Measurement of the half-life of ^{198}Au in a nonmetal: High-precision measurement shows no host-material dependence

J. R. Goodwin, N. Nica, V. E. Iacob, A. Dibidad,* and J. C. Hardy†
Cyclotron Institute, Texas A&M University, College Station, Texas 77843, USA
(Received 24 August 2010; published 29 October 2010)

We have measured the half-life of the β^- decay of ^{198}Au to be 2.6948(9) d, with the nuclide sited in an insulating environment. Comparing this result with the half-life we measured previously with a metallic environment, we find the half-lives in both environments to be the same within 0.04%, thus contradicting a prediction that screening from a "plasma" of quasifree electrons in a metal increases the half-life by as much as 7%.

DOI: 10.1103/PhysRevC.82.044320 PACS number(s): 21.10.Tg, 23.40.−s, 27.80.+w

I. INTRODUCTION

This experiment was undertaken to investigate if the half-life of the β^- decay of ^{198}Au depends on whether the decaying nucleus is located in a metallic or an insulating environment. The "Debye plasma model," which was originally invoked [1] to explain observed cross-section anomalies in the $d(d,p)t$ reaction, was later applied to radioactive decays by Limata *et al.* [2]. According to this model, the conduction electrons present in a metal comprise a sort of plasma, which is referred to as a Debye plasma. It has been argued that this plasma changes the phase space available for radioactive decay and increases (for β^- or electron-capture decay) or decreases (for β^+ decay) the nuclide's half-life. If this model were correct, this change in phase space would occur only in metals—not in insulators—and would be enhanced if the metal were to be cooled to very low temperatures.

In their subsequent study of the β^- decay of ^{198}Au sited in a pure-gold host material, Spillane *et al.* [3] claimed to have observed both these effects, albeit to a lesser extent than the theory predicted. The theory predicts that at room temperature the half-life of ^{198}Au sited in a metal should be 7% longer than it is in an insulator, while at 12 K the difference should increase even further to 32%. The corresponding measured numbers as reported by Spillane *et al.*, were 0.4(7)% and 4.0(7)%. We repeated their measurement in a metal at two different temperatures and have already reported [4] that any temperature dependence must be less than 0.04%, two orders of magnitude below the value claimed by Spillane *et al.* However, we have not yet addressed the possibility that there might be a difference between a ^{198}Au source distributed in a metal and one in an insulator. We do so now by reporting a measurement of the ^{198}Au half-life, for which the decaying nuclei were sited in Au_2O_3.

Both measurements of the half-life of ^{198}Au in gold metal—ours [4] and that of Spillane *et al.* [3]—were performed with sources prepared by neutron activation of natural gold, ^{197}Au. To obtain comparable conditions and statistics for our measurement in a nonmetal, we wished to use neutron activation again and sought a suitable gold compound that is also an insulator. Although strictly speaking it is not an insulator, we did identify Au_2O_3—gold (III) oxide—as a suitable candidate. It is considered to be a semiconductor [5] but, with a calculated band gap higher than 0.85 eV, it should behave like an insulator at room temperature. In fact, it does: Its room-temperature resistivity has been measured to be at least five orders of magnitude higher than that of pure gold [6], undoubtedly sufficient to ensure the absence of a conduction-electron plasma.

II. APPARATUS AND SETUP

Gold has two important advantages for precise half-life measurements: It is monoisotopic (^{197}Au), so neutron activation produces only ^{198}Au, and its decay spectrum is dominated by a single strong β-delayed γ ray at 412 keV. No corrections are required for contaminant activities and the peak-to-background ratio is very high. Although we used Au_2O_3 as the material to be activated in this experiment, all other aspects of the measurement were identical to those of our previous experiment [4], in which we activated pure gold. We can thus directly compare the ^{198}Au half-lives measured at room-temperature with two different host materials, one an insulator and the other a conductor.

We used a gold (III) oxide sample obtained from the Alfa Aesar Corporation. It was in the form of powder with a purity of 99.99%. A 170-mg quantity of this powder was held onto an aluminum disk by adhesive Mylar tape, 56 μm thick, and the assembly was activated in a flux of $\sim 10^{10}$ neutrons/cm^2 s for 10 s at the Texas A&M Triga reactor. The irradiated Au_2O_3 sample was then fastened on the cold head of a CryoTorr 7 cryopump, precisely as had been done previously for our pure gold measurement [4]. Although we did not cool the Au_2O_3 sample to a low temperature in this measurement, for consistency we nevertheless followed the same procedure as in the previous measurement, including the use of the cryopump as a location for our sample.

