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THE EFFECT OF LATTICE STRAINS ON THE ACOUSTIC RELAXATION LOSSES IN
DIELECTRIC CRYSTALS DUE TO MAGNETIC IONS

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Abstract: Recently information on the lattice strains present at Fe$^{2+}$ sites
in KMgF$_3$ has been obtained from electron spin resonance linewidth data.
The use of this data in an empirical model previously used with success on
Fe$^{2+}$:MgO shows that this model also gives an excellent description of the
acoustic relaxation losses in Fe$^{2+}$:KMgF$_3$. In order to place this empirical
model on a more sound theoretical footing a theory of relaxation losses in
multi-level systems based on the rate equations for the population of each
level has been studied. The empirical model results only if simplifying
assumptions are made and the implications of these are described.

Acoustic relaxation losses occur when the strain of an acoustic wave
modulates the energy levels of a system such as that consisting of a number of
magnetic ions embedded in a dielectric lattice. The dynamic repopulation of the
levels of such a system results in a loss to the acoustic wave. For a two-level
system the form of the loss $\alpha$ is:

$$\alpha = \text{constant} \cdot \frac{G^2}{v^3 kT} \cdot \frac{\omega^2 \tau}{1 + \omega^2 \tau^2}$$

where $\omega$ is the acoustic angular frequency, $v$ is the acoustic velocity, $G$ is a
coupling factor related to the energy level shift per unit strain and $\tau(T)$ is the
relaxation time. It may be shown that the expression for a n-level system reduces
to the sum of $n-1$ such expressions above. Random lattice strains influence the
magnitude and acoustic mode dependence of $\alpha$ via their effect on $G$ and on $\tau$, and
recently a successful treatment of the effect of strains on the relaxation loss
due to the Jahn-Teller ion Ni$^{3+}$ in the Al$_2$O$_3$ lattice has been given.

It is interesting to study Fe$^{2+}$ in cubic environments such as those provided
by MgO or KMgF$_3$, since quite different acoustic relaxation behaviour is found in
the two systems. In MgO the acoustic loss of $T_2$ symmetry modes is much
weaker than the predictions of a strain free model would suggest, while in KMgF$_3$
it is the E-symmetry modes which are experimentally less attenuated than on such
a model.

An empirical model due to King and Monk treats the effect of lattice
strains by considering a single two-level expression for the three-level ground
state but with a coupling factor $G^2 = (G_1 - G_2)^2 + (G_2 - G_3)^2 + (G_3 - G_1)^2$ where
$G_1, G_2, G_3$ are the shifts in the energies of the three ground state levels due to a unit acoustic strain in the presence of a much larger static lattice strain. Factors which represent reductions in the unstrained values of $G^2$ were computed as a function of the ratio of a mean compressional to a mean shear lattice strain, using the known form of the Hamiltonian.

The anisotropy of the electron paramagnetic resonance linewidth gives a measure of this ratio and for Fe$^{2+}$:MgO use of the empirical model then gives excellent predictions of the acoustic losses experimentally observed. Recently Grimshaw has measured the anisotropy of the Fe$^{2+}$ electron paramagnetic resonance linewidths for KMgF$_3$. The anisotropy is very different from that in MgO, but again use of the empirical model gives very good estimates of the acoustic relaxation losses of the various acoustic modes.

In order to understand why this simple empirical model is so successful a treatment of the acoustic relaxation loss in a n-level system has been developed and then applied to the Fe$^{2+}$ ion. This treatment uses the driven rate equations for the populations of the n-levels. The energies of the n-levels are displaced by the acoustic wave, the dynamic repopulations may be calculated from the rate equations, and the overall loss derived. This involves solving the equations:

$$ j \omega N_i \sum_s \frac{(G_i^s - G_1^s)}{kT} \frac{N_s^0}{s} = j \omega n_i + \sum_r n_r P_{ri} - n_i \sum_r P_{ir} $$

for the $n_i$ and then obtaining the attenuation from the expression:

$$ \alpha = \frac{8.686}{2\beta \nu} \sum_i \text{Im}(n_i) G_i^1 $$

In these equations $N_i^0$ is the thermal equilibrium population of the $i^{th}$ level, $n_i$ is the dynamic deviation from the equilibrium which would be obtained if the levels were frozen in energy at a particular instant in time, and the $P_{ij}$ are the probabilities of transitions between the $i^{th}$ and $j^{th}$ levels per unit time. The $G_i^1$ are linearly related to the $G_i^1$ of the empirical model. This treatment gives the usual expression for a two-level system and for a three-level system yields:

$$ \alpha \propto \frac{1}{(\omega^2 + \gamma_1^2)(\omega^2 + \gamma_2^2)} \left\{ \omega^2 [P_1(G_1^1 - G_3^1)^2 + P_2(G_1^1 - G_2^1)^2 + P_3(G_1^1 - G_2^2)^2] \
+ (P_1 P_2 + P_2 P_3 + P_3 P_1) \{P_1[(G_1^1 - G_3^1)^2 + (G_1^1 - G_2^1)^2] + P_2[(G_1^2 - G_2^1)^2 + (G_1^2 - G_3^1)^2] + P_3[(G_3^1 - G_1^1)^2 + (G_3^1 - G_2^1)^2] \} \right\} $$

where $P_1 = P_{23} = P_{32}$ etc. $\gamma_1$ and $\gamma_2$ are combinations of $P_i$. The above expression which can be decomposed into two, two-level expressions, has to be summed over all ions. Since each ion has a different strain each has in general a different set
of $G_1$ and $P_1$. If, however, we suppose that the $P_1$ are equal then the above expression reduces to the empirical expression and the two relaxation peaks coincide.

We note that experimentally the relaxation peak is observed in MgO and $\text{KMgF}_3$ in a region where an Orbach term involving the first group of excited states dominates the relaxation\(^2,3,4\). A single peak is observed in each case, the form of which gives a good prediction of the energy of the excited states ($\text{MgO}, 110 \text{ cm}^{-1}$, $\text{KMgF}_3, 96 \text{ cm}^{-1}$). This would not occur were it described by the sum of two different two-level expressions and the single peak is consistent with the $P_1$ being equal. Is it true then that all the $P_1$ are equal or are there another set of simplifying circumstances? Rough calculations suggest that although the $P_1$ will have the same temperature dependence they are not closely equal in magnitude.

We note that the rate equation method is equivalent to ignoring the off-diagonal elements in a density matrix formulation such as that by Isawa et al. The use of the rate equations assumes that $T_2$ relaxation times are much shorter than $T_1$ relaxation times. At the temperature at which the relaxation peaks are observed this approximation may not be strictly valid. The general expressions for a three-level system involving such terms are, however, very complex and difficult to handle. It should also be noted that current Jahn-Teller theories have so far failed to predict the equal coupling of $T_2$ and $E$ acoustic modes to the Fe$^{2+}$ ion.

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References

7. King, P. J. and Monk, D. J., to be published.