†Work supported by the U. S. National Science Foundation and the U. S. Energy Research and Development Administration.

[1]H. D. Jones, N. Onishi, T. Hess, and R. K. Sheline, Phys. Rev. C 3, 529 (1971).

[2]T. L. Khoo, J. C. Waddington, R. A. O'Neil, Z. Preibisz, D. G. Burke, and M. W. Johns, Phys. Rev. Lett. 28, 1717 (1972); T. L. Khoo, J. C. Waddington, and M. W. Johns, Can. J. Phys. 51, 2307 (1973).

[3]R. G. Helmer and C. W. Reich, Nucl. Phys. A114, 649 (1968).

[4]H. Massmann, J. O. Rasmussen, T. E. Ward, P. E. Haustein, and F. M. Bernthal, Phys. Rev. C 9, 2312 (1974).

[5]T. L. Khoo, F. M. Bernthal, R. G. H. Robertson, and R. A. Warner, to be published.

[6]The decay of a $K^{\pi}=(16^{+})$ isomer was previously observed (Ref. 3) in ^{178}Hf, but its exact location was not determined.

[7]T. L. Khoo, G. F. Bertsch, and G. Hamilton, to be published.

[8]G. F. Bertsch, *The Practioner's Shell Model* (North-Holland, Amsterdam, 1972).

[9]N. D. Newby, Phys. Rev. 125, 2063 (1962).

L_3 Conversion of the 2.38-keV Isomer in ^{90}Nb

J. L. Campbell* and B. Martin
Max Planck Institut für Kernphysik, Heidelberg, Germany

and

W. D. Schmidt-Ott and F. Smend
II Physikalisches Institut, Universität Göttingen, Göttingen, Germany

and

U. Raff
Institut fur Physik, Universität Basel, Basel, Switzerland

(Received 2 September 1975)

The 3.8% decrease in half-life of the 2.38-keV ^{90m}Nb isomer when Nb is converted from metal to a fluoride complex is one of the largest reported chemical effects on the internal conversion process. We present experimental and theoretical evidence that this effect is not due, as had been conjectured, to the "switching off" of L_3 conversion through increased L_3 binding energy.

Perturbations of nuclear decay rates (λ) due to changes in chemical environment[1] are typically 0.01–0.1%; for low-energy isomers the effects are attributed to altered valence-electron population and to the related change in electron density at the nucleus. The very large $\Delta\lambda$ of 3.8% observed[2,3] when the environment of ^{90m}Nb is changed from metallic Nb to a fluoride complex cannot be understood on this basis. This effect was seen by perturbing the decay chain ^{90}Mo(5.7 h) $\rightarrow$ ^{90m}Nb(18.8 sec) $\rightarrow$ ^{90g}Nb(14.6 h) through dissolution of activated Nb foils in acid, and a 1.9% effect was seen similarly for Nb/Nb_2O_5. However various workers[4-6] have prepared the 18.8-sec isomer directly and find no significant differences in half-lives measured for various chemical states; there exists no such measurement for the pair Nb/Nb(fluoride complex), but for the pair Zr/Zr(fluoride complex) the reported[6] upper limit on $\Delta\lambda$ is 0.18%.

The transition energy $E_\gamma = 2.38 \pm 0.36$ keV inferred[2] from energy balance of γ rays in ^{90}Mo decay has a large uncertainty and may lie above or below the L_3 binding energy of Nb. Olin[3] suggested that if in the metal $E(L_3) < E_\gamma$, and in the fluoride complex $E(L_3) > E_\gamma$, a large change in λ would result from the "switching off" of L_3 conversion. Smend, Borchert, and Langhoff[8] evaluated theoretical conversion coefficients for $E_\gamma = 2.38$ keV, and found that α_{L_3} contributed 3.8% of the total decay rate; they used the threshold value of α_{L_3} given by O'Connell and Carroll.[7]

In an attempt to clarify this situation we have performed measurements to determine whether or not L_3 conversion occurs in the free Nb atom. For comparison with theory we have computed α_{L_3} to within 10 eV of threshold.

