Using Eq. (53) from AK we have

$$\sum_l \tfrac{1}{2}\mu_l p_{lk} = \sum_{lk} \epsilon_k p_{0k} p_{lk} p_{lK}$$
$$= \sum_K \epsilon_K p_{0K} \delta_{kK}$$
$$= \epsilon_k p_{0k}. \quad \text{(C8)}$$

Thus

$$\Gamma_{kq} = (\epsilon_k - \epsilon_{k+q})\Theta_{kq} - \sum_{k_1} \epsilon_k p_{0k} p_{0k_1} (C_{k+q\uparrow}{}^\dagger C_{k_1\uparrow} - C_{k+q\downarrow}{}^\dagger C_{k_1\downarrow}) + \sum_{k_1} \epsilon_{k+q} p_{0\,k+q} p_{0k_1} (C_{k_1\uparrow}{}^\dagger C_{k\uparrow} - C_{k_1\downarrow}{}^\dagger C_{k\downarrow}). \quad \text{(C9)}$$

Transforming back to the scattering-state representation,

$$\Gamma_{kq} = (\epsilon_k - \epsilon_{k+q})\Theta_{kq} - \epsilon_k p_{0k} \sum_l p_{l\,k+q} (a_{l\uparrow}{}^\dagger a_{0\uparrow} - a_{l\downarrow}{}^\dagger a_{0\downarrow}) + \epsilon_{k+q} p_{0\,k+q} \sum_l p_{lk} (a_{0\uparrow}{}^\dagger a_{l\uparrow} - a_{0\downarrow}{}^\dagger a_{l\downarrow}). \quad \text{(C10)}$$

Taking averages in the perturbed state

$$\langle \Gamma_{kq} \rangle = (\epsilon_k - \epsilon_{k+q}) \langle \Theta_{kq} \rangle + (\epsilon_{k+q} - \epsilon_k) p_{0k} p_{0\,k+q} \times \langle a_{0\uparrow}{}^\dagger a_{0\uparrow} - a_{0\downarrow}{}^\dagger a_{0\downarrow} \rangle. \quad \text{(C11)}$$

Inserting this into Eq. (18) yields Eq. (36). In the final step we have used the result

$$\langle a_{l\uparrow}{}^\dagger a_{0\uparrow} - a_{l\downarrow}{}^\dagger a_{0\downarrow} \rangle = \langle a_{0\uparrow}{}^\dagger a_{0\uparrow} - a_{0\downarrow}{}^\dagger a_{0\downarrow} \rangle \delta_{l0}, \quad \text{(C12)}$$

which may be proven quite simply from consideration of the equation of motion of the operator.

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Influence of Radio-Frequency Magnetic Fields on the Mössbauer Effect in Magnetic Co^{57} Sources*

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Experiments are described that demonstrate the destruction of the Mössbauer hyperfine pattern by the action of a radio-frequency magnetic field. Two possible mechanisms for the effect are considered: magnetostriction and domain-wall passage. Calculations make magnetostriction seem unlikely, leaving as the probable cause the fluctuations in the γ-ray energy resulting from the alteration in the direction of the hyperfine field after the passage of a domain wall. This is calculated with the aid of the theory of motional narrowing in NMR and a demonstration by Peshkin of the correspondence between the γ ray and the NMR case. The hypothesis of 180° walls is found to be inadequate, and a less restrictive assumption is needed.

