

Accurate Values of Nuclear Magnetic Moments of Francium Isotopes

Mina Vajed-Samii,^(a) J. Andriessen,^(b) B. P. Das, S. N. Ray,^(c) Taesul Lee,^(d) and T. P. Das
Department of Physics, State University of New York, Albany, New York 12222

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A relativistic many-body perturbation-theoretic investigation has been carried out of the hyperfine field in the francium atom which when combined with recent measurements of hyperfine constants leads to $(3.39 \pm 0.09)\mu_N$ for the magnetic moment of the neutron magic isotope ^{213}Fr and equally accurate values for the other isotopes $^{208-212}\text{Fr}$.

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Among the alkali-atom nuclei, francium nuclei are the only ones for which magnetic moments are not available. This is because these nuclei including the natural isotope ^{223}Fr are unstable, making it difficult to carry out measurements of their nuclear-magnetic-resonance frequencies by conventional techniques. However, accurate measurements have recently become available¹ for the hyperfine constants of $^{208-213}\text{Fr}$ nuclei by laser spectroscopy techniques. In the present work, the hyperfine field at the nucleus in francium atom has been calculated to an accuracy of 0.5% with use of relativistic many-body perturbation theory² (RMBPT), which has been demonstrated to provide results of this accuracy in other alkali atoms.³⁻⁶ The combination of our calculated hyperfine field with the recently measured hyperfine constants has provided for the first time accurate values of the magnetic moments of francium isotopes, which should be valuable both for the testing of nuclear structure theories as well as for employing the francium nucleus as a probe for study of electronic properties of solid-state systems⁷ containing francium by means of hyperfine-interaction related data^{7,8} such as Knight shifts and relaxation times.

The relativistic many-body perturbation procedure that we have employed in our work on francium and also on the rest of the alkali atoms is described in detail in earlier papers,³⁻⁶ the calculations on lithium³ and sodium⁴ atoms, because of their lightness, having been carried out by the nonrelativistic version of the procedure. We shall only briefly point out here the main features of the relativistic method for the sake of completeness and to facilitate the discussion of our results.

Thus, the relativistic Hamiltonian for the atoms is given by^{9,10}

$$\mathcal{H} = \sum_i (c \vec{\alpha}_i \cdot \vec{p}_i + \beta_i m c^2) - \sum_i \frac{\xi_N e^2}{r_i} + \sum_{i>j} \frac{e^2}{r_{ij}} \quad (1)$$

α_i and β_i referring to the Dirac matrices of the i th electron, r_i its distance with respect to the nucleus, and r_{ij} the separation between the i th and j th electrons. The electron-nuclear magnetic hyperfine-interaction Hamiltonian \mathcal{H}_N' is given by⁵

$$\mathcal{H}_N' = \sum_i \vec{\alpha}_i \cdot \vec{\mu} \times \vec{r}_i / r_i^3. \quad (2)$$

If one could obtain the exact eigenfunction Ψ_0 of the total electronic Hamiltonian, \mathcal{H} , then the hyperfine field at the nucleus would be given by

$$H(0) = \mu^{-1} \langle \Psi_0 | \mathcal{H}_N' | \Psi_0 \rangle \quad (3)$$

which is independent of the magnitude μ of the nuclear moment. The hyperfine constant A measured conventionally in megahertz is related to $H(0)$ by the relation

$$A = [\mu H(0) / I J (2\pi\hbar)] \times 10^{-6}. \quad (4)$$

Thus, if A is measured experimentally and an accurate theoretical value of $H(0)$ is available, one can obtain the nuclear moment μ by use of Eq. (4).

The major problem in atomic theory is that the exact eigenfunction Ψ_0 cannot be obtained by directly solving the equation $\mathcal{H}\Psi_0 = E\Psi_0$. One has therefore to resort to alternate methods and in RMBPT,² we define first a neighboring Dirac Hartree-Fock type Hamiltonian:

$$\mathcal{H}_0 = \sum_i (c \vec{\alpha}_i \cdot \vec{p}_i + \beta_i m c^2) - \sum_i \left(\frac{\xi_N e^2}{r_i} - V_i \right), \quad (5)$$

where V_i is a one-electron potential for which it has been found convenient in many-body perturbation theory to choose the V^{N-1} approximation⁵ defined by

$$\langle a | V^{N-1} | b \rangle = \sum_{n=1}^{N-1} \left\{ \left\langle a n \left| \frac{e^2}{r_{ij}} \right| b n \right\rangle - \left\langle a n \left| \frac{e^2}{r_{ij}} \right| n b \right\rangle \right\}, \quad (6)$$

