using  $B$ -spline basis set in the framework of the no-pair Dirac-Coulomb-Breit Hamiltonian, as well as to the basic theory of Fock space multireference coupled-cluster method, is given in Sec. II. Numerical results are presented in Sec. III, together with comparisons with available experimental and theoretical data. Finally, a summary is given in Sec. IV. Atomic units are used in this paper.

## II. THEORETICAL METHOD

The no-pair Dirac-Coulomb-Breit Hamiltonian can be written in the form

$$H = \sum_i \{c\alpha_i \cdot \mathbf{p}_i + c^2(\beta_i - 1) + V_N(\mathbf{r}_i)\} + \Lambda_{++} \sum_{i < j} g_{ij} \Lambda_{++}, \quad (1)$$

where  $\Lambda_{++}$  is the projection operator onto the subspace spanned by the positive energy eigenfunctions,  $c$  is the speed of light,  $\mathbf{p}_i$  is the momentum operator for  $i$ th electron, and,  $\alpha_i$  and  $\beta_i$  are the usual Dirac matrices. Also in the above,  $V_N(\mathbf{r}_i)$  is the Coulomb potential between  $i$ th electron and nucleus, and the last part  $g_{ij}$  represents two-body potential including the electron-electron Coulomb interaction and Breit interaction, namely

$$g_{ij} = \left[ \frac{1}{r_{ij}} - \frac{\alpha_i \cdot \alpha_j + (\alpha_i \cdot \hat{\mathbf{r}}_{ij})(\alpha_j \cdot \hat{\mathbf{r}}_{ij})}{2r_{ij}} \right]. \quad (2)$$

The consideration of the Dirac-Coulomb-Breit Hamiltonian without quantum electrodynamic (QED) corrections is sufficient for the present paper. All effects through order  $\alpha^2$  are included in the zeroth-order Hamiltonian.

It is well known that the exact wave function and total energy of a many-electron atomic system can not be obtained directly because of the electron-electron interaction potential. In general, the Hamiltonian is divided into the zeroth-order part  $H_0$ , where the eigenfunctions of  $H_0$  can be solved precisely, and the residual part  $V$  (or perturbation part). In the present paper,  $H_0$  is chosen to be the Dirac-Fock-Breit Hamiltonian, and  $V$  is incorporated through the coupled-cluster theory. Thus, the first step is to perform a Dirac-Fock-Breit calculation using  $B$ -spline basis set. By doing so one can generate the energies and wave functions of single-particle orbitals. Then

the coupled equations of the single- and double-excitation cluster amplitudes can be solved iteratively.

### A. Dirac-Fock-Breit calculations in $B$ -spline basis

The zeroth-order approximation  $H_0$  is chosen to be

$$H_0 = \sum_i [c\alpha_i \cdot \mathbf{p}_i + c^2(\beta_i - 1) + V_N(\mathbf{r}_i) + U(\mathbf{r}_i)] = \sum_i h_0. \quad (3)$$

The eigenfunctions of  $H_0$  are the determinantal functions based on the occupied one-electron eigenfunctions  $|\varphi_v\rangle$  of  $h_0$ , where  $|\varphi_v\rangle$  and the corresponding eigenvalue  $\epsilon$  satisfy the following equation:

$$[c\boldsymbol{\alpha} \cdot \mathbf{p} + c^2(\beta - 1) + V_N(\mathbf{r}) + U(\mathbf{r})]|\varphi_v\rangle = \epsilon|\varphi_v\rangle \quad (4)$$

with

$$U|\varphi_v\rangle = \sum_b^{\text{occ}} [\langle \varphi_b | g_{vb} | \varphi_b \rangle |\varphi_v\rangle + \langle \varphi_b | g_{vb} | \varphi_v \rangle |\varphi_b\rangle]. \quad (5)$$

$|\varphi_v\rangle$  can be written as

$$|\varphi_v\rangle = \frac{1}{r} \begin{pmatrix} P_{n\kappa}(r)\Omega_{\kappa m}(\hat{\mathbf{r}}) \\ i Q_{n\kappa}(r)\Omega_{-\kappa m}(\hat{\mathbf{r}}) \end{pmatrix}, \quad (6)$$

where the radial wave functions  $P_{n\kappa}(r)$  and  $Q_{n\kappa}(r)$  are the large and small components, respectively, and  $\Omega_{\kappa m}(\hat{\mathbf{r}})$  and  $\Omega_{-\kappa m}(\hat{\mathbf{r}})$  are the corresponding angular components. The large and small components are expanded in terms of an  $N$ -dimensional  $B$ -spline basis of order  $k$

$$P_{n\kappa}(r) = \sum_{i=1}^N C_i^{P,n} B_{i,k}(r), \quad (7)$$

$$Q_{n\kappa}(r) = \sum_{i=1}^N C_i^{Q,n} B_{i,k}(r). \quad (8)$$

The  $B$  splines in the domain  $[0, R_{\max}]$  are defined on a nondecreasing knot sequence  $[t_i]$  satisfying an exponential distribution [20]. The order index  $k$  of a  $B$  spline will be omitted in following expressions.

Using the Galerkin method with the relativistic wave function boundary conditions proposed by Johnson *et al.* [21], a  $2N \times 2N$  generalized symmetric eigenvalue equation can be obtained

$$A\mathbf{v} = \epsilon O\mathbf{v}. \quad (9)$$

The Hamiltonian matrix  $A$  is

$$A = \mathbf{A}_{\text{DC}} + \begin{pmatrix} \mathbf{B}^{QQ} & \mathbf{B}^{QP} \\ \mathbf{B}^{PQ} & \mathbf{B}^{PP} \end{pmatrix}, \quad (10)$$

and the overlap matrix  $O$  is

$$O = \begin{pmatrix} \mathbf{S}^{PP} & 0 \\ 0 & \mathbf{S}^{QQ} \end{pmatrix}. \quad (11)$$

Here  $\mathbf{A}_{\text{DC}}$  corresponds to the matrix of the Dirac-Fock equation in the no-pair Dirac-Coulomb Hamiltonian. The explicit expressions for  $\mathbf{A}_{\text{DC}}$ ,  $\mathbf{S}^{PP}$ , and  $\mathbf{S}^{QQ}$  can be found in our previous work [41].  $\mathbf{B}^{QQ}$ ,  $\mathbf{B}^{QP}$ ,  $\mathbf{B}^{PQ}$ , and  $\mathbf{B}^{PP}$  are from the Breit interaction, the explicit expressions are given in the Appendix.

All matrix elements are evaluated using Gaussian-Legendre quadratures, and the single-particle orbitals and energies are obtained by solving the generalized eigenvalue equation with a completely self-consistent iteration method.

The method of  $B$ -spline basis was first adopted to solve the Dirac and Dirac-Fock (DF) equations by Johnson *et al.* [21], which is called the Notre Dame (ND) method. In the ND method, the DF equation is solved using a point-by-point numerical integration scheme, and the matrix  $A_{DC}$  is constructed by combining the obtained core DF orbitals with  $B$ -spline basis set. We note that there exist other types of  $B$ -spline basis sets satisfying more complex conditions in order to avoid spurious-state problem [42–47]. For example, Shabaev *et al.* introduced a dual kinetic balance basis using  $B$  splines for solving the Dirac equation with a finite nuclear-charge distribution [42]. Fischer and Zatsarinny proposed a stable ( $B^k, B^{k\pm 1}$ ) basis for the Dirac equation [47]. Among all these schemes, the ND one is the simplest form.

In the present paper, we expand the large and small components of the wave functions using 50  $B$  splines of order 13 defined on the finite domain [0,280]. We find that the calculations do not suffer from the spurious-state problem when we use  $B$  splines of order 13 or higher.

