Redetermination of ^{198}Au and ^{192}Ir γ-Ray Standards between 0.1 and 1.0 MeV

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Prominent γ-ray reference lines including ^{198}Au 411 and 675 keV and ^{192}Ir 205, 295, 308, 316, 468, 484, 588, 604, and 612 keV have been measured with respect to an I_2-stabilized He-Ne laser. The ^{198}Au 411-keV line, a *de facto* γ-ray standard, is reported at 3.010 7788 pm (0.37 ppm) or 411 804.41 eV, which is 25 ppm higher in energy and forty times more accurate than previous values.

Uncertainty in present-day Ge(Li) spectrometer measurements of mesonic x-ray wavelengths is dominated by the contribution of the γ-ray standards used for calibration (except those using preliminary results of this study).[1] The situation will worsen when more precise measurements using bent-crystal instruments coupled to high-current proton machines and meson production facilities become available. The problem is typified by the current situation in muonic x-ray spectra where the large differences between theoretical and experimental wavelengths which existed several years ago have been reduced by improved experimental techniques to the point that the remaining discrepancies are comparable with the uncertainty in the γ-ray standards, for example, the energy of ^{198}Au 411 (17 ppm).[2]

Such comparisons between theory (QED) and experiment require the ratio m_μ/m_e which is available from the muonium hyperfine structure interval.[3,4] One also needs γ-ray markers evaluated with respect to the electronic Rydberg, R_∞. This Letter presents redeterminations of an important group of γ-ray lines using a procedure which connects them with R_∞ in a direct and accurate way.

The most recent high-precision determination of R_∞ (for H and D) used as a reference a 633-nm He-Ne laser stabilized with respect to a saturated absorption feature (n) in the $^{129}I_2$ spectrum,[5] which has been related to the absorption feature (B) by a beat-frequency measurement.[6] Characteristics of this laser have been well established as has its relation to the 606-nm ^{86}Kr standard.[6] It has also been related to the CH_4-stabilized (3.4 μm) He-Ne device[7] which is connected to the Cs oscillator which defines the frequency scale.[8]

Our measurement chain to γ-ray wavelengths began with such a $^{129}I_2(B)$-stabilized He-Ne laser whose emission is assigned a wavelength of 632 990.079 pm (4 ppb).[6,9] It illuminated a high-finesse optical Fabry-Perot interferometer whose elements were attached to the separated parts of a two-crystal Laue case x-ray interferometer made of Si.[10] From scans of a common baseline we obtained a value for the lattice period in terms of the visible wavelength (in vacuum). Subsequent to the initial report,[10] improved procedures[11] and corrections for certain systematic problems have yielded a current value for the Si sample's cell edge dimension of 0.543 102 80 nm (0.15 ppm) at 22.5°C.[12]

From this interferometrically calibrated specimen, we determined interplanar spacings for other Ge and Si specimens more suitable for the γ-ray measurements using a modification of the quasi-nondispersive procedure of Hart.[13] For the Si γ-ray crystals the spacing of the (440) planes was directly compared to that of the calibrated specimen. For the Ge γ-ray crystals the spacing of the (800) planes was compared to that of the (355) spacing of an intermediate Si sample.[14] The measured lattice spacings at 22.5°C are a_0(Si) = 0.543 102 71 nm (0.2 ppm) and a_0(Ge) = 0.565 782 16 nm (0.2 ppm). Corrections for temperature changes were made using[15] $\alpha_{Si} = (2.56 \pm 0.03) \times 10^{-6}$ K^{-1} and[16] $\alpha_{Ge} = (5.95 \pm 0.11) \times 10^{-6}$ K^{-1}.

We used these calibrated crystals in a transmission double-flat-crystal instrument as indicated in Fig. 1. For rays parallel to the plane of dispersion, the Bragg-Laue equation $\lambda = 2d \sin\theta$ has no known error. Adequate sensitivity for measurement of the small diffraction angles ($\sim 10^{-2}$ rad) was obtained with Michelson angle interferometers[17] in which a 90° phase change corresponded to 0.03 arc sec. The fractional order was encoded as a polarization azimuth[1] and the system was totally servoed with null stabilities near 0.05 arc msec. Absolute calibration of the angle interferometers was obtained by summing to closure the interfacial angles of a 72-sided optical polygon using a sensitive (0.2 arc msec) autocollimator. Precision of these calibrations was about 0.05 ppm while their accuracy,

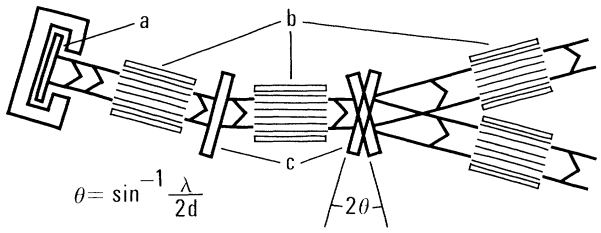

FIG. 1. Outline of γ-ray wavelength measurement. Radiation from a radioactive source *a* is collimated by the slits *b* and diffracted by crystals *c*.

