AMPLIFIED FERRIMAGNETIC ECHOES*

D. E. Kaplan, R. M. Hill, and G. F. Herrmann
Lockheed Palo Alto Research Laboratory, Palo Alto, California
(Received 15 April 1968)

Strongly amplified magnetostatic mode echoes in a ferrite are observed at X-band frequencies and temperatures from 300 to 1.6°K. Echo formation is discussed in terms of a mechanism in which the resonance frequency of a mode is a function of its amplitude of excitation.

We have observed strongly amplified echoes in a ferrite following typical two- and three-pulse echo sequences. Two-pulse echoes were generated with intensities exceeding that of the first pulse by a factor of 10^5. An important property of these echoes is that their amplitudes initially increase with increasing pulse separation, reaching a maximum value and then decreasing as relaxation becomes dominant. This behavior contrasts sharply with that of the usual spin echo[1] and previously observed magnetostatic mode echo,[2] which decay monotonically with pulse separation, with an echo intensity always smaller than that of either exciting pulse.

The experimental observations were made on single crystals of yttrium iron garnet of several arbitrary shapes, in a dc magnetic field. The microwave pulses were generated and detected with an apparatus previously described.[3] We have been able to find amplified echoes throughout the X-band frequency range (8.2-12.4 GHz) with echo intensity 10^3 to 10^5 greater than the intensity of the first pulse. Inhomogeneous microwave magnetic fields were used to excite preferentially volume magnetostatic modes of short wavelength. The effect was observed from 300 down to 1.6°K with only a slight increase in decay time at lower temperature due to a corresponding decrease in mode linewidth. A two-pulse echo sequence is shown in Fig. 1(a). The first pulse, increased by 10^3 for comparison, is displayed on the receiver trace using a double exposure technique. The second pulse, which is 10 nsec in duration, is electronically blanked from the display. Echoes could be observed with a first-pulse power as low as 5×10^{-12} W, which is close to the thermal noise level under our experimental conditions. The peak power of the second pulse for maximum amplification is typically 1-5 W with a duration of 10-100 nsec. As much as 0.1% of the second-pulse power may be returned in the echo. An echo envelope obtained by multiple exposure of an oscilloscope trace for a variable pulse separation is presented in Fig. 1(b) and shows the typical rise and decay pattern

We believe that the energy-storage process is similar to that of spin echo, namely, storage of the excitation among a large number of oscillation modes within the Fourier spectrum of the excitation pulses. The net magnetic moment produced by the pulses vanishes rapidly as the modes lose their phase coherence. An echo results from the return of phase coherence at specified times. In this case the mechanism for restoring coherence at echo time must be quite different from that of spin echo. A number of alternative nonlinear mechanisms, all of which in principle can result in echoes, have been suggested[3-6] in connection with the discovery of cyclotron echo.[3] The observed behavior of this ferrimagnetic echo suggests that the amplification results from a dependence of the mode frequency upon mode amplitude.[5,6] Such amplitude dependence can arise in principle from any of the nonlinear interactions in the spin system, but

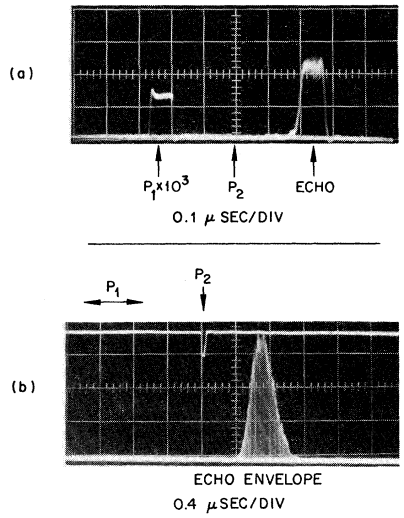

FIG. 1. (a) Two-pulse echo in single-crystal yttrium iron garnet illustrating amplification of the first pulse by ~10^3. The second pulse is electronically blanked. (b) Envelope of amplified two-pulse echo as the pulse interval is varied. The upper trace monitors the position of Pulse 2. The position range of Pulse 1 is indicated by the double-ended arrow.

the dipolar interaction seems the most likely candidate. An easily visualized mechanism is provided, for example, by the decrease in parallel dipolar fields with increased precession angles.

