On the absence of appreciable half-life changes in alpha emitters cooled in metals to 1 Kelvin and below

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- The recent suggestion that dramatic changes may occur in the lifetime of alpha and beta decay when the activity, in a pure metal host, is cooled to a few Kelvin, is examined in the light of published low temperature nuclear orientation (LTNO) experiments on such sources cooled to as low as 25 mK, with emphasis here on alpha decay. In LTNO observations are made of the anisotropy of radioactive emissions with respect to an axis of orientation. Correction of data for decay of activities in metallic samples held at temperatures at and below 1 Kelvin for periods of several days has been a routine element of LTNO experiments for many years. No evidence for any change of half-life on cooling, with an upper level of order 1%, has been found, in striking contrast to the predicted changes, for alpha decay, of several orders of magnitude. The proposal that such dramatic changes might alleviate problems of disposal of long-lived radioactive waste is shown to be unrealistic.

- It is the objective of this presentation to report aspects of already published experiments of significance to these suggestions, clearly indicating that the predictions are far from experimental reality. The experiments concern the decay of relatively long lived 224Rn, 225Ra and 227Ac and their daughter decay chains implanted in an iron metal foil and studied at temperatures down to below 20 mK. The original objectives of this work were to study the polarization of the decaying isotopes through measurement of the angular properties of their alpha and gamma emissions - the method of low temperature nuclear orientation (LTNO) - leading to results on the hyperfine interactions of the isotopes involved, their magnetic dipole moments and the magnetic hyperfine fields they experienced as implants in the ferromagnetic iron lattice.

- The experiments which provide the evidence given here were performed at ISOLDE, CERN and Oxford over a period of several years. Other LTNO groups, in particular those from Leuven and Bonn, have published work on implanted alpha emitters cooled to millikelvin temperatures in metals, containing similar evidence.

- From Ref. [1] the enhancement of alpha decay rates in a metal is associated with the presence around the metallic ion of a screening charge, due to free electrons, which is taken to reduce the barrier height by the screening energy UD
- According to this picture, the reduced barrier height enhances the rate of alpha tunneling through the barrier to separate from its parent nucleus, thus reducing the lifetime. Since ionic charge screening by free electrons occurs only in metals, it is predicted that large differences of alpha decay lifetime will be observed depending upon whether the decay takes place in an insulator or a metal. An expression is given for UD in terms of the charges of the alpha particle Z_ and the daughter nucleus Zt, the temperature of the lattice electrons and a parameter Ue(d+d) which relates to a d + d fusion reaction in a metal

$$U_D = Z_\alpha Z_t U_e (d+d)_{290} \left(\frac{290}{T}\right)^{1/2}$$

- where T is the absolute temperature of the metal in which the decay takes place. A ‘typical’ value of Ue(d+d)290 is given to be 300 eV at 290 Kelvin. The temperature dependence arises from taking the Drude model for the ‘quasi-free’ valence electrons as having an (average) kinetic energy of 0.5 kT. Based on this expression it is suggested that by cooling the activity in a metal sample to liquid helium temperature (4.2 K), the half-life of 210Po can be reduced from 138 d to 0.5 d and of 226Ra from 1600 y to 1.3 y, factors of order 1000, in each case through a reduction of the effective barrier potential by about 420 keV.

- Low temperature nuclear orientation experiments involve cooling radioactive isotopes in samples in which they experience strong hyperfine interactions to temperatures at which they become, in the case of a magnetic interaction, polarised along the direction of the magnetic field. The appropriate temperature range is typically between 1 and 100 mK for activities in, for example, a ferromagnetic iron foil sample.

