EXTRANUCLEAR EFFECTS WITH FAST RECOILING NUCLEI

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Summary

Experimental results are given on angular distributions in the reaction $^{19}F\ (p,p'\ \gamma)$ proceeding via the second excited state of ^{19}F, with targets in different physical and chemical states. It is noted that effects of extranuclear interactions in flight, in solid or liquid media, should be negligible. Tentative information is obtained about the fate of recoil nuclei in the solids NaF and CaF_2. The interpretation is consistent with some effects observed in α-γ angular correlations from ^{241}Am in solid sources.

I. Introduction

In the study of angular correlations of successive nuclear radiations, it is well known that, if an intermediate state is formed whose lifetime is sufficiently long ($<10^{-9}$ sec), the nucleus may interact with extranuclear fields in such a way that the degree of anisotropy observed may be considerably attenuated. The process may be viewed as one in which the intermediate state has been prepared with spatial orientation of its nuclear spin with respect to some axis. This spin orientation may subsequently be made to change via the interaction of the nuclear magnetic dipole or electric quadrupole moment with an appropriate external field, with corresponding attenuation of the angular distribution of the second radiation about the preferred axis. The degree of attenuation can be related to the magnitude of the interaction nuclear moment. The theory of these phenomena (Abragam and Pound 1953) explains well most experimental results, as is discussed in the review article by Steffen (1955).

The present problem deals with the special case where the intermediate nucleus recoils far from its original site, and extranuclear forces are related to energy loss phenomena in the medium. These interactions may be treated very similarly to those obtaining with a nucleus in thermal equilibrium with a liquid medium, a problem dealt with explicitly by Abragam and Pound. This is discussed in Section III of the present paper. In Section IV a further discussion is given of the effect of the environment when the intermediate nucleus comes to rest before emission of its radiation, and because the recoil is so violent one must investigate what chemically stable ions may be formed, whether they are magnetic or not, and whether they come to rest in a region of strong electric field gradient. In the present case it is suggested that strong attenuations of the anisotropy of radiation observed in certain solids are due to formation of free paramagnetic atoms or ions, whose spins must be expected to relax very quickly.

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II. Experimental Data

In the reaction $^{19}F(p,p'\gamma)$ at a proton energy of 873 keV, the ^{19}F recoil nucleus is formed preferentially in its second excited state (Ajzenberg and Lauritsen 1955) whose lifetime (Fiehrer *et al.* 1955) is $(1\cdot24\pm0\cdot04)\times10^{-7}$ sec, i.e. sufficiently long for extranuclear effects to be important. Measurements on the nuclear magnetic moment of this state have been made in liquid (Lehmann, Lévêque, and Fiehrer 1955) and solid targets (Treacy 1956), and we are here concerned with interpreting the attenuations, in the absence of externally applied fields, in solids. In Table 1 are listed values of the attenuation coefficient G_2 in different target media. Here G_2 is defined by the angular distribution of 197 keV γ-radiation

$$W(\theta)=1+G_2B_2P_2(\cos\theta),$$

where B_2 is the " true " undisturbed coefficient of the Legendre polynomial P_2.

The polycrystalline targets were made by vacuum evaporation on copper. By microscopic examination they were found to be of crystal size about 10^{-4} cm.

Table 1

Attenuation coefficient for γ-rays from $^{19}F(p,p'\gamma)$

Target Material	Form	G_2 (Error $\pm0\cdot05$)	Reference*
CF_4	Solid, temp. —196 °C	0	1
CF_4	Gas, press. 2·5 cm Hg	0·06	2
MnF_2	Polycrystalline	0·16	2
CaF_2	Polycrystalline	0·27	1 (cf. 3)
NaF	Polycrystalline	0·61	2

* References: 1, Treacy (unpublished data); 2, Treacy (1956); 3, Barnes (1955).

Table 1 shows that in three cases the value of G_2 is less than the minimum theoretically possible (0·2) in the presence of a static perturbing field. Reasons will be given below why time-dependent fields are to be expected in some of these.

III. Attenuations during Recoil

The ^{19}F nucleus recoils initially with a mean kinetic energy of 70 keV and, as is discussed by Treacy (1956), must undergo many collisions, successively capturing and losing electrons several hundred times, before coming to rest. The perturbation suffered by the nucleus can be viewed as a time-dependent one like that experienced by a nucleus from thermal agitation in a liquid. In such a problem one considers the possibility of precession of the nuclear spin with frequency ω, which remains coherent only within a correlation time τ_c. If $\omega\tau_c\ll1$, then the attenuation coefficient G_k of $P_k(\cos\theta)$ in the expansion of $W(\theta)$ is

$$G_k=\frac{1}{\tau_N}\int_0^\infty \exp(-t/\tau_N)\exp(-\lambda_k t)\mathrm{d}t, \qquad \ldots\ldots\ldots\ldots (1)$$

where τ_N is the nuclear mean lifetime and λ_k is proportional to $\omega^2\tau_c$. Equation (1) applies to magnetic or electric relaxation phenomena, and explicit forms for λ_k

are known in each case (Abragam and Pound 1953). In the present case τ_c and hence λ_2 is not constant as the nucleus slows down, so the integral

$$G_2=1/(1+\lambda_2\tau_N) \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (2)$$

has meaning only for an average value of λ_2.

