EFFECT OF MATERIAL ENVIRONMENT ON A CLASS OF NUCLEAR LIFETIMES

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G. J. Perlow

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Physics Division, Argonne National Laboratory, Argonne, IL 60439, USA

Abstract

The connection between internal conversion of a nuclear transition and EXAFS is pointed out. A prediction is made of sizable variations in lifetimes of nuclear states depending on the surrounding material environment, provided that the transition energy is just above threshold and the internal conversion coefficient is appreciable.

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If the x-ray absorption coefficient of a material is scanned in energy in the region just greater than an absorption edge of one of the constitutent atoms, one observes a quasi-periodic variation in the absorption.[1] This x-ray absorption fine structure, (EXAFS), is understood to be due to interference between the outgoing electron wave and one elastically back-scattered by surrounding atoms [2,3,4,5]. The purpose of this report is to show that similar interference is expected to occur in the case where an electron is emitted in the internal conversion of a nuclear transition. In both cases, the phase of the scattered wave and therefore the position of extrema depend on the product kR, where k is the electron wave-number and R is the interatomic distance. In EXAFS, one varies k and determines R thereby. In internal conversion, on the other hand, k is fixed by the transition energy. The variation of R in different material environments will produce variations in the internal conversion coefficient, α , defined as the ratio: (number of electron emissions) / (number of gamma-ray emissions), and if chemical effects on nuclear lifetimes that have been measured in the past [6,7]. In general "chemical effects" are due to to a change in the population of the electron shells, while here it is only the local crystallography which is involved. For example, a high pressure experiment would show an approximately periodic variation of lifetime with pressure. In this paper, we discuss the nature and size of the phenomenon, the question of multipole order, and display a few favorable cases called from the Brookhaven data base. No measurements have been made. The expected magnitude of the effect is given not from first principles but as a comparison with an assumed EXAFS measurement on a stable isotope.

The single-scattering description of EXAFS is adequate for our purpose [2,3]. The atom to be ionized is at the origin and contains

2 electrons in the shell which, to simplify the discussion, is restricted

to ns>. An isolated atom's absorption cross-section for an x-ray energy above

the shell edge is determined by the squared matrix element:

$$|M_o|^2 = |\langle p|H'|ns\rangle|^2 \tag{1}$$

Here H' is a dipole perturbation, acting only on coordinates, and $\langle p|$

represents an outgoing electron in a continuum p-state. The result of elastic

back-scattering from a neighboring atom at R is to alter eq. 1 to:

$$
\begin{aligned}
|M|^2 &= |\langle p + \phi_s|H'|ns\rangle|^2 \\
&= |M_o|^2 + \{M_o \cdot \langle\phi_s|H'|ns\rangle + cc.\} + \cdots
\end{aligned}
\tag{2}
$$

where ϕ_s is the back-scattered wave in the atomic potential of the ionized

atom, and the small term is neglected. It can be shown that the matrix

element is approximately unchanged by replacing $\langle\phi_s|$ by $B\langle p|$, where B is a

complex constant: $|B|e^{i\beta}$, Hence:

$$|M|^2 = |M_o|^2 \{1 + 2|B|\cos\beta\} \tag{3}$$

$$\chi \equiv \frac{|M|^2 - |M_o|^2}{M_o^2} = 2|B|\cos\beta$$

A specific model is needed to proceed further. The plane-wave approximation

[2] yields a useful result which can be written in condensed form:

$$\chi(k,R) = F(k) \frac{\sin(2kR + \gamma)}{R^2} \tag{4}$$

Here $F(k)$ contains the back-scattering amplitude as reduced by various effects such as shake-off, Debye-Waller factor, multiple scattering, and inelastic scattering. The phase γ includes all phase shifts that do not depend on R. A sum over all atoms has been omitted.

INTERNAL CONVERSION

In this process, [8] there is a transition between two nuclear states that sometimes results in the emission of a gamma ray, and sometimes in the ejection of an electron from one of the atomic shells. If J_1, Π_1 is angular momentum quantum number and parity of the upper nuclear state and J_2, Π_2 that of the lower, then the photon carries off angular momentum L $\hbar$ with any value allowed by: $|J_1 - J_2| < L < |J_1 + J_2|$. The radiation will have (+) parity if the parity is unchanged in the transition and (−) if there is a change. The transition can further be described as electric, E(L), or magnetic M(L), or mixed, depending on selection rules and dynamics. The following rules apply: a) The transition rate T rapidly decreases with increasing multipole order L. b) The transition rate is much greater for E(L) than for M(L). c) The parity of an E(L) photon is $(-1)^L$ and of M(L), $(-1)^{L+1}$. d) The angular distribution of the energy of E(L) and M(L) are identical, − the radiations differ by the interchange of electric and magnetic fields.

