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Absence of low-temperature dependence of the decay of ^{7}Be and ^{198}Au in metallic hosts

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The electron-capture (EC) decay rate of ^{7}Be in metallic Cu host and the β^{-}-decay rate of ^{198}Au in the host alloy Al-Au have been measured *simultaneously* at several temperatures, ranging from 0.350 K to 293 K. No difference of the half-life of ^{198}Au between 12.5 K and 293 K is observed to a precision of 0.1%. By utilizing the special characteristics of our double-source assembly, possible geometrical effects that influence the individual rates could be eliminated. The ratio of ^{7}Be to ^{198}Au activity thus obtained also remains constant for this temperatures range to the experimental precision of $0.15 \pm 0.16\%$. The resulting null temperature dependence is discussed in terms of the inadequacy of the often-used Debye-Hückel model for such measurements.

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The issue of the possible dependence of β-decay and electron-capture (EC) rates of radioactive nuclei on the nature and temperature of the host matrix and environment is a longstanding subject [1,2]. Numerous recent experiments have claimed that the half-life of radioactive nuclei embedded in metals would be significantly affected by screening of the electrons in the metal and this effect further be strengthened at low temperatures [3–7]. Several of the most recent articles cite a longer half-life for the EC of ^{7}Be ($0.8 \pm 0.2\%$ [8]) and a shorter half-life for the β^{+}-decay of ^{22}Na ($1.2 \pm 0.2\%$ [9]), where these nuclides were implanted in the metals Pd and In and cooled to $T = 12$ K. For the β^{-}-decay of ^{198}Au in a Au metallic environment the half-life was observed to be longer by $0.4 \pm 0.7\%$ at room temperature and by $4.0 \pm 0.7\%$ when the metal was cooled to $T = 12$ K, both compared to the literature value of the half-life [10]. The half-life of ^{7}Be in C_{60} at $T = 5$ K was observed to be 0.34(11)% shorter than that in C_{60} at $T = 293$ K and 1.5(1)% shorter than that inside Be metal at $T = 293$ K [11]. These errors are calculated using the half-life values as provided in [11]. For α-decay, the half-life of ^{210}Po in Cu metal at $T = 12$ K was measured to be shorter by $6.3 \pm 1.4\%$ as compared to room temperature [12]. However, such effects could not be confirmed by other experiments to the accuracy level of parts of a percent [13–16].

On the theoretical side, we note that the Debye-Hückel screening model, used in several previous publications to explain the apparent temperature dependence, is not applicable for a solid (strong-coupled plasma) but rather for a weak-coupled plasma. Furthermore, even within the Debye-Hückel screening model itself, the explicit temperature dependence of U_D—the Coulomb energy of the Debye cloud—of the form $1/\sqrt{T}$, is valid only for temperatures much higher than the Fermi temperature, E_F, being of the order of thousands of degrees for Cu [15,17,18]. Below the Fermi temperature (certainly for 293 K and lower), there should be no temperature dependence.

In light of this experimental and theoretical situation, we have recently carried out a precision measurement of the half-life of ^{7}Be in different host materials, where we found a null (or very small) change for conductors versus insulators at room temperature [19]. In this Communication, we report on the *simultaneous* measurement of the temperature dependence of the EC decay rate of ^{7}Be in metallic Cu host and of the β^{-}-decay rate of ^{198}Au in the host alloy Al-Au, ranging from 0.350 K to 293 K. Due to the relatively short half-life of ^{198}Au, it is possible to determine precisely the temperature dependence of its half-life within the time constraints of a low-temperature experiment. Subsequently, by utilizing the special characteristics of our double-source assembly (see below), possible geometrical and systematic errors that influence the individual rates could be eliminated by examining the *ratio* of ^{7}Be to ^{198}Au activities that decay by close-energy γ-rays.

