Spin Hamiltonian Parameter D versus Axial Crystal Field for S-State Ions*

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Measurements have been made, in a Mössbauer absorption experiment, of the nuclear quadrupole splitting of the $^6S_{5/2}$ state ion Fe^{3+} substituted in several compounds. The compounds chosen were those for which electron spin resonance measurements had previously been made of the quantity D in the electron spin resonance Hamiltonian. The measured quadrupole coupling eqQ/h is directly proportional to the axial crystal field, V_{ax}. Hence, it is possible to compare the results of the dependence of D on the axial crystal field with theoretical predictions of Watanabe and Pryce. It is found that considerable scatter of data points occurs about any smooth curve one might draw. This scatter is possibly the result of covalent bonding or the distortion of the iron wave functions by the surrounding ions. Moreover, a functional dependence of any smooth curve through the experimental points disagrees with the theoretical calculations. If the data are to be even crudely fit by a smooth curve, the sign of the coefficient of V_{ax}^2 should be positive and not negative as theory indicates. Finally, there is serious disagreement between theory and experiment on the dependence of the D vs V_{ax} curve on the particular paramagnetic ion (i.e., on whether the substituted ion is Fe^{3+} or Mn^{2+}).

INTRODUCTION

THE problem of estimating the magnitude of the ground-state splitting of $^6S_{5/2}$ state ions under the influence of a crystal field was first considered by Van Vleck and Penney.[1] Although a pure S state will not be split by either the crystalline field or by spin-orbit coupling, they demonstrated that admixtures of excited states into the ground state could cause a splitting. Using perturbation theory they obtained estimates of some processes involving spin-orbit coupling and the crystal field interaction in order to explain anisotropic magnetic susceptibilities of salts containing manganous ions. While Van Vleck and Penney's work was of a preliminary nature, it did indicate the complexity of the origin of the ^6S state splitting.

Because of the complicated nature of the interaction between a paramagnetic ion and its environment, electron spin resonance results have been interpreted in terms of a phenomenological spin Hamiltonian which for S-state ions is

$$\mathcal{H}=g\beta\mathbf{H}\cdot\mathbf{S}+D[S_z^2-\tfrac{1}{3}S(S+1)]+\tfrac{1}{6}a[S_x^4+S_y^4+S_z^4-\tfrac{1}{5}S(S+1)(3S^2+3S-1)]+\cdots. \quad (1)$$

Recently Watanabe[2] has reawakened interest in the relation between the spin Hamiltonian parameters, D and a, and the coefficients, A_2^0 and A_4^0, of the crystal field potential,

$$V_{\text{crys}}=A_2^0r^2Y_2^0+A_4^0r^4\times[Y_4^0+(5/14)^{1/2}(Y_4^4+Y_4^{-4})]+\cdots. \quad (2)$$

The first two terms (the axial and the cubic terms) of the crystal field potential are usually designated as V_{ax} and V_{cub}, respectively. Watanabe's calculation follows some earlier work of Pryce[3] who calculated a term contributing to D which was linear in V_{ax} and in the spin-spin interaction, W_{SS}. Watanabe estimated an additional contribution to D involving the square of V_{ax} and the spin-orbit interaction, W_{LS}. In addition, he calculated the contribution to a of several excited state configurations admixed into the ground state through either quadratic of quartic powers of the cubic potential. However, later work by Powell, Gabriel, and Johnston[4] indicated that terms in addition to those calculated by Watanabe must be considered in determining the variation of a with V_{cub}.

The object of this work was to experimentally measure the relation between D and V_{ax}. This was done using the Mössbauer absorption technique to measure the quadrupole coupling constant, eqQ/h, of Fe^{3+} in several host lattices for which D of Fe^{3+} had been determined previously by electron spin resonance methods. Since Fe^{3+} is in a spherically symmetric S state, the field gradient is directly related to the crystal field parameter A_2^0 by the relation

$$q=(1-\gamma_\infty)q_u=(1-\gamma_\infty)(5/\pi)^{1/2}A_2^0=(1-\gamma_\infty)\times\sum_i\frac{3\cos^2\theta_i-1}{r_i^3}e_i. \quad (3)$$

* A preliminary account of this work was given at the New York meeting of the American Physical Society, 1962 [W. J. Nicholson and G. Burns, Bull. Am. Phys. Soc. 7, 85 (1962)].