The principle of our experiment is to determine the intensity ratio L/K of L and K vacancies created in ^{90m}Nb decay (Fig. 1), through measurement of the LX/KX x-ray intensity ratio. However at $Z \sim 40$, L-shell fluorescence yields

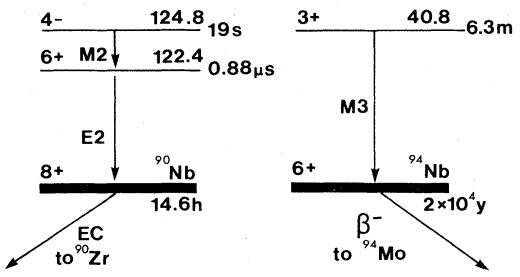

FIG. 1. Decay schemes of ^{90m}Nb and ^{94m}Nb.

are poorly known (50% difference between two main calculations whose results are tabulated in the review of Bambynek *et al.*[8]) and absolute efficiency calibration of a Si(Li) detector for 2-keV x rays involves large errors.[9,10] Therefore the *LX/KX* ratio is not measured absolutely but relative to the ratio for ^{94m}Nb, where the vacancy population created by the 40.8-keV *M*3 transition is well known. Efficiencies and *K* and *L* fluorescence yields cancel. The different subshell vacancy distributions matter little since the individual yields ν_1, ν_2, ν_3 are approximately equal to another in both the theoretical treatments mentioned.

For ^{90m}Nb, we have

$$\left(\frac{LX}{KX}\right)_{90} = \left[\left(\frac{\alpha_L + \alpha_K N_{KL}}{1+\alpha_T}\right)_{122} + \left(\frac{\alpha_{L_3}}{1+\alpha_T}\right)_{2.4}\right] \times \left[\left(\frac{\alpha_K}{1+\alpha_T}\right)_{122}\right]^{-1} \times C \qquad (1)$$

and for ^{94m}Nb

$$\left(\frac{LX}{KX}\right)_{94} = \left(\frac{\alpha_L + \alpha_K N_{KL}}{\alpha_K}\right)_{41} \times C. \qquad (2)$$

Using these, the numerical relation

$$R = \frac{(LX/KX)_{90m\,\mathrm{Nb}}}{(LX/KX)_{94m\,\mathrm{Nb}}} = 0.728 + 2\left(\frac{\alpha_{L_3}}{\alpha_T}\right)_{2.4} \qquad (3)$$

between the measured *LX/KX* ratios and the conversion coefficients of the 2.38-keV isomeric transition is obtained.

^{90m}Nb was produced by the $(d, 2n)$ reaction on Zr. A 20-nA beam of 19-MeV deuterons from the Göttingen synchrocyclotron passed through a stack of ten 0.02-mm Zr foils. Ethylene gas swept recoiling ^{90m}Nb atoms rapidly to a counting chamber where a gas jet[12] impinged on one of six positions on an aluminum wheel. During deposition of one source spot, the previous one was viewed with a Kevex Si(Li) x-ray detector of resolution 175 eV at 5.9 keV. The spots were almost invisible and the method ensured minimal self-absorption of 2 keV *L* x rays, since each essentially massless spot[12] was deposited on top of the previous one. Since the Nb atoms on the spots are embedded in organic material we are inclined to regard them as free atoms.

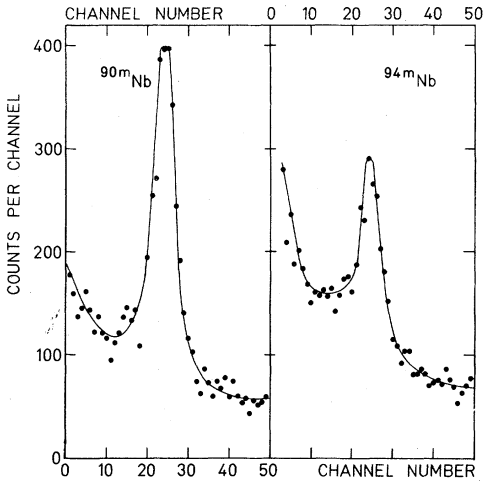

FIG. 2. *L* x-ray spectra recorded in the Si(Li) detector from jet-deposited sources of Nb isomers.

X-ray spectra from ^{90m}Nb were recorded using 30-sec deposition and counting periods. After each run the deuteron beam was switched off and the spectrum from accumulated 6.3-min ^{94m}Nb recorded for correction purposes. In the main measurement on ^{94m}Nb a 10-min period was used, after a 3-min interval to allow ^{90m}Nb to decay to a negligible level. The total deposition times were approximately equal for the two isomers. Typical *L* x-ray spectra are shown in Fig. 2.

The ^{94m}Nb spectra contained Zr x rays resulting from the 14.6-h decay of the ^{90g}Nb remaining after decay of ^{90m}Nb. The Zr *K* x rays were well resolved from the Nb *K* x rays of interest, but the Zr *L* x-ray peak was not separated from the Nb *L* peak. Using the ^{90g}Nb decay scheme[13] the theoretical Zr (*LX/KX*) intensity ratio was calculated in similar fashion to that of ^{94m}Nb (see above). With allowance for the slight difference in efficiencies and fluorescence yields between $Z = 40$ and 41, one finds

$$S = \frac{(LX/KX)_{90g\,\mathrm{Nb}}}{(LX/KX)_{94m\,\mathrm{Nb}}} = 0.59 \qquad (4)$$

for the theoretical ratio of the two (*LX/KX*) values. Then the denominator required for Eq. (3)

is obtained from

$$\left(\frac{LX}{KX}\right)_{94m} = \frac{L}{SK_{Zr} + K_{Nb}}, \quad (5)$$

where L, K_{Zr}, and K_{Nb} are measured x-ray intensities. The value obtained for this experimental ratio was 0.048 ± 0.003. The corresponding result for spectra from ^{90g}Nb decay is 0.028.

