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THE STUDY OF BETA ENVIRONMENTAL FINE STRUCTURE APPLIED TO THE β–DECAY OF ^{187}Re

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ABSTRACT

The theoretical formalism of the atomic effect, termed Beta Environmental Fine Structure, has been evaluated for the beta decay of ^{187}Re. This is an oscillatory spectral distortion which arises from the beta emission occurring within a crystal lattice environment and is predicted to be of a magnitude, experimentally observable for the very first time, in the particular case of a cryogenic measurement. A comprehensive description is given of all steps required in the calculation of the Beta Environmental Fine Structure for a perfect single crystal of rhenium and a polycrystalline rhenium foil structure.

1. INTRODUCTION

A cryogenic experiment[1] is currently in progress to measure the mass of the electron anti-neutrino from the spectral study of the ^{187}Re beta decay, of end point energy 2.6 keV. A high sensitivity measurement of the entire beta spectrum is made possible by means of the cryogenic thermal detection mode. In detail, a single crystal of rhenium (the beta isotope ^{187}Re has a natural occurrence of 63 %) is coupled thermally to a Neutron Transmutation

Doped (NTD) germanium thermistor. The interaction of a beta electron within the rhenium substrate generates a temperature rise and this thermal signal is detected by the NTD-Ge thermistor, the resistance of which is strongly dependent on temperature. This composite system is operated at a temperature at which rhenium is in a superconducting state, of 100 mK, where the thermal response to an energy deposition is maximal. An excellent detector performance has been achieved with this arrangement; an energy resolution σ of 13 eV and a low energy threshold of 100 eV.

In order to be sensitive to spectral distortions arising from a finite anti-neutrino mass, it is imperative that one has a complete understanding of all residual spectral distortions which are of atomic origin. The contribution of one particular atomic effect, *Beta Environmental Fine Structure*, arising from the ^{187}Re beta decay occurring not as an isolated atom but rather within a rhenium crystal lattice potential will be reported in this article. This atomic effect, termed *Beta Environmental Fine Structure (BEFS)* was first hypothesized by S.Koonin[2]. It is an effect which should occur theoretically in low energy nuclear beta-decay in analogy to the *Extended X-ray Absorption Fine Structure (EXAFS)* effect[3,4]. In X-ray absorption experiments oscillatory fine structure is observed on the photo-absorption cross-section due to the emitted photo-electron undergoing oscillations within the *crystal lattice* potential. In direct analogy, the emitted beta electron (out going electron wavefunction) will be reflected by the scattering potentials of the surrounding atoms in the crystal, and from the incoming waves a modification to the final state wavefunction of the beta electron will result. Thus an energy dependent modulation in the probability of beta emission will occur and should be evident as an oscillatory structure in the detected beta energy spectrum.

2. THE DESCRIPTION OF *BEFS* IN A RHENIUM SINGLE CRYSTAL

The description of the fractional *BEFS* modulation in β decay, $\chi(E)$ is given by,

$$\chi(E) = \sum_i \frac{N_i |f_i(\pi)|}{kR_i^2} sin\left(2kR_i + \phi_i + 2\delta_0\right) e^{-\gamma R_i} e^{-2\sigma_i^2 k^2} \tag{1}$$

where the sum is over all neighbouring shells of atoms (labeled by i), $f_i(\pi)$ is the amplitude for an electron of energy $E = \hbar^2 k^2/2m$ to backscatter from atom i, $\phi_i = $ Arg $f_i(\pi)$, δ_0 is the s-wave phase shift induced by the decaying central atom, γ is the energy dependent inverse

beta electron mean free path, R_i is the distance between the central decaying atom and the surrounding shell of atoms and σ_i is the correction for zero-point and thermal lattice motion occurring in the Debye-Waller exponential term.

To date, this effect has *not* been observed experimentally in any existing beta spectral study. However there are many features of the cryogenic beta spectral study of ^{187}Re that should prove favorable to such a positive observation. The first important aspect to note is that the cryogenic experimental conditions allow efficient electron detection down to an extremely low energy threshold of 100 eV. The *BEFS* effect should dominate in the energy range, less than a few keV, as at higher energies and corresponding shorter wavelengths the effects of zero-point and thermal motion (Debye-Waller effect) become more critical in the reduction of overall coherence. In addition, the temperature of operation, 100 mK and the high atomic mass of rhenium (186.2 a.m.u) should lead to respectively negligible thermal and zero point motion of the lattice which would otherwise also reduce the coherence. The high charge of the surrounding rhenium nuclei ($Z = 75$) should create a large backscattering amplitude and with the rhenium crystal lattice being hexagonally *close packed* there is also a high density of neighbouring atoms (scattering centres). Both these factors should result in an enhancement of the amplitude of *BEFS*. Finally, an important attribute of the detector will be the ability to resolve the structure of the individual oscillations, in a detailed way, due to the outstanding energy resolution of 13 eV.

