

## $M_F$ -Dependent Lifetimes due to Hyperfine Induced Interference Effects

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We report on the first theoretical investigation of  $M_F$ -dependent lifetimes due to interference between a magnetic octupole transition and a hyperfine induced electric quadrupole transition. Extensive multi-configuration Dirac-Fock calculations are used to model the hyperfine quenching of the magnetic octupole decay of  $3d^9 4s\ ^3D_3$  and the state mixing between the  $^3D_3$  and  $^3D_2$  due to hyperfine interaction in nickel-like  $\text{Xe}^{26+}$ .

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Forbidden transitions provide fundamental information about atomic structure and are important tools for diagnostics and modeling of astrophysical and laboratory plasmas. In this Letter we focus on a magnetic octupole transition ( $M3$ ) and an electric quadrupole transition ( $E2$ ) to investigate the effect of hyperfine quenching on the lifetime of the  $^3D_3$  metastable level in nickel-like  $\text{Xe}^{26+}$ .

In nickel-like ions, the ground state and first excited states are  $3d^{10}\ ^1S$  and  $3d^9 4s\ ^1,^3D$ , outside a  $1s^2 2s^2 2p^6 3s^2 3p^6$  core. The lowest excited level  $^3D_3$  can decay to the ground state only via an  $M3$  transition, while the decays of the two  $J = 2$  levels are dominated by  $E2$  transitions (see Fig. 1). There are extensive theoretical [1,2] and experimental [3–8] investigations of nickel-like ions. However, very few studies, either theoretical or experimental, have dealt with the more “exotic”  $M3$  decay. It was first reported in the x-ray region of the spectra of the highly charged ions  $\text{Th}^{62+}$  and  $\text{U}^{64+}$  [9] at the LLNL EBIT, followed by a theoretical investigation by Biémont *et al.* [10]. Recently, Träbert *et al.* [11] did a more detailed investigation of the  $^3D_3$  in nickel-like xenon, reporting a lifetime of  $11.5 \pm 0.5$  ms. This is about 20%–60% shorter than the theoretical results in the same publication, using an average level multiconfiguration Dirac-Fock (MCDF) method, and the results by Safronova *et al.* [12] and Träbert *et al.* [13], using a many-body perturbation theory (MBPT). The  $^3D_3$  lifetime studies of  $\text{Cs}^{27+}$  and  $\text{Ba}^{28+}$  were reported also in [11]. The measured lifetimes in the latter two cases,  $8.2 \pm 2.0$  ms and  $4.3 \pm 3.6$  ms, respectively, have larger error bars. These measurements are more than 60% shorter than what has so far been predicted by theory [12,13]. We suggest that these discrepancies are due to the hyperfine induced state mixing between the  $^3D_3$  and  $^3D_2$  levels, opening a new decay process. We also propose for the first time that the interference between the  $M3$  transition and the hyperfine induced  $E2$  transition results in lifetimes which are dependent on the magnetic quantum number of the initial level. Our arguments are supported by large-scale MCDF calculations and by simu-

lations of the resulting complex  $^3D_3$  decay by one-exponential fitting.

The basic ideas behind hyperfine quenching is that in the presence of a nuclear spin  $I$ , an additional hyperfine operator  $H_{\text{hpf}}$  is introduced into the otherwise electronic Hamiltonian. This Hamiltonian commutes with the total angular momentum  $F (= J + I)$  and  $F_z$ , instead of the total electronic angular momentum  $J$  and  $J_z$ . This hyperfine interaction therefore not only splits  $J$  levels, but also introduces a wave function mixing between levels of different  $J$  [14]. An experimental indication of hyperfine quenching was reported by Gould *et al.* [15]. The mixing can be represented as

$$|\Gamma JIF\rangle = |\Gamma_0 J_0 IF\rangle + \sum_i \epsilon_i |\Gamma_i J_i IF\rangle, \quad (1)$$

where  $\Gamma$  denotes all other quantum numbers needed to completely specify the state, and the main contribution represents the “pure” state  $|\Gamma_0 J_0 IF\rangle$ , in the absence of the hyperfine interaction. According to first order perturbation theory, the mixing coefficient can be computed as

$$\epsilon_i \approx \frac{\langle \Gamma_i J_i IFM_F | H_{\text{hpf}} | \Gamma_0 J_0 IFM_F \rangle}{E(\Gamma_0 J_0) - E(\Gamma_i J_i)}. \quad (2)$$

