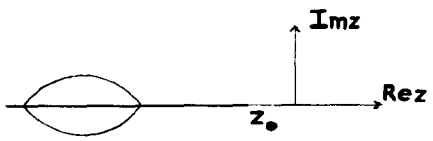

Fig. 1.

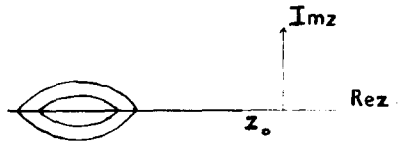

Fig. 2.

exactly three solutions to eq. (2) of largest real part for real z. One of these must be real and so (3) does not apply. This case corresponds to $c < 1$ and has been studied by Byckling [3] and Niemeyer [4]. Niemeyer has also studied the case $c = 1$ and states that the zeros lie on the real axis, but our rigorous results indicate otherwise.

The author would like to thank the Science Research Council for financial support and Professor O. Penrose for helpful discussions.

References

[1] C. N. Yang and T. D. Lee, Phys. Rev. 87 (1952) 404.
[2] O. Penrose and J. S. N. Elvey, J. Phys. A (1968) 661.
[3] E. Byckling, Phys. Rev. 140 A1165.
[4] Th. Niemeyer, Phys. Letters 29A (1969) 231.

* * * * *

SECOND AND FOURTH ORDER SPIN-LATTICE COEFFICIENTS FOR Gd^{3+} IN THORIA *

S. B. OSEROFF, R. CALVO ** and C. FAINSTEIN
Centro Atómico Bariloche, Instituto de Física "Dr. José A. Balseiro", Comisión Nacional de Energía Atómica, Universidad Nacional de Cuyo, San Carlos de Bariloche, Argentina

Received 7 July 1970.

We measured the EPR lineshifts for Gd^{3+} : ThO_2 under uniaxial stress. Values are given for the second and fourth order spin lattice strain coefficients.

The measurement of the shifts of the EPR lines of paramagnetic impurities in a crystal under uniaxial stress provides information about the spin-lattice interaction. The strength of this interaction is given by the spin-lattice coefficients whose values depend on the coupling between the lattice and the ions. Values for these coefficients have been obtained for S-state ions of the 3d group [1, 2] (Mn^{2+} and Fe^{3+}) and for the 4f group [3] (Gd^{3+} and Eu^{2+}), in cubic crystals. In these cases they can be defined from the spin-lattice Hamiltonian [3]:

$$H' = \sum_{n,i,\alpha,\xi} G_i^{(n,\xi)} O_{i,\alpha}^{(n,\xi)} \epsilon_{i,\alpha} \qquad (1)$$

where $\epsilon_{i,\alpha}$ is the linear combination of the components of the strain tensor transforming like the α-component of the i-irreducible representation of the group of symmetries of the ion; $O_{i,\alpha}^{(n,\xi)}$ is a linear combination of n-order Stevens' operators transforming accordingly, and the $G_i^{(n,\xi)}$ are the spin-lattice strain coefficients; ξ stands for the case where more than one n-order Stevens' operator transforms like $\Gamma_{i,\alpha}$.

For the $3d^5$ ($^6S_{5/2}$) ions, terms with $n = 2$ and $n = 4$ are allowed in eq. (1) but only $n = 2$ terms have been measured in uniaxial stress experiments [1, 2]. Terms with $n = 2$, $n = 4$, and $n = 6$ are allowed for the $4f^7$ ($^8S_{7/2}$) ions, and previous work in Eu^{2+} and Gd^{3+} in CaF_2 and CaO shows that $n = 4$ terms give an important contribution to the experimental shifts [3]. However, very small changes in the orientation of the sample when the stress is applied introduce spurious

* Research sponsored in part by the Consejo Nacional de Investigaciones Científicas y Técnicas, República Argentina under grant No. 2823/66.
** Member of the "Carrera del Investigador Científico", Consejo Nacional de Investigaciones Científicas y Técnicas, República Argentina.

Table 1
Second and fourth order spin-lattice strain coefficients for Gd^{3+} : ThO_2 in units of cm^{-1} measured at 295°K.

