is always smaller than the frozen-lattice gap as expected.

The dispersion curve of the magnons starts very slowly in the small-q region. So in an inelastic neutron scattering or a high-frequency ferromagnetic resonance experiment the free-lattice–mode contribution is totally buried under that of the frozen-lattice mode because the latter has an overwhelmingly larger density of states. However, at low enough frequencies and with an applied field near the spin-flip value, the number of thermally excited free-lattice magnons may be large enough so that they produce a sizable amount of resonance absorption of the microwave energy. The rf field at lower frequencies penetrates deeper into the specimen, so it couples more effectively with the free-lattice mode. Since the magnetoelastic interaction is stronger at lower temperatures, it helps to separate the two gaps so the free-lattice mode will stand out more clearly. All of these factors indicate that the free-lattice mode should be observable at low frequencies, low temperatures, and near the spin-flip field. The details of our calculation of the microwave absorption as a function of frequency, dc field, and temperature will be published elsewhere.[14] In the following Letter Hart and Stanford[15] compare their experimental results with our calculation for Tb metal and low microwave frequencies. The two results are in excellent agreement, providing definitive support for the free-lattice mode proposed above.

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Free-Lattice–Model Ferromagnetic Resonance in Terbium at 24 GHz*

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Our results experimentally confirm free-lattice–model behavior in Tb at low microwave frequencies, as predicted by Vigren and Liu. The resonances below a temperature of 140 K were only observable when the external magnetic field was not precisely aligned along an a axis in the hexagonal plane of the sample.

Apparent contradictions between the high- and low-frequency ferromagnetic resonance results in Tb and Dy have prompted much theoretical speculation.[1] Vigren and Liu[2] have proposed a theory explaining these apparent contradictions by predicting free-lattice–model behavior in Tb and Dy at low microwave frequencies and frozen-lattice–model behavior at high microwave frequencies. The experimental data presented in this Letter provide strong evidence in support of their theory, supplying a satisfying resolution to the apparent contradictions which have existed for some time in the literature. (Previous work on Tb at low frequencies did not go below $T = 140$ K which is the critical region for testing the theory of Vigren and Liu.)

Ferromagnetic resonance studies in Tb at 100 GHz by Wagner and Stanford[3] have provided the definitive experimental evidence for deciding between several spin-wave models proposed by Cooper.[1] The results showed conclusively that the frozen-lattice model was appropriate for Tb at high frequencies. In contrast, lower-frequency results, for example the 37-GHz work of Rossol and Jones,[4] supported the free-lattice model for Dy. Recent 100-GHz work on Dy by Wagner and Stanford[5] has shown that again, at high microwave frequencies, the frozen-lattice model is adequate to explain the experimental data. In this Letter we present experimental evidence similar to the work of Rossol and Jones[4] in Dy, supporting the free-lattice model in Tb at low microwave frequencies as predicted by Vigren and Liu.

The contrast between the 100-GHz Tb data of Wagner and Stanford[3] and our 24-GHz data in Fig. 1 is especially evident at $T = 100$ K where, for similar experimental configurations (given in Fig. 1 by the closed triangles), Wagner and Stanford observe a resonance at 6 kOe (close to the domain alignment field), while Fig. 1 shows a strong resonance at almost 12 kOe. These field differences for different microwave frequencies are adequately accounted for by assuming that the frozen-lattice model applies at 100 GHz and the free-lattice model at 24 GHz as predicted by Vigren and Liu. If the frozen-lattice model is applied at 24 GHz, all of the closed triangles for $T < 140$ K in Fig. 1 would be due to off-resonance absorption, and the 100-GHz Tb data for $T < 140$ K would be expected to have almost the same field dependence as the closed triangles in Fig. 1, but with even greater intensity,[1] which is obviously not the case.

Figure 2 shows tracings of power absorption versus applied magnetic field (H_{dc}) for H_{dc} aligned close to the a axis and along the b axis (compare with Fig. 1).