The "double-source assembly" in the present experiment was based on the 2 mm diameter ^{7}Be source, prepared by direct implantation at ISOLDE (CERN) onto a copper disk of 12 mm diameter and 1.5 mm thickness, whose characteristics have been provided in detail in [20–22]. ^{7}Be decays by EC to the ground and first excited state of ^{7}Li at 477.6 keV. The branching ratio to $^{7}Li^{*}$(477.6) is 10.44(4)% [23]. The half-life 53.353(50) d of ^{7}Be in copper was measured in a previous study [19]. Adjacent to the 2 mm spot of ^{7}Be activity on the Cu disk, a ^{198}Au ($T_{1/2} = 2.6956(3)$ d [24]) source was attached. This radionuclide disintegrates by 100%β^{-} emission with a 95.58(12)% branch of a 411.8 keV γ-ray [25], very close in energy to the γ line of ^{7}Be. A 0.51 mm thick Al-Au (0.135% of Au) alloy wire of mass 13.7 mg was irradiated in the nuclear reactor at the Soreq Nuclear Research Centre, Israel, to produce ^{198}Au by neutron activation—^{197}Au$(n,\gamma)^{198}$Au. The 20 min irradiation in the pneumatic transfer tube facility ("Rabbit") of the reactor induced an activity of approximately 2.8×10^{4} Bq at the end of irradiation, one to two days prior to the commencement of the γ measurements. The active Al-Au wire was glued to the copper disk, adjacent to the ^{7}Be spot, by a heat-conducting, low-temperature adhesive, thus assuring identical thermal as well as geometrical (thermally affected expansion and contraction) properties for both ^{7}Be and ^{198}Au radionuclides.

Decay γ-rays were measured by a p-type coaxial HPGe portable detector of 65% relative detection efficiency, 2.05 keV energy resolution (FWHM) and 70.4 peak/Compton ratio, all specified at the 1332.5 keV γ-ray of ^{60}Co. The detector was enveloped by a 5.1 cm mercury cylinder placed within

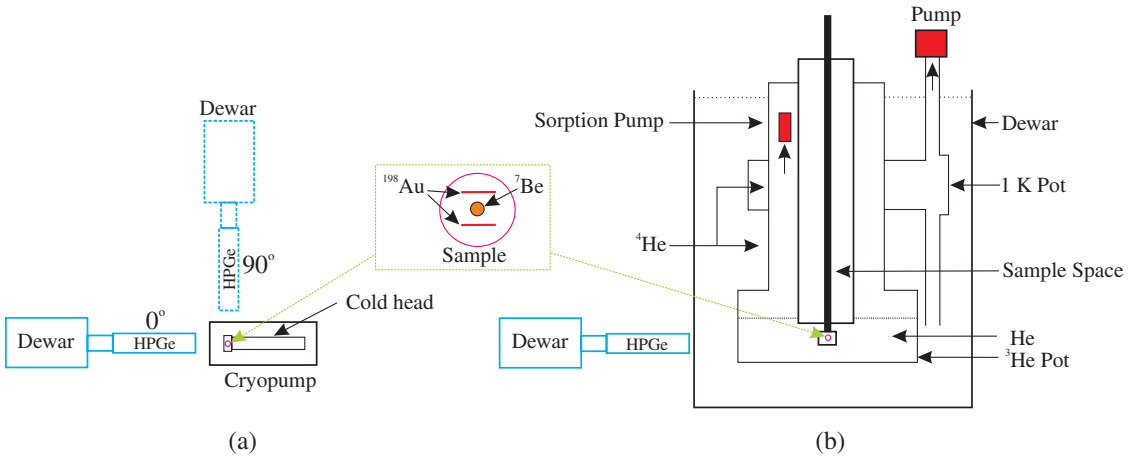

FIG. 1. (Color online) (a) Schematics of the setup for the measurements at 12.5 K and 293 K ("experiment 1") at the 0° and 90° geometry assemblies. (b) Schematics of the setup for the measurement at 0.350 K and 4 K ("experiment 2") at the 90° geometry assembly.

a 5.1 cm thick lead shield to suppress the environmental background.