### B. Multireference coupled-cluster method

According to Lindgren [10], the exact wave function  $|\Psi\rangle$  for a system can be generated by a normal-ordered wave operator  $\Omega$  acting on a model function

$$|\Psi\rangle = \Omega|\Phi\rangle, \quad (12)$$

where

$$\Omega = [e^S], \quad (13)$$

and  $S$  is called the cluster operator, which is defined with respect to a closed-shell reference determinant. In the framework of multireference coupled-cluster method [12–14], the cluster operator  $S$  is partitioned into

$$S = \sum_{m \geq 0} \sum_{n \geq 0} \left( \sum_{\ell \geq m+n} S_\ell^{(m,n)} \right), \quad (14)$$

where  $m$  and  $n$  are, respectively, the number of valence holes and the number of valence particles to be excited with respect to the reference determinant, and  $\ell$  is the number of excited electrons. The coupled equations for the cluster operators are derived from the generalized Bloch equation by considering only the connected terms

$$Q[S_\ell^{(m,n)}, H_0]P = Q[(V\Omega - \Omega H_{\text{eff}})_\ell^{(m,n)}]_{\text{conn}} P \quad (15)$$

$$H_{\text{eff}} = P(V\Omega)_{\text{conn}}^{(m,n)} P, \quad (16)$$

where  $P$  and  $Q$  are the usual projection operators, which act on the model space and its orthogonal complement, respectively, and  $H_{\text{eff}}$  represents the effective Hamiltonian.

For atomic systems, spherical symmetry allows for the separation of angular and radial parts. Using angular diagram theory, the angular part can further be simplified to some factors containing the  $3j$  and  $6j$  symbols. Thus, we only need to evaluate the radial integrals. In practice, the equations for

the sector  $S^{(0,0)}$  are first solved iteratively until a convergence is reached. The sector  $S^{(0,1)}$  or  $S^{(1,0)}$  is then solved using the known  $S^{(0,0)}$ , and so on. In this paper, we use the  $(m,n) = (0,0)$  and  $(0,1)$ , and  $\ell$  is truncated to 2, which corresponds to single and double excitations (CCSD). The cluster operators  $S^{(0,0)}$  and  $S^{(0,1)}$  are similar to the  $T$  and  $S$  operators in the traditional coupled-cluster method [29,30].

### C. Transition matrix elements

After obtaining the cluster amplitudes, the transition matrix element of operator  $\hat{O}$  can be evaluated according to

$$\begin{aligned} O_{wv} &= \frac{\langle \Psi_w | O | \Psi_v \rangle}{\sqrt{\langle \Psi_w | \Psi_w \rangle \langle \Psi_v | \Psi_v \rangle}} \\ &= \frac{\langle \Phi_w | e^{S^\dagger} O e^S | \Phi_v \rangle}{\sqrt{\langle \Phi_w | e^{S^\dagger} e^S | \Phi_w \rangle \langle \Phi_v | e^{S^\dagger} e^S | \Phi_v \rangle}}, \end{aligned} \quad (17)$$

where  $|\Psi_w\rangle$  represents a state with a closed core and a valence orbital  $w$ , and  $|\Phi_w\rangle = a_w^\dagger |\Phi\rangle$ . In calculating atomic properties, we only consider the linearized expansion of the coupled-cluster wave functions with the single and double excitations (LCCSD)

$$\begin{aligned} e^{S^\dagger} O e^S &\approx O + [S^{(0,0)\dagger} O + \text{H.c.}] + [S^{(0,1)\dagger} O + \text{H.c.}] \\ &\quad + S^{(0,0)\dagger} O S^{(0,0)} + [S^{(0,0)\dagger} O S^{(0,1)} + \text{H.c.}] \\ &\quad + S^{(0,1)\dagger} O S^{(0,1)} \end{aligned} \quad (18)$$

and

$$e^{S^\dagger} e^S \approx 1 + S^{(0,0)\dagger} S^{(0,0)} + S^{(0,1)\dagger} S^{(0,1)}, \quad (19)$$

where H.c. stands for the Hermitian conjugate part.

## III. RESULTS

### A. Energies and reduced matrix elements

The energy levels for the  $ns$  ( $n = 7-13$ ),  $np$  ( $n = 7-13$ ),  $nd$  ( $n = 6-12$ ),  $nf$  ( $n = 5-9$ ), and  $ng$  ( $n = 5-8$ ) states of Fr are calculated using relativistic coupled-cluster method in single and double approximation. To understand the role of electron correlation effects, we present the energies of these states in Table I using the DF (labeled  $E_{\text{DF}}$ ), LCCSD (labeled  $E_{\text{LCCSD}}$ ), and CCSD (labeled  $E_{\text{CCSD}}$ ) methods, and compared with available experimental values from NIST [48].  $\delta$  represents the relative difference between calculated and experimental values  $\delta_1 = |E_{\text{Expt}} - E_{\text{LCCSD}}|/E_{\text{Expt}}$ , and  $\delta_2 = |E_{\text{Expt}} - E_{\text{CCSD}}|/E_{\text{Expt}}$ .

From Table I, one can find that: (i) The electron correlation effects of the  $6d$  states are the largest among all states. (ii) The DF energies of the  $d_{5/2}$  states are lower than  $d_{3/2}$ , but the LCCSD and CCSD energies of  $d_{3/2}$  are lower than  $d_{5/2}$ . (iii) The LCCSD method overestimates the electron correlation effects. The contributions from the nonlinear terms of the cluster operator have opposite sign with respect to the LCCSD values. (iv) The energies and fine structure energies from the CCSD method are in better agreement with available experimental values than from the LCCSD method for all states; in other words, the CCSD method improves substantially the LCCSD method. Therefore, the nonlinear terms are important in precise prediction of the

TABLE I. Theoretical and experimental energy levels of Fr in  $\text{cm}^{-1}$ .  $E_{\text{DF}}$  denotes the lowest-order Dirac-Fock energy;  $E_{\text{LCCSD}}$  and  $E_{\text{CCSD}}$  are the energies obtained using the LCCSD and CCSD approximations, respectively.  $\delta_1 = |E_{\text{Expt}} - E_{\text{LCCSD}}|/E_{\text{Expt}}$ , and  $\delta_2 = |E_{\text{Expt}} - E_{\text{CCSD}}|/E_{\text{Expt}}$ . Values marked by *a* are our recommended values that can not be found from NIST database.

