

Effect of the Jahn-Teller Interaction on the g Value of Co^{2+} in MgO

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Abstract The effect of the Jahn-Teller interaction on the gromagnetic factor g is calculated for the ground state of a substitutional Co^{2+} in crystalline MgO . In the model employed, only the interaction between Co^{2+} and the six nearest O^{2-} is considered, and their displacements from the equilibrium position are expressed as linear combinations of all the phonon modes of the crystal. The phonon spectrum employed is an analytic approximation to the results of a detailed calculation of another author. The effects of covalence are also taken into account in the present model.

1. INTRODUCTION

Historically the Jahn-Teller (JT) theorem applies to a non-linear molecule or a crystalline defect with orbital degeneracy and it states that the nuclear configuration is expected to be unstable with respect to a static distortion which lifts the degeneracy. In this paper we discuss a dynamic form of the JT effect that has been extensively discussed by Ham in a report of 1968¹. According to this effect, even when the spin-orbit interaction stabilizes the configuration of higher symmetry, the coupling of the electrons of the crystalline defect to the symmetric modes of vibration of the neighbouring ions still affects the properties of the system. It is the strength of this coupling which determines whether one should observe a static distortion or a dynamic effect. The dynamic JT effect may become important when some of the energy differences between the spin-orbit levels are close to phonon frequencies with an appreciable density of states. The Born-Oppenheimer approximation is invalid in this case and the electronic and vibrational parts of the wavefunction for each eigenstate of the coupled system are inextricably mixed.

The mixing of states due to the JT effect implies a reduction in the values of matrix elements of orbital operators between the elec-

tronic states, and produces a shift in the g values for the defect. In the cases of Co^{2+} and Fe^{2+} ions placed as substitutional impurities in MgO , the magnitude of these shifts for the respective ground states has been already computed with a molecular cluster model^{2,3}. In that model the vibrational spectrum of the crystal has been replaced by a single effective frequency whose value, together with the values of the covalency parameters and of the JT coefficients, have been chosen so as to satisfy consistently the EPR data as well as, in the case of Fe^{2+} , the infrared spectrum of the system. In the present work we are concerned with the calculation of the g shift of the ground state of Co^{2+} in MgO with a model in which the JT interaction is restricted to the six nearest O^{2-} , but employing an extended phonon spectrum. In this way we eliminate one of the adjustable parameters (the cluster frequency) and provide a more realistic test for the use of the JT mechanism.

Koloskova and Kochelaev⁴ have performed this type of calculation with a Debye spectrum. In the present paper we employ a density of states that represents more closely the phonon spectrum calculated by Peckham⁵ for pure MgO , and we also consider simultaneously effects which were neglected by Koloskova and Kochelaev. A study of covalency is important because it competes with the JT effect, and their relative magnitudes must be considered in the assignment of the values of the JT coefficients. The effects of covalency are introduced in the next section, where we describe the problem in the absence of the JT effect. Section 3 is devoted to the description of the JT Hamiltonian and of its effects on the wavefunctions of Co^{2+} in MgO . In section 4 we discuss the available experiments of EPR of $\text{MgO}:\text{Co}^{2+}$ under external stresses, which can be used to find a range of values for the JT coefficients. Finally the expression for the JT shift of the lowest spin-orbit doublet is obtained in section 5 and the results and discussions are given in section 6.

2. THE ZEROth ORDER PROBLEM

The relative positions of the spin-orbit levels which are split from the ground electronic triplet ${}^4T_{1g}$ of the Co^{2+} in the presence of a cubic crystal field are shown in Fig. 1. Two coefficients α and β are introduced to take into account the crystal field admixture

of the states ϕ_i of ${}^4T_{1g}$ with the states ϕ'_i of 4P

$$\phi_i = \alpha \phi_i + \beta \phi'_i \quad (1)$$

An appropriate diagonalization of the Hamiltonian including the crystal field interaction in this weak field representation gives $\alpha = 0.981$ and $\beta = 0.194$ (ref. 6). The effects of σ and π covalent bonding of the states ${}^4T_{1g}$ and ${}^4T_{2g}$ (see Fig. 1) with linear combinations of the $2p$ orbital of the six nearest O^{2-} ions can be introduced through the orbital reduction factors k_0 , k_1 and k_2 ^{3, 7}

