[a]On leave from Institute of Nuclear Physics, Cracow, Poland.
[b]Present address: Department of Physics, University of Jyväskylä, Finland.

[1]S. Landowne, C. H. Dasso, R. A. Broglia, and A. Winther, Phys. Lett. 70B, 292 (1977).
[2]P. Kleinheinz *et al.*, Z. Phys. A 290, 279 (1979).
[3]R. Broda *et al.*, Z. Phys. A 293, 135 (1979).
[4]P. J. Daly *et al.*, Z. Phys. A 298, 173 (1980).
[5]M. Luontama, J. Kantele, R. Julin, A. Passoja, T. Poikolainen, and M. Pyvänäinen, Nucl. Instrum. Methods 159, 339 (1979).
[6]P. Kleinheinz *et al.*, Z. Phys. A 286, 27 (1978).
[7]A. Bohr and B. Mottelson, *Nuclear Structure* (Benjamin, New York, 1975), Vol. II, p. 564 ff.

Effect of Laser Radiation on the Decay of ^{235m}U

G. C. Baldwin and S. A. Wender
Los Alamos National Laboratory, Los Alamos, New Mexico 87545
(Received 15 March 1982)

Optical excitation of an atom containing an isomeric nucleus should affect its internal conversion coefficient through modification of its bound-state electronic wave functions as well as by interaction with the outgoing electron. The extreme case of ^{235m}U, which decays entirely by internal conversion and for which pronounced environmental effects on the half-life have been observed, is considered. Resonant (bound-state) effects on the lifetime and electron emission spectrum are predicted to occur with moderate-power cw laser irradiation.

PACS numbers: 23.20.Nx, 32.90.+a

Although most nuclear processes are insensitive to environmental parameters, exceptions are known in which measurable changes in lifetimes and decay energies of radioactive nuclei are found. Chemical bonds, ionization, strong fields, etc. affect the interaction between the nucleus and the electronic structure of the atom or solid in which it is immersed.[1-3] We here suggest that tuned cw laser irradiation may give rise to a relatively large and easily observed effect in the isomer ^{235m}U.

Recently Becker *et al.*[4] have suggested that intense optical laser irradiation should affect decay rates and spectra of beta-emitting nuclei by modification of the density-of-states term for the ejected electron in the Fermi "golden-rule" formula.[5] That effect is more sensitive for nuclei that emit lower-energy electrons. When Becker *et al.*[4] estimated its magnitude for the beta emitter ^{3}H (end-point energy 18 keV), they found that an extremely intense, and therefore pulsed, laser would be needed for appreciable alteration of its decay rate. There are, moreover, grave doubts whether the predicted effect could be observed.[6]

Seeking other cases in which an excited nucleus decays by emission of electrons of much lower energy than in the case of ^{3}H, we note that the (73 ± 5)-eV isomer ^{235m}U decays with a 26-min half-life soley by emission of conversion electrons.[7-12] Interaction of these electrons with a laser field, in which one or more photons of the order of 1 eV are exchanged, should affect not only the density-of-final-continuum-states factor, but also the transition matrix element.

In the elementary theory of internal conversion with a point-nucleus approximation,[3] the transition probability, given by the Fermi "golden-rule" formula,[5] can be factored into a product of three terms: One is purely nuclear, one is purely electronic, and the third is the density of final states in phase space for the converted electron.

If the electronic configuration of the atom is changed (by chemical combination, implantation, ionization, or optical excitation), then the atomic factor in the transition probability is altered, and, with it, the probability for internal conversion. The conversion rate is greatest for those electronic configurations having greatest amplitudes in the vicinity of the nucleus; i.e., for s-electron orbitals. If, then, an s electron is removed by photoionization, or excited to a state of higher angular momentum, a particularly large change should occur in the decay constant for internal conversion. This expectation is confirmed[12] by experimental observation of changes in the lifetime, approaching 5% in extreme cases, accompanying changes in the chemical environment of ^{235m}U.

In addition to the effect of changed bound-state electron configuration, the effect discussed for beta decay by Becker *et al.*[4] should also exist; viz., a change of conversion probability when an electromagnetic field is present from which the electron can absorb, or into which it can emit, photons. One expects the final-state effect[4] to be insensitive to the precise wavelength of the laser, while the effect of perturbed bound states of the irradiated atom should show pronounced resonances, enabling the magnitudes of the two effects to be determined separately.