Levels	$E_{\text{DF}}$	$E_{\text{LCCSD}}$	$E_{\text{CCSD}}$	NIST [48]	$\delta_1(\%)$	$\delta_2(\%)$
$7s_{1/2}$	-28761.22	-33339.50	-32908.18	-32848.87	1.5	0.18
$7p_{1/2}$	-18840.94	-20800.19	-20603.78	-20611.46	0.9	0.03
$7p_{3/2}$	-17650.88	-19067.97	-18923.02	-18924.87	0.8	0.01
$6d_{3/2}$	-13837.93	-17293.25	-16628.45	-16619.00	4.1	0.05
$6d_{5/2}$	-13938.52	-16991.53	-16423.95	-16419.23	3.5	0.03
$8s_{1/2}$	-12279.50	-13187.18	-13125.19	-13108.89	0.6	0.13
$8p_{1/2}$	-9235.22	-9777.02	-9731.64	-9735.91	0.4	0.04
$8p_{3/2}$	-8809.57	-9225.50	-9188.72	-9190.56	0.4	0.02
$7d_{3/2}$	-7730.28	-8706.81	-8603.36	-8604.84	1.2	0.02
$7d_{5/2}$	-7752.98	-8597.42	-8511.93	-8515.94	1.0	0.05
$9s_{1/2}$	-6857.11	-7202.08	-7183.27	-7177.87	0.3	0.08
$5f_{7/2}$	-6867.13	-6969.24	-6960.38	-6960(4) <sup>a</sup>		
$5f_{5/2}$	-6866.22	-6968.35	-6959.47	-6959(4) <sup>a</sup>		
$9p_{1/2}$	-5518.28	-5753.03	-5734.63	-5730.66	0.4	0.07
$9p_{3/2}$	-5317.75	-5502.33	-5486.92	-5482.67	0.4	0.08
$8d_{3/2}$	-4859.89	-5279.00	-5245.87	-5248.21	0.6	0.04
$8d_{5/2}$	-4865.73	-5227.67	-5200.41	-5203.50	0.5	0.06
$10s_{1/2}$	-4380.15	-4549.49	-4540.86	-4538.25	0.2	0.06
$6f_{7/2}$	-4396.66	-4457.31	-4451.78	-4452(3) <sup>a</sup>		
$6f_{5/2}$	-4395.91	-4456.59	-4451.03	-4451(3) <sup>a</sup>		
$5g_{7/2}$	-4389.51	-4402.47	-4401.44	-4401(1) <sup>a</sup>		
$5g_{9/2}$	-4389.51	-4402.47	-4401.44	-4401(1) <sup>a</sup>		
$10p_{1/2}$	-3673.79	-3797.79	-3787.31	-3784.69	0.3	0.07
$10p_{3/2}$	-3563.59	-3662.42	-3653.23	-3650.78	0.3	0.07
$9d_{3/2}$	-3325.07	-3545.38	-3530.75	-3532.37	0.4	0.05
$9d_{5/2}$	-3326.74	-3517.18	-3505.12	-3507.05	0.3	0.05
$7f_{7/2}$	-3053.39	-3091.20	-3087.66	-3088(2) <sup>a</sup>		
$7f_{5/2}$	-3052.87	-3090.69	-3087.14	-3087(2) <sup>a</sup>		
$6g_{7/2}$	-3048.28	-3056.68	-3056.01	-3056(1) <sup>a</sup>		
$6g_{9/2}$	-3048.28	-3056.68	-3056.01	-3056(1) <sup>a</sup>		
$11s_{1/2}$	-3040.22	-3136.15	-3131.47	-3129.96	0.2	0.05
$11p_{1/2}$	-2622.38	-2696.05	-2690.05	-2687.80	0.3	0.08
$11p_{3/2}$	-2555.43	-2614.64	-2609.42	-2607.27	0.3	0.08
$10d_{3/2}$	-2414.87	-2545.52	-2537.78	-2538.91	0.3	0.04
$10d_{5/2}$	-2415.30	-2528.36	-2521.96	-2523.27	0.2	0.05
$12s_{1/2}$	-2233.49	-2293.20	-2290.38	-2289.37	0.2	0.04
$8f_{7/2}$	-2243.18	-2268.01	-2265.65	-2265(2) <sup>a</sup>		
$8f_{5/2}$	-2242.84	-2267.68	-2265.31	-2265(2) <sup>a</sup>		
$7g_{7/2}$	-2239.55	-2245.18	-2244.73	-2245(1) <sup>a</sup>		
$7g_{9/2}$	-2239.55	-2245.18	-2244.73	-2245(1) <sup>a</sup>		
$12p_{1/2}$	-1965.84	-2013.21	-2009.75	-2007.89	0.3	0.09
$12p_{3/2}$	-1922.19	-1960.45	-1957.45	-1955.71	0.2	0.09
$11d_{3/2}$	-1832.32	-1916.25	-1911.64	-1912.55	0.2	0.05
$11d_{5/2}$	-1832.32	-1905.01	-1901.21	-1902.23	0.1	0.05
$13s_{1/2}$	-1710.14	-1749.95	-1748.12	-1747.33	0.1	0.05
$9f_{7/2}$	-1717.27	-1734.35	-1732.72	-1733(1) <sup>a</sup>		
$9f_{5/2}$	-1717.12	-1734.18	-1732.54	-1733(1) <sup>a</sup>		
$8g_{7/2}$	-1714.66	-1718.58	-1718.26	-1718(1) <sup>a</sup>		
$8g_{9/2}$	-1714.64	-1718.56	-1718.24	-1718(1) <sup>a</sup>		
$13p_{1/2}$	-1528.22	-1560.49	-1558.14	-1557.15	0.2	0.06
$13p_{3/2}$	-1498.28	-1524.37	-1522.33	-1521.41	0.2	0.06
$12d_{3/2}$	-1437.44	-1494.48	-1491.49	-1492.37	0.1	0.06
$12d_{5/2}$	-1437.25	-1486.69	-1484.25	-1485.22	0.1	0.06

energy levels. The biggest difference between the CCSD results and experimental values is 0.18%. Most of the CCSD ionization energies agree with experiment values within 0.1%. If the contributions from QED effects, triple and higher-order excitation corrections are included, the differences may be further reduced. For example, Dzuba *et al.* estimated the QED contribution of the  $7s$  state as  $77\text{cm}^{-1}$  [49]. If we add this QED value to our CCSD result of the  $7s$  state, the difference 0.05% instead of 0.18%. In NIST database [48], we do not see data for  $f$  and  $g$  states. We include in Table I our recommended values for these states as labeled by index *a*.

The electric and magnetic multipole transition matrix elements among above-mentioned states can be calculated using our code in Eq. (18) and Eq. (19). Here we just list some interesting reduced dipolar matrix elements, which are relevant to the lifetimes and polarizabilities. The final recommended values (absolute values) of reduced electric dipolar matrix elements are given in Table II. To estimate the uncertainties of our results, we perform the calculations using four different approximation models: LCCSD, CCSD, and LCCSD and CCSD with semiempirical scaling process [28] (labeled by LCCSD<sub>s</sub> and CCSD<sub>s</sub>). In the LCCSD<sub>s</sub> and CCSD<sub>s</sub> calculations, the single-excitation coefficients of the valence electron are replaced by new single-excitation coefficients that are equal to the old single-excitation coefficients multiplied by the factor  $\delta(E_v^{\text{Expt}})/\delta(E_v^{\text{Theory}})$ . Here  $\delta(E_v^{\text{Expt}})$  represents the correlation energy of the valence electron  $v$ , i.e.,  $\delta(E_v^{\text{Expt}}) = E_v^{\text{Expt}} - E_v^{\text{DF}}$ .

In the present work, we take the LCCSD results as the recommended values for  $s$ - $p$  transitions and take the CCSD results as the recommended values for other transitions. The uncertainties are evaluated as the maximum differences between the recommended values and the results from LCCSD<sub>s</sub> or CCSD<sub>s</sub>. For all transitions, the CCSD values agree with the CCSD<sub>s</sub> ones at the level of 2%. For most of the transitions, the results from the CCSD, LCCSD<sub>s</sub>, and CCSD<sub>s</sub> are in agreement within 3%. For the transitions  $8p$ - $6d$  and  $nf$ - $(n+1)d$ , however, the LCCSD<sub>s</sub> values have larger uncertainties. More detailed tabulation for the electric dipolar matrix elements using the DF method and the four approximation models are given in the Supplemental Material [50].

## B. Lifetimes

The lifetime of a state  $|\Psi_v\rangle$  is defined by

$$\tau_v = \sum_w \frac{1}{A_{vw}}, \quad (20)$$

where  $A_{vw}$  is the transition rate and the summation includes all possible electromagnetic transitions from  $|\Psi_v\rangle$ . For the states considered here, the contributions from other electromagnetic multipolar transitions, except the electric dipole transitions, are very small and they can thus be neglected. The electric dipole transition rate  $A_{vw}$  is evaluated according to

$$A_{vw} = \frac{2.02615 \times 10^{18}}{\lambda^3(2j_v + 1)} S_{wv} \quad (21)$$

where  $\lambda$  represents the transition wavelength in  $\text{\AA}$  and  $S_{wv}$  is the line strength that equals the square of electric dipolar reduced matrix element.