the sum of the conventional Coulomb and ex-

change potentials, the summation extending over all occupied states except the highest one, corresponding to the $7s_{1/2}$ valence state in the case of francium. The eigenfunction Ψ_0 is then derived from the eigenfunction Φ_0 of \mathcal{H}_0 by the linked-cluster expansion¹¹:

$$\Psi_0 = \sum_k^L [\mathcal{H}' / (E_0 - \mathcal{H}_0)]^k \Phi_0, \quad (7)$$

where E_0 is the ground-state eigenvalue of \mathcal{H}_0 and

$$\mathcal{H}' = (\mathcal{H} - \mathcal{H}_0) = \sum_{i>j} \frac{e^2}{r_{ij}} - \sum_i V_i^{N-1}. \quad (8)$$

With use of Eq. (3) and (7), the hyperfine field $H(0)$ is given by

$$H(0) = \frac{1}{\mu} \sum_{m,n}^L \left\langle \Phi_0 \left| \left(\frac{\mathcal{H}'}{E_0 - \mathcal{H}_0} \right)^m \mathcal{H}'_N \left(\frac{\mathcal{H}'}{E_0 - \mathcal{H}_0} \right)^n \right| \Phi_0 \right\rangle, \quad (9)$$

the L in the summation in (9) indicating that only terms represented by linked diagrams are to be included.

As has been discussed in our earlier work on hyperfine interactions in atoms²⁻⁶ studied by the many-body perturbation-theoretic procedure, the various diagrams corresponding to the terms in the perturbation expansion determined by the indices m and n in Eq. (9) are referred to as (m, n) order diagrams. Typical diagrams in alkali atoms that represent the various physical effects,⁵ direct, electron-core polarization (ECP), consistency, and correlation, are represented by the diagram in Fig. 1.

The diagram (a) in Fig. 1 represents the direct contribution⁵ which is of order (00) and its contribution to $H(0)$ is given by

$$H_{\text{direct}}(0) = \mu^{-1} \langle \varphi_{7s_{1/2}} | \mathcal{H}'_N | \varphi_{7s_{1/2}} \rangle. \quad (10)$$

Diagram (b) represents the ECP effect, the core states n , primarily the $s_{1/2}$ states (but with small contributions also from p states in relativistic theory) of same spin as the valence $7s_{1/2}$ electron getting perturbed by exchange interaction with the latter and contributing to the hyperfine field. The contribution from this diagram is given by

$$H_{\text{ECP}}(0) = \frac{2}{\mu} \sum_n \sum_k \frac{\langle \varphi_{7s_{1/2}} \varphi_n | (e^2/r_{ij}) | \varphi_k \varphi_{7s_{1/2}} \rangle \langle \varphi_k | \mathcal{H}'_N | \varphi_n \rangle}{(\epsilon_k - \epsilon_n)}, \quad (11)$$

where ϵ_n and φ_n are the one-electron energy and wave function for the core states n and ϵ_k and φ_k those for the unoccupied excited states, both bound and continuum.

Diagram (c) is a typical consistency diagram, of order (02). Physically it can be described as the contribution to the hyperfine field from a core state n' when the latter is perturbed by the exchange potential due to a core state n of the same spin which in turn has been perturbed by exchange interaction with the valence $7s_{1/2}$ electron. Its contribution is given by the equation

$$H_{\text{const}}(0) = \frac{2}{\mu} \sum_n \sum_{n'} \sum_k \sum_{k'} \frac{\langle \varphi_{7s_{1/2}} \varphi_n | (e^2/r_{ij}) | \varphi_k \varphi_{7s_{1/2}} \rangle \langle \varphi_k \varphi_{n'} | (e^2/r_{ij}) | \varphi_{k'} \varphi_n \rangle \langle \varphi_{k'} | \mathcal{H}'_N | \varphi_{n'} \rangle}{(\epsilon_k - \epsilon_n)(\epsilon_{k'} - \epsilon_{n'})}. \quad (12)$$

The diagram (d) is the direct counterpart of diagram (c). The diagrams (c) and (d) make the major contribution among the consistency diagrams, the consistency effect itself being rather weak, less