- The decay correction, fundamental to these measurements, is directly relevant to the present work, as it involves the decay constant of a isotope in question through a correction factor
- where λ_=ln(2)/T1/2 is the decay constant, _ is the real time of the counting period and t is the difference between the arbitrary normalisation time and the start of the measurement.

$$\Xi = \frac{\lambda\delta \exp(+\lambda t)}{1 - \exp(-\lambda\delta)}$$

- Physical parameters are extracted from measured anisotropies by comparison of the experimental angular distribution and the theoretical expression

$$W(\Theta, T) = 1 + f \sum_{k=2}^{k_{max}} B_k(T) U_k A_k Q_k P_k(\cos\Theta)$$

- where *Bk* , *Uk* and *Ak* are the usual orientation, deorientation and angular distribution coefficients, *Qk* are the detector solid angle corrections, *Pk* are Legendre polynomial and *f* is the fraction in good sites . Depending on the nature of a particular experiment, selected information contained in one of the coefficients can be obtained by such a comparison, assuming all the other coefficients are known.
- In the work referred to in this paper, the orientation coefficients *Bk* , dependent on the product of magnetic dipole moment and the hyperfine field acting on the studied nuclei in the sample, were used to extract the magnetic dipole moment of oriented states and compare them with either the same moments obtained by a different technique or with systematics and theory.
- It is obvious that should the experimental anisotropy be determined incorrectly, i.e. if the correction for the radioactive decay, made by using decay constants known at room temperature, would not apply at temperatures of 1 K and lower, the extracted magnetic dipole moment would be wrong.

- Additional sensitivity to the appropriateness of the decay correction arises from other detailed properties of the measured anisotropic distributions. The first sensitive feature is that, at the lowest temperatures, the anisotropy must become independent of temperature as the nuclei become fully polarized. Since there is a time sequence in such measurements as the sample temperature is changed, any incorrectness in the source decay correction will impose a false slope or scatter upon the extracted temperature dependence.

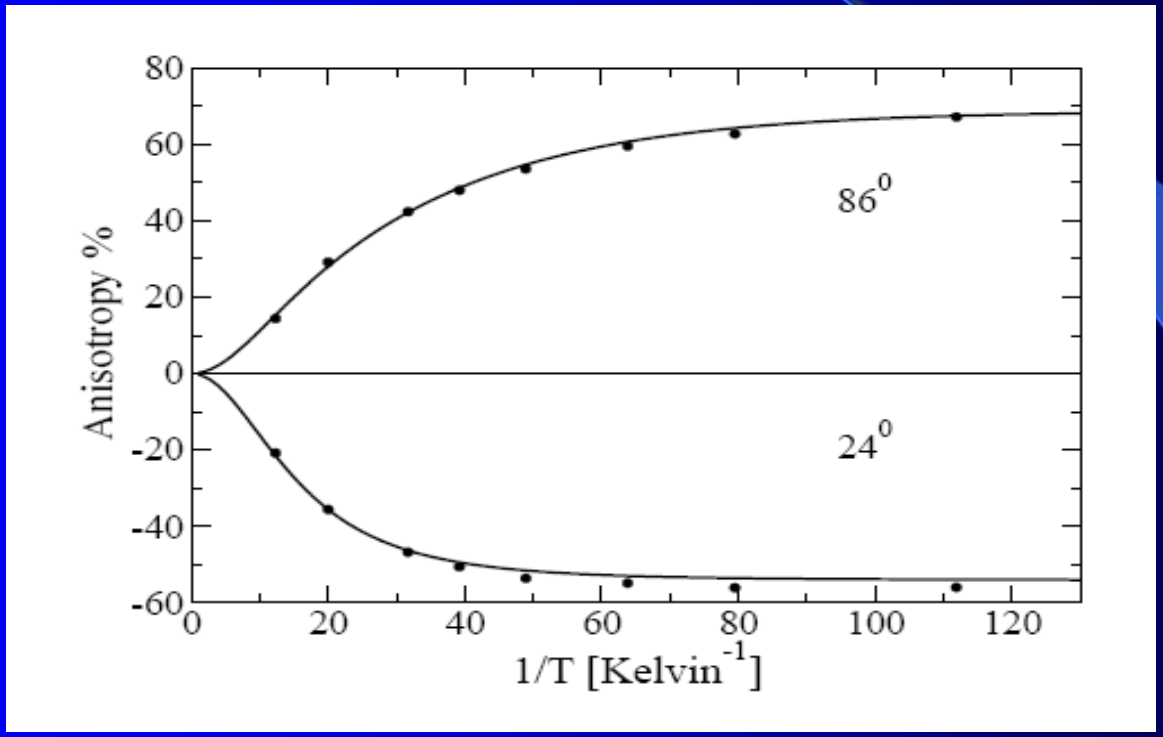
Table 1

Predicted alpha decay lifetime reductions and alpha fraction changes based upon Ref. [1]

