Hyperfine Quenching and Precision Measurement of the 2^3P_0-2^3P_1 Fine-Structure Splitting in Heliumlike Gadolinium (Gd^{62+})

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The hyperfine-quenched transition 2^3P_0-1^1S_0 has been observed in heliumlike gadolinium (Gd^{62+}) in the two isotopes ^{155}Gd and ^{157}Gd. The lifetime for the transition (τ_0) has been measured for each isotope and found to be $\tau_0(155)=13.43(27)\times10^{-12}$ sec and $\tau_0(157)=7.65(55)\times10^{-12}$ sec. From the measured lifetimes a value is inferred for the absolute value of the 2^3P_0-2^3P_1 fine-structuring splitting $|\Delta E_{0\text{-}1}|$ in Gd^{62+} with the result $|\Delta E_{0\text{-}1}|=18.57(19)$ eV, where the error represents 1 standard deviation. This result is compared with calculations based on the multiconfiguration Dirac-Fock method and on the unified method.

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The splittings of the 2^3P_J levels [1] in two-electron ions ($\Delta E_{J\text{-}J'}$) are determined largely by the electron-electron interaction. The measurement of these splittings constitutes, therefore, a test of theories of this interaction in the simplest atomic system in which it can be observed. Because of the rapid scaling of the relativistic part of this interaction with atomic number Z, measurements at high Z provide a particularly sensitive test of the relativistic theory. Calculations of the fine-structure splittings generally include the effects of relativity by a perturbation expansion in the parameter $(Z\alpha)^2$. The problems associated with this approach at high Z are illustrated by the fact that in ordinary helium ($Z=2$) the calculations [2] are accurate at the ppm level, whereas the most elaborate calculations [3,4] at $Z=64$ are accurate at only the few percent level. Accurate experimental measurements performed at high Z can therefore act to stimulate further theoretical work in including relativistic effects in the calculation of two-electron energy levels.

Accurate experimental measurements of the two-electron fine-structure splittings in the 2^3P_J multiplet have been performed in He and Li^+ using radio-frequency spectroscopy [5] and laser spectroscopy [6]. The extension of laser spectroscopy to the heliumlike ion F^{7+} was achieved by Myers *et al.* [7], who measured the interval 2^3P_1-2^3P_2 ($\Delta E_{1\text{-}2}$). However, extension of the laser method to much higher Z is experimentally not feasible. Indirect measurements using x-ray and UV spectroscopy have been performed and are summarized in review articles by Martin [8] and Desequelles [9], but such measurements have not been very precise. So far there have been no measurements reported with sufficient precision to examine the theoretical predictions at high Z. In this Letter we describe the first measurement at high Z with a precision smaller than the current theoretical accuracy.

The measurement reported here is of the interval $\Delta E_{0\text{-}1}$ in the two-electron ion Gd^{62+}. Gadolinium has a number of stable even-even isotopes with nuclear spin $I=0$. For these isotopes, the radiative decay 2^3P_0-1^1S_0 is strictly forbidden and the 2^3P_0 state undergoes $E1$ decay to the 2^3S_1 state with a calculated lifetime [10] of $\tau(2^3P_0)=0.339\times10^{-9}$ sec. Gadolinium also has two stable isotopes (^{155}Gd and ^{157}Gd) with nuclear spin $I=\frac{3}{2}$ and nuclear magnetic moments $\mu(155)=-0.2591\mu_N$ and $\mu(157)=-0.3397\mu_N$ [11]. These isotopes exhibit hyperfine coupling which causes a mixing of the 2^3P_0 and 2^3P_1 states. The 2^3P_1 state decays rapidly to the 1^1S_0 ground state with an unperturbed lifetime [10] $\tau(2^3P_1)=1.536\times10^{-16}$ sec. The mixing induced by the hyperfine coupling therefore causes the perturbed 2^3P_0 state to decay directly to the 1^1S_0 ground state. A transition induced in this way is referred to as a hyperfine-quenched transition. Such transitions are extremely rare in atomic physics [12] and have not been extensively studied. The possibility of using the hyperfine-quenched lifetime of the 2^3P_0 state to make precision measurements of $\Delta E_{0\text{-}1}$ was first demonstrated [13] in Ag^{45+}. This method was subsequently applied to Ni in a recent work [14].