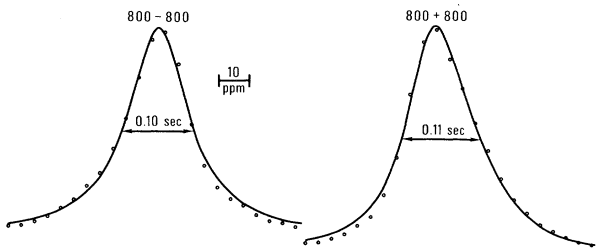

FIG. 2. Nondispersive and dispersive profiles for Ge crystals at Au (411 keV). The dots are data points and the curve is the computer fit. The asymmetry in the dispersive profile seems primarily due to vertical divergence effects.

as estimated from the time dependence of the output, was limited to about 0.1 ppm because of growth of the Invar material (0.5 ppm/yr) in the angle interferometers.[18]

In practice, radiation cannot be confined parallel to the plane of dispersion. The finite opening angle requires that a "vertical divergence correction" be introduced to convert observed diffraction angles to Bragg-Laue angles. We generated an explicit spectral window function which included an (asymmetric) vertical divergence function obtained analytically[19] from geometry, a Lorentzian component obtained from dynamical diffraction theory,[20] and a residual Gaussian determined empirically,[21] and fitted this to data, extracting from the fitting procedure the Bragg-Laue angle and statistical measures of goodness of fit. An example of the results of this procedure is shown in Fig. 2.

Measurements on the ^{198}Au 411-keV line have extended over a two-year period in which seven samples were irradiated. Only measurements associated with the latter four irradiations are included because the earlier irradiations were hampered by weak source strength, insufficient knowledge of the zero angle, and an extremely temperature-sensitive optical element. Wavelength measurements from each of the four irradiations exhibit a standard deviation, σ_i', of about 0.3 ppm. This would lead to estimates of σ_m' (for the mean) of about 0.18 ppm. On the other hand, the intergroup variance suggests the presence of an uncontrolled systematic characterized by $\sigma_s = 0.13$ ppm. As suggested by Eisenhart,[22] we combined this quadratically with the effective σ_m' to obtain an overall estimate of $\sigma_m = 0.22$ ppm. Both Si and Ge results are included in the reckoning; no significant difference was noted between the separate averages of Ge and Si data. The Si crystals were used in the configurations (220 ± 220), (440 ± 440), and (880 ± 880) while the Ge crystals were used in (400 ± 400), (400 ± 800), (800 ± 800), (800 ± 12 0 0), and (12 0 0 ± 12 0 0). The gold sources had an initial activity of 5000 Ci.[23]

We also report here new results for the ^{198}Au 675-keV line and the following lines from ^{192}Ir: 205, 295, 308, 316, 468, 484, 588, 604, and 612 keV. The Ir source had an initial activity of 2000 Ci. The upper part of Table I summarizes our results for these lines. The energies are obtained from the wavelengths via the voltage-wavelength conversion factor, namely $V\lambda = 1.239\,8520 \times 10^{-6}$ eV · m (2.6 ppm)[24]; this uncertainty is properly ignored except in cases where comparison is needed with results from electrical data. Although the Au 411-keV line is the only line for which there are sufficient data to estimate the uncontrolled systematic mentioned above, a 0.13-ppm error was also combined with the statistical uncertainties of the other lines to obtain the measurement uncertainties. The lower part of Table I gives energies obtained by summation of measured lines after correction for recoil. Where energy-level relations lead one to expect equality, namely 295 + 308 = 604 and 295 + 316 = 612, equality is obtained within 0.8 and 0.9 ppm with 1σ estimates of 0.8 and 1.3 ppm.

Our result for ^{198}Au 411 differs from that of Murray, Graham, and Geiger[2] by 1.5 times the quoted σ of their determination. An alternative route to ^{198}Au is via x rays. This combines a measurement of λ(Mo $K\alpha_1$),[10] the ratio λ(Mo $K\alpha_1$)/λ(W $K\alpha_1$) of Bearden *et al.*,[25] and the ratio $E(^{198}\text{Au}\ 411)/E(\text{W}\ K\alpha_1)$ from Borchert, Scheck, and Schult[26] to obtain $E(^{198}\text{Au}\ 411) = 411\,800$ eV (3.1 ppm), in significant disagreement with our new value. We have remeasured W $K\alpha$ and the result resolves the problem with a difference of 2.7 ± 3.1 ppm.[27]

A second kind of comparison that can be made is of the relative values implied by the data of Table I. Bent-crystal measurements have been,

TABLE I. Measured energies, measurement uncertainties, σ_m, and total uncertainties, σ_T. The systematic errors included calibration, 0.1 ppm; lattice spacing, 0.2 ppm; and vertical divergence, 0.2 ppm. Direct observations are given in the upper part while the entries in the lower part are obtained by summation of the direct observations. Energies were obtained from wavelengths by use of $V\lambda = 1.239\,8520 \times 10^{-6}$ eV · m.