A 70% high-purity germanium detector was placed directly facing the sample on the cryopump axis just outside the pump's cover plate, into which a cavity had been bored so that only 3.5 mm of stainless steel remained between the sample

*Research Experience for Undergraduates summer student from Florida A&M University, Tallahassee, FL 32307, USA.
†hardy@comp.tamu.edu

and the face of the detector. This arrangement was not altered in any way throughout the decay measurement. Sequential 6-h γ-ray spectra were acquired and recorded for a total period of 27 days—10 half-lives of ^{198}Au. The detector signals were amplified and sent to an analog-to-digital converter, which was an Ortec TRUMP-8k/2k card [7] controlled by Maestro software, which was installed on a PC operating under WINDOWS-XP.

During the entire period of the measurements, our computer clock was synchronized daily against the signal broadcast by WWVB, the radio station operated by the US National Institute of Standards and Technology. The TRUMP card uses the Gedcke-Hale method [8] to correct for dead-time losses, so by keeping our system's dead time below about 3% and recording all our spectra for an identical preset live time, we ensured that our results were nearly independent of dead-time losses. However, to achieve a precision better than 0.1%, a further small correction is required to account for residual rate-dependent effects such as pulse pileup. As described in Ref. [4], we have experimentally determined the fractional residual loss for our system to be $5.5(2.5) \times 10^{-4}$ per 1% increase in dead time. We applied this correction to the present results as we also did for the measurement to which this one is being compared: the half-life of ^{198}Au in gold metal.

III. ANALYSIS AND RESULTS

We analyzed the 412-keV γ-ray peak in each of the recorded spectra by using the least-square peak-fitting program GF3 from the RADware series [9]. Use of this program allowed us to be very specific in determining the correct background for a peak, and we visually inspected the peak of interest in each spectrum to ensure that the background was handled satisfactorily. So far as possible, the same criteria were applied to each of the 107 recorded spectra. The peak areas thus obtained for the 412-keV peak were then corrected for residual losses as described in Sec. II. The results are plotted as a function of time in Fig. 1.

The decay curve was then analyzed by a maximum-likelihood fit with a single exponential. The code we used, which is based on ROOT [10], has previously been tested to a precision of 0.01% with Monte Carlo-generated data. The result of the fit for the gold oxide half-life measurement is shown in Fig. 1, where the fitted decay curve is compared with the data in the top panel and the normalized residuals are plotted in the bottom panel. The ^{198}Au half-life obtained from this fit (with statistical uncertainty only) is 2.6948(9) d. The corresponding normalized χ^2 is 0.74, which gives a confidence level of 99%.

The equivalent room-temperature result for the ^{198}Au half-life, as measured in a pure-gold host material, was reported by us [4] to be 2.6949(5) d. The difference between these two results is 0.0001(10) d or 0.004(38)%. Both measurements were made under the same conditions and the data from both have been corrected for residual losses; however, the uncertainty in that correction has not been applied because it is correlated for the two measurements and does not contribute to the difference between them.

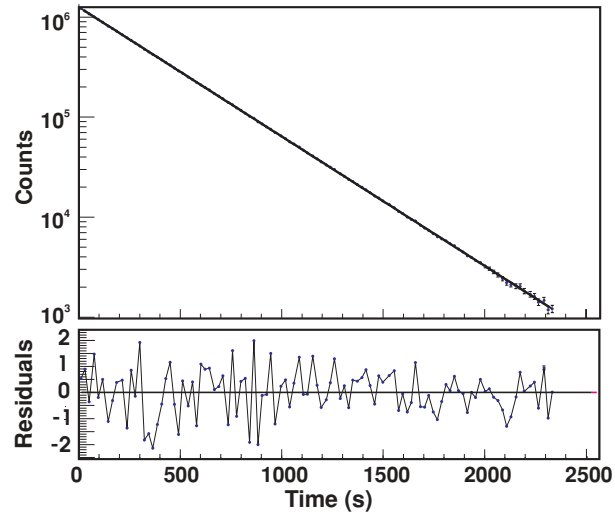

FIG. 1. (Color online) The decay of ^{198}Au in gold (III) oxide, at room temperature. Experimental data appear as dots in the upper portion of the figure; the straight line is the fit to these data. Normalized residuals are shown at the bottom.

