Angular Dependence of the Intensities of "Forbidden" Transitions of Mn^{2+} in MgO†

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There have been numerous studies of the positions and intensities of $\Delta M=\pm1$, $\Delta m=\pm1$ ESR lines of the divalent manganese ion in a variety of hosts. However, in determining expressions governing the line intensities, it has been assumed that the microwave field $\mathbf{H}_1$ is perpendicular to the static field $\mathbf{H}_0$. Depending upon the cavity mode and the placement of the sample within a microwave cavity, one may have significant $\mathbf{H}_1$ components parallel to $\mathbf{H}_0$. This paper gives expressions for the intensities of the $\Delta M=\pm1$, $\Delta m=\pm1$ lines of Mn^{2+} in MgO (octahedral symmetry) as a function of orientation of the field $\mathbf{H}_0$ relative to crystal axes and $\mathbf{H}_1$. The predicted ratio of intensities of the $(\frac{1}{2}, m)\leftrightarrow(-\frac{1}{2}, m-1)$ and the $(\frac{1}{2}, m-1)\leftrightarrow(-\frac{1}{2}, m)$ lines is $(0.87\cos\alpha-\sin 4\theta \sin\alpha)^2/(\sin4\theta \sin\alpha)^2$. This prediction is found to be in accord with experiment. In most of the hosts previously studied, the Mn^{2+} ion experiences a significant axial electric field component; for these the intensities of the $\Delta m\pm1$ lines would be nearly identical.

I. INTRODUCTION

THE usual selection rule for the lines observed in paramagnetic resonance spectra is $\Delta M=1$, $\Delta m=0$, where M and m denote the z components of electronic and nuclear spin, respectively. However, it is well known that "forbidden" transitions corresponding to the selection rules $\Delta M=1$, $\Delta m=\pm1$ are often observed. These transitions are caused by the admixture into the unperturbed wave functions $|M, m\rangle$ of wave functions having a z component of nuclear spin equal to $m\pm1$; this admixture arises via the combined action of the off-diagonal matrix elements of the hyperfine- and crystal-field operators.[1–5] Examples of Mn^{2+} spectra in which this mechanism is operative can conveniently be divided into two categories: (1) Those in which the Mn^{2+} ion is in a noncubic environment, and hence crystal-field terms of the type $DS_z{}^2$ are principally responsible for the admixture (e.g., Mn^{2+} in $CaCO_3$,[3] $ZnSiF_6\cdot6H_2O$,[6] $MgAl_2O_4$,[7] ZnO,[8] Al_2O_3,[9] ZnS,[10] $BaTiO_3$,[11] apatite and $ZnCO_3$,[12] β-Ga_2O_3,[13] $CaWO_3$,[14] and powdered MgO [15]). (2) The cubic cases, for which the cubic-field term of the type $\frac{1}{6}a[S_{x'}{}^4+S_{y'}{}^4+S_{z'}{}^4]$ is responsible for the admixture (e.g., Mn^{2+} in MgO,[15] CaO,[16,17] SrO,[17] and ZnSe [18]). Since the parameter D for the first category is typically an order of magnitude or more greater than the cubic-field parameter a in the second category, the $\Delta m=\pm1$ lines are far more intense in the noncubic cases, and can rival even the intensity of the $\Delta m=0$ lines.[4]

Expressions for the intensities of the $\Delta m=\pm1$ lines for all of the examples quoted above are based on the assumption that the microwave field $\mathbf{H}_1$ is perpendicular to the static magnetic field $\mathbf{H}$. Since this is not necessarily always the case, it is the purpose of this paper to discuss the effect upon the intensities of the "forbidden" $\Delta m=\pm1$ lines of a component of $\mathbf{H}_1$ parallel to $\mathbf{H}$. The particular system under discussion is Mn^{2+} in MgO, corresponding to category (2).

The Mn^{2+} spectrum consists of a set of six hyperfine lines (corresponding to the $\Delta m=0$ transitions), each being split into five fine-structure components. Approximately midway between the m and $(m+1)$ hyperfine lines one observes the forbidden transitions $(M=\frac{1}{2}, m)\leftrightarrow(-\frac{1}{2}, m+1)$ and $(\frac{1}{2}, m+1)\leftrightarrow(-\frac{1}{2}, m)$. The field separation of this doublet is approximately

$$\delta H=\tfrac{17}{2}(A^2/g^2\beta^2H)+2(g_n\beta_n/g\beta)H.$$

(See Drumheller and Rubins[5] for a more accurate expression.) The forbidden lines for the other fine-structure transitions ($M=\frac{3}{2}\leftrightarrow\frac{1}{2}$, etc.) are not observed, presumably because they are less intense and broader than the $M=\frac{1}{2}\leftrightarrow-\frac{1}{2}$ transitions; in addition, for given directions of the crystal axes and microwave field, only a few of the $M=\frac{1}{2}\leftrightarrow-\frac{1}{2}$ forbidden transitions can be measured, because of the multitude of observed lines in this region.

