Problems with the γ-ray spectrum in the Fleischmann *et al.* experiments

SIR—Fleischmann, Pons and Hawkins[1] recently announced the observation of significant heating in their cold-fusion experiments, a result that they attribute to copious fusion reactions. As compelling evidence that fusion had occurred, they reported the observation of the 2.22-MeV γ-ray line that originates from neutron capture by hydrogen nuclei[2,3]

$$n + p \rightarrow d + \gamma\ (2.22\ MeV) \qquad (1)$$

(Here d represents a deuteron.) They contend that the neutron in reaction (1) is generated by the reaction

$$d + d \rightarrow n + {}^3He \qquad (2)$$

and conclude, therefore, first that the 2.22-MeV γ-ray confirms that the fusion process (2) is occurring, and second that a neutron production rate of the order of 4×10^4 neutrons s^{-1} is derivable from their γ-ray signal rate. They further state that most of the heat generation occurs not through process (2), but through a hitherto unknown nuclear-fusion process.

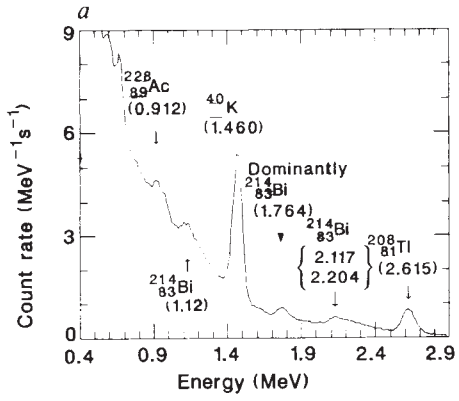
Here we focus solely on the identity of the reported γ-ray line, which we shall henceforth call the signal line. We argue that the claim of Fleischmann *et al.* to have observed the 2.22-MeV line characteristic of reaction (1) is unfounded. We do so on the basis of three quantitative considerations: (1) that the linewidth is a factor of two smaller than their instrumental resolution would allow; (2) that a clearly defined Compton edge[4], which should be evident in their published data at 1.99 MeV, is not in fact present; and (3) that their estimated neutron production rate is too large by a factor of 50. In addition, from a consideration of the terrestrial γ-ray background, we argue that their purported γ-ray line actually resides at 2.5 MeV rather than 2.22 MeV. These conclusions are, in part, based on our studies of neutron capture by hydrogen, using a neutron source submerged in water. These measurements allow us to compare the results of Fleischmann *et al.* directly with a controlled experiment.

We measured terrestrial γ-ray background spectra in order to compare our detector characteristics with those of Fleischmann *et al.* Figure 1*a* shows a typical terrestrial γ-ray background spectrum obtained with a 3 in. × 3 in. NaI(Tl) crystal spectrometer system (see ref. 5 for details). The main features of the background spectrum are quite similar throughout the terrestrial environment[6,7]. Fleischmann *et al.* showed a similar γ-ray spectrum on television (Fig. 1*b*). (We believe that we have viewed all the cold-fusion γ-ray spectra that have been shown on KSL-TV (Utah) up to 19 April. This information was obtained from Utah News Clips, Inc., Utah. As far as we can tell, all spectra are identical to that of Fig. 1*b*.) This spectrum was obtained in the course of the Fleischmann *et al.* experiments (M. Hawkins, personal communication). Their spectrometer system consisted of a Nuclear Data ND-6 portable analyser with a 3 in. × 3 in. NaI(Tl) crystal (ref. 1 and M. Hawkins and R. Hoffmann, personal communications). A ⅜-in.-thick Pb annulus encompassed the scintillator. It is clear from Fig. 1*a* and *b*, and particularly from the ^{40}K (1.46 MeV) and ^{208}Tl (2.61 MeV) lines, that our resolution is comparable to or better than that of the spectrometer used by Fleischmann *et al.*, a point to which we return later.