[1] J. H. Van Vleck and W. G. Penney, Phil. Mag. 17, 961 (1934).

[2] H. Watanabe, Progr. Theoret. Phys. (Kyoto) 18, 405 (1957). A very recent consideration of the ^6S state problem is that of Germanier, Gainon and Lacroix (see reference 25). This paper is commented on briefly at the conclusion of the discussion section.

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Here γ_∞ is the antishielding factor[5] appropriate to iron.[6]

RESULTS

The resonant absorption measurements were made using Co^{57} diffused into either stainless steel or copper. A loudspeaker velocity drive was used in conjunction with a multichannel analyzer to obtain a velocity spectrum. The various crystals were prepared by standard techniques. The oxide crystals were made by reacting the appropriate oxides and Fe_2O_3 at high temperature. The water soluble crystals were grown from a solution containing the appropriate iron salt and the host lattice salt. In each case approximately 1 to 2% of Fe^{57} was substituted into the host lattice. X-ray analysis was used to check that the solid state reaction with Fe_2O_3 yielded only the single phase desired.

The absorption results are shown in Fig. 1 and listed in the Table I along with the measured values of D. In nearly all cases the two components of the quadrupole splitting pattern were clearly resolved. In the cases of yttrium gallium garnet and beryl, the quadrupole interaction was estimated from the broadening of the principal absorption line in a comparison with an absorption line measured using natural iron. In the case of iron, an absorption linewidth of less than 0.35 mm/sec was obtained. That Fe^{3+} had been substituted into the various compounds (rather than Fe^{2+}) can be verified by the fact that the isomer shift corresponds to that of a $3d^5$ configuration.[7] In several spectra, most noticeably in the case of beryl, additional small peaks occur at -0.75 and 1.8 mm/sec. These correspond to traces of Fe_2O_3 that did not react with the host oxides. In the case of yttrium gallium garnet there is an additional absorption line at -0.2 mm/sec. This can possibly be attributed to iron atoms which have entered substitutionally into the tetrahedral site. The principal absorption line of yttrium gallium garnet is believed to originate from iron atoms located at octahedral sites. This follows from the results of Geschwind[8] who found that approximately ten times as much Fe^{3+} entered the octahedral site as the tetrahedral site. Results on Al_2O_3 have previously been reported by Wertheim[9] and our results are in agreement with his.

In several cases an asymmetry with respect to the amount of resonant absorption is observed. This is most evident in the case of Al_2O_3. A possible explanation of the asymmetry can be found in effects resulting from the alignment of the iron nuclei by the electric field gradient produced by the host crystal lattice. Using the principal axis of the field gradient tensor as an axis of quantization, the $m=\pm\frac{3}{2}$ magnetic substates will be oriented predominantly parallel to the electric field gradient (and hence will have a higher energy) and the $m=\pm\frac{1}{2}$ substates predominantly perpendicular to the crystal field gradient (with a concomitantly lower

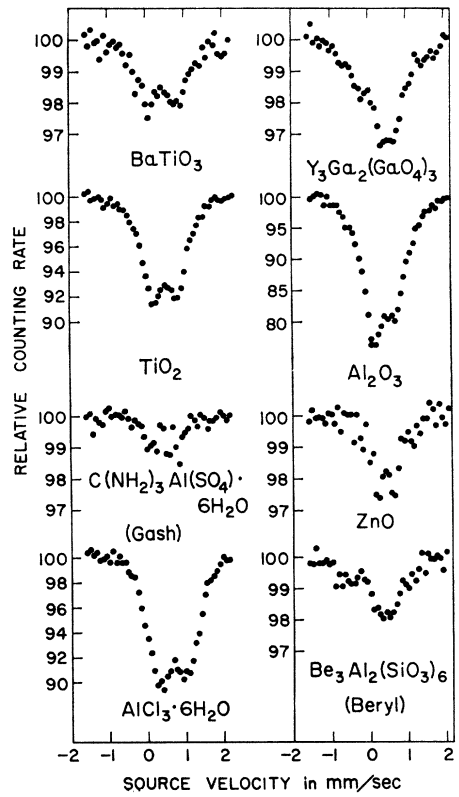

FIG. 1. Mössbauer absorption spectra for Fe^{3+} ions substituted into various host lattices.