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II. INTENSITIES OF "FORBIDDEN" LINES

A. Derivation of Intensity Formulas

The spin Hamiltonian for Mn^{2+} in a cubic field is

$$\mathcal{H}=g\beta\mathbf{H}\cdot\mathbf{S}+A\mathbf{S}\cdot\mathbf{I}+V_{\mathrm{cub}}-g_n\beta_n\mathbf{H}\cdot\mathbf{I}. \quad (1)$$

Here $S=\frac{5}{2}$, $I=\frac{5}{2}$, and the cubic field term is given by

$$\begin{aligned} V_{\mathrm{cub}}&=\tfrac{1}{6}a[S_{x'}{}^4+S_{y'}{}^4+S_{z'}{}^4-\tfrac{1}{5}\mathbf{S}^2(3\mathbf{S}^2-1)] \\ &\equiv(a/24)[7S_{z'}{}^4-(6\mathbf{S}^2-5)S_{z'}{}^2 \\ &\quad+\tfrac{3}{5}\mathbf{S}^2(\mathbf{S}^2-2)+\tfrac{1}{2}(S_{+'}{}^4+S_{-'}{}^4)], \end{aligned} \quad (2)$$

relative to the crystallographic axis x', y', and z'; we write $\mathbf{S}^2$ for $S(S+1)$.

The experimental results are obtained for the case in which the applied magnetic field $\mathbf{H}$, whose direction defines the z axis, lies in the crystallographic $x'z'$ plane. We can therefore define the y axis to be parallel to the y' axis; θ is taken as the angle between the z and z' axes, such that a positive rotation of θ about the y axis transforms z into z'. It can be seen that, since the crystallographic axes x', y', and z' are equivalent, this definition of axes is not unique; however, no ambiguity arises in the final expression for the transition probabilities.

The applied microwave field is also arranged to lie in the xz (or $x'z'$) plane, with components $H_1{}^{\parallel}$ and $H_1{}^{\perp}$ in the z and x directions, respectively. The Hamiltonian for the microwave field is taken to be

$$\mathcal{H}_1=g\beta(H_1{}^{\parallel}S_z+H_1{}^{\perp}S_x). \quad (3)$$

In terms of the spin operators in the x, y, z coordinate system, V_{cub} can be evaluated by the substitutions

$$S_{x'}=S_x\cos\theta-S_z\sin\theta,$$

$$S_{y'}=S_y,$$

and

$$S_{z'}=S_z\cos\theta+S_x\sin\theta; \quad (4)$$

however, it is unnecessary to make a complete evaluation.

The zeroth-order wave functions are denoted by $|M,m\rangle$. These wave functions are perturbed by the off-diagonal matrix elements of the spin Hamiltonian of Eq. (1); we denote the perturbed wave function as $\psi(M,m)$, and the off-diagonal operator as $\mathcal{A}+\mathcal{V}$, where $\mathcal{A}=\frac{1}{2}A(S_+I_-+S_-I_+)$, and $\mathcal{V}$=off-diagonal part of V_{cub}. We shall restrict our interest to those states admixed into $\psi(M,m)$ for which the denominator is of order $g\beta H$; furthermore, since we are interested in forbidden transitions for which $\Delta M=\pm1$, $\Delta m\neq0$, we need only consider admixtures of states for which these selection rules apply. It is therefore necessary to go only to second order in perturbation theory, so that the only states in $\psi(M,m)$ which we need consider are the following:

$$\begin{aligned} \psi(M,m)=&\,|M,m\rangle-(\langle M-1,m+1|\mathcal{A}|M,m\rangle/-g\beta H)\,|M-1,m+1\rangle \\ &-(\langle M+1,m-1|\mathcal{A}|M,m\rangle/g\beta H)\,|M+1,m-1\rangle+[|M,m+1\rangle/g\beta H(AM-g_n\beta_nH)] \\ &\times[\langle M,m+1|\mathcal{A}|M+1,m\rangle\langle M+1,m|\mathcal{V}|M,m\rangle-\langle M,m+1|\mathcal{V}|M-1,m+1\rangle\langle M-1,m+1|\mathcal{A}|M,m\rangle] \\ &+[|M,m-1\rangle/g\beta H(AM-g_n\beta_nH)][\langle M,m-1|\mathcal{A}|M-1,m\rangle\langle M-1,m|\mathcal{V}|M,m\rangle \\ &-\langle M,m-1|\mathcal{V}|M+1,m-1\rangle\langle M+1,m-1|\mathcal{A}|M,m\rangle]. \end{aligned} \quad (5)$$