$$\begin{aligned} \langle \psi_i | \vec{L} | \psi_j \rangle &= k_0 \langle \phi_i | \vec{L} | \phi_j \rangle \\ \langle \psi_i | \lambda \vec{L} | \psi_j \rangle &= k_1 \langle \phi_i | \lambda \vec{L} | \phi_j \rangle \\ \langle \psi_i | \lambda \vec{L} | \psi'_j \rangle &= k_2 \langle \phi_i | \lambda \vec{L} | \phi'_j \rangle \end{aligned} \quad (2)$$

The ψ_i and ψ'_i are molecular orbitals constructed with the ${}^4T_{1g}$ and ${}^4T_{2g}$ atomic orbitals, respectively, \vec{L} is the angular momentum operator and λ is the free ion spin-orbit coefficient of Co^{2+} . Instead of taking $k_1 = k_2$ as it was done by Tinkham ⁷, we shall leave k_1 and k_2 as independent parameters because the ${}^4T_{2g}$ states of Co^{2+} are expected to give a greater admixture with states of the neighbouring ions than the ${}^4T_{1g}$ states, so that $k_1 > k_2$. Nevertheless, we expect that the difference will not be too large. Using the effective spin-orbit Hamiltonian (H_{s_0}) and the effective Hamiltonian for the Zeeman effect deduced (both with respect to the states in the subspace ${}^4T_{1g}$) by Kanamori ⁸, we can express the energy differences of the states Γ_7 , Γ_8 and Γ'_8 from Γ_6 (see Fig. 1) and the value of the ground doublet Γ_6 as

$$E_7 - E_6 = 4 \left(-\frac{3}{2} \alpha^2 + \beta^2 \right) k_1 \lambda \quad (3a)$$

$$E_8 - E_6 = \frac{3}{2} \left(-\frac{3}{2} \alpha^2 + \beta^2 \right) k_1 \lambda - \frac{33}{20} \left(\frac{\sqrt{15}}{2} \alpha + \beta \right)^2 k_2^2 \lambda^2 / \Delta \quad (3b)$$

$$E'_8 - E_6 = 4 \left(-\frac{3}{2} \alpha^2 + \beta^2 \right) k_1 \lambda - \frac{18}{5} \left(\frac{\sqrt{15}}{2} \alpha + \beta \right)^2 k_2^2 \lambda^2 / \Delta \quad (3c)$$

$$g = \frac{5}{3} g_e + \left(\alpha^2 - \frac{2}{3} \beta^2 \right) k_0 + 2 \left(\frac{\sqrt{15}}{2} \alpha + \beta \right)^2 k_2^2 |\lambda| / \Delta \quad (3d)$$

where Δ is the energy separation of the ${}^4T_{2g}$ from the ${}^4T_{1g}$. On substituting the numerical values $\lambda = -180 \text{ cm}^{-1}$, $\Delta = 6800 \text{ cm}^{-1}$ and $g_e = 2.0023$ (see reference 3) we obtain the zeroth order energy differences

and g value in terms of the three covalency parameters.

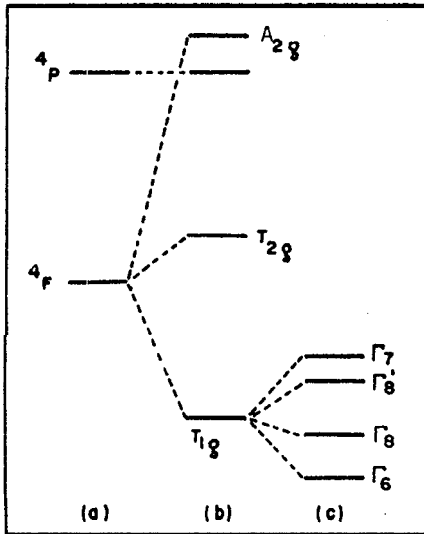


Fig.1 - (a) Energy level scheme of the free Co^{2+} ; (b) orbital levels in the presence of a cubic crystal field; (c) splitting of the ground orbital level in the presence of the spin-orbit interaction.

