PHYSICAL REVIEW B

CONDENSED MATTER

THIRD SERIES, VOLUME 35, NUMBER 7 1 MARCH 1987

EPR and optical-absorption studies of Cm^{3+} in YPO_4 and $LuPO_4$ single crystals

M. M. Abraham, L. A. Boatner, and C. B. Finch
Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831

W. K. Kot, J. G. Conway, G. V. Shalimoff, and N. M. Edelstein
Materials and Molecular Research Division, Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720

(Received 22 August 1986)

$^{244}Cm^{3+}$ has been incorporated as a dilute impurity in single crystals of the tetragonal-symmetry lanthanide orthophosphate hosts, $LuPO_4$ and YPO_4. EPR spectroscopic observations were made of all of the excited doublets in the ground $^8S_{7/2}$ multiplet of Cm^{3+} in both host crystals. Excited-state resonances were observed at both 77 and 4.2 K by use of *X*-band frequencies. The observed axial g values were used to compute the admixed wave functions for all four doublet levels in the ground manifold of Cm^{3+} and to calculate the g value expected for the lowest-lying doublet that could not be observed by EPR spectroscopy. Optical-absorption and Zeeman-effect investigations were carried out for the case of Cm^{3+} in YPO_4, and a ground-state g value was obtained for the lowest-lying doublet in the ground-state manifold that was in excellent agreement with the value predicted by use of EPR results for the three excited-doublet levels. The Mohr-circle construction was used to obtain additional information regarding the nature of the crystal-field interaction for Cm^{3+} in the two crystal systems. In particular, the tetragonal crystal-field interaction was found to be stronger in the YPO_4 host than in $LuPO_4$. Additionally, the crystal-field-induced admixtures were found to be smaller at 77 K than at 4.2 K for both hosts, and the signs of the Steven's parameters were determined to be positive for b_4^0 and negative for b_2^0.

I. INTRODUCTION

The trivalent lanthanide ion Gd^{3+} has a $4f^7$ electronic configuration, and, to first order, its associated $^8S_{7/2}$ ground state has no orbital angular momentum. The spin-orbit interaction for Gd^{3+} is ≈ 1600 cm^{-1}, and this interaction leads to so-called "intermediate-coupling effects" that result in admixtures of other states with $J=\frac{7}{2}$ but with different values of L and S into the $^8S_{7/2}$ ground state. When intermediate-coupling effects are taken into account, the Gd^{3+} ground state still retains a 97.4% S-state character. Therefore, in a crystalline host, the local crystal field will have a relatively small effect on the Gd^{3+} ground-state level. Ground-state splittings on the order of 0.1 to 1.0 cm^{-1} are observed experimentally, and the spin-lattice relaxation times are sufficiently long that Gd^{3+} paramagnetic resonance spectra can generally be observed at room temperature. In the case of the $5f^7$ configuration ion Cm^{3+}, however, the free-ion spin-orbit coupling parameter is ≈ 3000, cm^{-1}, i.e., almost twice as large as the value of ≈ 1600 cm^{-1} characteristic of Gd^{3+}.[1] Accordingly, intermediate-coupling effects are significantly larger for Cm^{3+}, and the ^8S content of the corresponding $5f^7$ ion is only 79%. This increased non-S-state character of the Cm^{3+} $5f^7$ ground state is responsible, at least in part, for a relatively larger interaction of the crystalline-electric field with the ground state of the Cm^{3+} ion, and splittings on the order of 10 to 50 cm^{-1} are observed. As a further consequence, the spin-lattice relaxation times are shorter for $5f^7$ configuration ions such as Cm^{3+} and, accordingly, low temperatures are generally required in order to observe their EPR spectrum.

In the first reported electron paramagnetic resonance (EPR) study of Cm^{3+}, its EPR spectrum was observed at 4.2 K in the hexagonal-symmetry hosts, lanthanum trichloride and lanthanum ethylsulfate.[2] In these hosts, the Cm^{3+} resonance line with $g_{\parallel}=g_J$ and $g_{\perp}\simeq 4g_J$ (where g_J is the Landé g factor) was due to the ground $|\pm\frac{1}{2}\rangle$ doublet which contained no observable crystal-field admixtures of the other M_s doublets. (An axial crystal field will split the predominantly $^8S_{7/2}$ ground state into four separate doublets of the form $|\pm\frac{1}{2}\rangle$, $|\pm\frac{3}{2}\rangle$, $|\pm\frac{5}{2}\rangle$, and $|\pm\frac{7}{2}\rangle$.) Since the hexagonal symmetry of the crystal field in $LaCl_3$ and lanthanum ethylsulfate only allows $\Delta M_s=\pm 6$

admixtures, an admixture could only occur between the $|\pm\frac{5}{2}\rangle$ and $|\mp\frac{7}{2}\rangle$ doublets in this case.