TABLE I. The measured values of eqQ/h and of the isomer shift for Fe^{3+} in several host lattices along with previously measured values of the quantity D of the electron spin resonance Hamiltonian.

Host lattice	eqQ/h (Mc/sec)	Isomer shift at 300°K (mm/sec)	D (cm^{-1})	Reference
$BaTiO_3$	17.9±0.6	0.47±0.04	±0.097	a
$Y_3Ga(GaO_4)_3$	7.5±0.8	0.50±0.02	−0.1294(octa) −0.0880(tetra)	b
TiO_2	13.8±0.4	0.50±0.02	+0.6780	c
Al_2O_3	11.6±0.3	0.39±0.02	+0.1679	d
$C(NH_2)_3Al(SO_4)_2 \cdot 6H_2O$ (77°K)	14 ±3	0.50±0.10	−0.18902 −0.19506	e
ZnO	9.6±0.4	0.41±0.03	−0.0594	f
$AlCl_3 \cdot 6H_2O$ (77°K)	15.8±0.8	0.51±0.05	±0.150	g
$Be_3Al_2(SiO_3)_6$	4 ±2	0.42±0.05	+0.0140	h

[a] A. W. Hornig, R. C. Rempel, and H. E. Weaver, J. Phys. Chem. Solids **10**, 1 (1959).
[b] S. Geschwind, Phys. Rev. **121**, 363 (1961).
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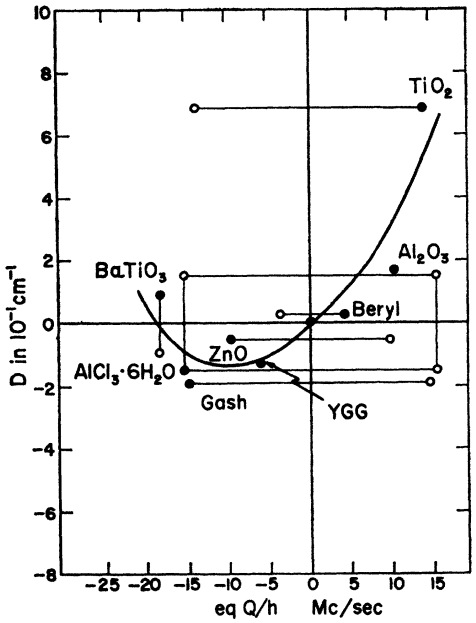

FIG. 2. Experimental values of the electron spin resonance parameter D plotted against measured values of the quadrupole coupling eqQ/h for Fe^{3+} in several crystal lattices.

energy). Since the 14.4-keV gamma rays of iron are from a magnetic dipole transition, the absorption to the $\pm\frac{3}{2}$ states will occur primarily from gamma rays incident perpendicular to the crystal field direction and to the $\pm\frac{1}{2}$ states from gamma rays incident parallel to the crystal field direction. If the relative strengths of the crystal bonds in these two directions are unequal, it is conceivable that the amount of recoilless absorption could be different for transitions to the $\frac{3}{2}$ and $\frac{1}{2}$ states, respectively. This interpretation is supported by measurements of the temperature dependence of the spectrum shape. It will be further studied in absorption measurements on single crystals of iron doped Al_2O_3.

The measured values of eqQ/h are plotted in Fig. 2 against the measured values of D. In several cases the sign of q was calculated using the point charge model and performing the appropriate lattice sum.[10] Knowledge of the sign of q and taking the sign of Q to be positive[11,12] enables the point in the figure to be placed in the appropriate quadrant. However, one must bear in mind the calculation of q is for the lattice without Fe^{3+} substituted and the interest here is q at Fe^{3+}. Even in the case with no Fe^{3+} present the result can be very inaccurate due to the sensitivity of the lattice sum to the charge distribution and charge position parameters. This might occur particularly in a complicated structure such as beryl. The addition of Fe^{3+} instead of the host atom, usually Al^{3+} here, will further cause the result to be more inaccurate. This has been observed previously,[13] particularly in hydrated compounds. With these restraining thoughts in mind the signs will be discussed.