The ^{90m}Nb spectra contain Zr and Nb x-rays from both ^{90m}Nb and ^{94m}Nb decays. The K x-ray contribution from ^{94m}Nb was obtained by switching off the beam after a series of runs totaling 90 min duration, recording the ^{94m}Nb spectrum, and calculating from this the equilibrium intensity of K x rays from ^{94m}Nb during the main run. The undesired L x-ray contributions were obtained by multiplying the Zr and the ^{94m}Nb K x-ray intensities by 0.028 and 0.048, respectively. The resulting numerator for Eq. (3) is 0.094 ± 0.009.

The final result is $R = 1.96 \pm 0.2$, from which we get

$$\alpha_{L_3}/\alpha = 0.61 \pm 0.10.$$

The error is purely statistical, representing the spread of results in three ^{94m}Nb and four ^{90m}Nb experiments.

Subsidiary measurements were made of the K conversion coefficients of the 122.37-keV $E2$ and the 40.8-keV $M3$ transitions. The KX/γ ratios were measured with a 10-mm-diam Ge(Li) low-energy photon spectrometer and corrected by the K fluorescence yield[8] to give α_K. In the ^{90m}Nb case the photon spectra from jet-deposited sources was recorded. Absolute efficiencies were determined with ^{241}Am and ^{57}Co intensity standards supplied by the Physikalisch-Technische-Bundesanstalt in Braunschweig. Data on photons per decay for these standards were taken, respectively, from Campbell and McNelles[9] and from Hansen *et al.*[10]

For ^{94m}Nb, activated Zr foils of 0.002 cm thickness were transferred manually from irradiation chamber to detector. Calibration here was by a vacuum-evaporated ^{241}Am source which was later calibrated against Laboratoire de Métrologie des Rayonnements Ionisants and International Atomic Energy Agency primary standards. The detector was collimated to ensure essentially constant efficiency from 26 to 60 keV, these being the two highest energy points measured. Both results are compared with predictions from the new program CATAR[11] in Table I; the good agreement justifies our earlier use of theoretical values to obtain R_1 and R_2. The uncertainties could be much reduced by more rigorous efficiency calibration[9] and improved statistics, but that diversionary effort was not thought to be necessary in the context of the main experiment.

The theoretical value of α_{L_3}/α_T for the 2.38-keV $M2$ transition was computed as a function of E_γ with CATAR. One motivation for CATAR was the need for more accurate calculations at energies within a few keV of threshold. The numerical accuracy achieved is 2% at 10 eV above threshold. The ratio is essentially independent of E_γ over the energy range 2.38 to 2.8 keV, its value being

$$\alpha_{L_3}/\alpha_T = 0.66, \quad E_\gamma > E(L_3),$$
$$= 0, \quad E_\gamma < E(L_3).$$

Thus the ratio does not provide a determination of E_γ but comparison of experimental and theoretical ratios shows clearly that L_3 conversion takes place. Should the transition energy be within 10 eV of threshold, the result provides no evidence for a precipitous drop in α_{L_3} as would be indicated by Ref. 7.

Turning to the reported 3.8% change[2,3] in decay rate of ^{90m}Nb between metal lattice and fluoride complex environments, it appears that this is not to be explained by "switching off" L_3 conversion in the fluoride complex since that would produce a 66% change. The explanation of the 3.8% effect may be that in the Nb^{n+} ion, $E(L_3)$ increases such that 3.8% of the area under the Lorentzian shape of the L_3 level profile (level width ~2 eV) lies above E_γ. Relativistic Hartree-Fock-Slater calculations yield 33 and 70 eV for the shifts in

TABLE I. K conversion coefficients.

Nucleus	Energy (keV)	Multipolarity	α_K Theory	α_K Measured
^{90}Nb	122.37	$E2$	0.465	0.45 ± 0.03
^{94}Nb	40.8 ± 0.1	$M3$	771 ± 8	790 ± 70

$E(L_3)$ from Nb to Nb^{3+} and Nb^{5+} respectively.

