Mössbauer sidebands from a single parent line*

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Mössbauer sidebands from a single parent line have been produced by using a composite sample made of two materials. A nonmagnetic 310 stainless-steel (ss) foil plated with Ni was used as a single-line Mössbauer-effect probe, and the ferromagnetic and magnetostrictive Ni provided the driving force for the production of ultrasound in the sample. The unequivocally measured sideband intensities agree qualitatively with Abragam's sideband theory. However, no existing theory at present can account quantitatively for the sideband intensities. Sidebands in the Ni-plated 310 ss sample decrease sharply with increasing frequency. No sideband was observed for 310 ss alone, nor 310 ss plated with Cu or Ag. These results further support the view that the driving mechanism for sidebands is magnetostriction and not eddy currents.

INTRODUCTION

When an electromagnetic wave with frequency ω_0 is frequency modulated at a different frequency ω_d, the resulting frequency spectrum consists of an infinite set of sidebands of frequency $\omega_0 \pm n\omega_d$, where $n = 0, 1, 2, 3 \ldots$. Two kinds of experiments have been devised to show this phenomenon. In one, Ruby and Bolef[1] and others[2,3] were able to produce FM sidebands in a Mössbauer spectrum by *mechanically* vibrating the source or absorbers with an external piezoelectric transducer. In the second kind, Heiman and co-workers[4,5] and others[6,7] reported a different type of experiment in which *no mechanical transducer was employed.* FM sidebands appeared when ferromagnetic samples were subjected to a radio-frequency (rf) magnetic field. Detailed studies of this latter phenomenon have indicated that the dominant mechanism for the rf sidebands is magnetostriction which, in turn, produces acoustic waves in the sample. The large acoustic Q of these samples provides a remarkable amplification of its vibration amplitude.[8]

Experimental determination of the sideband intensity has been difficult because of the requirement that the sample be ferromagnetic. In the case of ferromagnetic ^{57}Fe there are at least six parent lines. When sidebands appear for each of the six lines, the resultant spectrum unavoidably involves severe overlapping problems. For high-rf fields, the spectrum becomes completely washed out.

We report in this work an experiment in which one can observe sidebands from a *single* parent line by using a composite sample consisting of two materials of different properties. The sideband intensities can be unequivocally measured and compared with existing theories. The results provide a further test of the magnetostriction model proposed earlier.[4,5]

EXPERIMENTAL

Conventional Mössbauer spectroscopy using the 14.4-keV γ ray of ^{57}Fe was employed. A rf magnetic field was applied via an eight-turn flat coil surrounding the sample which was sandwiched between two sheets of Mylar to ensure mechanical stability. To avoid rf heating of the sample, the rf power was pulsed at a rate of 2.5 kHz with a 50% duty cycle. The sample was essentially at room temperature.

RESULTS AND DISCUSSION

According to the magnetostrictive model, under the influence of rf magnetic fields acoustic waves should exist for all magnetostrictive materials whether or not they contain ^{57}Fe. The presence of ^{57}Fe is required only to permit one to observe the resultant sidebands. We therefore used a sample which consists of two materials of different properties. The first material is ferromagnetic and highly magnetostrictive, but contains no Fe. To serve as a probe, nonmagnetic (therfore single-parent-line) material containing ^{57}Fe was bonded to the first foil so that acoustic vibrations could be detected by the production of sidebands on a single spectral line.

Ni metal and 310 stainless steel (ss) were chosen for the present work. Ni is ferromagnetic at room temperature and is highly magnetostrictive. 310 ss is nonmagnetic and shows a narrow single line. As expected, the 310-ss foil alone shows *no* sidebands from the rf field.

As a first attempt to observe sidebands, a 5-μm 310-ss foil was glued to a 2-μm Ni foil, by epoxy resin. Sidebands, although small, indeed appear, as shown in Fig. 1. Obviously, the epoxy resin is not very efficient in coupling the two foils acoustically. We then used a 5-μm