Subsequent to the initial studies on Cm^{3+} in $LaCl_3$ and lanthanum ethylsulfate,[2] Cm^{3+} resonances have also been observed for cubic symmetry sites in the fluorite-structure hosts: CeO_2, ThO_2, CaF_2, SrF_2, and $SrCl_2$.[3–8] In these hosts, the eight levels of the $J=\frac{7}{2}$ ground state are split by the cubic crystal field into two doublets and a quartet. The resulting levels are: a ground Γ_6 doublet, an excited Γ_8 quartet, and an even higher lying Γ_7 doublet. For the cubic fluorite-structure hosts, the Cm^{3+} EPR transition occurs between the Zeeman levels of the ground Γ_6 doublet. In these hosts, the applied magnetic field produced an admixture of the Γ_8 quartet into the Γ_6 ground doublet that resulted in an orientational anisotropy in the Γ_6 EPR transition. By studying this anisotropy, it was possible to determine the energy separation between the ground Γ_6 and first excited Γ_8 quartet levels for Cm^{3+} in all five fluorite-structure crystals. Additionally, in the case of $SrCl_2$, a magnetic resonance transition was also observed between the levels of the higher lying Γ_7 doublet. Analogous anisotropy measurements were carried out for this transition at a temperature of approximately 25 K, and a value was obtained for the Γ_8-Γ_7 splitting.[7] This measurement, together with the earlier determination of the Γ_6-Γ_8 splitting, was used to establish the values of the crystal-field parameters b_4^0 and b_6^0 for Cm^{3+} in $SrCl_2$.[5,7]

For the case of Cm^{3+} in the zircon-structure host $ThSiO_4$,[9] no Cm^{3+} EPR signals were detected. This result was not unexpected since the dominant crystal-field parameter b_2^0 is negative for the $4f^7$ ion Gd^{3+} in $ThSiO_4$,[9] and it was anticipated that b_2^0 for Cm^{3+} in $ThSiO_4$ would also be negative. A negative b_2^0 would reverse the order of the four ground doublets relative to that found for the trichloride and ethylsulfate hosts where b_2^0 is positive.[2] The lowest doublet for Cm^{3+} in $ThSiO_4$ would therefore be primarily a $|\pm\frac{7}{2}\rangle$ state with a value of $g_\perp \approx 0$. This would account for the absence of Cm^{3+} EPR signals in $ThSiO_4$.

Recently, in another zircon structure host, $LuPO_4$ doped with both ^{243}Cm ($I=\frac{5}{2}$) and ^{244}Cm ($I=0$), a positive identification was made[10] of a Cm^{3+} resonance. This resonance was identified as an excited-state transition by observing that lowering the sample temperature reduced the intensity of the EPR signal. The experimentally determined axial g values of this doublet were fitted to a wave function of the form $\alpha|\pm\frac{5}{2}\rangle+\beta|\mp\frac{3}{2}\rangle$. In this case, the tetragonal symmetry of the crystal field produces $\Delta M_s=\pm 4$ admixtures in the wave functions resulting in nonzero $g_\perp$ values for each of the four doublets that arise from the crystal-field splitting of the nominal $^8S_{7/2}$ ground state. Therefore, transitions within each doublet become allowed. The possibility of observing such transitions for excited states and, thereby obtaining more detailed information regarding the electronic properties of Cm^{3+} in orthophosphate hosts has led to the investigations reported here.

In the present work, EPR and optical absorption techniques have been used to observe and investigate all of the excited doublets of the ground $J=\frac{7}{2}$ multiplet for the case of $^{244}Cm^{3+}$ incorporated as a dilute impurity in a single crystal of YPO_4 (xenotime). The axial g values experimentally determined for the three excited doublets were then used in a self-consistent calculation of the ground-state wave functions and g values for all four ground-state doublets. This calculation resulted in a prediction of the axial g values for the lowest-lying Cm^{3+} doublet whose EPR spectrum was not observed. Zeeman effect studies of the Cm^{3+} optical absorption in YPO_4 were used to verify the value for $g_{\parallel}$ calculated as noted above for the lowest-lying doublet of the ground multiplet. The previous observations[10] of an excited state resonance for Cm^{3+} in a single crystal of $LuPO_4$ have also been extended and new EPR results are reported for the two remaining excited doublets (i.e., for the doublets $\alpha|\pm\frac{3}{2}\rangle-\beta|\mp\frac{5}{2}\rangle$ and $\gamma|\pm\frac{1}{2}\rangle-\delta|\mp\frac{7}{2}\rangle$). The axial g values for these doublets were used along with the previous results for Cm^{3+} in $LuPO_4$ (Ref. 10) to carry out a self-consistent calculation of all the ground-state doublet wave functions for this system, and to predict the axial g values for the lowest lying doublet $\gamma|\pm\frac{7}{2}\rangle+\delta|\mp\frac{1}{2}\rangle$, which was not observed by EPR.