TABLE II. Recommended values of the reduced matrix elements (denoted by  $D$ ) for the electric dipole transitions in Fr. Uncertainties are given in parentheses.

Transitions	$D$	Transitions	$D$	Transitions	$D$	Transitions	$D$	Transitions	$D$
$7s_{1/2}-7p_{1/2}$	4.27(7)	$7s_{1/2}-7p_{3/2}$	5.87(10)	$9s_{1/2}-9p_{1/2}$	17.40(5)	$8d_{5/2}-12p_{3/2}$	2.07(3)	$8f_{5/2}-9d_{3/2}$	3.02(46)
$7s_{1/2}-8p_{1/2}$	0.327(20)	$7s_{1/2}-8p_{3/2}$	0.930(21)	$9s_{1/2}-9p_{3/2}$	22.65(4)	$8d_{5/2}-13p_{3/2}$	1.37(2)	$8f_{5/2}-10d_{3/2}$	94.07(19)
$7s_{1/2}-9p_{1/2}$	0.110(6)	$7s_{1/2}-9p_{3/2}$	0.434(7)	$9s_{1/2}-10p_{1/2}$	1.83(29)	$9d_{3/2}-11p_{1/2}$	17.64(3)	$8f_{5/2}-11d_{3/2}$	54.06(28)
$7s_{1/2}-10p_{1/2}$	0.0554(40)	$7s_{1/2}-10p_{3/2}$	0.269(7)	$9s_{1/2}-10p_{3/2}$	3.87(3)	$9d_{3/2}-12p_{1/2}$	5.29(6)	$8f_{5/2}-12d_{3/2}$	11.59(6)
$7s_{1/2}-11p_{1/2}$	0.0340(38)	$7s_{1/2}-11p_{3/2}$	0.190(6)	$10s_{1/2}-9p_{1/2}$	16.09(13)	$9d_{3/2}-13p_{1/2}$	2.81(2)	$8f_{5/2}-8d_{5/2}$	1.03(3)
$7s_{1/2}-12p_{1/2}$	0.0234(30)	$7s_{1/2}-12p_{3/2}$	0.144(5)	$10s_{1/2}-9p_{3/2}$	27.00(18)	$9d_{5/2}-11p_{3/2}$	18.37(9)	$8f_{5/2}-9d_{5/2}$	1.17(10)
$7s_{1/2}-13p_{1/2}$	0.0173(19)	$7s_{1/2}-13p_{3/2}$	0.115(3)	$10s_{1/2}-10p_{1/2}$	26.61(1)	$9d_{5/2}-12p_{3/2}$	5.98(7)	$8f_{5/2}-10d_{5/2}$	25.47(3)
$8s_{1/2}-8p_{1/2}$	10.03(7)	$8s_{1/2}-8p_{3/2}$	13.26(10)	$10s_{1/2}-10p_{3/2}$	34.30(1)	$9d_{5/2}-13p_{3/2}$	3.24(3)	$8f_{5/2}-11d_{5/2}$	13.83(7)
$8s_{1/2}-9p_{1/2}$	0.988(10)	$8s_{1/2}-9p_{3/2}$	2.241(10)	$11s_{1/2}-9p_{1/2}$	3.12(2)	$10d_{3/2}-12p_{1/2}$	25.49(30)	$8f_{5/2}-12d_{5/2}$	3.08(1)
$8s_{1/2}-10p_{1/2}$	0.382(7)	$8s_{1/2}-10p_{3/2}$	1.049(6)	$11s_{1/2}-9p_{3/2}$	3.80(5)	$10d_{3/2}-13p_{1/2}$	7.58(9)	$8f_{7/2}-6d_{5/2}$	2.22(3)
$8s_{1/2}-11p_{1/2}$	0.214(6)	$8s_{1/2}-11p_{3/2}$	0.652(6)	$11s_{1/2}-10p_{1/2}$	24.60(24)	$10d_{5/2}-12p_{3/2}$	26.63(50)	$8f_{7/2}-7d_{5/2}$	3.24(7)
$8s_{1/2}-12p_{1/2}$	0.142(4)	$8s_{1/2}-12p_{3/2}$	0.462(4)	$11s_{1/2}-10p_{3/2}$	40.88(19)	$10d_{5/2}-13p_{3/2}$	8.60(9)	$8f_{7/2}-8d_{5/2}$	4.61(16)
$8s_{1/2}-13p_{1/2}$	0.104(2)	$8s_{1/2}-13p_{3/2}$	0.352(2)	$11s_{1/2}-11p_{1/2}$	37.66(3)	$11d_{3/2}-13p_{1/2}$	34.68(19)	$8f_{7/2}-9d_{5/2}$	5.27(44)
$7p_{1/2}-8s_{1/2}$	4.20(4)	$8p_{1/2}-8s_{1/2}$	10.03(6)	$11s_{1/2}-11p_{3/2}$	48.23(8)	$11d_{5/2}-13p_{3/2}$	36.30(40)	$8f_{7/2}-10d_{5/2}$	113.9(2)
$7p_{1/2}-9s_{1/2}$	1.018(6)	$8p_{1/2}-9s_{1/2}$	9.30(5)	$12s_{1/2}-9p_{1/2}$	1.543(12)	$5f_{5/2}-6d_{3/2}$	7.08(60)	$8f_{7/2}-11d_{5/2}$	61.79(31)
$7p_{1/2}-10s_{1/2}$	0.544(5)	$8p_{1/2}-10s_{1/2}$	1.941(6)	$12s_{1/2}-9p_{3/2}$	1.780(21)	$5f_{5/2}-7d_{3/2}$	24.82(4)	$8f_{7/2}-12d_{5/2}$	13.78(6)
$7p_{1/2}-11s_{1/2}$	0.361(3)	$8p_{1/2}-11s_{1/2}$	0.990(5)	$12s_{1/2}-10p_{1/2}$	4.56(4)	$5f_{5/2}-6d_{5/2}$	1.97(14)	$9f_{5/2}-6d_{3/2}$	1.41(2)
$7p_{1/2}-12s_{1/2}$	0.266(3)	$8p_{1/2}-12s_{1/2}$	0.645(4)	$12s_{1/2}-10p_{3/2}$	5.40(8)	$5f_{5/2}-7d_{5/2}$	6.674(4)	$9f_{5/2}-7d_{3/2}$	1.93(4)
$7p_{1/2}-13s_{1/2}$	0.207(3)	$8p_{1/2}-13s_{1/2}$	0.470(2)	$13s_{1/2}-9p_{1/2}$	0.990(17)	$5f_{7/2}-6d_{5/2}$	8.83(63)	$9f_{5/2}-8d_{3/2}$	2.59(7)
$7p_{1/2}-6d_{3/2}$	7.15(9)	$8p_{1/2}-6d_{3/2}$	2.25(45)	$13s_{1/2}-9p_{3/2}$	1.120(13)	$5f_{7/2}-7d_{3/2}$	29.84(2)	$9f_{5/2}-9d_{3/2}$	3.18(17)
$7p_{1/2}-7d_{3/2}$	3.53(6)	$8p_{1/2}-7d_{3/2}$	18.29(5)	$6d_{3/2}-9p_{1/2}$	0.725(24)	$5f_{5/2}-8d_{3/2}$	11.59(11)	$9f_{5/2}-10d_{3/2}$	1.77(50)
$7p_{1/2}-8d_{3/2}$	1.839(27)	$8p_{1/2}-8d_{3/2}$	5.035(9)	$6d_{3/2}-10p_{1/2}$	0.401(5)	$6f_{5/2}-6d_{3/2}$	3.79(7)	$9f_{5/2}-11d_{3/2}$	124.6(3)