When hyperfine structure is present the perturbed lifetime of the 2^3P_0 state can be obtained with high accuracy by solving

$$\begin{vmatrix} E_0+i\Gamma_0/2-\lambda & W_{10} \\ W_{10} & E_1+W_{11}+i\Gamma_1/2-\lambda \end{vmatrix}=0, \qquad (1)$$

where $W_{10}=\langle 2^3P_0|H_{\rm hfs}|2^3P_1\rangle$, $W_{11}=\langle 2^3P_1|H_{\rm hfs}|2^3P_1\rangle$, and E_0, Γ_0 (E_1, Γ_1) are the unperturbed energies and radiative widths of the 2^3P_0 (2^3P_1) levels, respectively, and $H_{\rm hfs}$ is the hyperfine structure Hamiltonian. The real and

TABLE I. Parameter values (in eV) for Eq. (1).

A	W_{10}	W_{11}	Γ_0	Γ_1
155	−0.06216	−0.03519	1.942×10^{-6}	4.286
157	−0.08150	−0.04615	1.942×10^{-6}	4.286

imaginary parts of λ_0 (λ_1) provide the perturbed energies and lifetimes of the 2^3P_0 (2^3P_1) levels. A fully relativistic calculation of the matrix elements has been made and the parameters Γ_0 and Γ_1 evaluated [10,11]. Values of these parameters associated with Gd^{62+} are given in Table I. Using these results with Eq. (1), a value of $|\Delta E_{0\text{-}1}|$ can be determined from the measured 2^3P_0 lifetimes.

This experiment was performed using the beam-foil time-of-flight technique [15] on the 35-MeV/A gadolinium beam provided by the GANIL accelerator (Caen, France). The extracted beam from the accelerator was passed through a 9.5-mg/cm^2 Be stripping foil. The beam emerging from this foil was measured to contain approximately 5.7% hydrogenlike gadolinium (Gd^{63+}). The Gd^{63+} beam was then separated from the other charge states present in the beam by a high-resolution magnetic spectrometer system (LISE). The pure Gd^{63+} beam emerging from LISE was then passed through a 422-μg/cm^2 carbon capture foil which produced the 2^3P_0 excited state of Gd^{62+} of interest. The 2^3P_0-1^1S_0 hyperfine-quenched transition from this state at 41.8 keV is observed downstream of the foil by two solid-state detectors mounted on opposite sides of the beam (see Fig. 1). One of the detectors is fixed and is used for normalization purposes. The second detector is movable and the time-of-flight data are obtained by measuring the ratio of counts in the movable detector to counts in the fixed detector as a function of the position of the movable detector. With this arrangement, we normalize directly to the ion population in the excited state of interest. Moreover, changes in the shape or state of the capture foil during the experiment do not produce any errors in the lifetime measurement.

The measurement of the beam velocity was performed in two steps using the alpha and LISE spectrometers. The LISE spectrometer was first calibrated against the highly precise alpha spectrometer using the gadolinium beam extracted from the accelerator but prior to stripping. The velocity of the unstripped beam was determined to be $\beta=0.2708$ for ^{155}Gd and $\beta=0.2671$ for ^{157}Gd. The 9.5-mg/cm^2 Be stripping foil was then placed in the beam in front of the LISE spectrometer. The energy loss in the stripping foil was measured to be 411 MeV for ^{155}Gd and 417 MeV for ^{157}Gd, in good agreement with an interpolation based on published tables [16]. The beam velocity after stripping was $\beta=0.2612$ for ^{155}Gd and $\beta=0.2574$ for ^{157}Gd. Using the same tables a semiempirical correction was made for the energy loss of the beam in the carbon capture foil with the result $\beta=0.2607$ for ^{155}Gd and $\beta=0.2569$ for ^{157}Gd.