The M, N, and O shells do not appear to be responsible for the 3.8% effect. We computed relativistic Hartree-Fock-Slater potentials for the configurations Nb, Nb^{1+}, Nb^{3+}, Nb^{5+}, and used these to generate α_{MNO}. The results are only approximate since eigenvalues were used in place of the experimental binding energies, but they do indicate that changes in α_{MNO} contribute less than 0.5% to $\Delta\lambda$.

In addition the half-life of the 122-keV radiation of ^{90m}Nb on the spots was measured yielding $T_{1/2} = 18.6 \pm 0.5$ sec. This value agrees well with the earlier result[5,6] $T_{1/2} = 18.8$ sec for ^{90m}Nb in several chemical environments. Thus even "free" Nb atoms show within the limits of error the same $T_{1/2}$ as atoms bound in metals or molecules indicating that internal conversion takes place in the L_3 and higher shells.

In conclusion this work appears to eliminate the intriguing possibility that ^{90m}Nb is a unique case where conversion in a specific inner subshell could be switched on and off by changes in chemical bonding. Only a steep drop in α_{L_3} within 10 eV of threshold would permit this, and we find no evidence for such a drop. The inconsistencies among direct and indirect measurements[1-6] of lifetime changes remain unexplained and further work in that area appears necessary. It should at least be possible to repeat the indirect measurement by Cooper, Hollander, and Rasmussen[2] of transition energy with about ± 12 eV precision. Finally the present theoretical calculation of conversion coefficients appears to be much more successful in the energy region close to threshold than its predecessors.[14]

We thank Professor A. Flammersfeld and Professor U. Schmidt-Rohr for their support and Professor H. M. Weiss for supplying standards. One of us (J.L.C.) thanks the Canadian National Research Council for a travel grant.

*On leave from University of Guelph, Guelph, Ontario, Canada.

[1]G. T. Emery, Annu. Rev. Nucl. Sci. 22, 165 (1972).

[2]J. A. Cooper, J. M. Hollander, and J. O. Rasmussen, Phys. Rev. Lett. 15, 680 (1965), and Nucl. Phys. A109, 603 (1968).

[3]A. Olin, Phys. Rev. C 1, 1114 (1970).

[4]W. Weirauch, W. D. Schmidt-Ott, F. Smend, and A. Flammersfeld, Z. Phys. 209, 289 (1968).

[5]J. S. Geiger, R. L. Graham, and M. W. Johns, Can. J. Phys. 47, 949 (1969).

[6]F. Smend, I. Borchert, and H. Langhoff, Z. Phys. 248, 326 (1971).

[7]R. F. O'Connell and C. O. Carroll, Phys. Rev. 138, B1042 (1965).

[8]W. Bambynek, B. Crasemann, R. W. Fink, H. U. Freund, H. Mark, C. D. Swift, R. E. Price, and P. V. Rao, Rev. Mod. Phys. 44, 716 (1972).

[9]J. L. Campbell and L. A. McNelles, Nucl. Instrum. Methods 125, 205 (1975).

[10]J. S. Hansen, J. C. McGeorge, D. Nix, W. D. Schmidt-Ott, I. Unus, and R. W. Fink, Nucl. Instrum. Methods 106, 365 (1973).

[11]H. C. Pauli and U. Raff, "A computer program for internal conversion coefficients and particle parameters" (to be published).

[12]W. D. Schmidt-Ott, to be published.

[13]J. B. Ball, M. W. Johns, and K. Way, Nucl. Data Sect. A 8, 407 (1970).

[14]J. L. Campbell and B. Martin, Z. Phys. A274, 9 (1975).

Hydrogen 1S-2S Isotope Shift and 1S Lamb Shift Measured by Laser Spectroscopy*

S. A. Lee, R. Wallenstein,† and T. W. Hänsch‡
Department of Physics, Stanford University, Stanford, California 94305
(Received 22 September 1975)

We measured the isotope shift of the 1S-2S transition in atomic hydrogen and deuterium to be 670.933 ± 0.056 GHz, using Doppler-free two-photon spectroscopy with a frequency-doubled, pulsed, dye laser. The Balmer-β line is simultaneously observed in saturated absorption with the fundamental dye-laser output. A comparison of the intervals 1S-2S and $2P_{3/2}$-$4D_{5/2}$ determines the Lamb shift of the 1S ground state to be 8.20 ± 0.10 GHz (H) and 8.25 ± 0.11 GHz (D), in agreement with the theoretical values.

We report on the first accurate measurement of the Lyman-α isotope shift for atomic hydrogen and deuterium, using Doppler-free two-photon spectroscopy of the 1S-2S transition with a frequency-doubled, pulsed, dye laser near 2430 Å.[1] The fundamental dye-laser output near 4860 Å allowed the simultaneous observation of the Balmer-β line by high-resolution saturation spec-