$7p_{1/2}-9d_{3/2}$	1.192(15)	$8p_{1/2}-9d_{3/2}$	2.663(15)	$6d_{3/2}-11p_{1/2}$	0.269(2)	$6f_{5/2}-7d_{3/2}$	5.24(50)	$9f_{5/2}-12d_{3/2}$	73.60(27)
$7p_{1/2}-10d_{3/2}$	0.862(9)	$8p_{1/2}-10d_{3/2}$	1.745(9)	$6d_{3/2}-12p_{1/2}$	0.199(2)	$6f_{5/2}-8d_{3/2}$	44.43(10)	$9f_{5/2}-6d_{5/2}$	0.384(6)
$7p_{1/2}-11d_{3/2}$	0.665(7)	$8p_{1/2}-11d_{3/2}$	1.273(5)	$6d_{3/2}-13p_{1/2}$	0.156(2)	$6f_{5/2}-9d_{3/2}$	23.10(17)	$9f_{5/2}-7d_{5/2}$	0.546(9)
$7p_{1/2}-12d_{3/2}$	0.535(5)	$8p_{1/2}-12d_{3/2}$	0.988(4)	$7d_{3/2}-9p_{1/2}$	6.02(29)	$6f_{7/2}-6d_{5/2}$	4.67(17)	$9f_{5/2}-8d_{5/2}$	0.757(18)
$7p_{3/2}-8s_{1/2}$	7.416(38)	$8p_{3/2}-8s_{1/2}$	13.26(10)	$7d_{3/2}-10p_{1/2}$	1.855(3)	$6f_{7/2}-7d_{5/2}$	7.07(49)	$9f_{5/2}-9d_{5/2}$	1.02(4)
$7p_{3/2}-9s_{1/2}$	1.40(2)	$8p_{3/2}-9s_{1/2}$	15.87(6)	$7d_{3/2}-11p_{1/2}$	1.007(8)	$6f_{7/2}-8d_{5/2}$	53.62(7)	$9f_{5/2}-10d_{5/2}$	0.94(11)
$7p_{3/2}-10s_{1/2}$	0.715(7)	$8p_{3/2}-10s_{1/2}$	2.468(13)	$7d_{3/2}-12p_{1/2}$	0.668(5)	$6f_{7/2}-9d_{5/2}$	26.38(20)	$9f_{5/2}-11d_{5/2}$	33.78(4)
$7p_{3/2}-11s_{1/2}$	0.467(5)	$8p_{3/2}-11s_{1/2}$	1.198(7)	$7d_{3/2}-13p_{1/2}$	0.491(3)	$7f_{5/2}-6d_{3/2}$	2.494(3)	$9f_{5/2}-12d_{5/2}$	18.83(5)
$7p_{3/2}-12s_{1/2}$	0.341(4)	$8p_{3/2}-12s_{1/2}$	0.767(5)	$6d_{5/2}-9p_{3/2}$	0.230(24)	$7f_{5/2}-7d_{3/2}$	3.55(16)	$9f_{7/2}-6d_{5/2}$	1.72(3)
$7p_{3/2}-13s_{1/2}$	0.265(4)	$8p_{3/2}-13s_{1/2}$	0.555(2)	$6d_{5/2}-10p_{3/2}$	0.417(26)	$7f_{5/2}-8d_{3/2}$	4.13(46)	$9f_{7/2}-7d_{5/2}$	2.44(3)
$7p_{3/2}-6d_{3/2}$	3.27(7)	$8p_{3/2}-6d_{3/2}$	0.67(19)	$6d_{5/2}-11p_{3/2}$	0.279(13)	$7f_{5/2}-9d_{3/2}$	67.37(14)	$9f_{7/2}-8d_{5/2}$	3.40(8)
$7p_{3/2}-7d_{3/2}$	2.19(2)	$8p_{3/2}-7d_{3/2}$	8.30(4)	$6d_{5/2}-12p_{3/2}$	0.206(9)	$7f_{5/2}-10d_{3/2}$	37.22(23)	$9f_{7/2}-9d_{5/2}$	4.59(16)
$7p_{3/2}-8d_{3/2}$	1.012(13)	$8p_{3/2}-8d_{3/2}$	3.407(5)	$6d_{5/2}-13p_{3/2}$	0.160(8)	$7f_{5/2}-11d_{3/2}$	9.52(6)	$9f_{7/2}-10d_{5/2}$	4.27(48)
$7p_{3/2}-9d_{3/2}$	0.629(8)	$8p_{3/2}-9d_{3/2}$	1.573(18)	$7d_{5/2}-9p_{3/2}$	6.19(26)	$7f_{5/2}-12d_{3/2}$	3.90(2)	$9f_{7/2}-11d_{5/2}$	151.1(3)
$7p_{3/2}-10d_{3/2}$	0.445(5)	$8p_{3/2}-10d_{3/2}$	0.979(5)	$7d_{5/2}-10p_{3/2}$	2.043(17)	$7f_{7/2}-6d_{5/2}$	3.052(4)	$9f_{7/2}-12d_{5/2}$	84.19(26)
$7p_{3/2}-11d_{3/2}$	0.339(4)	$8p_{3/2}-11d_{3/2}$	0.696(5)	$7d_{5/2}-11p_{3/2}$	1.124(20)	$7f_{7/2}-7d_{5/2}$	4.61(15)	$11s_{1/2}-12p_{1/2}$	4.15(4)
$7p_{3/2}-12d_{3/2}$	0.270(3)	$8p_{3/2}-12d_{3/2}$	0.532(3)	$7d_{5/2}-12p_{3/2}$	0.749(11)	$7f_{7/2}-8d_{5/2}$	6.16(46)	$11s_{1/2}-12p_{3/2}$	8.25(3)
$7p_{3/2}-6d_{5/2}$	10.07(17)	$8p_{3/2}-6d_{5/2}$	2.24(50)	$7d_{5/2}-13p_{3/2}$	0.550(5)	$7f_{7/2}-9d_{5/2}$	81.47(11)	$12s_{1/2}-12p_{1/2}$	50.57(6)
$7p_{3/2}-7d_{5/2}$	6.21(6)	$8p_{3/2}-7d_{5/2}$	25.24(8)	$8d_{3/2}-10p_{1/2}$	11.15(15)	$7f_{7/2}-10d_{5/2}$	9.52(6)	$12s_{1/2}-12p_{3/2}$	64.45(19)
$7p_{3/2}-8d_{5/2}$	2.945(30)	$8p_{3/2}-8d_{5/2}$	9.517(10)	$8d_{3/2}-11p_{1/2}$	3.38(3)	$7f_{7/2}-11d_{5/2}$	8.02(5)	$12s_{1/2}-13p_{1/2}$	5.64(4)
$7p_{3/2}-9d_{5/2}$	1.845(15)	$8p_{3/2}-9d_{5/2}$	4.512(12)	$8d_{3/2}-12p_{1/2}$	1.81(1)	$7f_{7/2}-12d_{5/2}$	4.67(2)	$12s_{1/2}-13p_{3/2}$	11.02(5)
$7p_{3/2}-10d_{5/2}$	1.313(11)	$8p_{3/2}-10d_{5/2}$	2.837(8)	$8d_{3/2}-13p_{1/2}$	1.197(14)	$8f_{5/2}-6d_{3/2}$	1.82(2)	$13s_{1/2}-13p_{1/2}$	65.30(5)
$7p_{3/2}-11d_{5/2}$	1.003(8)	$8p_{3/2}-11d_{5/2}$	2.026(7)	$8d_{5/2}-10p_{3/2}$	11.56(9)	$8f_{5/2}-7d_{3/2}$	2.54(4)	$13s_{1/2}-13p_{3/2}$	82.89(6)
$7p_{3/2}-12d_{5/2}$	0.801(6)	$8p_{3/2}-12d_{5/2}$	1.553(7)	$8d_{5/2}-11p_{3/2}$	3.79(3)	$8f_{5/2}-8d_{3/2}$	3.39(16)		

