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Measurement of the Energy Spectrum of ^{60}Ni Recoils following ^{60}Co Decay using Nuclear Resonance Fluorescence

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Abstract

The energy spectrum of ^{60}Ni recoils from ^{60}Co β decay in a vapour $CoBr_2$ source has been studied by measuring the profile of the 1·33 MeV γ rays emitted by ^{60}Ni. The profile was deduced from the measured cross sections for resonance scattering of the 1·33 MeV γ rays in coincidence with the preceding 1·17 MeV γ rays. The results show fair agreement with the profile calculated for free ^{60}Co atoms. The influence of molecular Coulomb fragmentation and chemical binding are discussed.

Introduction

The energy spectrum of recoil ^{60}Ni nuclei from the β decay of ^{60}Co is determined primarily by the kinematics of the β-decay process (Wu and Moszkowski 1966) and by molecular effects in the source. Possible molecular effects include chemical binding and Coulomb fragmentation of the molecule. It is well known that electron-capture decays cause the atoms in a molecule to become highly charged by multiple Auger processes (Snell *et al.* 1961). Coulomb repulsion of these highly charged atoms will cause the molecule to virtually explode, leading to high recoil velocities of the component atoms (Carlson and Milford-White 1965). These high recoil velocities can cause Doppler broadening of γ rays emitted from the recoil nucleus which can sometimes be observed as an enhancement of the resonance scattering of γ rays. This effect has been observed in the electron-capture decay of a number of nuclei including ^{65}Zn (Metzger 1968), ^{114}In^{m} (Schumacher *et al.* 1971) and ^{75}Se (Borchert *et al.* 1972).

Beta decay causes much less ionization in the daughter atom than is the case for electron capture. In a β decay, multiple ionization of the atom is produced mainly by 'shake-off' of electrons by the adiabatic change in nuclear charge (Carlson *et al.* 1961). The probability of Coulomb fragmentation of the molecule is much lower than for electron-capture decays and the Coulomb repulsion is weaker. The only radioisotope for which enhancement of resonance scattering due to this effect has been observed is ^{131}I (Berman and Beard 1970; Langhoff 1971).

A better understanding of the processes involved in Coulomb fragmentation following β decay could extend the range of elements which can presently be analysed using γ-ray resonance scattering (Sowerby 1973; Sowerby *et al.* 1975). For example, in the analysis of Co, Zr and W there is insufficient energy available in the β decay of ^{59}Fe, ^{91}Y and ^{182}Ta respectively to compensate for recoil energy losses. Coulomb fragmentation in a molecular source could possibly supply the required energy.

The chemical binding energies of $CoBr_2$ and $NiBr_2$ are about 3 eV (Cottrell 1958). Even though the energy in the β decay of ^{60}Co is 318 keV, the ^{60}Ni recoil has a maximum recoil energy of only 2·9 eV. Measurement of the energy spectrum of ^{60}Ni recoils will give information on possible chemical binding effects.

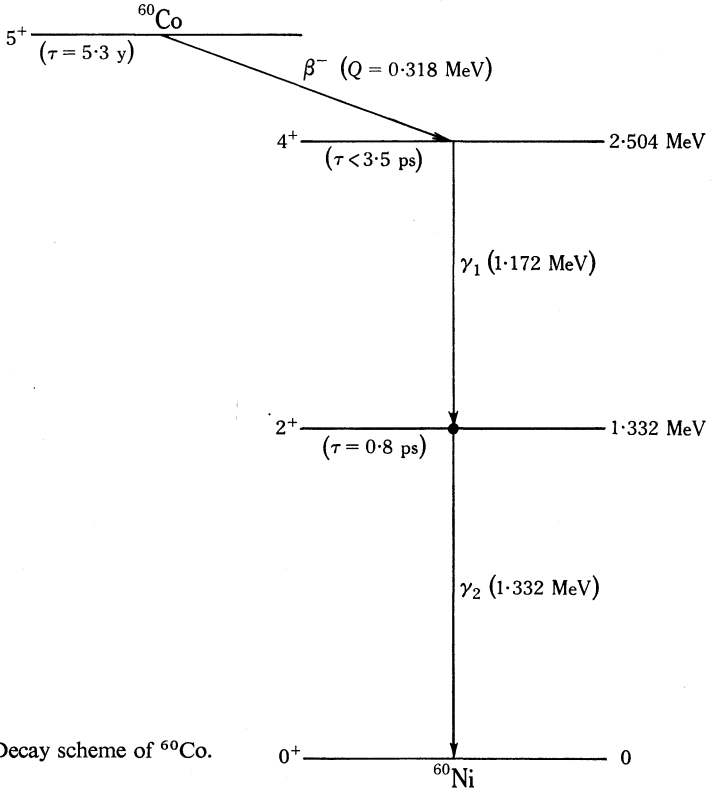

Fig. 1. Decay scheme of ^{60}Co.

