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Relaxation of Gallium Nuclei in Ga^{3+}-Doped YIG*

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We report a study of the NMR of gallium impurities in YIG. In this magnetic system a nonmagnetic impurity is added to a ferrimagnetic host where the observed nucleus is located at the magnetic defect site. This nucleus is coupled to the surrounding magnetic icns by a transferred hyperfine interaction, similar in form to the Heisenberg interaction, giving rise to local impurity states. These local states have been of much theoretical interest in recent years. Spin echo techniques were used to measure the relaxation behavior of ^{71}Ga and ^{69}Ga in single-domain samples of $Y_3Fe_{5-x}Ga_xO_{12}$ for x between 0.05 and 1.0. The observed linewidth at $x=0.25$ shows a strong inhomogeneous broadening due mainly to the interaction between impurity sites. T_1 is very sensitive to concentration, ranging from 1 sec for $x=0.25$ to about 10 msec for $x=1.0$ and varies as $1/T$ for T between 1.9° and 4.2°K. In contrast T_2^{-1} varies almost linearly with concentration and is temperature independent.

THE NMR of gallium impurities in YIG, first reported by Streever and Uriano,[1] represents an interesting example of a *local mode* resonance. The nonmagnetic gallium impurity replaces a ferric ion on a crystallographic d site creating a defect in the ordered magnetic structure. However, the nuclear moments of the abundant ^{69}Ga and ^{71}Ga nuclei are coupled to the surrounding magnetic ions by a transferred hyperfine interaction which is presumably of the same form, i.e., $A\mathbf{I}\cdot\mathbf{S}$, as the Heisenberg interaction between the ions of the host lattice. Thus, from a formal point of view, the gallium nuclei represent spin impurities coupled to the surrounding spins by a greatly reduced effective exchange interaction. The theoretical description of this situation is quite distinct from that of more common cases of NMR in ordered magnetic systems since here the nucleus forms an intrinsic part of the local defect.

The impurity states of a magnetic impurity in a Heisenberg ferromagnet have been treated theoretically in considerable detail[2–5] and these considerations may be applied, with due circumspection, to this problem.

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[1] R. L. Streever and G. A. Uriano, Phys. Rev. 139, A305 (1965).

[2] T. Wolfram and J. Calloway, Phys. Rev. 130, 2207 (1963).

[3] Y. A. Izyumov, Advan. Phys. 14, 569 (1965).

[4] Y. A. Izyumov and M. V. Medvedev, Soviet Phys.—JETP 21, 1155 (1965).

[5] H. Ishii, J. Kanamori, and T. Nakamura, Progr. Theoret. Phys. (Kyoto) 33, 795 (1965).

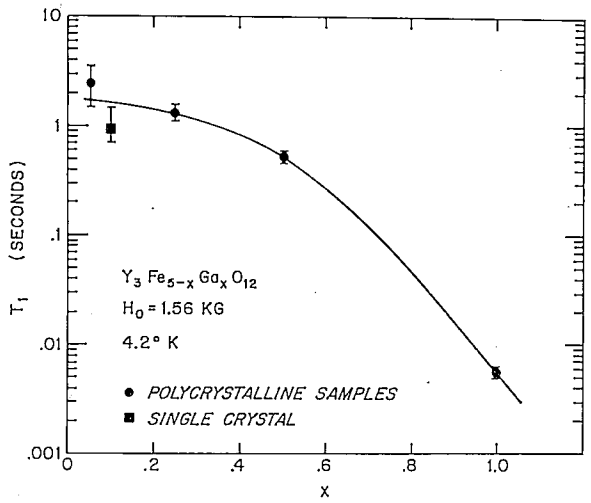

FIG. 1. Semilog plot of longitudinal relaxation time T_1 vs x for ^{71}Ga.

