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Research Article

On the connection between K_α X-rays and energetic alpha particles in Fleischmann–Pons experiments

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Abstract

X-ray emission at K_α energies has been reported in Fleischmann–Pons experiments, and alpha particle emission has been reported in others. It is possible for energetic alpha particles to result in K_α radiation following impact ionization. As a result, one might imagine that K_α radiation is a signature of energetic ions in these experiments. We have calculated yields for K_α X-rays as a function of the energetic alpha particle energy in PdD. As a result of these calculations, we conclude that it is unlikely that these X-rays are a result of energetic alpha particles. We note that energetic alpha particles can produce excitation in lithium at 478 keV, and that the relative line strength of the Pd K_α, the Pt K_α and the Li 478 keV line can be used as a consistency check for energetic alpha particles.

Keywords: Fleischmann–Pons experiments, K-alpha emission, Gamma-emission, 478 keV line of ^{7}Li

1. Introduction

Spectroscopy is a key diagnostic in a variety of fields, including plasma physics, astrophysics, chemistry, and materials science. If a system is in thermal equilibrium, then it is may be possible to determine the plasma temperature from the relative intensity of different emission lines. At low density the atomic and ionic levels may not be in equilibrium, but in some cases it possible to determine both the temperature and electron density from the relative strengths of strong emission lines.

Spectroscopy has so far not played a particularly important role in Fleischmann–Pons experiments, which is unfortunate since it is such a powerful tool. There have been reported observations of K_α radiation from Pd [1] and from Pt [1,2] in Fleischmann–Pons experiments. Such observations are important because they give us the possibility of learning new things about the physical mechanisms behind the anomalies.

At present we do not understand why such radiation should occur. One possibility is that they are produced as a result of impact ionization from energetic particles. If so, then the X-ray emission could be considered as a secondary

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process which can be used as a diagnostic for the presence of energetic particles. Alternatively, it may be that X-ray emission occurs for some other reason, in which case it would be of interest to understand better what energetic process could result in the production of K_α emission.

For other reasons we have examined issues associated with nuclear radiation in general that would be expected in energetic alpha particles were present in large quantities in the Fleischmann–Pons experiment. It had been conjectured that the excess heat comes about from new reactions which result in energetic alpha particle emission in a way such that the energetic alpha particles are "hidden" deep within the PdD. We calculated the yields for a variety of reactions which would result in neutron, X-ray and γ-ray production. From a comparison with experiments in which excess heat was observed while nuclear emissions were being detected, we were able to conclude that the upper limit on the alpha particle energy must be less than 10 keV in order to be consistent with experiment.

As a result of this exercise, it became clear that spectroscopic diagnostics were available for energetic alpha particles. In particular, energetic alpha particles should produce K_α X-rays as well as the 478 keV line in ^{7}Li. Moreover, it should be possible to develop an estimate for the energy from measurements of the ratios of the different transitions. Since energetic alpha particles would also produce neutrons, one would expect a correlation between all of the different signals.

Now, there are experiments in which energetic alpha particles have been reported [3,4]. There are also experiments in which neutrons and X-rays have been reported. Previously we were interested in the correlation between excess power and nuclear radiation. Here, we are instead interested in the possible correlation between different low-level X-ray, gamma-ray and neutron emission. In what follows we will discuss results for yields of X-rays and γ-rays from energetic alpha particles. Based on these calculations, and from the few available experimental results, we conclude that the K_α radiation is not due to energetic alpha particles. Some other mechanism should be sought to account for them.

2. Impact ionization and K_α emission

X-ray emission resulting from bombardment by energetic ions is a subject that has been well studied for many years. In the case of alpha particle-induced K-shell X-ray emission, we can make use of the measurements of Wilson et al. [5], which is available in a relevant energy range for Rh and Ag (which are close to Pd).