In the present experiment, resonance scattering of 1·33 MeV ^{60}Ni γ rays is used to study the ^{60}Ni recoil energy spectrum in a vapour $^{60}CoBr_2$ source by means of the coincidence technique described below. ^{60}Co is favourable for this study as the energies in the decay permit a substantial fraction of the recoil spectrum to be measured and the ^{60}Co decay scheme is well understood. Also the ^{60}Ni recoil spectrum is very difficult to measure by any other technique.

Description of Coincidence Method

In the decay of ^{60}Co shown schematically in Fig. 1, the ^{60}Ni nucleus receives a recoil velocity $\boldsymbol{v}_r$ from the electron and neutrino emission and an additional velocity $\boldsymbol{v}_{\gamma 1}$ from the emission of γ_1. The velocity $v_{\gamma 1} = E_{\gamma 1}/M_f c$, where $E_{\gamma 1}$ is the energy of γ_1, M_f is the mass of the radioactive fragment and c is the velocity of light. The energy shift of γ_2 will be

$$\Delta E_{\gamma 2} = \{(\boldsymbol{v}_r . \boldsymbol{n}) - v_{\gamma 1} \cos \phi\} E_{\gamma 2}/c, \qquad (1)$$

where $\boldsymbol{n}$ is the direction of γ_2 and ϕ is the angle between the directions of γ_1 and γ_2. For resonance scattering to take place

$$\Delta E_{\gamma 2} = E_{\gamma 2}^2 / M_{\mathrm{Ni}} c^2 , \tag{2}$$

where M_{Ni} is the mass of the ^{60}Ni nucleus. Therefore the energy shift from the β component of the nuclear recoil is

$$E = (\boldsymbol{v}_r . \boldsymbol{n}) E_{\gamma 2}/c = E_{\gamma 2}^2 / M_{\mathrm{Ni}} c^2 + (E_{\gamma 1} E_{\gamma 2} / M_f c^2) \cos\phi . \tag{3}$$

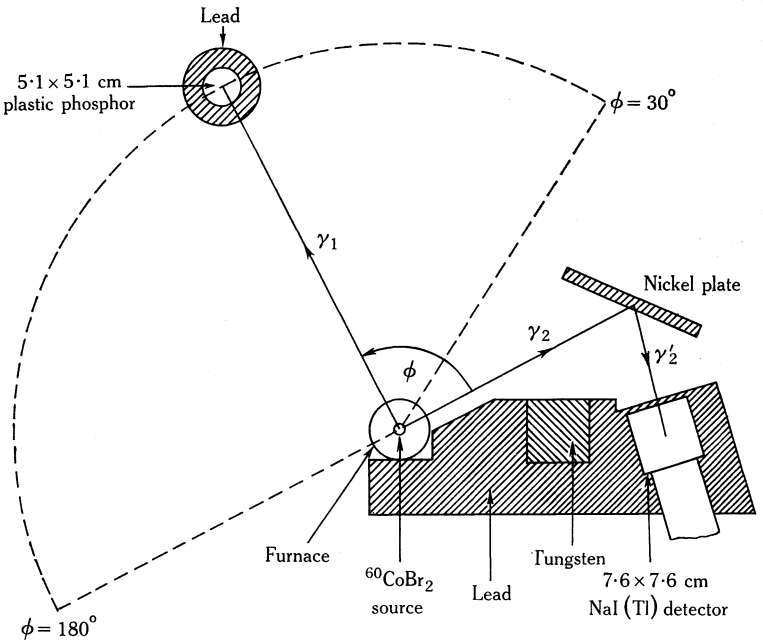

Fig. 2. Scale drawing of the experimental apparatus. The energy spectrum of the scattered γ_2 radiation is registered in coincidence with γ_1 radiation.

In the present experiment, coincidences are measured between γ_1 and resonantly scattered γ_2 as a function of angle ϕ. This yields the cross section for resonance scattering $\sigma(\phi)$ which is directly proportional to $N(E)$, the profile of the γ_2 emission line due to ^{60}Ni recoil from the β decay only.