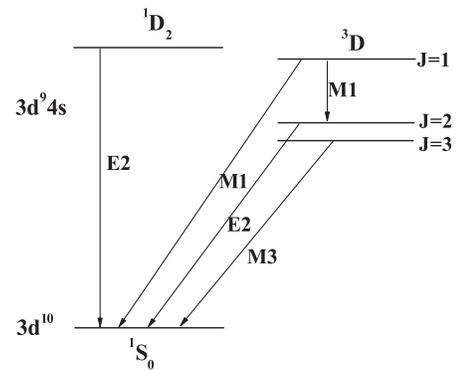


FIG. 1. Schemes of the ground level  $3d^{10}\ ^1S_0$  and first excited levels  $3d^9 4s\ ^1,^3D$  in Ni-like ions. The main decays involved are also given. LS term designation is used.

In this Letter we will consider only the most important contribution to the hyperfine interaction, the magnetic dipole interaction, which will mix different  $J$  levels only with an angular momentum difference  $\Delta J \leq 1$ . Since the significant contributions will come only from within the same configuration, this interaction will mix the  ${}^3D_3$  level only with  $J = 2$  levels of the same parity and then open a new decay process for the impure “ ${}^3D_3$ ” state besides its own  $M3$  transition. Since the  $E2$  transition rates,  $A({}^1S_0-{}^3D_2)$  and  $A({}^1S_0-{}^1D_2)$ , involved here are more than 6 orders of magnitude larger than the  $M3$  transition rate, it is clear that even a very small state mixing will cause significant influence on the lifetime of the  ${}^3D_3$  level. Our

calculation shows that the hyperfine induced mixing of  ${}^3D_3$  with  ${}^1D_2$  is over 2 orders of magnitudes lower than that with  ${}^3D_2$ , so it can be neglected. The mixing between  ${}^3D_2$  and  ${}^1D_2$  is already included in relativistic calculation. The mixing of the “ $3d^94s\ {}^3D_3$ ” state can then be written as

$$|\Gamma_0^3D_3IF\rangle = |\Gamma_0^3D_3IF\rangle + \epsilon|\Gamma^3D_2IF\rangle. \quad (3)$$

Normally a transition rate is proportional to the square of the reduced transition matrix element, with coefficients  $K_{p,k,\lambda}$  depending on the type  $p$ , order  $k$ , and wavelength  $\lambda$  of the transition. The transition rate of a specific  $FM_F$  sublevel in the mixed “ ${}^3D_3$ ” to all the  $M'_F$  sublevels of ground state  $|\Gamma^1S_0IF'\rangle$  can then be expressed as

$$\begin{aligned} A(F, M_F) &= \sum_{M'_F, q} |\langle \Gamma^1S_0IF'M'_F | \sqrt{K'_{M,3,\lambda}} M_q^{(3)} + \sqrt{K'_{E,2,\lambda}} E_q^{(2)} | \Gamma_0^3D_3IFM_F \rangle|^2 \\ &= K_{M,3,\lambda} \langle \Gamma^1S_0 || M^{(3)} || \Gamma_0^3D_3 \rangle^2 + \epsilon^2 \cdot K_{E,2,\lambda} \langle \Gamma^1S_0 || E^{(2)} || \Gamma^3D_2 \rangle^2 + 2\epsilon(2F+1) \\ &\quad \times \sqrt{K_{M,3,\lambda} K_{E,2,\lambda}} \langle \Gamma^1S_0 || M^{(3)} || \Gamma_0^3D_3 \rangle \langle \Gamma^1S_0 || E^{(2)} || \Gamma^3D_2 \rangle \sum_{M'_F, q} \begin{pmatrix} I & 3 & F \\ -M'_F & q & M_F \end{pmatrix} \begin{pmatrix} I & 2 & F \\ -M'_F & q & M_F \end{pmatrix}, \quad (4) \end{aligned}$$