The evaluation of each term of $\chi(E)$ in Equation 1 for *BEFS* applied to a perfect single crystal of rhenium will be comprehensively described in the following sections. The analysis will be made in analogy to that of *EXAFS* and Low Energy Electron Diffraction (*LEED*).

2.1 The Debye-Waller Effect

The lattice motion from zero point and thermal effects serves to reduce the coherence and results correspondingly in a reduced amplitude of the *BEFS* modulations. This damping is given by the Debye-Waller formula, $DW(k)$,

$$DW(k) = e^{-\sigma(\vec{k}-\vec{k}')^2} = e^{-\sigma(2k)^2} \text{ for } \vec{k}' = -\vec{k} \tag{2}$$

where $\vec{k}$ is the emitted beta wavevector and $\vec{k}'$ is the reflected wavevector and where σ is defined[5],

$$\sigma = \frac{3\hbar^2}{2Mk_B\Theta_D} \left[\frac{1}{4} + \left(\frac{T}{\Theta_D} \right)^2 \int_0^{\Theta_D/T} \frac{x \, dx}{e^x - 1} \right] \qquad (3)$$

where M is the mass of the atom, k_B is the Boltzmann constant and Θ_D is the Debye temperature (210 K for Re). The factor 1/4 in the brackets corresponds to the zero-point lattice motion. In this calculation, the mass was taken as the atomic mass of rhenium (186.2 a.m.u), and therefore averaged over the ^{185}Re and ^{187}Re isotopes. In the evaluation a numerical integration procedure was used. The results of $DW(E)$ for Re at a temperature of 100 mK are plotted in Figure 1. These are compared to the case for tritium embedded in a germanium counter at 80 K. The relevance of the tritium experiment is that it represents the only existing alternative study of low energy beta spectral properties. The motivation for such a study also lies in the sensitivity of the tritium beta spectrum to the mass of the electron anti-neutrino. The less significant effect of reduction in rhenium in comparison to germanium results from the lower temperature of operation and the higher mass of Re atoms. In conclusion, the $BEFS$ effect in rhenium should be considerably less suppressed from the Debye-Waller effect, with respect to tritium in germanium.

2.2 The Energy Dependence in the β Electron Mean Free Path

The description of the decay of the beta electron within the lattice is based on the arguments of Ashley and Doniach[4]. The electron wavelength will not be as in free space but will be influenced by the inner potential well inside the crystal. The assumption is made that the main effects of multiple scattering will be to attenuate the amplitude of the beta electron outgoing wave by transferring it to other partial waves in an incoherent way. For each shell of atoms (*shell* refers to a group of atoms located at a given distance from the central decaying atom, ie. the first shell corresponds to the first nearest neighbours, the second shell to the next nearest neighbours etc. it does *not* correspond to electron shells with an atom) one defines a spherically averaged transmission function, given by

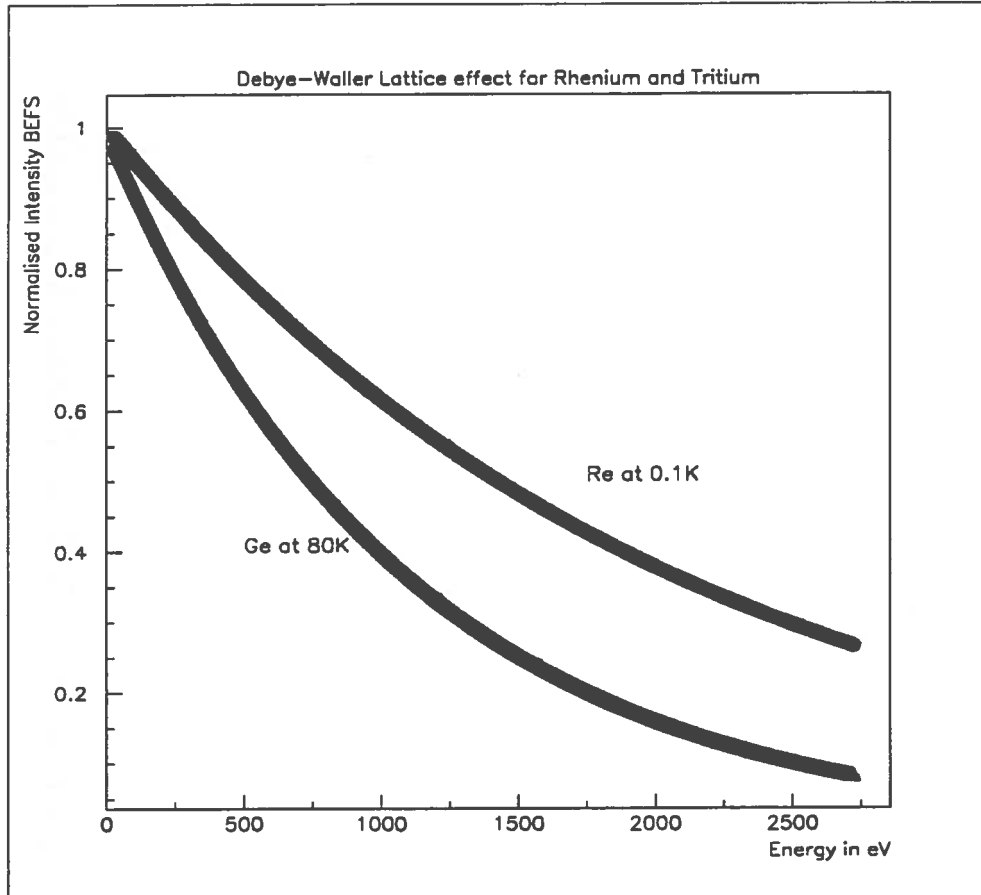$$W_i(k) = 1 - N_i \frac{\sigma_T(k)}{4\pi R_i^2}. \qquad (4)$$