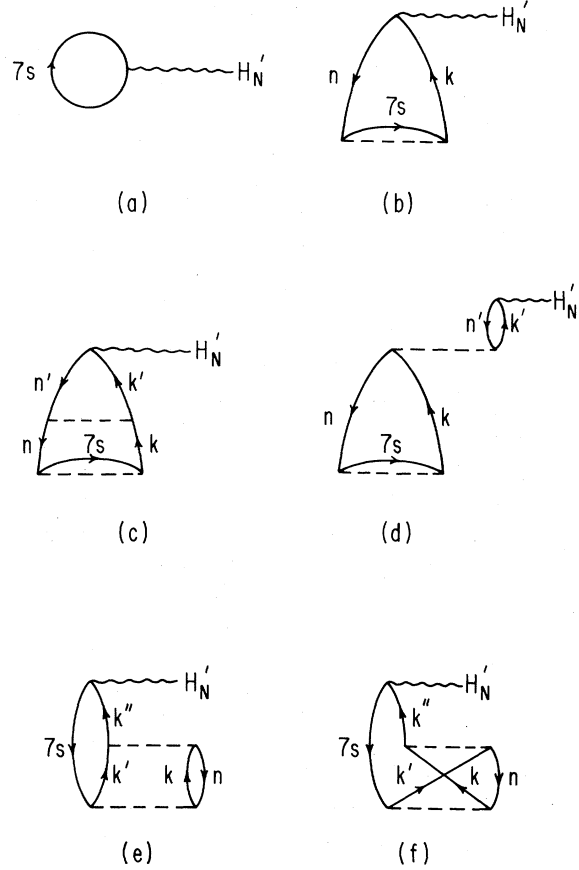


FIG. 1. Zero-, first-, and second-order diagrams contributing to hyperfine interactions in francium.

than 1% of the net hyperfine field.

Diagrams (e) and (f) represent the major correlation diagrams, (f) being the exchange counterpart of (e). Diagram (e) represents the influence of pair-correlation interaction between the valence electron and all core electrons, not just the s electrons, on the hyperfine field. In fact, the major correlation effect occurs from pair interaction between the valence electron and the outermost p shell electrons in all the alkali atoms except lithium which does not have p cores. The contribution from the diagram (e) is given by

$$H_{\text{corr}}(0) = \frac{2}{\mu} \sum_n \sum_k \sum_{k'} \sum_{k''} \frac{\langle \varphi_{7s\ 1/2} \varphi_n | (e^2/r_{ij}) | \varphi_{k'} \varphi_k \rangle \langle \varphi_k \varphi_{k'} | (e^2/r_{ij}) | \varphi_n \varphi_{k''} \rangle \langle \varphi_{k''} | \mathcal{H}_{N'} | \varphi_{7s\ 1/2} \rangle}{(\epsilon_{k''} - \epsilon_{7s})(\epsilon_{k'} + \epsilon_k - \epsilon_n - \epsilon_{7s})}. \quad (13)$$

We consider first the contributions from the various physical effects to the hyperfine field in francium before discussing the total hyperfine field needed for the evaluation of the evaluation of the magnetic moments of the francium isotopes. The direct contribution to the hyperfine field, $H_{\text{direct}}(0)$, in francium comes out as 5278 kOe. This is about 69% of the total hyperfine field, which is comparable to the corresponding ratios of direct³⁻⁶ and experimentally observed¹² hyperfine fields in the other alkali atoms, these ratios all lying between 65% and 70%. The ECP contribution $H_{\text{ECP}}(0)$ to the hyperfine field was found to be 891 kOe, with about 64% of this contribution arising from the outermost core shell $6s_{1/2}$ and the balance from the other core s shells, a feature in common with other alkali atoms.³⁻⁶ The net consistency contribution represented by diagrams (c) and (d) and related diagrams is rather small, relative to the direct and ECP contributions, only about 80 kOe, again a feature in common with the rest of the alkali atoms.³⁻⁶ The correlation contribution obtained by evaluating the diagrams (e) and (f), and other diagrams which were found to make nonnegligible contributions in rubidium and cesium, was found to be 1377 kOe. Of this, 1257 kOe, or about 91%, is contributed by the correlation diagram (e), with the state n being $6p$, corresponding to $6p-7s$ correlation. Of this $6p-7s$ correlation contribution, about 74%, that is 930 kOe, is contributed by the $6p-d$ mode of excitation in diagram (e), about 21%, that is 264 kOe, by the $6p-p$ excitation, the balance of 5% or 63 kOe arising from other modes of excitations of the $6p$ shell. Other correlation diagrams including the contributions to diagram (e) from $5d-7s$ and $4f-7s$ correlation and from diagram (f) not only make small contributions but also involve substantial mutual cancellations among themselves, leading to a net contribution of about 120 kOe.