Parent nucleus	Normal alpha decay half-life at 290 K*	Alpha energy (keV)	Normal fraction (%) at 290 K	U_D (keV)	Half-life reduction factor at 1 K	Predicted alpha fraction (%) at 1 K**	Predicted decay half-life (%) at 1 K***
$A = 224$ decay chain, headed by alpha emitter ^{224}Ra							
^{224}Ra	3.66 days	5686	94	898	6.3×10^4		4.7 s
		5449	5.5	898	1.3×10^5		
^{220}Rn	55.6 s	6228	100	878	1.2×10^4		4.7 ms
^{216}Po	0.15 s	6779	100	857	2.7×10^3		55×10^{-6} s
^{212}Bi	60.6 m	6090	27.2	847	1.2×10^4	26	0.82 s
		6051	69.9	847	1.3×10^4	74	
Predicted missing transitions if enhanced alpha decay of ^{212}Bi weakens beta decay branch							
^{212}Po	0.30×10^{-6}	8784	100	Not seen if beta decay of ^{212}Bi ceases			

Parent nucleus	Normal alpha decay half-life at 290 K*	Alpha energy (keV)	Normal fraction (%) at 290 K	U_D (keV)	Half-life reduction factor at 1 K	Predicted alpha fraction (%) at 1 K**	Predicted decay half-life (%) at 1 K***
$A = 225$ decay chain, headed by ^{225}Ra > 99% beta <1×10^{-4}% alpha $T_{1/2}$ 14.8 days							
^{225}Ac	10.0 days	5830	50.7	889	3.8×10^4	46	20.6 s
		5794	24.3	889	4.3×10^4	25	
		5732	10.1	889	5.0×10^4	12	
		5637	4.4	889	6.5×10^4	7	
^{221}Fr	4.8 m	6341	83.4	867	7.7×10^3	77	34 ms
		6126	15.1	867	1.3×10^4	23	
^{217}At	32.3 ms	7067	99.9	867	1.8×10^3	100	18×10^{-6} s
^{213}Po	4×10^{-6} s	8375	99.9	857	3.3×10^2	100	1.2×10^{-8} s
Predicted additional transitions via enhanced alpha decay of ^{225}Ra							
^{225}Ra	>4×10^4 yr	4950	not seen	898	5.0×10^5	30% of all	28 days
^{221}Rn	125 m	6035	strongest	878	2.0×10^4	strongest	0.38 s
^{217}Po	<10 s	6537	>95	857	4.4×10^3	100	<2 ms
Predicted additional transitions via enhanced alpha decay of ^{213}Bi							
^{213}Bi	38 h	5869	93	847	2.1×10^4	85	5.9 s
		5549	7	847	4.8×10^4	15	

Parent nucleus	Normal alpha decay half-life at 290 K*	Alpha energy (keV)	Normal fraction (%) at 290 K	U_D (keV)	Half-life reduction factor at 1 K	Predicted alpha fraction (%) at 1 K**	Predicted decay half-life (%) at 1 K***
$A = 227$ decay chain headed by ^{227}Ac, normal decay; 98.6% beta, 1.4% alpha, $T_{1/2}$ 21.8 yr							
^{227}Th	18.72 days	6038	24.5	918	3.2×10^4	19.7	40.7 s
	100% alpha	5979	23.4	918	3.5×10^4	20.5	
		5756	20.3	918	6.8×10^4	34.7	
		5713	4.9	918	7.6×10^4	9.3	
		5709	8.2	918	7.6×10^4	15.6	
^{223}Ra	11.44 days	5748	9.1	898	5.4×10^4	7	14.0 s
	100% alpha	5717	53.7	898	6.1×10^4	46.7	
		5607	26	898	7.9×10^4	29.1	
		5539	9.1	898	9.9×10^4	12.7	
		5434	2.3	898	13.5×10^4	4.4	
^{219}Rn	4 s	6819	81	878	3.1×10^3	62.9	1.1 ms
	100% alpha	6553	11.5	878	5.3×10^3	16.6	
		6425	7.5	878	7.2×10^3	14.7	
^{215}Po	1.8 ms	7386	100	857	1.0×10^3	100	1.8×10^{-6} s
^{211}Bi	2.15 m	6623	84	847	3.3×10^3	71	33 ms
		6279	16	847	7.1×10^3	29	
Predicted additional transitions via enhanced alpha decay of ^{227}Ac							
^{227}Ac	21.77 yr	4950	47	889	5.7×10^5	45.4	1.00 days
		4938	40	889	5.7×10^5	38.8	
		4870	6.1	889	7.5×10^5	7.8	
		4853	3.7	889	7.5×10^5	4.7	
^{223}Fr	21.8 m	5340	strongest	867	1.3×10^5	100	3.4 m
^{219}At	0.9 m	6275	strongest	867	4.0×10^3	100	13.5 ms

