

gain per cm is provided by the surface electric field of the electron beam; thus if E is the energy gain in units of electron rest energies per cm, one should satisfy

$$2\lambda r_e / aE \gg 1.$$

In both the above relations the ion is taken to be singly charged. The quantity λr_e is the beam current in units of approximately 17 000 amperes.²

Some solutions have been worked out under nonrelativistic conditions for a sinusoidal traveling wave; these have been shown to produce acceleration when the wave is accelerated through variation of the frequency of a deflecting field applied to the beam. Similar condi-

tions of acceleration of a filamentary traveling wave might exist in transverse two-stream instabilities arising in currents of relativistic electrons traversing a plasma. Work is in progress on a more detailed solution of Eqs. (1) and (3) in order to examine more carefully the motion of the accelerated particle within the beam and to find the relativistic behavior when the acceleration is provided by a sinusoidal traveling wave.

¹V. I. Veksler, Proceedings of the CERN Symposium on High-Energy Accelerators and Pion Physics, Geneva, 1956 (CERN Scientific Information Service, Geneva, Switzerland, 1956), p. 80.

²G. J. Budker, ibid., p. 69.

MAGNETIC RELAXATION AND ASYMMETRIC QUADRUPOLE DOUBLETS IN THE MÖSSBAUER EFFECT*

M. Blume

Physics Department, Brookhaven National Laboratory, Upton, New York

(Received 17 December 1964)

Goldanskii, Makarov, and Khrapon¹ have proposed an explanation for the asymmetry of quadrupole lines in Mössbauer spectra from isotropic polycrystalline sources. This explanation is based on the anisotropy of the Debye-Waller factor (recoil-free fraction) for a nucleus in a site with less-than-cubic symmetry. It predicts that the asymmetry should become more pronounced as the temperature is raised.

It is the purpose of this note to point out that such asymmetries can also arise as a result of the fluctuating electric and magnetic fields produced by the relaxation of paramagnetic ions or by the fluctuation of the environment surrounding the nucleus. These relaxation effects can lead to a variety of temperature dependences for the asymmetry of the quadrupole lines, including the opposite to that expected on the basis of a Goldanskii effect. The effects of fluctuating fields have been considered in great detail in the theory of nuclear magnetic resonance,² and we can apply the results of these analyses, with slight modification, to the Mössbauer effect.

The phenomena are best illustrated by considering a specific situation. Let us take the case of an Fe⁵⁷ nucleus in a fixed electric field gradient, along the z axis, which arises from

distant charges. The energy levels of the nucleus (for a negative gradient) are shown in Fig. 1. In an isotropic polycrystalline source, in the absence of an anisotropic Debye-Waller factor, the two lines shown should have the same width and overall intensity. We now introduce the effect of paramagnetic relaxation. Assume for simplicity that the ion is in an electronic state with $S = \frac{1}{2}$ and $g_{\perp} = 0$, so that the effects of the hyperfine interaction can be treated as an effective internal magnetic field. As the ion makes the transitions between the $+\frac{1}{2}$ and $-\frac{1}{2}$ electronic levels, the internal field jumps between the values $+\hbar_0$ and $-\hbar_0$ (both

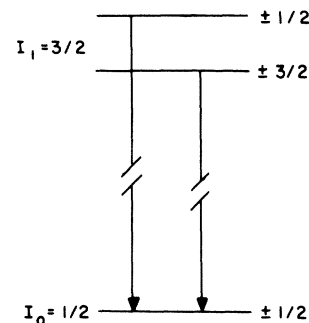


FIG. 1. Energy levels for the Fe⁵⁷ nucleus in a negative electric field gradient along the z axis.

