

acting electron gas. The $q=0$ roots at $\omega = m\omega_C$ found from these equations are actually the zero-sound modes $\omega = \gamma_n m \omega_C$ (for $n > 1, n > |m|$) which all coalesce when $\gamma_n = 1$ for all values of n . This can be seen by substituting back into Eq. (11) and noting that Γ_n^m vanishes for these modes⁷; thus there is no electric field or current density associated with them at $q=0$. When all the A_n are finite, the solution for $n=1$ is the transverse or longitudinal plasma frequency. The "plasma waves" observed by Walsh and Platzman⁴ are predominantly zero-sound waves with $m=1$ for small values of X . For a given m , the "zero-sound" wave with the smallest possible value of n is most strongly coupled to an electric field for long wavelengths. Thus for $m=1$, the $n=2$ mode is most strongly excited for polarization parallel to the dc field, and the $n=3$ mode is for the other polarization.

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²The notation used in this note differs slightly from that of Ref. 1. In particular, our coefficient A_n corresponds to Silin's $A_n/(2n+1)$.

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⁵S. C. Ying and J. J. Quinn, to be published.

⁶Silin does not make use of the equation of continuity to eliminate the $n=0, m=0$ harmonic from the secular equation.

⁷It should be noted that, despite the rather complicated definitions [Eq. (10)] of Γ_n^m , these functions turn out to be identical to the spherical harmonics used by Silin.

TRANSFERRED HYPERFINE INTERACTION OF ISOELECTRONIC Yb³⁺ AND Tm²⁺ IN CaF₂

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ENDOR measurements of the ¹⁹F nuclei in the first four shells in CaF₂ containing Yb³⁺ ions in the cubic site are reported. The results are compared with those of isoelectronic Tm²⁺ ions.

Transferred hyperfine interactions in rare earths have been measured so far only in a few cases.^{1,2} The interpretation of these results is, to date, not very conclusive because the various different contributions such as overlap, covalency, dipolar interaction, and configuration interaction are difficult to evaluate. Some light could be shed on this problem if isoelectronic systems would be studied, in which the paramagnetic ion is contained in the same crystal host. This possibility exists for the alkaline earth fluorides such as CaF₂, SrF₂, or BaF₂. It has been shown by numerous authors that stable divalent rare-earth ions can be produced in this crystal host,³ where the rare-earth ions take the place of the cation and preserve the cubic symmetry. On the

other hand, trivalent rare-earth ions can be substituted for the calcium ion. A fraction of these ions, which depends of the nature of the ion—the manner in which the crystals were grown and the subsequent heat treatment—can be substituted at the cubic site with charge compensation far removed.

We have studied the ENDOR fluorine spectrum of the first four shells in the single crystal of CaF₂ containing approximately 0.1% of Yb³⁺ ions. The results are contained in Table I. They are to be compared with Bessent and Hayes' measurements on Tm²⁺ in CaF₂.² Also listed in Table I are the g factors, the cubic crystal-field parameters as determined from the optical spectra,^{4,5} and the deviation of the g factor from the

theoretical values. The ENDOR spectrum can be fitted to a simple dipolar form of a Hamiltonian:

$$\mathcal{H}_{\mathbf{F}} = [A_S + A_p (3 \cos^2 \theta - 1)] S_z I_z^{\mathbf{F}} + 3A_p \sin \theta \cos \theta S_z I_x^{\mathbf{F}} - g_{\mathbf{F}} \beta_n H_z I_z^{\mathbf{F}};$$

$$S = \frac{1}{2}, \quad I^{\mathbf{F}} = \frac{1}{2}.$$

The relative signs of the isotropic and anisotropic parts A_S and A_p are found to be same. The absolute sign cannot be determined. If the sign is negative, the subtraction of the positive dipolar energy A_D would give a very large negative value. The second, third, and probably even the fourth shells have values of A_p which are very close to those of A_D and at the same time have a finite value of A_S with the same sign as A_p . We are, therefore, inclined to assume that both A_S and A_p have a positive sign, that is to say, a spin direction parallel to that of the rare-earth ion. This is opposite to what has been found for Eu^{2+} in CaF_2 .¹

A complete theory of these isoelectronic spectra would have to account for the following differences of the $\text{CaF}_2:\text{Yb}^{3+}$ and $\text{CaF}_2:\text{Tm}^{2+}$ spectra: (a) a larger value of the anisotropic part of the transferred hyperfine interaction A_p in the first three shells for Yb^{3+} ; (b) a smaller value of the isotropic part A_S at least for the first shell; (c) small but finite value of A_S for the second, third, and probably the fourth shells; (d) significantly larger values of the cubic crystal field parameters; and (e) slightly larger deviations of the measured g factor from that computed on the

basis of static-crystal-field considerations. This deviation is expressed as an orbital reduction factor $1-k$.

The increase of the cubic crystal-field parameters, the increase of the anisotropic part A_p , and the spatial extent of the isotropic part A_S (this cannot be compared with Tm^{2+} since the accuracy in these measurements is not sufficient) are consistent both with a smaller $\text{Yb}^{3+}-\text{F}^-$ distance caused by polarization of the surroundings by the excess charge on Yb^{3+} and also with an increase in covalency and overlap. It is difficult to disentangle these two effects. A point-charge model with a distance reduced by about 3%-4% of the internuclear distance of $\text{Ca}^{2+}-\text{F}^-$ would account for the increased crystal field and in part for the increased value of A_p . The existence and extent of the isotropic part A_S and the large value of A_p suggest covalency and overlap effects. These two effects, however, do not explain why A_S is smaller for Yb^{3+} compared with that of Tm^{2+} . Apparently the contracted wave function and the reduced overlap may be more important and overcome the effect of contracted internuclear distance.