where  $K' = (2J+1)K$ . In this expression, the first term on the rightmost side represents the pure  $M3$  transition rate  $A(M3)$ . The second term represents the hyperfine induced  $E2$  transition rate  $\epsilon^2 A(E2)$ , which is  $F$  dependent due to the  $F$ -dependent mixing coefficient  $\epsilon$ . The last term is the interference term  $A(\text{inter})$  between the  $M3$  and the hyperfine induced  $E2$  transition. In this case, when the  $A(M3)$  and  $\epsilon^2 A(E2)$  are comparable in size, the interference term cannot be neglected. Since the sum of the product of two 3- $j$  symbols depends on  $M_F$ , the total transition rate  $A(F, M_F)$  will be  $M_F$  dependent.

We use the GRASP VU package, a modified version of GRASP92 [16], which is based on the fully relativistic MCDF method [17] to determine transition rates and energies. In the MCDF method, the atom is represented by an atomic state function  $\Psi(\Gamma J)$ , a linear combination of configuration state functions (CSFs)  $\Phi(\alpha_i J)$ ,  $\Psi(\Gamma J) = \sum_i c_i \Phi(\alpha_i J)$ . The CSFs are constructed from the sum of products of one-electron spin orbitals. The coefficient  $c_i$  and spin orbitals are optimized by minimizing an energy functional according to the self-consistent field method.

We use a restricted active space approach [18], which is based on the active set (AS) [19,20] of orbitals, to include correlations. In this approach, the active set is increased in a systematic way. Considering the relative importance of different excitations from the viewpoint of order-by-order expansion of energy in perturbation theory [21], we allowed single and double replacements from the reference configuration to an active set. Valence correlation and core-valence correlation are both included in the calculation. In the first step of our calculation, which we label Dirac-Fock (DF) in Table I, all orbitals of  $3d^{10}$  and  $3d^94s$  are optimized in independent extended average calculations [17]. In the second step, we include single excitation from the  $3s$  and  $3p$  subshells, and single and double excitations from the  $3d$  and  $4s$  subshells, to the active set AS1 =  $\{4s, 4p, 4d, 4f\}$ . Only the orbitals added in this step are optimized. In the third step, the same excitations are allowed to the active set AS2 = AS1 +  $\{5s, 5p, 5d, 5f\}$ , followed by optimization of the new orbitals. In the fourth step, in addition to the excitations in the previous step, single and double excitations from  $3d$  and  $4s$  are allowed to the active set AS3 = AS2 +  $\{6s, 6p, 6d, 6f\}$ . Finally, single  $s$ -electron excita-

TABLE I.  $\text{Xe}^{26+}$  excitation energies (in  $\text{cm}^{-1}$ ) from different calculations (see text) and experiment.

	Method	${}^3D_3$	${}^3D_2$	${}^3D_1$	${}^1D_2$
This work	DF	4 757 839	4 766 207	4 869 177	4 875 505
	AS1	4 765 543	4 774 172	4 873 759	4 880 026
	AS2	4 758 245	4 766 993	4 866 560	4 872 984
	AS3	4 757 727	4 766 513	4 866 033	4 872 502
	AS4	4 757 588	4 766 359	4 865 900	4 872 355
[12]	MBPT	4 758 506	4 766 899	4 866 417	4 873 769
[11]	MCDF	4 749 680	4 758 520	4 861 113	4 868 001
[11]	Experiment	4 761 859	4 770 247		4 876 308

TABLE II. Computed transition rates (without hyperfine quenching) in  $\text{Xe}^{26+}$  ( $\text{s}^{-1}$ ). For the two  $E2$  transitions (from  ${}^3D_2$  and  ${}^1D_2$  states), results from both Coulomb and Babushkin (in brackets) gauges are presented.