The emitted electron and the residual ion must conserve the energy, angular momentum, and parity of the converted transition. Whereas in EXAFS only E(1), transitions occur with sufficient strength to be important, the

reverse is true for low enegy (less than ~ 100 keV) nuclear transitions. M(1) and E(2) are more common.

Gamma ray emission and internal conversion are independent modes of decay of the state, - the separate transition rates add. Thus

$$T = T_\gamma + T_{ic} = T_\gamma(1 + \alpha) = \frac{1}{\tau} \tag{5}$$

where τ is the mean-life of the upper nuclear state. Therefore if $\alpha \gg 1$, the decay rate is dominated by the internal conversion process and whatever changes its value will change the observed lifetime appropriately. We can show that an EXAFS-like alteration of the final electronic state will occur if the decaying atom is in a material environment. For the largest effect the following conditions should hold: a) The transition energy should be not greater than about 1 keV above threshold, that is the electron energy should be less than 1 keV, preferably considerably less. b) As mentioned, the internal conversion coefficient should be large compared to unity. c) The transitions from the upper state to other states than the one of interest should be infrequent (but may be useful for measuring the lifetime). d) The conversion coefficient for higher shells than the one of interest should be much smaller. A similar situation prevails in EXAFS, for if the x-ray quantum has energy just greater than the K-edge, the photo-effect in the L and higher shells will decrease the value of χ.

The matrix element for the internal conversion in the isolated atom can be written:

$$\left| M_o^{ic} \right|^2 = \left| \langle J_2, \phi_o | H' | J_1, ns \rangle \right|^2 \tag{6}$$

$$= \left| \langle J_2 | H_n' | J_1 \rangle \cdot \langle \phi_o | H_e' | ns \rangle \right|^2$$

where the subscript n refers to nuclear and e to electron quantities, and $H' = H'$ (electric) $+ H'$ (magnetic). The factorization of H' into atomic and nuclear parts results from the large ratio of the respective radial coordinates. Hence,

$$\alpha = \text{const} \cdot \frac{\left| M_o^{ic} \right|^2}{\left| M_n \right|^2} = \text{const} \cdot \left| \langle \phi_o | H_e' | ns \rangle \right|^2 \tag{7}$$

$$= \text{const} \cdot \left| M_o^e \right|^2$$

The constant involves the ratio of nuclear to electron phase space quantities. The internal conversion coefficient to very good approximation depends only on energy and multipole order and on atomic quantities. For an E(1) nuclear transition, the latter are essentially the same as in EXAFS. One can therefore again write for the atom in a material:

$$\left| M^e \right|^2 = \left| \langle \phi_o + \phi_s | H' | ns \rangle \right|^2 = \left| M_o^e \right|^2 \cdot \{ 1 + 2 \ B \cos \beta \} \tag{8}$$

$$\frac{\Delta \alpha}{\alpha} = F(k) \frac{\sin (2 \ kR + \gamma)}{R^2}$$

as in eq. 4.

In the case of an M(1) transition, the outgoing wave ϕ_o is an s-state (with an irrelevant spin flip), and for E(2), a d-state. There will be a difference both in the phase γ and in the magnitude F(k). To estimate the effect of multipole order, we can consider the returning wave from a single scatterer to be plane at the atomic boundary r, and take the effect in question to be proportional to the coefficient j_ℓ (kr) of the ℓ-th partial wave. The quantities depend critically on r, but as we are not concerned with the location of the extrema but with their range of values, we can add an arbitrary phase to kr and compare the maximum values of j_ℓ. In this way we find that M(1) conversion in a state ns> to be superior to E(1) by about a factor of 2.3 and E(2) to give only 0.7 of the E(1) effect. However, if the scatterers can be considered as grouped in coordination shells, then adding the amplitudes coherently produces an approximation to an s- wave for all multipoles and the ratios are nearer to unity. The details are beyond the scope of this report.

In the case of conversion in shells other than ns> more than one component of the plane wave must be considered. Their corresponding contributions to the wave function in the neighborhood of the parent shell will have different phases and will interfere. This may invalidate the estimate for the single scatterer.