The value of q in Al_2O_3 is calculated[10] and experimentally determined[14] to be positive and is similar to that found in[11] Fe_2O_3 so q at the Fe^{3+} site is undoubtedly positive. In yttrium iron garnet q at the Fe^{3+} site is negative[11] and it also is negative at the Ga^{3+} site in yttrium gallium garnet. Hence, it is undoubtedly negative for Fe^{3+} in the garnet. For the two hydrated compounds one calculates very small values for q at the Al^{3+} site which are in agreement with the observations of eqQ/h for Al,[15] but the values for Fe^{3+} are much larger due to local distortions of the H_2O groups.[13] Thus, one cannot predict the sign of q at Fe^{3+} in the hydrated compounds. For $BaTiO_3$ one calculates a negative value for q at the Ti^{4+} site and assumes Fe^{3+} has the same sign. Most of the contribution to q comes from point dipoles, is negative, and probably will not be affected too much by the presence of Fe^{3+} in place of Ti^{4+}. Calculations were carried out for q at the Ti^{4+} site in TiO_2 and many of its isomorphous compounds (ZnF_2, FeF_2, etc.). It was found that agreement between the calculations and various electron spin resonance or quadrupole resonance results could not be obtained with respect to either the magnitude of the splittings or the direction of principal axes. This may be due to the lack of good x-ray data. However, due to this lack of agreement and the further complication that the interest here is q at Fe^{3+} which has a different charge from Ti^{4+}, no prediction was made for TiO_2. Furthermore, no calculations were made for beryl since the lattice is quite complicated and the x-ray data are poor. No prediction was made for ZnO because of the lack of x-ray data on the atomic coordinates.[16]

Where the calculation of the sign of eqQ/h was felt to be unreliable, points on either side of the axis are shown connected by a light solid line. Similarly in the two cases where the sign of D is unknown, $BaTiO_3$ and $AlCl_3 \cdot 6H_2O$, positive and negative values of D are shown. In each case where an uncertainty in sign exists, the requirement that D be a relatively sensible function of eqQ/h suggests the choice of sign which is indicated by the solid circles and by which the heavy solid line is drawn. For instance, a negative sign for eqQ/h of TiO_2 would make it impossible to draw a curve for which D would be a single valued function of eqQ/h.

Figure 3 reproduces the theoretical calculations of Watanabe for the ^6S state ion Mn^{2+}.[2] The parameter, Δ, used by Watanabe is related to the usual crystal field

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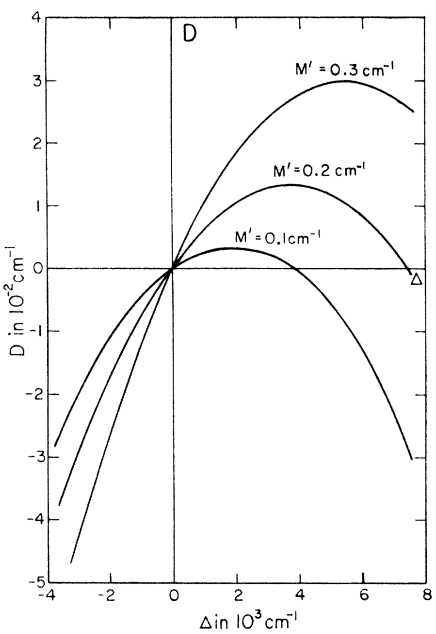

FIG. 3. Calculated values of D vs Δ of Mn^{2+} for several values of M' [taken from H. Watanabe (reference 2)].

parameter by the equation

$$\Delta=\tfrac{1}{4}(5/\pi)^{1/2}\langle r^2\rangle A_2{}^0=\tfrac{1}{4}\langle r^2\rangle q_{\mathrm{u}}. \tag{4}$$

The relation between D and Δ arises from two interactions. One suggested by Price[3] mixes states of the ${}^6D(3d^44s)$ configuration by the spin-spin interaction and can be represented schematically as $\langle {}^6S|W_{SS}|{}^6D\rangle \times\langle {}^6D|V_{\mathrm{ax}}|{}^6S\rangle$. The second interaction considered by Watanabe involves mixing into ${}^6S$ state some ${}^4P$ and ${}^4D$ wave functions through the interaction

$$\langle {}^6S|W_{LS}|{}^4P\rangle\langle {}^4P|V_{\mathrm{ax}}|{}^4D\rangle\langle {}^4D|V_{\mathrm{ax}}|{}^4P\rangle\langle {}^4P|W_{LS}|{}^6S\rangle.$$

These are all states of the d^5 configuration. The relative magnitudes of the two interactions could not be calculated accurately because of inadequate knowledge of the applicable wave functions. Hence, three representative curves are shown in Fig. 3, designated by the quantity M', which involves the overlap integral of $4s$ and $3d$ wave functions and is a measure of the strength of the interaction linear in V_{ax} [see Eq. (5) of discussion section].