In the interval 1.46–2.61 MeV, the energy resolution of a NaI(Tl) spectrometer, which determines the γ-ray linewidth, can be well described by the formula[8,9]

$$R(E) = \frac{\Delta E}{E} \approx R(E_n)\ \sqrt{E_n/E} \qquad (3)$$

Here ΔE is the full width at half maximum (FWHM) of the line, E is the energy of the photon and $R(E_n)$ is the measured 'reference' resolution at energy E_n. $R(E_n)$ can be accurately determined using a ^{60}Co source (that is, the ^{60}Co line at 1.33 MeV), or it can be fairly well approximated by the ^{40}K decay line at 1.46 MeV. (From Fig. 1*b*, the ^{40}K decay line allows one to estimate Fleischmann *et al.*'s resolution as ~ 8%.)



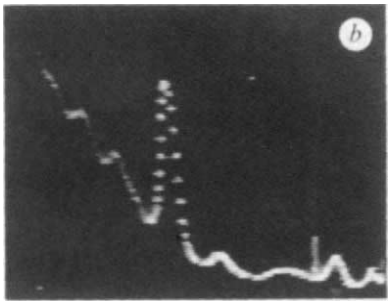


FIG. 1 *a*, The γ-ray background spectrum measured with a 3 in. × 3 in. NaI(Tl) detector at MIT. Some important terrestrial γ-ray lines have been identified in this figure[6,7,12]. (As explained in ref. 12, the immediate parent of the final decay product is identified. For example, ^{40}K β^+ decays into an excited nuclear state of ^{40}Ar, which actually then emits the 1.460-MeV photon discussed in the text.) The spectrum is averaged over an 84-hour run. *b*, The γ-ray spectrum shown on television by Fleischmann *et al.* The main characteristics of the two spectra are similar; one can also tell that the two detectors have comparable spectral resolution. In *b*, note the curious structure at about 2.5 MeV and that beyond the ^{208}Tl peak (2.61 MeV), which appear to be artefacts. (The spectrum can also be obtained from KSL-TV in Utah (M. Hawkins, personal communication).)

TABLE 1 Comparison of energy resolutions of the γ-ray spectrometers

(*a*) *Resolution of MIT spectrometers*

Energy (MeV) / Origin	1.17 ^{60}Co	1.33 ^{60}Co	1.46 ^{40}K	2.22 n(p,γ)d	2.61 ^{208}Tl
Natural background			0.055		0.043 (0.041)
^{60}Co	0.056	0.051			
Pu/Be neutron source				0.05 (0.045)	

(*b*) Resolution of the Fleischmann *et al.* spectrometer[1]

Energy (MeV) / Reference	1.33 ^{60}Co	1.46 ^{40}K	2.22 n(p,γ)d	2.61 ^{208}Tl
Hoffman*	0.056	0.065		
TV news†		~0.08		~0.05 (0.049)
Ref. 1 (errata)			0.025 (0.053)	

The resolution is defined as the full width at half maximum (FWHM) divided by the peak energy. Numbers in parentheses are predicted values based on the detector resolution at 1.46 MeV (see text). In *b*, the prediction is based on the resolution value (0.065 at 1.46 MeV) provided by R. Hoffman (personal communication).
*R. Hoffman (personal communication).
†Derived from images of the televised news broadcasts.

Table 1 lists the resolution data for our detectors and for that of Fleischmann *et al.*.

We now compare the signal line of Fleischmann *et al.* (Fig. 1*a* of ref. 1 (errata), shown as Fig. 2 here) with our measured spectrum obtained from the experiments on neutron capture by hydrogen (Fig. 3 here, and Fig. 4 of ref. 5). In these experiments, a Pu/Be neutron source was placed in a water tank. $^{239}_{94}$Pu emits energetic α-particles, which produce neutrons through (α, n) reactions with Be (refs 4,9). The neutrons are thermalized in water, and we observe the emitted neutron-capture γ-rays with our spectrometers. The measured resolution at 2.22 MeV is ~ 5% (Table 1*a*), and is reasonably well predicted by equation (3). As a consequence, this calls into immediate question the identity of Fleischmann *et al.*'s signal line as a γ-ray line. Specifically, Fig. 2 shows the signal line to have a resolution of 2.5%. This is about a factor of two smaller than that predicted by equation (3) on the basis of the known resolution (Table 1*b*) from either the ^{40}K decay line (1.46 MeV) or from the ^{60}Co source (1.33 MeV) (R. Hoffman, personal communication). But we know from Table 1 that the spectrometer used by Fleischmann *et al.* has a resolution that is at best comparable to our own for the entire region from 1.46 to 2.61 MeV (see also Fig. 1), so it is inconsistent that their linewidth at 2.22 MeV is a factor of two below the predicted value.