INTRODUCTION

A REPORT[1] at the Allerton House Conference on the Mössbauer effect described a magnetic-resonance method of measuring the g factor of the 14.4-keV excited state of Fe^{57} in iron metal. The technique was to apply a steady field of a few hundred oersteds parallel to the plane of a foil and a rf magnetic field of a few oersteds also in the plane but perpendicular to the dc field. A change in the transmission of the radiation from a Co^{57} source in an iron lattice was sought for at the frequency of 26 MHz, which corresponds to the separation between successive hyperfine levels of the excited nuclear state. The possibility of such an effect arises from the fact that the hyperfine field of 330 kOe lies parallel to the atomic moment, and thus even quite weak external rf fields, by changing the direction of the magnetization, cause directional changes in the hyperfine field and hence equivalent large hyperfine rf fields. Theory shows[2] that such fields produce a resonant line splitting, equivalent to a frequency modulation of the γ-ray line. This would result in an increase in transmission. Such an effect was apparently seen, but an obscuring nonresonant effect resulted in a large change in transmission between rf on and rf off, at frequencies well away from 26 MHz. The present work arose in part in an effort to understand this phenomenon.

An additional aim in starting the research was to attempt to perform a series of simple experiments in γ-ray optics similar in principle to the sort of optical experiments discussed by Righi.[3] For example, if the linearly polarized radiation emerging perpendicular to

* Work performed under the auspices of the U. S. Atomic Energy Commission.

[1] G. J. Perlow, University of Illinois, Allerton Park Conference Report, 1960 (unpublished). This talk presents work done by E. C. Avery, C. Littlejohn, G. J. Perlow, and B. Smaller. A more recent experiment showing the resonance by rf means is reported by E. Matthias, University of California, Lawrence Radiation Laboratory Report No. UCRL-17877 (unpublished).

[2] M. N. Hack and M. Hamermesh, Nuovo Cimento 19, 546 (1961).

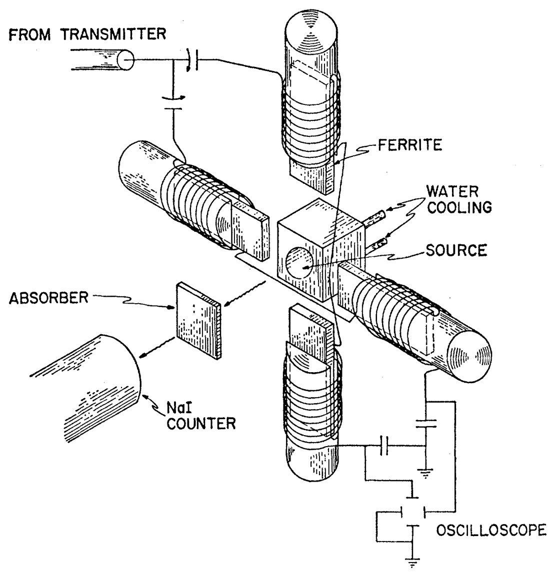

FIG. 1. Apparatus for applying a rotating rf magnetic field to the source. The two series LC circuits are fed in parallel and are tuned $\frac{1}{2}\pi$ apart, straddling the resonance frequency.

a magnetized foil containing a source be decomposed into its counter-rotating circular polarizations, and if the plane of the linear polarization be rotated at constant angular speed, one circular component will increase and the other decrease in frequency. In the Mössbauer effect this would show up as a line splitting. The rotation of the magnetization direction in the source by means of a rotating rf field, if it is appreciably slower than the Larmor frequencies, provides a simple way of rotating the plane of polarization. This aim influenced the nature of the apparatus, but no successful conclusion was reached. As we shall see, the application of a rotating external rf field causes a relatively violent change in the hyperfine pattern, and this change is not related in any simple way to the idea of a rotating hyperfine field.

The experiments to be reported here were done on Fe^{57} in a variety of ferromagnetic environments and at a variety of frequencies away from resonance. The results are qualitatively similar for all cases although the causes are not necessarily so. The report is mainly on Supermalloy (79% Ni, 15% Fe, 5% Mo, trace Mn)[4] at 4.2 MHz, with which most of the data were obtained, and on Permalloy (78.5% Ni, 21.5% Fe) at 6.5 MHz.

