Letters to the Editor

Paramagnetic Resonance Spectra of Some Ions of the 3*d* and 4*f* Shells in Cubic Crystalline Fields*

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Paramagnetic resonance spectra are useful in determining the low-lying energy levels in crystals. The degeneracy and splittings of these levels are a function of the crystalline symmetry. Most of the paramagnetic ions of the iron group have been investigated in crystals like Tutton salts or alums, crystals which show deviations from cubic symmetry. Results for the rare earth group have been obtained using crystals of trigonal symmetry.[1] These deviations from cubic symmetry, in particular tetragonal and rhombic components of the crystalline symmetry, remove the orbital and spin degeneracies of the ground state.

We have investigated a number of paramagnetic ions in single crystals of cubic symmetry. A summary of the results is presented here. The detailed results of the spectra of the individual ions will be presented at a later date.

Ni^{2+}: Nickel was diffused into single crystals of MgO at 1100°C. The outer layers of the crystal were cleaved off in order to reduce the line width. In a cubic field, the $^3F_4$ ground state splits into two threefold-degenerate levels and one singlet, the latter being the lowest level. The threefold spin degeneracy is not removed in a cubic field even with spin-orbit interaction. Only one line was observed, with the following g-values: $g=2.225\pm0.005$, $T=290$°K; $g=2.227\pm0.002$, $T=70$°K; $g=2.234\pm0.002$, $T=4$°K. Assuming a reduced spin-orbit coupling constant $\lambda=-250$ cm^{-1},[2,3] and an average value of $g=2.23$, one finds the separation between the single and the next higher lying triplet to be 8700 cm^{-1}.

Mn^{2+}: Crystal MgO, concentration of Mn 0.01–0.001% by weight. The ground state is $^6S_{5/2}$ and the sixfold degenerate level is split into close-lying twofold and fourfold levels by higher order interactions. The nuclear spin of Mn^{55} of 5/2 splits each level into 6 components. In a pure cubic field the hyperfine structure splitting is larger than the fine structure splitting. The spectrum consists, therefore, of 6 groups of 5 lines of relative intensity 8:5:9:5:8. The separation of the fine structure lines is a function of the angle between the external magnetic field and the crystalline axes. The spectrum is described by $g=2.0014\pm0.0005$, $A=86.8\pm0.2$ gauss, a ground-state splitting of 59.8 gauss, line width of about 4 gauss. A very slight deviation from cubic symmetry (presumably rhombohedral) was found. The ground-state splitting is larger than that found in ZnS:Mn which is about 20 gauss.[4] This may be because of the reduced spin-orbit coupling in covalent ZnS (the hyperfine structure in ZnS is also reduced to 68 gauss), and because of stronger crystalline fields in MgO.

Eu^{2+}: Crystal $SrCl_2$ (fluorite structure), concentration of Eu$\sim$0.001%. As in the manganese spectrum, the $^8S_{7/2}$ state is lifted by higher order interactions of the combined cubic field and spin-orbit coupling and is split into twofold, fourfold, and twofold degenerate levels which are separated in the ratio of 3:5. The known value of 5/2 for the spin of the two isotopes of Eu gives a complicated spectrum of 94 lines. The spectrum can be represented by $g=1.995_0\pm0.001$, $A_{151}=34.5\pm0.3$ gauss, $A_{153}=15.5\pm0.3$ gauss, half-width about 4–5 gauss. The hyperfine structure, as in Mn, is caused by configurational interaction, and to a first approximation should be independent of the crystal host. The results should be compared with the spectrum found[5] in powdered SrS (NaCl structure): $g=1.991$, $A_{151}=32.61$ gauss. These small changes in the hyperfine structure constant are possibly due to differences in the amount of covalent bonding.

V^{2+}: Crystal MgO, concentration of V $\sim$0.0005% (determined spectroscopically). A cubic field splits the $^4F_{3/2}$ state similarly to the Ni, leaving a singlet as the lowest lying level. The spin quadruplet remains degenerate in a cubic field including spin-orbit interaction. The hyperfine structure due to the dominant isotope 51 with spin 7/2 splits the levels. Experimentally one finds that each hyperfine-structure line is flanked by 2 satellites with intensity ratios of 2:3:2.[6] The parameters are $g=1.9803\pm0.0005$, $A=80.3\pm0.2$ gauss, $T=290$°K, line width of about 3–4 gauss. The maximum separation between the two satellites is about 15 gauss. This splitting can possibly be ascribed to the small deviation from cubic symmetry that has been noticed for Mn and spin-spin interaction.[7] It is interesting to compare the ratio of the hyperfine-structure constants of Mn and V for similar environments. In the Tutton salt $(NH_4)_2ZnSO_4\cdot6H_2O$ this ratio is 1.06 ± 0.03,[8] while in MgO the corresponding ratio is 1.09 ± 0.01, the hyperfine splittings in the Tutton salts being about 10% larger.