This is easily reduced to the form

$$\begin{aligned} \psi(M,m)=&\,|M,m\rangle+(A/2g\beta H)[S_-{}^MI_+{}^m|M-1,m+1\rangle-S_+{}^MI_-{}^m|M+1,m-1\rangle] \\ &+[A/2g\beta H(AM-g_n\beta_nH)][I_+{}^m(S_+{}^M\langle M+1|\mathcal{V}|M\rangle-S_-{}^M\langle M|\mathcal{V}|M-1\rangle)\,|M,m+1\rangle \\ &+I_-{}^m(S_-{}^M\langle M-1|\mathcal{V}|M\rangle-S_+{}^M\langle M|\mathcal{V}|M+1\rangle)\,|M,m-1\rangle]. \end{aligned} \quad (6)$$

Here $S_\pm{}^M\equiv[\mathbf{S}^2-M(M\pm1)]^{1/2}$, $I_\pm{}^m\equiv[\mathbf{I}^2-m(m\pm1)]^{1/2}$. We therefore require the matrix elements $\langle M|\mathcal{V}|M\pm1\rangle$ which are independent of m; thus, we need only those parts of $\mathcal{V}$ which shift the M value by ±1. Since V_{cub} is composed of fourth-order spin tensors $O_q{}^{(4)}$ (definitions of these are given, for example, in Ref. 3), we look for the coefficients of the parts which have the form of $O_{\pm1}{}^{(4)}$, namely,

$$[\mp14S_z{}^3+21S_z{}^2\mp(19-6\mathbf{S}^2)S_z+(6-3\mathbf{S}^2)]S_\pm. \quad (7)$$

In fact, we have merely to pick out the coefficients of $S_z{}^3S_+$ and $S_z{}^3S_-$ after substituting Eq. (4) into Eq. (1); these turn out to be $(7a/48)\sin4\theta$ for both $S_z{}^3S_+$ and $S_z{}^3S_-$. It follows that

$$S^M\langle M\pm1|\mathcal{V}|M\rangle-S_\mp{}^M\langle M|\mathcal{V}|M\mp1\rangle=\mp(a/48)\sin4\theta[35M^4+(25-30\mathbf{S}^2)M^2+3\mathbf{S}^2(\mathbf{S}^2-2)]. \quad (8)$$

This can be substituted into the last part of Eq. (6). The observed "forbidden" transitions correspond to the transitions $M=\frac{1}{2}\leftrightarrow-\frac{1}{2}$; the relevant perturbed wave functions are therefore

$$\psi(\tfrac{1}{2},m)=|\tfrac{1}{2},m\rangle+(A/2g\beta H)[3I_+^m|-\tfrac{1}{2},m+1\rangle-2\sqrt{2}I_-^m|\tfrac{3}{2},m-1\rangle]$$
$$-[5aA\sin 4\theta/2g\beta H(A-2g_n\beta_n H)][I_+^m|\tfrac{1}{2},m+1\rangle-I_-^m|\tfrac{1}{2},m-1\rangle]\quad(9)$$

and

$$\psi(-\tfrac{1}{2},m')=|-\tfrac{1}{2},m'\rangle+(A/2g\beta H)[-3I_-^{m'}|\tfrac{1}{2},m'-1\rangle+2\sqrt{2}I_+^{m'}|-\tfrac{3}{2},m'+1\rangle]$$
$$+[5aA\sin 4\theta/2g\beta H(A+2g_n\beta_n H)][I_+^{m'}|-\tfrac{1}{2},m'+1\rangle-I_-^{m'}|-\tfrac{1}{2},m'-1\rangle].\quad(10)$$

The required transition probabilities are given by the squares of the matrix elements between the above states of the microwave Zeeman operator [Eq. (3)]; thus, we find

$$\langle\psi(\tfrac{1}{2},m)|\mathcal{H}_1/g\beta|\psi(-\tfrac{1}{2},m')\rangle=\tfrac{3}{2}H_1^{\perp}\delta_{m',m}+\tfrac{3}{2}H_1^{\perp}\delta_{m',m-1}(5aI_-^m\sin 4\theta/g\beta H)[A^2/(A^2-4g_n^2\beta_n^2H^2)]$$
$$-\tfrac{3}{2}\delta_{m',m+1}(I_+^m/g\beta H)\{AH_1^{\|}+5aH_1^{\perp}\sin 4\theta[A^2/(A^2-4g_n^2\beta_n^2H^2)]\}.\quad(11)$$

This expression is correct up to terms involving the first power of $1/g\beta H$.