fields being along the z axis) where, for Fe, h_0 can be of the order of several hundred kilogauss. If the fluctuation rate is very slow compared to the precession frequency of the nucleus in the field h_0 , the full six-line hyperfine pattern should be observed.³ On the other hand, if the fluctuation rate is extremely rapid the nucleus will see only the time-average field, which is zero if the ionic states are degenerate, and the symmetric quadrupolar pattern will be observed. This is the familiar "motional-narrowing" effect. In the Mössbauer effect, however, there are a number of Zeeman precession frequencies corresponding to the different transitions between magnetic sublevels of the excited and ground nuclear states. In fact, the $\pm\frac{3}{2} \rightarrow \pm\frac{1}{2}$ transitions, which make up one of the lines of the quadrupolar doublet, have a larger splitting in a magnetic field than do the $\pm\frac{1}{2} \rightarrow \pm\frac{1}{2}$ and $\pm\frac{1}{2} \rightarrow \mp\frac{1}{2}$ transitions, which make up the other line. This means that the fluctuations of the paramagnetic ion have a different effect on the two components of the quadrupole-split line. In particular, we may expect to find a range of relaxation times in which the $\pm\frac{1}{2} \rightarrow \pm\frac{1}{2}, \mp\frac{1}{2} \rightarrow \mp\frac{1}{2}$ quadrupole line is fully narrowed, while the $\pm\frac{3}{2} \rightarrow \pm\frac{1}{2}$ line is still broad. That this does in fact happen is shown in Fig. 2. The calculations on which these figures are based make use of Anderson's stochastic model of motional narrowing,² adapted to the Mössbauer effect. In 2(a) the relaxation time τ_R (i.e., the mean time between transitions from the internal field h_0 to the field $-h_0$) is long compared to the Zeeman frequencies of any of the lines in the field h_0 . (In these calculations h_0 is taken to be 300 kilogauss.) In 2(b) the relaxation time is shorter, and the two central lines of the magnetic pattern have collapsed onto their center of gravity, which is the position of one of the quadrupole doublets. The other magnetic lines are still present, although they are considerably broadened. In 2(c) through 2(e) the relaxation time becomes progressively shorter, so that the remaining magnetic lines collapse. The quadrupole pattern starts to appear, but the left-hand line (consisting of the $\pm\frac{3}{2} \rightarrow \pm\frac{1}{2}$ transitions) narrows more slowly than the right-hand line, since the precession frequency in the field h_0 is larger for the $\pm\frac{3}{2} \rightarrow \pm\frac{1}{2}$ transition. When the relaxation time gets shorter still, the symmetrical pattern is eventually found. It should be noted that the asymmetric pattern persists for relaxa-

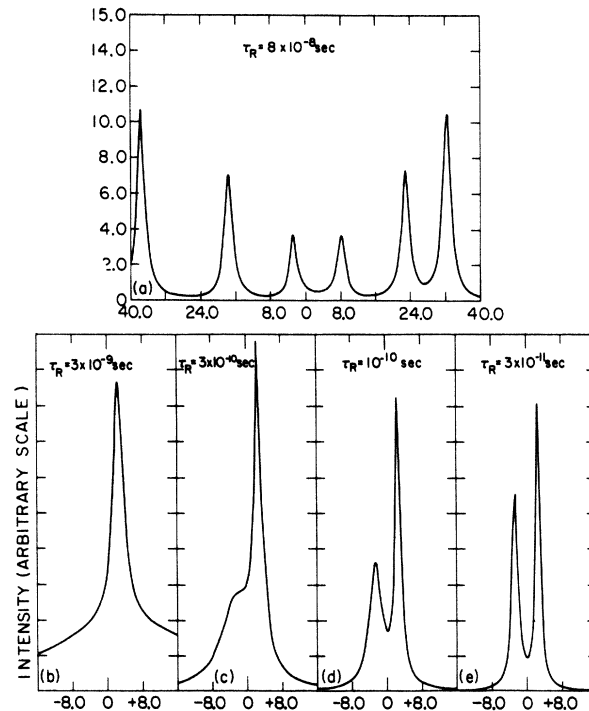


FIG. 2. Line shapes for an Fe^{57} nucleus in a fluctuating magnetic field and a fixed electric field gradient, for different values of the electronic relaxation time τ_R . The vertical scale differs from one curve to another.

tion times varying over two decades. If the fluctuations in the internal magnetic field are due to electronic spin-lattice relaxation, we expect the relaxation times to decrease with increasing temperature. This means that raising the temperature will decrease the asymmetry of the quadrupolar pattern, which is opposite to the temperature dependence of the Goldanskii mechanism. On the other hand, if the fluctuations are due to electronic spin-spin relaxation, the asymmetry will depend on the concentration of paramagnetic ions, but it will be essentially independent of temperature.

