Collapse of Mössbauer spectra in strong applied radio-frequency fields

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(Received 29 July 1987; revised manuscript received 29 February 1988)

We consider two possible model time dependences of the local magnetization produced by the rf field in radio-frequency collapse experiments. For the first model—coherent, periodic reversal of the magnetization—we derive the absorption line shape and show that it conflicts with the absence of sidebands in collapsed spectra of nonmagnetostrictive and vibrationally damped magnetostrictive materials, but that it is roughly compatible with experiments on undamped magnetostrictive foils provided that the symmetry properties of magnetostriction are correctly accounted for. The second model, which assumes that the local magnetization fluctuates stochastically with a small, periodic part, appears to be more compatible with experimental results. In addition to explaining how sidebands arise, this model correctly predicts that in some materials the spectra will remain collapsed for short times after the rf field is switched off. The second model is based on a magnetic cluster picture of some soft magnetic materials and assumes that the rf field destroys cluster-cluster order without affecting magnetic ordering within clusters. We conclude that the primary effect of the rf field is not to cause fast switching of the magnetization but rather is to destroy long-range magnetic order below the Curie temperature.

I. INTRODUCTION

During attempts to observe nuclear magnetic resonance (NMR) with Mössbauer spectroscopy two new effects were observed.[1-4] In the first effect absorption sidebands were found to appear in the Mössbauer spectra of magnetic foils when a radio-frequency (rf) magnetic field was applied in the plane of the foil.[1,5] In the second effect the magnetically split spectra of soft magnetic materials were found to collapse in strong, high-frequency rf fields to single or double lines typical of paramagnetism.[6-8] These effects were named the "rf sideband effect"[9] and the "rf collapse effect,"[8] respectively, and they are illustrated in Fig. 1. In both effects a spectrum consists of a central "carrier" spectrum plus sideband spectra displaced from the carrier by $\pm n\omega_{\rm rf}$, where n is an integer and $\omega_{\rm rf}$ is the frequency of the applied rf field.[1,2,5-8,10-12] In the sideband effect the carrier spectrum is the usual magnetically split pattern at all frequencies (six lines for ^{57}Fe), while in the collapse effect the carrier spectrum is a single line at high frequencies and a magnetically split pattern at low frequencies.[6,8] The phenomenology of these effects is understood to a degree, but neither effect is understood in detail. The present work represents an attempt to understand the collapse effect, and in particular to infer from the known experimental facts the effect of the radio-frequency field on the local magnetization. In this section the experimental and theoretical background needed for our discussion is briefly reviewed, and in the following sections two models of the time dependence of the magnetization in the collapse effect are considered, and the Mössbauer spectra which they would give rise to compared with experiment.

In order for the sideband effect to be observed in hard magnetic materials the absorber must have nonzero magnetization,[1,7,9] nonzero magnetostriction,[13,14] and transverse dimensions larger than the wavelength of sound at $\omega_{\rm rf}$.[1,9] From this and other evidence[1,7,9,10,15] it is clear that the sidebands result from vibrations of the foil produced by the magnetoelastic coupling between the lattice and an induced, periodic component of the magnetization, $\delta M(t)$. (For details, see Refs. 1, 7, 8, 10, and 11.) Because both odd- and even-order sidebands are observed and magnetostriction is symmetric with respect to inversion of the magnetization, the induced component $\delta M(t)$ must be small compared to the saturation magnetization.[1,4,9]

It is almost certain that the sidebands sometimes observed in the collapse effect are also associated with magnetostriction. Certainly this has been the assumption of the majority of workers in the field[7-9,10,15,16] (but see Olariu *et al.*[17,18]). Strong evidence in favor of this idea is that no sidebands appear in zero magnetostriction materials when the collapse effect occurs.[13,14] In addition, it has been found that if an Invar foil is coated with tape, grease, or varnish then in the high-frequency regime the sidebands are damped out but the spectrum remains collapsed.[19]

In the collapse effect the entire magnetization has been assumed to be reversing coherently with period $\omega_{\rm rf}$, under the control of the radio-frequency field. The evidence which originally led to this conclusion is that the effect is only seen in materials with low anisotropy fields H_a, and to see a collapsed spectrum the time-averaged magnetic field at the nucleus must obviously be zero. In this model if the magnitude of the applied rf field is sufficiently larger than the anisotropy field then the total magnetization will switch periodically to follow the rf field, so each nucleus will see a time-dependent magnetic hyperfine field, $\mathbf{H}_{\rm hf}(t)$. This field reverses its direction periodically with frequency $\omega_{\rm rf}$, and if $\omega_{\rm rf}$ is larger than the Larmor precession frequency of the nucleus then the spectrum

will collapse.[6,7,9,16,20] If ω_{rf} is less than the Larmor precession frequency no effect is expected, which supposedly explains why low-frequency spectra in the collapse and sideband effects are so similar (Fig. 1).[7] The central feature of this picture is that the magnetization must reverse its direction quickly compared with the period of the rf field,[6,7,20] and it must do so at all points in the foil.

More recent evidence that the magnetization is controlled by the rf field is due to Kopcewicz,[16] who investigated the dependence of the collapse effect in Permalloy and Invar on the intensity of the rf field. As the intensity is increased from less than to greater than H_a, one expects a region where the field is not large enough to cause the magnetization to switch on every cycle, but large enough to cause occasional reversals. In this region, if the switching of the magnetization is random, one expects to see a decreased magnetic hyperfine field, but not complete collapse, and this is observed. This was also observed in $Fe_{40}Ni_{40}Ba_{20}$.[13,14,20] Albanese, *et al.*[6] studied the collapse effect in an electrically insulating, planar ferrite, $Ba_2Zn_2Fe_{12}O_{22}$ (Zn_2Y). To ensure that any collapse was due to fluctuations driven by the applied field, and not due to heating, the rf field was pulsed on and off, and two spectra collected, one with the field pulsed on, and one with the field pulsed off. With the field pulsed off a partial collapse of the spectrum due to heating of the sample was observed, but the spectrum collected with the rf pulsed on showed a further collapse to a fairly narrow line. This further collapse was ascribed to the rf field driving the magnetization. The authors found that the heating was due to frictional losses by the rotating magnetization. No sidebands were observed, but the spectra are quite noisy, so weak sidebands may be present.

