Temperature Dependence of the Hypersonic Absorption in MgO Containing Fe^{++}†

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The ultrasonic absorption at 3000 Mc/sec was measured in MgO with Fe^{++} impurities as a function of temperature. In contrast to the behavior of other dielectric crystals, there is in MgO a pronounced absorption maximum at about 17°K. The height of this maximum—not its position—is very sensitive to external magnetic fields. In this paper it is shown that this anomalous maximum is caused by the Fe^{++} impurities and probably arises from an ultrasonic saturation of the spin system for temperatures below 17°K.

INTRODUCTION

In a previous paper,[1] we have investigated the absorption of 3 kMc/sec ultrasonic waves in Al_2O_3 and MgO and, in particular, the temperature dependence of the absorption. In sapphire (Al_2O_3), the absorption was found to increase with about the fourth power of the temperature, which is as expected if only thermal phonons are responsible for the ultrasonic absorption.

The same T^4 dependence is found in MgO, but *only* if a sufficiently large magnetic field ($H \geqq 4000$ G) is applied at all temperatures (see dashed line in Fig. 1). After removing the magnetic field the absorption increases considerably, exhibiting a maximum at about 17°K as shown in Fig. 1. This anomaly occurs for both longitudinal and transverse waves in MgO.

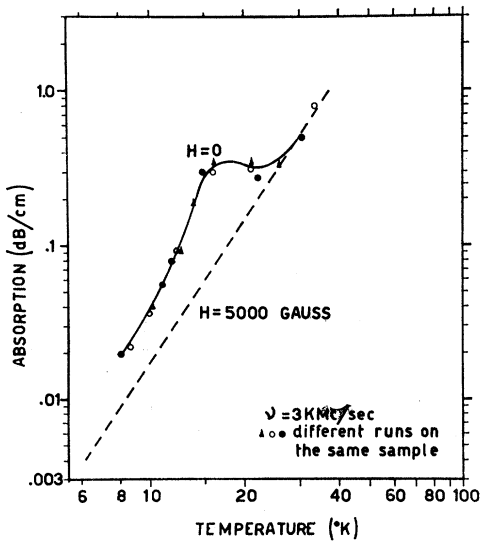

FIG. 1. Attenuation of longitudinal ultrasonic waves at 3 kMc/sec in MgO+Fe^{++} as a function of temperature. The dashed line gives the absorption in a magnetic field of about 4500 G; as shown in Ref. 1, this absorption is due to the scattering of the sound waves by thermal phonons. The full line gives the absorption in zero magnetic field.

In this paper we report experiments on the variation of this extra absorption with magnetic field, demonstrating that Fe^{++} impurities are responsible for the extra absorption. Finally, we discuss the temperature dependence of this extra ultrasonic absorption which, in principle, should occur in most paramagnetic crystals.

Experiments and Results

The MgO samples used were single crystals (from R. Meller and Company, Providence, Rhode Island) of rectangular (4×4×40 mm) and of cylindrical (diameter 3 mm, length 25 mm) shape. The end faces were polished optically flat and parallel within a few seconds, the rod axis being parallel to the (100) crystal axis. Sound was excited and detected piezoelectrically by two thin x-cut quartz disks, with a fundamental frequency of 10 Mc/sec, attached to each side of the MgO rod. Nonaq was used as a bonding material.

The absorption was measured using a pulse-echo technique as in the earlier experiments.[1] The acoustic pulses of 0.5-μsec duration had a peak power of about 50 mW inside the MgO rod, and a repetition rate of a few hundred times per second. The dependence of the absorption on magnetic field and temperature, as described below, was calculated from the observed variation of the acoustic echoes with H and T, respectively.

In order to investigate the nature of the extra absorption in Fig. 1 occurring at low magnetic fields, we studied at a fixed temperature of $T=15$°K the way in which this extra "magnetic" absorption varied with magnetic field. The results are presented in Fig. 2, for various orientations of the magnetic field **H** with respect to the direction of the acoustic propagation vector **k**. The line with **H** perpendicular to **k** corresponds to a g value of 6.8, and the two lines observed with **H** at 45° to **k** represent g values of 6.8 and 3.4, respectively. These are the same g values as previously found[2,3] in MgO with Fe^{++} impurities, and they correspond to the $\Delta m=2$ and $\Delta m=1$ transitions of this paramagnetic center. Furthermore, in agreement with the expected angular dependence,[4,5] the transition $\Delta m=2$ is strongest

