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γ-ray irradiation in the early Solar System and the conundrum of the ^{176}Lu decay constant

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Abstract

When recent geological calibrations of the ^{176}Lu decay constant are used, the ^{176}Lu–^{176}Hf ages of chondrites are consistently 4% too old (∼4.75 Ga). Here, we suggest that this discrepancy reflects the photoexcitation of the long-lived ^{176}Lu ground state to the short-lived isomeric state ($T_{1/2} = 3.7$ h) by γ-rays irradiating early condensates. Irradiation may have been of solar origin and taking place at the inner edge of the nebular disk. Alternatively, the source of γ-rays could have been one or more supernova(e) exploding in the vicinity of the solar nebula. Such photoexcitation has been experimentally observed, but requires γ-ray photons that have energies in excess of 838 keV. At this stage, we cannot assess whether the Hf isotope composition of the Bulk Silicate Earth differs from that of chondrites, eucrites, and the 4.56 Ga old Martian meteorite ALH84001, and therefore, whether the precursor material for these different planetary bodies received comparable fluences of γ-rays.

1. Introduction

The actual value of the ^{176}Lu decay constant has so far eluded consistent determinations. The half-life of the decay of ^{176}Lu to ^{176}Hf (Norman, 1980; Sguigna et al., 1982; Sato et al., 1983; Gehrke et al., 1990; Dalmasso et al., 1992; Nir-El and Lavi, 1998; Grinyer et al., 2003), as is true for other nuclides decaying by β-emission, such as ^{87}Rb and ^{187}Re, is notoriously difficult to measure directly using standard counting techniques. Typically, physical measurements provide precise but inconsistent values (Fig. 1, see also Amelin and Davis, 2005, Fig. 1). As with most other beta decay chronometers, the hope was that age comparisons of whole-rock isochrons based on ancient objects, typically meteorites for which the age is known with an error smaller than the current spread of the decay constant values, could help make headway with this problem. In Fig. 1 we have compared existing determinations of $\lambda_{^{176}\mathrm{Lu}}$, including those obtained by physical measurements, from meteorite isochrons, and from well-dated terrestrial minerals and rocks. Because of the large spread in Lu/Hf ratios among eucrites, it was originally thought that they would be particularly well suited for deriving a precise $\lambda_{^{176}\mathrm{Lu}}$ value and this potential was explored by different groups (Patchett and Tatsumoto, 1980; Tatsumoto et al., 1981; Blichert-Toft et al., 2002; Scherer et al., 2003). In addition to these ^{176}Lu decay constant determinations using eucrites, Bizzarro et al. (2003) used a combination of chondrites and eucrites. These studies achieved reasonably good alignments on ^{176}Lu–^{176}Hf isochron diagrams, yielding estimates of $\lambda_{^{176}\mathrm{Lu}}$ of ∼1.93–1.98×10^{-11} a^{-1}, consistent with the early counting experiment value of Sguigna et al. (1982). However, chondrites as a group do not define a statistically meaningful isochron (Patchett et al., 2004) and cumulate eucrites, which appear to have formed or re-equilibrated somewhat later than basaltic eucrites, plot significantly off the Lu–Hf array of basaltic eucrites (Blichert-Toft et al., 2002). Such a scatter

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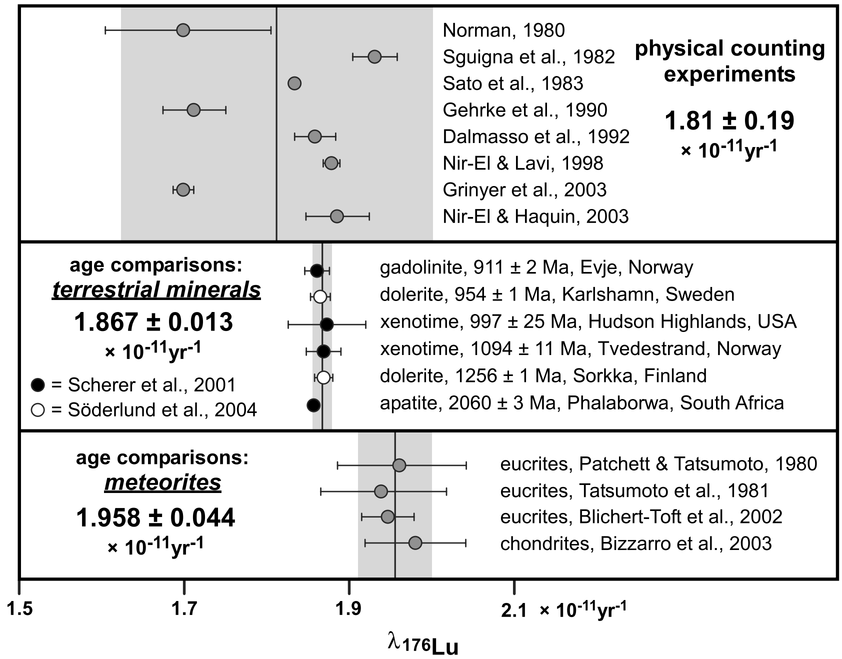

Fig. 1. Compilation of the values of $\lambda_{^{176}\mathrm{Lu}}$ obtained by physical laboratory measurements (top), compared with values determined by age comparison for precisely dated terrestrial minerals and rocks (middle), and for meteorite suites (bottom). The mean $\lambda_{^{176}\mathrm{Lu}}$ from terrestrial samples includes data of Scherer et al., 2003. Shaded vertical bars indicate the two s.d. of the values in each category.

indicates either some Hf isotopic heterogeneity at the time the parent bodies of these meteorites formed or severe perturbation at a later stage.