An experiment which appears to contradict the model of coherently reversing magnetization was reported by Kotlicki.[21] The pulse technique was applied on Permalloy and Invar foils, and the unexpected result was obtained that in the high-frequency regime, where one observes a single collapsed peak plus sidebands when the field is on, with the field pulsed off the spectrum *is still collapsed, but the sidebands are absent.* This was interpreted as meaning that the collapse effect takes place due to rf heating to above the Curie point, but this interpretation was ruled out by Kopcewicz *et al.*[13,14,22] who used a surface pyrometer and verified that the sample temperatures are well below the Curie point when the spectra are collapsed. In addition, in separate experiments they (a) superposed large static fields to eliminate magnetic effects of the rf (Ref. 13) and (b) increased the sample anisotropy field by applying external stresses.[22] Both techniques should not interfere with rf heating, and the collapse was inhibited, so that rf heating is not the source of the collapse.

As noted by Olarui *et al.*,[17] there is an obvious theoretical problem with the accepted model of the collapse effect and the associated sidebands: if the entire magnetization is reversing coherently, then only even-order magnetostrictive sidebands should appear, but both even- and odd-order sidebands are observed. There are only two general classes of solutions to this problem, which we shall refer to as hypotheses I and II: either (I) the magnetization is reversing coherently in which case the odd-order sidebands cannot be magnetostrictive, or (II) all sidebands are magnetostrictive, in which case the collapse cannot occur because the magnetization is reversing coherently under the influence of the rf field. It would appear that no one has ever discussed the existence or implications of hypothesis II. Olariu *et al.*,[17] who did not have the benefit of all of the currently known experimental facts, attempted to demonstrate that hypothesis I is correct by showing that a periodically reversing local moment on a Mössbauer atom will produce sidebands in the Mössbauer spectrum. Like previous theoretical studies Olariu *et al.* performed perturbative calculations of the amplitude of the two-photon process involving an rf photon and a γ ray.[17,18,23–25] Specifically the hyperfine field was assumed to vary sinusoidally and it was found that the line shape consists of a single line plus sidebands, and, at high frequencies, where only the first-order sideband is large (i.e., the cross section for three or more photon processes is small), good agreement with experimental data was obtained. Their calculation only holds at high frequency, so comparison with low-frequency experimental results was not possible. Nevertheless, it was suggested that multiphoton absorption explains the sidebands at *both* high and low frequencies, so it is not necessary to invoke the magnetostrictive mechanism of sideband creation, which makes hypothesis I consistent with the experiments and also internally consistent.

We felt that it was not acceptable to take agreement in the high-frequency limit as proof that magnetostriction is not needed to explain sidebands seen at low frequencies. Futhermore, we were interested in a possible connection between slow fluctuations due to cluster-type excitations of the magnetization (Rancourt *et al.*,[26,27] and the collapse effect. For that reason we have done a nonperturbative calculation of the Mössbauer absorption line shape under the assumptions of hypothesis I, using an adiabatic approximation. The advantage of our approach is that it allows us to calculate the line shape at "all" frequencies, but it has the disadvantage that the results cannot apply in the rather restricted but very interesting case where the applied frequency is resonant with a transition between hyperfine levels of the ground or excited states, since in this case the adiabatic approximation fails. We compare the derived line shape with experiments and find poor agreement. A possible model compatible with hypothesis II is then shown to be in better agreement with experimental results.

II. CALCULATION OF COHERENT REVERSAL LINE SHAPE

In this section we calculate the absorption line shape assuming that the magnetization follows the rf field coherently, with a negligible time required for it to reverse direction. We start from the usual correlation-function expression for the intensity of emission of frequency ω by a system with ground state $|I_0 m_0\rangle$ and excited state $|I_1 m_1\rangle$,[28–30]

$$W(\mathbf{k}) = \frac{2}{\Gamma} \mathrm{Re} \int_0^\infty G(\tau) e^{[i\omega - (\Gamma/2)]\tau} d\tau \,, \tag{1}$$

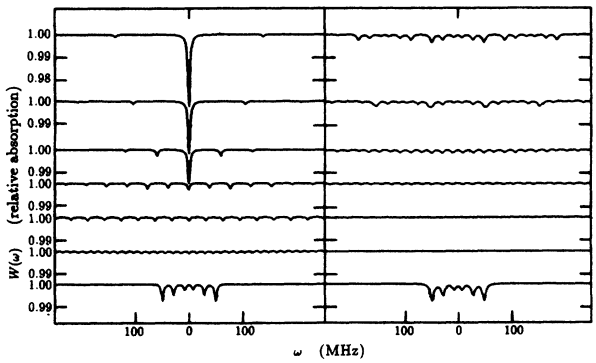

FIG. 1. A comparison of the spectra observed in the rf collapse effect (left column) with those observed in the rf sideband effect (right column). Going from bottom to top, the applied frequencies are $\omega_{rf}=0$, 15, 32, 60, 106, and 140 MHz, for a system with half-separation of lines 1 and 6 in the unperturbed spectrum $\alpha(\frac{3}{2},\frac{1}{2})=50$ MHz. These spectra are based on Ref. 7.

where Γ is the lifetime of the excited state.

The central problem is to calculate the correlation function

$$G(\tau)=\langle H^{(-)}H^{(+)}(\tau)\rangle \ , \tag{2}$$

where $H^{(+)}$ describes the interaction of the system with a γ ray which is being emitted, and the brackets denote averaging over quantum states

$$\langle \hat{O}\rangle=\sum_{\lambda} p_{\lambda}\langle \lambda \mid \hat{O} \mid \lambda\rangle \ .$$