We have observed the spectrum of Fe^{2+} in MgO ($g\sim3.428$, 6.90) and in ZnS ($g\sim2.26$). A detailed calculation will be presented in the future. Work is in progress on Cu^{2+} and Cr^{2+} in MgO.

Spectra of divalent Mn in NaCl ($g=2.0022$, $A=86.6$ gauss) and in KBr ($g=2.0041$, $A=94.96$ gauss), of divalent Eu in KCl, and of trivalent Gd in $CaCl_2$ and in $SrCl_2$ will be reported in a separate paper.

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[5] B. Bleaney and W. Low, Proc. Phys. Soc. (London) **A68**, 55 (1955).

[6] Professor J. Wertz has independently observed the spectrum of V and Mn in MgO. We are grateful to Dr. Wertz for communication on this subject.

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[8] The data are taken from reference 1 for Tutton salts at a dilution of 1000 and at a temperature of 20°K.

Possible Method of Measuring Magnetic Moments of *V* Particles

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It is reasonable to assume that those *V* particles which have a spin $I>0$ will, as a rule, be produced partially polarized with respect to their line of flight or their production plane. If their decay is observed before they have interacted with other particles or external fields, their "memory" of the polarization should remain intact and should, for *V* particles with $I>1/2$, lead to angular distributions of the decay products which are not isotropic. In fact, the study of such angular distributions, or some of their characteristic features, e.g., the distribution of dihedral angles between the decay planes and the production planes of *V* particles, has recently been discussed in detail as a promising method of determining the spins of *V* particles.[1–3] There are strong indications in the experiments of Fowler, Shutt, Thorndike and Whittemore[4] and of Walker and Shepard[5] that the decay planes of Λ^0's, produced in the reaction $p+\pi^-\rightarrow\Lambda^0+\theta^0$, are oriented in such a way as to form preferentially small dihedral angles with their production planes. Tentatively it thus appears that the Λ^0 has a spin $I\geqslant 3/2$.[6] If this should be confirmed it would seem possible to measure, besides the spin, the sign and magnitude of the magnetic moment of Λ^0 by studying the effect of a magnetic field on the distribution of decay planes. In the nonrelativistic limit ($v/c\ll 1$), we find that in a magnetic field of strength B, a particle with a magnetic moment μ will carry out Larmor precessions around the axis of the field with an angular frequency

$$\omega=\mu B/I\hbar,$$

in a sense determined by the sign of the magnetic moment. Any discernible characteristic of its angular distribution at decay which can be established in field free space (e.g., a plane of symmetry) will precess around the axis of the magnetic field through an angle

$$\varphi=\omega t=2.744\times 10^5\, gBt \text{ (degrees)},$$

where the gyromagnetic ratio $g=\mu/I$ (μ measured in nuclear magnetons), and t is the time (in sec) spent in the magnetic field, e.g., before entering a cloud chamber or inside one. Thus, with the perhaps optimistic, but not completely unrealistic assumptions of a magnetic field $B\approx 18\,000$ gauss, a time of flight in this field of the order of 10^{-9} sec, and a gyromagnetic ratio $g\approx 2$, we should find a precession angle $\varphi\approx 10°$.

Similar methods could be applied to obtain spins and magnetic moments of specific hypernuclei with $I>1/2$, especially if they could be produced in well-defined reactions, such as

$$K^-+He^4\rightarrow {}_{\Lambda}H^4+\pi^-,$$

or

$$K^-+He^4\rightarrow {}_{\Lambda}H^3+n, \text{ etc.}$$

Because of the smaller distances involved here, the use of more intense (pulsed[7]) magnetic fields may be considered.

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hfs Separations and hfs Anomaly in the $6\ ^2P_{3/2}$ Metastable Level of Tl^{203} and Tl^{205}†

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Precision measurements at weak field of the frequency of the hfs line ($F=2$, $m_F=0\leftrightarrow F=1$, $m_F=0$) in the metastable $6\,^2P_{\frac{3}{2}}$ level of the thallium