A sample spectrum obtained with the movable detector is shown in Fig. 2. The large peak (41.8 keV) contains the counts from the decay of interest (2^3P_0-1^1S_0) and possible unresolved counts arising from cascades through the 2^3P_1 level. A smaller peak (at 42.6 keV) contains counts from the cascade decays 2^3P_2-1^1S_0 and 2^1P_1-1^1S_0. The raw data used to determine the lifetimes and the fine structure consist of similar spectra taken at many fixed-detector–movable-detector separations for each of the two isotopes $^{155}Gd^{62+}$ and $^{157}Gd^{62+}$.

Before forming the ratio of total integrated counts under the peak in the movable detector to the same in the fixed detector, it is necessary to subtract those counts due to background cascades. Background counts under the peak are determined from the background rate on the low-energy and high-energy sides of the peak and interpolating. The correction for cascade counts under the peak is based on the following. Previous work [17] shows that the decay of cascade peaks in time is linear on a log-log plot of counts versus times. Hence the decay in time is characterized by an exponent. It is also observed

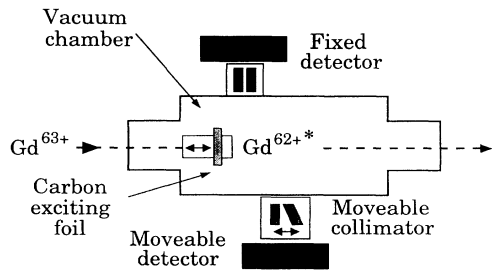

FIG. 1. Schematic design of the time-of-flight apparatus.

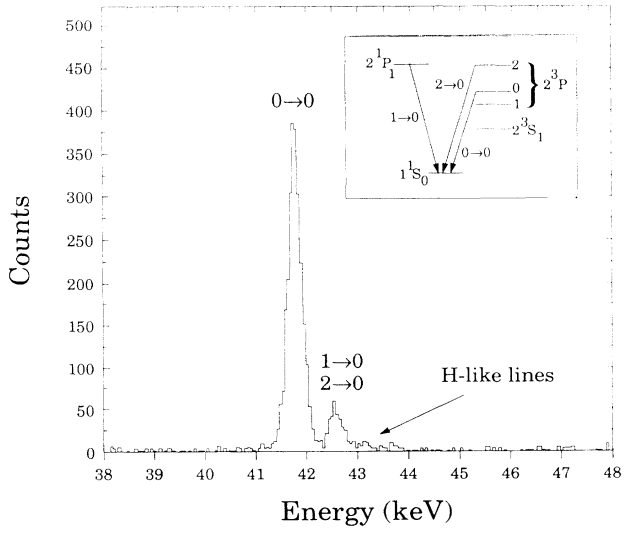

FIG. 2. Sample spectrum obtained by the movable detector downstream of the exciting foil.

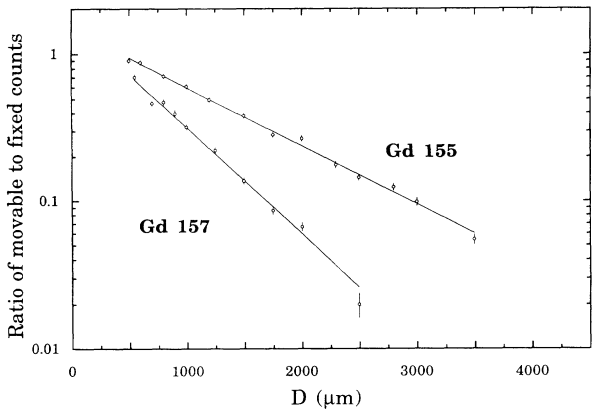

FIG. 3. Radiative decay of the 2^3P_0 state observed in each of the two isotopes ^{155}Gd and ^{157}Gd.