In the discussion to this point the nuclear state has been assumed not to be oriented with respect to the crystallographic axes. In the case of an un-oriented nucleus and a mixed transition the multiple components are incoherent, and can be treated by separately considering the parts (for example M1 and E2) and adding intensities. Therefore it suffices to consider pure multipoles.

Finally, we insert all the factors important in estimating the size

of the expected effect:

$$\{\text{range of } ^{\Delta\tau}/\tau \} = b_n \cdot b_\alpha \cdot C \cdot \{\text{range of } \chi \} \qquad 9$$

Here the branching ratio b_n gives the fraction of desired transitions compared to all transitions originating in the upper nuclear state. Similarly b_α is the ratio of the internal conversion coefficient in the shell of interest to the total internal conversion coefficient. The constant C takes into account the effect of multipole order discussed above. For an un-oriented nuclear state in a location of high symmetry, and conversion in ns>, C ~ 1.

In order to give some idea of the ranges involved we shown in Fig. 1, a k-shell EXAFS spectrum of metallic Ni [9]. The spectrum has been normalized first by extrapolating and subtracting the background from events below the edge and then fitting and subtracting a smooth curve above the edge corresponding to the absorption of the isolated atom. The quotient χ so obtained is independent of absorber thickness, of photo-effect in higher shells, and of the contribution to the absorption coefficient from scattering of the x-ray beam. This is exactly what should be used in eq. 9, as in internal conversion there are no thicknesses, no x-rays to scatter, and the conversion in higher shells is explicitly contained in b_α.

FAVORABLE CASES

Fig. 1 shows three favorable cases. The selection process extracted all listed transitions in the Brookhaven Data Base [10] with energies below 100 keV, and of these selected further those with transition energies greater than but within 1 keV of an edge. Internal conversion coefficients were taken from theory [11]. The tables give listings to a minimum electron energy of

1 keV. Electric transitions vanish at threshold [12] as k, but as there was no evidence of a maximum at 1 keV or higher, it is not believed that any conclusions drawn from the conversion coefficients we have used is seriously in error. If the electron energy is taken as the only criterion, a number of k-shell cases at higher Z would be included, but these have been excluded by consideration of too short K-shell lifetime [5].

ACKNOWLEDGEMENTS

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FIGURE CAPTIONS

Fig. 1   Normalized (see text) k-shell nickel EXAFS spectrum, (ref. 9). The
         normalized corrects for absorption in higher shells, pseudo-
         absorption due to x-ray scattering by the sample, and absorber
         thickness. It is appropriate for use in Eq. 9.