Figure 1. The Debye-Waller lattice influence on the *BEFS* effect for a rhenium crystal and a tritium doped germanium counter.

where N_i are the number of atoms in shell i, R_i is the distance between the decaying atom and the atom shell i and σ_T is the total electron-atom cross-section. For rhenium the details of the atom *shells* were obtained from the Landbolt-Bornstein New Series and these are tabulated in Table 1 for the first 8 nearest neighbours. Rhenium is hexagonally close packed (hcp) and at room temperature has lattice parameters $a = 2.7609$ Å and $c = 4.4576$ Å and their ratio $c/a = 1.614$ compares well to the ideal ratio of hcp crystals, $c/a = 1.633$. There is a weak temperature dependence[8] in the amplitude of the lattice parameters ($T = 200$ K : $a = 2.7594$ Å, $c = 4.4552$ Å and $T = 100$ K : $a = 2.7586$ Å, $c = 4.4533$ Å) and assuming a linear dependence this extrapolates to $T = 0.01$ K to give $a = 2.7578$ Å, $c = 4.4514$ Å.

Table 1

The characteristics of the rhenium hcp lattice and the shells of neighbouring atoms extrapolated to 100 mK.

Shell i	Coordinates	Radius R_i Å	No. of Atoms N_i
1	$(a/\sqrt{3}, 0, c/2)$	2.7366	6
2	$(0, a, 0)$	2.7578	6
3	$(-2a/\sqrt{3}, 0, c/2)$	3.8851	6
4	$(0, 0, c)$	4.4514	2
5	$(5a/2\sqrt{3}, a/2, c/2)$	4.7644	12
6	$(a\sqrt{3}, 0, 0)$	4.7766	6
7	$(0, a, c)$	5.2364	12
8	$(0, 2a, 0)$	5.5156	6

The total electron-atom cross-section (inelastic and elastic) σ_T can be approximated by the total *elastic* cross-section σ_E as the inelastic contribution will be small, and thus σ_T can be obtained from the Optical theorem,

$$\sigma_E = \left(\frac{4\pi}{k}\right) \Im f(0). \tag{5}$$

The evaluation of the imaginary component of the forward scattering amplitude $\Im f(0)$ will be discussed in detail in Section 2.4 below where a detailed description of the calculation of the scattering amplitude is included. The attenuation factor $A_i(k)$ of the beta electron in the lattice is then given by the product of the transmission functions $W_i(k)$, where $A_1(k) = 1$ is defined for the first shell. As the factor $A_i(k)$ is defined only at 8 discrete atom shell

distances R_i, it is necessary to fit $[A_i(k)$ vs. $R_i]$ at each energy. In Figure 2 the range dependence of the attenuation factor is plotted at two given energies, 675 eV and 2700 eV.
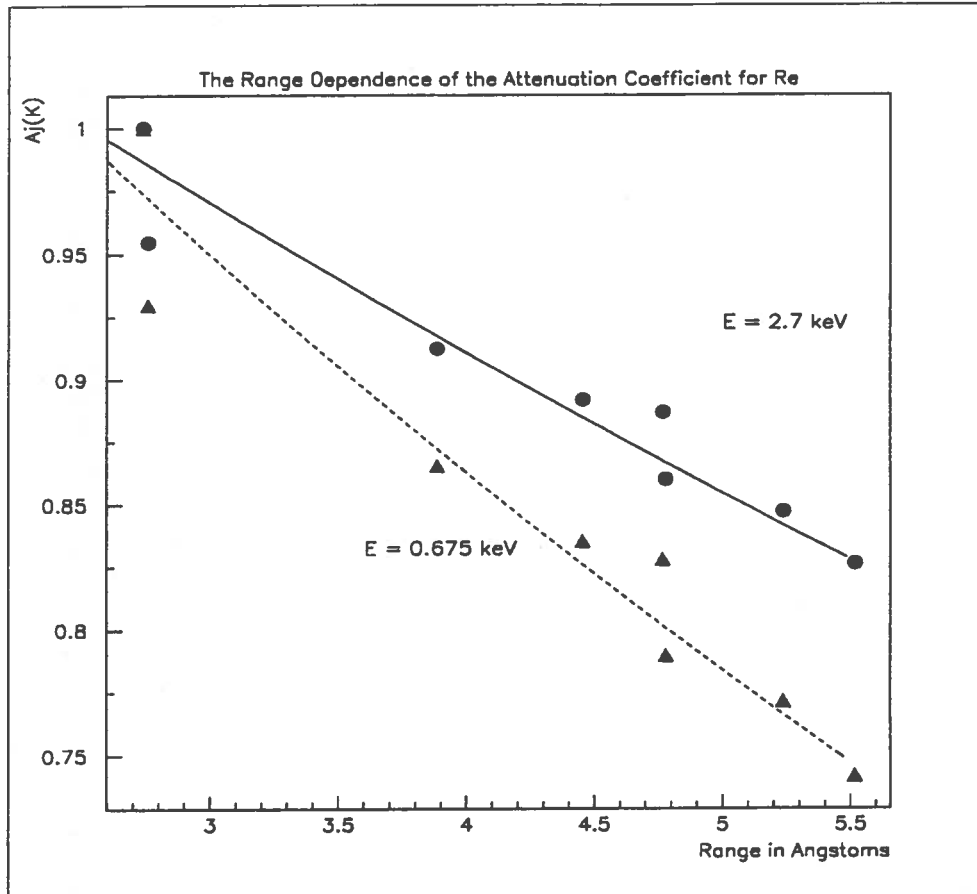