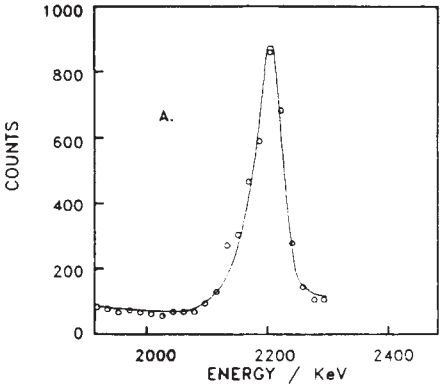


FIG. 2 A reproduction of the purported 2.22-MeV γ-ray signal line of Fleischmann *et al.* (Fig. 1*a* of errata to ref. 1). The resolution, based on the linewidth, is about 2.5%. With such resolution, one would expect to see a clearly defined Compton edge at 1.99 MeV. No edge is evident. Also, a resolution of 2.5% is inconsistent with their spectral resolution. Furthermore, we argue that the signal line may reside at 2.5 MeV, not at 2.22 MeV as is claimed by Fleischmann *et al.* and depicted here.

There is a second crucial inconsistency with the published signal line (Fig. 2). If we assume a resolution of 2.5% at 2.22 MeV, then there should be a clearly defined Compton edge[4] at 1.99 MeV. For example, in Fig. 3 the Compton edge is evident even for our measured resolution of only 5%. For a resolution of 2.5%, the definition of the Compton edge would be distinctly sharper. The lack of a Compton edge at 1.99 MeV for the signal line therefore negates the conclusion of Fleischmann *et al.* that they have observed the 2.22-MeV γ-rays from neutron capture by hydrogen.

We also point out that in our (Pu/Be) neutron-capture experiments, a conspicuous $e^+ - e^-$ annihilation single-escape peak exists at 1.71 MeV (Fig. 3), as well as a double-escape peak at 1.20 MeV. (The full spectrum from the Pu/Be experiment, as well as the background spectrum, can be found in ref. 5.) Such features unambiguously identify the primary γ-rays as having an energy of 2.22 MeV, and are a necessary consequence of the physical processes of detection of γ-rays in a finite-sized NaI scintillator.

Based independently on both their γ-ray and neutron measurements, Fleischmann *et al.* claim to have observed a neutron production rate of ~ 4×10^4 neutrons s^{-1} (ref. 1). This claim is clearly inconsistent with their γ-ray signal line, for the following quantitative reasons. The Pu/Be neutron source used in our experiment is absolutely calibrated to within 10% of 1.5×10^6 neutrons s^{-1} (ref. 10 and MIT Reactor Radiation Protection Office). In obtaining the data in Fig. 3, we used an experimental setup similar to that of Fleischmann *et al.* (ref. 1; televised broadcasts; and M. Hawkins and R. Hoffman, personal communications). Our Pu/Be source was submerged 6 in. into a large water tank. The rate at the 2.22-MeV peak, after subtracting the background continuum, is about 1.4×10^3 MeV^{-1} s^{-1} (see Fig. 3). Scaling this rate to a neutron source of 4×10^4 neutrons s^{-1} (the level given by Fleischmann *et al.*), and integrating over the linewidth, gives a total 2.22-MeV γ-ray rate of about 4.5 counts per second. This value is a factor of 50 times higher than the rate that would be calculated on the basis of the results in Fig. 1*a* (that is, 0.081 counts per second). (Fleischmann *et al.* state that their neutron count rate is measured with a BF_3 neutron counter over a 0.4 mm × 10 cm Pd cell, and that the γ-ray measurement is over a 0.8 mm × 10 cm Pd cell[1]. If the total reaction rate is proportional to the volume of Pd rod, as they state, the inconsistency in the reported neutron rate is by a factor of 200 rather than 50.) While differences in rates of a factor of two might possibly be explained by geometrical considerations, a factor of 50 is inexplicable.