To obtain results for Pd, we could interpolate data taken for nearby elements. There is considerable literature on the closely related problem of scaling laws for impact ionization of K-shell electrons [6]. The basic idea is that there exists an underlying scaling law that depends on a universal function, from which K-shell impact ionization cross sections for many elements can be obtained over a wide range of energies [7].

2.1. Universal function

To proceed, we would like to take advantage of ideas based on the notion that there exists a universal function for alpha particle-impact K-shell ionization. Certainly there is considerable literature both of a theoretical nature as well as experimental devoted to this problem. There are a number of specific universal functions which have been put forth based on different data sets or theoretical models.

For our computations in particular, we have the problem that the data sets which are close to Pd do not go up to high enough energy for our application. Hence our interest is in making an empirical extension of the model in which we used scaled experimental data in the higher energy regime as a way to predict what the cross section would be for Pd.

To proceed, we write

$$U\sigma_{\mathrm{K}} = \omega_{\mathrm{K}} f\left(\frac{E}{U}\right), \tag{1}$$

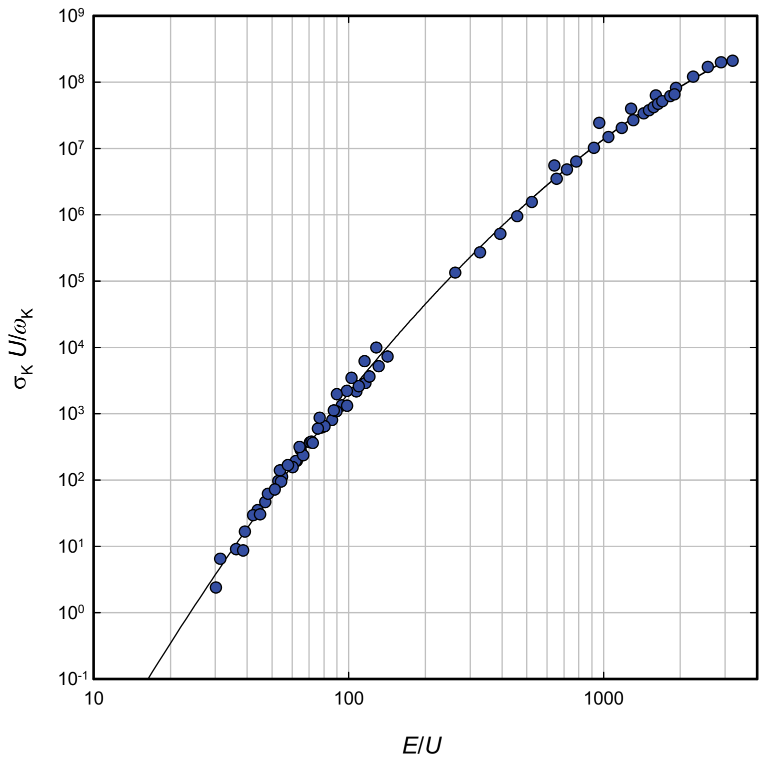

Figure 1. Universal function f (in units of barns-eV) built up from K-shell X-ray production cross sections for collisions with alpha particles.

where f is the presumed universal function, U is the K-shell binding energy and where ω_K is the fluorescence yield.

To determine whether this approach will be helpful given the data sets that we have available, we can plot the data in terms of the universal function according to

$$f\left(\frac{E}{U}\right) = \frac{U\sigma_K}{\omega_K} \tag{2}$$

as a function of E/U. The results of this are shown in Fig. 1.