Another approach was applied to terrestrial samples by Scherer et al. (2001), who dated old, high-Lu/Hf minerals by both U–Pb and Lu–Hf, and Soderlund et al. (2004), who obtained internal Lu–Hf isochrons and baddeleyite U–Pb ages on Proterozoic dikes. To verify the emerging dichotomy between meteorite and terrestrial data sets, Scherer et al. (2003) confirmed their own original results on pegmatite minerals (gadolinite) from different localities. The mean value of the ^{176}Lu decay constant obtained from the analysis of these terrestrial samples ($1.867 \times 10^{-11}\ \mathrm{a}^{-1}$) is ~4% lower than the meteorite value, and, when applied to meteorites, makes their Lu–Hf ages unacceptably old (~4.75 Ga). In contrast, the 'terrestrial' decay constant, when applied to 3.7 Ga supracrustal rocks from Isua, to 2.1 Ga old rocks from West Africa (Blichert-Toft et al., 1999), and to apatites from the Eocene Gardiner intrusion (Barfod et al., 2003), yields Lu–Hf ages that agree with ages from other isotope systems, whereas, the meteorite value results in ages that are conspicuously too young. The alternatives therefore are that either all the terrestrial values are consistently wrong or the ^{176}Lu–^{176}Hf isochrons defined by chondrites and eucrites are biased by an early process that acted exclusively before Earth's accretion.

2. Previous attempts to explain the ^{176}Lu decay constant conundrum

The question of why terrestrial and meteoritic samples yield different apparent $\lambda_{^{176}\mathrm{Lu}}$ values is currently the subject of much speculation and, as expressed by Soderlund et al. (2004), 'unanswered questions remain as to why the terrestrial versus meteoritic discrepancy exists.' This topic has been recently reviewed in detail by Soderlund et al. (2004) and Amelin and Davis (2005). Only a brief summary is needed here where we will consider the potential explanations provided so far for the $\lambda_{^{176}\mathrm{Lu}}$ conundrum:

(i) It is observed that the ^{176}Lu–^{176}Hf array of meteoritic material does not satisfy the statistical criteria for an isochron (this will later prove to be informative). The statistical properties of the array can be evaluated by calculating the initial ^{176}Hf/^{177}Hf values of all the chondrites and achondrites measured so far (Blichert-Toft and Albarède, 1997; Blichert-Toft et al., 2002; Bizzarro et al., 2003; Patchett et al., 2004). A few clear outliers such as Orgueil in Blichert-Toft and Albarède (1997) and the cumulate eucrites from Blichert-Toft et al. (2002) have been excluded. Since most of these rocks are known to have formed within the first 50 My of the Solar System (which is much less than the observed age discrepancy of 4%), pooling these whole-rock samples to test the hypothesis of initial Hf isotopic homogeneity of planetary objects is legitimate. The standard deviation s of the initial ^{176}Hf/^{177}Hf values is calculated in Table 1 and Fig. 2 using both the 'terrestrial' decay constant ($1.867 \times 10^{-11}\ \mathrm{a}^{-1}$) and the conventional 'meteoritic' ($1.94 \times 10^{-11}\ \mathrm{a}^{-1}$) decay constant. The minimum value of s (0.000022 or 0.8 ε units) lies outside 1-sigma reproducibility on the ^{176}Hf/^{177}Hf ratios (~0.18 ε units). This is in agreement with previous conclusions that a unique isochron does not exist. For an age of 4.56 Ga, the $1.867 \times 10^{-11}\ \mathrm{a}^{-1}$ makes s even larger (0.000025 or almost 6 times the in-run statistics). Chondrite-only and

Table 1
Initial Hf isotope compositions for meteorite samples calculated for two values of the ^{176}Lu decay constant at different times