II. MAGNETIC RESONANCE RESULTS

A. Cm^{3+} in $LuPO_4$ single crystals

The EPR spectrum arising from the excited doublet in the ground multiplet of the form $\alpha|\pm\frac{5}{2}\rangle+\beta|\mp\frac{3}{2}\rangle$ has

TABLE I. Wave functions and g values for the ground-state doublets of Cm^{3+} in $LuPO_4$.

Sample temperature	Ground-state doublets	Calculated $g_{\parallel}$	Calculated $g_\perp$	Measured by EPR $g_{\parallel}$	Measured by EPR $g_\perp$
77 K	$0.9837|\pm\frac{1}{2}\rangle-0.1798|\mp\frac{7}{2}\rangle$	1.424	7.436	1.424(2)	
	$0.9463|\pm\frac{3}{2}\rangle-0.3233|\mp\frac{5}{2}\rangle$	4.155	4.07	4.15(1)	4.07(1)
	$0.9463|\pm\frac{5}{2}\rangle+0.3233|\mp\frac{3}{2}\rangle$	8.00	4.07	8.00(2)	4.07(1)
	$0.9837|\pm\frac{7}{2}\rangle+0.1798|\mp\frac{1}{2}\rangle$	12.95	0.25		
4.2 K	$0.98205|\pm\frac{1}{2}\rangle-0.1886|+\frac{7}{2}\rangle$	1.373	7.402	1.373(1)	7.402(5)
	$0.9455|\pm\frac{3}{2}\rangle-0.3255|\mp\frac{5}{2}\rangle$	4.13	4.096	4.12(1)	4.10(1)
	$0.9455|\pm\frac{5}{2}\rangle+0.3255|\mp\frac{3}{2}\rangle$	7.976	4.096	7.98(1)	4.096(4)
	$0.98205|\pm\frac{7}{2}\rangle+0.1886|\mp\frac{1}{2}\rangle$	12.9	0.27		

been investigated previously[10] for the case of Cm^{3+} in $LuPO_4$. Subsequently, it has been possible to observe not only the EPR spectrum of this excited state but those of the two remaining excited doublets in the ground-state manifold of Cm^{3+} as well. All three excited-state resonances were observed at sample temperatures of either 77 or 4.2 K. In the case of the $\gamma\,|\pm\frac{1}{2}\rangle-\delta\,|\mp\frac{7}{2}\rangle$ state, the resonance transition could not be followed to the $g_\perp$ orientation at 77 K due to line broadening and subsequent loss of the signal intensity. However, the $g_\perp$ value of this transition could be determined at 4.2 K. The experimentally determined axial g values at both temperatures are listed in Table I for the three excited doublets of Cm^{3+} in $LuPO_4$. A resonance associated with the lowest-lying doublet was not detected at either sample temperature. Small tetragonal, crystal-field-induced admixtures of the excited $|\mp\frac{1}{2}\rangle$ wave function into the ground $|\pm\frac{7}{2}\rangle$ level produce a finite, but relatively small, $g_\perp$ value for the ground doublet. This small $g_\perp$ value will result in a much lower transition probability for a magnetic resonance transition between the levels of the $\gamma\,|\pm\frac{7}{2}\rangle+\delta\,|\mp\frac{1}{2}\rangle$ doublet than that associated with the other higher-lying doublets. The failure to observe a resonance transition for the lowest-lying state can be attributed to this effect. The wave function and axial g values of the lowest-lying state can be computed using the measured g values for the three excited doublets in the ground-state manifold, and a set of coefficients and g values can be calculated which represents a self-consistent "best fit" to the data used. The results of such a calculation are given in Table I for Cm^{3+} in $LuPO_4$. Using data for a sample temperature of 77 K, the predicted g values for the lowest-lying doublet are $g_{\parallel}=12.95$ and $g_\perp=0.25$, with a wave function given by $0.9837\,|\pm\frac{7}{2}\rangle+0.1798\,|\mp\frac{1}{2}\rangle$. The corresponding results for $T=4.2$ K are listed in Table I.