So far this discussion has been based on the adiabatic effects of the electronic relaxation, in that we have assumed that the fluctuating fields are along the same direction as the electric field gradient, so that they do not cause transitions between the nuclear sublevels. In case these fields are perpendicular to the quadrupolar axis they may cause transitions between the nuclear levels, giving rise to nuclear spin-lattice relaxation. These effects can be considered from a different point of view. Any transitions between nuclear sublevels will af-

fect the lifetimes of the sublevels, and will give them an additional width, which must be added to the gamma-ray and internal-conversion contributions in determining the overall width of the observed line. Nuclear-relaxation effects differ from the other contributions to the widths in that they can be different for different sublevels. Thus, if the nuclear relaxation is due to magnetic-dipole effects, a nucleus in the $+\frac{3}{2}$ state can make transitions only to the $+\frac{1}{2}$ state. One in the $+\frac{1}{2}$ state, on the other hand, can make transitions to the $+\frac{3}{2}$ state and also to the $-\frac{1}{2}$ state, which is degenerate with the $+\frac{1}{2}$ state. The width of a level n is given by the sum of the transition probabilities out of that level. Hence, for magnetic-dipole relaxation,

$$1/\tau_{3/2} = P_{3/2 \rightarrow 1/2},$$

and

$$1/\tau_{1/2} = P_{1/2 \rightarrow 3/2} + P_{1/2 \rightarrow -1/2}.$$

If the temperature is sufficiently high so that $P_{3/2 \rightarrow 1/2} = P_{1/2 \rightarrow 3/2}$, we expect that the $\pm\frac{1}{2}$ levels in the excited state in Fig. 1 will be broader than the $\pm\frac{3}{2}$ levels. This too will give rise to an asymmetry in the quadrupole pattern. The temperature dependence of this effect depends, however, on the temperature dependence of the nuclear spin-lattice relaxation time. If this time decreases as the temperature increases, than the asymmetry will become more pronounced with increasing temperature, as is the case for Goldanskii's effect. In order to observe effects of nuclear relaxation, the relaxation times must be of the order of, or shorter than, the radiative lifetime of the excited nuclear state, i.e., about 10^{-8} sec. Such times are quite short for nuclear-relaxation

processes, but they are not unreasonable for nuclei in magnetic materials.

Experimentally, it is possible to distinguish relaxation-induced asymmetries from the Goldanskii mechanism, apart from the temperature dependence, by observing the quadrupole pattern from a single-crystal source. The relaxation effects will give a broadening which affects the same line regardless of the direction of observation. The anisotropic Debye-Waller factor, on the other hand, will broaden only those gamma rays which are emitted in a specific direction, namely, along the direction in which the ion can most easily recoil.

The phenomena described here are a part of a large number of interesting and unusual line shapes which should be observable whenever fluctuating fields are present. A theoretical treatment which can be applied to ferromagnetic and antiferromagnetic substances, as well as to paramagnets, and which includes the effects of fluctuating electric field gradients, has been worked out, and details of the theory and the experimental results to which it is applicable will be given separately.

Many colleagues at Brookhaven National Laboratory and Bell Telephone Laboratories, too numerous to mention individually, have made helpful comments and suggestions.

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

¹V. I. Goldanskii, E. F. Makarov, and V. V. Khrapov, *Phys. Letters* **3**, 344 (1963).

²P. W. Anderson, *J. Phys. Soc. Japan* **9**, 316 (1954).

³A. M. Afanasev and Yu. Kagan [*Zh. Eksperim. i Teor. Fiz.* **45**, 1660 (1963) [translation: *Soviet Phys.-JETP* **18**, 1139 (1964)]] have given a detailed treatment of this aspect of the problem for one-phonon electronic relaxation processes.

MAGNETIC FIELD DEPENDENCE OF THE MICROWAVE SURFACE IMPEDANCE IN PURE SUPERCONDUCTORS*

Kazumi Maki

The Enrico Fermi Institute for Nuclear Studies, The University of Chicago, Chicago, Illinois
(Received 3 December 1964)

There have been a number of experimental works¹⁻⁶ on the microwave surface impedance of superconductors in the presence of static magnetic fields. They have found, especially, that the surface impedance decreases as the static field increases for temperatures close to the critical temperature. The difficulty in accounting for this phenomenon in terms of the

two-fluid model has been stressed by Pippard.⁷

In the present note we will show that many of the features of the experimentally observed field dependence of the surface impedance follow directly from the current microscopic theory. We calculate the surface impedance in pure superconductors in the presence of magnetic fields by using the techniques developed