2.2. Parameterization

We see that the data seems to describe a universal curve, which we have fit to

$$\ln f = a + bx + cx^2 \tag{3}$$

with

$$x = \ln\left(\frac{E}{1\ \text{MeV}}\right) \tag{4}$$

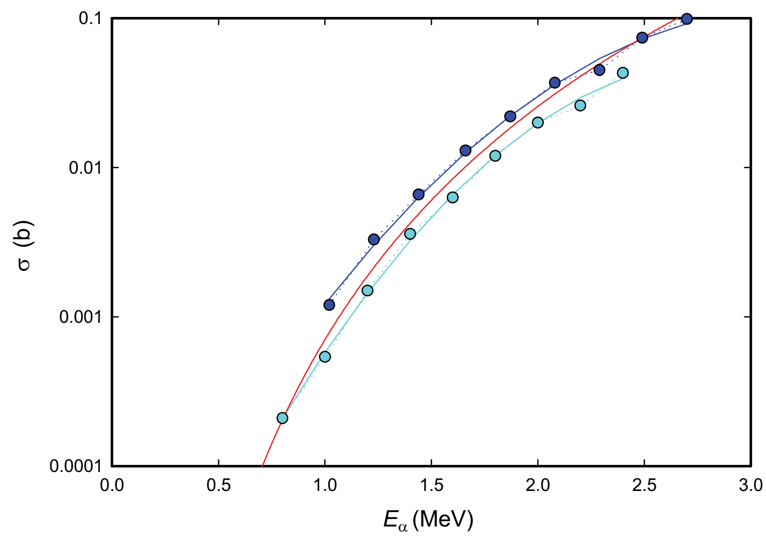

Figure 2. Comparison of the empirical X-ray cross section (red line) and data of Wilson et al. [5] for Rh (upper) and Ag (lower) data.

and

$$a = -22.7185, \qquad b = 8.4285, \qquad c = -0.3995 \tag{5}$$

for f in barns-eV.

2.3. Comparison with experimental results

We compare the empirical model with the data of Wilson et al. [5] in Fig. 2. Taking the Wilson data alone, one might think that the empirical fit was a bit off at both high and low energy. However, since there is reasonable agreement between different data sets (with data taken between Al and Gd) in the construction of the universal function, it seems that our empirical K_α cross section for Pd should be reasonable outside of the range where we have relevant data.

3. Yield of Pd K_α X-rays

We have computed the yield function for alpha particles in Pd (see Fig. 3).

3.1. Experimental result for the yield in Pd

There is an experimental data point available for the yield function of energetic alpha particles from the decay of ^{252}Cf given in a paper by Watson [8]. The yield function is given as $(0.93 \pm 0.08) \times 10^{-2}$ for the associated long range alpha particles in Pd. From the yield curve, we would compute that the associated alpha particle energy should be 15.0 MeV in order to be consistent. The long-range alpha particle energy spectrum for ^{252}Cf has been measured by Fraenkel [9], and one sees that the peak occurs roughly at 15 MeV.

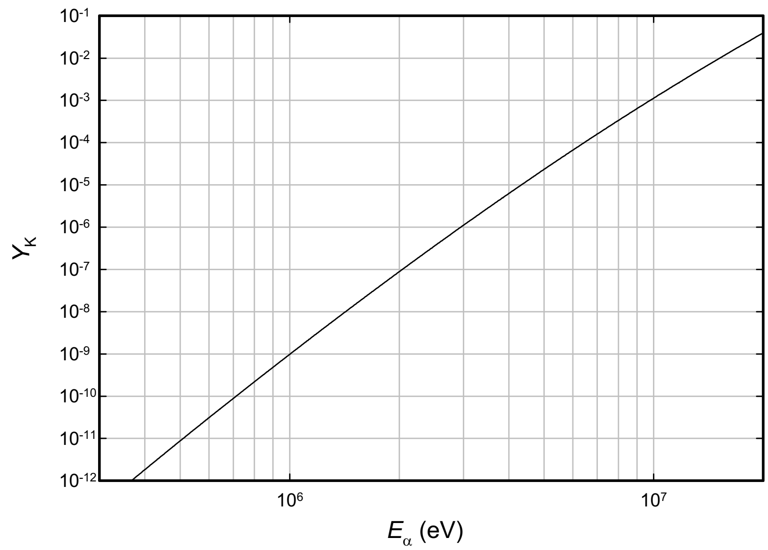

Figure 3. Yield function for Pd K-shell X-ray emission due to energetic alpha particles as a function of alpha particle energy.