Sample	Type	Ref.	^{176}Lu/^{177}Hf	^{176}Hf/^{177}Hf	Initial ^{176}Hf/^{177}Hf ($\lambda_{^{176}Lu} = 1.867 \times 10^{-11}$ a^{-1})				Initial ^{176}Hf/^{177}Hf ($\lambda_{^{176}Lu} = 1.94 \times 10^{-11}$ a^{-1})			
Initial time (Ga)					4.55	4.65	4.75	4.85	4.55	4.65	4.75	4.85
Béréba	EUC	1	0.0289	0.282367	0.279802	0.279743	0.279684	0.279625	0.279697	0.279636	0.279574	0.279513
Bouvante	EUC	1	0.0243	0.281908	0.279751	0.279701	0.279652	0.279602	0.279663	0.279611	0.279559	0.279508
Cachari	EUC	1	0.0315	0.282587	0.279793	0.279729	0.279665	0.279601	0.279679	0.279612	0.279545	0.279478
Caldéra	EUC	1	0.0355	0.282947	0.279800	0.279728	0.279656	0.279583	0.279672	0.279597	0.279521	0.279446
Camel Donga	EUC	1	0.0279	0.282287	0.279812	0.279756	0.279699	0.279642	0.279711	0.279652	0.279593	0.279533
Ibitira	EUC	1	0.0322	0.282677	0.279820	0.279755	0.279689	0.279623	0.279704	0.279635	0.279567	0.279498
Jonzac	EUC	1	0.0295	0.282404	0.279789	0.279729	0.279669	0.279609	0.279683	0.279620	0.279557	0.279495
Juvinas	EUC	1	0.0305	0.282471	0.279764	0.279702	0.279640	0.279577	0.279654	0.279589	0.279524	0.279459
Lakangaon	EUC	1	0.0286	0.282319	0.279782	0.279723	0.279665	0.279607	0.279678	0.279617	0.279556	0.279495
Millbillillie	EUC	1	0.0296	0.282398	0.279778	0.279718	0.279658	0.279597	0.279671	0.279608	0.279546	0.279483
Padvarninkai	EUC	1	0.0323	0.282633	0.279771	0.279705	0.279640	0.279574	0.279654	0.279586	0.279517	0.279448
Palo Blanco Creek	EUC	1	0.0262	0.282135	0.279815	0.279762	0.279708	0.279655	0.279720	0.279664	0.279609	0.279553
Pasamonte	EUC	1	0.0295	0.282393	0.279777	0.279717	0.279656	0.279596	0.279670	0.279607	0.279544	0.279482
Peramiho	EUC	1	0.0330	0.282752	0.279823	0.279755	0.279688	0.279621	0.279703	0.279633	0.279563	0.279492
Sioux County	EUC	1	0.0288	0.282333	0.279780	0.279721	0.279662	0.279603	0.279675	0.279614	0.279553	0.279492
Stannern	EUC	1	0.0253	0.282016	0.279773	0.279721	0.279670	0.279618	0.279681	0.279628	0.279574	0.279520
Harayia	EUC	1	0.0282	0.282343	0.279845	0.279788	0.279731	0.279673	0.279743	0.279684	0.279624	0.279564
Nuevo Laredo	EUC	1	0.0285	0.282317	0.279794	0.279736	0.279678	0.279620	0.279691	0.279630	0.279570	0.279509
Emmaville	EUC	1	0.0289	0.282336	0.279777	0.279719	0.279660	0.279601	0.279673	0.279611	0.279550	0.279489
Alfianello	L6	2	0.0355	0.282982	0.279834	0.279761	0.279689	0.279617	0.279705	0.279630	0.279554	0.279479
Barrata	L3.8	2	0.0326	0.282732	0.279843	0.279776	0.279710	0.279643	0.279724	0.279655	0.279586	0.279517
Belle Plaine	L6	2	0.0292	0.282382	0.279794	0.279735	0.279675	0.279615	0.279688	0.279626	0.279564	0.279502
Edmonson	H4	2	0.0339	0.282841	0.279836	0.279767	0.279698	0.279629	0.279714	0.279642	0.279570	0.279497
Ella Island	L6	2	0.0312	0.282569	0.279806	0.279743	0.279679	0.279616	0.279693	0.279627	0.279561	0.279495
Julesbourg	L3.6	2	0.0320	0.282641	0.279802	0.279737	0.279672	0.279606	0.279686	0.279618	0.279550	0.279482
Hedjaz	L3.7	2	0.0329	0.282726	0.279813	0.279747	0.279680	0.279613	0.279695	0.279625	0.279555	0.279485
Herredia	H5	2	0.0341	0.282821	0.279799	0.279729	0.279660	0.279590	0.279675	0.279603	0.279530	0.279458
M'Bale	L6	2	0.0264	0.282127	0.279785	0.279732	0.279678	0.279624	0.279690	0.279634	0.279578	0.279521
Tennesalim	L4	2	0.0329	0.282720	0.279805	0.279738	0.279671	0.279604	0.279686	0.279616	0.279546	0.279476
Waltman	L4	2	0.0316	0.282626	0.279824	0.279760	0.279696	0.279631	0.279710	0.279643	0.279576	0.279508
Allende	CV3	2	0.0331	0.282783	0.279847	0.279779	0.279712	0.279644	0.279727	0.279656	0.279586	0.279515
Allende	CAI	2	0.0324	0.282684	0.279816	0.279750	0.279684	0.279618	0.279699	0.279630	0.279561	0.279492
Allende	Matrix	2	0.0337	0.282792	0.279809	0.279740	0.279672	0.279603	0.279687	0.279615	0.279544	0.279472
Murchison	CM2	2	0.0323	0.282679	0.279820	0.279754	0.279688	0.279622	0.279703	0.279634	0.279566	0.279497
Murchison	CM2	3	0.0338	0.282804	0.279807	0.279738	0.279670	0.279601	0.279685	0.279613	0.279541	0.279469
Murray	CM2	3	0.0330	0.282742	0.279815	0.279748	0.279681	0.279613	0.279696	0.279626	0.279556	0.279485
Mighei	CM2	3	0.0339	0.282798	0.279792	0.279723	0.279654	0.279585	0.279670	0.279598	0.279526	0.279453
Kainsaz	CO3	3	0.0351	0.282942	0.279826	0.279755	0.279683	0.279612	0.279699	0.279625	0.279550	0.279475
Allende piece	CV3	3	0.0341	0.282847	0.279827	0.279758	0.279688	0.279619	0.279704	0.279632	0.279559	0.279487
Allende pdrb	CV3	3	0.0341	0.282828	0.279806	0.279736	0.279667	0.279597	0.279682	0.279610	0.279537	0.279465
Karoonda	CK4	3	0.0354	0.282951	0.279813	0.279741	0.279669	0.279597	0.279685	0.279610	0.279535	0.279459
Bjurböle	L/LL4	3	0.0324	0.282714	0.279846	0.279780	0.279714	0.279648	0.279729	0.279660	0.279591	0.279522
Homestead	L5	3	0.0332	0.282810	0.279867	0.279800	0.279732	0.279664	0.279747	0.279677	0.279606	0.279536
Holbrook	L6	3	0.0303	0.282483	0.279794	0.279732	0.279670	0.279608	0.279684	0.279620	0.279555	0.279491
Bruderheim	L6	3	0.0302	0.282518	0.279837	0.279775	0.279714	0.279652	0.279727	0.279663	0.279599	0.279535
Girgenti	L6	3	0.0313	0.282597	0.279821	0.279757	0.279694	0.279630	0.279708	0.279641	0.279575	0.279508
Avanhandava	H4	3	0.0324	0.282669	0.279795	0.279730	0.279664	0.279597	0.279678	0.279609	0.279540	0.279471
Ankober	H4	3	0.0317	0.282661	0.279850	0.279785	0.279720	0.279656	0.279735	0.279667	0.279600	0.279533
Ochansk	H4	3	0.0301	0.282444	0.279772	0.279710	0.279649	0.279587	0.279663	0.279599	0.279535	0.279470
Allegan	H5	3	0.0352	0.282951	0.279829	0.279758	0.279686	0.279614	0.279702	0.279627	0.279552	0.279477
Pultusk	H5	3	0.0337	0.282810	0.279825	0.279756	0.279688	0.279619	0.279703	0.279631	0.279560	0.279488
Richardton	H5	3	0.0310	0.282593	0.279843	0.279780	0.279716	0.279653	0.279730	0.279665	0.279599	0.279533
Forest City	H5	3	0.0336	0.282782	0.279804	0.279736	0.279667	0.279598	0.279682	0.279611	0.279540	0.279468
Indarch	EH4	3	0.0335	0.282797	0.279829	0.279761	0.279692	0.279624	0.279707	0.279636	0.279565	0.279494
Mean					0.279809	0.279745	0.279681	0.279616	0.279695	0.279628	0.279561	0.279494
s.d.					0.000025	0.000023	0.000022	0.000022	0.000022	0.000022	0.000024	0.000026

EUC, eucrites. The rest is chondrites. [1]Blichert-Toft et al. (1999); [2]Bizzarro et al. (2003); and [3]Patchett et al. (2004).