We have used two experimental arrangements for the series of temperature dependence measurements, one for a $T = 293$ K to $T = 12.5$ K comparison ("experiment 1") and one for a $T = 4$ K to $T = 0.350$ K comparison ("experiment 2"). In "experiment 1", the sample was attached [see Fig. 1(a)] to a copper block, fixed to the cold head of a liquid-helium driven cryopump, at a minimum temperature of $T = 12.5$ K and a typical pressure of 3×10^{-5} mbar. As can be seen in Fig. 1(a), the Ge detector was placed at 0° and 90°, alternatively, to the axis of the cryopump, mounted on a massive stand at a distance of 50 mm from the sample for both geometries, ensuring mechanical stability of better than 0.1 mm. The geometrical correction to the effective detection efficiency due to thermal contraction of the cold head is sizably reduced when the detector is placed at 90° (see discussion below). The temperature was measured with a silicon diode, mounted close to the source and was recorded continuously.

For the 0° geometry, three measurements of the ^{7}Be and ^{198}Au activities at room temperature commenced approximately 24 h after the end of irradiation in the reactor with a typical statistical precision of the 411.8 and 477.6 keV peaks of ^{198}Au and ^{7}Be of 0.04 and 0.35%, respectively. The cryopump was then turned on, reaching the temperature limit of $T = 12.5$ K in approximately 3 h; the counting time of each of the next seven measurements at 12.5 K was set to about 12 h with a typical statistical precision of 0.05 and 0.32%. Following this first cycle of room- and low-temperatures, the cryopump was then turned off, with room temperature being reached in approximately 4 h after flushing with dry nitrogen gas. A similar cycle of six measurements at 293 K and eight measurements at 12.5 K was then repeated, finally followed by four measurements at room-temperature with precisions of 0.20 and 0.34%. In the 90° measurements, there was one cycle of room- and low-temperatures and finally, 4 measurements at room-temperature with typical precisions of 1.08 and 0.72% of the ^{198}Au and ^{7}Be peaks, respectively. The 90° measurements resulted in lesser precision data and were stopped when the typical statistical uncertainty of the ^{198}Au activity became comparable to that of the ^{7}Be activity, thus significantly affecting the combined uncertainty on the activity ratio. The analysis of the half-life of ^{198}Au (see below) was based only on the 0° measurements.

In order to probe the temperature dependence at yet lower temperatures, we have performed a second series of experiments in which the copper disk with the double source assembly was immersed in a Cryofab CMSH-100 [26] ^{3}He-^{4}He cryostat [Fig. 1(b)—"experiment 2"]. The complete fridge includes a ^{4}He dewar which supplies liquid ^{4}He to a 1 K Pot where the ^{4}He is pumped on, and a separate but thermally coupled ^{3}He system (Pot). A sorption pump (a molecular sieve) is used to pump on the liquid ^{3}He and cool it down to 0.350 K. In this way, the sample was cooled down by being thermally coupled to the pumped-on liquid ^{3}He to reach 0.350 K and 4 K alternately [see Fig. 1(b)]. Even though such a dewar is not well suited for γ detection, it nevertheless allowed probing the temperature dependence down to 0.35 K. The Ge detector was placed horizontally outside the dewar in the 90° geometry. Due to the relatively large distance of the Ge detector from the sources and due to considerable absorption of the γ radiation in the dewar's wall, the statistical precision of the 411.8 and 477.6 keV peaks of ^{198}Au and ^{7}Be was not as good as in "experiment 1", typically 0.25 and 1.04%, respectively. We quote below only the *ratio* of ^{7}Be to ^{198}Au activities, a procedure that is evermore important for this setup.

The half-life ($T_{1/2}$) of ^{198}Au at the two temperatures of 12.5 and 293 K at the 0° detector position was derived by a weighted linear regression of the logarithm of the net peak count rate versus time. The decay curve of ^{198}Au is shown in Fig. 2 and the calculated two straight lines fit well the measured results. The weighted averages of the measured half-lives of the ^{198}Au in the host alloy Al-Au are presented in Table I, where n is the number of data points, r is the correlation coefficient, χ^2_ν is the reduced chi-square, the number of degrees of freedom ν is equal to $n - 2$, and $P_\chi(\chi^2, \nu)$ is the probability that any random set of n data points would yield a value of chi-square not smaller than χ^2. The goodness of the linear fits is clearly

TABLE I. The details of the half-life determination of ^{198}Au and the linear regression fits at 12.5 and 293 K temperatures.