Quantitative prediction of the magnitude of an electron-configuration effect requires foreknowledge of the relative contributions of various electron orbits to the total conversion rate, which are not known experimentally. Calculating them requires knowledge of wave functions that are not accurately known for uranium. If they were, then the purely nuclear part of the transition probability could also be determined from the measured decay constants.

Grechukhin and Soldatov[13] have nevertheless attempted such an analysis. The electronic configuration of the free uranium atom,

$$(6s_{1/2})^2(6p_{1/2})^2(6p_{3/2})^2(6d_{3/2})^1(5f_{5/2})^3(7s_{1/2})^2,$$

was assumed to be perturbed by implantation into a medium,[12] altering the occupation numbers (viz., 3 in the $5f_{5/2}$ orbit). Using relativistic wave functions for the bound states, obtained by numerical integration of the Dirac equation with either Hartree-Fock-Slater or Thomas-Fermi-Dirac potential, they estimated the relative contributions of each orbital to the total conversion probability for the normal atom and for various assumed configurations of an environmentally modified atom. Adding, they found that changes in the decay rate approaching 10% are possible. However, they cautioned that this calculation depends upon uncertain input quantities, and that the conversion spectrum has yet to be resolved experimentally into the separate lines contributed by each orbital.

Taking Grechukhin and Soldatov's values[13] for the partial contributions, we estimate that excitation of one $7s_{1/2}$ electron to a p orbital should increase the conversion lifetime by approximately 1%.

Grechukhin and Soldatov employed asymptotic free-particle wave functions for the converted electron. According to Becker *et al.*,[4] the laser-perturbed problem must be analyzed with use of relativistic wave functions for an unbound electron in an electromagnetic field.[14]

An experiment involving selective modification of the electron configuration by tunable optical radiation should allow isolation of the partial contributions. In such an experiment, an isomer would be prepared and its half-life and conversion electron spectrum observed while it is irradiated with a laser that is tuned, successively, to various optical absorption lines.

The optical spectrum of the uranium atom is extremely complex.[15] According to Blaise and Radziemski[16] there are over 92 000 emission lines in the wavelength range between 310 and 900 nm; over 360 odd-parity levels and 1240 even-parity levels have been identified.

Although we can find no tabulations of the absorption spectrum, and no information concerning the optical linewidths of uranium deposits on a metallic substrate, we assume that the former include all observed emission lines that terminate on the ground state and that these lines, although they may be shifted, will not be unduly broadened. This second assumption is supported by observation[12] that surface deposits give consistent values of half-life and conversion spectrum, independent of the substrate material.

Table I combines information from several sources. Entries, arranged in order of increasing term value, include all transitions that terminate on the f^3ds^2 ground state and that are listed in Refs. 15, 16, or both. The remaining column headings are the electronic configurations given by Blaise and Radziemski.[16] The entries are Einstein A coefficients for each transition, in relative units, calculated by Maceda[17] from tabulated line intensities. For example, an observed line of wavelength 742.55 nm originates from an even-parity level at 13 463.392 cm^{-1} with electronic configuration f^3ps^2. The corresponding absorption transfers one electron from a d orbital to a p orbital. Similarly, the transition at 639.278 nm has the character $f \rightarrow d$, while that at 635.928 is an $s \rightarrow p$ transition. In the range of this table, there are nine transitions of this latter type, for which an effect on the internal conversion probability should be observable; three of these lines are quite strong.

Note also that many lines are to excited states of unknown electronic configuration. Observing the effect of saturating those levels on the internal conversion rate should assist in characterization of those states.

The intensity required to saturate an optical transition is found by solving the Einstein rate

TABLE I. A values for transition to the ground state from states of given electron configuration.[a]