DISCUSSION

Two observations are immediately evident from a comparison of the calculated and experimental curves relating eqQ/h and D. First, there is considerable scatter in the experimental data about any smooth curve that one might conceivably draw. Second, and of greater consequence, if one is to draw any curve, there is disagreement between theory and experiment in the functional dependence of D on V_{ax}.

Some scatter in the data might be expected. Effects can occur from the distortion of the wave functions of the paramagnetic ion by the presence of the various neighboring ions. D would appear to be particularly sensitive to the distortion of electron charge produced by neighboring ions as it is a function of the overlap integrals of the ground state and various excited state wave functions which have considerable radial extent. q, on the other hand, is dependent primarily on the position of the neighboring ions. Since these distortion effects will be different in the several crystals we have studied, there will be deviations from any simple theory.

In addition, small amounts of covalent bonding can lead to further scatter. Several calculations[17] in the past few years have demonstrated the importance of covalency in the estimate of the crystal field parameter $10\ Dq$. That some covalent bonding exists in our samples is suggested by the isomer shift data listed in Table I. From their analysis of early Mössbauer isomer shift data, Walker, Wertheim, and Jaccarino[7] concluded that a pure d^5 state of iron should have a shift of 0.57 mm/sec. The values for our samples deviate as much as 0.18 mm/sec from this value. The direction of deviation is such as to indicate fewer d electrons and more s electrons are present than in purely ionic compounds. It may be of interest that the largest deviations (Al_2O_3 and ZnO) occur in those samples having the greatest departure from the smooth curve drawn in Fig. 2.

Furthermore, effects specific to a particular lattice may be important. For instance, in TiO_2, $BaTiO_3$, and ZnO, Fe^{3+} is replacing either a Ti^{4+} or a Zn^{2+}. The difference in charge is probably compensated by vacancies in the case of the first two compounds and by the addition of an appropriate amount of Li^{1+} in the latter instance. These vacancies or the Li^{1+} could have some effects on our results. However, they are probably small as the electron resonance spectrum in each case had the symmetry characteristics of the host lattice and appeared to be uninfluenced by charge compensation effects. The use of larger amounts of Fe^{3+} in the Mössbauer measurements than in the electron resonance experiments was also found to be unimportant. This point was checked by comparing similar samples containing different amounts of substituted iron. In each case the same quadrupole splitting was observed indicating concentration effects were relatively small.

Of more importance than the distribution of data points about a smooth experimental curve is disagreement between theory and experiment in the functional dependence of D on V_{ax}. The experimental results can possibly be crudely fit by a dependence of D on V_{ax} and $V_{\mathrm{ax}}{}^2$ as suggested by Watanabe. However, the sign of the coefficient of $V_{\mathrm{ax}}{}^2$ must be positive and not negative as given by the theory. The calculations of Watanabe have been checked and found to be correct with respect to the sign of $V_{\mathrm{ax}}{}^2$ contribution. In addition, no interactions within the d^5 configuration other than that considered by Watanabe were found to contribute

[17] For a recent review paper and references to earlier work see S. Sugano, J. Appl. Phys. 33, 303S (1962).

to D.[18] It should, however, be pointed out that the choice of a positive sign for the coefficient of V_{ax}^2 depends only on one point, TiO_2. There is little doubt that experimentally the sign of D is, indeed, positive. However, TiO_2 is the only compound studied that had a measurable asymmetry parameter, E ($E=2.21$kMc/sec; $D=20.35$ kMc/sec).[19] The effect of this asymmetry on the measured value of eqQ should be small. It was taken into account in the tabulation of the data listed in the Table I using the relation

$$q'=q(1+\tfrac{1}{3}\eta^2)^{1/2}, \tag{4}$$

where q' is the apparent field gradient calculated from the observed splitting. η can be estimated from the relation, $\eta=3E/D$. As mentioned previously, D (and similarly, E) is probably more sensitive to the electronic charge distribution than q. Hence, the relation $\eta=3E/D$ is not strictly valid. However, it is unlikely that the small asymmetry of the axial crystal field in TiO_2 would give rise to a sign reversal in the relation between D and V_{ax}. If one ignores the point due to TiO_2, the best curve for D vs eqQ/h would be some sort of wide horizontal line.