A further point concerning the identification of the signal line is the precise value of the energy at which the peak occurred. From Fig. 2, the background in the neighbourhood of the peak is seen to be ~ 80 counts per channel, a level that corresponds to ~ 400 counts per channel for a 48-hour accumulation time (the data in Fig. 2 were accumulated for a period of 10 hours[1]). On the other hand, in the Utah measurements of terrestrial γ-ray background, the level in the vicinity of the 2.22-MeV feature was found to be ~ 4,000 counts per channel (R. Hoffman, personal communication). The only relevant part of the entire γ-ray spectrum (between 1.46 and 2.61 MeV) in which the background was as low as 400 counts was at an energy in the vicinity of 2.5 MeV (R. Hoffman, personal communication). Thus, we argue that the peak in the spectrum shown in Fig. 2 may be at 2.5 MeV, not at 2.22 MeV.

The importance of properly identifying the energy of the feature claimed by Fleischmann *et al.* can hardly be overemphasized. Thus, it is extremely unfortunate that they chose to display only the energy range 1.9–2.3 MeV in their pub- ▶

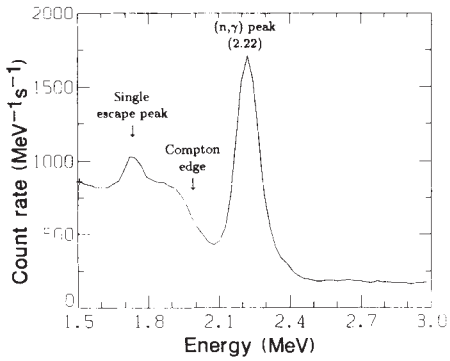


FIG. 3 The γ-ray spectrum measured by a 3in. × 3in. NaI(Tl) spectrometer during a neutron-capture-by-hydrogen experiment using a (Pu/Be) neutron source submerged in water. Because of the finite size of the crystal (which is identical to that of Fleischmann *et al.*[1]), we also see an escape peak[2-4] and, of particular importance here, the Compton edge[4]. In this figure, the digitization energy width is 0.024 MeV per channel. The full Pu/Be and background spectra are shown in Fig. 4 of ref. 5.

1. Fleischmann, M., Pons, S. & Hawkins, M. *J. electroanalyt. Chem.* **261**, 301–308 (1989); and errata.
2. Hamermesh, B. & Culp, R. J. *Phys. Rev.* **92**, 211 (1953).
3. Greenwood, R. C. & Black, W. W. *Phys. Lett.* **6**, 702 (1966).
4. Knoll, G. F. *Radiation Detection and Measurements* (Wiley, New York, 1979).
5. Petrasso, R. D. *et al.* MIT Plasma Fusion Center Report PFC/JA-89-24 Rev. (1989).
6. Eisenbud, M. *Envir. Radioactivity* (Academic, New York, 1973).
7. Adams, J. A. S. & Lowder, W. M. *The Natural Radiation Environment* (Univ. of Chicago Press, 1964).
8. *Harshaw Radiation Detectors Scintillation Counting Principles*, Solon, Ohio, 44139 (1984).
9. Crouthamel, C. E. *Applied Gamma-Ray Spectrometry* 2nd edn (eds Adam, F. & Dams, R.) (Pergamon, Oxford, 1970).
10. Reilly, W. F. thesis, Massachusetts Institute of Technology (1959).
11. Koonin, S. E.; Bailey, D. C. *Am. phys. Soc. Meet.* Special Session on Cold Fusion, Baltimore, Maryland, 1–2 May (1989).
12. Lederer, M. C., Hollander, J. M. & Perlman, I. *Table of Isotopes* 6th edn (Wiley, New York, 1967).