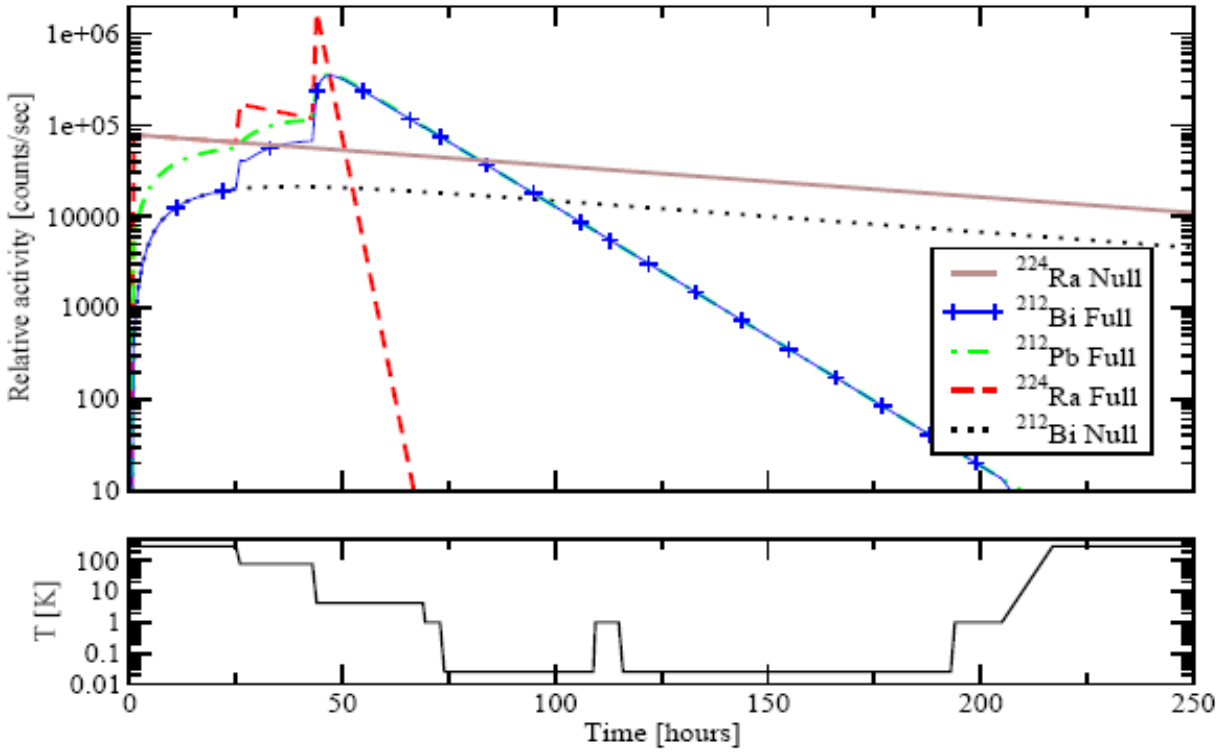


^{227}Ac decay chain: ^{211}Bi 6623 keV alpha transition anisotropy vs inverse temperature

- The second sensitive feature is that the anisotropy involves an increase in observed intensity in some directions to the orientation axis, and in others a decrease, the actual quantitative changes being determined by well established theory of the distribution. The application of an incorrect adjustment for source decay will falsify the relationship between observations in detectors in different directions to the axis. Thus, if the decay correction assumes too short a lifetime, the ‘warm’ unoriented distribution will be taken to fall too fast, so that any observed increase in intensity, measured at a time later than the ‘warm’ distribution, will be analysed as too large relative to its true value by a certain factor, and any simultaneously observed decrease would be analysed as smaller than its true value, by the same factor. These failures would lead to distortion of the relation between the analysed anisotropies in the two directions, which can usually be readily identified.