A convenient way of understanding the experimental method is to think in terms of vector additions of the recoil velocities. The ^{60}Ni recoil velocity $\boldsymbol{v}$ prior to the emission of γ_2 is the vector sum of the recoil velocities $\boldsymbol{v}_r$ and $\boldsymbol{v}_{\gamma 1}$ following β decay and γ_1 emission respectively. The magnitude and direction of $\boldsymbol{v}_{\gamma 1}$ and $\boldsymbol{v}$ are fixed for a given experimental configuration, and so for each value of ϕ there corresponds a unique value of $\boldsymbol{v}_r$ as determined by equation (3).

The present experimental results were analysed using the assumption $M_{\mathrm{Ni}} = M_f$. The experimental arrangement shown in Fig. 2 allows variation in ϕ from 30° to 180°, and therefore $N(E)$ is accessible over the range $E = 3{\cdot}8$–$55{\cdot}3$ eV. The maximum γ-ray energy shift expected from β decay is $13{\cdot}5$ eV.

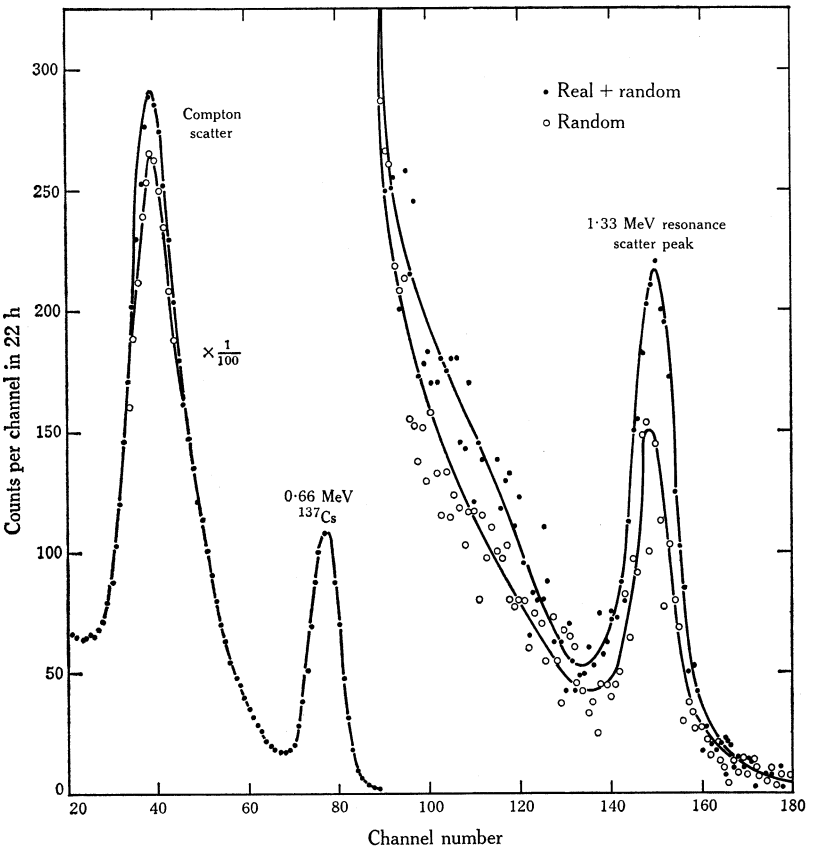

Fig. 3. Spectra of scattered γ_2 radiation coinciding with γ_1 obtained with a vapour $^{60}CoBr_2$ source for $\phi = 180°$.

Experimental Details

Source Preparation

The experiment was carried out using a 12·1 mCi ($4{\cdot}5 \times 10^8$ Bq) gaseous $^{60}CoBr_2$ source. A vapour source is required to ensure that slowing down times of the recoil nuclei are long compared with the lifetimes of the ^{60}Ni excited states. The source ampoule was doubly enclosed in welded stainless steel cans and heated to about 1000°C. Scans of the heated ampoule, using a lead slit assembly, indicated that >95% of the activity was in vapour state at this temperature. The internal pressure in the ampoule at the operating temperature of 1000°C was about 130 kPa which corresponds to a vapour density of about 8×10^{18} molecules cm^{-3}.

