${}^{i}\rho(c) \simeq {}^{i}\rho(0)/(1-c\,{}^{i}s)$, where ${}^{i}s = 1-({}^{i}\sigma_{sc}/{}^{0}\sigma_{sc})$, depending on whether ${}^{i}s$ is positive or negative. On balance, $\mathbf{E}_{1/2}(c)$ goes through a minimum if ${}^{i}s<0$ (cf. Figs. 3 and 4), but diminishes monotonically if ${}^{i}s>0$: The qualitative behavior of $\mathbf{E}_{1/2}(c)$ reflects the relative magnitude of ${}^{i}\sigma_{sc}$ and ${}^{0}\sigma_{sc}$.

In conclusion, small concentrations of impurities can induce Ps formation by subexcitation positrons and thereby change the Ps yield of a substance significantly. At a given impurity concentration, the increase of the Ps yield depends on the ratio of the Ps-formation cross section of the impurity and the energy-loss cross section of the subexcitation positrons [cf. Eq. (7) and Fig. 2]. The further rise in the impurity Ps yield under the influence of an electric field is regulated, in addition, by the elastic scattering cross section of subexcitation positrons [cf. Eqs. (12) and (15)]. The energy dependence of the cross sections can be studied to some extent through the choice of impurity. The expected magnitude of the impurity-induced changes in the positron lifetime spectra suggests that they can give experimental access in Ps-forming substances to scattering and energy-loss rates relevant for an understanding of electronic subexcitation processes in matter.

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Selective Excitation of Nuclear Sublevels*

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A method is described for selectively populating any one of the sublevels of a nuclear excited state by means of the Mössbauer effect. Specifically, a single hyperfine level of the 14.4-keV state of Fe^{57} was populated using a constant-velocity drive system, a single-line Co^{57} source, and a metallic Fe^{57} scatterer arranged in 90° scattering geometry. Polarization was detected by analyzing the scattered radiation with a second Mössbauer drive and a single-line absorber. Two applications of the selective excitation process are also reported. In the first, the energy distribution of the scattered radiation was studied as a function of the Doppler shift of the incident radiation. In the second, NMR transitions between the hyperfine sublevels of the 100-nsec state of Fe^{57} were directly observed.

THIS paper reports the development of a technique for producing and detecting excited-state *nuclear* polarization by means of nuclear resonance radiation (the Mössbauer effect). This is the first successful extension to nuclei of the concept of selective excitation of magnetic sublevels that was the basis of numerous experiments using atomic levels.[1] Two applications of the technique are also reported here. In the first, the energy distribution of the resonantly scattered radiation was measured as a function of the Doppler shift of the incident γ rays. In the second, NMR transitions in the 14.4-keV state of Fe^{57} were directly observed for the first time. This second application demonstrates that the problems associated with magnetically induced rf sidebands,[2] which completely obscured the results of earlier resonance experiments,[3,4] have been overcome, that the general technique of Mössbauer-effect detection of excited-state NMR is feasible, and that therefore investigations into the effects of resonant transitions upon the electromagnetic line shape can be performed. The results reported here indicate that such effects, if they exist at all, are smaller than predicted either by Hack and Hammermesh[5] or by Gabriel.[6]

The basic idea of the polarization process is as follows. A constant-velocity transducer (CVT) and a constant-acceleration transducer are arranged in a right angle with a scattering foil at the apex (see Fig. 1). The CVT supplies the proper velocity to a monochromatic source in order to Doppler-shift the recoillessly emitted γ rays into resonance with a transition to only one of the resolved hyperfine levels of nuclei in the scattering

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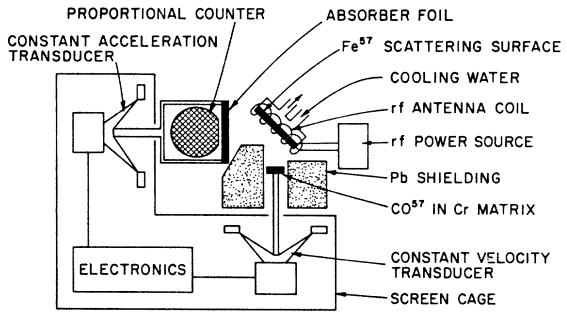

FIG. 1. Experimental geometry.

foil. Resonant absorption by nuclei in the foil results in the population of only this single hyperfine level. Observation of this excited-state polarization is accomplished by means of a standard transmission experiment in which the scatterer now plays the role of a stationary polarized source. The radiation, reemitted from the foil, is analyzed by a monochromatic absorber which is swept through a range of velocities by the constant-acceleration drive system. Transmission minima, indicating resonant absorption of the radiation from the scatterer, should occur only at the velocities corresponding to the transitions from the single hyperfine level of the nuclei in the scattering foil. Because of some apparent similarity between this procedure and that used for the production of polarized *radiation* (such as the experiments of Shtrikman[7]), it must be emphasized that we are describing a technique for polarizing excited-state *nuclei*, not just γ rays.