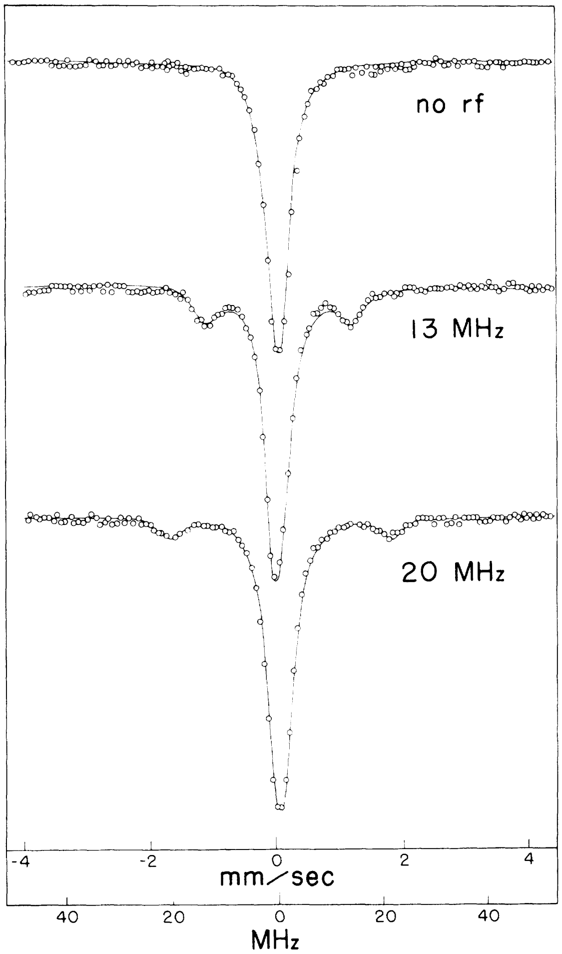

FIG. 1. Mössbauer sideband spectra of 5-μm 310-ss foil epoxied to 2-μm Ni foil.

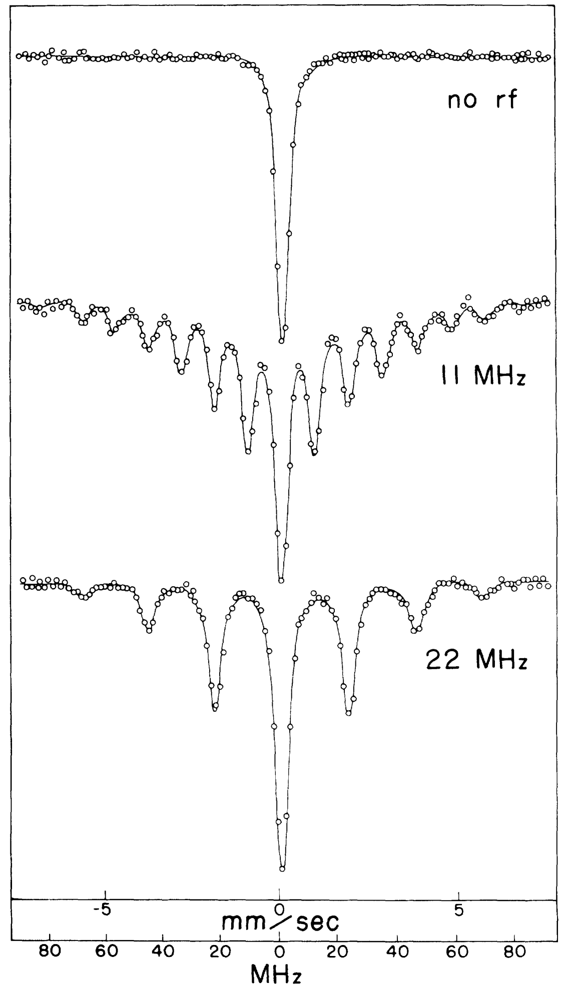

FIG. 2. Mössbauer sideband spectra of 5-μm 310-ss foil electroplated with 5-μm Ni.

310-ss foil with 5 μm of Ni electroplated on each side. Under the influence of a rf field, large sidebands can be easily observed, as shown in Fig. 2.

Since the foil (Ni) in which the acoustic waves are generated contains no measurable amount of ^{57}Fe, and since the probe foil (310 ss) has practically zero average hyperfine field, these results are clear evidence that sideband effect is purely an acoustic effect, that it is independent of nuclear properties, and that it is not due to disturbances of the magnetic hyperfine field, as has been suggested.

Because the number of parent lines is now one instead of six, the overlapping of the sidebands is eliminated. The sideband intensities can be directly measured and compared with theory.

Theoretical studies have indicated that the sideband spectrum can be described as[5]

$$W'(E)=\sum_{n=-\infty}^{\infty} W(E+n\hbar\omega_d)\int_0^{\infty} J_n^2\left(\frac{x_0}{\lambda}\right)P(x_0)\,dx_0\,, \quad (1)$$

where $W(E)$ is the Mössbauer transition probability, ω_d is the driving frequency, J_n is the nth-order Bessel function, x_0 is the peak vibration amplitude of the nucleus, $\lambda = 0.137$ Å for 14.4-keV γ rays, and $P(x_0)$ is the distribution function of the peak amplitude x_0. The nth-order sideband therefore has the intensity of

$$\int_0^{\infty} J_n^2\left(\frac{x_0}{\lambda}\right)P(x_0)\,dx_0\,. \quad (2)$$

Obviously,

$$\sum_{n=-\infty}^{\infty}\int_0^{\infty} J_n^2\left(\frac{x_0}{\lambda}\right)P(x_0)\,dx_0=1$$

to conserve transition probability.