	$G_{1g}^{(n)}$	$G_{3g}^{(n)}$	$G_{5g}^{(n)}$
$n = 2$	-	$(-2.5 \pm 0.8) \times 10^{-2}$	$(-5.0 \pm 0.5) \times 10^{-2}$
$n = 4$	$(4.9 \pm 1.5) \times 10^{-4}$	$(-1.7 \pm 1.0) \times 10^{-4}$	$(4.9 \pm 1.5) \times 10^{-4}$

contributions to the shifts which are specially important in the measurement of the spin-lattice coefficients [3].

In this work we tried successfully to measure the fourth order coefficients of H' for Gd^{3+} in ThO_2. This system was chosen because its small EPR linewidths (about 1 gauss) allows precise measurements of the lineshifts. We used a conventional 35 GHz EPR Spectrometer with provisions to apply uniaxial stress on the sample. The stress system was carefully designed and built in order to minimize the changes of the position of the sample with stress ‡.

The samples were grown by C. B. Finch of Oak Ridge National Laboratory ‡‡; they were cut as rectangular prisms of about $1 \times 1 \times 1.5$ mm^3 oriented in the [110] direction, the error in orientation being smaller than 1°. Stresses up to 700 kg/cm^2 were applied along this direction and the magnetic field was rotated in the [110] plane. The shifts of the seven fine structure lines were measured at 295°K for $H \parallel [001]$, $H \parallel [1\bar{1}0]$ and $H \parallel [1\bar{1}1]$; from these data we obtained the values for the second and fourth order spin-lattice coefficients given in table 1.

Data taken for a fourth direction of the magnetic field was in agreement with the values of table 1.

The spin-lattice strain coefficients $G_{3g}^{(2)}$ and $G_{5g}^{(2)}$ are equivalent to G_{11} and G_{44} as defined by Feher [1] in a second order theory and measured for the iron group and rare earth group S-state ions. They correspond to the coupling of the ion with the electric field originated by tetragonal and trigonal deformations of the lattice. On addition, we are reporting values for the three fourth order spin-lattice coefficients $G_{1g}^{(4)}$, $G_{3g}^{(4)}$ and $G_{5g}^{(4)}$ which correspond to completely symmetrical (hydrostatic), tetragonal and trigonal deformations. The origin of the large errors given in table 1 can be understood by inspection of the formulas given in section III of ref. [3] ‡

Wybourne [4] performed the first detailed calculation of the crystal field splittings of Gd^{3+}. He obtained a total contribution to the ground state splitting which is twice the observed value and of the opposite sign. Recently Detrio [5] reported values of the spin-lattice coefficients calculated with a point charge model using the free ion wave functions given by Wybourne [4]. He found again an agreement in magnitude, but the wrong sign for these coefficients. Probably other mechanisms like these considered by Sharma et al. [6] for Mn^{2+} and the influence of covalency should be considered in future calculations. We think that our data on the fourth order in addition to the second order spin-lattice coefficients can be of help on this purpose.

It is interesting to report that preliminary data taken by us at 77°K shows an unexpected variation with temperature of the spin-lattice coefficients. Further data and discussion of the theory will be presented elsewhere in the near future.

Referenees

[1] E. R. Feher, Phys. Rev. 136 (1964) A145.
[2] R. Calvo, Z. Sroubek, R. S. Rubins and P. Zimmermann, Phys. Letters 27A (1968) 143.
[3] R. Calvo, R. A. Isaacson and Z. Sroubek, Phys. Rev. 177 (1969) 484.
[4] B. G. Wybourne, Phys. Rev. 148 (1966) 317.
[5] J. A. Detrio, Bull. Am. Phys. Soc., Series II, 15 (1970) 335.
[6] S. S. Sharma, T. P. Das and R. Orbach, Phys. Rev. 171 (1968) 378.

‡ We are very grateful to Mr. C. Lulich who built the stress head.
‡‡ We are indebted to Dr. M. M. Abraham for the loan of the sample.

‡ There is a misprint in eq. (14) of ref. [3]; the numerical coefficient that multiplies $C_{5g}^{(4)}$ should be -1/18 and not -1/8.

* * * * *