Following Anderson and Weiss,[31,32] time-dependent effects of the environment of the absorbing nucleus are included via an explicitly time-dependent hyperfine Hamiltonian $\mathcal{H}(t)$, in which case $H^{(+)}$ describes the interaction of the γ ray with the unperturbed nucleus, and

$$\begin{aligned} H^{(+)}(\tau)=&\left[\hat{T}\exp\left[i\int_0^{\tau}\mathcal{H}(t)dt\right]\right] \\ &\times H^{(+)}\left[\hat{T}\exp\left[-i\int_0^{\tau}\mathcal{H}(t)dt\right]\right] \\ \equiv& U^{\dagger}(\tau)H^{(+)}U(\tau) \ , \end{aligned} \tag{3}$$

where $\hat{T}$ is the time-ordering operator. Random fluctuations in the environment (for example fluctuating hyperfine fields) are treated by adding a random time-dependent term to $\mathcal{H}(t)$ in Eq. (3), and performing an average over the stochastic degrees of freedom as well as the average over quantum states in Eq. (2). That is,

$$G(\tau)=(\langle H^{(-)}H^{(+)}(\tau)\rangle)_{av} \ . \tag{4}$$

We want to consider the effect of a magnetic hyperfine field with constant magnitude but time-dependent direction, so we write the hyperfine Hamiltonian as (following Blume and Tjon[33,34])

$$\mathcal{H}(t)=\mathcal{H}_0+Q(3I_z^2-\mathbf{I}^2)+g\mu_N H_{hf}I_z f(t) \ , \tag{5}$$

where $f(t)$ takes on the values $+1$ or -1, g is the nuclear g factor, μ_N is the nuclear magneton, and H_{hf} is the magnitude of the magnetic hyperfine field. This is appropriate for an ^{57}Fe nucleus in a magnetic hyperfine field constrained to an axis parallel to the electric field gradient and is intended to reflect the assumption that the local magnetization is always parallel to the rf field. The discrete nature of $f(t)$ in Eq. (5) represents a dramatic improvement over the assumption, made by Olariu *et al.*,[17] that the magnitude of the magnetic field at the nucleus varies sinusoidally. The magnetization and the applied field might vary continuously, but the magnetic hyperfine field is produced by the atomic electrons, and is therefore constant in magnitude.

In the general case $f(t)$ could vary from one site to the next. If we write the magnetization as

$$\mathbf{M}(\mathbf{r},t)=m_0\hat{\mathbf{f}}(\mathbf{r},t) \ , \tag{6}$$

where $\hat{\mathbf{f}}(\mathbf{r},t)$ is a vector of unit length, then the assumption of hypothesis I, that the magnetization everywhere switches coherently with a reversal time much shorter than the period of the rf field, means that $\hat{\mathbf{f}}(\mathbf{r},t)$ is a square wave and is independent of $\mathbf{r}$, so that $f(t)$ has identical square-wave behavior at all sites. If the magnetic switching time were longer $f(t)$ would be less periodic. In addition to periodic reversals, in the general case there might be random fluctuations of the local moments due to, say, cluster excitations (Rancourt *et al.*[26,27]), in which case $\hat{\mathbf{f}}(\mathbf{r},t)$ has an additional random time dependence which is not related to the rf field. In the extreme of this case, where the local magnetization fluctuates completely independently of the rf field, the line shape will reduce to a stochastic relaxation line shape. In the present calculation it is assumed that any such random fluctuations of the local moments are much faster than the Larmor precession frequency of the nucleus, so they can be included in the size of H_{hf}.

Hypothesis I thus reduces to the assumption that $f(t)$ is a square wave with period $p=2\pi/\omega_{rf}$. If we specify the origin of time at a given site as an arbitrarily chosen step of $f(t)$ and write all subsequent times as $t=n_t p+\Delta t$, where n_t is an integer and $0\le\Delta t\le p$, then $f(t)$ in Eq. (5) can be written as

$$\begin{aligned} f(t)&=f(n_t p+\Delta t) \\ &=f(\Delta t) \\ &=\begin{cases} +1 \text{ if } 0\le\Delta t\le p/2 \\ -1 \text{ if } p/2\le\Delta t\le p \ . \end{cases} \end{aligned} \tag{7}$$

The advantage of using a Hamiltonian in the form of Eq. (5) is that (i) it is diagonal and, therefore, (ii) it commutes with itself at different times (i.e., $[\mathcal{H}(t), d\mathcal{H}(t)/dt]=0$), so that we can drop the time-ordering operator $\hat{T}$ from our expression for $U(t)$ [Eq. (3)]. In that case it can be shown[34] that

$$G(\tau)=\tfrac{1}{4}\sum_{m_0,m_1}|\langle I_0m_0|H^{(+)}|I_1m_1\rangle|^2\exp(-i\omega_0\tau-iQ(3m_1^2-\tfrac{15}{4}\tau))\left[\exp\left[i(g_0m_0-g_1m_1)\mu_N H_{\rm hf}\int_0^\tau f(t)dt\right]\right]_{\rm av}. \tag{8}$$

Thus, defining

$$G_{m_0m_1}(\tau)\equiv\left[\exp\left[i\alpha\int_0^\tau f(t)dt\right]\right]_{\rm av}, \tag{9}$$

where $\alpha=(g_0m_0-g_1m_1)\mu_N H_{\rm hf}$ is one-half of the magnetic splitting of the pair of lines associated with the transitions $\pm m_1$ to $\pm m_0$, we obtain

$$W(\mathbf{k})=\frac{2}{\Gamma}\sum_{m_0,m_1}\tfrac{1}{4}|\langle I_0m_0|H^{(+)}|I_1m_1\rangle|^2{\rm Re}\int_0^\infty e^{z\tau}G_{m_0m_1}(\tau)d\tau, \tag{10}$$

where $z=i[\omega-\omega_0-Q(3m_1^2-\frac{15}{4})]-\Gamma/2$.

The sole stochastic degree of freedom which must be averaged over Eq. (4) is the phase of the rf field relative to $\tau=0$, the time at which the excited nuclear state is created. This average can be performed by writing

$$\begin{aligned}G_{m_0m_1}(\tau)&=\frac{1}{p}\int_0^p\exp\left[i\alpha\int_t^{t+\tau}f(t')dt'\right]dt\\&=\frac{1}{p}\sum_{n=0}^{1}\int_0^{p/2}\exp\left[(-1)^n i\alpha\int_t^{t+\tau}f(t')dt'\right]dt\\&=\frac{1}{p}\sum_{n=0}^{1}\begin{cases}\left[\frac{p}{2}-\Delta\tau\right]e^{\gamma\Delta\tau}-\frac{1}{2\gamma}(e^{-\gamma\Delta\tau}-e^{\gamma\Delta\tau}), & \text{if } 0\le\Delta\tau<p/2\\ +\left[\frac{p}{2}-\Delta\tau\right]e^{\gamma(\Delta\tau-p)}-\frac{1}{2\gamma}(e^{\gamma(\Delta\tau-p)}-e^{\gamma(p-\Delta\tau)}), & \text{if } p/2\le\Delta\tau<p,\end{cases}\end{aligned} \tag{11}$$

where $\gamma\equiv(-1)^n i\alpha$.