In particular, Izyumov and Medvedev[4] have derived expressions for the excitation spectrum for a magnetic impurity with a g factor different from that of the host ion. Their results are valid even though the g value and effective exchange constant are significantly different from those of the host, provided only that the range of the perturbation is short. In the limit of very small impurity concentrations and for a finite applied magnetic field, each gallium ion contributes one mode to a localized impurity level or state lying well below the spin wave band of the undisturbed crystal. From a more conventional view point, this local mode represents, of course, the NMR mode at a frequency $A\langle S\rangle/\hbar$. It is our view that recasting this NMR problem in the language of impurity states is not merely a semantic exercise, but rather the means of systematizing a difficult problem. For example, Izyumov and Medvedev[4] have given susceptibility expressions for the response of the local mode to an oscillatory magnetic field. The character of this response is sensitive to the disposition of the local mode with respect to the spin wave band and gives a direct picture of the enhancement effects of NMR in this material.

The local mode picture is perhaps most useful in the discussion of the line breadth and relaxation characteristics of the NMR. We report here some observations of these characteristics. Spin-echo experiments have been performed in the helium temperature range on both mono- and polycrystalline samples of $Y_3Fe_{5-x}Ga_xO_{12}$. Perhaps the most remarkable characteristic of these observations is the extreme breadth of the resonance lines in single crystals and for small Ga concentrations. For example, a sample with $x=0.25$ gives for the magnetization in the [111] direction a width of 5 MHz for the ^{71}Ga line at 26 MHz. The quadrupole interaction can, at most, account for 1.5 MHz of this width.[6] In contrast the homogeneous width, as measured by the coherence time T_2, is only about 12 Hz. The peculiar shape of the observed echo is a further indication that the line is inhomogeneously broadened.[7] We attribute the extreme breadth of the line to the strong interaction between localized impurity states. Lifshitz[8] has discussed the broadening of the impurity mode as the interaction between impurities is taken into account. This broadening is not associated with the reduction in the lifetime of the state but rather a broadening of the spectral distribution of the impurity states. The observed lineshape for these single domain samples is similar to that observed in powders by Streever and Uriano[1] who found that the width of the Ga line has only a slight dependence on Ga concentration for $x>0.25$. In comparison, the ^{57}Fe line of the host ion is found to be much narrower in width for a given Ga concentration and to increase steadily as the Ga concentration increases. We conclude that the localized states are more sensitive to the range of the interaction between impurities and may show significant narrowing at concentrations smaller than $x=0.25$. Unfortunately, because of signal to noise problems we have not been able to measure the linewidth at smaller concentrations.

The longitudinal relaxation T_1 was measured by observing the stimulated echo following a three-pulse sequence. The echo decay was not exponential, although the initial fall to half the initial height exhibited a constant rate of decrease. When the external field is zero or very small, the impurity modes are degenerate with the spin waves and domain wall excitations. In this situation the local mode becomes a virtual state of finite lifetime. Thus, as might be expected the observed T_1 is quite short when domain walls are present and increases rapidly as the sample is magnetized. Some experimental data is shown in Fig. 1 for magnetically saturated samples. It is to be noted that T_1 is extremely concentration dependent. In addition T_1 is found to vary as T^{-1} between 1.9° and 4.2°K and to increase by a factor of two as the drive frequency is changed from 27 to 19 MHz. These results suggest that the dominant process is a direct one in which the nuclei relax through a phonon modulation of the interaction between impurities. For the direct process T_1^{-1} varies directly as the temperature and the square of the energy separation as observed.

Measurements of the transverse relaxation time show that T_2 is independent of temperature between 1.9° and 4.2°K. T_2 decreases linearly with Ga concentration, decreasing from 16 msec at $x=0.25$ to 4.5 msec at $x=1.0$. This behavior for T_2 is similar to that observed in most magnetic materials.

[6] R. H. Tancrell, Bull. Am. Phys. Soc. 10, 317 (1965).
[7] W. B. Mims, Rev. Sci. Instr. 36, 1472 (1965).
[8] I. M. Lifshitz, Advan. Phys. 13, 485 (1964).