Two models have been proposed for the distribution function $P(x_0)$. If one assumes that all the nuclei vibrate with the same peak amplitude a, i.e., $P(x_0)=\delta(x_0-a)$, then the sideband intensity varies as

$$J_n^2(m)\,, \tag{3}$$

where $m = a/\lambda\!\!\!^-$ is the modulation index. This description, however, is not even qualitatively correct. From Fig. 2, it is clear that experimentally one observes that the $(n+1)$th sideband is *always* less than the nth sideband. But Eq. (3) predicts exactly the opposite for certain values of the modulation index m (Fig. 3). For instance, for $m \approx 2.5$, Eq. (3) predicts that the first and second sidebands are larger than the zeroth order. This is never observed in any sideband experiments.

Abragam[9] has proposed that $P(x_0)$ has a Rayleigh distribution of the form

$$P(x_0) = (x_0/a^2)e^{-x_0^2/2a^2}\,. \tag{4}$$

The sideband intensity then varies as

$$e^{-m^2}I_n(m^2)\,, \tag{5}$$

where I_n is the modified Bessel function of the first kind. This form agrees with the experimental observation that the nth-order is always less than all lower-order sidebands (Fig. 4). Unfortunately, this theory does not provide close *quantitative* agreement with experiment.

In Fig. 5, we plot the theoretical ratio of nth order to zeroth order according to Eq. (5). The bars on the vertical lines indicate experimental results. If only first and second orders of sidebands are observed for small rf fields, one can find a value of m which allows Eq. (5) to satisfactorily describe the sideband intensities. For larger rf fields where more than two orders of sidebands appear, there is no value of m which can closely describe the sideband intensities. In general, Eq. (5) predicts sideband intensity too large for lower n and too small for higher n.

The disagreement with either theoretical model is not too surprising in view of the assumptions made in each case. Equation (3) was obtained by assuming that the phonon excitation is completely coherent. Equation (5) assumes that the phonon excitation is thermalized and incoherent. As pointed out by Mishory and Bolef,[3] these results correspond to very long and very short phonon-relaxation-time limits. In actual physical systems, the situation lies between these extremes. Typically, relaxation times are of the order of 10 μsec.[8]

It should be pointed out that in a different but related experiment, Cranshaw and Reivari[2] found rather good agreement with the Rayleigh-distribution model of Abragam. In their experiment, the acoustic waves were generated by means of a piezoelectric transducer to which the Mössbauer absorber was fastened. The absorber was FeAl powder glued to a quartz transducer. One might expect the phonon relaxation times in such a system to be much shorter than those observed for our magnetostrictively driven samples. For short relaxation times one would then expect to obtain much better agreement with the Rayleigh-

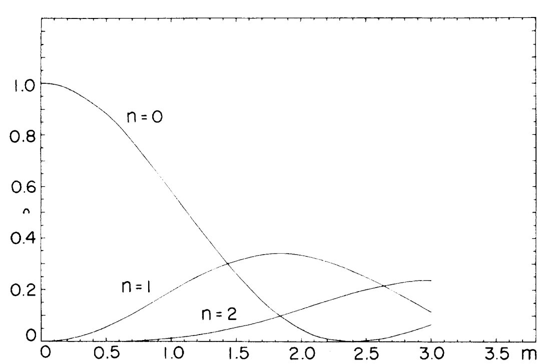

FIG. 3. Sideband intensity as predicted by $J_n^2(m)$, for $n = 0$, 1, and 2.

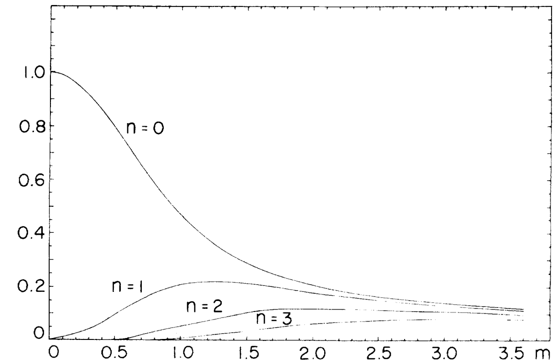

FIG. 4. Sideband intensity as predicted by $e^{-m^2}I_n(m^2)$, for $n = 0$, 1, 2, and 3.