Figure 2. The range dependence of the attenuation coefficient shown at energy 2700 eV and 675 eV.

The applied fit to $A_i(k)$ is of the form $exp(\gamma_1 R_i + \gamma_2)$ and is fitted for each energy value considered, in intervals of 1.35 eV. The range dependence is defined in reference (4) as a simple exponential $exp(-\gamma_1 R_i)$ for copper and germanium lattices however for the rhenium lattice a more complex expression was required to fit the data. Despite differing attenuation expressions for (Cu, Ge) and Re, the tendency for longer mean free paths at higher energies is the same for all cases.

2.3 The Electron-Atom Phase Shifts

The critical aspect of calculating *BEFS* relies on the evaluation of the electron-atom phase shifts and the corresponding scattering amplitude within rhenium. The modulus of the back scattering amplitude, $|f(\pi)|$ determines the amplitude of the *BEFS* and the argument $Arg[f(\pi)]$ contributes to the period of the modulations.

From elementary scattering theory, to describe the magnitude of scattering of the emitted beta electron from the surrounding rhenium atoms, the calculation of the phase shift is required. The electron-atom phase-shifts have been calculated following the prescription of J.B.Pendry[3,6]. The computer program to calculate phase shifts by Pendry is included in the appendix of reference (6) and this has been used to evaluate the rhenium phase shifts. Prior to working with rhenium, a study of the energy dependence of the phase shifts of aluminium and copper was undertaken to allow a comparison to published phase shifts and good agreement was found. In order to calculate the corresponding phase shifts for rhenium, the input parameters include the atomic wavefunctions of rhenium. These atomic wavefunctions have been tabulated by Herman and Skillman[7] for all elements.

The rhenium atom is represented as $[Xe]4f^{14}5d^56s^2$ where $[Xe]$ corresponds to the electron *core* states and $4f^{14}5d^56s^2$ to the *valence* electron states. The effect of electron screening of the rhenium nucleus on the potential observed by the emitted electron has to be considered. It is included in the program by taking into account the wavefunctions of the electrons in the *core* states and representing the *valence* electrons as a uniform distribution of charge density within the potential sphere (the atom potential is termed 'muffin tin' potential). The *core* states of rhenium, $[Xe]$ are equivalent to $1s^22s^22p^63s^23p^63d^{10}4s^24p^64d^{10}5s^25p^6$. The *core* state wavefunctions (as a function of radius) of each electron shell were supplied as input parameters to the program. The charge density ρ of the *n valence* electrons ($n = 21$), was defined as $\rho = n/(4/3\pi R_{max}^3)$. The radius R_{max} was assigned the value at which the rhenium atom potential $U_R < 0.1$, and the specific value of R_{max} for this condition was found in the tables of Herman and Skillman. For rhenium, $\rho = 7.679$ and $R_{max} = 0.8675$. The program then calculates self-consistently within the Hartree-Fock approximation, the phase shift of the electron being scattered including interaction with the atomic wave functions.