Since $g_n\beta_nH\sim10^{-4}$ cm^{-1}, while $A=-81.3\times10^{-4}$ cm^{-1}, one can neglect $(g_n\beta_nH)^2$ as compared to A^2. Thus, we obtain the ratios of the strengths of the $\Delta m=0$ and $\Delta m=\pm1$ transitions as

$$(\tfrac{1}{2},m)\leftrightarrow(-\tfrac{1}{2},m):(H_1^{\perp})^2$$
$$(\tfrac{1}{2},m)\leftrightarrow(-\tfrac{1}{2},m-1):(25a^2\sin^2 4\theta/g^2\beta^2H^2)[I(I+1)-m(m-1)](H_1^{\perp})^2$$
$$(\tfrac{1}{2},m)\leftrightarrow(-\tfrac{1}{2},m+1):(25a^2/g^2\beta^2H^2)[I(I+1)-m(m+1)][(-AH_1^{\|}/5a)-H_1^{\perp}\sin 4\theta]^2.\quad(12)$$

Since[19] $a=+18.6\times10^{-4}$ cm^{-1}, the coefficient of $H_1^{\|}$ in the final bracket of Eq. (12) is 0.87.

B. Experimental Procedure

An undoped MgO crystal 5 mm long was ground to form a cylinder of 5-mm diameter. It was mounted in the center of a TE_{011} cylindrical reflection cavity having the longitudinal axis horizontal, so that the microwave field $\mathbf{H}_1$ lies in the plane of rotation of the applied magnetic field $\mathbf{H}$; one thus has variable microwave field components $H_1^{\|}$ and $H_1^{\perp}$. This type of cavity was selected for maximum homogeneity of the microwave field at the sample. The crystal could be rotated about a vertical $\langle001\rangle$-type axis, giving a variable angle θ between the applied field and one of the horizontal cubic axes. The x-band spectrometer operating at ~9.2 GHz used a balanced mixer with phase-sensitive detection at 390 Hz. The experiments were performed at room temperature.

C. Experimental Results

It can be seen that the inclusion of a component of the microwave field $\mathbf{H}_1$ parallel to the static magnetic field causes an additional term to appear in the intensity expression for the $\Delta M=-1$, $\Delta m=+1$ transition. This is the transition which conserves the total angular momentum component S_z+I_z, and it corresponds to the lower field component in each of the five doublets of "forbidden" lines which occur between the $\Delta m=0$ lines.

In our experiments, we have measured the ratio of the intensities of the components of the doublets which correspond to the transitions $(\frac{1}{2},m)\leftrightarrow(-\frac{1}{2},m-1)$ and $(\frac{1}{2},m-1)\leftrightarrow(-\frac{1}{2},m)$; we denote the intensities of the two types of lines as $P(m)$ and $Q(m-1)$, respectively. From Eq. (12), it follows that

$$P(m):Q(m-1)=[0.87\cos\alpha-\sin 4\theta\sin\alpha]^2:[\sin 4\theta\sin\alpha]^2,\quad(13)$$

where α denotes the angle between $\mathbf{H}_1$ and the static magnetic field. Table I gives a summary of the results

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TABLE I. Comparison of measured and predicted values of intensity ratios for the $\Delta m=\pm1$ doublets.