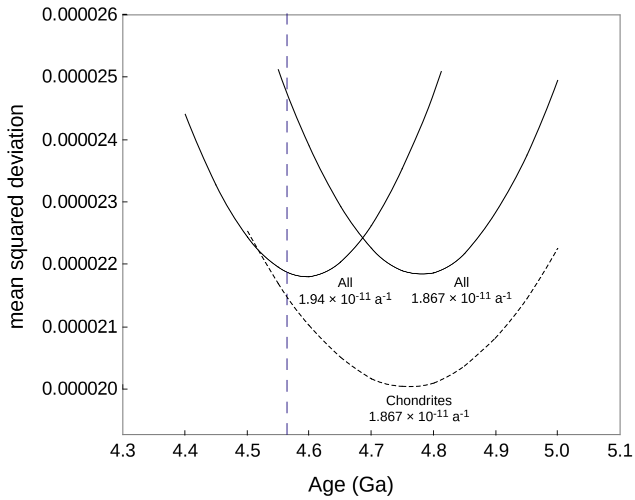

Fig. 2. Variations of the standard deviation of the initial ^{176}Hf/^{177}Hf ratios as a function of age. Chondrite data from Blichert-Toft and Albarède (1997), Bizzarro et al. (2003), and Patchett et al. (2004). Basaltic eucrite data from Blichert-Toft et al. (2002) and Bizzarro et al. (2003). Solid lines: eucrites and chondrites together using $\lambda_{^{176}Lu} = 1.867 \times 10^{-11}$ a^{-1} (left), $\lambda_{^{176}Lu} = 1.94 \times 10^{-11}$ a^{-1} (right). Dashed line: chondrite subset. The terrestrial decay constant $\lambda_{^{176}Lu} = 1.867 \times 10^{-11}$ a^{-1} cannot be reconciled with a ~4.56 Ga age of meteorites.

chondrite + achondrite arrays do not signal that the choice of material introduces a particular bias (Fig. 2). We therefore reject the explanation that the discrepancy is due to a poor statistical alignment in the isochron diagram.

(ii) Patchett et al. (2004) suggested Hf isotopic heterogeneities in the Early Solar System as a possible contribution to the complex behavior of the ^{176}Lu–^{176}Hf system. A variable contribution of s-, r-, and p-processes may in principle result in variable ^{176}Hf/^{177}Hf isotopic compositions (e.g., Table 7 in Käppeler et al., 1989 and Fig. 3, this work), but nucleosynthetic anomalies are almost invariably restricted to refractory inclusions (Birck, 2004), which, contrary to the bulk silicate material hosting them, have retained their geochemical identity through the turbulent mixing affecting the collapsing solar nebula. In addition, how a nucleosynthetic ^{176}Hf excess could be correlated with ^{176}Lu is unclear. A neutron irradiation-induced ^{176}Hf excess would require irradiation of the short-lived p-process ^{175}Hf nuclide (Fig. 3), and such an effect is likely to be negligible. Effective shielding of ^{176}Lu and ^{176}Hf by ^{176}Yb also rules out an r-process effect. A p-process contribution at mass 176 is still possible but, as attested to by the very small abundance of ^{174}Hf (which accumulated for the entire age of the universe), these effects are usually very small. We therefore conclude that the ^{176}Hf excess observed in meteorites is almost certainly not inherited from nucleosynthetic anomalies.

(iii) Another possibility is a branched decay of ^{176}Lu to ^{176}Yb by β^+ emission. Amelin and Davis (2005) investigated the isotope composition of Yb in old terrestrial zircons. Even in relatively high-Lu/Yb samples, no ^{176}Yb excess could be detected and these authors concluded that 'branching decay can therefore be eliminated as the cause of the discrepancy in ^{176}Lu decay constant estimates.'

There is no known physics that would allow the rate of radioactive decay of any nuclide to vary spontaneously with time. Lundgaard et al. (2004) on the eucrite Juvinas, Bouvier et al. (unpublished data) on the eucrite Millbillillie, and Y. Amelin (pers. comm.) on both the chondrite Richardton and the achondrite Acapulco found internal Lu–Hf isochrons consistent with the 'terrestrial' value of the decay constant inferred by Scherer et al. (2001, 2005). Other eucrites produce no mineral alignments at all, which reflects that these meteorites are badly shocked. Overall, therefore, the processes responsible for the discrepancy between the 'terrestrial' and the 'meteoritic' values of the ^{176}Lu decay constant seem to belong within the first million years of the history of the Solar System.

We can conclude with another citation from Soderlund et al. (2004) that the ^{176}Lu decay constant discrepancy has 'as yet undetermined causes' and move on to introduce a different hypothesis.

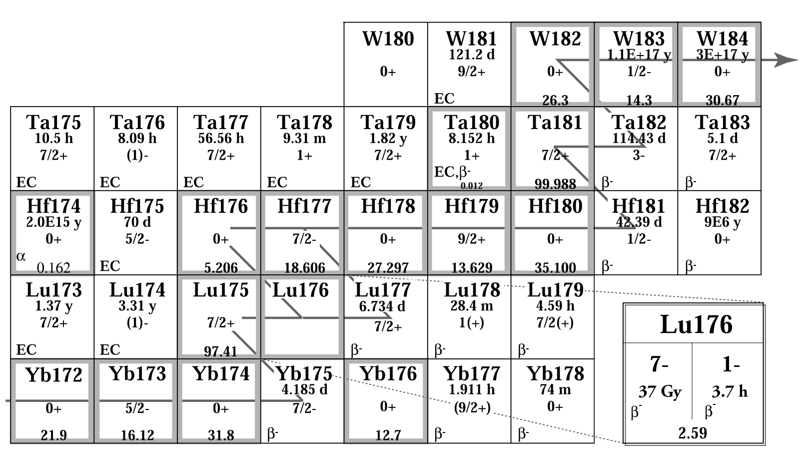

Fig. 3. The chart of the nuclides in the vicinity of ^{176}Lu. The two isomeric states of this nuclide are shown in inset with their different half-life and nuclear spin/parity configurations. Shaded cells: stable isotopes. The s-process pathway is shown as a grey line. ^{176}Lu is shielded from r-process additions by ^{176}Yb.