Experimental Procedure and Results

The coincidence count rate was measured as a function of ϕ over the range 30° to 180° using the apparatus shown in Fig. 2. The time resolution (FWHM) of the coincidence circuit was less than 3 ns. A pulse height spectrum of the coincidence scattered radiation in the NaI(Tl) detector is shown in Fig. 3 for $\phi = 180°$. The gain of the NaI(Tl) detector was stabilized on the 0·66 MeV γ-ray peak from a small ^{137}Cs source placed near the detector.

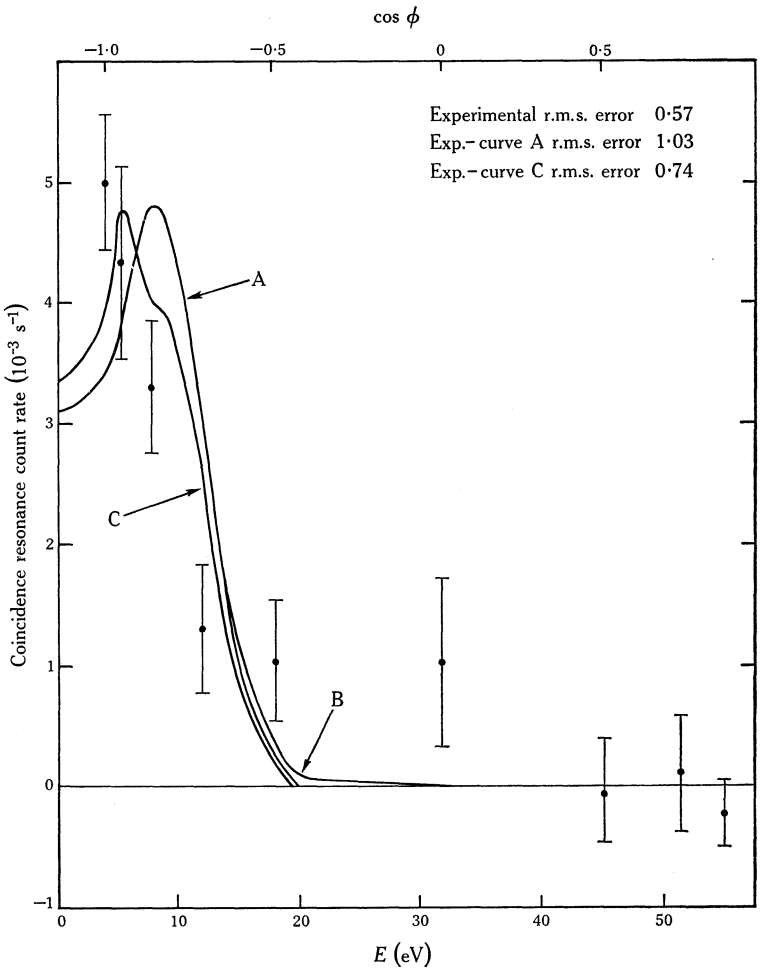

Fig. 4. Experimental and calculated results for the profile of the 1·33 MeV ^{60}Ni γ ray from a $^{60}CoBr_2$ source. The energy E is the γ-ray energy shift from the β component of the nuclear recoil and it is related to the angle ϕ by equation (3). The experimental results are the differences between the real coincidence count rates with vapour and solid sources. The calculated curves A, B and C are discussed in the text.

At each angle, measurements were made with a vapour source for a counting time in excess of 22 h. Measurements in excess of 60 h were made for about half the angles. At each angle, the measurements were repeated for an equal time using a cooled (i.e. solid) source. Real and random coincidence count rates were determined at the same time. The experimental results shown in Fig. 4 were obtained by differencing the real coincidence count rates obtained with vapour and solid sources. Counting statistics could be improved significantly only by increasing times to a minimum of several weeks per angle.

The results in Fig. 4 have been corrected for the angular distribution of γ_2 in coincidence with γ_1. This angular distribution has been previously measured to be (Raman 1968)

$$W(\phi) = 1 + 0{\cdot}102\,P_2(\cos\phi) + 0{\cdot}009\,P_4(\cos\phi),$$

where $P_2(\cos\phi)$ and $P_4(\cos\phi)$ are Legendre polynomials. This angular distribution was verified in the present experiment by replacing the nickel plate with a shielded NaI(Tl) detector and measuring coincidences between it and the plastic phosphor as a function of ϕ.