Now $G(\tau)=G(n_\tau p+\Delta\tau)=G(\Delta\tau)$, and $e^{z\tau}=(e^{(zp)})^n e^{z\Delta\tau}$, so that

$$\int_0^\infty e^{z\tau}G_{m_0m_1}(\tau)=\left[\sum_{N=0}^{\infty}(e^{zp})^N\right]\int_0^p e^{z\Delta\tau}G_{m_0m_1}(\Delta\tau)d\Delta\tau. \tag{12}$$

The integration and sum are trivial, giving a final result

$$W(\mathbf{k})=-\tfrac{1}{8}\sum_{m_0,m_1}|\langle I_0m_0|H^{(+)}|I_1m_1\rangle|^2{\rm Re}\left[\sum_{n=0}^{1}\frac{1}{z+\gamma}\left\{1+\frac{8\gamma e^{zp/2}}{(z^2-\gamma^2)(1-e^{zp})}\left[\cosh\left[\frac{p}{2}\gamma\right]-\cosh\left[\frac{p}{2}z\right]\right]\right\}\right]. \tag{13}$$

It appears that the first term inside the large bold parentheses brackets, $1/(z+\gamma)$, corresponds to an unperturbed absorption pattern, while the rest is the rf part. In fact, it can be shown that there is *no* absorption corresponding to the positions of the unperturbed peaks. Expanding Eq. (13) in a power series the term in large bold parentheses is found to be

$$\left[\frac{1}{1-e^{zp}}\right]\left\{\frac{1}{p}\sum_{n=0}^{1}\sum_{m=0}^{\infty}\frac{(z+\gamma)^n}{(m+1)!}\left[\frac{p}{2}\right]^{m+1}\left[\frac{p/2}{m+2}+(-1)^m e^{zp}\left[\frac{p/2}{m+2}-\frac{4}{\gamma}\right]\right]\right\}. \tag{14}$$

All absorption peaks correspond to the $(1-e^{zp})$ term in the denominator [i.e., to the term $\sum_N \exp(zp)^N$ in Eq. (12)], so that resonances occur only when $\mathrm{Im}(zp)$ is an integer multiple of π. That is, whenever $(\omega-\omega_0)$ is an integral multiple of the applied frequency ω_{rf}. Hence, all peaks are separated from $\omega=\omega_0$ by an integral multiple of ω_{rf}, although their intensities depend on γ in such a way that sidebands are enhanced when their position coincides with that of a peak in the unperturbed spectrum.

III. COMPARISON OF COHERENT REVERSAL LINE SHAPE WITH EXPERIMENT

Our line shape [Eq. (13)] is plotted in Fig. 2 for several values of ω_{rf}, assuming that the excited state splitting of the ^{57}Fe sublevels is 21 Mhz [so $\alpha(\frac{3}{2},\frac{1}{2})$, the half separation of lines one and six, is approximately 50 MHz]. The collapse of the spectrum at high frequency is obvious (see Fig. 2), as are sidebands. We stress that these sidebands are not magnetostrictively generated, but rather arise from the periodic local moment reversals. Their presence is strongly suggested by the way we have written the line shape in Eq. (14). Thus we conclude, in agreement with Olariu *et al.*,[17] that the assumption of previous workers[6,7,8,16,19] that sidebands can only arise through magnetostrictively generated vibrations, is not correct.

It is immediately clear that this line shape is not consistent with experiments on nonmagnetostrictive materials[13,14] and vibrationally damped magnetostrictive materials[19] in which rf collapse was observed with no sidebands, since in this model the collapse cannot occur without sidebands appearing. But given the scarcity of such experiments compared with those on magnetostrictive materials, it is worth our while to compare this line shape with data on undamped magnetostrictive foils as well.

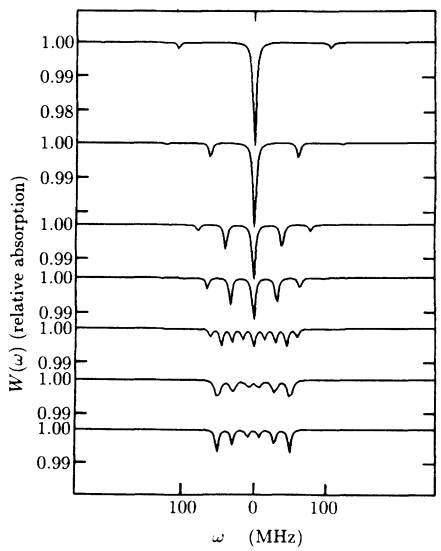

FIG. 2. A plot of the coherent reversal line shape, Eq. (13), for $\alpha(\frac{3}{2},\frac{1}{2})=50$ MHz and $\omega_{rf}=0$, 4, 15, 32, 39, 60, and 106 MHz, going from bottom to top.

Comparing Figs. 1 and 2 reveals a gross lack of agreement between Eq. (13) and the experiments on rf collapse in magnetostrictive materials. Especially the sidebands in Eq. (13) are basically nonexistent for n greater than 4 at 32 MHz, while they appear to be strong up to perhaps $n=20$ or 30 in Fig. 1. In Fig. 3 we have plotted as a function of ω_{rf} the ratio of the first sideband to the carrier from (a) Eq. (13), (b) the calculation of Olariu *et al.*,[17] and (c) three data points due to Pfeiffer.[7] There is good agreement at high frequency (which is what led Olariu *et al.* to state that the magnetostrictive mechanism of sideband generation is unnecessary), but the low-frequency agreement is again quite poor. (Note that the error in frequency in Fig. 3 is negligible and that the theory of Olariu *et al.* does not apply at low frequency.)

This lack of agreement between Eq. (13) and the data on magnetostrictive materials is not an artifact of our hyperfine Hamiltonian. That is, no other periodic time dependence of the magnetic hyperfine field will produce better agreement, because the square-wave behavior of $H_{hf}(t)$ should generate high-order sidebands better than any other reasonable model, since a square wave has large harmonic content. (By reasonable model, we mean one that preserves the magnitude of the magnetic hyperfine field at the nucleus.) It is furthermore obvious that any random deviations from periodicity in $H_{hf}(t)$ will degrade the sideband intensity, leading to further divergence between this model and the data.