From the selected input parameters, the phase shifts were calculated in the energy range 1.3 eV – 2700 eV for angular momentum values $L = 0 - 11$. The energy range of validity associated with the phase shifts $L = 0 - 11$ is in fact approximately 2.7 keV from the
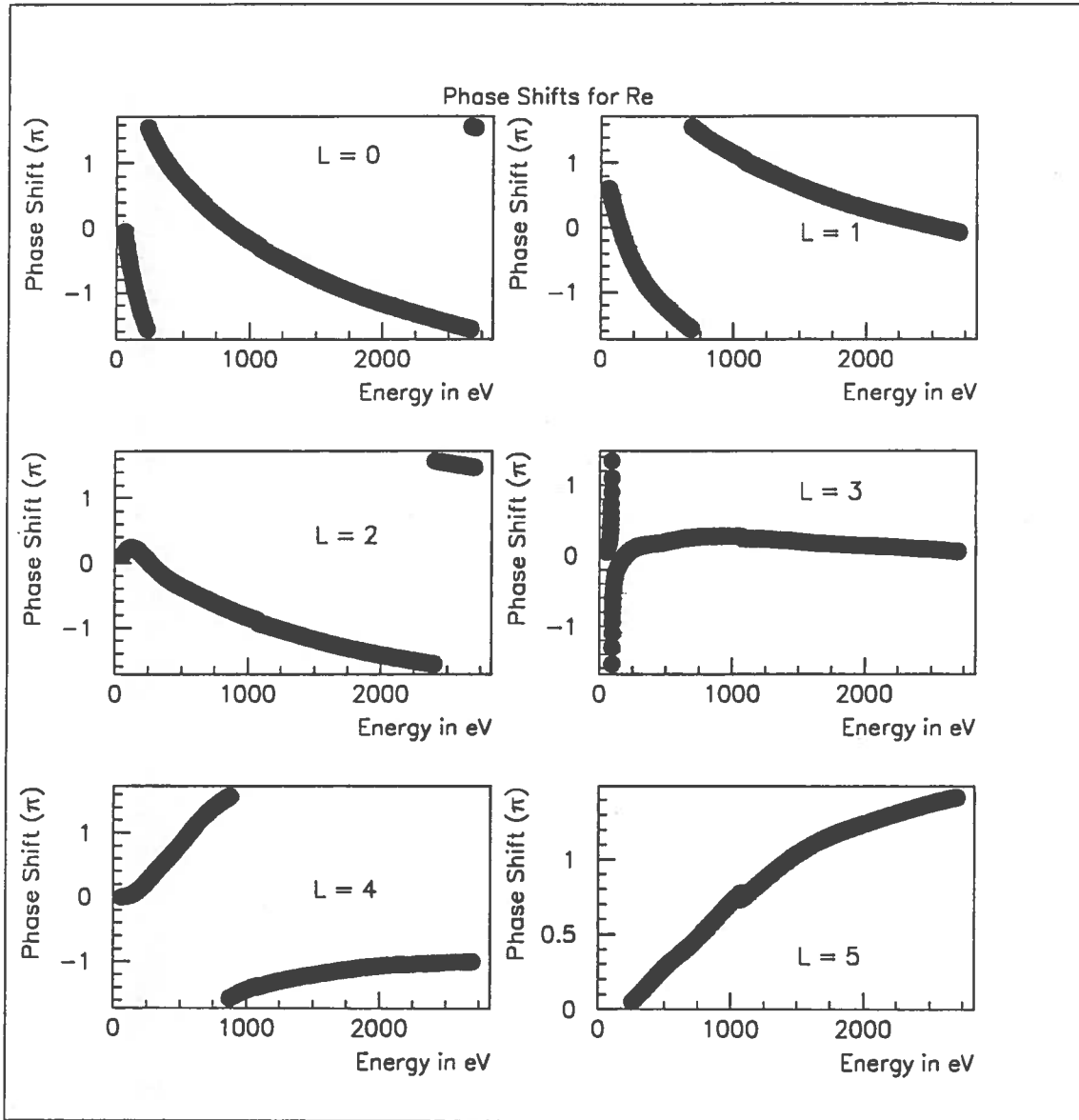
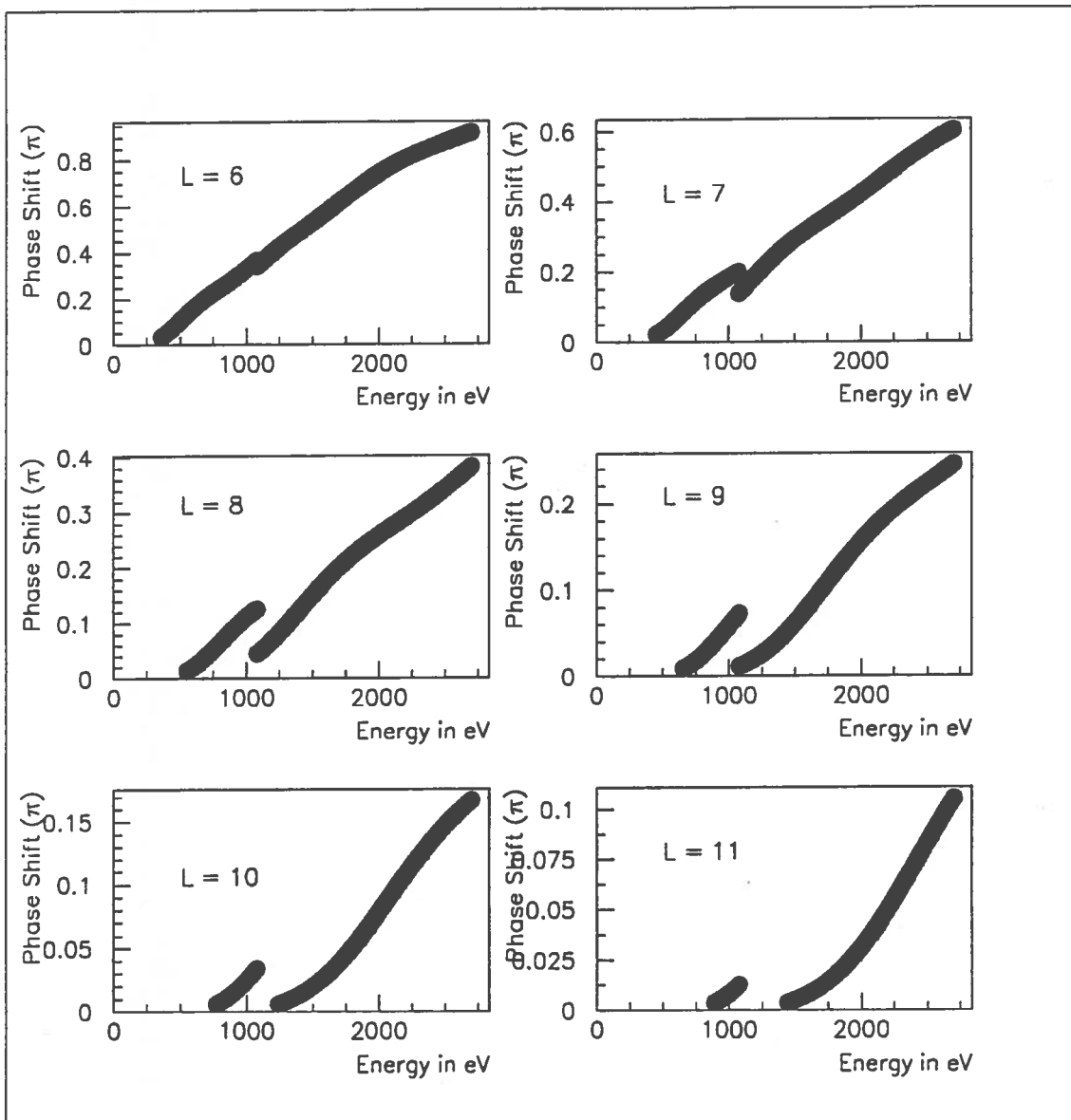Figure 3. The atom-electron phase shifts for angular momentum from 0 to 5.