	Method	${}^3D_3$	${}^3D_2$	${}^3D_1$	${}^1D_2$
This work	AS1	62.90	$2.58(2.62) \times 10^8$	80.45	$2.36(2.39) \times 10^8$
	AS2	64.93	$2.63(2.65) \times 10^8$	94.91	$2.40(2.41) \times 10^8$
	AS3	66.20	$2.65(2.67) \times 10^8$	90.44	$2.41(2.43) \times 10^8$
	AS4	66.15	$2.65(2.67) \times 10^8$	94.12	$2.41(2.43) \times 10^8$
[11]	MCDF	71.30	$2.72 \times 10^8$	89.20	$2.49 \times 10^8$
[12]	MBPT	53.70	$2.26 \times 10^8$	136.0	$2.06 \times 10^8$

tions from the reference configurations to the active sets  $\text{AS4} = \{4s, 5s, 6s, 7s, 8s\}$  are included, to take care of the main contribution from the spin polarization effect [22,23]. The resulting expansion is over 79 363 CSFs. In this last step, only the  $7s$  and  $8s$  orbitals are optimized. In the final configuration interaction calculation, we add transverse photon contributions, vacuum polarization, and normal and specific mass shifts to the Dirac-Coulomb Hamiltonian as perturbations.

Table I presents the transition energies calculated in this work at different configuration expansions, along with

those from other calculations and the experiment. The computed transition energies in this work are consistently smaller than the experimental results by about  $4000 \text{ cm}^{-1}$ , due to the slower convergence of correlation in the ground state. But this is less than 0.1% of the excitation energy. More importantly, our “fine structure” energy splittings  $E({}^3D_2) - E({}^3D_3) = 8771 \text{ cm}^{-1}$  and  $E({}^1D_2) - E({}^3D_3) = 114767 \text{ cm}^{-1}$  are in excellent agreement with the experimental values. It is clear from Eq. (2) that this energy splitting is most important for computing hyperfine quenching. The transition rates are shown in Table II. For

TABLE III. Contribution to the  $M_F$ -dependent rates  $A(\text{s}^{-1})$  for the  $3d^{10} {}^1S_0 \rightarrow 3d^9 4s {}^3D_3$  transition [according to Eq. (4)] and the final lifetime of “ ${}^3D_3$ ” sublevels of  $\text{Xe}^{26+}$ .

	Isotope	$I$	$F$	$M_F$	$A(M3)$	$\epsilon^2 A(E2)$	$A(\text{inter})$	$A(F, M_F)$	$\tau(\text{ms})$		
This work	Xe	Even isotopes			66.15	0.0	0.0	66.15	15.12		
			${}^{129}\text{Xe}$	1/2	7/2	66.15	0.0	0.0	66.15	15.12	
				5/2	+5/2	66.15	286.34	-104.04	248.45	4.02	
					+3/2	66.15	286.34	-27.56	324.93	3.08	
					+1/2	66.15	286.34	-7.98	344.51	2.90	
					-1/2	66.15	286.34	7.98	360.47	2.77	
					-3/2	66.15	286.34	27.56	380.05	2.63	
					-5/2	66.15	286.34	104.04	456.53	2.19	
		${}^{131}\text{Xe}$	3/2	9/2		66.15	0.0	0.0	66.15	15.12	
				7/2	+7/2	66.15	96.36	92.19	254.70	3.93	
					+5/2	66.15	96.36	43.78	206.30	4.85	
					+3/2	66.15	96.36	19.39	181.90	5.50	
					+1/2	66.15	96.36	5.51	168.02	5.95	
					-1/2	66.15	96.36	-5.51	157.00	6.37	
					-3/2	66.15	96.36	-19.39	143.12	6.99	
					-5/2	66.15	96.36	-43.78	118.73	8.42	
					-7/2	66.15	96.36	-92.19	70.32	14.22	
					5/2	+5/2	66.15	92.53	104.22	262.90	3.80
						+3/2	66.15	92.53	44.27	202.95	4.93
						+1/2	66.15	92.53	10.15	168.83	5.92
				-1/2	66.15	92.53	-10.15	148.53	6.73		
				-3/2	66.15	92.53	-44.27	114.41	8.74		
				-5/2	66.15	92.53	-104.22	54.46	18.36		
			3/2	+3/2	66.15	45.95	69.19	181.29	5.52		
			+1/2	66.15	45.95	11.31	123.41	8.10			
			-1/2	66.15	45.95	-11.31	100.80	9.92			
			-3/2	66.15	45.95	-69.19	42.91	23.30			
	Fitted lifetime:				11.80 ms (population I)		11.85 ms (population II)		11.65 ms (population III)		
Experiment	[11]						$87 \pm 4$	$11.5 \pm 0.5$			
Theory	[11]				71.30				14.03		
Theory	[12]				53.70				18.62		