EXPERIMENTAL METHOD

The substances examined were used as sources in the form of thin circular disks 1 cm in diameter. The Supermalloy source was a rolled foil[5] 3.2 μ thick into which carrier-free Co^{57} was diffused in a hydrogen atmosphere at 1000°C for 2 h. It was annealed more or less according to the prescription that results in minimum anisotropy field. There appear to be only small differences among the results obtained with different samples. The Permalloy sample was a special melt prepared by Karasek and rolled to 1.3 μ. The Co^{57} was diffused into it in hydrogen at about 800°C.

The active foil was held in a Lucite holder which was constructed to allow the sample to be cooled by a gentle current of water. A rotating rf field was obtained by means of coils at right angles (Fig. 1) in each of which the phase of an rf current could be controlled with external tuning. The magnetic field was enhanced by placing ferrite slabs in the coils. As a guide in minimizing the eccentricity of the rotating field, signals proportional to the coil currents were impressed on an oscilloscope to form a Lissajous pattern. The field could be measured by use of a small coil shielded against electrostatic influence. Radio-frequency power of about 50 W was available, but only some fraction of this could be used without overheating the insulating parts. Very little of the power (less than 1 W) was dissipated in the sample as measured by the temperature rise of the cooling water.

The Mössbauer absorber was either sodium ferrocyanide or stainless steel, both enriched in Fe^{57}. They were used at room temperature.

The results of a series of runs are shown in Fig. 2. Plots (a)–(f) were done with the Supermalloy source and ferrocyanide absorber at a frequency of 4.2 MHz. The amplitude of the rf field was held constant during a run and was varied from 0 to 5.4 Oe over the series. The spectrum without field has relatively broad lines because of the complexity of the alloy. The fact that the second and fifth lines have the greatest intensity shows that the magnetization lies fairly well in the plane of the foil. The foil is almost fully magnetized in the earth's field since Supermalloy attains 90% of saturation in only 0.1 Oe. In the progression from (a)–(f) it is seen that the hyperfine lines broaden, so that all that remains of the pattern at 2.7 Oe is a slight dip between the two halves of the spectrum. The effect is virtually saturated by 2.7 Oe—there is no significant increase in broadening upon doubling of the rf field strength. Plot (g) shows that increasing the frequency to 6.5 MHz makes no appreciable change in the final pattern.

The lines in the plot were obtained by a least-squares fitting process from which areas could be obtained. After correction for background, it was found that all areas were equal within ±3%, which may be regarded as the uncertainty in the determination. Thus the recoilless fraction f_s is unchanged by the rf and no measurable

[3] A. Righi, J. Phys. **2**, Ser. II, 437 (1883).
[4] R. M. Bozorth, *Ferromagnetism* (D. Van Nostrand, Inc., Princeton, N. J., 1951), p. 140.
[5] Supplied by Arnold Engineering Co., Marengo, Ill.

part of the spectrum is beyond the velocity limits of the experiments.

A series of auxiliary experiments was performed to eliminate various possible causes of the phenomena observed.