On combining the contributions from all the mechanisms, the net hyperfine field in francium

comes out as 7628 kOe. In arriving at the confidence limit of this result, we have used our experience with other alkali atoms to ascribe a range of ± 50 kOe due to higher order diagrams³⁻⁵ beyond second order and another ± 150 kOe from considerations of computational accuracy and the use of a finite basis set of states for the continuum states in the complete set of states of the zero-order Hamiltonian involved in the perturbation analysis. Thus our net result for the hyperfine field is

$$H_{\text{tot}}(0) = 7628 \pm 200 \text{ kOe}. \quad (14)$$

We have utilized this value of the calculated hyperfine field together with the experimental value¹ of the magnetic hyperfine constant of 8744.9 ± 10.5 MHz for $A(^2S_{1/2})$ for ^{213}Fr and its nuclear spin of $I = \frac{9}{2}$ to derive its magnetic moment employing Eq. (4). The value we have derived this way is

$$\mu(^{213}\text{Fr}) = (3.39 \pm 0.09) \mu_N. \quad (15)$$

This value is about 80% of the value estimated recently¹ with use of results of g -factor measurements⁸ for $(h_{9/2})^n$ proton states of $N = 126$ nuclides and the assumption that g_I is independent of I and n , indicating that this assumption,¹ while reasonable for semiquantitative estimates, is not rigorously valid.

Using the experimental ratios¹ of the hyperfine constants between ^{213}Fr and the other isotopes $^{208-212}\text{Fr}$ for which they are available, their nuclear spins¹ as well as $I = \frac{9}{2}$ for ^{213}Fr , and the magnetic moment of ^{213}Fr in Eq. (15), we obtain $\mu(^{208}\text{Fr}) = (4.00 \pm 0.10) \mu_N$, $\mu(^{209}\text{Fr}) = (3.32 \pm 0.09) \mu_N$, $\mu(^{210}\text{Fr}) = (3.71 \pm 0.10) \mu_N$, $\mu(^{211}\text{Fr}) = (3.37 \pm 0.09) \mu_N$, and $\mu(^{212}\text{Fr}) = (3.90 \pm 0.10) \mu_N$.

Before concluding, we would like to point out that the results of our many-body investigations of francium indicate that the ratio of H_{corr} to H_{direct} is 0.28, significantly smaller than the ratio 0.38 observed in cesium, while the ratio

of H_{ECP} to H_{direct} appears to be nearly constant at 0.17 in both systems. These interesting trends as well as those with respect to the whole alkali-atom series will be discussed elsewhere.

^(a)Present address: Department of Physics, Johns Hopkins University, Baltimore, Md. 21218.

^(b)Permanent address: Technische Natuurkunde, Technische Hogeschool, Delft, Netherlands.

^(c)Present address: Systems and Applied Sciences Corporation, Riverdale, Md. 20737.

^(d)Present address: Computer Science Corporation, Silver Spring, Md. 20910.

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¹⁰For our investigations here, we have used a point charge for the nucleus. The departure from a point nucleus is important for the study of hyperfine-structure anomaly effects and can also significantly influence the absolute values of the nuclear magnetic moment derived from experimental hyperfine data. From our investigations on cesium atom [B. P. Das, Ph.D. thesis, State University of New York at Albany, 1981 (unpublished)], we expect the finite size of the cesium nucleus to influence the hyperfine field by about 0.1%. For francium, which is a heavier nucleus, the finite-size effect could be significantly larger.

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Excitation of the Positronium $1^3S_1 \rightarrow 2^3S_1$ Two-Photon Transition

Steven Chu and Allen P. Mills, Jr.

Bell Laboratories, Murray Hill, New Jersey 07974

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The $1^3S_1 \rightarrow 2^3S_1$ transition in positronium has been observed using two-photon Doppler-free excitation. Transitions were induced by a pulsed dye laser, and detected by photoionization of the 2^3S_1 state. The resonance signal has a linewidth of 1.5 GHz and a signal-to-noise ratio of 20:1. The frequency agrees with theory to 0.5 GHz. The resonance signal has an amplitude proportional to the square of the laser intensity and displays the expected Stark shift to higher frequency under an applied electric field.

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Positronium (Ps), first observed in 1951 by Deutsch,¹ is the purely leptonic atom consisting of an electron and its positron antiparticle. The atom provides a unique opportunity for studying a bound-state two-body system and the quantum electrodynamic (QED) corrections to that system. These corrections to the energy levels of Ps are of particular interest because they contain virtual annihilation terms not found in hydrogen or muonium. Furthermore, unlike these

atoms or hydrogenlike ions, the Dirac equation is not an adequate starting point for deriving the QED corrections in Ps, and the Bethe-Salpeter formalism² must be used. Unfortunately, the fully covariant Bethe-Salpeter equation has no known analytic solution and presents formidable calculational difficulties. Nevertheless, Ps is one of the few bound-state systems where the exact Hamiltonian is believed to be known to great accuracy, and precision experiments pro-