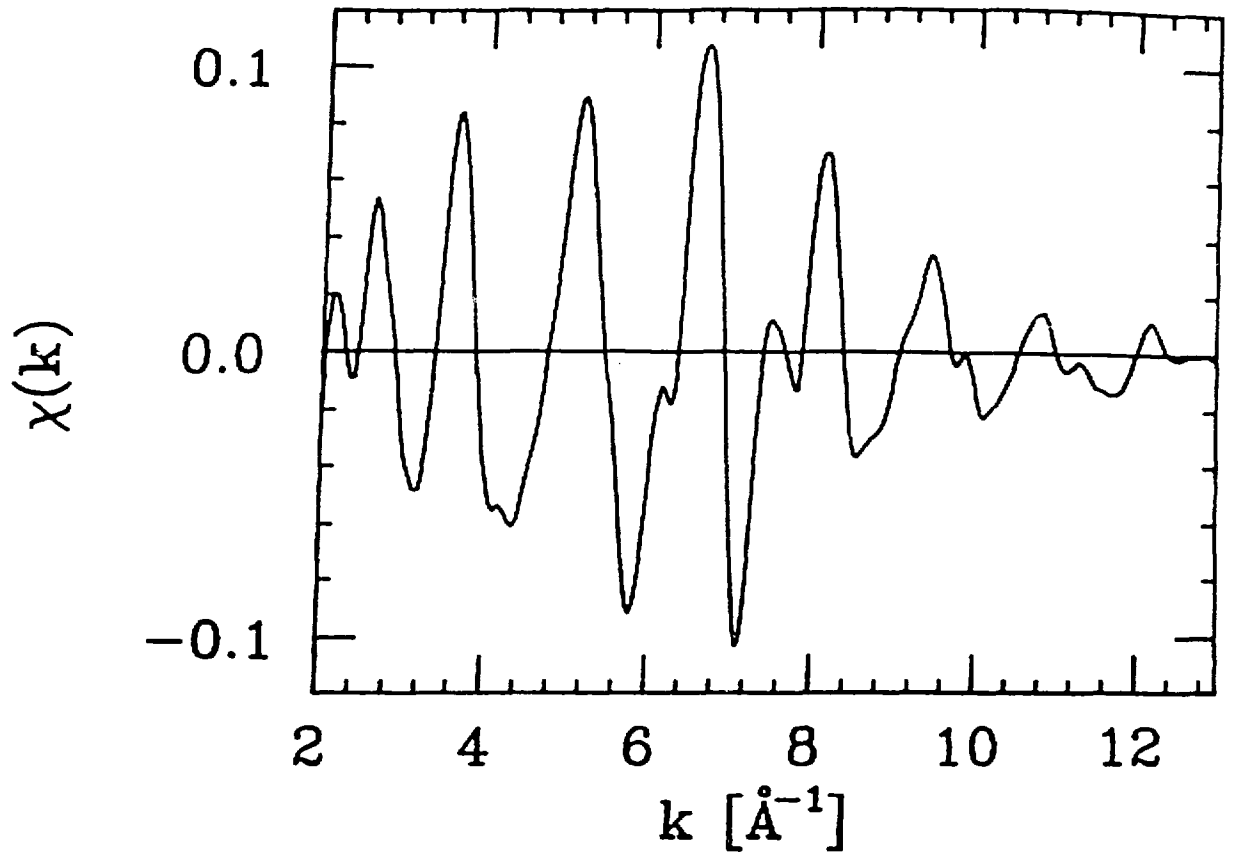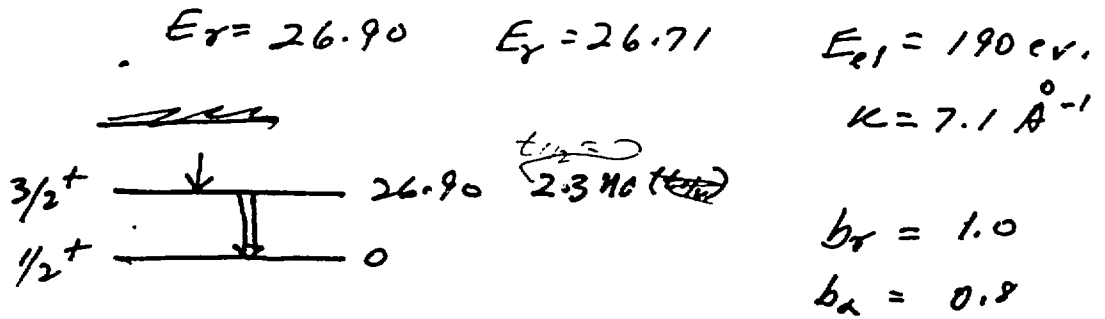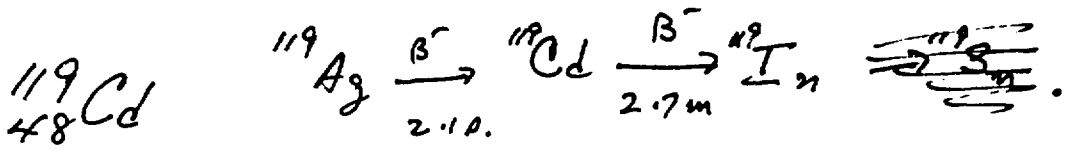Fig. 2   Three cases culled from a large data base (ref. 10).

Ni−metal, K−EXAFS

Fig. Perlow

$^{119}_{48}Cd$

$^{119}Ag \xrightarrow[2.1 D.]{\beta^-} \; ^{119}Cd \xrightarrow[2.7m]{\beta^-} \; ^{119}I_n$

$E_r = 26.90$ $E_\gamma = 26.71$ $E_{el} = 190\, ev.$

$K = 7.1\, \AA^{-1}$

$3/2^+$ ⎯⎯⎯⎯ 26.90 $t_{1/2} = 2.3\, Me$

$1/2^+$ ⎯⎯⎯⎯ 0

$b_r = 1.0$

$b_\alpha = 0.8$

$M1$

Far from stability—difficult to work with.

