We have attempted to estimate the value of G by employing Gladney's wave functions,[2] combined with a nearest-neighbor point-charge model of the crystal field, and the assumption that in CoF_2 the electronic excitations associated with the Co^{++} ion can be treated as completely localized to the Co site. As remarked earlier, this calculation produces a value of $|G|$ some six or seven times smaller than the value 0.09 estimated from the experimental data. Before our work, Moriya and coworkers also found a value of $|G|$ far smaller than that required to fit the data, on the basis of a similar computation.[10] Thus, it appears as if one cannot apply Gladney's wave function for Co-doped MgF_2 directly to CoF_2 without modification. Since our crystal-field calculation is long and tedious, but employs only well-known straightforward techniques, we omit the details of the calculation from the present note.

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[6] We use a convention where the ground state is labeled with $s_z = +\frac{1}{2}$ and the excited state with $s_z = -\frac{1}{2}$.

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COMMENTS AND ADDENDA

The Comments and Addenda section is for short communications which are not of such urgency as to justify publication in Physical Review Letters and are not appropriate for regular Articles. It includes only the following types of communications: (1) comments on papers previously published in The Physical Review or Physical Review Letters; (2) addenda to papers previously published in The Physical Review or Physical Review Letters, in which the additional information can be presented without the need for writing a complete article. Manuscripts intended for this section may be accompanied by a brief abstract for information-retrieval purposes. Accepted manuscripts will follow the same publication schedule as articles in this journal, and galleys will be sent to authors.

Comment on Determination of Nuclear Quadrupole Interactions: Copper-Doped Rutile*

H. So and R. Linn Belford

Noyes Chemical Laboratory and Materials Research Laboratory, University of Illinois, Urbana, Illinois 61801

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A complete diagonalization of the spin Hamiltonian is necessary to obtain consistent principal values of the nuclear quadrupole coupling tensor from $\Delta M_I = 0$ and $\Delta M_I = \pm 2$ lines in EPR spectra. As an example, proper recalculation for $TiO_2 : Cu^{++}$ shows greatly revised values, $Q' \approx 0.0010\ cm^{-1}$, $\eta \approx 0.1$.

In the extraction of quadrupole coupling constants for Cu^{++} and other paramagnetic ions from orientation-dependent allowed and forbidden hyperfine transitions,[1] we have been struck by the inadequacies of the second-order perturbation expressions for positions of certain of the lines in some ranges of orientation of the magnetic field.[2] As Lyons and Kedzie[3] have already demonstrated, the perturbation expressions are inadequate for the forbidden hyperfine lines with $\Delta M_I = \pm 1$ when the magnetic field lies between the principal axis and its perpendicular. Here we point out that even for magnetic field perpendicular to the principal axis of a uniaxial or near-uniaxial site, the perturbation-theory