There seems to be no way to reconcile the existence of high-order sidebands in the experimental spectra of magnetostrictive materials with hypothesis I as it stands. However, we have found that good agreement is possible if we invoke a *modified* magnetostrictive model which satisfies symmetry requirements. To do this we use a phenomenological expression due to Pfeiffer to generate sidebands to Eq. (13) at *even* integer multiples of the applied frequencies, i.e., at $\pm 2n\omega_{rf}$. In this model, the intensity of the $2n$th sideband to a line is given by

$$I_{2n}=e^{-m^2}\mathcal{J}_{2n}(m^2)\ , \tag{15}$$

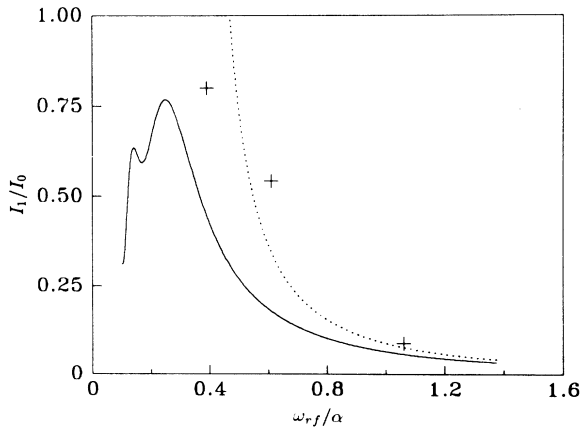

FIG. 3. Ratio of the intensity of the first sideband (I_1) to the carrier (I_0) vs ω_{rf} for Eq. (13) (solid line), the expression of Olariu *et al.* (Ref. 17) (dotted line), and the data of Pfeiffer (Ref. 7, Fig. 12) (crosses).

where $\mathcal{J}_n$ is the modified Bessel function of the first kind of order n and the "modulation index" m depends on ω_{rf}, but, at a given ω_{rf} not on the sideband order n.[7] The results are shown in Fig. 4. The carrier spectra are those shown in Fig. 2, and we choose modulation indices m that give good qualitative agreement with the results of Ref. 7. The modulation indices appear to be physically reasonable. With this model we can thus, for the first time, explain the intensity of the sidebands observed in the collapse effect in magnetostrictive materials in a consistent and plausible way, and hypothesis I can lead to spectra similar the those observed in vibrationally undamped magnetostrictive materials.

There is, however, a further experiment which this hypothesis cannot explain. If the rf field is indeed controlling the magnetization, then as soon as the rf field is turned off the magnetization should be constant and stable, in conflict with the result[21] that the local magnetization continues to reverse with zero time average for at least short times after the field is pulsed off. This effect is not observed in all materials.[6]

Our feeling is that the coherent reversal model is likely wrong, because its acceptance requires that we disregard (a) experimental evidence that sidebands are absent in collapsed spectra of nonmagnetostrictive materials and vibrationally damped magnetostrictive materials, and (b) evidence that at least in some materials the local magnetization is not stable for short times after the rf field is turned off.

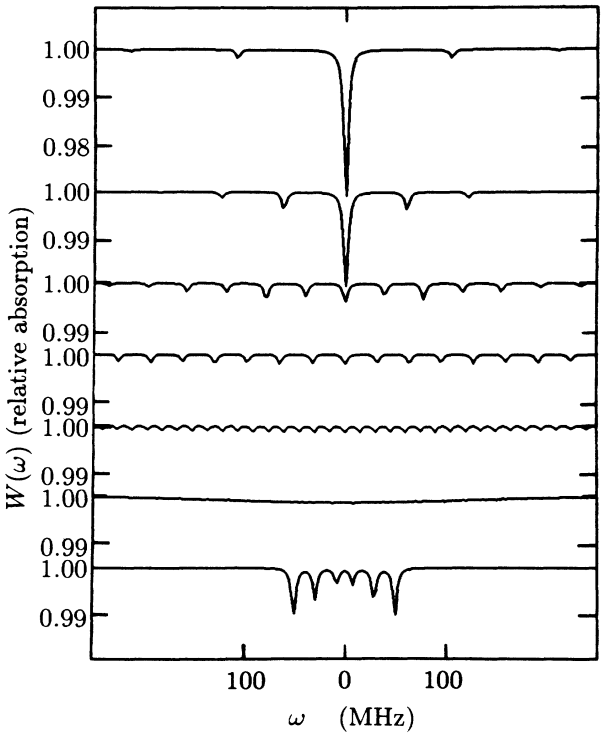

FIG. 4. A plot of Eq. (13) plus the effect of even-order magnetostrictive sidebands with values of ω_{rf} and m (0,0), (4,800), (15,200), (32,100), (39,30), (60,1.4), and (106,0.8) going from bottom to top, with $\alpha(\frac{3}{2},\frac{1}{2})=50$. The values of the modulation index were chosen so that the resulting spectra agree with experimental spectra (Ref. 7).

IV. SIDEBANDS IN SPECTRA WITH AN INCOHERENTLY FLUCTUATING MAGNETIZATION (HYPOTHESIS II)

In this section we investigate the possibility that hypothesis II, proposed in the Introduction, can account for the experiments. In particular we want to show that a model can be constructed which predicts both collapse and odd- and even-order magnetostrictive sidebands. Recall that hypothesis II amounts to the assumption that magnetostriction is responsible for all sidebands in the spectra. For this to be true requires three things: (1) as discussed by Olariu *et al.*[17] and in the Introduction, and as explicitly demonstrated below, production of odd-order sidebands means that the entire magnetization cannot reverse coherently with frequency ω_{rf}, (2) production of sidebands by magnetostriction requires that at least some part of the magnetization follow the rf field at frequency ω_{rf}, and (3) for the spectra to collapse the time average hyperfine field at each nucleus must be zero and the fluctuations must be faster than the inverse of one half of the splitting of the spectrum. (That this last point is true for a periodically reversing moment is demonstrated in the next section, while for a stochastically reversing moment it has been proven by Blume and Tjon.[34]) A magnetization which satisfies these requirements may be written as the sum of a stochastic and periodic part with frequency ω_{rf}

$$\mathbf{M}(\mathbf{r},t)=m_s\hat{\mathbf{f}}_s(\mathbf{r},t)+m_p[\hat{\mathbf{z}}\sin(\omega_{rf}t)+\hat{\mathbf{y}}\cos(\omega_{rf}t)]\ , \qquad (16)$$

where m_s is the magnitude of the stochastic part of the magnetization, $\hat{\mathbf{f}}_s(\mathbf{r},t)$ is a unit vector with stochastic time dependence and zero time average in any direction, and m_p is the magnitude of the periodic part of the magnetization. In general, the periodic part could include higher harmonics, but these do not contribute to magnetoelastic waves at ω_{rf}, and so do not help to generate odd order sidebands, so we are not interested in such terms.