In order to avoid a lengthy mathematical discussion, we confine ourselves to a qualitative exposition of the process by which amplitude-dependent frequencies result in echoes. We treat the problem in a random phase approximation; i.e., it is assumed that at a time $t=\tau$ sufficiently long after the incidence of the first pulse of amplitude A_1 the mode density $n(\theta)$ is a constant, independent of the phase angle θ [see Fig. 2(a)]. The resultant transverse magnetization, obtained by summing over all modes, equals zero. At $t=\tau$ a second pulse of amplitude A_2 and duration ϵ is introduced. Under linear conditions, at $t=2\tau$, $n(\theta)$ will again be a constant, Fig. 2(a). If the first pulse is very small compared to the second, the angle θ_k of a particular mode will be essentially the same at $t=2\tau$ as at $t=\tau$, just prior to the incidence of the second pulse, as the phase of each mode drifts by the same angle during the intervals $0<t<\tau$ and $\tau<t<2\tau$. Superposition of the two excitations on a particular mode following the second pulse at $t=\tau+\epsilon$ may be represented in the form $A^2=A_1{}^2+A_2{}^2+2A_1A_2\cos\theta_k$. The explicit form of the frequency dependence will vary with the experimental conditions. To illustrate a simple case we choose the oscillator frequency ω to vary with $-A^2$, which is the contribution to the frequency shift from the lowest order terms in the Hamiltonian expansion discussed below. Under this assumption one obtains a θ-dependent frequency shift $\Delta\omega(\theta)=-\alpha A_1A_2\cos\theta$, Fig. 2(b), where α is an arbitrary coefficient. The arrows indicate schematically the sense and magnitude of the additional phase drift imposed due to $\Delta\omega(\theta)$. Thus, at $t=2\tau$ a given mode will arrive not at the angle θ_k but at $\theta_k-\alpha\tau A_1A_2\cos\theta_k$. The mode density $n(\theta)$ is therefore bunched toward $\theta=-\pi/2$, Fig. 2(c), resulting in a net transverse magnetic moment in that direction, and hence an echo. As τ equals $\sim10^4$ precession cycles, a very small frequency shift can produce a large echo. Echo energy is drawn from the second pulse, the first pulse providing the θ dependence of the phase shift. Large amplification is consequently possible. Note also that since the phase shift at $t=2\tau$ is proportional to τ the echo will increase monotonically with τ until degraded by relaxation effects.

It is necessary to add some remarks concern-

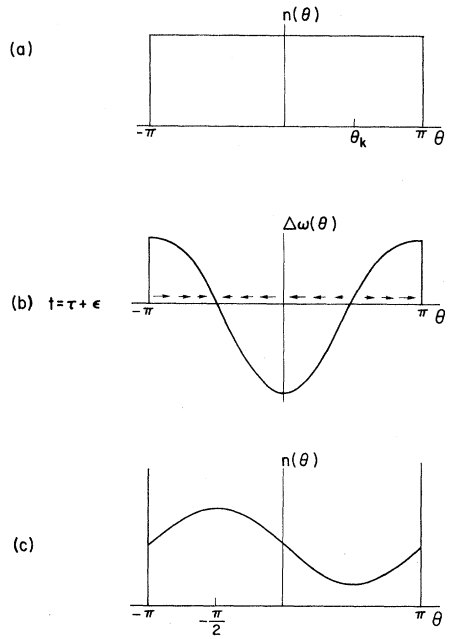

FIG. 2. Echo formation due to an amplitude-dependent frequency. (a) Mode density $n(\theta)$ at $t=\tau$, and at $t=2\tau$ in the linear case. (b) Amplitide-dependent frequency shift $\Delta\omega(\theta)$ following the second pulse. (c) Mode density $n(\theta)$ at $t=2\tau$ resulting from phase bunching induced by frequency shift.

ing the nature of the modes and their relation to the magnetic Hamiltonian.[7] The Hamiltonian is usually given as an expansion in terms of canonical spin-wave variables u_k. At low excitation levels $\mathcal{H}=\sum_k\omega_k u_k u_k{}^*$, and u_k represents the amplitude of a normal mode at frequency ω_k. At high signal level higher order terms arising from dipolar and exchange interactions and from crystalline anisotropy must be included, e.g., $u_k u_{k'}{}^* u_{k''} u_{k'''}{}^*$, etc. It is easily seen that terms of the form $u_k u_k{}^* u_k u_k{}^*$, and higher even-order "self-interaction" terms, represent (in a dissipationless system) an amplitude-dependent frequency shift. However, the mere presence of such terms in the Hamiltonian does not guarantee the appearance of an echo, as they must compete with all the interactions among modes of different k. In fact, if the modes are true "momentum" states of the form $\exp(ikr)$, then it can be shown on the basis of complementarity arguments that no echo can occur.[8] On the other hand, if the modes are spatially localized, the self-interaction terms for which $k=k'=k''=k'''$ are favored and echoes will result. In the present experiment localization is obtained by coupling to short-wavelength magnetostatic modes ($\lambda\sim0.001$-0.01 cm) whose group velocity is small

over a wide range of wavelengths, and by providing inhomogeneous internal fields by the choice of irregularly shaped samples. Some mode overlap must remain because of exchange and will contribute to the echo decay by dephasing the echo components.

We would like to acknowledge valuable assistance of Mr. Steve Ichicki in these experiments.

*This work was supported jointly by the Lockheed Independent Research Fund and the U. S. Office of Naval Research under Contract No. NOnr-2541(00).