in work on heliumlike Xe^{52+} that cascade decays through the $(1s)(2p_{1/2})^3P_{0,1}$ and $(1s)(2p_{3/2})^3P_2, ^1P_1$ states are characterized by the same exponent to within the experimental error. Hence the number of cascade counts under the peak of interest can be taken to be a constant (C) multiplied by the number of counts in the cascade peak at 42.6 keV. The constant C will be independent of foil-detector separation. Moreover, since the cascade transitions (mainly along the yrast chain and from highly excited, long-lived s states) are not affected by hyperfine structure, we require that C be independent of isotope. The data for both isotopes were then fitted with a single value of C, which was chosen so as to minimize χ^2, with the result that $C=0.856$ and $\lambda(155)=1087(22)$ μm and $\lambda(157)=610(44)$ μm. In Fig. 3, the decay of the 2^3P_0 state is shown for each of the isotopes ^{155}Gd and ^{157}Gd. The different lifetimes for the decay of the same state is very evident. After correcting the measured decay lengths by the appropriate Lorentz contraction factors the lifetimes of the 2^3P_0 states are determined to be $\tau(155)=13.43(27)\times10^{-12}$ sec and $\tau(157)=7.65(55)\times10^{-12}$ sec. The errors given here represent 1σ and are limited mainly by the cascade correction. From the measured lifetimes it is possible to deduce values for $|\Delta E_{0\text{-}1}|=|E_1-E_0|$ by using Eq. (1). We obtain $|\Delta E_{0\text{-}1}(155)|=18.60(20)$ eV and $|\Delta E_{0\text{-}1}(157)|=18.23(67)$ eV. The agreement between the two values of $|\Delta E_{0\text{-}1}|$ for the two different isotopes within the experimental error is an important check on the consistency of the experiment and the theoretical interpretation of the data. Taking a weighted average of $|\Delta E_{0\text{-}1}|$ for the two isotopes yields our final value $|\Delta E_{0\text{-}1}|=18.57(19)$ eV. We note that this uncertainty is less than 5% of the natural linewidth of 4.29 eV for this transition.

There have been two widely different approaches used to calculate precise eigenvalues for heliumlike ions. The first of these is the unified method [3] which has as its starting point high-precision variational results which are used for the nonrelativistic eigenvalues and matrix elements of the Breit interaction. The second of these is the multiconfiguration Dirac-Fock (MCDF) method [4,18] in which the effects of relativity are built into the calculation at the starting point. In Table II a comparison is given between our experimental result and calculations based on the two methods. Drake has noted that the uncertainty in the calculations based on the unified method due to higher-order relativistic terms can be estimated [3] to be $1.2(Z/10)^4$ cm^{-1} which, at $Z=64$, is equal to 0.25 eV. Drake further noted, however, that the calculations should be somewhat more accurate for the 2^3P_J fine-structure splittings because of cancellations. Clearly, more theoretical and experimental work is called for.

TABLE II. Experimental results and comparison with theory (values in eV).

	Experiment (This work)	MCDF method (Ref. [10])	Unified method (Ref. [3])
$\lvert\Delta E_{0\text{-}1}\rvert$	18.57 ± 0.19	18.530	19.082

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[1] In this paper we use as a matter of convenience the L-S coupling notation 2^3P_1 to denote the state $(1s_{1/2})(2p_{1/2})$ $J=1$. Strictly speaking, the actual state in Gd^{62+} is much closer to the j-j coupling limit than it is to the L-S coupling limit.

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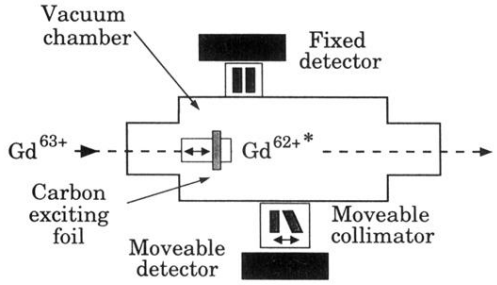

FIG. 1. Schematic design of the time-of-flight apparatus.