3.2. K X-rays per unit energy

In order to compare with experiment, we require a computation of the yield divided by the energy, we require the K-shell X-ray yield divided by the alpha particle energy. This is shown in Fig. 4.

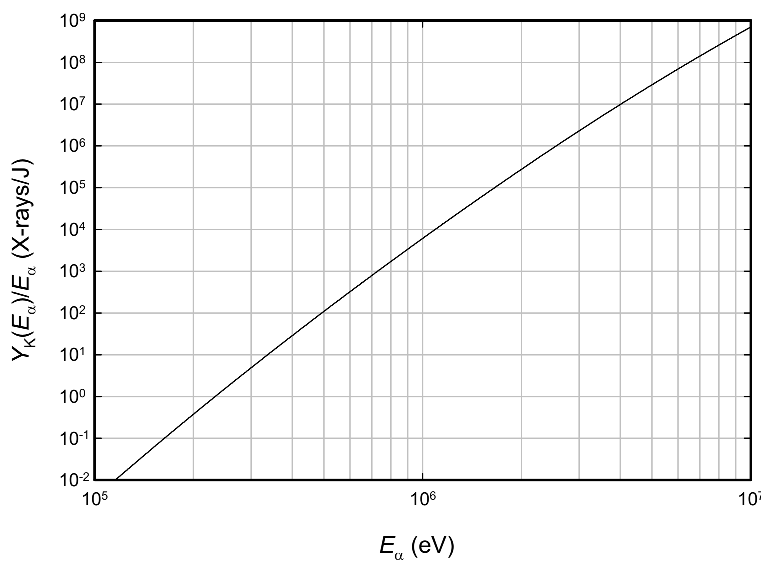

Figure 4. Yield function for K-shell X-rays in Pd produced by energetic alpha particles divided by the alpha particle energy, in X-rays/J.

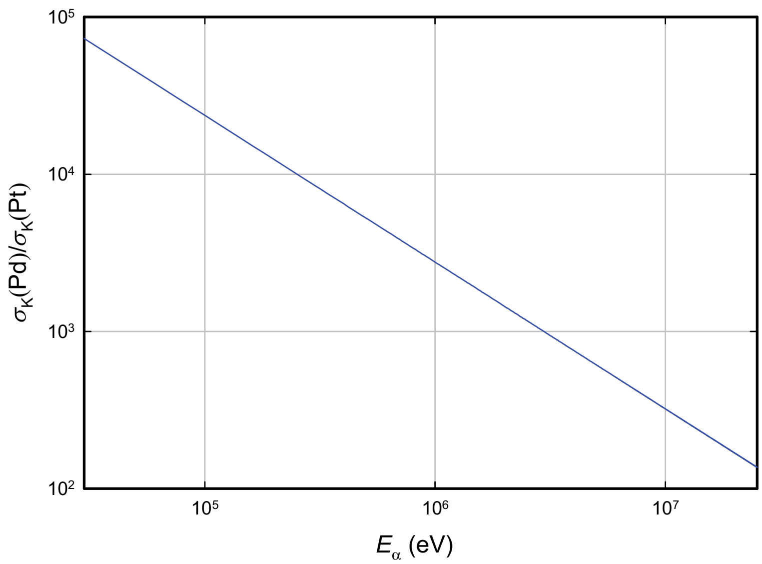

Figure 5. Ratio of K_α production cross sections for Pt relative to Pd as a function of alpha particle energy.

3.3. Ratio of Pd to Pt K_α cross sections

We mentioned above the possibility of gaining information about the alpha particle energy from the ratio of K_α emission from Pt as compared with Pd. There are few experiments where both have been measured at the same time, and the only work we could find reporting such a result was [1].