3. THE VIBRONIC PROBLEM

The total Hamiltonian for the system $MgO:Co^{2+}$ can be written as

$$H_S = H_0 + H_{JT} = H_{S_0} + H_V + H_{JT} \quad (4)$$

where H_V describes the vibrational states of the MgO and H_{JT} describes the JT interaction. The eigenfunctions of the zeroth-order Hamiltonian H are products of the electronic eigenfunctions Γ_6 , Γ_7 , Γ_8 and Γ_8' times the vibrational eigenfunctions of H_V . In the low temperature limit we may consider vibrational states with no more than one-photon excited, so that in the occupation number representation they can be described by $|0\rangle$ when $n_{qs}^{\rightarrow} = 0$ for all wavevector and polarization and by $|n_{qs}^{\rightarrow}\rangle$ when only the occupation number corresponding to the wavevector \vec{q} and polarization s is equal to one. The JT interaction mixes each $|\Gamma_n n_{qs}^{\rightarrow}\rangle = |\Gamma_n\rangle |n_{qs}^{\rightarrow}\rangle$ with all the other eigenfunctions of H_0 and these linear combinations are the vibronic eigenfunctions of H_S which describe the states of the coupled system. H_{JT} is given by^{1,2,3}

$$H_{JT} = V_A \tau^{\alpha} Q_{\alpha} + V_E (\tau^{\theta} Q_{\theta} + \tau^{\epsilon} Q_{\epsilon}) + V_T (\tau^{\xi} Q_{\xi} + \tau^{\eta} Q_{\eta} + \tau^{\zeta} Q_{\zeta}) \quad (5)$$

where V_A , V_E and V_T are the JT coefficients, τ^Y are electronic operators which can be defined in the subspace of the functions ${}^4T_{1g}$ as

$$\begin{aligned}
 \tau^\alpha &= I \\
 \tau^\theta &= (-3/2) l_z^2 + I \\
 \tau^E &= (\sqrt{3}/2) (l_x^2 - l_y^2) \\
 \tau^\xi &= l_y R_z - l_z l_y \\
 \tau^\eta &= l_z l_x - l_x l_z \\
 \tau^\zeta &= R_x R_y - l_y l_x
 \end{aligned} \tag{6}$$

and the Q_Y are normal modes of the seven ions complex composed by the impurity and its six nearest O^{2-} . In eq.(6) the I is the identity operator and the l_x , R_Y and R_z are components of an effective angular momentum operator with $l=1$, defined for the orbital part of the subspace generated by the ground ${}^4T_{1g}$ states of Co^{2+} . We see from eq. (5) that the JT interaction is limited to modes of vibration with symmetries A_{1g} , E_g and T_{2g} , the partners of which are $\alpha(A_{1g})$, θ and E (E_g) and ξ , η and ζ (T_{2g}), respectively. We shall see that only the modes E_g and T_{2g} contribute effectively in our calculation.

The coordinates Q_Y are linear combinations of the cartesian coordinates of the six nearest O^{2-} which can be expressed in terms of the normal modes Q_{qs}^+ of the crystal (phonons). Using the corresponding creation and annihilation operators a_{qs}^+ and a_{qs} , we find

$$H_{JT} = i \sqrt{\frac{\hbar}{2Nm_0}} \sum_{\vec{q}s} V_Y \tau^Y T_{qs}^Y \frac{1}{\sqrt{\omega_{\vec{q}s}}} (a_{qs}^+ - a_{qs}) \tag{7}$$