This time dependence [Eq. (16)] may appear peculiar, but we have a specific model in mind, which is as follows. There are good indications that observed cluster behavior in iron-nickel alloys[35,36,37] and competing ferromagnetic and antiferromagnetic interactions produce magnetic clusters[26,27] with the property that spins within a cluster are strongly coupled but the cluster-cluster coupling is weak (for a discussion see Ref. 27). We propose that the effect of the rf field is to break up the cluster-cluster order, without disturbing the magnetic order within clusters. The clusters then behave superparamagnetically, fluctuating stochastically, and possess a macroscopic moment which could interact with the rf field giving rise to a small periodic component. Since the clusters have finite size, they can produce magnetoelastic waves (unlike the paramagnetic state, which would give a collapsed spectrum but no vibrational sidebands).

We now follow the discussion of Pfeiffer *et al.*[9] to calculate the time-dependent lattice strain produced by

$\mathbf{M}(\mathbf{r},t)$. The strain measured in a direction l is

$$\delta l(\mathbf{r},t)/l = \tfrac{3}{2}\Lambda \cos^2\theta(\mathbf{r},t) , \tag{17}$$

where $\theta(\mathbf{r},t)$ is the angle between l and $\mathbf{M}(\mathbf{r},t)$, and Λ is the magnetostrictive constant of the material (which in an inhomogeneous material would show some spatial dependence, which we ignore). Now using

$$\cos\theta(\mathbf{r},t) = \frac{l\cdot\mathbf{M}(\mathbf{r},t)}{|l|\ |\mathbf{M}(\mathbf{r},t)|} \tag{18}$$

we investigate two obvious limits of $\mathbf{M}(\mathbf{r},t)$ which are of interest: $m_s \gg m_p$ and $m_s \ll m_p$. In the latter limit the coherent rotation part dominates. For both limits the strain is easily calculated to the lowest-order periodic term.

In the limit $m_s \ll m_p$ we find

$$\delta l(\mathbf{r},t)/l = \tfrac{3}{2}\frac{\Lambda m_p^2}{|l|^2}\{[l\cdot\hat{\mathbf{z}}\sin(\omega_{\rm rf}t)+l\cdot\hat{\mathbf{y}}\cos(\omega_{\rm rf}t)]^2 + O(m_s/m_p)\} . \tag{19}$$

The right-hand side is a periodic function with frequency $2\omega_{\rm rf}$, so vibrations must be created at this frequency, and only even order vibrational sidebands are produced.

In the limit where the stochastic part of the magnetization dominates ($m_s \gg m_p$) we find

$$\delta l(\mathbf{r},t)/l = \tfrac{3}{2}\frac{\Lambda}{|l|^2}\left[[l\cdot\hat{\mathbf{f}}_s(\mathbf{r},t)]^2+\frac{2m_p}{m_s}[l\cdot\hat{\mathbf{f}}_s(\mathbf{r},t)]\{l-[l\cdot\hat{\mathbf{f}}_s(\mathbf{r},t)]\hat{\mathbf{f}}_s(\mathbf{r},t)\}[\hat{\mathbf{z}}\sin(\omega_{\rm rf}t)+\hat{\mathbf{y}}\cos(\omega_{\rm rf}t)]+O((m_p/m_s)^2)\right] . \tag{20}$$

The first term produces stochastic strains which do not contribute to sidebands. The next term is periodic with frequency $\omega_{\rm rf}$, and we propose that scattering of these vibrations into the direction perpendicular to the plane of the foil, perhaps by the mechanisms suggested by Pfeiffer *et al.*[9] would produce vibrations at this frequency. This would produce odd- and even-order vibrational sidebands in the spectra. In nonmagnetostrictive materials, of course, Λ is zero so no magnetostrictive sidebands appear, and, equally important, because the periodic component of the local magnetization is small the periodic part of the magnetic hyperfine field at any given site will be negligible compared with the stochastic part, so no sidebands arise from this source either.

It may be objected that the term periodic in $\omega_{\rm rf}$ in Eq. (20) is odd in $\hat{\mathbf{f}}_s(\mathbf{r},t)$, so it has both time-averaged and instantaneous spatial average of zero, so that this term may contribute nothing to coherent vibrations of the foil. This may be so, but until it is proven so—which requires that we first understand the mechanisms that scatter vibrations out of the plane of the foil—or until a better explanation without this difficulty is advanced, we feel that this objection should not be accepted. It may be relevant that, due to domain structure in hard magnetic materials, the spatial average of this term is also zero in the calculation of Pfeiffer *et al.*[9] in the absence of an external field.

A magnetization of the form of Eq. (16), with $m_s \gg m_p$, further explains in an obvious way the experiments of Kotlicki.[21] When the rf field is suddenly switched off, the small periodic part dies, so the sidebands disappear, but the large stochastic part continues to fluctuate until long-range cluster-cluster order is reestablished, so the spectrum remains collapsed for a certain amount of time after the field is switched off. The fact that the magnetic splitting re-appears in the ferrite studied by Albanese *et al.*[9] but not in the Invar foil studied by Kotlicki[21] could be ascribed to a faster relaxation time for long-range order to be established in the ferrite. It would be very interesting to collect several spectra, at several delay times after the rf field is pulsed off, to see how long it takes for the stable local magnetization to be reestablished. The dynamics of this process may be related to spinodal decomposition. It has been demonstrated that in some materials it takes hours for long range cluster-cluster order to be established, despite the fact that the order within cluster is well defined.[38]

This model can thus explain how the collapse effect can occur and yet be compatible with (a) the absence of sidebands in zero magnetostriction and vibrationally damped magnetostrictive materials, (b) the presence of odd-order sidebands in undamped magnetostrictive materials, and (c) experiments with pulsed rf fields in which the spectrum was found to be collapsed both with the field pulsed on and off, but with sidebands present only when the field was pulsed on.[21]

We finally comment that our picture in which the magnetic clusters fluctuate stochastically with a small component that follows the rf field seems more compatible with the strong, random, uniaxial anisotropy that is often found in soft magnetic iron-nickel foils.[39] It is difficult to believe that relatively weak rf fields could control the magnetization of all clusters, including holding the majority of them along hard directions, as is required in the coherent reversal model. It is important to distinguish here between rotating the net magnetization of the foil and the local magnetization at all points in the foil, since the two are not necessarily equivalent. It is the local magnetization that Mössbauer spectroscopy observes. It is far more plausible that the cluster magnetizations fluctuate in direction along their easy axes.