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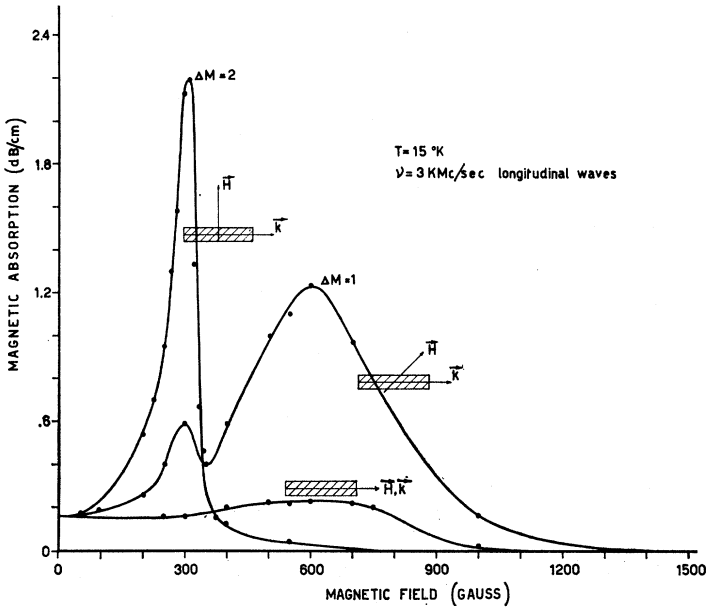

FIG. 2. Attenuation of ultrasonic waves in MgO+Fe^{++} as a function of the magnetic field. The absorption due to the scattering by the thermal phonons is subtracted. The 3 curves represent 3 different orientations of the magnetic field.

for **H** perpendicular, while $\Delta m=1$ processes absorb mainly for **H** at 45° to the **k** direction. An electron-spin-resonance (ESR) investigation of the same samples showed an Fe^{++} concentration of not more than 0.01%. It should be mentioned also that for $\Delta m=2$ both the ultrasonic and the ESR line are asymmetrical around the maximum.

It is interesting that some absorption in Fig. 2 exists already in zero magnetic field, giving rise to the peak in Fig. 1. This finite absorption at zero magnetic field is perhaps caused by the same strain-induced crystal fields which are responsible for the large linewidth of the $\Delta m=1$ transition in Fig. 2.

The absorption peak of Fig. 1 can be much enhanced by working either at 600 G with **H** at 45° to **k** or at 300 G with **H** perpendicular to the **k** direction. In Fig. 3 we have plotted the "magnetic" absorption only, i.e., the absorption measured at a field H, minus the pure lattice contribution observed at large fields ($H>5000$ G). Its temperature dependence is indicated for 3 cases: (a) $H=0$, (b) $H=600$ G, **H** under 45° to **k**, (c) $H=300$ G, **H** perpendicular to **k**. For case (c), the absorption was too strong to be measured very accurately, but its maximum occurred within ± 1°K at the same temperature as for case (a) and (b); thus, its position is not very sensitive to the orientation and magnitude of the magnetic field. At the high-temperature side of the maximum, as can best be seen for curve (b) taken at 600 G, the absorption decreases approximately as T^{-n}, where n is between 2 and 3, while below 17°K it increases with temperature much more rapidly than T^3. This characteristic temperature dependence of the "magnetic" absorption will be discussed below.

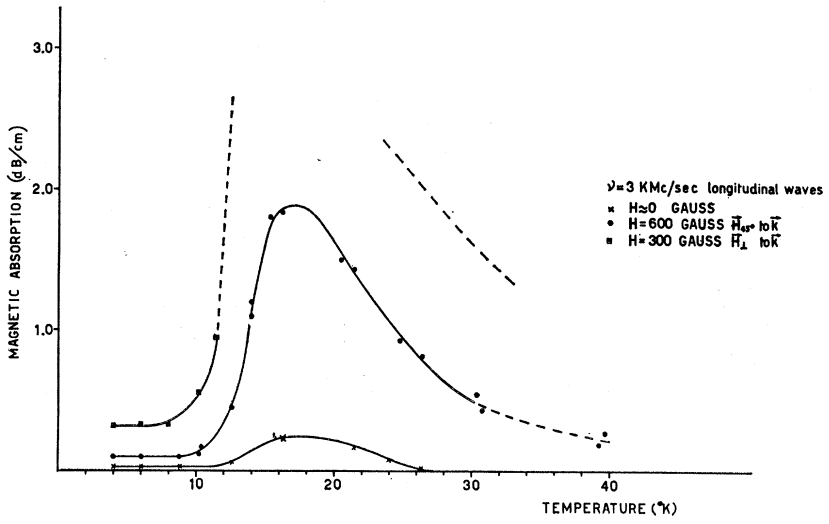

FIG. 3. Magnetic absorption of ultrasonic waves in MgO+Fe^{++} as a function of the temperature for different values of the external magnetic field.