where m_0 is the mass of the O^{2-} , N is the number of cells in the crystal, $V_\theta = V_\epsilon = V_E$, $V_\xi = V_\eta = V_\zeta = V_T$ and the T_{qs}^Y are the coefficients connecting the Q_Y to the Q_{qs}^+ ; expressions for the T_{qs}^Y are found, for example, in reference 1. In terms of a_{qs}^+ and a_{qs} we have

$$H_v = \sum_{\vec{q}s} \hbar \omega_{\vec{q}s} a_{qs}^+ a_{qs} \tag{8}$$

With eqs. (7), (8) and the spin-orbit hamiltonian⁸

$$H_{s_0} = \left(-\frac{3}{2} \alpha^2 + \beta^2\right) \vec{l} \cdot \vec{S} - \left(\frac{15}{2} \alpha + \beta\right)^2 \frac{\lambda^2}{\Delta} \left[2(\ell_x^2 \ell_x^2 + \ell_y^2 \ell_y^2 + \ell_z^2 \ell_z^2) - (\vec{l} \cdot \vec{S})^2 \right] \quad (9)$$

where S_x , S_y and S_z are the components of the spin operator \vec{S} , we complete the definition of all terms appearing in the vibronic Hamiltonian H_s given in Eq. (4).

4. THE EFFECTS OF APPLIED STRESSES

To derive the JT coupling coefficients from measurements of g values in the presence of external deformations we shall assume that the interaction caused by these displacements is described by the Hamiltonian

$$H' = V_3(\tau^\theta e_\theta + \tau^E e_E) + V_5(\tau^\xi e_\xi + \tau^\eta e_\eta + \tau^5 e_5) \quad (10)$$

where the e_γ transform like the irreducible representations of the cubic group and are linear combinations of the usual strains. The V_i and V_j are coupling coefficients related to the JT coefficients by $V_E = \sqrt{3} V_3/2$ and $V_T = V_5/2R$ (R is the next smallest distance of the MgO lattice). Taking H' as a perturbation to H_s , retaining only terms which are linear in the magnetic field and neglecting contributions of the third and higher order, we obtain the corrections δ to the tensor g for the doublet Γ_6 . Using the Voigt notation⁹ we can then write

$$\delta g_i = F_{ij} e_j \quad (11)$$

It is found that from the 36 possible elements F_{ij} the only non-zero ones satisfy the relations $F_{11} = F_{22} = F_{33}$, $F_{44} = F_{55} = F_{66}$ and $F_{12} = F_{13} = F_{23}$ ($F_{ji} = F_{ij}$). The calculation for our model gives

$$F_{11} = -F_{12} = \frac{4}{3} \frac{(-3/2) \alpha^2 + \beta^2 - g_e}{E_6 - E_8} \frac{2R}{\sqrt{3}} V_E \quad (12)$$

$$F_{44} = -\frac{2}{3} \frac{(-3/2) \alpha^2 + \beta^2 - g_e}{E_6 - E_8} 2R V_T$$

Applying static uniaxial stresses along the crystal axis $[1, 0, 0]$ and $[1, 1, 0]$ - in the first case with magnetic fields parallel and normal to the strain and in the second case with a magnetic field parallel to the strain - Tucker⁹ has measured the values of F_{11} , F_{12} and F_{44} . Substi-

tuting his results into eq.(12) with E_6-E_8 given by eq. (13-b) and taking $k_1=k_2=1$, we find

$$\begin{aligned} V_E &= 2.04 \times 10^{11} \text{ cm}^{-1}/\text{cm} \\ V_T &= 0.34 \times 10^{11} \text{ cm}^{-1}/\text{cm} \end{aligned} \quad (13)$$

The estimated experimental error is 20%, so that we shall let the value of V_E to vary within an interval of $\pm 20\%$ around the result above. As the value of V_T is rather smaller than that of V_E , we shall neglect the effects of 20% changes of its value, and we shall employ the value quoted for V_T in Eq. (13).