3. A nuclear origin of the ^{176}Hf excesses

We now examine the possibility that irradiation by photons, the source of which will be discussed in the next section, has accelerated ^{176}Lu decay during the first few million years of the existence of the Solar Nebula. The odd–odd ^{176}Lu has both a ground state and a first isomer that are unstable with respect to β^- decay. The level structure of the ^{176}Lu system is peculiar in having an exotic ground state of high spin (7^-) (Fig. 4) (Klay et al., 1991; Lesko et al., 1991). The presence of a rapidly decaying isomer next to the ground state is a rare occurrence, but its very different spin (1^-) prevents its direct decay back to the ground state. The half-life of the isomer (3.7 h) is strikingly shorter than that of the ground state (37.1 Gy or $\lambda = 1.867 \times 10^{-11}\ a^{-1}$). The isomer energy level is 122.9 keV above the ground state (Firestone et al., 1996). At stellar temperatures in excess of $2–3 \times 10^8$ K, the isomeric state becomes populated, which causes the rate of decay of ^{176}Lu to increase by ~11 orders of magnitude. This process has been proposed as a potential thermometer for helium burning in stars (Klay et al., 1991), but is irrelevant for the Solar System.

Direct electromagnetic transition to the isomer is 'forbidden' by nuclear selection rules of spin. Among the permitted transitions through levels of higher energy, one is clearly dominant (Klay et al., 1991; Lesko et al., 1991) and consists of pumping the isomer level by the gamma transition from the ground state through the lowest-lying mediating (5^-) level at 838.6 keV. The resulting short-lived isomer of ^{176}Lu promptly de-excites by β^- emission to ^{176}Hf. The half-life of the mediating level is on the order of ~10 ps (Doll et al., 1999).

This unusual decay pattern has been nicely demonstrated by laboratory experiments. Veres and Pavlicsek (1970) and Norman et al. (1985) exposed ^{176}Lu to high-energy γ-rays produced by ^{60}Co (1332 keV) and ^{24}Na (1369 keV) sources. They observed production of the short-lived ^{176}Lu isomer, which is identifiable by its decay gamma spectrum following β^--decay to ^{176}Hf, whereas irradiation by ^{137}Cs (662 keV) was ineffective. A fast decay pathway between ^{176}Lu and ^{176}Hf therefore exists. Many other such possible mediating levels exist at higher energies but their population probabilities are substantially lower.

Two nuclides of chronometric importance, ^{176}Lu and ^{182}Hf, are prone to such nuclear effects upon irradiation by ~1 MeV γ-rays. They seem to be the only nuclides for which evidence of nuclear effects can be found in the mineral isotopic record. The reason for such a rarity is that, in general, photons do not induce (γ, n) or (γ, p) reactions unless their energy exceeds the >5 MeV particle evaporation threshold (Rauscher and Thielemann, 2004) required to separate a neutron or a proton from its nucleus. The (γ, α) reaction threshold can be much lower, but in this case the photon cross-sections are far too small ($<10^{-12}$ barn) (Rauscher and Thielemann, 2004) for isotopic anomalies to be measurable. Excitation reactions to levels that decay to the ground state by internal transition have no measurable isotopic effect. The only cases for which detection is possible involves coexisting isomeric states which decay to the daughter isotope at different rates. This is the unusual case of ^{176}Lu and ^{182}Hf. Experiments have been consistently reproduced for ^{176}Lu, but the large quantities of ^{182}Hf that would be needed to demonstrate a similar effect seems to have never been assembled.

γ-irradiation of ^{176}Lu can therefore account for the excess ^{176}Hf in meteorites. Because the number of ^{176}Hf atoms created in such a process would be proportional to the amount of ^{176}Lu present at the time of irradiation, apparent isochronous relationships among meteorites

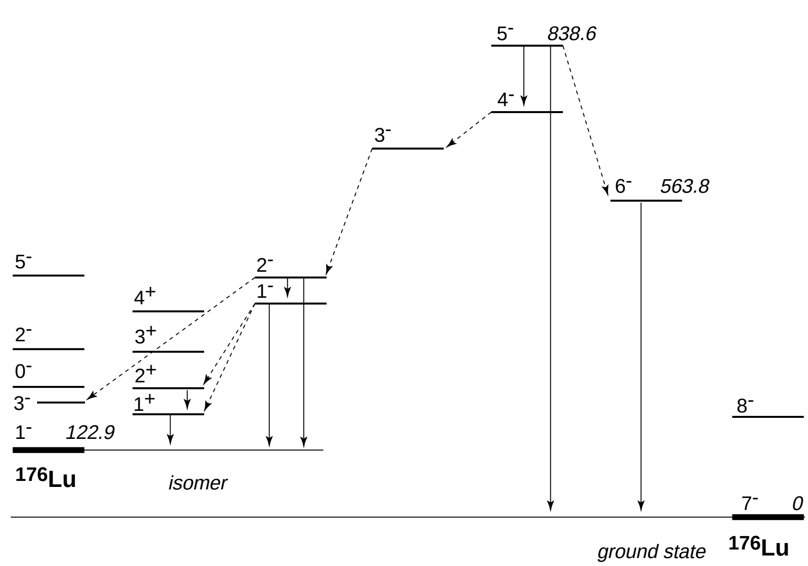

Fig. 4. The level scheme of ^{176}Lu with the ground state (right) and the isomer (left). This figure is redrawn from Klay et al. (1991) and simplified. The spin, parity, and energy (in keV) of each level are shown. Photoexcitation of the ground state to the isomer by direct transition from 7^- to 1^- requires a very large spin jump and is therefore 'forbidden'. The level 5^- at 838.6 keV is by far the most efficient mediating level (Klay et al., 1991; Lesko et al., 1991).

would be preserved albeit with slopes that reflect the compounded effect of spontaneous and γ-ray-induced radioactive decay.