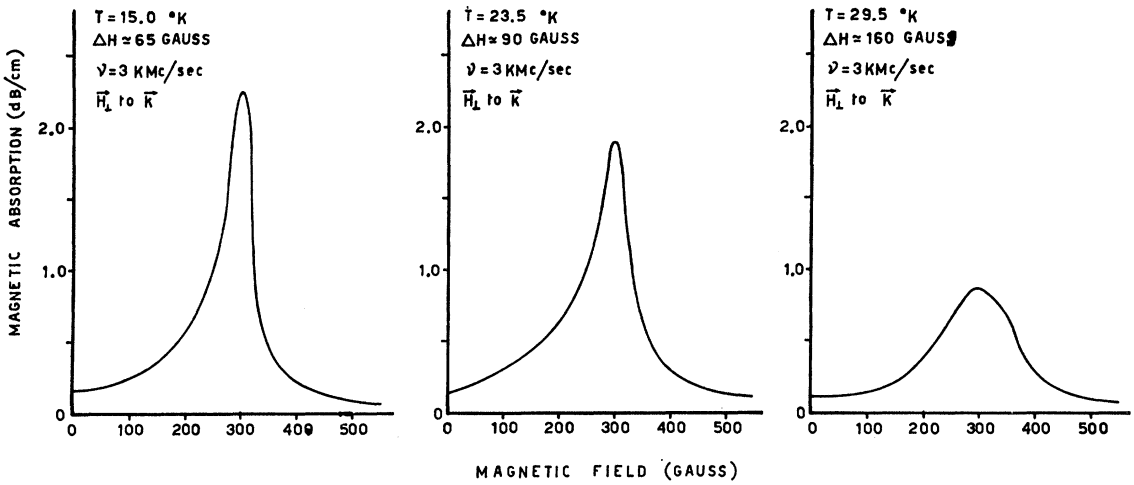

FIG. 4. Ultrasonic absorption by $\Delta m=2$ transitions at different temperatures.

Both longitudinal and transverse waves showed the same qualitative behavior; however, the data reported here were taken with longitudinal waves.

DISCUSSION

The ultrasonic absorption caused by N paramagnetic ions, having two energy levels of separation $h\nu$, with a linewidth $\delta\nu$ is

$$\alpha \propto \Delta N/\delta\nu, \tag{1}$$

where ΔN is the excess population in the lower state, which in thermal equilibrium at a temperature $T \gg h\nu/k$ is

$$\Delta N = (\tfrac{1}{2}N)(h\nu/kT). \tag{2}$$

The linewidth $\delta\nu$ also depends on temperature: In Fig. 4, the ultrasonic absorption due to $\Delta m=2$ transitions is plotted for 3 temperatures between 15° and 30°K. We note from this that the linewidth $\delta\nu$ varies with temperature between T and T^2, probably due to spin-lattice relaxation. Similiarly, the linewidth for the $\Delta m=1$ transitions probably also increases with temperature. According to Eqs. (1) and (2), it seems therefore reasonable that the absorption should decrease with temperature more rapidly than $(1/T)$, as observed above 17°K (see Fig. 3). The exact temperature dependence may, of course, be somewhat different for the two transitions.

In the previous argument we have neglected ultrasonic saturation of the spin system which would decrease ΔN in Eq. (1) below the thermal value given in (2) and thereby reduce the ultrasonic absorption. This is expected to occur at high acoustic power levels, or, if on cooling, the spin-lattice relaxation time becomes too long to keep the spin system at the lattice temperature. It seems likely that the rapid decrease of the absorption below 17°K in Fig. 3 is caused by a rapid increase of the spin-lattice relaxation time below 17°K. If this picture is correct, the temperature at which the maximum of the absorption in Fig. 3 occurs should be sensitive to the acoustic power level used. Corresponding experiments are now in progress.

At still lower temperatures, $T<10$°K, when the saturation of the spin system is nearly complete, the acoustic power P absorbed depends on the spin-lattice relaxation time T_1:

$$P \propto (\Delta N \times h\nu)/T_1{}^n,$$

where $n=1$ refers to homogeneously broadened lines and $n=\frac{1}{2}$ to inhomogeneous lines.[5,6] Noting Eq. (2) and assuming that T_1 at low temperatures varies as T^{-1}, as expected for "direct processes," the power absorbed should be independent of the temperature for homogeneous lines as evident in Fig. 3 below 10°K. For inhomogeneous lines, the absorption may be expected to show a minimum below 10°K, increasing again (as $T^{-1/2}$) at still lower temperatures.

It should be mentioned, however, that the spin saturation at these low temperatures apparently causes an irregular change of the relative size of the echoes, absent at high temperatures, which is not yet well understood and makes the measurements below 10°K less accurate.

ACKNOWLEDGMENTS

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