(1) The magnetic source was replaced by a non-

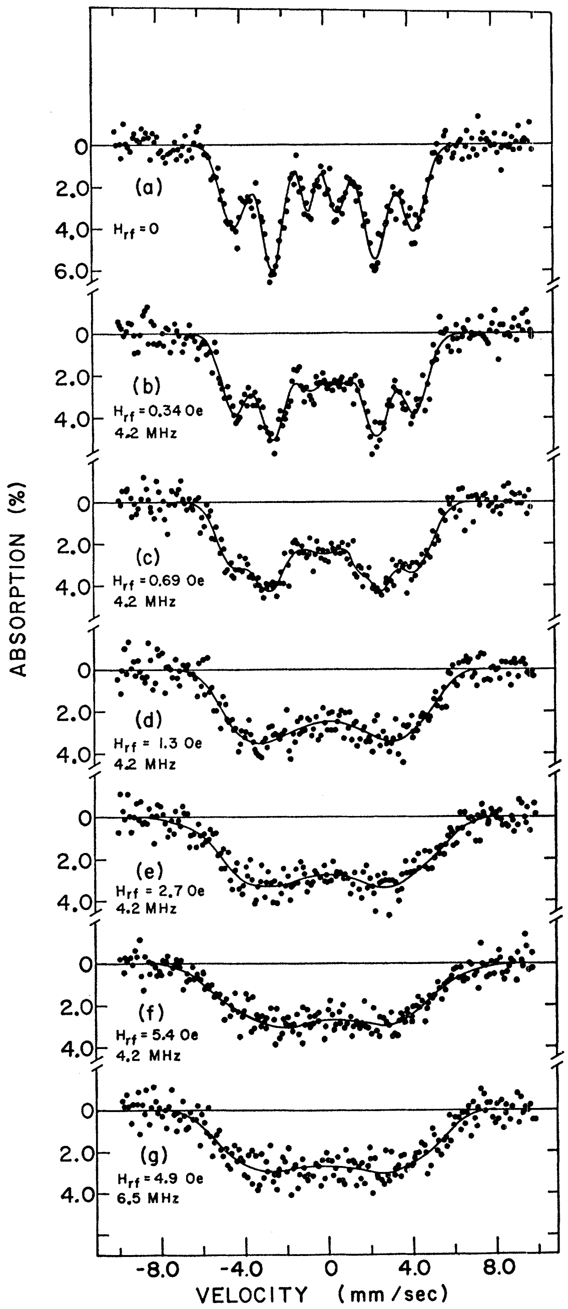

FIG. 2. The effect of the rf magnetic field. In runs (a)–(f), the field amplitude was increased from 0 to 5.8 Oe at 4.2 MHz. Run (g) was at 4.9 Oe and 6.5 MHz. The source was Co^{57} diffused into Supermalloy; the absorber was sodium ferrocyanide.

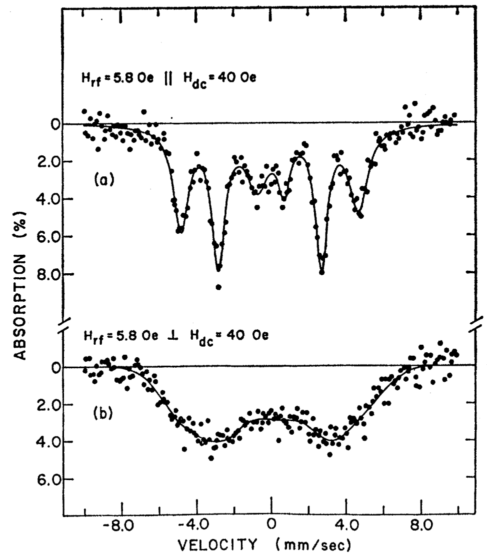

FIG. 3. Oscillating rf field, 5.8 Oe peak at 6.4 MHz, either parallel (a) or perpendicular (b) to a fixed magnetic field of 40 Oe. Both fields lie in the source plane. The source is Co^{57} in Permalloy, and the absorber is type 310 stainless steel.

magnetic one and spectra were obtained with and without rf. The rf had no effect on the spectrum.

(2) The circularly rotating rf field was replaced by a linearly oscillating one, by disconnecting one of the coils. The same sort of progressive broadening with field strength occurred.

(3) A dc magnetic field in the plane of the foil was added to the linear rf field of auxiliary experiment No. 2. It was oriented parallel to the latter in one set of runs and perpendicular in a second set. Figure 3 shows the results for the two cases with $H_{dc}=40$ Oe and $H_{rf}=$ 5.8 Oe (peak). The frequency is 6.4 MHz. The source is Permalloy in this case but the effects are similar for Supermalloy. The hyperfine pattern is substantially unaffected by the parallel fields but suffers strong broadening in the perpendicular case.

(4) The entire source assembly was rotated so that the plane of the foil made an angle of 45° to the axis along which the γ rays were observed. The results of a series of runs with varying rf rotating field strength were quite similar to those of the original experiments of Fig. 1.