For the present measurement the systematic uncertainty associated with the residual-loss correction is small compared to the statistical uncertainty, so the total uncertainty is unchanged from the statistical one. Our final result is thus 2.6948(9) d. This is in excellent agreement with 2.695 00(27) d, the weighted average of all previous measurements of the gold half-life (see Ref. [4]), most of which were performed at an unrecorded temperature and in an unspecified host medium.

IV. CONCLUSIONS

We have measured the half-life of ^{198}Au in gold (III) oxide at room temperature. This result obtained with the decaying nuclei sited in this insulating medium is consistent with a half-life result we published previously for ^{198}Au sited in pure gold, a conductor. We established that the difference between the half-lives measured in an insulator and in a conductor is less than 0.04%, with a confidence level of 68% (one standard deviation). This limit is more than two orders of magnitude lower than the 7% difference predicted by the Debye plasma model [3]. Our result, together with previous measurements of ours [4,11] and others [12], effectively refutes all the predictions of the Debye plasma model as they apply to β^-, β^+, and electron-capture decays and also contradicts the measurements that initially supported those predictions [2,3,13].

Our concern in undertaking these measurements was for the integrity of precise half-lives measured in the past. Because physical conditions were believed to have no influence on half-lives, no care was taken in the past to select a particular host material or even to specify the temperature at which a measurement was made. Our main concern was with the half-lives of superallowed $0^+ \rightarrow 0^+\beta^+$ emitters, which are essential to fundamental tests of the standard model [14]. Their precision has typically been quoted to less than 0.05%, well below the temperature and host-material

dependence claimed by the measurements in Refs. [2,3,13]. We can now state with confidence that, at the level of 0.05%, half-lives are affected neither by temperature changes between 19 K and 295 K nor by the resistivity of the host medium in which they are located. There is no need to revisit past measurements of half-lives quoted to high precision.

ACKNOWLEDGMENTS

We thank R. Watson for his interest and for helpful discussions. We also appreciate the assistance of the staff at the Texas A&M Nuclear Science Center, where we had our source activated. This work was supported by the US Department of Energy under Grant No. DE-FG03-93ER40773 and by the Robert A. Welch Foundation under Grant No. A-1397.

[1] F. Raiola *et al.*, Eur. Phys. J. A **19**, 283 (2004).
[2] B. Limata *et al.*, Eur. Phys. J. A **28**, 251 (2006).
[3] T. Spillane *et al.*, Eur. Phys. J. A **31**, 203 (2007).
[4] J. R. Goodwin, V. V. Golovko, V. E. Iacob, and J. C. Hardy, Eur. Phys. J. A **34**, 271 (2007).
[5] H. Shi, R. Asahi, and C. Stampfl, Phys. Rev. B **75**, 205125 (2007).
[6] F. Machalett, K. Edinger, J. Melngailis, M. Diegel, K. Steenbeck, and E. Steinbeiss, Appl. Phys. A **71**, 331 (2000).
[7] [http://www.ortec-online.com/Solutions/multichannel-analyzers.aspx].
[8] R. Jenkins, R. W. Gould, and D. Gedcke, *Quantitative X-ray Spectrometry* (Dekker, New York, 1981), p. 266.
[9] D. Radford (private communication) [http://radware.phy.ornl.gov/main.html].
[10] R. Brun and F. Rademakers, Nucl. Instrum. Methods Phys. Res. A **389**, 81 (1997).
[11] J. R. Goodwin, V. V. Golovko, V. E. Iacob, and J. C. Hardy, Phys. Rev. C **80**, 045501 (2009).
[12] G. Ruprecht, C. Vockenhuber, L. Buchmann, R. Woods, C. Ruiz, S. Lapi, and D. Bemmerer, Phys. Rev. C **77**, 065502 (2008); **78**, 039901(E) (2008).
[13] B. Wang *et al.*, Eur. Phys. J. A **28**, 375 (2006).
[14] J. C. Hardy and I. S. Towner, Phys. Rev. C **79**, 055502 (2009).