ACKNOWLEDGEMENTS. We thank V. Kurz, J. S. Machuzak, F. F. McWilliams and Dr S. C. Luckhardt. For the use of a spectrometer system, we thank Professor G. W. Clark. For discussions, we thank M. Hawkins and R. Hoffman of the University of Utah. For suggestions and criticisms, we are grateful to Dr G. R. Ricker Jr and Professor D. J. Sigmar. We are indebted to J. K. Anderson for assembling this document. For locating important references, we thank K. A. Powers. Supported in part by the US Department of Energy.

lished Fig. 1*a*, thereby not providing the supporting evidence of the ^{40}K (1.46-MeV) and ^{208}Tl (2.61-MeV) features which must be present in their spectra in order for their identification to be correct.

Therefore, although Fleischmann *et al.* may have observed a change in their γ-ray spectra that bears some relation to detector location, we conclude that it is unrelated to the 2.22-MeV neutron-capture γ-rays, and that it is also unrelated to the background $^{214}_{83}Bi$ line (2.20 MeV; Fig. 1*a*), as has been suggested elsewhere[11]. We can offer no plausible explanation for the feature other than it is possibly an instrumental artefact, with no relation to a γ-ray interaction.

R. D. PETRASSO
X. CHEN
K. W. WENZEL
R. R. PARKER
C. K. LI
C. FIORE

Plasma Fusion Center
Massachusetts Institute of Technology
Cambridge,
Massachusetts 02139, USA

Fusion in from the cold?

SIR—Recent experiments involving the loading of deuterium (D) into palladium[1] via a 0.1 M $LiOD/D_2O$ electrolyte may have created something not encountered in the absence of current flow. The voluminous body of older work on electrolytic D–Pd loading is based on thin wires or foils of high aspect ratio which allow only small voltage differentials. But the reported chemical potential across a thick deuterated palladium negative elctrode could give rise to metastable solid phases alien to the existing equilibrium Pd–D phase diagram. The theoretical existence of palladium analogues to hydrogen-rich[2] compounds like Li_2ReH_9 is as tantalizing as the limit of solid solubility of D in Pd is finite. And beyond PdD_3 (by analogy to the known compound Li_3Pd) the thermodynamic distinction between the higher palladium deuterides and metallic deuterium grows dim. The mechanism of their formation might involve the intersection of high deuteron mobility grain boundaries with the Cottrell clouds of H and D that decorate dislocations in many transition metals.

A small mass of (mostly) deuterons transforming into molecular deuterium as they recapture electrons previously delocalized into the S- and D-bands of palladium could release roughly 1 MJ mol^{-1}. Apart from explaining the electrode meltdown and vaporization reports[1] (although this could be due to lithium's remarkable ability to lower the melting point of palladium), this could raise local temperatures to very high values.

Could the so-called 'cold fusion' environment in fact involve local temperatures of greater than 10^5 K generated by the detonation of deuteron clusters (R. G. Gordon, personal communication) or of a metallurgical precipitate, coarse or fine-grained, of a high D/Pd ratio intermetallic compound inside a deuterated palladium electrode? The energy of reassociation of electrons and deuterons is roughly 1 Rydberg (13.6 eV) minus the work-function of Pd (4.9 eV); to this must be added the roughly 4.7 eV liberated when two deuterium atoms pair. Obviously, these energies must be scaled down to compensate for the difference between reactions in free space and the solid state. Nevertheless, the 20 eV energy of formation of D_2 from deuterons could thus produce hot and highly compressed deuterium plasma bubbles of small (>0.01μm–<100μm) size. As to the objection that the surrounding cool metal will quench these bits of pale fire in a nanosecond or so, I believe it answers a serious question: how come the Fleischmann and Pons[1] claimed neutron yield is 9–14 orders of magnitude short of their claimed heat flux? At present, one can only speculate as to by how many orders of magnitude a reflected spherical shock front might raise the temperature and pressure of the D-plasma.