5. THE JT OF THE g VALUE

The matrix elements of H_z are calculated here between pairs of vibronic states. Denoting the two Γ_6 states by $|a\rangle$ and $|-a\rangle$ and the corresponding vibronic states by $|A\rangle$ and $|-A\rangle$, the following reduction factors can be defined^{1,2}

$$\begin{aligned} k_s &= \langle -A | S_z | -A \rangle / \langle -a | S_z | -a \rangle \\ k_l &= \langle -A | l_z | -A \rangle / \langle -a | l_z | -a \rangle \\ k_A &= \langle -A | (\vec{l} \cdot \vec{S}) l_z + l_z (\vec{l} \cdot \vec{S}) | -A \rangle / \langle -a | (\vec{l} \cdot \vec{S}) l_z + l_z (\vec{l} \cdot \vec{S}) | -a \rangle \\ k_B &= \langle -A | l_z^2 S_z | -A \rangle / \langle -a | l_z^2 S_z | -a \rangle \end{aligned} \quad (14)$$

and the expression for the g value can then be written as

$$g = \frac{5}{3} g_e k_s + (\alpha^2 - \frac{2}{3} \beta^2) k_0 k_l - \frac{2}{3} \left(\frac{\sqrt{15}}{2} \alpha + \beta \right)^2 \frac{|\lambda|}{\Delta} k_2^2 (5k_A - 8k_B) \quad (15)$$

We see that each reduction factor is the ratio of the matrix element of an electronic operator calculated between vibronic states to the matrix element of the same operator calculated between the corresponding electronic states and *with the lattice in the ground vibrational state*. When the JT effect is neglected we have $k_s = k_l = k_A = k_B = 1$, and eq. (15) reduces to eq. (3-d).

In the perturbation calculation the state $|-A\rangle$ is obtained as a correction to $|-a\rangle = |-a0\rangle$, with H_{JT} given by eq. (7). To perform the calculation we have employed a few approximations: we replace the sum over \vec{q} by an integral over $d^3\vec{q}$ with the density of states $V/8\pi^3$, where

V is the volume of the crystal, and we take the phonon frequency as independent of the direction of \vec{q} and of the polarization. If we assume that the phonon polarization is strictly transversal or longitudinal, the following relation⁴

$$\int_0^{4\pi} \sum_s \frac{T_{qs}^{\gamma*}}{q_s} \frac{T_{qs}^{\gamma'}}{q_s} d\Omega_q = 4\pi \left(1 - \frac{\sin 2Rq}{2Rq} \right) \delta_{\gamma\gamma'}, \quad (16)$$

can be used separately for the acoustical and optical phonons. With this relation the following expression is obtained in second order of perturbation

$$\langle -A | \Omega | -A \rangle = \langle -a | \Omega | -a \rangle + \frac{1}{2NVm_0} \sum_{\gamma j} V_{\gamma} \int A_j^{\gamma}(w_q) \left(1 - \frac{\sin 2Rq}{2Rq} \right) \frac{\rho_j(w_q)}{w_q} dw_q \quad (17)$$

where the index j is used to distinguish the contributions of the optical phonons from that of the acoustical phonons, and where

$$\begin{aligned} A_j^{\gamma}(w_q) = & \sum_{mn} \left[\frac{\langle -a | \tau^{\gamma} | \Gamma_m \rangle \langle \Gamma_m | \Omega | \Gamma_n \rangle \langle \Gamma_n | \tau^{\gamma} | -a \rangle}{(w_{m_6} + w_q)(w_{n_6} + w_q)} \right. \\ & + \frac{\langle -a | \Omega | \Gamma_m \rangle \langle \Gamma_m | \tau^{\gamma} | \Gamma_n \rangle \langle \Gamma_n | \tau^{\gamma} | -a \rangle}{w_{m_6}(w_{n_6} + w_q)} \\ & \left. + \frac{\langle -a | \tau^{\gamma} | \Gamma_m \rangle \langle \Gamma_m | \tau^{\gamma} | \Gamma_n \rangle \langle \Gamma_n | \Omega | -a \rangle}{(w_{m_6} + w_q)w_{n_6}} \right] \\ & - \sum_n \frac{\langle -a | \tau^{\gamma} | \Gamma_n \rangle \langle \Gamma_n | \tau^{\gamma} | -a \rangle}{(w_{m_6} + w_q)^2} \langle -a | \Omega | -a \rangle \end{aligned} \quad (18)$$