The lifetimes for the  $ns$  ( $n = 8-13$ ),  $np$  ( $n = 7-13$ ), and  $nd$  ( $n = 6-12$ ) states are determined using our recommended values of the matrix elements and the transition energies from available experiments [48]. The results are listed in Table III, and compared with experimental and other theoretical values. Our results are in excellent agreement with the experimental values for the  $7p_{1/2,3/2}$  [51],  $8p_{1/2,3/2}$  [52],  $8s$  [53],  $9s$  [52], and  $7d_{3/2,5/2}$  [54] states. Safronova *et al.* [55] calculated the

lifetimes for the  $7p_{1/2,3/2}$ ,  $8p_{1/2,3/2}$ ,  $8s$ , and  $7d_{3/2,5/2}$  states using a relativistic all-order method. Our results are very close to theirs. Sahoo *et al.* reported the lifetimes of the  $6d$  states using the relativistic coupled-cluster method in Gaussian basis [56], where the differences between our results and theirs are about 8%. The reason for the discrepancy is that their electric dipole reduced matrix elements for the  $6d-7p$  transitions are larger than ours by about 4%. Wijngaarden *et al.*

TABLE III. Radiative lifetimes of Fr in nanoseconds. Uncertainties are given in parentheses.

States	Present	Other	Expt.
$7p_{1/2}$	29.57(96)	28.44 [57] 29.62 [55]	29.45(11) [51]
$7p_{3/2}$	21.23(71)	20.90 [57] 21.28 [55]	21.02(11) [51]
$6d_{3/2}$	583(15)	540(10) [56] 559.2 [57]	
$6d_{5/2}$	1854(61)	1704(32) [56] 1915 [57]	
$8s_{1/2}$	53.40(67)	54.36 [55] 63.06 [57]	53.30(44) [53]
$8p_{1/2}$	145(12)	102.5 [57]	149.3(3.5) [52]
$8p_{3/2}$	81.8(2.3)	61.13 [57]	83.5(1.5) [52]
$7d_{3/2}$	72.3(2.0)	73.08 [55] 75.90 [57]	73.6(3) [54]
$7d_{5/2}$	67.8(1.3)	67.93 [55] 70.29 [57]	67.7(2.9) [54]
$9s_{1/2}$	107.6(9)	124.3 [57]	107.53(90) [52]
$9p_{1/2}$	319(10)	233.4 [57]	
$9p_{3/2}$	186(2)	130.7 [57]	
$8d_{3/2}$	105(3)	99.87 [57]	
$8d_{5/2}$	101(3)	95.89 [57]	
$10s_{1/2}$	200(2)	230.4 [57]	
$10p_{1/2}$	566(7)	424.3 [57]	
$10p_{3/2}$	346(5)	231.3 [57]	
$9d_{3/2}$	164(4)	144.9 [57]	
$9d_{5/2}$	159(2)	141.7 [57]	
$11s_{1/2}$	337(5)	393.5 [57]	
$11p_{1/2}$	905(4)	691.2 [57]	
$11p_{3/2}$	573(8)	374.8 [57]	
$10d_{3/2}$	248(4)	208.4 [57]	
$10d_{5/2}$	241(4)	205.8 [57]	
$12s_{1/2}$	531(10)	624.4 [57]	
$12p_{1/2}$	1351(12)	1029 [57]	
$12p_{3/2}$	878(11)	562.4 [57]	
$11d_{3/2}$	360(6)	291.7 [57]	
$11d_{5/2}$	352(5)	289.8 [57]	
$13s_{1/2}$	787(12)	935.5 [57]	
$13p_{1/2}$	1926(40)	1482 [57]	
$13p_{3/2}$	1278(30)	799.9 [57]	
$12d_{3/2}$	507(6)	396.7 [57]	
$12d_{5/2}$	496(4)	395.6 [57]	

computed the lifetimes and polarizabilities for 60 low-lying  $S$ ,  $P$ ,  $D$ , and  $F$  states using a Coulomb approximation [57]. It is found that there are obvious differences between ours and their results. Since our approach is an *ab initio* one, our results are expected to be more reliable.

### C. Dipolar scalar and tensor polarizabilities

The dynamic dipole polarizability of a state  $|\Psi_v\rangle$  at photon energy  $\omega$  is defined by

$$\alpha_1(\omega) = \sum_i \frac{f_{vi}^{(1)}}{\varepsilon_{vi}^2 - \omega^2}, \quad (22)$$

where  $f^{(1)}$  represents the dipole oscillator strength, summation includes all intermediate states  $|\Psi_i\rangle$ . The dipole polarizability

has a tensor component if  $J > 1/2$ , which can be written as

$$\alpha_1^T(\omega) = 6 \left( \frac{5J_g(2J_g - 1)(2J_g + 1)}{6(J_g + 1)(2J_g + 3)} \right)^{1/2} \times \sum_{J_i} (-1)^{J_g + J_i} \begin{bmatrix} J_g & 1 & J_i \\ 1 & J_g & 2 \end{bmatrix} \frac{f_{gi}^{(1)}}{\varepsilon_{gi}^2 - \omega^2}. \quad (23)$$

The dynamic polarizability is reduced to the static one if  $\omega = 0$ . The polarizability for a state with nonzero angular momentum  $J$  depends on the magnetic projection  $M_g$

$$\alpha_{1,M_g} = \alpha_1 + \alpha_1^T \frac{3M_g^2 - J_g(J_g + 1)}{J_g(2J_g - 1)}. \quad (24)$$

The dynamic polarizability includes contributions from the core that is represented by a pseudo-oscillator strength distribution [41]. In this work,  $\alpha_{\text{core}} = 20.4$  was used. The summation includes the contributions from all bound states and the continuum. For the low-lying intermediate states, we used the recommended reduced matrix elements given in the above section. For the contributions from higher-lying states and the continuum, we used the reduced matrix elements obtained by the Dirac-Fock plus core potential method (DFCP) [41]. In DFCP, 80  $B$  splines of order 15 were used. The size of the  $B$ -spline domain was set to be 500 a.u. All polarizabilities were computed using available experimental transition energies.

Table IV lists the static dipole polarizabilities for  $S$ ,  $P$ , and  $D$  states (in multiples of 1000) and a comparison with other theoretical results. For the ground state, our value for  $\alpha^{(1)}$  is 315(9), which is in agreement with the value 317.8(2.4) of Derevianko *et al.* using an all-order method [58], with the value 316.81 of Singh *et al.* using the RCC method [59], with the value 315.2 of Lim *et al.* [60] and with the value 311.5 of Borschevsky *et al.* [61] using the RCC and a finite field method. For the  $7p$  states, our results are different from the *ab initio* results of Singh *et al.* [59] by about 5%. Wijngaarden *et al.* reported the polarizabilities for the  $S$ ,  $P$ ,  $D$ , and  $F$  states using a Coulomb approximation [57]. Similar to the case of lifetimes of excited states, our results have larger discrepancy from theirs. For the  $D$  states, we find that the contributions from the  $f$  states have large cancellation with the  $p$  states, leading to larger uncertainties.