This polarization process can be understood by examining the results of the application of this technique to Fe^{57} (shown in Fig. 2). The top plot is an Fe^{57} calibration spectrum showing the location of the six normal Mössbauer absorption lines which result when all four sublevels of the 14.4-keV state are equally populated. When the CVT is operated at +5.3 mm/sec, the 14.4-keV γ rays recoillessly emitted from the source (Co^{57} in Cr) are Doppler-shifted into resonance with the transition from the $-\frac{1}{2}$ level of the ground state to the $-\frac{3}{2}$ level of the first excited state of the Fe^{57} nuclei in a metallic-iron scattering foil. Only the $-\frac{3}{2}$ excited level is populated. When the radiation, reemitted from the foil, is analyzed with a sodium ferrocyanide absorber, only one line at +5.3 mm/sec appears (the second spectrum in Fig. 2). If the CVT is operated at +3.1 mm/sec, the source radiation is in resonance with the $-\frac{1}{2} \rightarrow -\frac{1}{2}$ transition, thus populating only the $-\frac{1}{2}$ excited sublevel. In this case, two lines appear (the third spectrum in Fig. 2), since now the excited nucleus may make transitions to both the $+\frac{1}{2}$ and $-\frac{1}{2}$ ground-state sublevels. Similar results (bottom spectrum in Fig. 2) occur if the CVT is operated at +0.8 mm/sec, exciting only the $+\frac{1}{2}$ excited level. One noticeable feature of the bottom two spectra in Fig. 2 is that the ratio of peak intensities is not the same in both cases, as would be predicted on the basis of pure Mössbauer scattering. This is due to the presence of recoilless Rayleigh scattering, which increases the depth of lines on the right but leaves the lines on the left unaffected.

At this point it is interesting to ask what the intensity distribution of the resonantly scattered radiation would be if the intensity distribution of the incident radiation were not centered on the resonant energy of the scatterer. Straightforward calculations[8] indicate that as the incident radiation is shifted progressively farther from resonance, the scattered radiation line should shift about one-half as much and become broader and more shallow. Sufficiently far from resonance, the line should bifurcate, and ultimately the spectrum should consist of two very shallow lines centered on the resonant energy of the source and the scatterer, respectively.

One can study these predicted effects with the polarization technique simply by setting the CVT progressively farther off resonance. The results of such an

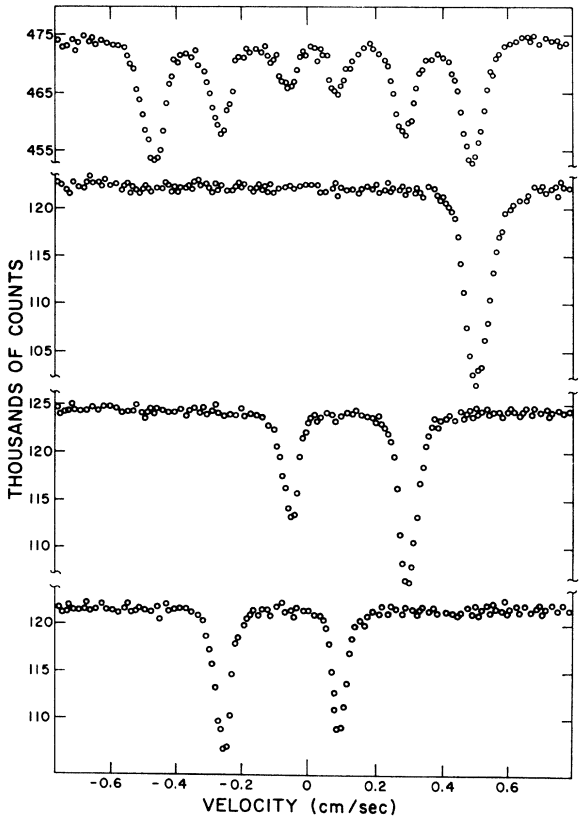

FIG. 2. Results showing polarization of excited Fe^{57} nuclei. The top spectrum is an iron calibration spectrum. The next three show, respectively, that only the $-\frac{3}{2}$, $-\frac{1}{2}$, and $+\frac{1}{2}$ excited-state sublevels have been populated.

[7] S. Shtrikman, Solid State Commun. 5, 701 (1967).