Figure 4. The atom-electron phase shifts for angular momentum from 6 to 11.

relation, $kR_{max} < L$. The computed phase shifts δ_L for Re are plotted in Figure 3 and Figure 4 for the angular momentum values $L = 0 - 11$.

The s-wave phase shift, δ_0 is included in the *BEFS* expression in Equation 1. There appears to be an apparent problem of discontinuity in the phase shifts at 1.076 keV and the origin of this requires to be found.

2.4 The Scattering Amplitude

The amplitude $f(\theta)$ to scatter the beta electron through an angle θ due to the potential of the Re crystal is given by,

$$f(\theta) = \frac{1}{k} \sum_{L=0}^{\infty} (2L + 1)\, e^{i\delta_L}\, sin(\delta_L) P_L(cos\theta) \tag{6}$$

and the forward scattering amplitude, $\Im f(\theta = 0)$ required in the Optical theorem (see Section 2.2) can be found, where the Legendre Polynomial $P_L(cos\theta) = 1$. The backscattering amplitude $f(\pi)$ relevant to *BEFS* in both modulus and argument form was calculated for $L = 0 - 11$ and they are plotted in Figure 5 and Figure 6 respectively, where the region of validity is less than 2.7 keV.

3. THE COMPUTED *BEFS* EFFECT IN A SINGLE CRYSTAL OF RHE-NIUM

The *BEFS* structure applicable to ^{187}Re beta decay in a single perfect crystal was calculated from Equation 1 by the simple combination of the terms described in the preceding sections. The structure is illustrated in Figure 7 in the energy range 50 eV to 2.6 keV and in more detail in Figure 8 in the energy range 250 eV to 1.6 keV.