### D. Tune-out and magic wavelengths

Recently, Dammalapati *et al.* reported the tune-out wavelengths of  $7s$  and the magic wavelengths of  $7s$ - $8s$  and  $7s$ - $7p$  transitions in Fr [62]. In their calculations, they ignored the contributions from atomic core part and from the transitions  $8s$ - $np$  ( $n = 7$ - $10$ ) and  $7p$ - $6d$ . Thus, Singh *et al.* recalculated the magic wavelengths for the  $7s$ - $7p$  transition using the RCC method, and reported a few different results [59]. In this paper, we evaluated the tune-out wavelengths for  $7s$  and the magic wavelengths for the transitions  $7s$ - $8s$  and  $7s$ - $7p$  using our recommended reduced matrix elements and experimental transition energies. For the reduced matrix elements of  $7s$ - $7p_{1/2}$  and  $7s$ - $7p_{3/2}$ , we used 4.277 and 5.898 that are extracted from the lifetime measurements [51].

Table V lists the tune-out wavelengths of the ground state in the range of 340–800 nm. A comparison is made with

TABLE IV. Static dipole scalar and tensor polarizabilities (in multiples of 1000) for Fr. Uncertainties are given in parentheses.

States	$\alpha^{(1)}$		$\alpha_t^{(1)}$	
	Present	Others	Present	Others
$7s_{1/2}$	0.315(9)	0.31681 [59] 0.3178(24) [58] 0.315 [60] 0.305 [57]		
$8s_{1/2}$	4.700(71)	4.600 [57]		
$9s_{1/2}$	26.33(21)	27.0 [57]		
$10s_{1/2}$	95.27(87)	105.9 [57]		
$11s_{1/2}$	262.0(4.1)	319.6 [57]		
$12s_{1/2}$	606.9(20.0)	815.2 [57]		
$13s_{1/2}$	1201(45)	1840 [57]		
$7p_{1/2}$	1.154(22)	1.225 [59] 1.106 [57]		
$8p_{1/2}$	22.68(11)	22.490 [57]		
$9p_{1/2}$	160.1(8)	148.9 [57]		
$10p_{1/2}$	710(4)	636.0 [57]		
$11p_{1/2}$	2385(16)	2061 [57]		
$12p_{1/2}$	6658(56)	5671 [57]		
$13p_{1/2}$	16231(71)	13390 [57]		
$7p_{3/2}$	2.155(60)	2.304 [59] 2.102 [57]	-0.446(7)	-0.467 [59] -0.403 [57]
$8p_{3/2}$	43.10(31)	42.930 [57]	-6.572(27)	-6.463 [57]
$9p_{3/2}$	313.9(1.4)	285.9 [57]	-42.59(37)	-38.91 [57]
$10p_{3/2}$	1417(5)	1250 [57]	-181.2(1.3)	-160.7 [57]
$11p_{3/2}$	4824(19)	4045 [57]	-591(7)	-501.7 [57]
$12p_{3/2}$	13583(50)	11020 [57]	-1623(18)	-1334 [57]
$13p_{3/2}$	33326(61)	26420 [57]	-3884(6)	-3137 [57]
$6d_{3/2}$	-0.274(29)	-0.330 [57]	0.242(12)	0.254 [57]
$7d_{3/2}$	-0.336(78)	-0.270 [57]	4.078(41)	4.016 [57]
$8d_{3/2}$	-20.07(86)	-12.56 [57]	31.23(28)	28.94 [57]
$9d_{3/2}$	-141.5(4.9)	-97.17 [57]	145.0(2.0)	130.2 [57]
$10d_{3/2}$	-598.0(18.0)	-400.4 [57]	502.0(7.0)	445.6 [57]
$11d_{3/2}$	-1917(54)	-1298 [57]	1434(20)	1286 [57]
$12d_{3/2}$	-5039(153)	-3411 [57]	3530(47)	3103 [57]
$6d_{5/2}$	-0.610(8)	0.269 [57]	0.872(15)	-0.048 [57]
$7d_{5/2}$	-7.398(50)	-7.367 [57]	18.38(11)	18.36 [57]
$8d_{5/2}$	-73.42(83)	-59.69 [57]	142.8(4)	128.1 [57]
$9d_{5/2}$	-386.0(5.0)	-308.3 [57]	666.0(2.0)	578.9 [57]
$10d_{5/2}$	-1442(18)	-1075 [57]	2315(7)	1907 [57]
$11d_{5/2}$	-4310(55)	-3102 [57]	6607(21)	5266 [57]
$12d_{5/2}$	-10951(140)	-7787 [57]	16353(55)	12760 [57]

the results reported by Dammalapati *et al.* [62]. Our results are in reasonable agreement with theirs except those lying between the  $7s$ - $8p_{1/2}$  and  $7s$ - $8p_{3/2}$  transition energies, which have about 2.6 nm difference.

Table VI lists the magic wavelengths for  $7s$ - $7p$  in the range of 600–1500 nm and for  $7s$ - $8s$  in the range of 800–1500 nm, together with a comparison with other theoretical results. The major differences between our results and the values from Ref. [62] and Ref. [59] are highlighted in bold. For the  $7s$ - $7p$  transition, some significant discrepancies between ours and the results of Ref. [62] are observed. It is noted that ours are consistent with the those by Singh *et al.* [59]. The contributions from the  $7p$ - $6d$  and  $7p$ - $8s$  transitions were not included in

TABLE V. Tune-out wavelengths of the ground state of Fr in nanometers. Uncertainties are given in parentheses. The values with an asterisk (\*) are from the experimental lifetimes of the  $7p_{1/2,3/2}$  states.

Resonance	$\lambda_{\text{Res.}}$	$\lambda_{\text{Tune}}$	Ref. [62]
$7s$ - $7p_{1/2}$	817.1664	782.084(7)	782.96, 781.65*
$7s$ - $7p_{3/2}$	718.1846		
$7s$ - $8p_{1/2}$	432.6577	433.70(15) 427.11(14)	433.87 424.49
$7s$ - $8p_{3/2}$	422.6845		
$7s$ - $9p_{1/2}$	368.7559	368.87(2) 366.57(3)	368.96 365.76
$7s$ - $9p_{3/2}$	365.4143		
$7s$ - $10p_{1/2}$	344.0661	344.09(1)	
$7s$ - $10p_{3/2}$	342.4881	342.97(2)	

Ref. [62] that may explain the observed discrepancies. The slight discrepancies between our magic wavelengths and the results by Singh *et al.* [59] could be attributed to the different reduced matrix elements used by us and by Singh *et al.* [59].

For the  $7s$ - $8s$  transition, the magic wavelength in the range of 800–1500 nm occurs near the resonant position of  $8s$ - $np$  ( $n = 10$ -18). In this range, 19 magic wavelengths were determined in our calculations, while Dammalapati *et al.* only gave two magic wavelengths. This may be due to the fact that they ignored the contributions from  $8s$ - $np$  ( $n = 7$ -10). The determinations of these magic wavelengths require knowledge of the transition matrix elements from  $8s$  to higher  $p$  states. In our CC calculations, we obtained  $np$  ( $n = 7$ -13) bound states. The higher-lying  $np$  ( $n = 14$ -22) bound states and the continuum were obtained using the DFCP method.

#### IV. SUMMARY

In this paper, we have presented a relativistic Fock space multireference coupled-cluster method for atomic structure calculations. The no-pair Dirac-Coulomb-Breit Hamiltonian is our starting point, together with a finite  $B$ -spline basis set to expand the large and small components of the Dirac wave functions. We have used this method to calculate the Fr atomic properties, including ionization energies, reduced matrix elements, lifetimes, and static electric dipolar scalar and tensor polarizabilities. To investigate the role of electron correlation effects and evaluate the uncertainties of our results, we have performed the calculations using the LCCSD, CCSD, LCCSD<sub>s</sub>, and CCSD<sub>s</sub> approximation models. Our results have shown that the nonlinear terms of the cluster operators are important for precise determinations of atomic properties, especially for the ionization energies. Most of our CCSD ionization energies agree with experimental values at the level of 0.1%. Our lifetimes for the  $8s$ ,  $9s$ ,  $7p$ ,  $8p$ , and  $7d$  states are in excellent agreement with the experimental values.