We have taken $w_{\ell_6} = (E_{\ell} - E_{\epsilon})/\hbar$, where E_{ℓ} is the energy of the unperturbed electronic states $|\Gamma_{\ell}\rangle$. It is readily verified that, when $\gamma = A_{1g}$, the last term cancels the first one. Since the cases $E_m = E_6$ or $E_n = E_6$ are excluded in the second or third terms, we can conclude that the A_{1g} modes do not contribute to our calculation when we limit ourselves to the subspace of states of the ground ${}^4T_{1g}$. The contributions coming from excited ${}^4T_{2g}$ states can be neglected, because then w_{R_6} would be at least an order of magnitude greater than when it belongs to ${}^4T_{1g}$. Expression (18) can be simplified, if we take into account that the

operators Ω we employ couple $|-a\rangle$ only with states $|\Gamma_g\rangle$; in this case, according to the table of Koster et al.¹⁰, the double sum over γ and m (or n) in the second and third terms of eq. (18) is zero and these do not contribute.

We employ simple analytical expressions to simulate the one-phonon density of acoustical phonons $\rho_a(w)$ and of optical phonons $\rho_o(w)$ calculated by Peckham⁵. We have selected six phonon frequency intervals, and expressed $\rho_a(w)$ and $\rho_o(w)$ in each interval by a function of the type

$$\rho_j(w) = \left[N a_{ij} + b_{ij} (w - w_j) c_{ij} \right] \quad (19)$$

Each of these contributions is shown in Fig. 2, together with their sum and with the curve of Peckham. We have introduced dispersion relations $q_j(w)$ that satisfy the relation

$$(V/2\pi^2) \int q_j^2(w) dq(w) = \int \rho_j(w) dw \quad (20)$$

and its expression in the i -th interval is of the type

$$q_j(w) = \frac{1}{R} \left[d_{ij} + e_{ij} w + f_{ij} (w - w_1)^{g_{ij}} \right]^{1/3} \quad (21)$$

Substituting eqs. (19) and (21) into eq. (17) we had to calculate integrals of the type

$$I_{mn} = \sum_{ij} \int \frac{a_{ij} + b_{ij} (w - w_j)^{c_{ij}}}{(w_{m6} + w)(w_{n6} + w)} \left[1 - \frac{\sin 2 \left[d_{ij} + e_{ij} w + f_{ij} (w - w_j)^{g_{ij}} \right]^{1/3}}{2 \left[d_{ij} + e_{ij} w + f_{ij} (w - w_j)^{g_{ij}} \right]^{1/3}} \right] \frac{dw}{w} \quad (22)$$

which have been obtained numerically. In terms of these integrals we can write

$$\langle -A | \Omega | -A \rangle = \langle -a | \Omega | -a \rangle + \frac{1}{2\pi N m_0} \sum_{\gamma mn} V^2 \langle -a | \tau^\gamma | \Gamma_m \rangle \left[\langle \Gamma_m | \Omega | \Gamma_n \rangle - \langle -a | \Omega | -a \rangle \delta_{mn} \right] \cdot \Gamma | \tau^\gamma | -a \rangle I_{mn} \quad (23)$$

The matrix elements of the electronic operators Ω and τ^γ can be calculated with the functions $|\Gamma_\ell\rangle$ given, for example, in ref. (1), and with

aid of the table of Koster et al.. To calculate the integrals I_{mn} it is necessary to know the values of $E_R - E_6$ as given by eqs. (3-a, b, c), with appropriate choice of the covalency parameters k_1 and k_2 . This choice and also that of the parameters k_0 and V_E which satisfy the experimental result $g = 4.278$ ¹¹ is discussed in the next section.