Temperature (K)	n	$T_{1/2}$ (d)	$(r+1)10^5$	χ^2_ν	$P_\chi(\chi^2,\nu)$ (%)
293	13	2.6971(20)	2.3	0.75	69.0
12.5	15	2.6976(23)	4.9	1.07	38.4

shown in Table I. The ratio of the half-life values at 12.5 to 293 K is calculated to be 1.0002(11) for the 0° geometry measurement. Hence, there is a temperature null-effect of the half-life of ^{198}Au in the host alloy Al-Au (0.135% of Au) at the range 293 to 12.5 K. The weighted average of half-life of ^{198}Au 2.6973(15) d observed in the experiment is in agreement within 1σ with the published value 2.6956(3) d [24]. The half-life values for ^{198}Au in agreement with the above have been obtained also for the 90° geometries in both "experiment 1" and "experiment 2", albeit with lesser precision due to the decaying source strength.

Due to the relatively low-intensity of the ^{7}Be and the short time span of the experiment (about 0.2 of the half-life; a dedicated experiment for the half-life determination requires six months [19]), we have not opted for a similar half-life determination for ^{7}Be, but have rather chosen the *normalized ratio* of ^{7}Be to ^{198}Au activities as a measure of the temperature dependence of the ^{7}Be EC rate. The count rates of ^{7}Be and ^{198}Au were calculated for every measured spectrum and corrected for decay using the published half-lives 53.353 and 2.6956 d. The two reference times for the 0° and 90° measurements were the middle times of the first room-temperature runs in each. The weighted averages of the ratios of count rates at different temperatures are presented in Fig. 3 and in Table II.

The individual normalized count rates of ^{7}Be and ^{198}Au at the 0° position for 293 K were 2.8462(44) and 236.05(6). For 12.5 K the normalized rates were 2.8121(29) and 232.59(20), respectively. These results demonstrate that at this position, the

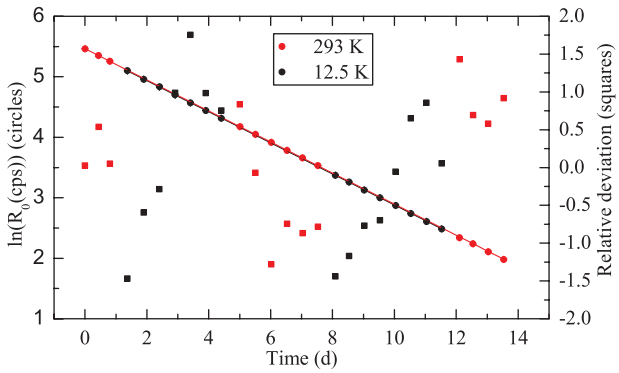

FIG. 2. (Color online) Left axis (circles): Decay curve of the 412 keV γ-ray from the ^{198}Au in Al-Au. The red and black colors correspond to the measured points and to the χ^2 fitted straight lines at 293 K and 12.5 K temperatures, respectively. Right axis (squares): The deviation between the measured and the fitted count rates, divided by the corresponding individual uncertainty. The color code is identical to that of the decay-curve values (left axis).