4. Production of ^{176}Hf excesses by γ-ray irradiation of nebular material

To assess which natural process may be responsible for the observed ^{176}Hf excesses, photon doses must be calculated. Because silicate material and metal absorb ~1 MeV photons to only within 5 cm of the exposed surface (Berger and Hubbell, 1993), such irradiation could not have affected rocks of larger dimensions. Thus, the ^{176}Hf excesses were most likely produced prior to coalescence of the condensed dust and grains and in the immediate vicinity of the radiation source. For energies in the meV range, Gardner et al. (1988) calculated cross-sections of ~20 μbarn (or 2×10^{-33} m^2) for γ-ray excitation of the natural ^{176}Lu ground-state to the isomer. The $^{176}Hf_{excess}/^{176}Lu$ ratio of chondrites and achondrites with respect to non-irradiated material is ≈0.005 and therefore the fluence J_γ of photons with energy in excess of the reaction threshold (838.6 keV) is $0.005/2 \times 10^{-33}$ or 2.5×10^{30} photons m^{-2}.

The decay of radioactive nuclides evenly distributed within the volume of the accreting material is too weak a source of γ-rays to account for the observed ^{176}Hf excesses. Because terrestrial rocks were not affected, we only need to consider those short-lived nuclides with half-lives $<10^8$ years. The most powerful sources of γ-rays are ^{60}Fe, via ^{60}Co, to ^{60}Ni (Shukolyukov and Lugmair, 1993) ($T_{1/2} = 1.5$ My), with two photons emitted at 1332 and 1173 keV, and ^{26}Al to ^{26}Mg (Lee et al., 1976) ($T_{1/2} = 0.73$ My), with one photon emitted at 1809 keV. A canonical $^{26}Al/^{27}Al$ ratio of 5×10^{-5} corresponds to a $^{26}Al/^{176}Lu$ ratio of ~3000. A $^{60}Fe/^{56}Fe$ ratio of 5×10^{-6} would also correspond to a $^{60}Fe/^{176}Lu$ value of ~3000. Given the photoexcitation cross-section of ^{176}Lu for γ-rays, these radioactive nuclides were simply not abundant enough to account for the observed ^{176}Hf excesses.

A more plausible scenario is that the accreting material was exposed to γ-rays emitted by the young Sun. The exposure of material to irradiation by magnetic reconnection flares at the inner edge of the protosolar accretion disk has recently received much attention (Shu et al., 1997). Although the X-wind model does not explicitly consider the production of γ-rays, it is possible that the ^{176}Hf excesses are carried by silicate material that was exposed to heavy doses of such radiation. The Reuven Ramaty High Energy Solar Spectroscopic Imager (RHESSI) observed that γ-rays are indeed produced by the Sun itself (Hurford et al., 2003; Lin et al., 2003). Because electrons and protons accelerated by solar flares interact with the solar atmosphere, significant bursts of >1 MeV γ-rays accompany modern solar activity (Chupp, 1984; Murphy et al., 1987). In addition, bremsstrahlung of high-energy electrons creates photons with a continuum spectrum in the energy range of interest here, while Compton scattering of the gammas created by pion decay, de-excitation of nuclides up to Fe, and absorption of thermalized neutrons, notably in the $^1H(n,\gamma)^2H$ reaction at 2.223 MeV, also contribute to γ-ray radiation. Feigelson et al. (2002) estimated that the <1 Myr old pre-main sequence Sun exhibited X-ray flares that are ~32 times more powerful and ~320 times more frequent than the most powerful flares seen on the contemporary Sun. As in the X-wind model, chondrules and refractory inclusions, which represented a substantial proportion of the Solar Nebula material at the inner edge of the accretion disk, may have been copiously irradiated by γ-rays, possibly during the flash-heating events that produced these condensates in the vicinity of the Sun. Irradiation in the neighborhood of the young Sun was previously invoked to explain the observation of the decay products of ^{10}Be ($T_{1/2} = 1.5$ My) (Chaussidon and Robert, 1995) and was later corroborated by the correlation between $^{10}B/^{11}B$ and the B/Be ratio in Allende refractory inclusions (McKeegan et al., 2000). Likewise, Hf isotopic data may provide a potential constraint on the relative doses of γ-rays emitted by the nascent Sun. It is not clear at this point whether the ^{176}Hf excesses were introduced by refractory inclusions, by chondrules, or by a matrix rich in presolar material. Because of the opacity of the nebular material, we suggest that irradiation took place relatively close to the source.

Alternatively, γ-rays could be emitted from galactic sources located outside of the solar nebula. Modeling of the protostellar accretion disk indicates that dissipative accretion on the equatorial plane is capable of driving convection (Bell et al., 1997) and therefore provides an efficient mechanism for mixing the isotopic anomalies. Chevalier (2000) reviewed evidence that low-mass stars like the Sun formed in association with massive stars, which quickly collapse into supernovae. The existence of live ^{60}Fe (Shukolyukov and Lugmair, 1993; Tachibana and Huss, 2003) attests to the incorporation of debris from such a nearby supernova into the solar nebula (Timmes et al., 1995). Let us calculate the γ-ray fluence inferred from the ^{176}Hf excesses by assuming the production of ~1 MeV photons. The γ-ray energy release E_{GRB} of the gamma-ray bursts (GRB) associated with supernovae can reach 2×10^{51} erg (Bloom et al., 2003). This energy is not spread isotropically but beamed by narrow jets of opening angles $\theta \ll 4\pi$ steradians (Frail et al., 2001). A 'standard candle' distance d between the solar nebula and the supernova can be estimated from the fluence J_γ of γ-rays and their energy E_γ of ~1 MeV as

$$d = \sqrt{\frac{E_{GRB}}{\theta J_\gamma E_\gamma}}.$$

For a typical value $\theta = 5°$, we obtain $d = 2 \times 10^{14}$ m = 0.006 parsec (pc).