A possible explanation for the divergence between theory and experiment is that there are some excited states in a configuration other than d^5 contributing to V_{ax}^2. However, because of the high excitation energy of these states, it is unlikely that any possible contribution would be significant. Thus, it appears that the divergence must be attributed to the failure of the simple crystal field theory used by Watanabe for this calculation. Further evidence for this conclusion is found in the comparison of the theoretical and experimental dependence of the slope (at $D=0$) of the D vs Δ curve on the particular paramagnetic ion (i.e., on whether the substituted ion is Fe^{3+} or Mn^{2+}). While the magnitude of the slope cannot be accurately calculated, its dependence on species should be reproduced by theory. In actual fact it is not.

Experimental values of $(dD/d\Delta)_0$ can be found from data of D and eqQ/h for Mn^{2+} and Fe^{3+} and using a linear extrapolation of the D vs Δ curve to the origin. Folen[20] has observed the effect of an electric field gradient on the hyperfine spectra of Mn^{2+} in Al_2O_3 and finds a value of 0.7 ± 0.2 G for the quantity $Q'=3eqQ/4I(2I-1)$. Using appropriate values for Q and I[21] and Eqs. (3) and (4), Δ is calculated to be $+39\times10^2$ cm^{-1}. A value of D of 0.0194 cm^{-1} has been obtained by Low and Suss[22] for this same material. Using a linear extrapolation to zero of the D vs Δ curve, a value of $(dD/d\Delta)_0$ of 5×10^{-6} is obtained in reasonable agreement with Watanabe's theoretical estimates. However, for Fe^{3+} a value of $(dD/d\Delta)_0$ is obtained from Fig. 3 that is 20 times the above value for Mn^{2+}. The simple ionic model of Watanabe indicates $(dD/d\Delta)_0$ is proportional to a quantity, $M'\Delta'/E\Delta$. Here E is the energy separation between the ground state and the excited $^6D(3d^44s)$ state and

$$M'=\beta^2\left[3\int_0^\infty (1/r)\psi_{3d}(r)\psi_{4s}(r)\int_0^r r'^2\psi_{3d}^2(r')dr'dr - 2\int_0^\infty (1/r^3)\psi_{3d}^2(r)\int_0^r r'^4\psi_{3d}(r')\psi_{4s}(r')dr'dr\right], \tag{5}$$

$$\Delta'=\left(\frac{5}{\pi}\right)^{1/2}A_2^0\int_0^\infty r^4\psi_{3d}(r)\psi_{4s}(r)dr, \tag{6}$$

and

$$\Delta=\frac{1}{4}\left(\frac{5}{\pi}\right)^{1/2}A_2^0\int_0^\infty r^4\psi_{3d}^2(r)dr. \tag{7}$$

Using Slater wave functions[23] for ψ_{3d} and ψ_{4s}, the quantity $M'\Delta'/\Delta$ is calculated to be 15% larger for Mn^{2+} than for Fe^{3+}. To obtain an estimate of E, the known energies[24] of corresponding excited states of Mn^{2+} and Fe^{3+} were compared. In all cases the excited states of Mn^{2+} lie lower, the amount varying from 16% for the first excited state to 41% for the highest known excited state. Hence, E should be at least 16% smaller for Mn^{2+}. From these estimates of $M'\Delta'/\Delta$ and E, it is clear that the theoretical ratio $(dD/d\Delta)_0(\mathrm{Fe})/(dD/d\Delta)_0(\mathrm{Mn})$ is considerably smaller than unity and not 20 as observed experimentally.

