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SQUID ACOUSTOMAGNETIC PARAMAGNETIC RELAXATION STUDIES OF MgO:Fe$^{2+}$

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Abstract - A Superconducting Quantum Interferometer Device (SQUID) which is part of a SQUID acoustomagnetic spectrometer has measured a magnetization change in MgO:Fe$^{2+}$ which corresponds to a non-resonant acoustic phonon-electron spin coupling to the paramagnetic spin system.

I. INTRODUCTION

With the use of a high sensitivity SQUID (Superconducting Quantum Interference Device) acoustomagnetic spectrometer, it has been possible to observe at 4.2 K the non-resonant ($\Delta m = 0$) absorption of acoustic energy coupled to the dilute paramagnetic ion Fe$^{2+}$ in MgO as a change in magnetization ($\Delta M_z$) along an external magnetic field in the $\hat{z}$ direction. An earlier investigation [1] has also investigated the non-resonant absorption in MgO:Fe$^{2+}$ using ultrasonic attenuation and dispersion measurements. The ultrasonic frequencies chosen in both experiments are much less than the resonant frequencies for the paramagnetic spin system in the applied magnetic fields but are comparable to spin-lattice relaxation rates. The non-resonant acoustomagnetic experiments are very similar to Paramagnetic Relaxation experiments except that the time varying external magnetic field along the $\hat{z}$ direction in a Paramagnetic Relaxation experiment is replaced by a time varying effective magnetic field due to the spin-phonon interaction.

In MgO:Fe$^{2+}$, the lowest lying energy state is a triplet $100$ cm$^{-1}$ below the next highest energy level [3]. At magnetic fields less than 1 T, the Zeeman splittings of the triplet state are less than 1 cm$^{-1}$ and the Fe$^{2+}$ impurity can be described by an effective spin of $S = 1$. In an external magnetic field, the spin energy levels will be described by the quantum numbers $m = +1, 0,$ and $-1$ and the equilibrium population of electronic spins in these states is described by the Boltzman distribution for the magnetic field $H$ and the lattice temperature $T$. If a spin-phonon coupling is present giving rise to an effective field in the direction of $H$, the energy levels will vary in spacing at the frequency of the acoustic wave. If the frequency of the acoustic wave is much less than the relaxation rate $\Gamma$, relaxation occurs within the period of the acoustic wave and an isothermal situation occurs with no net absorption of acoustic energy and no magnetization change. If the frequency of the acoustic wave is much greater than the relaxation rate $\Gamma$, then an adiabatic situation occurs and again there is no net magnetization change or acoustic absorption. Using the theory of Fedders [4], Yuhas et al. [11] have shown when $\omega \approx \Gamma$ that there is a net acoustic absorption for longitudinal acoustic waves propagating in a [100] direction of this cubic crystal given by

$$\Delta \alpha = \frac{NG_{11}^2}{16k_B T \rho \omega^3} \frac{(3\cos^2 \theta - 1)^2 \omega^2 \Gamma}{\omega^2 + \Gamma^2}$$

(1)

where $N$ is the number of Fe$^{2+}$ cm$^{-3}$, $G_{11}$ the magneto-elastic coupling constant, $k_B$ the Boltzman constant, $\rho$ the mass density, $\nu$ the phase velocity of the acoustic waves, and $\theta$ the angle in the (010) plane between the direction of propagation and the direction of the magnetic field. The related spin-phonon transition probability is

$$W_{sp} = \frac{3\pi^2}{4} \frac{G_{11}^2 \epsilon^2}{h^2} \frac{(3\cos^2 \theta - 1)^2 \Gamma}{\omega^2 + \Gamma^2}$$

(2)

where $\epsilon$ is the elastic strain amplitude. At 4.2K, the dominant contribution to the relaxation rate $\Gamma$ is the direct process. In this case [1]...
where $\Gamma = \Gamma_0 + CH^2T$  

(3)

where $\Gamma_0$ and $C$ are field and temperature independent.

II. EXPERIMENTAL

From previous experimentation with the SQUID acoustomagnetic spectrometer, $\Delta M_z$ have been found for direct nuclear spin-phonon coupling \([5,6]\) as well as for the mechanical resonances within the fundamental and odd harmonics of the transducer responses, transmission line responses, sample cavity resonances, and whenever the acoustic power in the sample is changed. We have measured the radial distribution of acoustic energy for acoustic standing longitudinal waves in a crystal with flat and parallel faces whose dimensions are larger than the piezoelectric transducer radiating area ($\pi R^2$). The acoustic energy falls rapidly from a maximum value near the center to approximately 25% of the maximum value at $R$ and is near zero at $1.3R$. When this distribution of acoustic energy is present, it raises the temperature of the sample within the volume of the acoustic energy distribution, where $\Delta T$ is the spatial average of the temperature increase above the sample boundary and the surrounding sample holder.