The theory of Vigren and Liu assumes the crystal strains to be locally coupled to the ionic spins at all times. For zero-wave-number ($q = 0$) spin waves, all spins oscillate in phase, and the magnetoelastic contribution to the Hamiltonian is that due to a single spin multiplied by the number of spins. This energy is invariant under rotation of the magnetization in the hexagonal plane to first order, so the free-lattice model developed by Cooper[1] holds, which means there is essentially no magnetoelastic contribution to the spin-wave spectrum at $q = 0$. H_{dc} in

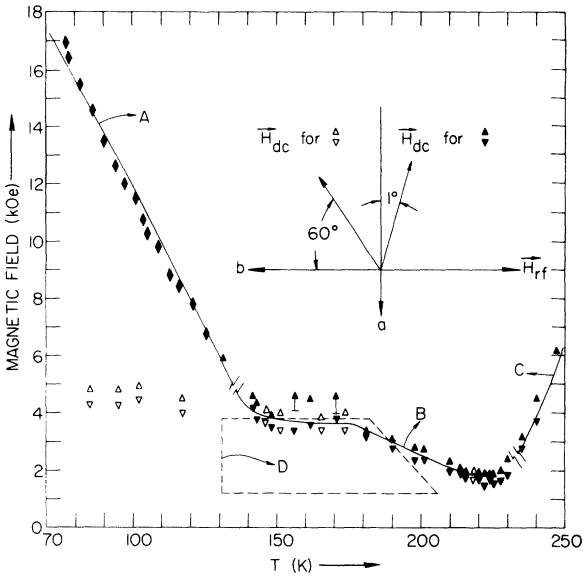

FIG. 1. Plot of the field values of power-absorption maxima as a function of temperature. The diagram gives the experimental configuration for the closed and open triangles. Triangles with the vertex pointing up or down represent data taken while increasing or decreasing H_{dc} with time, respectively. A, calculation by Vigren and Liu of Cooper's free-lattice–model absorption maxima at 24 GHz for $H_{dc} \| a$ axis. B, prediction by Vigren and Liu of off-resonance absorption maxima at the domain-alignment field in Cooper's free-lattice model for $H_{dc} \| a$ axis. Off resonance in this case means that the actual resonance conditions are satisfied for a value of H_{dc} below the domain-alignment field. Becauseof the large linewidth of the resonance, its trailing edge is observed for H_{dc} greater than the domain-alignment field. The open triangles are the domain-alignment field determined by the knee of off-resonance absorption when $H_{dc} \| b$ axis. ($H_{dc} \| b$ axis causes the $q = 0$ spin-wave energy to become a monotonic increasing function of H_{dc} with an energy too high for on-resonance absorption at 24 GHz at the domain-alignment field.) C, paramagnetic resonance. The amplitude of these resonances decreases rapidly with increasing temperature. D, region of nonresonance absorption maxima (not plotted) due to domain alignment when using the thicker sample with H_{dc} oriented 1° from the a axis.

the hard $\langle 11\bar{2}0 \rangle$ direction can lower the spin-wave energy gap to our experimental frequency in this model.

For $q \neq 0$ spin waves, however, the spins are not in phase; and this phase difference causes a cancelation of all but the equilibrium magnetoelastic energy, resulting in the frozen-lattice model proposed by Turov and Shavrov[6] and extended by Cooper,[1] giving a large magnetoelastic contribution to the spin-wave spectrum. An ap-

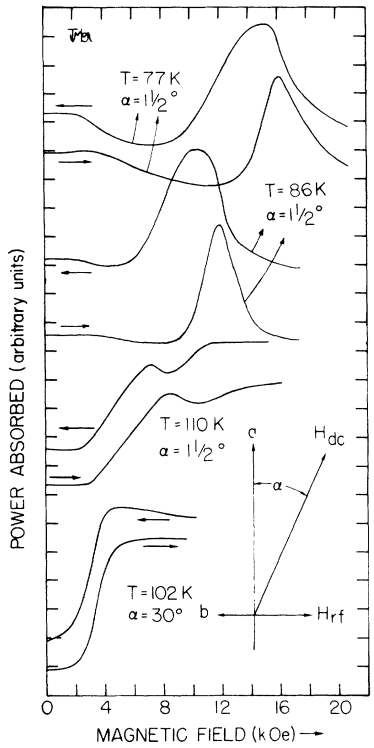

FIG. 2. Tracings of typical power absorption versus external-applied-field curves. The experimental configuration is shown in the diagram.

plied magnetic field cannot lower the spin-wave energy gap to our experimental frequency in this model.

In a ferromagnetic resonance experiment the largest value of q excited is $q \cong 1/\delta$, where δ is the microwave skin depth. In such an experiment, Vigren and Liu[7] show that the states of $q < 1/\delta$ behave essentially as $q = 0$ free-lattice states. If the experimental microwave frequency is low enough, only the $q = 0$ free-lattice mode should be observable because the applied field along $\langle 11\bar{2}0 \rangle$ lowers the energy of the $q = 0$ free-lattice mode to that of our microwave energy, while the $q = 0^+$ ($1/\delta \lesssim q \leq 2\pi/c$, where c is the $\langle 0001 \rangle$ lattice spacing) frozen-lattice mode remains well above our experimental frequency.

