

Brief Reports

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Analysis of hyperfine interactions in alkaline-earth oxides

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We analyze the hyperfine interactions between the trapped hole spin at a *V*-type center and a nearby nucleus in magnetism oxide and show that the leading correction for the size of the hole wave function involves its dipole moment, and that the hyperfine interaction is relatively insensitive to the size of the quadrupole moment of the spin density.

The most successful description of the measured magnetic hyperfine interaction between the trapped hole at a *V*-type center in alkaline-earth oxides¹ and a nearby nucleus is provided by the model of Wertz, Auzins, Griffiths, and Orton² in which the hole is localized on a single oxygen ion neighboring the cation defect. The simplest version of this model, in which the spatial distribution of the trapped hole spin density is neglected, was sufficient to describe the early experiments in which the nucleus of the substituted cation³ takes part in the hyperfine interaction. Recently, Unruh, Chen, and Abraham⁴ doped magnesium oxide crystals with ²⁵Mg, and measured the hyperfine interactions between trapped hole *V*-type centers and ²⁵Mg nuclei in a variety of nearby positions. We are proposing a simple refinement of the model of Wertz *et al.* to take account of the spatial distribution of the trapped hole in a manner consistent with the experimental results of Unruh *et al.*⁴

Our model is constructed using the following assumptions: (1) that the hole is localized in an *s-p* hybridized orbital on a single oxygen ion neighboring the magnesium vacancy; (2) that the axis of rotational symmetry of the orbital is directed towards the magnesium vacancy;⁵ (3) that the contact interaction is isotropic; and (4) that the traceless part of the magnetic hyperfine interaction arises from the classical dipole-dipole interaction.

For this model, the magnetic dipole-dipole interaction between the trapped hole spin, in a state with orbital wave function $\psi(\vec{r})$ and a nuclear spin *I* at an arbitrary point \vec{R} in the *x-z* plane, is

$$A_{\alpha\beta} = \frac{\mu_e \mu_n}{SI} \int d^3r |\psi(\vec{r})|^2 \left[3 \frac{(R_\alpha - r_\alpha)(R_\beta - r_\beta)}{|\vec{R} - \vec{r}|^5} - \frac{\delta_{\alpha\beta}}{|\vec{R} - \vec{r}|^3} \right], \tag{1}$$

where μ_e and μ_n are the magnetic moments of the hole and nucleus, respectively. This expression is most readily evaluated using the angular momentum representation of dipole-dipole coupling,⁶ which gives $A_{\alpha\beta}$ in terms of

$$\tilde{A}_M = \int d^3r |\psi(\vec{r})|^2 \frac{Y_{2M}(\vec{R} - \vec{r})}{|\vec{R} - \vec{r}|^3}. \tag{2}$$

Using the two-center expansion,⁷ and assuming that $r < R$, we find that

$$\frac{Y_{2M}(\vec{R} - \vec{r})}{|\vec{R} - \vec{r}|^3} = \frac{4\pi}{3} \sum_{l,m,m'} C_M(l,m,m')(2l+3) \times \frac{r^l}{R^{l+3}} Y_{l+2,m}(\vec{R}) Y_{lm}(\vec{r}), \tag{3}$$

where

$$C_M(l,m,m') = \int d\Omega Y_{2M}(\vec{r}) Y_{lm}(\vec{r}) Y_{l+2,m}(\vec{r})$$

is a Gaunt coefficient. Substituting this expression into Eq. (2) gives

$$\tilde{A}_M = \frac{4\pi}{3} \sum_{l,m,m'} C_M(l,m,m')(2l+3) \frac{Y_{l+2,m}(\vec{R})}{R^{l+3}} q_{lm}, \tag{4}$$

where

$$q_{lm} = \int d^3r r^l Y_{lm}(\vec{r}) |\psi(\vec{r})|^2.$$

As the hole state is an hybridized *s-p_z* orbital, the only nonzero values of q_{lm} are $q_{00} = 1/\sqrt{4\pi}$, $q_{10} = p\sqrt{3}/4\pi$, and $q_{20} = Q\sqrt{5}/16\pi$, where *p* and *Q* are the dipole and quadrupole moments of the hole spin distribution. We note that as $\psi(\vec{r})$ is an hybridized *s-p_z* orbital, which may be written as $\psi(\vec{r}) = f_s(r) + f_p(r) Y_{10}(\vec{r})$, then the quadrupole moment is $Q = \int dr r^4 f_p(r)^2/5$, and must be positive.