Since the writing of this paper, an article by Germanier, Gainon, and Lacroix[25] has come to our attention. This article is also concerned with the effect of the axial part of the crystal field on the $^6S_{5/2}$ state ion, Fe^{3+}. In particular, they report: (a) a measurement of the dependence of the D term in the spin Hamiltonian of Mn^{2+} and Fe^{3+} under uniaxial stress, (b) a theoretical calculation of two additional terms, each proportional to V_{ax}, that contribute to D and a recalculation of the two terms considered by Watanabe, and finally, (c) a comparison of D with a calculated axial contribution of the crystal field potential (calculated by performing the appropriate lattice sum) for several crystals. Their calculation of the sign of the coefficient of V_{ax}^2 is in disagreement with Watanabe's and our calculations. Moreover, the previously mentioned difficulties encountered in our calculation of the sign of the lattice sum in a particular crystal indicate that the calculation of the magnitude of V_{ax} can be quite unreliable. For example, in the lattice sum calculations of Germanier *et al.* V_{ax} for beryl is calculated to be about 10 times that of TiO_2, $BaTiO_3$, and Al_2O_3. However, as can be

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[25] A. M. Germanier, D. Gainon, and R. Lacroix, Phys. Letters **2**, 105 (1962).

seen by the results reported here, V_{ax} at an Fe^{3+} site in beryl is considerably smaller than for the other three crystals.

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Dielectric Constant of a Superconductor*

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The longitudinal dielectric constant has been calculated for the extended Bardeen-Cooper-Schrieffer model of a superconductor. An explicit formula is obtained which has been evaluated numerically. The competition between collective and single-particle effects is pronounced, so that the dielectric function differs remarkably from the most elementary approximations to it. However, the dielectric function of the superconductor does not differ greatly from that in the normal metal in either the high-frequency or static limit, regardless of the wavelength. This prevents the modifications due to superconductivity from being readily observed. In particular, the shift in the static polarizability should cause very small shifts in the phonon speed, so small that no effect on the lattice specific heat should be observed.

I. INTRODUCTION

THE dielectric function of a metal is of fundamental interest not only for its direct experimental relevance, but because of its close relationship to the correlation structure and excitation system of the metal. In a "normal" metal, the dielectric function is believed to be determined primarily by the single-particle excitations. In a superconductor, on the other hand, there exists a strong competition between the single-particle and collective effects, especially in the longitudinal dielectric function. We have studied the longitudinal dielectric function of a superconductor, in order to exhibit these collective effects, as well as to clarify some points which we shall mention below.

Anderson[1] and Rickayzen[2] have previously studied the dielectric function, and they have obtained the fundamental equations. More recently, Nishiyama[3] has discussed limiting cases of these equations. We have obtained numerical values for the dielectric function, and thus are in a position to discuss the dielectric function quite generally.

An elementary approximation to the dielectric function is obtained by restricting the possible excitations to those of two quasi-particles. In the normal case, this leads to the Lindhard[4] formula. We shall consider the irreducible polarization part, rather than the dielectric function. The wave number and frequency-dependent dielectric function is related to the irreducible polarization part, $N\Lambda(q,\omega)$, by

$$\epsilon(q,\omega)=1+v(q)N\Lambda(q,\omega), \tag{1}$$

where $v(q)$ is the Fourier transform of the Coulomb potential,

$$v(q)=4\pi e^2/q^2=3(\omega_p/v_0q)^2/N. \tag{2}$$

We have introduced the notation ω_p for the classical frequency, $\omega_p^2=4\pi ne^2/m$, and v_0 for the Fermi velocity. The density of states at the Fermi surface is denoted by $N=mp_0/\pi^2$. The irreducible polarization part is slightly more convenient to discuss theoretically than the dielectric constant itself. The function $\Lambda(q,\omega)$ is to a first approximation independent of the coupling. It depends weakly on the Coulomb potential $v(q)$, being much more strongly influenced by the "average" attractive force.

It, of course, should be realized, that ω_p/v_0q is very large for wavelengths of interest, namely, wavelengths such that $v_0q\lesssim k_B\Theta$ for $\Theta\lesssim 100°K$. As a result of this large, direct Coulomb coupling, great experimental difficulties stand in the way of a direct observation of the dielectric constant.

The polarization part is given by the sum of all irreducible graphs shown in Fig. 1. An irreducible graph is one which cannot be separated into two pieces by

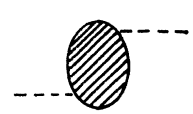

FIG. 1. General structure of the polarization part. The shaded area represents the sum of all graphs which cannot be separated into two disjoint graphs by breaking a single Coulomb line.

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