[1]E. L. Hahn, Phys. Rev. 80, 580 (1950).
[2]D. E. Kaplan, Phys. Rev. Letters 14, 254 (1965).
[3]G. F. Herrmann, R. M. Hill, and D. E. Kaplan, Phys. Rev. 156, 118 (1967).
[4]G. F. Herrmann and R. F. Whitmer, Phys. Rev. 143, 122 (1966).
[5]W. H. Kegel and R. W. Gould, Phys. Letters 19, 531 (1965); R. W. Gould, Phys. Letters 19, 477 (1965).
[6]J. M. Wachtel and J. L. Hirshfield, Phys. Rev. Letters 17, 348 (1966).
[7]E. Schlömann, Raytheon Co. Technical Report No. R-48, 1959 (unpublished).
[8]This result follows because the dipolar interaction is a function of position coordinates only. This point will be analyzed in detail elsewhere.

CRITICAL-POINT MAGNETIZATION OF AN IMPURITY IN AN ANTIFERROMAGNET

G. K. Wertheim, H. J. Guggenheim, and D. N. E. Buchanan
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received 8 April 1968)

The magnetization of Fe^{2+} in MnF_2, measured in terms of its magnetic hfs interaction, exhibits a critical exponent β equal to that of the host lattice. The large change in the coefficient D is ascribed to the effects of anisotropy and to the difference between the Fe-Mn and Mn-Mn exchange.

It is well known that the temperature dependence of the magnetization of a spin impurity in a magnetic solid may be quite different from that of the host. This effect can be demonstrated within the confines of molecular-field theory and arises whenever the impurity spin or its coupling to the molecular field differs from that of the host atoms. This effect has also been treated from the point of view of spin-wave theory.[1] However, neither of these two approaches provides a valid description of the temperature dependence of the magnetization of a pure substance in the critical region.[2] The behavior of an impurity in that region is therefore best approached by comparing it with the behavior of the host lattice.

For this experiment we chose MnF_2 as the host lattice because its critical behavior has been more thoroughly analyzed than that of any other magnetic material,[3,4] and Fe^{2+} as the impurity because it can be studied by Mössbauer effect. A recent examination of the magnetization of Fe^{2+} in MnF_2 in the spin-wave region[5] showed that the impurity magnetization drops much more slowly with increasing temperature than the magnetization of the host. This effect arises because the Fe-Mn exchange is larger than that between Mn atoms and also because of the greater anisotropy of the D-state impurity. The difference in spin, $\frac{5}{2}$ for Mn^{2+} and 2 for Fe^{2+}, also contributes. We here report the extension of these measurements into the critical region.

The experiment was carried out with MnF_2 into which radioactive $^{57}Co^{2+}$ had been introduced as a dilute impurity.[6] On the basis of the total activity, the crystal contained $\sim 10^{16}$ atoms/cm^3 of ^{57}Co. The cobalt decays by electron-capture to ^{57}Fe, which then emits the 14.4-keV gamma ray used in the Mössbauer-effect experiment. The spectrum was obtained with a conventional constant-acceleration spectrometer, using a single-line $K_4{}^{57}Fe(CN)_6 \cdot 3H_2O$ absorber. The MnF_2 sample was immersed in liquid nitrogen. Temperature was regulated by controlling the vapor pressure with a Cartesian diver manostat.

Experiments in the paramagnetic region show that the $^{57}Fe^m$ produced by the electron-capture decay of the divalent ^{57}Co is entirely in the divalent state when the Mössbauer gamma ray is emitted. This is in accord with the finding in a similar experiment on ^{57}Co in rutile structure ZnF_2.[7] The Mössbauer-effect hyperfine spectra obtained in the critical region are very similar to those in the critical region of FeF_2.[8] They

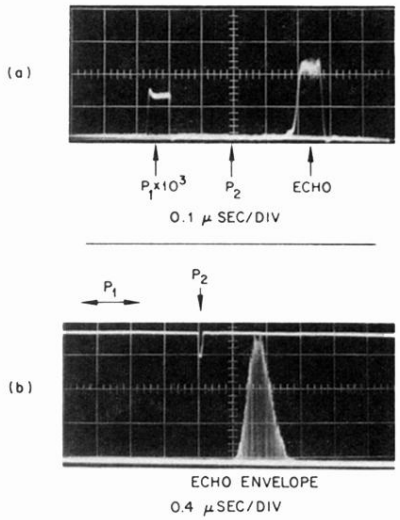

FIG. 1. (a) Two-pulse echo in single-crystal yttrium iron garnet illustrating amplification of the first pulse by $\sim 10^3$. The second pulse is electronically blanked. (b) Envelope of amplified two-pulse echo as the pulse interval is varied. The upper trace monitors the position of Pulse 2. The position range of Pulse 1 is indicated by the double-ended arrow.