V. DISCUSSION OF DETAILS OF COHERENT-REVERSAL LINE SHAPES

Although we feel that hypothesis I does not describe existing experiments, this does not mean that such experiments will never be done. Therefore we wish, as an aside, to point out some of the salient features of the line

shape which we have derived under hypothesis I [Eq. (13)]. Readers not interested in the details of this line shape should proceed to the next section.

It will be assumed for the sake of clarity that magnetostrictively generated sidebands are not present. We begin with a discussion of the frequency dependence of the collapse of the carrier spectrum due to a coherently reversing magnetization. The impression one gets from the literature[6-8,16,19] is that as the frequency of the rf field is increased the spectrum should be more-or-less unaffected up to some critical frequency, at which point the reversals occur so quickly that the nucleus can no longer follow the rf field, and the spectrum collapses. Alternatively, it has been suggested that there is an intermediate frequency regime in which the magnetic hyperfine field is degraded, so that the spectrum narrows and then collapses. It is generally stated (e.g., Ref. 7) that the critical frequency for collapse is related to the Larmor precession frequency of the nucleus in the hyperfine field, although the exact relation has not been presented previously.

There are two points to be made. First, in Eq. (13) it is true that there is a low-frequency regime where the spectrum is not collapsed, but it is not true that the rf field has *no* effect in this regime, since, as we have seen, the rf field determines the positions of the absorption peaks at all frequencies. This is to be contrasted with the rf sideband effect, where the applied field never affects the positions of the peaks in the carrier spectrum. Equally clearly, the peaks in the spectrum do not shift inwards before the collapse occurs. Furthermore, the collapse occurs gradually, not suddenly, and it occurs independently for the three pairs of lines (3-4, 2-5, 1-6), at a frequency depending roughly on the splitting of each pair of lines. This is illustrated in Fig. 5, where the absorption only by lines 1 and 6 is shown. If we define the spectrum as collapsed when the carrier peak is larger than the $n=1$ (and higher-order) sideband, then the collapse occurs when the applied frequency is somewhat less than $\alpha(\frac{3}{2},\frac{1}{2})$—half of the magnetic hyperfine splitting of the pair of lines in question. This frequency corresponds to 50 MHz in Fig. 5. The collapse process can be summarized by saying that as the applied frequency increases the intensity of the lower-order sidebands increases at the expense of the higher-order ones.

The detailed dependence of the intensity of the sidebands on the applied frequency is quite complicated, as can be seen in Figs. 6 and 7, where the intensities of some of the low-order sidebands due to the $\pm\frac{3}{2}\rightarrow\pm\frac{1}{2}$ transitions are shown. It is unlikely that the sideband intensity can be followed experimentally at low frequencies, but it is none the less interesting to describe some of the general features. The intensity of the nth order sideband I_n has its highest maximum at or below $\omega_{rf}=\alpha(\frac{3}{2},\frac{1}{2})/n$, at which frequency the sideband energy corresponds to the energy of the unperturbed absorption peak. The intensity minima on the other hand occur at frequencies $\alpha(\frac{3}{2},\frac{1}{2})/(2n)$ for even-order sidebands, for all n greater than the order of the sideband in question, and at $\alpha(\frac{3}{2},\frac{1}{2})/(2n+1)$ for odd-order sidebands. For example,

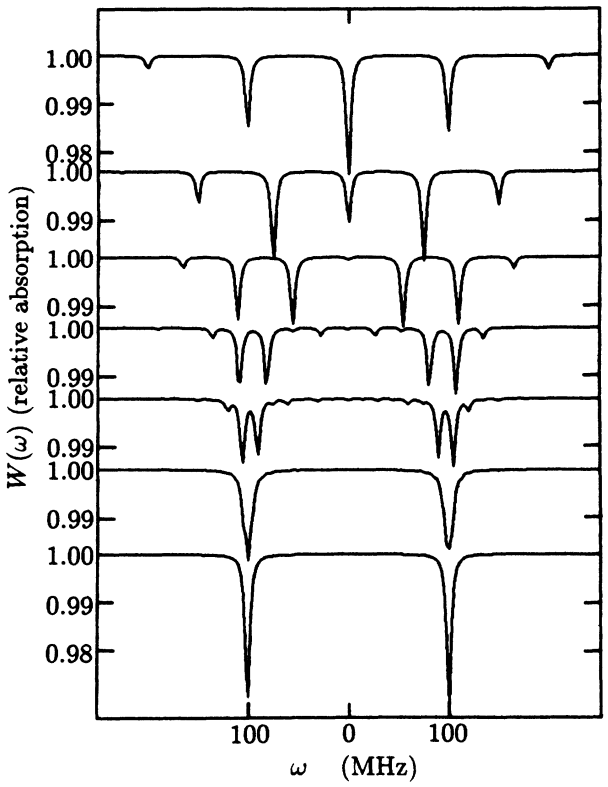

FIG. 5. Absorption due to the transitions $m_1=\pm\frac{3}{2}$ to $m_0\pm\frac{1}{2}$, from Eq. (13), for values of $\omega_{rf}/\alpha(\frac{3}{2},\frac{1}{2})=0$, 0.08, 0.16, 0.26, 0.6, 0.9, and 1.4, going from bottom to top, in the absence of magnetostrictive sidebands.

