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PRESSURE DEPENDENCE OF THE ELECTRIC FIELD GRADIENT AT THE Al NUCLEUS IN GdAl₂

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Abstract. – High pressure NMR measurements have been used to show that the temperature dependence of the electric field gradient at the Al sites of GdAl₂ cannot be due lattice expansion. Any pseudoquadrupole contribution to the electric field gradient is also negligible from a comparison of GdAl₂ and HoAl₂ measurements.

1. Introduction

The cubic Laves compounds RA₁₂, where R is a rare earth, have their Al atoms arranged in tetrahedral formation. NMR studies of the quadrupole frequencies ν₂ at the Al sites, reflecting their electric field gradients, have been of particular interest because of the strong temperature dependence of ν₂ observed in the ferromagnetic phase of GdAl₂ by Degani and Kaplan [1]. At the Al sites whose electric field gradients are directed along the (111) easy direction this dependence may be represented in GdAl₂ as:

\[ P_{1} = \frac{1}{2} \nu_{Q}(111) = P_{1}(0) \left(1 + 4.84 \times 10^{-7} T^{3/2}\right) \]  \(\text{(1)}\)

where \(P_{1}\) is the quadrupole parameter and \(P_{1}(0)\) is its zero temperature value of 271 kHz. \(P_{1}\) is defined by the equation

\[ P_{1} = 3 e^{2} q Q / 4 I (2 I - 1) \hbar \]

where \(e_{Q}\) is the principal component of the electric field gradient, \(Q\) is the quadrupole movement of the of the nucleus and \(I\) is the nuclear spin). Since the magnetisation also has a \(T^{3/2}\) dependence within the range measured (up to 0.7\(T_{c}\)) the possibility of a magnetically induced contribution linear in \(M(T) / M(0)\) has been the frequent subject of speculation. Zevin and Kaplan [2] suggested its origin to be a magnon in a magnetoquadrupole interaction and their calculations gave a temperature dependence which agreed with that observed within a factor of two. Gehring and Walker [3], however, disagreed with their model, suggesting that the pseudoquadrupole interaction would actually be too small to give an observable contribution to the temperature dependence of \(P_{1}\).

Some authors [3, 4] have suggested that the temperature dependence is due to lattice expansion rather than a magnetic effect. However we now show from high pressure measurements that this cannot be the case, although lattice vibrations may provide a mechanism. In addition we have looked for evidence of a magnetically induced contribution to the electric field gradient in HoAl₂ which has a far lower Curie point \(T_{c}\) than GdAl₂ (~30 K as opposed to 176 K).

2. Experimental

All NMR measurements were taken on a computer controlled spin-echo spectrometer [5]. GdAl₂ readings were made on a single crystal and HoAl₂ readings on a powdered polycrystalline sample. Accurate measurements of the quadrupole frequencies were made from the oscillatory decay of the echo as the pulse separation is varied [6], and analysed using the technique of linear prediction by single value decomposition (LPSVD) which is more accurate than the usual fast Fourier transform method [7]. Pressure measurements were made at 4.2 K in a liquid filled cell pressurised at room temperature, and the pressure measured using a semiconductor transducer.

3. Results and discussion

The zero field \(^{27}\text{Al}\) NMR spectrum of GdAl₂ at 4.2 K is shown in figure 1. It consists of a mixture of domain and domain wall signals [4, 8]. The high frequency end of the spectrum consists of signals from Al atoms having their axially symmetric [111] axis along the direction of magnetisation in domains, and this is also where the highest \(\nu_{Q}\) value, equal to 2\(P_{1}\) should be observed. This value was obtained at a series of hydrostatic pressures up to 9 kbar leading to

\[ \frac{\partial \ln P_{1}}{\partial P_{T}} = + (14 \pm 2) \times 10^{-4} \text{ kbar}^{-1} \]  \(\text{(2)}\)

and hence, using a value of \(12.6 \times 10^{-4} \text{ kbar}^{-1}\) for the compressibility \(- \left(\frac{\partial \ln V}{\partial P_{T}}\right)\) [9],

\[ \frac{\partial \ln P_{1}}{\partial \ln V} = -1.1. \]  \(\text{(3)}\)
Lattice expansion would therefore decrease the electric field gradient with temperature, as opposed to the increase observed by Degani and Kaplan [1]. The effect of thermal expansion would also be an order of magnitude smaller than the observed temperature dependence of $P_l$. We conclude that lattice expansion cannot explain Degani and Kaplan’s results. However, the amplitude of lattice vibrations generally contributes more to the temperature dependence of electric field gradients than does thermal expansion and this may still provide an explanation.

It is interesting to reconsider the possibility of a magnetically induced pseudoquadrupole contribution to $P_l$. The angular dependence of $\nu_q$, as reflected in its change with NMR frequency, shows that there is no appreciable contribution to $P_l$ in any direction other than along the axially symmetric (111) axes in GdAl$_2$ irrespective of the direction of magnetisation at 4.2 K, and this also appears to be the case at 77 K [8]. According to Zevin and Kaplan’s model [2], the pseudoquadrupole contribution to the quadrupole parameter is given by an expression of the form

$$ P_{pq} = A_{pq} \left[ 1 - \frac{M(T)}{M(0)} \right] $$

$$ A_{pq} = \text{const} \times \frac{\omega_{Al}^2}{T_c} $$

(4)

where $\omega_{Al}$ represents the hyperfine frequency of the Al nuclei at 0 K and $T_c$ the Curie point of the material.

There should therefore be no pseudoquadrupole contribution to the electric field gradient at low temperature in contrast to the model of Gehring and Walker [3] which gives a maximum pseudoquadrupole contribution at 0 K. The result for GdAl$_2$ at 4.2 K is therefore consistent with Zevin and Kaplan’s model, but that at 77 K is unexpected.

The value of $A_{pq}$ for HoAl$_2$ is almost identical to the GdAl$_2$ value of 43 kHz, so a similarly sized pseudoquadrupole contribution should be observed. The HoAl$_2$ spectrum at 4.2 K is shown in figure 2, and consists of two distinct peaks. The value of $\nu_q$ on the higher frequency peak was found to be $520 \pm 5$ kHz at all temperatures up to 14 K, where $M(T) / M(0) = 0.85$, whereas an increase of 13 kHz would be expected from equation (4). It therefore appears that there can be no pseudoquadrupole interaction following the model of Zevin and Kaplan.

4. Conclusion

Pressure measurements have shown that lattice expansion cannot explain the temperature dependence of the electric field gradient in GdAl$_2$. Measurements on HoAl$_2$ have also shown that there cannot be an appreciable pseudoquadrupole contribution to the temperature dependence. A mechanism involving lattice vibrations has not been ruled out.