Additional information can be obtained which relates to the crystal-field interaction by utilizing a procedure known as the Mohr circle construction.[11] It is possible to express the tetragonal crystal-field admixture as the quantity $\tan(2\theta)$, where $\tan(2\theta)$ is equal to twice the off-diagonal crystal-field matrix element divided by the difference in the diagonal elements. Therefore, for the case of admixtures of the $|\pm\frac{7}{2}\rangle$ and $|\mp\frac{1}{2}\rangle$ levels, $\tan(2\theta)$ is given by

$$\tan(2\theta)=\frac{2(\sqrt{35}/5)b_4^4}{(-12b_2^0+2b_4^0-6b_6^0-4g_J\mu_B H)}\,, \tag{1}$$

and the corresponding expression for admixtures of the $|\pm\frac{5}{2}\rangle$ and $|\mp\frac{3}{2}$ levels is

$$\tan(2\theta)=\frac{(2\sqrt{3})b_4^4}{(-4b_2^0+10b_4^0+14b_6^0-4g_J\mu_B H)}\,, \tag{2}$$

where the b_n^m are the crystal-field parameters introduced by Stevens.[12] The values of $\tan(2\theta)$ for the two different admixtures may be calculated from the wave-function coefficients listed in Table I. (The coefficients are merely $\cos\theta$ and $\sin\theta$.) In this manner, $\tan(2\theta)=0.7736$ and 0.7811 for the first admixture at $T=77$ and 4.2 K, respectively, and $\tan(2\theta)=0.3782$ and 0.3988 for the second admixture at $T=77$ and 4.2 K, respectively. The ratio of the two admixtures is found to be approximately 2, and not about 4, as would be expected if b_2^0 were the dominant parameter in the denominators of the expressions given above for $\tan(2\theta)$. It is clear that the parameters b_4^0 and b_6^0 cannot be neglected in Eqs. (1) and (2). Since b_2^0 is negative, it can be concluded that b_4^0 and probably b_6^0 are both positive for Cm^{3+} in $LuPO_4$.

B. Cm^{3+} in YPO_4 single crystals

The experimentally determined axial g values for the three excited doublets of Cm^{3+} in YPO_4 at 77 K are given in Table II, along with the calculated "best fit" wave functions and g values for all four levels at 77 K. The predicted g values for the lowest-lying doublet resulting from this procedure are seen to be $g_{\parallel}=13.3$ and $g_\perp=0.08$. While resonances from all three excited levels were observed at 77 K, only one resonance could be seen at 4.2 K, due to thermal depopulation of the two higher doublets. Table II therefore shows the experimental results and calculated values for only two doublets for Cm^{3+} in YPO_4 at 4.2 K. Using the Mohr-circle construction discussed previously, the $\tan(2\theta)$ values for the $|\pm\frac{5}{2}\rangle$ and $|\mp\frac{3}{2}\rangle$ admixture at 77 and 4.2 K were calculated to be 0.5924 and 0.5943, respectively, while the $\tan(2\theta)$ value for the $|\pm\frac{7}{2}\rangle$ and $|\mp\frac{1}{2}\rangle$ admixture is 0.2133 at 77 K. It can be seen that, since the admixtures

TABLE II. Wave functions and g values for the ground-state doublets of Cm^{3+} in YPO_4.

Sample temperature	Ground-state doublets	Calculated		Measured by EPR	
		$g_{\parallel}$	$g_\perp$	$g_{\parallel}$	$g_\perp$
77 K	$0.9945\,\lvert\pm\frac{1}{2}\rangle-0.1049\,\lvert\mp\frac{7}{2}\rangle$	1.75	7.60	1.745(5)	7.47(3)
	$0.9645\,\lvert\pm\frac{3}{2}\rangle-0.2642\,\lvert\mp\frac{5}{2}\rangle$	4.687	3.391	4.68(1)	3.391(5)
	$0.9645\,\lvert\pm\frac{5}{2}\rangle-0.2642\,\lvert\mp\frac{3}{2}\rangle$	8.53	3.391	8.53(1)	3.391(5)
	$0.9945\,\lvert\pm\frac{7}{2}\rangle+0.1049\,\lvert\mp\frac{1}{2}\rangle$	13.3	0.08		
4.2 K	$0.9643\,\lvert\pm\frac{3}{2}\rangle-0.2649\,\lvert\mp\frac{5}{2}\rangle$	4.687	3.401		
	$0.9643\,\lvert\pm\frac{5}{2}\rangle+0.2649\,\lvert\mp\frac{3}{2}\rangle$	8.53	3.401	8.53(1)	3.041(5)