There have been a number of theories regarding the origin of the isotropic transferred hyperfine interaction.⁶⁻⁸ It is generally conceded that the $4f$ overlap and covalency with the $2s$ and $2p_{\sigma}$ orbitals of the F^- ion would give rise to a positive but small value for A_S . However, a reduced value of A_S because of smaller overlap would also mean smaller values of crystal field parameters, contrary to the experimental results. Watson and Freeman⁷ have calculated the effects of exchange polarization and found a large negative

Table I. ENDOR fluorine parameters for Yb^{3+} and Tm^{2+} in CaF_2 cubic sites. The values of A_D are calculated by assuming that the $\text{Yb}^{3+}-\text{F}^-$ distances are the same as $\text{Tm}^{2+}-\text{F}^-$ distances. The values of b_4^0 and b_6^0 have been obtained from the absorption and fluorescence spectra. A full discussion is to appear in the forthcoming Racah Memorial Volume (to be published). The ENDOR spectrum of the first shell with somewhat reduced accuracy has also been obtained by U. Ranon and J. S. Hyde, Phys. Rev. 141, 259 (1966). The ENDOR results on Tm^{2+} are from Ref. 2.

Shells	Yb^{3+}			Tm^{2+}		
	A_S (Mc/sec)	A_p (Mc/sec)	A_D (Mc/sec)	A_S (Mc/sec)	A_p (Mc/sec)	A_D (Mc/sec)
1	1.667 ± 0.01	17.567 ± 0.01	9.783	2.584 ± 0.01	12.283 ± 0.01	9.809
2	0.015 ± 0.005	1.418 ± 0.005	1.393	0.010 ± 0.01	1.386 ± 0.005	1.397
3	0.006 ± 0.005	0.622 ± 0.005	0.613	0.00 ± 0.01	0.612 ± 0.005	0.615
4	0.005 ± 0.005	0.362 ± 0.005	0.362	0.00 ± 0.01	0.367 ± 0.001	0.363
		$g = 3.443 \pm 0.002$			$g = 3.452 \pm 0.002$	
		$b_4^0 = 51.6 \text{ cm}^{-1}$			$b_4^0 = 45.8 \text{ cm}^{-1}$	
		$b_6^0 = 6.39 \text{ cm}^{-1}$			$b_6^0 = 5.05 \text{ cm}^{-1}$	
		$1-k = 0.02 \pm 0.005$			$1-k = 0.01 \pm 0.005$	

value of A_S . Lewis et al.⁸ have also shown that configuration interaction, in which an electron is promoted to a $6s$, $6p$, or $6d$ shell, also gives rise to a negative value for A_S and is consistent with O^{17} NMR shifts in aqueous solutions of rare-earth ions. It is difficult to reconcile these theories with the experimental results listed above even if a negative value for A_S and A_p is assumed. Any decrease in the absolute value of A_S should be accompanied with a decrease of the crystal field parameters. A partial resolution of this problem could be obtained if matrix elements of the type $\langle 5p | h | 4f \rangle$ might be larger for Yb^{3+} . This may give rise to a larger crystal-field splitting, affecting the $t_{1\mu}$ levels (but not the ground state), and possibly an increased anisotropic exchange of the $5p$ shell mixing with $2p_\sigma$ and $2p_\pi$ resulting in a large negative value of Δ_p without effecting the smaller value of A_S .

Further work on other isoelectronic ions of rare-earth ions both in cubic and other symmetries is in progress.

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EXPERIMENTAL LIMITS ON THE EXISTENCE OF ANOMALOUS ELECTRONS*

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Experimental and theoretical evidence is presented against the existence of particles which differ from electrons by a conserved internal quantum number.

We wish to consider the possibility of the existence of anomalous electrons and positrons (denoted by e'^- and e'^+ , respectively) which differ from ordinary positrons and electrons by an unspecified conserved internal quantum number. A scheme in which this quantum number is strangeness has been proposed¹ recently to explain the apparent absence of neutral lepton currents. We shall discuss several experiments which can be used to set limits on the abundance of e'^{\pm} in matter and report on the results of one such experiment that we have recently carried out.

It is useful to begin by enumerating some of the properties of e'^{\pm} . We assume that at low energies e'^{\pm} have the same electromagnetic interactions as do e^{\pm} .² Conservation of lepton number requires that e'^{\pm} be stable, which implies that e'^{\pm} may be constituents of ordinary matter along with e^{\pm} . By virtue of the Pauli exclusion principle, the presence of e'^- will be most notice-

able in those systems where the low-lying energy levels are already occupied by ordinary electrons. The existence of an additional internal quantum number would give rise to a set of new elements in which each closed shell could have up to $2n$ particles, n electrons and n anomalous electrons. Elements with completely closed shells, such as $({}^4\text{Be}^0)'$, with the electronic configuration $1s^2 1s'^2$ would behave as inert gases and should be present in the atmosphere in trace amounts. We shall return shortly to discuss the results of a mass spectrometric search for Be' that we have recently completed. In stellar systems the presence of anomalous electrons could be detected, at least in principle, by the observation of white dwarfs more massive than about 1.4 solar masses.³

Direct evidence for the presence of e'^+ would be the observation of molecules in which a hydrogenic proton is replaced by an anomalous positron. Since the process $e'^+ + e^- \rightarrow \text{photons}$ is for-