For a single explosion, this is probably two orders of magnitude 'too close' and the solar nebula would have been blown off by the momentum of the exploding star

(Chevalier, 2000). Uncertainties on such calculations remain, however substantial. In particular, the value of the fluence J_γ depends on the poorly constrained cross-section of the photoexcitation reaction, and the energy of the supernova explosion may fall outside the range compiled by Bloom et al. (2003). It is also known that, in particular domains of nebular clouds thought to be good analogs of the Solar System birthplace (Hester et al., 2004), the density of stars and therefore of potential supernovae may be very high. For example, the Orion nebula may locally contain up to 20,000 stars per cubed pc (Hillenbrand and Hartmann, 1998). We therefore, consider that supernovae clusters may have provided a γ-ray fluence adequate to account for the observed ^{176}Hf excesses in meteorites.

5. The survival of Hf isotope heterogeneities in the solar nebula

The eucrite–chondrite array implies that, 4.56 Ga ago, Hf was isotopically heterogeneous within the solar nebula with ^{176}Hf/^{177}Hf ratios correlated with the ^{176}Lu/^{177}Hf ratio. As discussed by Blichert-Toft et al. (2002), the isotopic heterogeneities observed in these meteorites are inherited from their mantle sources. This is true even for the cumulate eucrites, in which the unusually broad range of Lu/Hf and Sm/Nd ratios cannot be assigned to the effect of crystal fractionation alone. An outstanding question is why further planetary processes were inefficient at resetting what seems to have remained a mixing array and why complete Hf isotopic homogenization was not achieved in the parent bodies. The various chondrite parent bodies (H, L, LL, E, and carbonaceous) were too small or formed too late to have been extensively melted, and in these cases the persistence of isotopic heterogeneities is not an issue. The planetary values are not well known for either Mars, the Moon, or the Earth, and we can only speculate about whether Hf isotopic homogeneity was ever achieved.

The Hf isotopic properties of the eucrite parent body are more difficult to explain. A first explanation is that convection in the parent body (Vesta?) was too slow for isotopic homogenization to occur. For a planet with a ~540 km radius, gravity will be a factor of ~20 smaller than on the Earth, the mantle will be thinner by a similar factor, and the Rayleigh number for internal heating (e.g., Schubert et al. (2001)),

$$Ra = \frac{\alpha \rho g h^5 H}{\nu \kappa}$$

(in which α is the thermal expansion coefficient, ρ the density, g gravity acceleration, h the depth of the mantle, H radioactive heat production, ν the viscosity, and κ the thermal diffusivity) should be 7–8 orders of magnitude smaller than for a planet the size of the Earth. Even if the planet started out partially molten, convection should have stopped very rapidly. Unless melting is very extensive, mantle convection cannot, therefore, erase the primordial isotopic heterogeneities at the scale of a small planet. Likewise, 'short-range' petrological processes, such as melt migration by porous flow or dyke injection (clearly important in the eucrite parent body (Barrat et al., 2000)), are unlikely to provide an efficient mechanism for the wholesale isotopic homogenization of small to medium planetary bodies.

A complementary factor for the survival of early Hf isotopic heterogeneities appeals to the slow diffusion of Hf^{4+} (the dominant ionic species in minerals) in geological material. It is remarkable that among all the available long-lived chronometers, with the exception of U–Pb in zircons, that of Lu–Hf is the most difficult to reset without recrystallizing the rocks (Scherer et al., 2000; Bedini et al., 2004). The diffusion coefficients of Hf^{4+} in solids have not been measured, except for zircon $ZrSiO_4$ in which it is six orders of magnitude smaller than that of the trivalent rare-earth elements (Cherniak et al., 1997). The crystal chemistry model of Van Orman et al. (2001) predicts that the diffusion coefficients of 4+ ions in diopside are almost three orders of magnitude smaller than those of 3+ ions. We therefore predict that the isotopic homogenization of Hf would be significantly more difficult to achieve than for other radiogenic isotopes such as divalent Sr and Pb and trivalent Nd and Lu.

If the proposed irradiation model applies, the Hf isotope compositions of early planetary rocks provide a measure of the dose of high-energy radiation they have received. We assume that the value of the initial ^{176}Hf/^{177}Hf ratio in Solar System material prior to irradiation by γ-rays corresponds to the intercept of the linear chondrite–eucrite array in a ^{176}Hf/^{177}Hf versus ^{176}Lu/^{177}Hf diagram (i.e., the Lu-free phase). We recalculated this ratio to be 0.279647 ± 69 by fitting a least-squares straight line through the most recent chondrite and achondrite data (Fig. 5), which is indistinguishable from the value of Bizzarro et al. (2003). As discussed above, this linear chondrite–eucrite array is not an isochron. Precursor materials of chondrites and eucrites residing in the inner protoplanetary disk likely had homogeneous Hf isotope compositions, but variable Lu/Hf as a result of thermal fractionation.