the two electric quadrupole transitions ( $3d^{10} 1S_0—3d^9 4s^3 D_2$  and  $3d^{10} 1S_0—3d^9 4s^1 D_2$ ), both Coulomb and Babushkin gauges are given; they are in excellent agreement. All the transition rates, except the  $M1$  transition which is not important in this discussion, change by less than 5% when correlation is included. Both energy and transition results are converged and give us confidence that the main correlations are included.

The hyperfine interaction was included as a perturbation after the MCDF calculations using the program HFSVU, a modified version of HFS92 [24], to compute the mixed atomic wave function of [ $\Gamma^3 D_3 IFM_F$ "]. Our calculation shows that the matrix element in Eq. (2) for hyperfine interaction between  $^3 D_3$  and  $^1 D_2$  is about 1/10 of that between  $^3 D_3$  and  $^3 D_2$ ; the energy splitting is more than 10 times larger than that between the latter, so the mixing coefficient  $\epsilon(^1 D_2)$  is about 2 orders of magnitude smaller than  $\epsilon(^3 D_2)$ . While their transition rates are comparable, the contribution from  $^1 D_2$  to the  $^3 D_3$  decay is negligible compared to that from  $^3 D_2$ . An overview of the  $M_F$ -dependent transition rate and lifetime for each individual sublevel along with our simulated lifetime results and previous results are shown in Table III. For the isotopes with zero nuclear spin, only the  $M3$  transition can occur. It is clear that the  $M_F$  dependence is introduced by the non-zero interference term between the  $M3$  transition and the hyperfine induced  $E2$  transition. It is worth noting that the  $M_F$  dependent lifetimes span over a wide range. The superposition of these decays will lead to a multiexponential behavior in the decay of the composite “ $^3 D_3$ ” level. This can be expressed as

$$I(t) = I_0 \sum_{i,F,M_F} W_i W_{i,F,M_F} \exp[-t/\tau(i, F, M_F)], \quad (5)$$

where  $I_0$ ,  $W_i$ , and  $W_{i,F,M_F}$  represent the initial intensity, isotope abundance, and the weight of sublevel population in a given isotope. For the natural abundance of xenon, we composed three synthetic decay curves based on expression (5) assuming three extreme populations: (I) a uniform population on the  $M_F$  sublevels, (II) an increasing population along  $M_F$ ,  $|M_F|f(F)$  on an  $M_F$  sublevel, and (III) a decreasing population  $(F+1-|M_F|)f(F)$ , to estimate any polarization effect.  $f(F)$  is an  $F$ -dependent normalization coefficient. Fitting the synthetic decay curves with single exponential leads to lifetimes of 11.80, 11.85, and 11.65 ms, respectively, corresponding to the three above mentioned population assumptions; see Table III. Our results agree much better with the experiment results  $11.5 \pm 0.5$  ms [11] compared to the previous calculations [11,12]. This supports our proposal that the interference between the  $M3$  transition and the hyperfine induced  $E2$  transition plays an important role in the decay of the “ $^3 D_3$ ” level in  $\text{Xe}^{26+}$ . Furthermore, the different lifetimes deduced from the three assumptions show the effect of polarization on  $M_F$  sublevel populations. The lifetime value of 11.65 ms is closest to the experimental value [11], possibly implying that the decreasing population is closest