DISCUSSION

The auxiliary experiments show that the phenomenon is entirely magnetic and that it occurs only when the applied field changes in direction as in case No. 2 and in the perpendicular case of No. 3. It is clear that the effect is not due to the uniform rotation of the magnetization; this would produce only a relatively small broadening of the lines. The data of Fig. 2(a) represent a mean hyperfine field of 260 kOe. With this value the excited-state and ground-state g values are 20.4 and 35.7 MHz, respectively. A doubling of each line with a separation between the components of only 4.2 MHz, while readily observable, would not give the pattern seen in Fig. 2(f). This argument depends on the assumption that the adiabatic approximation is valid for the nuclear moment in the rotating field. The rotation frequency is somewhat high for full validity, but a calculation by Peshkin[6] indicates that the line broadening due to this cause will be small under the conditions of the experiment.

Two possible explanations of the observed effect suggest themselves. The first is mechanical vibration due to magnetostriction and the second may be thought of as a modulation due to domain-wall motion. Both explanations are treated in detail below.

Magnetostriction

The result of magnetostriction would be to cause the emitting nuclei to oscillate about their equilibrium positions with a frequency double that of the applied rf frequency. The resulting Doppler effect causes the emitted γ radiation to be frequency modulated.[7] The modulation results in the production of sidebands separated by the modulation frequency ω_{mod}. The intensity of the nth sideband (n may have either sign) is proportional to $J_n^2(a)$, where the modulation index a is the ratio of the maximum frequency deviation to the modulation frequency. For modulation by the Doppler effect, we have $a=kx$, with $k=1/\lambdabar$ the wave number of the γ radiation, and x the amplitude of the displacement. The magnetostriction for a bar of length l in the unmagnetized state which changes by Δl upon saturation is characterized by the material constant $\Lambda_s=\Delta l/l$. The intensity of the nth sideband is therefore

$$I_n \propto J_n^2(\Lambda_s kl).$$

We may first consider magnetostriction along the direction of the foil thickness, for which (if we think of the central plane of the foil as being stationary) l has the value of 1.64×10^{-4} cm for an emitter on the surface. If we take[8] $\Lambda_s \leq 2\times10^{-6}$ and $k=7.30\times10^8$ cm, we obtain $a\leq0.23$. A trial shows that this value of the modulation index makes a scarcely perceptible change in the spectrum of Fig. 2(a).

It is of interest, however, to see whether the spectral shape can be fitted by magnetostriction if the modulation index is allowed to be as large as needed. This was done by Gimmestad, who programmed a computer to make the necessary convolution. The activity was assumed to be distributed uniformly throughout the foil and contributions were taken for modulation indices uniformly distributed between 0 and a value $a_{\max}$. The results appear in Fig. 4. The data points correspond to those with the same label in Fig. 2. The line is obtained from the computer calculation that is judged visually to fit best. The value of $a_{\max}$ appears to the right of each figure. The fits are remarkably good, at least to the eye, but require a value of the magnetostriction constant 25 times too large.

One possible way in which the modulation index could have been underestimated is in the choice of the factor l. There could be a transverse magnetostriction associated with motion along the diameters of the foil. In all of these experiments there is necessarily some divergence of the γ-ray beam; hence the diametral motion would have a component along the observation direction for some of the radiation. The effect of such a motion would be very strongly enhanced if the foil were turned so that its plane made some appreciable angle to the observation direction. As mentioned before (auxiliary experiment No. 4), the experiment was tried with negative results.

