

rected for the change in Boltzmann factor, is the same as that at room temperature in the tetragonal phase. Resonance has also been observed in the orthorhombic and rhombohedral phases.

<sup>1</sup> W. Low and D. Shaltiel, Phys. Rev. Lett. **1**, 51 (1958).

<sup>2</sup> Hornig, Rempel, and Weaver, J. Phys. Chem. Solids (to be published).

<sup>3</sup> L. M. Matarrese and C. Kikuchi, J. Phys. Chem. Solids **1**, 117 (1956); W. Low, Phys. Rev. **105**, 793 (1957).

<sup>4</sup> Because of the trisoctahedral symmetry of the nearest-neighbor oxygens at the barium site, in contrast to the octahedral symmetry at the titanium site, the sign of the cubic field is reversed. As Professor E. T. Jaynes originally pointed out (private communication), under these conditions it would be possible to see  $\text{Fe}^{2+}$ ,  $S=2$ , at room temperature. However, the existence of five main transitions eliminates this ion as a possibility.

### ELECTRON PARAMAGNETIC RESONANCE IN $\text{BaTiO}_3$ †

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In the preceding Letter, Hornig *et al.*<sup>1</sup> have reported detailed measurements of electron paramagnetic resonance (EPR) spectra in  $\text{BaTiO}_3$  which differ considerably from those reported by us.<sup>2</sup> Further work has revealed a number of points in agreement with those reported by Hornig *et al.* and a few in disagreement.

(a) In agreement with Hornig *et al.*, we have detected additional lines which make the spectrum compatible with a spin  $S=5/2$ . We have observed in addition so-called forbidden transitions corresponding to  $\Delta M = \pm 2, \pm 3$  and probably  $\pm 4$ . Our assignment of a spin of  $3/2$  in reference 2 is, therefore, in error. The spectrum could conceivably be assigned to  $\text{Fe}^{3+}$ .

(b) In order to verify that the EPR spectrum is caused by a  $\text{Fe}^{3+}$  impurity we have compared

the spectra of so-called pure crystals with those of crystals intentionally doped with  $\text{Fe}_2\text{O}_3$  (0.2-0.5 mole percent added to melt). The same spectrum was observed. The doped crystals showed a more intense spectrum, but not by a large factor, compared with the pure crystals obtained from many different sources. If the spectrum is assigned to trivalent iron, these results indicate that some of the iron may not be situated as trivalent iron at the titanium lattice sites.

(c) We have examined a number of *a*-domain crystals. In all those which were carefully prepared the EPR spectrum was a factor of ten less intense than that of *c*-domain crystals. In several particular cases the spectrum could not even be detected. The EPR spectrum may, therefore, be a sensitive indicator of the mixture of *a* and *c* domains present in  $\text{BaTiO}_3$ .

(d) We want to comment on reference 4 in the preceding letter. We have made detailed calculation of the energy level scheme of divalent iron in octahedral, trisoctahedral, tetrahedral, and axial symmetries taking into account spin-orbit coupling to the second order.<sup>3</sup> In the trisoctahedral field, the lowest level  $\Gamma_3$  splits under the action of spin-orbit coupling in the second order into two singlets, two triplets, and one doublet. The splitting of these levels, for any reasonable crystal field, is far too large to be detected at 3-cm wavelength, and to give a spin of 2 at room or lower temperatures. We have also calculated the energy level scheme of  $\text{Ti}^{3+}$  situated in a hypothetically dominant trisoctahedral symmetry. Transitions between the two nonmagnetic doublets are forbidden and a spectrum corresponding to a spin of  $3/2$  is unlikely. At any rate it is now ruled out by the results observed by Hornig *et al.* and by us.

Investigations are in progress in which the EPR spectra of  $\text{BaTiO}_3$  are observed while ac or dc electric fields are applied to the crystals while situated inside the cavity.

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<sup>1</sup> Hornig, Rempel, and Weaver, preceding Letter [Phys. Rev. Lett. **1**, 284(1958)].

<sup>2</sup> W. Low and D. Shaltiel, Phys. Rev. Lett. **1**, 51 (1958).

<sup>3</sup> W. Low and M. Weger (to be published).