of the wave functions for Cm^{3+} in YPO_4 are smaller than the corresponding admixtures for Cm^{3+} in $LuPO_4$, the tetragonal crystal field in the YPO_4 host is stronger than it is in the $LuPO_4$ host (i.e., the four doublets are farther apart in YPO_4 than they are in $LuPO_4$). Furthermore, since the ratio of the two different admixtures for Cm^{3+} in YPO_4 at 77 K is larger than the ratio for the LuPO4 host, it can be concluded that the effect of the b_4^0 and b_6^0 parameters relative to the b_2^0 parameter in the denominators of the expressions for $\tan(2\theta)$ is smaller for YPO_4. It is still true, however, that for Cm^{3+} in YPO_4 (just as in $LuPO_4$) the signs of b_4^0 (positive) and b_2^0 (negative) are different, and that the crystal-field-induced admixtures are smaller at 77 than at 4.2 K.

III. OPTICAL ABSORPTION RESULTS

The optical absorption spectrum of $^{244}Cm^{3+}$ in YPO_4 single crystals was measured photographically from approximately 16 000 cm^{-1} to 27 000 cm^{-1} for sample temperatures of 77 and 4.2 K. Linear polarization data were obtained with the electric vector oriented either parallel or perpendicular to the tetragonal *c* axis of the crystal. Zeeman spectra were obtained on a few of the strongest low-energy lines at 4.2 K with the magnetic field oriented parallel to the *c* crystal axis.

The optical spectra obtained were of rather poor quality. Although some lines were sharp, most of the lines were weak and diffuse. A major reason for this difficulty was related to the use of ^{244}Cm, an isotope with a 17.6 year half-life. Radiation damage produced significant broadening of the sharper lines in a few days, but this broadening could be reduced by annealing the crystals for 2 h at $\approx$500 °C. Orthophosphate crystals are now being grown which contain ^{248}Cm, an isotope with a 4.7×10^5 year half-life, and high-resolution experiments will be carried out on these samples in the future.

The major difference between the optical spectra of Cm^{3+} in YPO_4 at 77 and 4.2 K is a change in the relative intensities of pairs of lines that are separated by $\approx$11.7 cm^{-1}—with the lower-energy member of the pair being much weaker at 4.2 K. The positions of some of the lines which exhibit this intensity change are given in Table III.

The principal components of the ground-state eigenvector for the Cm^{3+} ion have been given as:[6]

$$\Psi(Cm^{3+})=0.891\,|\,{}^8S_{7/2}\rangle +0.414\,|\,{}^6P_{7/2}\rangle-0.090\,|\,{}^6D_{7/2}\rangle+\cdots\,.$$

TABLE III. Measured optical absorption lines in YPO_4:Cm^{3+}.

Cm^{3+} absorption lines (cm^{-1})	Difference (cm^{-1})
16 579.5	11.7
16 567.8	
19 817.0	11.5
19 805.5	
22 625.7	11.9
22 613.8	

TABLE IV. Experimental Zeeman data for Cm^{3+} in YPO_4.

J'	Level zero field (cm^{-1})	Level 26.3 kG (cm^{-1})	$\lvert g_{\parallel}\rvert$ ground state	$\lvert g_{\parallel}\rvert$ excited state
		16 590.7		
$\frac{7}{2}$	16 579.5	16 584.9	13.4	4.73
		16 636.3		
$\frac{7}{2}$	16 624.3	16 628.6	13.3	6.23
		19 826.4		
$\frac{5}{2}$	19 817.0	19 823.6	13.0	2.31

The intermediate coupled $J=\frac{7}{2}$ state is primarily $^8S_{7/2}$ and, as noted previously, this manifold should split into four levels (each level doubly degenerate) in an axial crystal field. Previous work on Cm^{3+} in various host crystal systems indicates that the overall splitting should be on the order of 10 to 50 cm^{-1}. The EPR data reported here for Cm^{3+} in YPO_4 are consistent with this limit to the total splitting. Additionally, the results suggest that the lower two levels are very close in energy. The observed optical splitting may be explained by assuming that the four levels (in zero field) are ordered in pairs with an energy difference of 11.7 cm^{-1} between the pairs. We estimate that there may be a splitting as large as 2 to 4 cm^{-1} between the lowest two levels.