If we make use of the universal function ideas above, we can estimate the ratio of K-shell emission cross sections using

$$\frac{\sigma_K(\mathrm{Pd})}{\sigma_K(\mathrm{Pt})} = \frac{\omega_K(\mathrm{Pd})}{\omega_K(\mathrm{Pt})} \frac{U_{\mathrm{Pt}}}{U_{\mathrm{Pd}}} \frac{f(E/U_{\mathrm{Pd}})}{f(E/U_{\mathrm{Pt}})}. \tag{6}$$

This ratio is shown in Fig. 5. We see that the cross section for X-ray production is much larger for the lower energy Pd K_α X-ray than for the higher energy Pt K_α X-ray, which is as expected since it is harder to ionize a more tightly bound electron.

3.4. Surface versus bulk effects

The discussion above requires additional comment, since Pt is found only within less than a micron from the outer surface of the Pd cathode. If the alpha particles are born inside of the cathode away from the surface, then one would not expect to see any Pt K_α X-rays. If the alpha particles are born near the cathode surface, then one might expect to see excitation of the Pt K_α, but the ratio of Pd to Pt K_α emission might be expected to be even larger than the ratios found above. In the single experimental measurement that shows both, there is not a big difference in the relative strength, as discussed later on in this work. With Pt restricted to a small region near the surface, the Pd K_α should be favored even more over the Pt K_α than in Fig. 5.

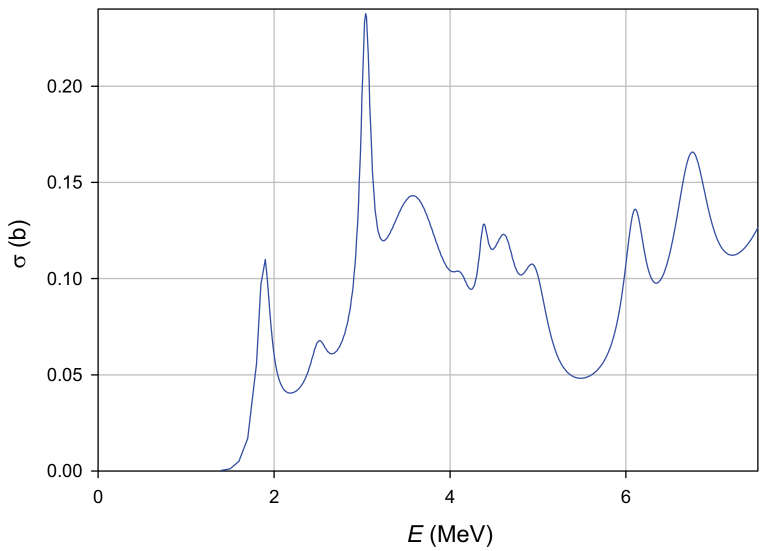

Figure 6. Cross section for ^{7}Li 478 keV gamma production as a function of alpha particle energy.

4. Excitation of the ^{7}Li nuclear state at 478 keV

If energetic alpha particles are present, one would expect to see excitation of nuclear states for low-Z nuclei that have bound excited states. In the Fleischmann–Pons experiment Li appears in the electrolyte, and presents an obvious candidate for possible nuclear excitation. In ^{7}Li the lowest excited state occurs at 478 keV, which makes it the prime candidate for nuclear excitation (other excited states in both ^{6}Li and in ^{7}Li are at significantly higher energy). This state has been the focus of numerous studies over the years precisely for this reason.

4.1. Cross section for gamma production

Because of its special status, the 478 keV gamma line has been studied experimentally in order to determine the gamma production cross section. This is somewhat different than an excitation cross section, as excitation to more highly excited states can lead to the 478 keV state as a product in a decay chain. We have made use of the measurements of Li and Sherr [10], and of Cusson [11]. The cross section is complicated, with more than a dozen resonances apparent. We have made use of the Li and Sherr data at low energy up to the first peak. Above the first peak, we have developed an approximate fit (made up of many resonance terms) to the Cusson measurements. The resulting cross section is shown in Fig. 6.