λ[b], nm	ν[c] cm^{-1}	f^3dsp	$f^2d^2s^2$	f^3s^2p	??[d]
860.794	11613.917				
817.585					
742.550	13463.392			178	
682.693	14643.867	0642			
639.545	15631.855		457		
639.278	15638.367		143		
635.928	15720.682	146			
605.680	16505.773			97	
591.540	16900.387	1511			
575.814	17631.895				
532.926	18759.179				53
528.083	18932.767	73			
?	19477.861		14		
508.829	19647.507	259			
502.738	19885.515	2285			
?	20114.300	172			
?	20218.830				77
?	20420.517		57		
488.515	20464.523				116
?	20766.506		15		
?	20943.428				85
?	21078.730				18
?	21265.094				833
463.162	21584.695				3888
?	21636.957				8329
?	21766.53				33
?	21767.971	247			
?	22056.302				136
446.932	22368.467				1106
?	22377.764				4
?	22464.293				72
442.694	22582.654				146
439.360	22754.061				7285
437.276	22862.451				1558
436.206	22918.555	1011			
433.573	23057.676				2942
?	23325.189				12
426.632	23432.795				1433
424.626	23543.508				6995
?	23572.086				4749
419.194	23848.625				1587
415.397	24066.566				27210
413.350	24185.803				6465
410.836	24333.793				224
409.164	24433.260				3664
?	24560.410				58
?	24671.388				292
394.845	25319.274				1254

[a]The Einstein A values are relative, taken from Maceda, Ref. 17. To obtain absolute values, they should be multiplied by 1380 s^{-1}. All transitions for which Maceda lists an A value for transition to the $f^3d^1s^2$ ground state from an even-parity excited state are included, although not all correspond to lines in the National Bureau of Standards tabulation (Ref. 15).

[b]Wavelengths are from Meggers, Corliss, and Scribner, Ref. 15. A question mark in the wavelength column indicates that, although an A value is given in Ref. 7 for the term listed in Ref. 16, the spectral line is not listed in the National Bureau of Standards tabulation, Ref. 15.

[c]Electron configurations and term values are taken from Blaise and Radziemski, Ref. 16. The ground-state electron configuration is $f^3d^1s^2$, odd parity. This reference also lists isotope shifts; the values have not been corrected to ^{235m}U, for which there is also an (unknown) isomer shift.

[d]This column lists A values for transitions included by Maceda (Ref. 17) for which Ref. 16 did not assign an electron configuration.

equation

$$\dot{N}_2 = AN_2 + BU(N_2 - wN_1) - 0$$

for equilibrium with resonant radiation of energy density per unit bandwidth, U. The latter is related to the laser-beam intensity J by

$$U = J/c\Gamma,$$

where Q, the effective bandwidth, is assumed to be the Doppler width

$$\Gamma = (2/\lambda)(2\ln 2)^{1/2}(kT/M)^{1/2}.$$

Introducing the well-known ratio of the Einstein coefficients

$$B = A\lambda^3/8\pi h$$

and defining

$$x = N_2/(N_1 + N_2),$$

one obtains

$$J = \frac{1.07(11)}{(\lambda_{nm})^4}\,\frac{T^{1/2}}{M}\,\frac{x}{1-2x}.$$

M here is the atomic mass number, T is the absolute temperature, and we have set the statistical weight factor equal to unity.

To reach a saturation level of $x = 0.25$ at $\lambda = 417$ nm (worst case), $T = 300$ K, $M = 235$, we obtain 2 W cm^{-2}; going to 860 nm, we need only 0.11 W cm^{-2}. If the lines are broadened, the intensity needed will, of course, be higher.

The majority of previous workers[8,10-12] have prepared samples of ^{235m}U by depositing recoils from alpha decay of ^{239}Pu on a metallic surface and detected the conversion electrons after integral energy analysis by a retarding potential applied between the collector and a grid. Nêve de Mévergnies found that when an atmosphere of inert gas (argon) serves to slow down the recoils, they do not penetrate below the surface of the collector, and the measured half-life values are then independent of the substrate material.[12] If the collected deposit is irradiated by a tunable laser while the lifetime and electron spectrum are measured as functions of laser intensity and wavelength, then, at selected optical resonances of uranium, changes of the order of 1% in the

half-life can be expected, as in the case of implantation.[12]

The single-photon energy of visible radiation does not exceed the electron work functions of common substrates. The photoemissive properties of a uranium deposit should not be affected by the state of its nucleus. The background will therefore be related uniquely to the laser intensity and wavelength, and since its spectrum should not approach the 73-eV maximum energy of the conversion electrons, these low-energy electrons can be eliminated with a small retarding potential.

If the absorption lines should prove to be broadened excessively by interaction with the substrate, it would of course be necessary to devise an alternative technique in which the uranium is kept in gaseous form and the electron emission rate measured with and without optical irradiation.