In the case of Permalloy, for which results quite similar to Supermalloy were obtained, the magnetostriction constant is also quite small ($\Lambda_s \approx 2\times10^{-6}$) and the same serious difficulties are raised by invoking

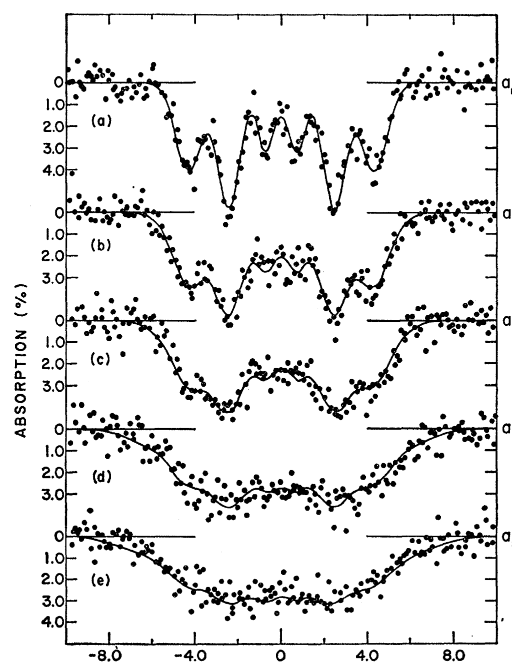

FIG. 4. Fit with magnetostriction mechanism. The points are from the correspondingly labeled experiment of Fig. 2. The line is calculated as the best visual fit for the maximum modulation index $a_{\max}$ shown at right.

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[6] M. Peshkin (private communication).

[7] S. L. Ruby and D. I. Bolef, Phys. Rev. Letters **5**, 532 (1960); T. E. Cranshaw and P. Reivari, Proc. Phys. Soc. (London) **90**, 1059 (1967).

[8] The magnetostriction constant for molybdenum Permalloy, which has similar composition, is quoted as 2×10^{-6} in Ref. 4, p. 675. Supermalloy with higher initial permeability is probably less.

magnetostriction as an explanation. Therefore if the destruction of the Mössbauer pattern is to be attributed to physical motion of the emitting nucleus, it is not magnetostriction as ordinarily understood that is responsible. On the other hand, there must be at least a small contribution from this mechanism.

Domain-Wall Motion

It would be naive to assume that under the rather primitive conditions of sample preparation used for these sources, the magnetization would change its direction in a uniform manner—always following the applied field. There is a considerable technology[9] on the alteration in the direction of magnetization in thin (usually 1000 Å or less) specially prepared films under the influence of external fields (usually of the switching type). In general, one finds that the alteration in magnetization direction is associated with a complex pattern of domain walls which form, move, and disappear. The magnetization has different directions on opposite sides of such a wall. Let us now imagine a radiating nucleus across which a number of such walls may move during its lifetime. The excited-state hyperfine energy $-\langle \boldsymbol{\mu}\cdot\mathbf{H}\rangle$ in general is different before and after the passage, and the same is true for the ground state. Therefore the transition energy changes during the emission and the spectrum is altered. It is likely that the spatial interval between successive walls is appreciably greater than the wall thickness. This leads to the approximation of considering the walls infinitesimally thin and the field direction as jumping abruptly at each passage. Its magnitude stays fixed. There is no reason to believe that there is any regular time interval between the traversals of successive walls—certainly not when averaged over all the radiating positions in the source. One must therefore consider a distribution of time intervals, subject to some average "jump time" τ. It is not the intent of this discussion to add to the considerable literature concerning detailed switching mechanisms in thin films. On the contrary, the aim is to avoid the subject and to characterize the details of the domain-wall motion by a single quantity, the jump time τ. It is chosen to fit the observed spectra if possible.