[8] A. J. F. Boyle and H. E. Hall, Rept. Progr. Phys. 25, 441 (1962); P. B. Moon, Proc. Roy. Soc. (London) A263, 309 (1961).

experiment are shown in Fig. 3, which provides the Mössbauer-effect analysis of the scattered radiation for four different settings of the CVT near 3.1 mm/sec, the $-\frac{1}{2}\rightarrow-\frac{1}{2}$ resonance. The results in general bear out the theoretical predictions, although there are some understandable departures from the ideal theory. Specifically, the center of the distribution of the scattered radiation tends to lie closer to the constant-velocity setting than to the resonant energy of the scatterer. This results from the source having a smaller linewidth than the scatterer. The stronger line on the right is seen to shift more dramatically than the other. This is because the weaker line on the left is unaffected by the Rayleigh scattering and therefore displays the shift due solely to Mössbauer scattering. The right-hand line, however, has a Rayleigh component with the same energy distribution as the incident radiation. The contribution of the Rayleigh component is most markedly displayed in the spectrum taken with a constant-velocity setting of 1.94 mm/sec. At this velocity, little Mössbauer scattering is occurring, and the right-hand peak is almost pure Rayleigh scattering and is centered very nearly on 1.94 mm/sec, the source velocity. In this case, one would also expect the left-hand portion of the spectrum to consist of two well-separated peaks. This departure from the ideal theory results because the source has a narrower line and because the velocity setting here is midway between two resonant levels of the scatterer, and the observed line is therefore the sum of the scattering from these two levels.

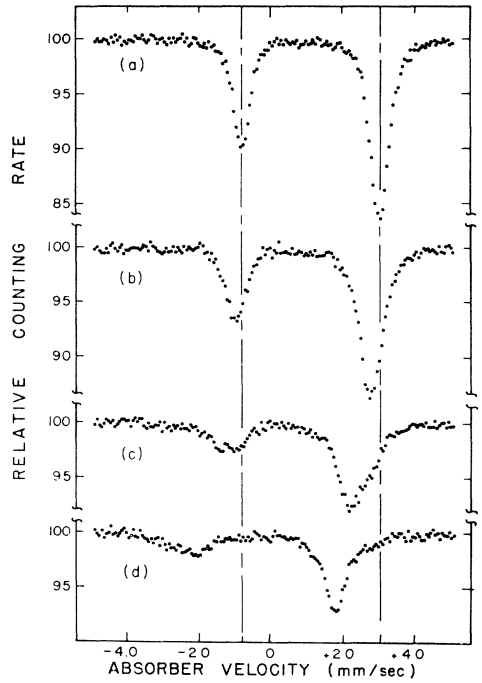

FIG. 3. Intensity distribution of resonantly scattered radiation as a function of the Doppler shift of the incident γ rays. Source velocities were (a) +3.10, (b) +2.88, (c) +2.40, and (d) +1.94 mm/sec.

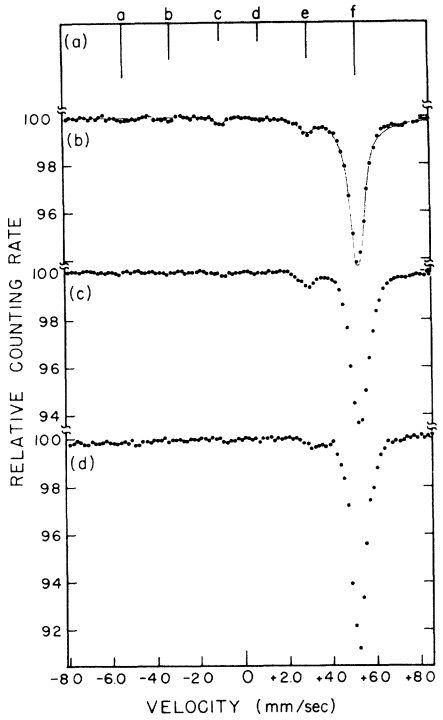

FIG. 4. Results showing excited-state NMR transitions for several static- and rf-field intensities: (b) $H_0\simeq1.7$ kG, $H_{rf}\simeq60$ G; (c) $H_0\simeq1.0$ kG, $H_{rf}\simeq50$ G; (d) $H_0\simeq1.0$ kG, $H_{rf}\simeq25$ G. (a) is a schematic showing the positions of the normal Mössbauer absorption lines. The solid line in Fig. 3(b) is a least-squares fit of the data to NMR theory. The rf and static fields were perpendicular to one another and both in the plane of the scatterer. All field values were measured with the sample in place.