to the real distribution under the experiment condition in [11]. We suggest an experiment on the lifetime of the “ $^3 D_3$ ” of a pure xenon isotope with nonzero nuclear spin, in which case the polarization effect would show more influence on the lifetime.

In conclusion, the excited level  $3d^9 4s^3 D_3$  in nickel-like xenon was investigated using MCDF with inclusion of the hyperfine interaction. For the first time, it is proposed that not only the mixing of the level of  $^3 D_3$  with  $^3 D_2$  by the hyperfine interaction but also the interference between the  $M3$  and the hyperfine induced  $E2$  transitions, which gives rise to  $M_F$  dependent sublevel lifetime, is important for the “ $^3 D_3$ ” level decay. This provides a possibility to study the polarization by measuring level lifetimes. This could be another useful tool for plasma condition diagnostics.

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- [1] C. Z. Dong *et al.*, Phys. Scr. **T92**, 314 (2001).
- [2] U. I. Safronova, W. R. Johnson, and J. R. Albritton, Phys. Rev. A **62**, 052505 (2000).
- [3] C. L. Cocke *et al.*, Phys. Rev. A **12**, 2413 (1975).
- [4] M. Klapisch *et al.*, Phys. Rev. Lett. **41**, 403 (1978).
- [5] J. F. Wyart *et al.*, Phys. Rev. A **34**, 701 (1986).
- [6] I. Y. Skobelev *et al.*, J. Phys. B **32**, 113 (1999).
- [7] A. Rahman *et al.*, Phys. Scr. **67**, 414 (2003).
- [8] A. Rahman *et al.*, Phys. Scr. **70**, 21 (2004).
- [9] P. Beiersdorfer *et al.*, Phys. Rev. Lett. **67**, 2272 (1991).
- [10] E. Biémont, J. Phys. B **30**, 4207 (1997).
- [11] E. Träbert *et al.*, Phys. Rev. A **73**, 022508 (2006).
- [12] U. I. Safronova *et al.*, At. Data Nucl. Data Tables **92**, 47 (2006).
- [13] E. Träbert *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **235**, 23 (2005).
- [14] T. Brage *et al.*, Astrophys. J. **500**, 507 (1998).
- [15] H. Gould *et al.*, Phys. Rev. Lett. **33**, 676 (1974).
- [16] F.-A. Parpia, C. F. Fischer, and I. P. Grant, Comput. Phys. Commun. **94**, 249 (1996).
- [17] I. P. Grant *et al.*, Comput. Phys. Commun. **21**, 207 (1980).
- [18] T. Brage and C. F. Fischer, Phys. Scr., T **47**, 18 (1993).
- [19] J. Olsen *et al.*, J. Chem. Phys. **89**, 2185 (1988).
- [20] B. O. Roos *et al.*, Chem. Phys. **48**, 157 (1980).
- [21] I. Lindgren and J. Morrison, *Atomic Many-Body Theory*, Springer Series in Chemical Physics Vol. 13 (Springer-Verlag, Berlin, 1982).
- [22] P. Jönsson, Phys. Scr. **48**, 678 (1993).
- [23] C. F. Fischer, T. Brage, and P. Jönsson, *Computational Atomic Structure an MCHF Approach* (Institute of Physics Publishing, Bristol, 1997).
- [24] P. Jönsson, F. A. Parpia, and C. F. Fischer, Comput. Phys. Commun. **96**, 301 (1996).