There is another ramification to the notion of nano-novas flashing out of per-deuteride grains decorating the grain boundaries of electrochemically over-wrought palladium nagative electrodes. The explosion of such precipitated grains will, above a critical radius, generate cracks in the adjacent metal. It has been postulated (G. Chapline, personal communication) that the surfaces of such cracks, as they open, could host a field gradient or a propagating array of plasmons down whose wake field a deuteron could accelerate. This presumes that the bulk Pd–D system has reached 1:1 stoichiometry and thus been restored to long-range order; this condition is very far from the disorderly and anharmonic state of Pd–D during the early stages of electrolytic D-loading. In this model some thousands of unit cells of travel would suffice to yield keV deuterons. So the propitiated shade of Rutherford may yet countenance not-so-cold fusion.

It has been noted in a News and Views article in the 27 April issue[3] that neutrons have been observed when cracks are generated in crystals of lithium deuteride[4].

RUSSELL SEITZ

Center for International Affairs,
Harvard University, Cambridge,
Massachusetts 02138,
USA

1. Fleischmann, M. & Pons, S. *J. electoanalyt. Chem.* **261**, 301–308 (1989).
2. Shaw, B.L. *Inorganic Hydrides*, 99 (Pergamon, Oxford, 1967).
3. Cohen, J.S. & Davies, J.D. *Nature* **338**, 705–707 (1989).
4. Klyuev, V.A. *et al. Sov. tech. Phys. Lett.* **12**, 551 (1986).

Mössbauer cancer therapy doubts

SIR—Mills *et al.*[1] present data to support the view that a dose of 10^{-5} Gy of 14.4-keV X-rays can ablate a population of malignant cells containing $^{57}Fe(III)$ · bleomycin. They suggest that such a regime may have the potential for the low-dose sterilization of superficial human tumours.

This is unlikely on simple physical grounds, basically because only a small fraction of the exposed cells will have received any energy deposition at all. The proportion of cells that would receive one or more energy depositions, assuming the statistical independence of such events, is obtained from the Poisson distribution and is $(1 - e^{-n})$; here n, the average number of energy depositions occurring in the sensitive site, is given by D/z_{1F}, where D is the absorbed dose and z_{1F} is the so-called 'frequency-averaged specific energy per event' — or simply, the mean energy per unit mass deposited by single energy deposition events in the sensitive site in the cell[2]. This mean specific energy will be similar, irrespective of whether the photon is ultimately absorbed in a photoelectric event or in a Mössbauer absorption (it would be slightly larger in the latter case); it has been measured for 12-keV photons in a tissue-equivalent material in spherical sites of various volumes[3].

Appropriate volumes for consideration are those of typical human cell nuclei (100–1,000 μm^3)[4] or, perhaps more relevantly, the volume of nucleotides in the mammalian nucleus (~3 μm^3)[5]. For volumes of 250 and 3.5 μm^3, for which measurements have been made[3], the corresponding values of z_{1F} are 4×10^{-3} Gy and 0.14 Gy. These numbers yield a probability of about 2.5×10^{-3} that a 250 μm^3 volume of cell nuclei will be subject to at least one energy deposition, and a corresponding probability of 7×10^{-5} for a 3.5 μm^3 volume of nucleotides. Thus, only about 1 in 400 cells (250 μm^3 volume) or 1 in 14,000 cells (3.5 μm^3 volume) would receive any energy deposition at all if exposed to a dose of 10^{-5} Gy.

To sterilize even a small tumour containing about 10^8 cells requires an appropriately small probability (less than 10^{-9}) that any cell will receive no energy-deposition events. Thus, an average number of energy depositions per cell of

1. Mills, R.L. *et al. Nature* **336**, 787–789 (1988).
2. *Microdosimetry ICRU Rep.* 36 (ICRU, Bethesda, 1983).
3. Kliauga, P.J. & Dvorak, R. *Radiat. Res.* **73**, 1–20 (1978).
4. Altman, P.L. & Katz, D.D. (eds) *Cell Biology* (FASEB, Bethesda, 1976).
5. Brenner, D.J. *Radiat. Envir. Biophys.* **27**, 189 (1988).
6. Goodhead, D.T., Thacker, J. & Cox, R. *Phys. med. Biol.* **26**, 1115–1127 (1981).