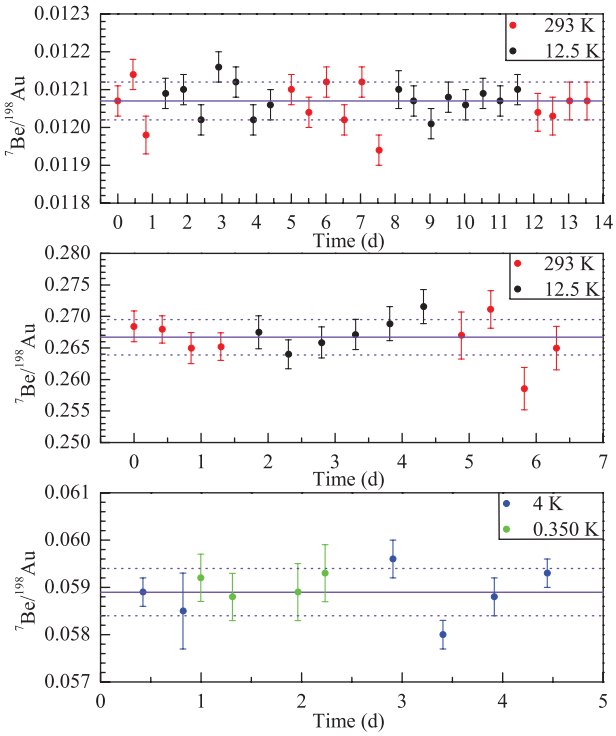

FIG. 3. (Color online) Ratio of rates (^{7}Be/^{198}Au). Top: 0° geometry for 12.5 K and 293 K; Middle: 90° geometry for 12.5 K and 293 K; Bottom: 90° geometry for 4 K and 0.350 K (see text). The solid lines represent the weighted averages and the broken lines correspond to an interval of $\pm 1\sigma$ standard deviation from the average—reflecting the scattering of the individual points and possible systematic errors.

count rates of both radionuclides are lower by about (1.20 ± 0.18) to (1.46 ± 0.09)% at the low temperature (12.5 K) compared to the room temperature (293 K). This finding can be qualitatively explained by the geometrical contraction during cooling down of the cold head of the cryopump (on which the copper disk was mounted) that is fixed by 70 mm long stainless steel rods. The distance between the sources and the Ge detector was 50 mm, inducing a 1.2% decrease of the count rate, which agrees very well with the measured decrease 1.2 to 1.5%. This effect may have influenced the previous

TABLE II. Weighted averages of the ratio of count rates of ^{7}Be and ^{198}Au, in the various temperatures and positions of the Ge detector. First four measurements were calculated from "experiment 1" and last two measurements were calculated from "experiment 2".

Temperature (K)	Geometrical configuration	Ratio (^{7}Be/^{198}Au)
293	0°	0.01206(2)
12.5	0°	0.01208(1)
293	90°	0.2663(11)
12.5	90°	0.2672(10)
4	90°	0.0589(3)
0.350	90°	0.0590(3)

measurements [3,9]. The corresponding count rates of ^{7}Be and ^{198}Au at the 90° position for 293 K were 0.8030(22) and 94.53(24) and 0.8092(29) and 95.57(44) for 12.5 K, respectively. This amounts to an opposite dependence at 90°, fully consistent with a null-effect that can be anticipated since the thermal contraction induces a negligible change in the distance between the detector and the sources. We emphasize again that these effects cancel out in very good approximation by the ^{7}Be/^{198}Au ratios.

The half-life of ^{198}Au in the alloy Al-Au (0.135% of Au) was measured and a null-effect of (0.02 ± 0.11)% was found at 12.5 K relative to 293 K. The count rate of ^{7}Be in copper was measured at these temperatures relative to that of ^{198}Au. The normalized ratio of ratios (ratio ^{7}Be/^{198}Au at 12.5 K to ratio ^{7}Be/^{198}Au at 293 K), at the 0° and 90° are 1.0015(16) and 1.0032(58), which are equal within the experimental uncertainty. Therefore, the half-life of ^{7}Be at 12.5 K is shorter by (0.15 ± 0.16)% than the value of 53.353 d at 293 K [19]. The same ratios of ^{7}Be to ^{198}Au yield 0.0590(3) and 0.0589(3) at 0.350 K and 4 K, respectively. The ratio of ratios is therefore 1.0028(71), again a null temperature dependence of (0.28 ± 0.71)% for these two temperatures.

The presently determined decay rates for ^{7}Be (EC) and ^{198}Au (β^{-}) for a wide temperature range of 293 K to 12.5 and 4 K to 0.35 K do not support earlier observations for metallic hosts and present a novel technique for probing subpercentage effects in the decay of, e.g., ^{7}Be and other nuclei. The results also validate the theoretical picture above.

Note added. After the submission of this manuscript, a new measurement on the T-dependence of the half-life of ^{198}Au was published [27], in excellent and full agreement with the present null effect.

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