Source	Energy (keV)	σ_m (ppm)	σ_T (ppm)
^{198}Au	411.804 41	0.22	0.37
^{198}Au	675.887 43	0.98	1.02
^{192}Ir	205.795 49	0.13	0.33
^{192}Ir	295.958 25	0.32	0.44
^{192}Ir	308.456 89	0.37	0.48
^{192}Ir	316.507 89	0.49	0.57
^{192}Ir	468.071 47	0.49	0.57
^{192}Ir	484.577 97	0.79	0.85
^{192}Ir	588.584 46	1.18	1.22
^{192}Ir	604.414 15	0.72	0.78
^{192}Ir	612.465 04	1.24	1.28
	Sum		
Au 411 + 675	1087.690 33	0.61	0.68
Ir 604 − 468	136.343 04	3.61	3.62
Ir 295 + 588 − 468	416.471 36	1.77	1.80
Ir 295 + 308	604.414 63	0.24	0.38
Ir 295 + 316	612.465 62	0.30	0.42
Ir 295 + 588	884.541 74	0.79	0.84

reported on the 675-keV line by Borchert[28] and on the Ir lines by Borchert, Scheck, and Wieder[29] and by Beer and Kern.[30] In addition, Helmer, Greenwood, and Gehrke[31] have developed an "adjusted" set of values for Ir including Ge(Li) data. For comparison all data have been renormalized to the presently reported ^{198}Au 411 value. The 675 result from Ref. 28 is 675.8898 keV ± 5.6 ppm which differs from the value in Table I by 3.6 ± 5.7 ppm. The Ir data are shown in Fig. 3 with the present results taken as origin values (the horizontal lines and rectangular 1σ estimates). The average slopes (dashed lines) suggest an angle-scale error or a source interchange problem. Comparison with our results would appear as the scatter about the dashed lines if the data were normalized to the average ^{192}Ir values. It appears that our data and other relative energy measurements are in reasonable agreement.

The discrepancies between theory and experiment for muonic x-ray spectra are dependent upon these new γ-ray reference values. Vuilleumier *et al.*[32] compile the differences between recent measurements and theory. When the energy scale is based on the Au 411 value reported here,

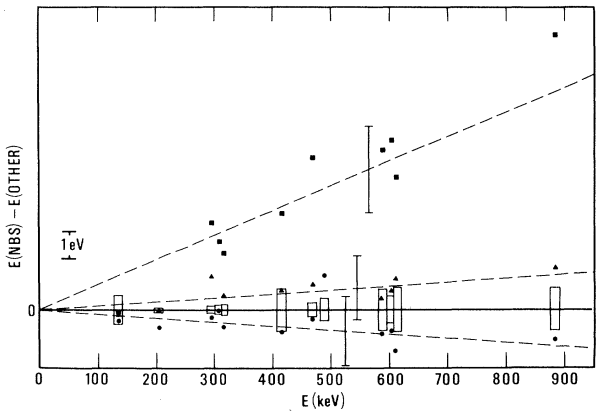

FIG. 3. Comparison of ^{192}Ir γ-ray energies from several sources. The average 1σ estimates are Ref. 29 (●) ± 1.2 eV, Ref. 30 (■) ± 1.5 eV, and Ref. 31 (▲) ± 1.1 eV.

the differences are reduced in two cases and increased in one case. However, the latest Ge(Li) data[33] which claim a higher accuracy than previous results appear to agree better with theory using the revised scale.

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Experimental Method for Testing the Potential of Moving Ions in Solids

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Molecular ions, which define the initial separation between nuclei, are used as a probe of the potential established by swift ions in solids. Employing 75–300-keV H_2^+ and HeH^+ ions one observes that a large fraction of clusters do not explode in solid targets whose thickness is much greater than the mean electron-loss distance. The results are consistent with the view that protons moving in solids carry bound electrons.

We report on measurements that were designed to probe the potential of ions moving in solids. This is accomplished by measuring the energy distribution of emerging ions when molecules in vibrational states are incident on solid targets. At our ion energies E_1 (75–300 keV), the electrons binding the molecules are torn off within ~5 to 10 Å.[1,2] The resulting clusters of ions explode under the influence of Coulomb repulsion between their constituents.[3] Aside from energy straggling, the width of the energy distribution of the emerging ions is determined by the conversion of Coulomb and vibrational energies into kinetic energy of the cluster ions over the range of the screened internuclear Coulomb force in the solid, and the cluster dwell time in the target. The relative intensities in this distribution are indicative of the fraction of clusters undergoing different degrees of explosion. Hence the members of the cluster can serve as test particles for the internuclear potential in clusters moving through solids.

We measure the energy distribution of emerging protons when H_2^+ and HeH^+ ions in the velocity range $v_1=(1.2 \text{ to } 2.45)v_0$ (75–300 keV) are incident on 70–500-Å (1.4 to 10 μg/cm^2) carbon foils. Hitherto only two-peak distributions have been observed.[4,5] We observe three-peak distributions.