The net result of the wall traversals is to produce a random frequency modulation of the γ radiation. The resultant spectrum depends on some further assumptions. The simplest assumption about the directional changes in H is that there is a 180° reversal in field at each wall passage. The radiation problem can then be solved by the Weisskopf-Wigner[10] method. This has been done by Peshkin,[11] who shows that the solution for the spectrum is identical to that well known in the theory of narrowing due to chemical exchange in nuclear magnetic resonance.[12–15] In the latter the resonance frequency is taken as jumping between two values, which correspond in Peshkin's solution to the γ-ray energies $E(m_{\text{exc}}\rightarrow m_{\text{gnd}})$ and $E(-m_{\text{exc}}\rightarrow -m_{\text{gnd}})$. The final spectrum would be the sum of the three pairs of such partial spectra corresponding to the six allowed lines of the Fe^{57} transition. Another assumption amenable to calculation is that the frequency jumps randomly among the six values. It is a less stringent specification of the change in the direction of magnetization upon passage of a wall. This calculation can be taken over from NMR. It neglects the preference of the magnetization to remain in the plane of the foil.

As described by Abragam,[15] the spectrum can be written

$$I(\omega)=\text{Re}\{\mathbf{W}\cdot\mathbf{A}\cdot\mathbf{1}\}, \quad (1)$$

where $\mathbf{W}$ is a vector whose six components are the observed line areas in the absence of the rf perturbation, and $\mathbf{1}$ is the vector each of whose components is numerically 1. The matrix A is defined by

$$A_{kl}=i\delta_{kl}(\omega_k-\omega+\Gamma_k)+\pi_{kl}, \quad (2)$$

where the ω_k are the six original line positions and the Γ_k are the linewidths. Frequencies are measured relative to the center of the pattern. The quantity $\pi_{kl}dt$ is the probability that the nucleus while radiating the kth line at time t will jump to the lth in the subsequent interval dt. The diagonal elements are chosen so that $\sum_l \pi_{kl}=0$.

If the lines in the Fe^{57} spectrum are labeled $1,\cdots,6$ in order of increasing energy, then the alteration of m_{exc} to $-m_{\text{exc}}$ and m_{gnd} to $-m_{\text{gnd}}$ interchanges line 1 with line 6, 2 with 5, and 3 with 4. With this ordering for the line indices k and l, we have for the 180° wall case

$$\begin{aligned}\pi_{kl}&=(1/\tau)\times(-1), && l=k\\ &=(1/\tau)\times(+1), && l=7-k\\ &=(1/\tau)\times(0) && \text{otherwise.}\end{aligned} \quad (3a)$$

And for the random jump case:

$$\begin{aligned}\pi_{kl}&=(1/\tau)\times(-5), && l=k\\ &=(1/\tau)\times(+1), && l\neq k.\end{aligned} \quad (3b)$$

The evaluation of Eq. (1) was programmed for the CDC-3600 computer by Taraba. The results for various

[9] See, for example, R. F. Soohoo, *Magnetic Thin Films* (Harper & Row, New York, 1965).

[10] See, for example, E. G. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, New York, 1947), 2nd ed., p. 111.

[11] M. Peshkin (private communication).

[12] H. S. Gutowsky, D. W. McCall, and C. P. Slichter, J. Chem. Phys. **21**, 279 (1953).

[13] P. W. Anderson, J. Phys. Soc. Japan **9**, 316 (1954).

[14] R. A. Sack, Mol. Phys. **1**, 163 (1958).

[15] A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon Press, Oxford, England, 1961), p. 447.

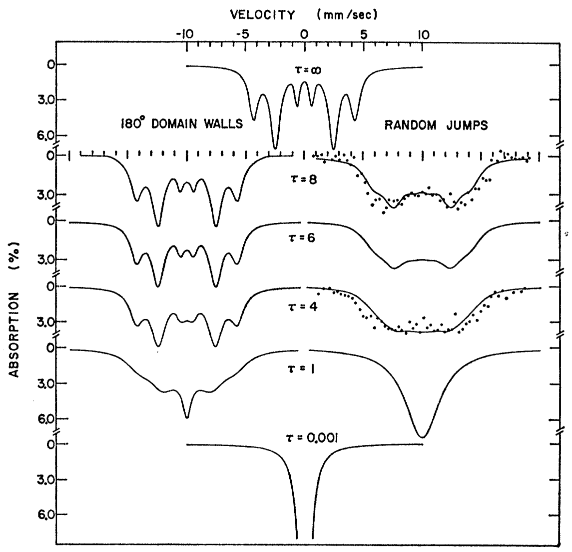

FIG. 5. Spectra calculated on the basis of 180° domain-wall passages (left side) and jumps among six emission frequencies (right side). The parameter τ is the jump time in appropriate units. The points shown at $\tau=8$ and $\tau=4$, correspond to those in Figs. 2(d) and 2(f), respectively.