Fig 2a Peelw

$^{171}_{70}Yb$ ⟵ ^{171}Tm $\xrightarrow[1.9y]{\beta^-}$ ^{171}Yb $\xleftarrow[8.3d]{\beta^+, ec.}$ ^{171}Lu

$E_\gamma = 9.15$ kev. $E_{L_{III}} = 8.94$ $E_{ce} = 210$ ev.

$5/2^-$ ———— 75.66 $\overbrace{t_{1/2} =}^{}$ 1.6 no. $(t_{1/2})$

$3/2^-$ ———— 66.72

$1/2^-$ ———— 0

$b_\gamma = 0.94$

$b_\alpha = 0.65$
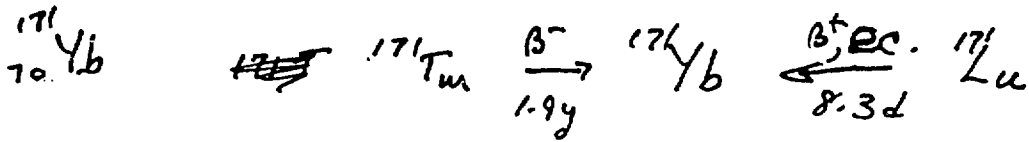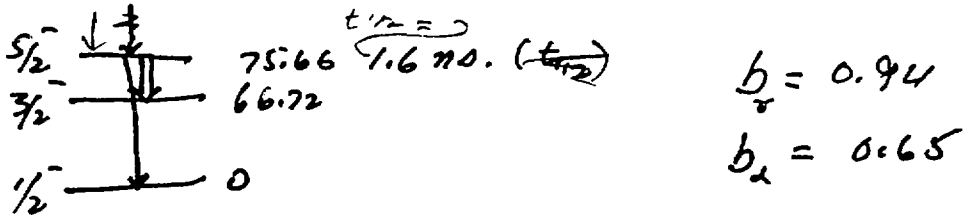
$M1 + 0.02\,E2$, $\alpha_{L_{III}}(E2) = 8.3 \times 10^4$

but ~~cont~~ int. conversion $\alpha_{L_{III}}(M1) = 5.6$

is overwhelmingly E2

$^{185}_{74}W$ ⟵ ^{185}Ta $\xrightarrow[50m.]{\beta^-}$ ^{185}W $\xrightarrow[75d.]{\beta^-}$ ^{185}Re

$E_\gamma = 69.70$ kev. $E_K = 69.53$ $E_{ce} = 170$ ev.

$3/2^-$ ———— 93.33

$9/2^-$ ———— 65.86

$1/2^-$ ———— 23.54

$3/2^-$ ———— 0

$b_\gamma = \sim 1$

$b_\alpha = 0.74$

$\alpha_K = 15.4$
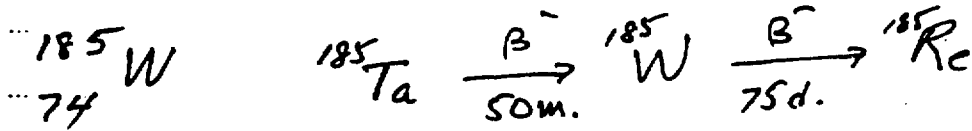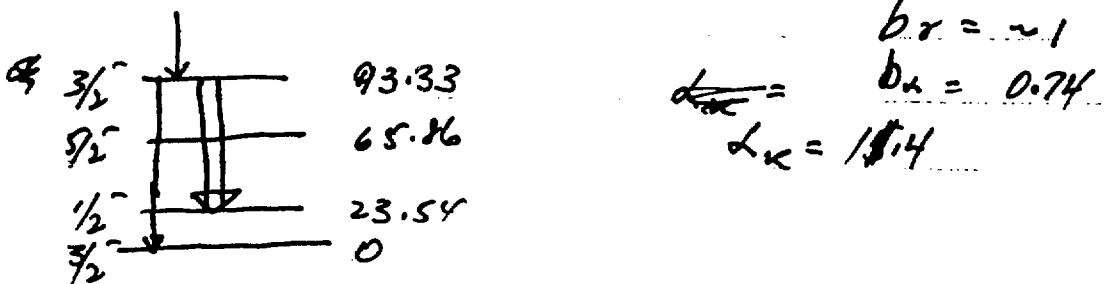
$M1 + 0.27\,E2$

Width of the K shell ~ 40 ev , a severe handicap to use of this isotope

Fig 2 b, c   Perlow