This case offers an unusually sensitive test for influence of atomic structure on a nuclear property. The laser experiment should also lead to improved knowledge of both the nuclear and the atomic properties of ^{235m}U. In particular, better understanding of the details of the internal conversion process, and better values for lifetime and conversion electron spectrum, energy levels, and wave functions, would be obtained. Determining the atomic contribution to the conversion probability would permit the nuclear contribution to be isolated, with more confidence in the result than was possible in Ref. 13. Isomer shifts in the atomic levels and hyperfine splittings (if they can be resolved) would provide information as to the nuclear volume change and the magnetic and quadrupole moments of ^{235m}U. Finally, one will be in a better position to analyze the inverse internal conversion process[18-20] in ^{235m}U, about which there are still unresolved questions.[21,22]

We gratefully acknowledge helpful discussions with R. M. DeVries, P. Dyer, M. Moore, C. D. Olsen, and L. Radziemski.

[1]For example, F. Pleiter and H. DeWaard, in *Mössbauer Isomer Shifts*, edited by G. K. Shenoy and F. E. Wagner (North-Holland, Amsterdam, 1978), Chap. 5c.

[2]M. S. Freedman, Ann. Rev. Nucl. Sci. 24, 209–247 (1974).

[3]H. C. Pauli, K. Alder, and R. M. Steffen, in *The Electromagnetic Interaction in Nuclear Spectroscopy*, edited by W. D. Hamilton (North-Holland, Amsterdam, 1975), Chap. 10.

[4]W. Becker, W. H. Louisell, J. D. McCullen, and M. O. Scully, Phys. Rev. Lett. 47, 1262–1266 (1981).

[5]W. Heitler, *The Quantum Theory of Radiation* (Oxford Univ. Press, New York, 1944), 2nd ed., Sec. 9, Eq. (10).

[6]J. Gersten and M. H. Mittleman, Phys. Rev. Lett. 48, 651 (1982).

[7]*Table of Isotopes*, edited by C. M. Lederer and D. E. Shirley (Wiley Interscience, New York, 1978), 7th ed.

[8]F. Asaro and I. Perlman, Phys. Rev. 107, 318–319 (1957).

[9]J. R. Huizenga, C. L. Rao, and D. W. Englekemeir, Phys. Rev. 107, 319–320 (1957).

[10]M. S. Freedman, F. T. Porter, F. Wagner, and P. P. Day, Phys. Rev. 108, 836–841 (1957).

[11]H. Mazaki and S. Shimizu, Phys. Rev. 48, 1161 (1966).

[12]M. Nève de Mévergnies, Phys. Lett. 26B, 615–617 (1968), and Phys. Rev. Lett. 23, 422–425 (1969), and Phys. Lett. 32B, 482–484 (1970).

[13]D. P. Grechukhin and A. A. Soldatov, Yad. Fiz. 23, 273–282 (1976), and 28, 1206–1222 (1978) [Sov. J. Nucl. Phys. 23, 143–147 (1976), and 28, 622–630 (1979)].

[14]David Liberman (private communication) has proposed a closely related theoretical study of photoabsorption rates, using Thomas-Fermi-Dirac wave functions.

[15]W. F. Meggers, C. H. Corliss, and B. F. Scribner, *Tables of Spectral Line Intensities*, National Bureau of Standards Monograph 32 (U.S. Government Printing Office, Washington, D. C., 1961), Part 1.

[16]J. Blaise and L. Radziemski, J. Opt. Soc. Am. 68, 649–658 (1976).

[17]E. Maceda, Ph.D. thesis, University of Illinois, 1977 (unpublished).

[18]M. Morita, Prog. Theor. Phys. 49, 1574 (1973).

[19]V. I. Gol'danskii and V. A. Namiot, Phys. Lett. 62B, 393 (1976).

[20]K. Okamoto, Laser Interact. Relat. Plasma Phenom. 4A, 283–303 (1977).

[21]Y. Izawa and C. Yamanaka, Phys. Lett. 88B, 59–61 (1979).

[22]V. I. Gol'danskii, R. N. Kuz'min, and V. A. Namiot, Yad. Fiz. 33, 319 (1981) [Sov. J. Nucl. Phys. 33, 169 (1981)], and in Proceedings of LASER-81 International Conference (to be published), paper No. 0-1.