4.2. Yield function

Lithium is present in the LiOD electrolyte as mentioned above, and it also is incorporated into the outer few thousand Angstroms of the cathode surface [12,13]. We expect the alpha particles to be sourced near the surface, so that there is some possibility of interaction with the Li incorporated into the Pd. However, alpha particles born this close to the surface will also have a 50% probability of going into the electrolyte. The computation of the yield function in the electrolyte is a preferable computation for us (since we need not worry about the profile of the lithium and the trajectory of the alpha particles), and we expect that it will provide the dominant signal under most conditions.

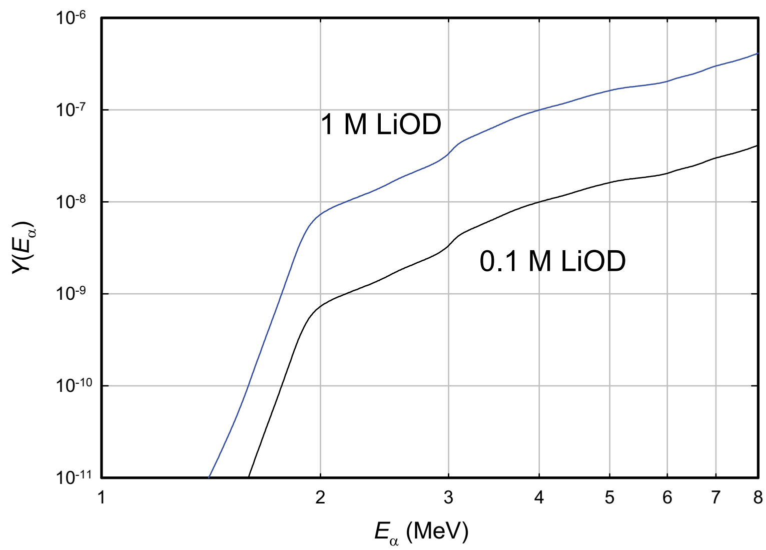

Figure 7. Yield of the ^{7}Li 478 keV gamma production as a function of alpha particle energy.

The resulting yield function for alpha particles slowing down in D_2O are shown in Fig. 7 for 0.1 M and 1.0 M LiOD. For alpha particles of energy greater than 2 MeV, we see that the yields are small, but respectable for this kind of process. Clearly a substantial flux of alpha particles would be required to observe this signal. However, large yields of alpha particles are now being claimed by Storms and Scanlan [14], and by the SPAWAR group [4]. It is probably worthwhile to look for the 478 keV line in these experiments. The associated yield per unit energy is shown in Fig. 8.

5. Discussion and conclusions

In the sections above, we have presented results for cross sections and yields of K_α X-rays and the ^{7}Li 478 keV gamma ray. Given the good accuracy of the stopping power models and cross sections, the resulting yields should be very good.

From these results, some conclusions can be drawn. In regard to ongoing experiments, it would be interesting to monitor the K_α signals and ^{7}Li signal, since they can provide independent information about the flux and energy of energetic particles. Although we have focused on energetic alpha particles here, similar yield functions for the K_α X-rays can be developed easily for other energetic ions. Excitation cross sections for the 478 keV line will be much more difficult to obtain for other light ions. These results also allow us to comment on the experiments showing K_α emission. Although important information about the Bush and Eagleton experiment [1] is not included in the write up, if we assume that the Rh, Pd and Ag K_α spectrum were taken in the same run as the Pt K_α spectrum, then we can be sure that these signals were not due to energetic alpha particle impact ionization. The reason for this is that the signal strength of the Pt appears to be comparable to that of Pd, which is inconsistent with what would be expected from alpha particle impact ionization (even if there were as much Pt as Pd available near the surface). Since the X-ray detection in this experiment has a relatively low resolution, there can be questions as to whether the signals are actually due to the K_α lines.