In MgO, the time domain response when the acoustic frequency is changed from the center of a mechanical resonance to an off-resonance position in a $\mu$-sec, the decay of the of $\Delta M_z$ has a time constant of seconds. This long decay suggests that mechanism by which $\Delta M_z$ decays is due to thermal conduction of acoustic power from the acoustic radial distribution discussed above radially outward to the sample boundary and the sample holder. This "indirect coupling" magnetization change is related to $\Delta T$ through Curie's Law

$$\Delta M_{z_{ic}} = -\chi_0 H \Delta T / T$$

(4)

where $\chi_0$ is the paramagnetic susceptibility.

If $P$ is the acoustic power supplied to the sample when the acoustic frequency is adjusted to a mechanical resonance, then steady state relationship between incident power and power transferred by thermal conduction radially is

$$P f_2 f_1 = K \Delta T$$

(5)

where $f_2$ is the transducer response at the fundamental or odd harmonic,

$$f_1 = \frac{\alpha a}{(\alpha a)^2 + (\omega - \omega_0)^2}$$

(6)

$K$ is a constant including the thermal conductivity and geometrical factors related to the thermal conduction, $\alpha$ is the total acoustic attenuation at the frequency $\omega$ and field $H$

$$\alpha = \alpha_b + \Delta \alpha$$

(7)

$a$ is twice the sample length, $\omega_0$ in the position of the $n$th mechanical resonance in the transducer response, and $\alpha_b$ is the "background" attenuation. The insertion of (5) into (4) gives

$$\Delta M_{z_{ic}} = \frac{-\chi_0 H f_2 f_1}{KT}$$

(8)

When there is a direct spin-phonon coupling, the magnetization change, $\Delta M_z$, from Abragam \([7]\) is

$$\Delta M_z = -\frac{W_{sp} T_1 \chi_0 H}{1 + W_{sp} T_1}$$

(9)

If a mechanical resonance is observed at the position of a direct spin-phonon coupling resonance, then the observed $\Delta M_z$ in the above equation is multiplied by $f_1 f_2$.

In the experiment described in this paper, what is actually measured is the fractional change at the center of a mechanical resonance when the angle $\theta = 0$ and for $W_{sp} T_1 << 1$ for two different, but close magnetic field values which is given by

$$F = -\Delta \Gamma \frac{B \omega^3 (\omega^2 - \Gamma^2) + A (2 \alpha_b \Gamma + B \omega^3) + \Delta \Gamma (A \alpha_b - B \omega^3 \Gamma)}{[\alpha_b (\omega^2 + (\Gamma + \Delta \Gamma)^2) + B \omega^2 (\Gamma + \Delta \Gamma)] [(\omega^2 + \Gamma^2)]}$$

(10)

where
Experimentally, we measured $F$ at the center of a mechanical resonances at the third through fifteenth transducer harmonics at the magnetic fields of 194.4, 402.3, 817.0, 2085.9, 4627.1, 6539.0, and 7670.1 G. Widths of mechanical resonances were also determined to provide a measure of $\alpha_h$.

Values of $\Delta M_z$ are proportional to $H$ and $F=0$ up to $H=817.0 G$. For $H>817.0$ and increasing $F$ becomes increasingly more negative indicating the presence of an additional absorption of acoustic energy in the sample which causes a decrease in the magnitude of $\Delta M_z$. With the use of the Yuhas determined values [8] for $C$ and $\Gamma_0$, the average of the two measured values of $G_{11}$ [9], and the measured value of the thermal conductivity in MgO at 4.2 K [10], we have determined a computed value of $F$.

Table I -- Comparison of the experimental, $F_{data}$, and computed $F_{model}$, fractional changes in magnetization. The precision of $F_{data}$ is $\pm 0.01$ and the uncertainty in $F_{model}$ is $\pm 0.02$.

<table>
<thead>
<tr>
<th>H (G)</th>
<th>$\Delta H$ (G)</th>
<th>$F_{data}$</th>
<th>$F_{model}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>815</td>
<td>1269</td>
<td>0.008</td>
<td>0.016</td>
</tr>
<tr>
<td>2084</td>
<td>1540</td>
<td>0.034</td>
<td>0.029</td>
</tr>
<tr>
<td>4623</td>
<td>1905</td>
<td>0.069</td>
<td>0.066</td>
</tr>
<tr>
<td>6538</td>
<td>1142</td>
<td>0.022</td>
<td>0.056</td>
</tr>
</tbody>
</table>

CONCLUSION

We have reported observation of acoustic properties, such as mechanical resonances, in MgO as a $\Delta M_z$ due to the heating of the sample and therefore the paramagnetic Fe$^{2+}$ ions by the acoustic wave. A physical model has been developed which allows calculation of the fractional change $F$ from acoustic coupling to $\Delta M_z$ from both thermally heating the lattice and a direct spin-phonon coupling. In Table I, the computed values of $F$ generally agree in magnitude and sign with the experimental values which (i) supports the two couplings to $\Delta M_z$ and (ii) confirms the direct relaxation rate in MgO:Fe$^{2+}$ as having a constant term ($\Gamma_0$) and a second term ($C$) multiplying $H^2$ as shown in (3), and (iii) is in general agreement with the values of $\Gamma_0$ and $C$ measured by Yuhas [8].

REFERENCES