α [a]	θ [b]	Observed ratio[c] $P(m):Q(m-1)$	Predicted ratio
30°	−30°	$P(\frac{5}{2}):Q(\frac{3}{2})=5.2$	6.7
45°	−30°	$\bar{P}:\bar{Q}=4$	4
22.5°	−22.5°	$P(\frac{1}{2}):Q(-\frac{1}{2})=1.2$	1.2
90°	−22.5°	$\bar{P}:\bar{Q}=1$	1
−45°	22.5°	$P(\frac{3}{2}):Q(\frac{1}{2})=3.4$	3.5
45°	22.5°	$\bar{P}:\bar{Q}=0$	0
−30°	37°	$Q(-\frac{5}{2}):P(-\frac{3}{2})=0.08$	0.06
−30°	45°	$\bar{Q}:\bar{P}=0$	0
−45°	60°	$\bar{P}:\bar{Q}=0$	0.003
90°	67.5°	$\bar{P}:\bar{Q}=1$	1

[a] α is the angle between the main field and the microwave field.
[b] θ is the angle between the main field and one of the principal cubic axes.
[c] $P(m)$ is the relative amplitude of the derivative of the transition $(\frac{1}{2},m)\leftrightarrow(-\frac{1}{2},m-1)$.
$Q(m-1)$ is the relative amplitude for $(\frac{1}{2},m-1)\leftrightarrow(-\frac{1}{2},m)$.

for various values of θ and α. Because of the complexity of the spectrum, since each $\Delta m=0$ transition consists of five fine-structure lines, it is frequently impossible to measure all the $\Delta m=\pm1$ transitions. Thus, in Table I, where a measurement is given for a particular value of m, this is the only value for which the corresponding doublet of lines was measurable; otherwise, the symbols $\bar{P}$ and $\bar{Q}$ denote an average over several doublets. It should be noted that $Q(m)=0$ for $\theta=0$, $\pm\frac{1}{4}\pi$, while $P(m)=0$ for $\tan\alpha\,\sin4\theta=0.87$.

III. DISCUSSION

It can be seen that there is very satisfactory agreement between theory and experiment, considering the experimental difficulties of accurate alignment of the crystal and of measuring lines of low intensity. It is interesting to point out that the intensities of forbidden lines can be much increased by the presence of local deviations from cubic symmetry; this effect is spectacularly demonstrated in powder samples of MgO[15] and CaO, where the forbidden lines are an order of magnitude larger relative to the $\Delta M=1$, $\Delta m=0$ lines than in single MgO crystals. Such effects can also arise in single crystals; if we assume that the effect of random distortions can be represented by second-rank spin tensors (of the form $S_z^2-\frac{1}{3}S^2$, etc.), it can be shown that the averaged effect in a single crystal gives an additive contribution

$$I'=(F+G\cos4\theta)[(H_1^{\perp})^2/g^2\beta^2H^2][I(I+1)-m(m\pm1)]$$

to the forbidden line intensities of Eq. (12) (F and G are constants describing the average strength and angular distribution of the local distortions, where $F>|G|\geq0$). However, we find that it does not appear necessary to include such terms in the analysis of our experimental results on single crystals.

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Quantum Theory of Spontaneous Parametric Scattering of Intense Light*

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A quantum theory is developed for the spontaneous optical parametric process in nonlinear, anisotropic, and dispersive media; the process is treated directly as a scattering problem rather than a problem involving equivalent coupled oscillators. Following the general procedure in quantum field theory, a general expression is obtained for the differential extinction coefficient, which contains all the information on the intensity of the spontaneous emission in the parametric scattering process. The tuning characteristics, beam divergence, and spectral properties of the spontaneous emission for colinear and noncolinear interactions are described in detail and illustrated with numerical examples on such standard nonlinear optical crystals as $LiNbO_3$, $NH_4H_2PO_4$, and KH_2PO_4 under various pumping conditions. Explicit formulas for the intensity of the spontaneous emission are also obtained. The spontaneous intensity calculated using the formulas given is shown to be in complete agreement with known experimental data on $NH_4H_2PO_4$ pumped at 3472 Å.

I. INTRODUCTION

PARAMETRIC interaction of light waves in optically transparent nonlinear media is an effective method for producing continuously tunable amplification and generation of coherent optical radiation throughout the visible and into the infrared region of the spectrum.[1] Since the initial observation[2] of the parametric effect in KH_2PO_4 (KDP), significant progress has been made in developing such amplifiers and oscillators.[3–5] The theory of optical parametric process has also been discussed extensively in the past.[6–9] The calculations dealt mainly with the gain

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[1] See, for example, the review article by S. A. Akhmanov and R. V. Khohklov, Usp. Fiz. Nauk. **88**, 437 (1966) [English transl.: Soviet Phys.—Usp. **9**, 210 (1966)]. *Note added in proof.* After our paper was submitted, a letter by D. N. Klyshko, Zh. Eksperim. i Teor. Fiz. Pis'ma v Redaktsiyu **6**, 490 (1967) [English transl.: Soviet Phys.—JETP Letters **6**, 23 (1967)], which considers the same subject, became available in English translation.

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