Because of the short mean life (100 nsec) of the 14.4-keV state of Fe^{57}, NMR transitions in the excited state would require impossibly large rf fields were it not for the fact[9] that in ferromagnetic materials the effective rf field at the nucleus is enhanced by a factor of the order of H_{hf}/H_0, the ratio of the nuclear hyperfine field to the applied static field. Since the hyperfine field in iron is approximately 333 kG, this enhancement factor can be quite large. Attempts to utilize the Mössbauer effect to detect the NMR of the 14.4-keV state of Fe^{57} have been made before—by Perlow[3] in 1960 and more recently by Matthias.[4] Both experimenters observed a change in the recoilless absorption of an iron foil at 26 MHz, the 14.4-keV hyperfine resonant frequency. We have since shown that these resonant effects were probably not due to NMR but rather to magnetostrictively induced acoustic sidebands. The results of our investigation into this sideband effect are reported elsewhere[2]; however, two observations are pertinent to this paper. First, the sideband effect in

[9] A. M. Portis and R. H. Lindquist, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1965), Vol. II-A, p. 357.

iron foils was observed to diminish with increasing static field, dropping off rapidly for fields above 1 kG. Second, although strong sideband effects which would completely obscure detection of NMR have been observed in iron foils, no sidebands have been observed with iron powder, even though evidence for such sidebands has been sought in experiments both with and without static fields and with rf fields as large as 50 G. Consequently, the NMR experiments described here were performed with powdered iron scatterers.

As has been shown, with the CVT set at +5.3 mm/sec, only the $-\frac{3}{2}$ excited-state sublevel is populated, and only a single line appears at +5.3 mm/sec. However, if the scatterer is subjected to a rf field at 26 MHz, the hyperfine resonant frequency of the 14.4-keV state, NMR transitions to other levels should be induced, mainly to the $-\frac{1}{2}$ excited-state sublevel. These transitions should manifest themselves as additional lines appearing at −0.8 and +3.1 mm/sec coupled with an attenuation of the peak at +5.3 mm/sec.

Typical results of this excited-state NMR experiment, performed with several static and rf-field intensities, are shown in Fig. 4. Figure 4(a) is a schematic diagram indicating the position of the six normal Mössbauer lines. In Figs. 4(b)–4(d), the application of a 26-MHz rf field perpendicular to the scattering plane is seen to result in the appearance of lines e and c, indicating that NMR transitions have occurred in the 14.4-keV state. The solid line in Fig. 4(b) is a least-squares fit, illustrating the agreement with the unsophisticated theory of the preceding paragraph. The depths of lines e and c in these three spectra indicate NMR transition probabilities ranging from 10 to 16%, in good agreement with the values calculated using the measured rf-field intensity and assuming ideal ferromagnetic enhancement. The application of an 18- or 34-MHz field does not produce such additional lines, proving that the results in Fig. 4 are indeed resonant phenomena.

The NMR effect is observed to be relatively constant for large static fields in the range 500–2000 G, applied in the scattering plane at 45° to both the incident and scattered beam; but the NMR effect falls off rapidly as the field is decreased below 500 G. This behavior is in marked contrast to static field dependence of the sideband effect, which drops off rapidly for high static fields. Not surprisingly, it also disagrees with the formula for ferromagnetic enhancement for small static fields, where this formula assumes that the main contribution to the NMR signal comes from nuclei located in the domain walls. In the present experiment, however, the scattering nuclei are located mainly within the domains. It is therefore necessary to magnetically saturate the scatterer in order to make the hyperfine field in the domains follow the applied rf field.

In addition to producing NMR transitions, the high-intensity rf field should produce a distortion in the electromagnetic line shape in accord with the theories of Hack and Hammermesh[5] and of Gabriel.[6] These theories predict that each line will become progressively broader as the rf-field intensity increases and will split into multiple components for sufficiently large fields. The present experiment provides a possibility for direct observation of these effects. The theories require a homogeneous rf field to produce a resolved line structure because the separation between the components depends directly on the rf-field intensity H_1. Even if the rf field is inhomogeneous, however, one still expects to observe a broadening of the line. No measurable broadening can be detected in the results shown in Fig. 4, in disagreement with the theories. In these results, however, there is a Rayleigh-scattered component which would tend to obscure this effect. To eliminate this obscuring influence, the NMR experiment was repeated using the $-\frac{1}{2} \rightarrow -\frac{1}{2}$ populating transition and $H_{rf}=75$ G and $H_0=750$ G. A slight distortion of the line shape was observed in this experiment but no measurable broadening. Theoretical calculations based on either the measured values of H_{rf} and H_0 or on the observed transition probabilities predict a detectable broadening. Consequently, if such line-shape effects exist at all, they are smaller than indicated by present theories.

It should be remarked that changes in the angular distribution of the scattered γ rays could have been used to detect the NMR. This method was not used at this stage of investigation because of the possibility of confusing other resonant effects (such as those due to sidebands) with NMR. This is an important consideration in view of the difficulties encountered in previous attempts. In addition to providing a clear picture of the excited-state NMR process, the Mössbauer detection method allows one to investigate simultaneously the line-shape predictions.

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