- A separate aspect of the LTNO technique with relevance to lifetime is the fact that for the observed anisotropies to be characteristic of the hyperfine interaction in a specific nuclear state, usually a ground state, the lifetime of that state must be long enough for the nuclei to reach thermal equilibrium with the metallic lattice and achieve a degree of polarisation characteristic of that temperature and interaction strength. The time for this to be reached, the nuclear spin-lattice relaxation time, T1, has been established in implanted, diffused and co-melted metallic samples as following the Korringa conduction electron mechanism. In iron samples of the type discussed in this paper T1's have been measured to range from many hours to below 100 ms at millikelvin temperatures. The relevant parameter is the ratio T1/ T1/2. There is abundant evidence that were a lifetime to become shorter than 0.1 ms thermal equilibrium will not be achieved. In situations involving a decay cascade, such as are described in this paper, a degree of polarisation may be 'inherited' from a precursor state higher in the decay chain. Reference to situations of this kind are made in the text where relevant, as the degree of equilibration will be affected if lifetimes were to be severely reduced on cooling.

- Finally, as a matter of course, OLNO experiments include measurement of the unoriented intensity at both the start and the end of the experiment, and quite often also at an inter-mediate time, when the temperature is at close to 1 K. It is a standard requirement that these normalisation measurements must, after due correction for decay of the source, yield a consistent value for the reference ‘warm’ intensity to be used in evaluation of all measured anisotropies. If the normalisation should not be consistent this would be immediately apparent and present a challenge to the experimenters. Thus we have, in effect, a measure of the source decay at 1 K over the period of the experiment, which is typically at least several days.



Relative activity for selected members of the ^{224}Ra decay chain in the Full and Null scenar-

DISCUSSION AND CONCLUSIONS

- In this paper evidence has been presented concerning the prediction that introducing alpha activities into metals and cooling them to temperatures of a few Kelvin and below should produce major changes in their decay rates and the appearance of their alpha spectra. On this evidence, no such changes have been observed with an upper level of one percent, at 1 K, a factor of about 105 smaller than the predicted changes. Two of the three nuclear moments deduced from these experiments, which depended upon correct source decay treatment, have been confirmed by independent subsequent measurements; the third, measured solely by this technique, agrees well with theoretical calculation.

The evidence presented has been of five types, namely

- 1. consistency of warm counts corrected using ’normal’ half-lives - a 1 K property.
- 2. absence of activity change between 1 K and millikelvin temperatures as shown by smooth temperature variation of observed anisotropies and the correct low temperature saturation behaviour of strong anisotropies. Large effects are predicted, even in the Partial scenario. However, these predictons depend upon accepting the $T^{-1/2}$ dependence of the screening energy UD as applicable down to millikelvin temperature - the original suggestions in Ref. [1] made no limit of temperature range and took as their example cooling to 4.2 K.
- 3. absence of anisotropy where isotope properties lead to no polarisation.
- 4. no loss of peaks or appearance of additional peaks in alpha spectra between room temperature and millikelvin temperatures.
- 5. evidence that ratios of relaxation time to half-life are not changed enough to prevent thermal equilibrium in some shorter lived ground states.

- Since Ref. [1] appeared there have been several follow-up papers. Amongst these the original proponents of the effect, Raiola et al. [22] claim to have observed a 6% lifetime decrease on cooling a sample of 210Po, implanted into Cu, to 12K – a result ‘consistent in sign but significantly smaller than expected’ [22] (i.e. 6% compared to 1000%, or 1 part in 150 of the prediction). Jeppesen et al. [6] report changes in half-life at room temperature between decay of 221Fr implanted into metals and non-metals of order 0.4%, with comparable errors, where the Full prediction would be reduction by a factor _2 (1 part in 500 of the prediction).