The relatively large experimental error bars shown in Fig. 4 are mainly due to the high random coincidence counting rate in the present experiment. The random coincidence count rate at the resonance photopeak was approximately 8×10^{-3} s^{-1} while the real coincidence count rate had a maximum value of about 5×10^{-3} s^{-1}, as shown in Fig. 4. The real coincidence count rate at the photopeak from a solid source was negligible for $\phi \geqslant 90°$ but at smaller angles it gradually increased to about $1{\cdot}2\times10^{-3}$ s^{-1} at $\phi = 30°$. This real coincidence count rate is caused by pair production in the lead shields and nickel plate.

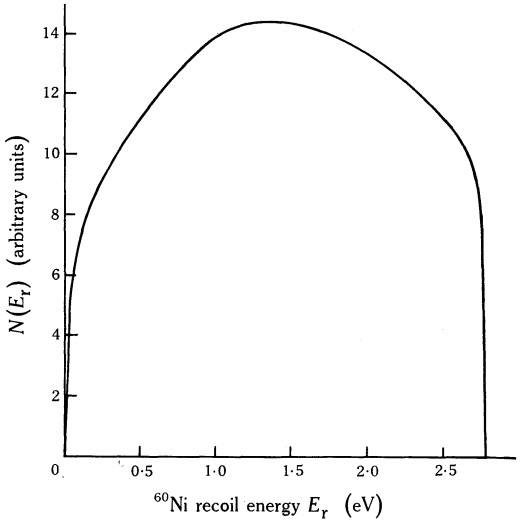

Fig. 5. Calculated energy spectrum of ^{60}Ni recoils following β and neutrino emission from ^{60}Co.

Discussion

To examine molecular effects on the energy spectrum of the ^{60}Ni recoils, the profile of the 1·33 MeV γ-emission line was calculated for free ^{60}Co nuclei. ^{60}Co decays by a pure Gamow–Teller interaction with an angular correlation coefficient $a = \frac{1}{3}$ and so its recoil energy spectrum can be calculated exactly (Johnson *et al.* 1963; Wu and Moszkowski 1966). The calculated energy spectrum of the ^{60}Ni recoils is shown in Fig. 5. The profile calculated for this energy spectrum is shown as curve A in Fig. 4. This profile has been smoothed for the effects of the angular resolution of the experimental apparatus of approximately $\pm12°$ (about ±4 eV) (mainly due to the size of the nickel plate) and the thermal width of the absorption line ($\pm1{\cdot}3$ eV FWHM). The results have been arbitrarily normalized to the experimental data.

The charge distribution on ^{60}Ni nuclei following ^{60}Co β decay is expected to be (Carlson *et al.* 1961): charge 1 (83%), 2 (10·7%), 3 (3·2%), 4 (2·0%), 5 (0·9%) and 6 (0·3%). The total energy release in the Coulomb fragmentation of a molecule with charges Z_1 and Z_2 is $Z_1 Z_2 e^2/4\pi r$, where r is the interatomic spacing. Calculation of the recoil energy spectrum due to Coulomb fragmentation is difficult for a triatomic

molecule. If we make the simplifying assumption that the Br_2 remains intact during the fragmentation, the calculations are much simpler even though the ^{60}Ni recoil velocities will be overestimated. The profile calculated on this assumption is shown as curve B in Fig. 4. Clearly, the present experimental results are not sensitive to small changes in the high energy tail of the profile.

If the molecule does not break up following the β decay, the recoil velocity of the fragment is much reduced and the maximum γ-ray energy shift from the β recoil is reduced to 7·1 eV, compared with 13·5 eV for atomic recoil. If the molecule also remains intact for the γ emissions, the energy range accessible to the experiment shrinks from 3·8–55 eV to 24–38 eV. Therefore, if the molecule remains intact throughout the decay scheme, the coincidence count rate observed in the present experiment will be zero. However, if the molecule breaks up after the β decay but before the emission of γ_1, the energy range accessible to the present experiment remains at 3·8–55 eV but the maximum energy shift from the β recoil will be 7·1 eV. The calculated lineshape for 20% of the molecules remaining intact until before γ_1 emission is shown in Fig. 4 as curve C. The r.m.s. deviation for curve C is 0·74 and for curve A 1·03. For a 'correct' profile calculation there is only a 5% probability that the r.m.s. deviation will exceed 1·03.

In summary, the experimental results show only fair agreement with calculations based on atomic recoil only. The effect due to molecular Coulomb fragmentation is small and would not be seen in the present experiment. There is some evidence that about 20% of the molecules remain intact during the β decay but break up before γ_1 emission.

Acknowledgment

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