After irradiation, such materials would produce a positive slope on a Lu–Hf isochron plot, with the *Y*-intercept indicating the initial Hf isotope composition of the non-irradiated material. The initial ^{176}Hf/^{177}Hf ratio is clearly independent of the decay constant and, for all practical purposes, the Lu–Hf ages of eucrites and chondrites should be indistinguishable. Fig. 5 shows the theoretical 4.56 Ga isochron for non-irradiated material together with the chondrite and eucrite data and the Bulk Silicate Earth. The mean ^{176}Hf/^{177}Hf for the Bulk Silicate Earth (0.282854 ± 3, 2s standard error) is given by the value of the ^{176}Hf/^{177}Hf versus ^{143}Nd/^{144}Nd correlation for more than 2000 terrestrial basalts (mantle array) at the relatively well-constrained chondritic ^{143}Nd/^{144}Nd value. The center of this Hf–Nd mantle array is about three parts per 10^4 above the mean ^{176}Hf/^{177}Hf value of meteorites (Blichert-Toft and Albarède, 1997). On the same diagram, our

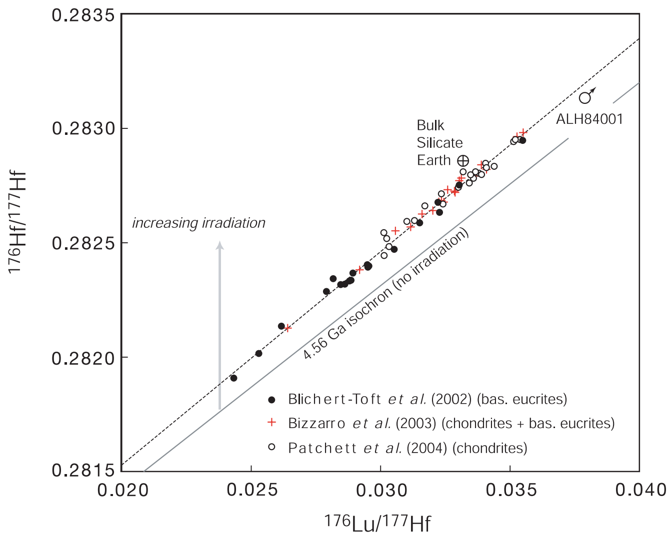

Fig. 5. Lu–Hf isochron plot of chondrites (Bizzarro et al., 2003; Patchett et al., 2004) and basaltic eucrites (Blichert-Toft et al., 2002; Bizzarro et al., 2003). Error bars are slightly larger than the symbol size. The dotted line is a linear fit through the chondrite and eucrite data and indicates a Lu–Hf age of 4.75 Ga. The solid 4.56 Ga isochron of non-irradiated material is calculated from the intercept (0.279647 ± 69) of the dotted line using the 'terrestrial' decay constant (Scherer et al., 2001; Scherer et al., 2003; Soderlund et al., 2004) of $1.867 \times 10^{-11}\ a^{-1}$. The excess ^{176}Lu decay observed in meteorites is ascribed to irradiation by γ-rays of early condensates in the solar nebula. The ^{176}Hf/^{177}Hf ratio of the Bulk Silicate Earth is calculated by regressing over 2000 Nd–Hf isotopic data of terrestrial basalts and assuming a chondritic ^{143}Nd/^{144}Nd value of 0.512638. The ^{176}Lu/^{177}Hf of the Bulk Silicate Earth is assumed to be identical to the mean ratio of chondrites (0.0332) (Blichert-Toft and Albarède, 1997; Patchett et al., 2004). Unpublished data for ALH84001 by Blichert-Toft are shown for reference: this ~4.56 Ga-old Martian meteorite (Jagoutz et al., 1995) represents the primordial lithosphere of Mars. Its position above the isochron of non-irradiated material indicates that at least some of the precursor material of Mars was also irradiated by γ-rays.

unpublished datum for the meteorite ALH84001 plots slightly below the meteorite array but still above the isochron of non-irradiated material. This sample is unique among Martian meteorites in that its Sm–Nd age is 4.56 Ga (Jagoutz et al., 1995). We speculate that the γ-ray fluence received by the material precursor to different planetary objects varied from place to place but the ranges apparently overlap. We see no systematic relationship to the distance to the Sun and there is clearly room for a common $^{176}Hf_{excess}/^{176}Lu$ value.

The answer to the question of whether irradiation varied across the nebula probably lies with the measurement of the ^{176}Lu/^{175}Lu ratios of a variety of planetary objects. Variable ratios would indicate a variable γ-ray fluence across the Solar System, while constant ratios would hint at a constant fluence and support a supernova-type rather than solar source of γ-rays situated outside of the Solar System. Scherer et al. (2005) measured the ^{176}Lu/^{175}Lu ratios of terrestrial and lunar rocks, chondrites, eucrites, and Allende CAI and found no resolvable (i.e., at the 0.1% level) isotopic differences among these samples. If the ^{176}Hf excesses were created by photoexcitation of ^{176}Lu, the 'burn-out' of this nuclide seems to be homogeneous across the accessible Solar System, which is probably in favor of a distant (GRB) rather than proximal (Sun) source of γ-rays.

6. A note on the ^{182}Hf–^{182}W chronometer

Hafnium-182 is a nuclide of considerable chronological importance which, in particular, provides a time scale for planetary core formation (Kleine et al., 2002; Yin et al., 2002). Next to its ground state (0^+), it also has an isomer (8^-) with a half-life much shorter (62 min) than that of the ground state (8.9×10^6 year) (Vockenhuber et al., 2004). The gap between the energy levels of the two states is larger (1173 keV) than in the case of ^{176}Lu and the difference in nucleus configuration and spin is very large. Both conditions are unfavorable to a nuclear transition. Unfortunately, neither cross-sections for photoexcitation nor possible mediating levels are known for ^{182}Hf. Nevertheless, ^{182}Hf photoexcitation should still be considered and there is a real possibility that irradiation by γ-rays may affect to some extent the ages of Hf/W fractionation events, notably those associated with core segregation in planetary bodies.

7. Conclusions

We considered that, so far, all existing models have failed to explain why the Lu–Hf age of meteorites is ~4% too old. We suggest that the discrepancy between the values of the ^{176}Lu decay constant obtained from meteorites and well-dated terrestrial rocks may be accounted for by the exposure of the nebular gas and dust to γ-rays. Such irradiation populates the fast-decaying isomeric level of ^{176}Lu and creates variable ^{176}Hf excesses in silicates that are proportional to the amount of ^{176}Lu present. The source of the γ-rays can be the young Sun itself or, perhaps more likely, one or several nearby supernovae exploding in the neighborhood of the solar nebula.

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Associate editor: Yuri Amelin

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