Furthermore, the tune-out wavelengths of the ground state and magic wavelengths for the  $7s$ - $8s$  and  $7s$ - $7p$  transitions have been determined by evaluating the dynamic polarizabilities of the  $7s$ ,  $8s$ , and  $7p$  states. Our magic wavelengths for the  $7s$ - $7p$  transition in the range of 600–1500 nm are

TABLE VI. Magic wavelengths for  $7s$ - $8s$  and  $7s$ - $7p$  transitions in Fr with linearly polarized light in the range of 600–1500 nm. Uncertainties are given in parentheses.

$7s$ - $7p_{1/2},  m_j  = 1/2$		$7s$ - $7p_{3/2},  m_j  = 1/2$		$7s$ - $7p_{3/2},  m_j  = 3/2$		$7s$ - $8s_{1/2},  m_j  = 1/2$	
Present	Others	Present	Others	Present	Others	Present	Others
<b>1484(4)</b>	<b>1479.49 [59]</b>	<b>1025(3)</b>	<b>1017.45 [59]</b> <b>1266.3 [62]</b>	<b>1098(6)</b>	<b>1076.60 [59]</b> <b>1117.7 [62]</b>	1356.5(1)	
<b>839.2(2)</b>	<b>838.08 [59]</b> <b>871.62 [62]</b>	968.79(1)	968.79 [59] 968.83 [62]	967.03(1)	967.03 [59] 967.19 [62]	1073.4(1) 1061.7(1)	
<b>771.9(1)</b>	<b>771.03 [59]</b> <b>797.75 [62]</b>	852.84(3)	852.84 [59] 853.93 [62]	783.17(1)	782.83 [59] 783.35 [62]	959.75(2) 953.39(2)	960.13 [62] 953.09 [62]
745.57(2)	745.6 [59] 745.36 [62]	<b>798.87(3)</b>	<b>798.74 [59]</b> <b>784.62 [62]</b>	731.91(4)	731.88 [59] 731.77 [62]	900.86(1) 897.02(2)	
<b>645.69(15)</b>	<b>646.05 [59]</b> <b>642.85 [62]</b>	731.21(4)	731.21 [59] 731.32 [62]	729.78(2)	729.77 [59] 729.73 [62]	865.68(2) 863.17(2)	
621.43(2)	621.48 [59] 621.11 [62]	729.65(2)	729.63 [59] 730.51 [62]	649.50(1)	649.50 [59] 649.51 [62]	842.75(2) 841.00(1)	
		694.91(1)	694.92 [59] 694.67 [62]	645.52(6)	645.64 [59] 645.95 [62]	826.87(1) 825.60(1)	
		649.65(1)	649.65 [59] 649.65 [62]	610.18(1)	610.18 [59] 610.20 [62]	815.37(1) 814.42(1)	
		646.42(7)	646.49 [59] 645.11 [62]	607.22(5)	607.59 [59] 606.66 [62]	806.77(1) 806.02(1)	
		632.79(1)	632.83 [59] 632.38 [62]			800.15(1) 799.54(1)	
		610.27(5)	610.27 [59]				
		607.90(4)	608.15 [59] 605.64 [62]				
		600.86(1)	600.89 [59] 600.33 [62]				

consistent with the results reported by Singh *et al.* [59]. For the  $7s$ - $8s$  transition, we have found that there are 19 magic wavelengths in the range of 800–1500 nm. We hope that the atomic properties and magic wavelengths for Fr reported in this paper will be useful for analyzing systematic uncertainties and for designing experiments.

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#### APPENDIX: MATRIX ELEMENTS OF BREIT INTERACTION

$$B_{ij}^{QQ} = \sum_{b,K} D_{b,K} [e^{Q_b Q_b}(K) R_{ij}^{Q_b Q_b}(K+1) + f^{Q_b Q_b}(K) R_{ij}^{Q_b Q_b}(K-1) + g(K) R_{ij}^{Q_b Q_b}(K)], \quad (A1)$$

$$B_{ij}^{PP} = \sum_{b,K} D_{b,K} [e^{P_b P_b}(K) R_{ij}^{P_b P_b}(K+1) + f^{P_b P_b}(K) R_{ij}^{P_b P_b}(K-1) + g(K) R_{ij}^{P_b P_b}(K)], \quad (A2)$$

$$B_{ij}^{QP} = \sum_{b,K} D_{b,K} \{ e^{Q_b P_b}(K) R_{ij}^{Q_b P_b}(K+1) + f^{Q_b P_b}(K) R_{ij}^{Q_b P_b}(K-1) + g(K) R_{ij}^{Q_b P_b}(K) + h [W_{ij}^{Q_b P_b}(K-1) - W_{ij}^{Q_b P_b}(K+1)] \} = B_{ij}^{PQ}, \quad (A3)$$

where

$$R_{ij}^{X_b Y_b}(K) = \sum_{k,l}^N C_k^{X_b} C_l^{Y_b} \int B_i(r_1) B_k(r_1) dr_1 \left[ \int_0^{r_1} \frac{r_2^K}{r_1^{K+1}} B_j(r_2) B_l(r_2) dr_2 + \int_{r_1}^{R_{\max}} \frac{r_1^K}{r_2^{K+1}} B_j(r_2) B_l(r_2) dr_2 \right], \quad (\text{A4})$$

$$W_{ij}^{X_b Y_b}(K) = \sum_{k,l}^N C_k^{X_b} C_l^{Y_b} \int B_i(r_1) B_k(r_1) dr_1 \left[ \int_0^{r_1} \frac{r_2^K}{r_1^{K+1}} B_j(r_2) B_l(r_2) dr_2 - \int_{r_1}^{R_{\max}} \frac{r_1^K}{r_2^{K+1}} B_j(r_2) B_l(r_2) dr_2 \right], \quad (\text{A5})$$

with

$$e^{XY}(K) = \begin{cases} -\frac{[(K+1)+(\kappa_v-\kappa_b)][(K+3)(\kappa_b-\kappa_v)+K(K+1)]}{2(K+1)(2K+3)}, & X = Y = Q \\ \frac{[K(K+1)^2+(3+K)(\kappa_v-\kappa_b)^2]}{2(K+1)(2K+3)}, & X = Q, Y = P \\ -\frac{[(K+1)+(\kappa_b-\kappa_v)][(K+3)(\kappa_v-\kappa_b)+K(K+1)]}{2(K+1)(2K+3)}, & X = Y = P \end{cases} \quad (\text{A6})$$

$$f^{XY}(K) = \begin{cases} \frac{[K+(\kappa_b-\kappa_v)][(K-2)(\kappa_v-\kappa_b)+K(K+1)]}{2K(2K-1)}, & X = Y = Q \\ -\frac{[(K-2)(\kappa_b-\kappa_v)^2+K^2(K+1)]}{2K(2K-1)}, & X = Q, Y = P \\ \frac{[K+(\kappa_v-\kappa_b)][(K-2)(\kappa_b-\kappa_v)+K(K+1)]}{2K(2K-1)}, & X = Y = P \end{cases} \quad (\text{A7})$$

$$D_{b,K} = (2j_b + 1) \begin{pmatrix} j_v & K & j_b \\ -1/2 & 0 & 1/2 \end{pmatrix}^2, \quad (\text{A8})$$

$$g(K) = \frac{(\kappa_b + \kappa_v)^2}{K(K+1)}, \quad (\text{A9})$$

$$h = \frac{(\kappa_v - \kappa_b)}{2}. \quad (\text{A10})$$

In the above,  $b$  represents a core orbital, running over all core orbitals.

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