The most important feature to note is the large amplitude of this oscillatory effect in rhenium that is predicted below 1 keV. This effect should be evident experimentally on the residual beta spectrum even with reasonably low statistics. In addition, the period of the oscillations should be resolvable with the experimental energy resolution of 13 eV.
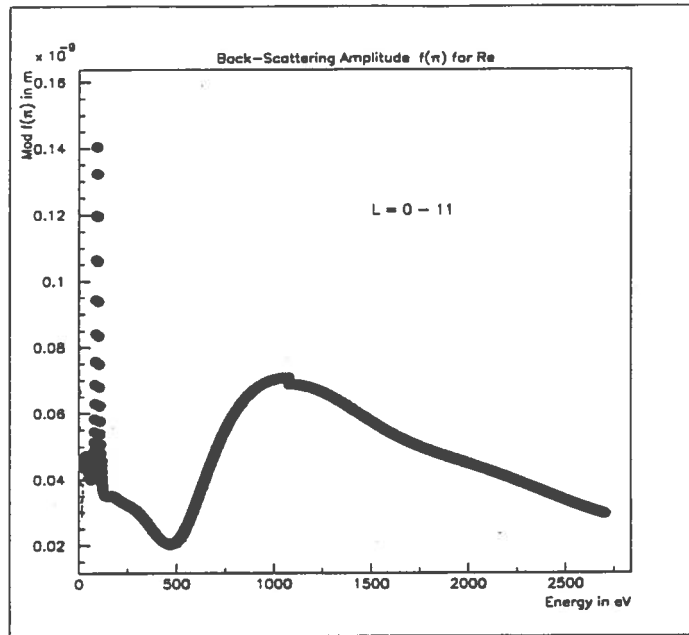
Figure 5. The energy dependence of the modulus of the back-scattering amplitude of an electron from a rhenium atom.
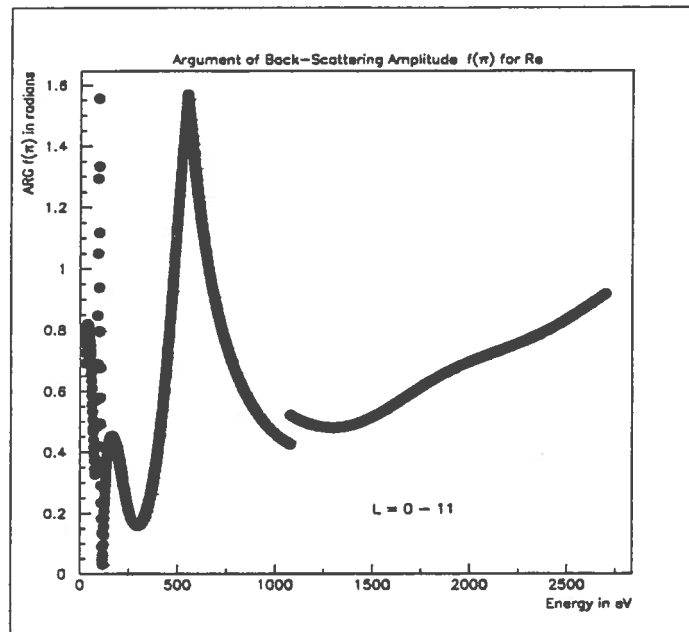

Figure 6. The energy dependence of the argument of the back-scattering amplitude of an electron from a rhenium atom.
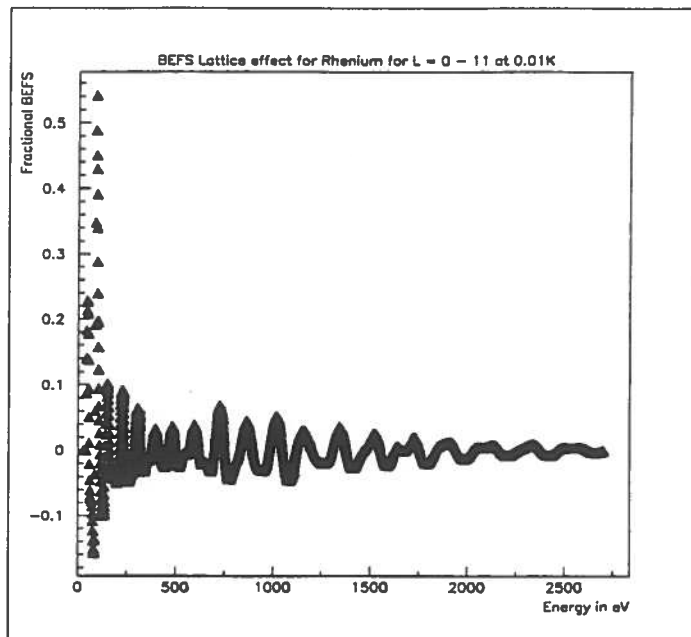
Figure 7. The *BEFS* lattice effect applicable to beta decay in a rhenium single crystal.
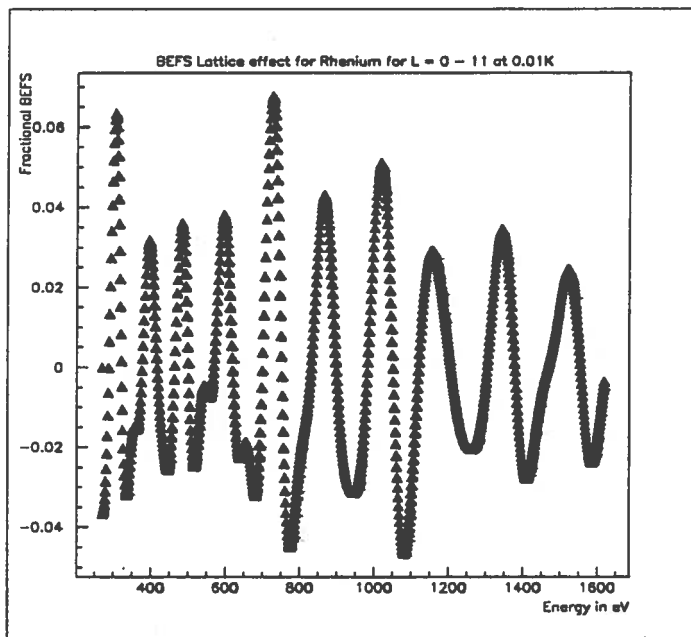