When \vec{R} lies in the *x-z* plane, there are only three independent nonzero components of $A_{\alpha\beta}$:

$$A_{zz} = \frac{2\mu_e \mu_n}{SI} \left[\frac{P_2(\cos\theta)}{R^3} + 3p \frac{P_3(\cos\theta)}{R^4} + 3Q \frac{P_4(\cos\theta)}{R^5} \right], \tag{5a}$$

$$A_{xz} = -\frac{\mu_e \mu_n}{SI} \left[\frac{P_2^1(\cos\theta)}{R^3} + 2p \frac{P_3^1(\cos\theta)}{R^4} + \frac{3}{2} Q \frac{P_4^1(\cos\theta)}{R^5} \right], \tag{5b}$$

and

$$A_{xx} - A_{yy} = \frac{\mu_e \mu_n}{SI} \left(\frac{P_2^2(\cos\theta)}{R^3} + p \frac{P_3^1(\cos\theta)}{R^4} + \frac{1}{2} Q \frac{P_4^1(\cos\theta)}{R^5} \right), \quad (5c)$$

where θ is the polar angle of \vec{R} and $P^m(x)$ an associated Legendre function.⁸ For equatorially placed ²⁵Mg nuclei, those for which $\theta \cong 90^\circ$, Unruh *et al.*⁴ found that $A_{xx} - A_{yy} = -0.7298$ MHz, $A_{zz} = 0.0248$ MHz, and $(2A_{zz} - A_{xx} - A_{yy})/3 = 0.2517$ MHz. The value of $\mu_e \mu_n / hSI$ is⁴ -4.843 MHz \AA^3 .

In Eqs. (5a)–(5c) there are four unknown quantities: R , θ , p , and Q . One of them must be specified in order that the other three might be determined from the experimental data. If, following Schirmer,⁹ we assume that $p = 0$, we find that $Q = -0.2944$ \AA^2 . However, as noted earlier, Q must be positive so that neglecting the dipole moment of the hole spin distribution is clearly unphysical.

If, on the other hand, we assign $Q = 0$, the solution to Eqs. (5a)–(5c), consistent with the data of Unruh *et al.*, is $R = 2.71$ \AA , $\theta = 94.9^\circ$, and $p = -0.0726$ \AA . The sign of the hole spin-density dipole moment may appear surprising, implying as it does that an extra electron added to the ion would have an electric dipole moment pointing away from the effectively negative vacancy. However, this result is consistent with the spin-density calculated using the wave functions of Wilson and Wood¹⁰ for an O^- ion neighboring a cation vacancy in MgO. The magnitude of the spin-

density dipole moment does not agree as well with the result calculated using the wave functions of Wilson and Wood, as they find $p = -0.0366$ \AA . The value of Q calculated using the wave function of Wilson and Wood is 0.136 \AA^2 ; when we use this value to solve Eqs. (5a)–(5c) the solution is $R = 2.70$ \AA , $\theta = 95.9^\circ$, and $p = -0.0874$ \AA .

It is clear that the equatorial nearest-neighbor distance deduced from the experiments⁴ of Unruh *et al.* is not very sensitive to the assumed size of the hole spin-density quadrupole moment, remaining at about 2.7 \AA , 29% longer than the nearest-neighbor distance in a perfect crystal, which is to be compared to an increase of 10% found in HADES calculations.¹¹ In addition, the nearest-neighbor polar angle and the hole spin-density dipole moment are only weakly dependent on the assumed value of Q .

Our calculations indicate that an accurate theoretical description of the magnetic hyperfine interaction between the trapped hole at a V -type center in an alkaline-earth oxide and a nearby nucleus must include the dipole moment of the hole spin density, and that it will often suffice to neglect entirely the quadrupole moment of the hole spin density.

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