in Fig. 6 it can be seen that the carrier peak ($n=0$) has minima at $\omega_{rf}/\alpha=\frac{1}{2}$, $\frac{1}{4}$, $\frac{1}{6}$, etc., while the $n=1$ sideband has minima at $\omega_{rf}/\alpha=\frac{1}{3}$, $\frac{1}{5}$, $\frac{1}{7}$, etc., and the intensities of the even and odd-order sidebands are reciprocal, so that peaks in the even-order sidebands occur at valleys in the

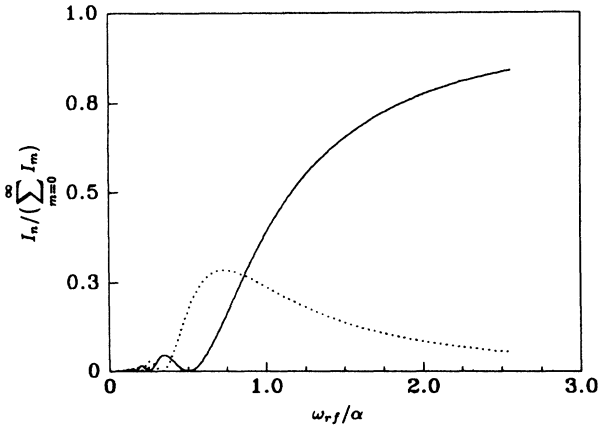

FIG. 6. Fractional intensity of the carrier I_0 (solid line) and the first sideband I_1 (dotted line) for the transition $\pm\frac{3}{2}$ to $\pm\frac{1}{2}$ plotted against $\omega_{rf}/\alpha(\frac{3}{2},\frac{1}{2})$, in the absence of magnetostrictive sidebands.

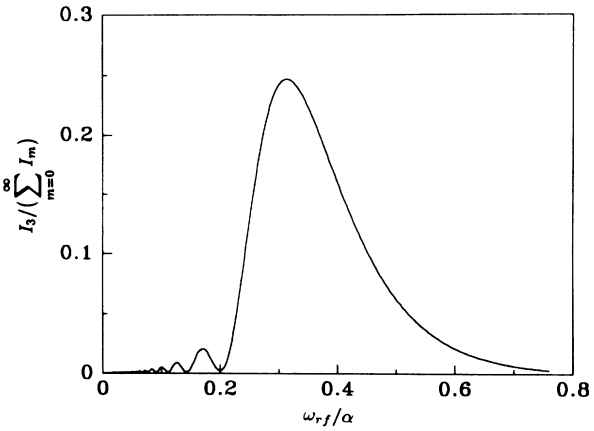

FIG. 7. Fractional intensity of the third sideband I_3 for the transition $\pm\frac{3}{2}$ to $\pm\frac{1}{2}$ plotted against $\omega_{rf}/\alpha(\frac{3}{2},\frac{1}{2})$ in the absence of magnetostrictive sidebands.

odd-order sidebands, and vice versa. The total absorption intensity in the spectrum is of source constant, and in the absence of all other sidebands and the carrier peak, it appears that a given sideband would increase monotonically up to $\omega_{rf}=\omega(\frac{3}{2},\frac{1}{3})/n$, and then decrease monotonically at higher frequencies. The added complication occurs because whenever a given sideband reaches its "central maximum" [the one closest to $\alpha(\frac{3}{2},\frac{1}{2})/n$], intensity is channeled into that sideband and out of the lower-order sidebands. Furthermore, as the carrier peak grows, it takes intensity from the rest of the spectrum so each central maximum is shifted to lower frequencies. This is particularly evident for small values of n. Thus the central maximum of I_4 is at $\alpha(\frac{3}{2},\frac{1}{2})/4$, but that of I_1 is at $0.73\alpha(\frac{3}{2},\frac{1}{2})$, due to the increased size of the carrier (I_0).

VI. CONCLUSIONS

We have looked at two solutions to the conflict between the supposed magnetostrictive origin of the odd-order sidebands observed in the rf collapse effect and the supposition that the spectra collapse because the magnetization reverses coherently at the radio frequency.

The first solution (hypothesis I), originally proposed by Olariu *et al.*,[17] is that the magnetization is indeed reversing coherently but that the sidebands are produced by the periodically reversing magnetic hyperfine field at each nucleus. Using an adiabatic approximation we have calculated the spectra for this physical situation and showed that (a) the sidebands produced by the reversing hyperfine fields are too strong to explain the absence of sidebands in the collapsed spectra of nonmagnetorestrictive and vibrationally damped magnetostrictive foils[13,14,19] although (b) they can roughly explain the sidebands observed in undamped magnetostrictive foils if even-order magnetostrictive sidebands are present (in agreement with symmetry requirements). We have also made the point that hypothesis I conflicts with the finding that in some situations the spectra remain collapsed for short times after the rf field is pulsed off.[21]

The second solution (hypothesis II) that was investigated assumed a local magnetization at each point in the foil that has a large stochastic part and a small periodic part. This hypothesis appears to give results that are consistent with all of the current experiments. The physical picture underlying this hypothesis is that the magnetic moments are tightly bound in small clusters that interact weakly with each other. The effect of the rf field is to break up the long-range cluster-cluster ordering, so that the clusters fluctuate superparamagnetically with a small component that follows the rf field. We thus conclude on the basis of experimental evidence that the effect of the strong rf field on a soft magnetic foil is to destroy the long range correlations of the local magnetization, as opposed to controlling the total magnetization.

We are currently investigating theoretical spectra for a magnetic hyperfine field with variable stochastic and periodic parts, with the intention of (a) putting a limit on the size of the periodic part, given a maximum size of the first sideband in vibrationally damped foils, and (b) of simulating experiments in which the rf intensity is varied.

Experimentally, it appears to be important to establish better limits on maximum sideband intensities in nonmagnetostrictive and vibrationally damped magnetostrictive foils. Better spectra are also required from the planar ferrite studied in Ref. 6, since existing spectra are very noisy but contain some indication that the rf field is in fact controlling the moments. Ideally, investigations of the collapse effect would be done in the pulse mode, and it would probably be interesting to investigate the re-establishment of a stable local magnetization (and hence long-range order) in Invar and Permalloy foils after irradiation with rf fields.

ACKNOWLEDGMENTS

The financial support of the Natural Sciences and Engineering Research Council of Canada (NSERC) is gratefully acknowledged. We are grateful to Professor Denis Rancourt for helpful discussions.

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