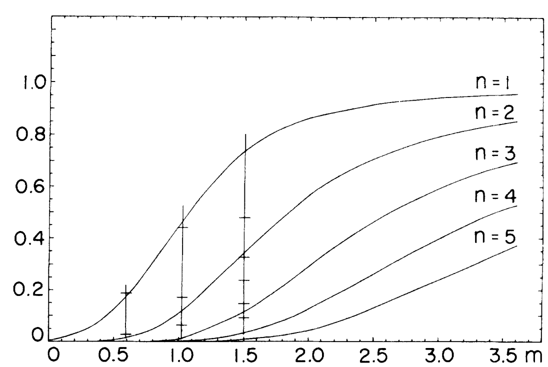

FIG. 5. Sideband intensity normalized by the zeroth-order intensity. The curves are predicted by Abragam's theory. The horizontal bars on the vertical lines are the measured results from the Ni-plated 310-ss foil.

distribution model.

The δ function and the Rayleigh-distribution functions for $P(x_0)$ are the only ones proposed thus far. They are certainly not the only two physically realizable situations. On the contrary, one can assume an infinite number of $P(x_0)$, and Eq. (2) would predict the sideband intensity accordingly. The $P(x_0)$ that satisfactorily describes the data is not known at present.

In order to further test the magnetostriction model and look for other possible mechanisms, such as eddy currents, we also plated the 310-ss foil with 5 μm of Cu or 5 μm of Ag. These foils, just as does the 310-ss foil alone, show *no* sidebands with the maximum rf power available to us. In Fig. 6, we show the result of monitoring the counting rate at zero Doppler velocity from 10 to 60 MHz. Any increase in the counting rate will be due to the appearance of sidebands. Only the Ni-plated 310-ss foil shows the effect. No sign of any change was observed for unplated 310 ss (shown) nor for 310 ss plated with Cu or Ag (not shown). Therefore eddy-current contributions will be much too small to be comparable to the magnetostriction contribution. Furthermore, the frequency dependence of the sideband intensity in the Ni-plated 310-ss case indicates that the effects cannot be due to eddy currents, since an eddy-current contribution would increase with increasing frequency, contrary to the results shown in Fig. 6.

Since one can measure frequency much more easily and to much higher accuracy than one can measure velocity, the single-line sideband technique can be used for accurate calibration of the energy scale of Mössbauer spectra for energy ranges much higher than that of ^{57}Fe. For the 14.4-keV energy of the transition in ^{57}Fe, an energy shift associated with a Doppler velocity of 5 mm/sec corresponds to a frequency of 60 MHz. Therefore a single-line sideband spectrum showing several orders of sidebands can be used as an absolute energy calibration corresponding to a

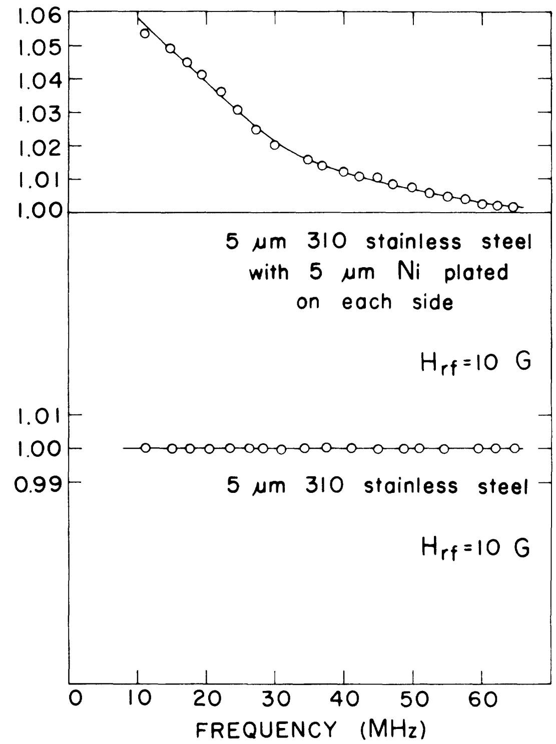

FIG. 6. The counting rate at zero Doppler velocity for Ni-plated 310 ss and unplated 310 ss.

Doppler-velocity range several times 5 mm/sec and therefore much higher than the calibration range of a normal Fe spectrum. A second application involves the use of 310 ss as a probe to detect magnetostriction in other nonferrous magnetic materials by making a composite sample by bonding, as in the present experiment. This is particularly useful for magnetic materials that can be made only in thin foil form, notably the splat-cooled amorphous ferromagnets, for which conventional magnetostriction measurements are difficult because of the small thickness of the sample.

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