Figure 8. The *BEFS* lattice effect applicable to beta decay in a rhenium single crystal.

4. CONCLUSION

The proposed experimental measurement of the ^{187}Re beta spectrum in the energy range, 100 eV $\leq E \leq$ 2.6 keV for anti-neutrino mass purposes allows one also the opportunity to study in parallel higher order atomic effects on the beta spectrum, hitherto unobservable. Such an effect termed *Beta Environmental Fine Structure* in the ^{187}Re beta decay has been evaluated to be of an amplitude experimentally observable in a low temperature environment.

The potential applications of such an atomic effect include quantitative analysis of the internal structure of materials in an analogous manner to the surface studies of materials using low energy electron diffraction. By implanting the ^{187}Re isotope within a sample one could extract structural information by studying the residual fine structure in the beta spectrum. This could provide a powerful diagnostic tool for material analysis inaccessible by low energy electron diffraction studies.

REFERENCES

(1) E.Cosulich, F.Fontanelli, G.Gallinaro, F.Gatti, A.M.Swift and S.Vitale, Nuc. Phys. A **592**, 59 (1995).

(2) S.E.Koonin, Nature **354**, 468 (1991).

(3) P.A.Lee and J.B.Pendry, Phys. Rev. B **11**, 2795 (1975).

(4) C.A.Ashley and S.Doniach, Phys. Rev. B **11**, 1279 (1975).

(5) D.W.Jepson, P.M.Marcus and F.Jona, Phys. Rev. B **5**, 3933 (1972).

(6) J.B.Pendry, *Low Energy Electron Diffraction*, Academic Press, (1974).

(7) F.Herman and S.Skillman, *Atomic Structure Calculations*, Prentice Hall Inc., (1963).

(8) V.A.Finkel *et al.* Journ. of Phys. and Chem. of Solids **31**, 1345 (1970).

APPENDIX 1 - ESTIMATION OF *BEFS* IN RHENIUM FOIL

The cryogenic measurement of the [187]Re beta decay will also be made with a rhenium foil substrate replacing the rhenium single crystal. The preceding analysis for *BEFS* applicable to an ideal single crystal of rhenium can be extended to the case of a polycrystalline rhenium foil. One can estimate that the rhenium foil structure will only have periodicity from the first two neighbouring shells and that beyond the second neighbouring shell, coherence will be lost. The validity of this first estimation can be verified by taking the Fourier transform of the experimental *BEFS* modulations and by constructing the corresponding rhenium lattice foil structure. With the above formalism and by replacing eight neighbouring shells with only the first two shells, the *BEFS* structure is predicted to be of the form illustrated in Figure 9.
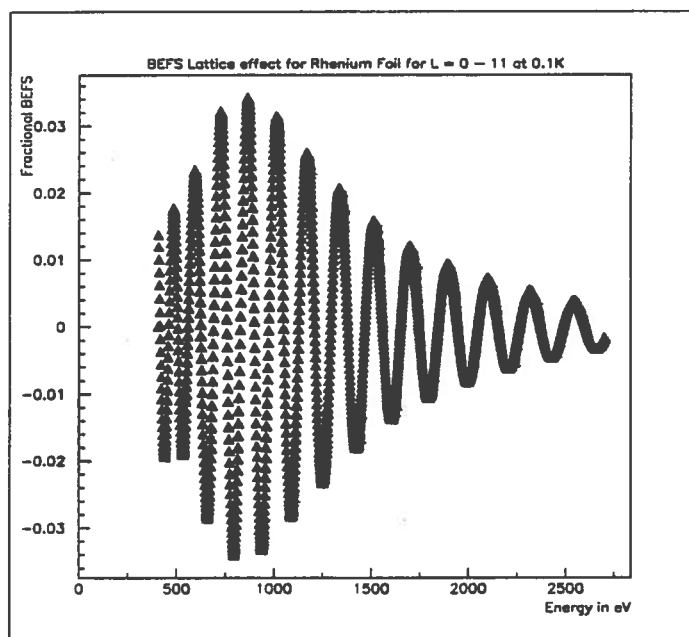

Figure 9. The *BEFS* modulations associated with a polycrystalline rhenium foil.

One observes, as expected, a smaller amplitude of modulations and less secondary structure in the oscillations in comparison to the single crystal of rhenium.