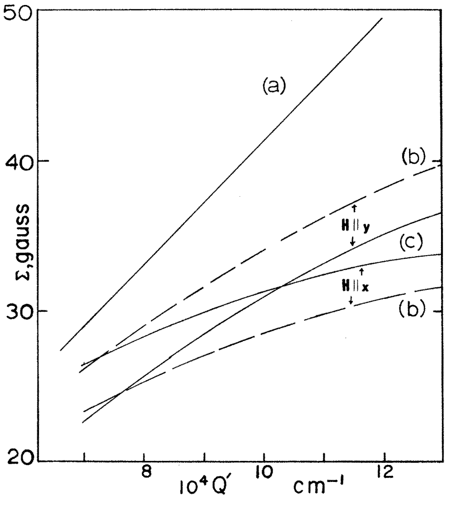

FIG. 1. Computed values of $\Sigma \equiv |a_i^2 - b_i^2| + |c_i^2 - d_i^2|$ for $i=x$ or y (i.e., the sum of the separations in the high-field and low-field pairs of $\Delta M_I = \pm 2$ lines; notation as in Ref. 4). (a) Calculated by the perturbation expressions (5.1c) of Ref. 4, $\Sigma = 4Q'h\nu/g_i\beta$; (b) computer diagonalization of $\mathcal{H}_{\text{spin}} = \beta\vec{H}\cdot\overleftrightarrow{g}\cdot\vec{S} + \vec{I}\cdot\overleftrightarrow{A}\cdot\vec{S} + \vec{I}\cdot\overleftrightarrow{P}\cdot\vec{I} - g_N\beta_N\vec{I}\cdot\vec{H}$, with $g_z = 2.346$, $g_x = 2.109$, $g_y = 2.094$, $A_z = -0.00875$, $A_x = 0.00187$, $A_y = 0.00272$ cm^{-1}, $Q' = 1.5P_z$, $\text{Tr}\,\overleftrightarrow{P} = 0$, $P_x = P_y$; (c) same as (b) except $\eta \equiv (P_y - P_x)/P_z = 0.87 \times 10^{-4}$ cm$^{-1}/Q'$. The experimental values of Σ for $TiO_2:Cu^{++}$ were in the vicinity of 30 G. See Ref. 4.

expressions are inadequate for certain transitions and that this inadequacy has severe consequences in the determination of quadrupole coupling parameters from EPR spectra.

An interesting case in point is provided by $TiO_2:Cu^{++}$, recently reported by Ensign, Chang, and Kahn.[4] They measured EPR spectra of ^{63}Cu and ^{65}Cu (natural-abundance mixture) doped into rutile and used the $\Delta M_I = \pm 2$ lines for $\vec{H}$ along the x and y axes, together with the perturbation expressions of Bleaney *et al*,[5] to extract a quadrupole tensor for ^{63}Cu: $Q' = 7.42 \pm 0.24 \times 10^{-4}$ cm^{-1}, $\eta = 0.03 \pm 0.03$. They also found that the results obtained from the $\Delta M_I = 0$ lines were not consistent with those from the $\Delta M_I = \pm 2$ lines. Applying their Eq. (5.1a) for the $\Delta M_I = 0$ lines in their x and y spectra, we obtain $Q' \approx 10.2 \times 10^{-4}$ cm^{-1} and $\eta \approx 0.15$.

We have examined the perturbation equations (5.1a) and (5.1c) of Ensign *et al*,[4] in comparison with exact 8×8 matrix-diagonalization expressions for the same quantities. Figure 1 shows a sample plot. Clearly, the perturbation expressions are apt to yield a quadrupole coupling tensor grossly in error.

Upon fitting the $TiO_2:Cu^{++}$ spectra published by Ensign *et al.*[4] by a complete diagonalization of the spin Hamiltonian, we obtained values of $Q' = 9.7 \times 10^{-4}$ cm^{-1} and $\eta = 0.09$ from the $\Delta M_I = \pm 2$ lines. These values are close to those obtained from the $\Delta M_I = 0$ lines. Considering all the data and errors, we recommend $Q' = +10 \times 10^{-4}$ cm^{-1} and $\eta = 0.1$. These values, moreover, are in the range of expected values[1,4] for cupric ion in a slightly distorted octahedral site.

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ERRATA

Theory of the Nuclear Magnetic Resonance Chemical Shift of Xe in Xenon Gas, Frank J. Adrian [Phys. Rev. 136, A980 (1964)]. In the discussion following Eq. (33) there is a misprint in the statement "$(S_{\sigma\sigma} + S_{\pi\pi})^2 = 0.061$ at $R = 4$ Å." This should have read "$(S_{\sigma\sigma} + S_{\pi\pi})^2 = 0.0061$ at $R = 4$ Å." This misprint does not affect any other result or conclusion, and the value of $(S_{\sigma\sigma} + S_{\pi\pi})^2$ as a function of R is correctly given at the top of p. A987. The author is indebted to Dr. A. K. Jameson and Dr. C. J. Jameson for calling this misprint to his attention.

Spin Correlation and Entropy, H. Falk and Masuo Suzuki [Phys. Rev. B 1, 3051 (1970)]. Several typographical errors (which do not propagate to