An approximate description has been given of the attenuation in the CF_4 gas target (Treacy 1956), in terms of a succession of static interactions due to the large number (effectively 150) of electrons captured and lost. As the values used for ω and τ_c were such that $\omega\tau_c$ was about 10^{-2}, the time-dependent theory should apply, and, using a mean calculated value of λ_2, the spatial average* of G_2 becomes approximately $0\cdot3$. In order to make it coincide with the value $0\cdot06$, it is necessary to assume that ^{19}F spends approximately 60 per cent. of the time as a magnetic atom, compared to the figure 10 per cent. taken from properties of neon ions in gases (Massey and Burhop 1952) and used by Treacy (1956). Considering the great uncertainty in applying the data to fluorine, the difference is of no significance. The approximate picture is not altered on any reasonable assumption as to the effect of elastic scattering in the gas. One may use a reasonable compromise (in the absence of experimental data) to the true situation by assuming the hard-sphere approximation (Massey and Burhop 1952) with energy loss cross section proportional to the recoil energy, up to that necessary ($5\frac{1}{2}$ eV) to ionize the negative ion of fluorine, and constant for higher energies. On this assumption the slowing down time is of order 10^{-7} sec, and λ_2 and consequently G_2 are not substantially different from those already quoted.

Turning to the case of a target of condensed matter, with density 10^4 times that of the gas, the slowing down time is reduced to about 10^{-11} sec, an insignificant interval compared to the nuclear lifetime. Therefore contributions in flight to G_2 (equation (1)) are negligible. The condition $\omega\tau_c \ll 1$ must of course be fulfilled, but could only be violated by a value of ω greater than 10^{14} sec^{-1}, which would be highly improbable in any of the usual collision processes.

IV. Attenuations due to Chemical Effects

The type and magnitude of attenuation experienced by the nucleus after it reaches thermal energy depend on the chemical properties of the ion formed. The problem may be viewed as one in radiation damage, and it is usually assumed (Seitz 1949; Kinchin and Pease 1955) that displaced atoms are likely to enter interstitial sites and stay there until allowed to return to lattice sites by annealing.

In the relatively simple problem of ^{19}F in the molecular solid CF_4 one may argue that in slowing down from the energy necessary to remove one electron from the atoms (28 eV) to that necessary to ionize the negative ion ($5\frac{1}{2}$ eV) the nucleus is likely to come to rest as a neutral atom. (The likelihood of this depends on the smallness of the hard-sphere scattering assumed in Section III.)

* As is shown by Treacy (1956), for some of the recoil time the ^{19}F nucleus is in a negative ion, of no magnetic moment, so during this time the contributions to the angular distribution are not attenuated.

If truly free, the resting atom should produce a " hard core " magnetic attenuation corresponding to $G_2=0{\cdot}69$, statistically weighted from the $^2P_{3/2,\frac{1}{2}}$ states of the fluorine atom. In fact, as Table 1 shows, attenuation is complete, i.e. $G_2=0$. This may be attributed to the familiar relaxation mechanisms observed in paramagnetic resonance (Bleaney and Stevens 1953). Here the stable ^{19}F nuclei in CF_4 molecules should produce a temperature-independent spin-spin relaxation time of 10^{-7} sec or less. Such an effect underlines the difficulty of obtaining free paramagnetic atoms in solids. Paramagnetic relaxation may also be responsible for the strong attenuations observed in the α-γ angular correlation from ^{241}Am in solids (Fraser and Milton 1954 ; Novey 1954). Here one expects the ^{237}Np intermediate state to be in one of its paramagnetic oxidation states $+4$ to $+6$ (Wahl and Bonner 1951). There are other α-γ correlations, in $^{228,\,230}$Th (Steffen 1955) and $^{224,\,226}$Ra and ^{238}Pa (Milton and Fraser 1954) which are appreciably less attenuated in solids, and which would not be expected to have paramagnetic ions in the intermediate states. These seem to show quadrupole attenuations of the same order of magnitude as those to be discussed below.

In the case of ^{19}F nuclei recoiling in polycrystalline solids with ionic bonding the fact that attenuations are relatively weak leads one to guess that the interstitial atoms must be non-magnetic. Assuming the G_2-values from Table 1 for NaF and CaF_2, we may use the theoretical estimate (Barker 1956 and personal communication) for the quadrupole moment of the 197 keV level of ^{19}F ($Q=8\times10^{-27}$ cm^2) to calculate the apparent field gradients at the ^{19}F nuclei at rest. The results are as follows :

NaF	Field gradient	$0{\cdot}9\times10^{14}$ e.s.u.
CaF_2	Field gradient	$4{\cdot}1\times10^{14}$ e.s.u.

The figure for CaF_2 is not very significant as the attenuation is nearer the " hard core ", which corresponds to infinite field gradient, and we have no knowledge from a magnetic decoupling experiment, as we have for NaF (Treacy 1956) that magnetic effects are absent.

The remaining case of MnF_2 quoted in Table 1, where G_2 is certainly less than the " hard core " value, is not amenable to any simple description similar to the idealized cases considered above.

V. Conclusions

It has been noted that, because of the usual magnetic interactions with stable nuclei, paramagnetic atoms suffer fast relaxation in solids. As was pointed out by Frauenfelder (1951) the ability of disturbed nuclei to exhibit strong angular correlations depends on their being non-paramagnetic. In one case, namely the α-γ correlation in ^{241}Am, the fast relaxation observed is probably due to magnetic interactions ; this could be verified by a magnetic decoupling experiment (Goertzel 1946).

It is evident that for a proper description of the slowing-down of ions to thermal energies in solids, more experimental data on energy-loss cross sections are required. The problem is of importance in radiation damage studies, where the effects of ionization should be important with recoil energies well below those at which it is usually neglected (Seitz 1949). Further studies are obviously needed of radiation damage effects on the microscopic, i.e. atomic, scale. The interesting experiments of Grace *et al.* (1955) on the effects of radiation damage on nuclear orientation, illustrate one possible approach to the problem.

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VII. References

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