The presence of the Rh K_α line at similar strength to the Ag K_α line is very hard to account for in this experiment since the cathode was a PdAg alloy. In any event, we will need a confirmation of this experiment before we can be sure of our conclusions. In future investigations of K_α emission it will be important to use a high resolution detector so that

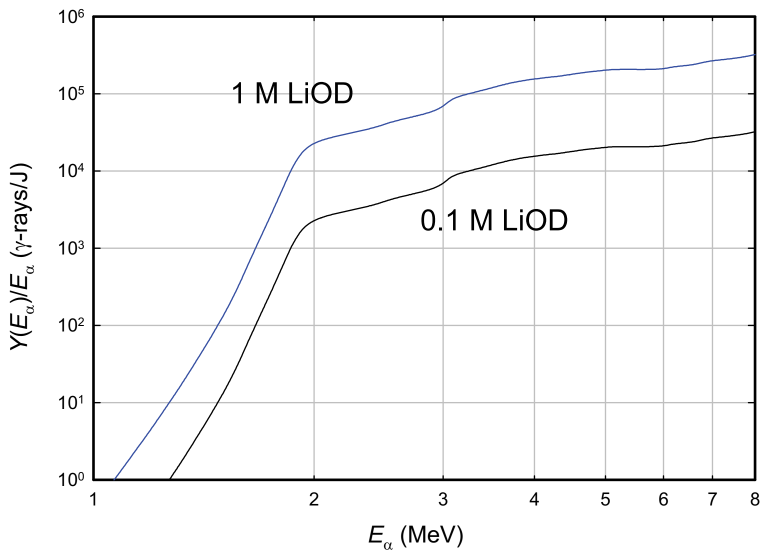

Figure 8. Yield of the ^{7}Li 478 keV gamma production divided by energy as a function of alpha particle energy.

the K_α and K_β lines can be resolved, so that we might have more confidence in the results.

Also of interest is the spectrum presented by Iwamura et al. [2], which shows clearly a dominant Pt K_α signal, but no 478 keV line. If produced by energetic alpha particles, probably the alpha particles would need to have an energy well above 2 MeV. Since there is no correlation with neutron emission, which was also measured, we are sure from other calculations that the Pt K_α signal is not associated with energetic alpha particle emission.

We draw attention to an earlier publication which contains a brief summary of these and earlier results [15].

References

[1] R. T. Bush and R. D. Eagleton, *Proc. ICCF3* 409 (1993).
[2] Y. Iwamura, N. Gotoh, T. Itoh and I. Toyoda, P̂roc. ICCF5, p. 197 (1995).
[3] A. G. Lipson, G. H. Miley, A. S. Roussetski and E. I. Saunin, *Proc. ICCF3*, p. 539 (2003).
[4] P. Mosier-Boss and L. P. G. Forsley, *Bull. APS* paper BAPS.2008.MAR.A17.10 (2008).
[5] S. R. Wilson, F. D. McDaniel, J. R. Rowe and J. L. Duggan, *Phys. Rev. A* **16** (1977) 903.
[6] J. D. Garcia, *Phys. Rev. A* **4** (1971) 955.
[7] G. Basbas, W. Brandt and R. Laubert, *Phys. Rev. A* **7** (1973) 983.
[8] R. L. Watson, *Phys. Rev.* **179** (1969) 1109.
[9] Z. Fraenkel, *Phys. Rev.* **156** (1967) 1283.
[10] C. W. Li and R. Sherr, *Phys. Rev.* **96** (1954) 389.
[11] R. Y. Cusson, *A study of the elastic and inelastic scattering of alpha particles by lithium-7*, CalTech Ph.D. Thesis (1965).
[12] O. Yamazaki, H. Yoshitake, N. Kamiya, K. Ota, *J. Electroanal. Chem.* **390** (1995) 127.
[13] H. Uchida, M. Sato, W. Cui, T. Tabata, M. Kumagai, H. Takano and T. Kondo, *J. Alloys and Compounds* **293–295** (1999) 30.
[14] E. Storms and B. Scanlan, presented at ICCF15.
[15] P. Hagelstein, *Naturwissenschaften* **97** (2010) 345.