Two disk-shaped Tb samples with the c axis normal were used in this experiment. One of them was the same as the thinner crystal reported in previous work,[8] except that its dimensions were changed to 5.9 mm diam and 0.3 mm thickness. This sample was mounted as in Ref. 7 and was used only for the data taken at $T = 143$ and 161 K. It was the thinner of the two samples made, and so was least subject to demagnetizing effects affecting domain alignment. (The resonance field depends on the domain alignment field at $T = 143$ and 161 K.) Data at all other temperatures were taken on a sample having 9 mm diam and 1.2 mm thickness cut from the same large crystal as the first sample. The external magnetic field H_{dc} was applied in the plane of the disk-shaped sample, and the sample was aligned with a b axis (easy magnetic direction) along the microwave magnetic field H_{rf}.

The data were taken by measuring direct absorption as a function of temperature and H_{dc} (where H_{dc} could be oriented at any angle with respect to H_{rf}). The closed triangles in Fig. 1 are in good agreement with the free-lattice model of Cooper[1] calculated by Vigren and Liu[2] using the experimental twofold and sixfold anisotropy constants measured by Rhyne and Clark.[9] On the basis of these data, we believe that the rise in resonance field with decreasing temperature observed by Bagguley and Liesegang[10] in Tb at 10 GHz for temperatures above 140 K was due to a combination of off-resonance-absorption and domain-alignment effects, and was not an indication of free-lattice-model behavior. The frozen-lattice-model interpretation of the data of Wagner and Stanford[8] at 10 GHz for temperatures above 175 K does not contradict the free-lattice-model interpretation of Vigren and Liu since the two models give similar predictions for $T > 140$ K.

In this Letter we are concerned with the low-temperature absorption peaks ($T < 110$ K) observed when H_{dc} is aligned about 1° from the a axis. We attribute these peaks to on-resonance absorption in the free-lattice model for the following reasons (some of which were discussed in more detail earlier): (1) The amplitudes are large and the linewidths are comparatively narrow, indicating on-resonance absorption. (2) The temperature dependence of the peak is that of the free-lattice model, which departs significantly from that of the frozen-lattice model for $T < 140$ K. (3) Anomalies in the magnetostriction[11] and magnetoresistance[12] of the heavy rare-earth metals (i.e., Tb, Dy, Ho, etc.) are associated with domain-alignment and magnetic phase transitions. Since our peaks in power absorption for $T < 140$ K occur exclusively in the ferromagnetic phase of Tb above the domain-alignment field, they should not be due to changes in background conductivity or magnetostriction. (4) The peak at $T = 86$ K (see Fig. 2) has a $\sin^2\varphi$ amplitude dependence (where φ is the angle between

H_{rf} and H_{dc}) as expected for a spin-wave resonance.

At $T = 110$ K a second absorption begins to occur at higher fields (see Fig. 2). The second absorption is very broad and occurs also at lower temperatures when H_{dc} is rotated several degrees from the a axis. Present theory offers no satisfactory explanation for the absorption peaks observed for H_{dc} aligned from 3° to 10° from the a axis in the basal plane of the sample when $T < 140$ K, which will be discussed in a later publication.

A second discrepancy between theory and experiment is that no absorption peak is observed when H_{dc} is aligned exactly along the a axis, contrary to the prediction of Cooper's free-lattice model.[1] Our experimental results reveal that peaks with the temperature dependence predicted by Cooper for the free-lattice model actually occur for H_{dc} from 1° to 2° off the a axis (see Fig. 1).

In conclusion, our results at 24 GHz experimentally confirm free-lattice–model behavior in Tb at low microwave frequencies where the $q = 0$ mode can be excited as predicted by Vigren and Liu, furnishing a satisfying resolution to the previously paradoxical ferromagnetic resonance data on terbium.

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Quadrupole Coupling Constants of Fe^{3+} in Yttrium Iron Garnet*

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The recently determined value of the quadrupole moment of Fe^{57m} has been shown to explain consistently the observed quadrupole coupling constants associated with two nonequivalent Fe sites in yttrium iron garnet.

In a previous publication[1] the molecular-orbital method for a complex imbedded in a lattice has been shown to resolve the ferrous-ferric anomaly in the quadrupole moment of Fe^{57m} by considering the cases of Fe_2O_3 and $Al_2O_3:Fe^{3+}$. It has also been established that the quadrupole moment of the iron nucleus is 0.18 b within 10% accuracy. However, it is still not clear whether this value of the quadrupole moment can also explain the observed quadrupole coupling constants in a variety of systems without fitting[2,3] with parameters such as polarizability and antishielding