values of τ are seen in Fig. 5 for both the 180°-wall and for the random-jump hypotheses. The parameter τ is in units appropriate to the measurement of angular frequency ω in mm/sec; i.e., τ (velocity units) $=k\tau$ (sec), with $k=1/\lambda\!\!\!^{-}$ the wave number of the γ ray. Then $\tau=1$ corresponds to 1.369×10^{-8} sec. The case $\tau=\infty$ is the same for both calculations; for values of τ greater than about 100, both calculations replicate the spectrum obtained with the rf turned off. Similarly, for very small values of τ the spectrum degenerates to a single line in the middle; the hyperfine magnetic field averages to zero. For intermediate values of τ the two cases show a considerable difference. It is not possible to get the flat spectrum of Fig. 2(f) with the restriction to 180° walls. The random-jump hypothesis, however, shows reasonable, if not perfect, agreement. Experimental points have been added to the $\tau=8$ and $\tau=4$ plots to demonstrate the fit. We find $\tau=4$ corresponds to Fig. 2(f), where $H_{\mathrm{rf}}=5.4$ Oe, and $\tau=8$ corresponds to Fig. 2(d), where $H_{\mathrm{rf}}=1.3$ Oe. However, the line narrowing that the theory predicts for higher jump frequency has never been seen.

Expressed in time units, $\tau=4$ corresponds to 5.5×10^{-8} sec. The excited-state Larmor frequency is 1.28×10^{8} rad/sec; hence $\tau=4$ represents an interruption at every 7 rad on the average. There are an average of 2.6 wall crossings during the mean life (1.4×10^{-7} sec) of the nuclear state.

The velocity v of a domain wall depends on many properties of the material, but an order-of-magnitude estimate can be made if it is assumed that the velocity is limited by eddy currents. In that case,[16,17]

$$v=[(3\times10^{9})/(B_s d)]\rho H,$$

where B_s is the saturation flux density, ρ is the resistivity, and d is the sample thickness. For the Supermalloy sample, $B_s=7900$ G, $\rho=60\times10^{-6}$ Ω cm, and $d=3.2\times10^{-4}$ cm. With these values, the velocity indicated by Fig. 2(f), for which $H=5.8$ Oe and $\tau=4$, is $v\approx4\times10^{4}$ cm/sec and the corresponding mean spacing between walls is $\approx2\times10^{-3}$ cm. Domain-wall spacings of comparable magnitude are often seen in thin films.[18]

In summary, one can fit the observed spectra by either a magnetostriction or a domain-wall hypothesis. The former requires exceptionally large values of the magnetostriction constant, and therefore probably supplies only a small part of the effect. The domain-wall mechanism does not require unrealistic values for domain-wall speeds and spacings and is therefore favored. On the other hand, any mechanism which interferes rapidly enough with the nuclear-spin precessions will lead to similar experimental findings. In that sense, it cannot be said that the domain-wall mechanism is proved. Yet it certainly must contribute a considerable part to the effect.

ACKNOWLEDGMENTS

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[16] H. J. Williams, W. Shockley, and C. Kittel, Phys. Rev. **80**, 1090 (1950).
[17] N. C. Ford, J. Appl. Phys. **31**, 300S (1960).
[18] See, for example, D. O. Smith, in *Magnetism* (Academic Press Inc., New York, 1963), Vol. 3, pp. 465–523.