In order to determine the ground-state *g* value, the optical spectrum was measured in a magnetic field of 26.3 kG applied parallel to the fourfold *c* axis of the crystal. Four lines exhibited a splitting as well as a significant shift from their zero-field position. If the ground level has a very large Zeeman splitting compared with the Zeeman splitting of the excited state, a large shift to higher energy of the center of gravity of the excited state would be expected. The $g_{\parallel}$ values of the ground and excited states derived from this data are given in Table IV. The optically determined values of $g_{\parallel}$ ranging from 13.0 to 13.4 for the ground state are in excellent agreement with the value of $g_{\parallel}=13.3$ derived from the EPR data.

IV. SUMMARY

Paramagnetic resonance spectra have been observed for all of the excited doublets in the crystal-field-split $J=\frac{7}{2}$ ground manifold of Cm^{3+} in the host single crystals $LuPO_4$ and YPO_4. The axial *g* values have been determined for Cm^{3+} in both host crystals at temperatures of 77 and 4.2 K, and these results have been used to calculate the wave functions for all four doublets and to predict the values of the axial *g* values appropriate to the lowest-lying doublet that is not observed by EPR spectroscopy. Information regarding the nature of the crystal field interaction has been obtained for the two systems investigated. In the case of YPO_4 doped with Cm^{3+}, optical absorption and Zeeman effect measurements have been used to investigate the ground-state properties of the trivalent ion, and a $g_{\parallel}$ value was obtained for the lowest-lying doublet that is in excellent agreement with the value computed with the

EPR results for the three excited doublets in the ground multiplet.

Preliminary calculations employing free-ion parameters of Cm^{3+} in $LaCl_3$ and the crystal-field parameters of Nd^{3+} in YPO_4 have been performed which give the correct ordering for the YPO_4:Cm^{3+} ground-state levels. These show two pairs of two levels, each of which is separated by about 6 cm^{-1} from the center of gravity of the pairs. The calculated $g_{\parallel}$ values for these levels agree very well with those determined by the EPR measurements. Assignments can be made for these excited states on the basis of the g values and polarization data, but a detailed analysis awaits measurements on crystals doped with the longer lived ^{248}Cm isotope, where the effects of radiation-induced line broadening will be significantly reduced.

ACKNOWLEDGMENTS

This research was sponsored by the Division of Materials Sciences, U.S. Department of Energy, under Contract No. DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc., and by the Director, Office of Energy Research, Office of Basic Energy Sciences, Chemical Sciences Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098. The authors are indebted to John Bigelow and other staff members of the transplutonium element production facilities at the Oak Ridge National Laboratory. The authors also acknowledge with thanks the excellent technical contributions of H. E. Harmon.

---

[1] B. G. Wybourne, *Spectroscopic Properties of Rare Earths* (Interscience-Wiley, New York, 1965).

[2] M. M. Abraham, B. R. Judd, and H. H. Wickman, Phys. Rev. **130**, 611 (1963).

[3] M. M. Abraham, G. W. Clark, and C. B. Finch, Phys. Rev. **168**, 933 (1968).

[4] N. Edelstein, W. Easley, and R. McLaughlin, J. Chem. Phys. **44**, 3130 (1966).

[5] M. M. Abraham, L. A. Boatner, C. B. Finch, R. W. Reynolds, and H. Zeldes, Phys. Rev. B **1**, 3555 (1970).

[6] N. Edelstein and W. Easley, J. Chem. Phys. **48**, 2110 (1968).

[7] W. Kolbe, N. Edelstein, C. B. Finch, and M. M. Abraham, J. Chem. Phys. **56**, 5432 (1972).

[8] W. Kolbe, N. Edelstein, C. B. Finch, and M. M. Abraham, J. Chem. Phys. **58**, 820 (1973); **59**, 1568E (1973).

[9] M. M. Abraham, G. W. Clark, C. B. Finch, R. W. Reynolds, and H. Zeldes, J. Chem. Phys. **50**, 2057 (1969).

[10] M. M. Abraham and L. A. Boatner, Phys. Rev. B **26**, 1434 (1982).

[11] J. J. Nye, *Physical Properties of Crystals* (University, Oxford, England, 1957), p. 43.

[12] K. W. H. Stevens, Proc. Phys. Soc., London, Ser. A **65**, 209 (1952).