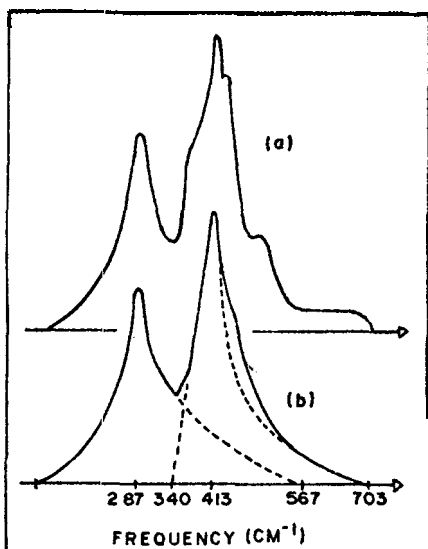


Fig.2 - (a) One-phonon density of states for the MgO obtained by Peckhan; (b) the full line represents a simulation of the Peckhan's curve used in our calculation, and it is obtained by the superposition of the acoustical and optical contributions (dashed lines)

6. RESULTS AND DISCUSSIONS

In a recent work¹², we have shown that the JT effect reduces the energy difference $E_7 - E_6$ and increases $E_8' - E_6$ by an amount of $30-40 \text{ cm}^{-1}$ when, in absence of the JT interaction, we take these differences as -300 cm^{-1} and -900 cm^{-1} , respectively. Since the corresponding experimental values are close to 305 cm^{-1} and 935 cm^{-1} , respectively (see ref. 12), we assume that in the absence of the JT interaction these differences are $330-340 \text{ cm}^{-1}$ and $890-900 \text{ cm}^{-1}$. Knowing the values of $E_7 - E_6$ and $E_8' - E_6$, we can then deduce k_1 and k_2 from eqs. (3-b) and (3-c). Imposing $0.8k_1$ as the lower limit of k_2 in the condition $k_2 < k_1$ (stated in section 2), we take $k_1 = 0.92 \pm 0.01$ and $k_2 = 0.82 \pm 0.01$ which correspond to $E_7 - E_6 = 330 \text{ cm}^{-1}$, $E_8' - E_6 = 890 \text{ cm}^{-1}$ and $E_7 - E_6 = 945 \text{ cm}^{-1}$. Substituting these numbers into eq. (22) we find by

numerical integration the following values for I_{mn}

$$\begin{aligned}
 I_{,,} &= 0.177 \times 10^{-36} (\text{rad/s})^{-3} & I_{8,8,1} &= 0.0347 \times 10^{-36} (\text{rad/s})^{-3} \\
 I_{77} &= 0.0325 \times 10^{-36} (\text{rad/s})^{-3} & I_{88,1} &= 0.0638 \times 10^{-36} (\text{rad/s})^{-3} \\
 I_{78} &= 0.0615 \times 10^{-36} (\text{rad/s})^{-3} & I_{78,1} &= 0.0337 \times 10^{-36} (\text{rad/s})^{-3}
 \end{aligned}
 \tag{24}$$

Substituting these values into eq. (22) and taking $V_T = 0.34 \times 10^{11}$ (cm^{-1}/cm) and $g = 4.278$, the following sets of pairs of values of V_E (within the limits of the experimental error) and k_0 satisfy eq.(15)

$V_E (10^{11} \text{ cm}^{-1}/\text{cm})$	1.60	1.80	2.00	2.40
k_0	0.88	0.89	0.93	0.96

The orbital reduction factor for Fe^{2+} in MgO has been estimated to be $k_0 = 0.95^2$ and if we assume, as emphasized by some authors^{7,8}, that the effects of covalency in the Fe^{2+} are greater than in the Co^{2+} , a natural choice of k_0 in the table above is 0.96. This is in the region predicted by Ray and Regnard³ and together with $k_1 = 0.92$ they are much closer to one than the values $k_0 = 0.89$ and $k_1 = 0.81$ obtained when the JT effect is neglected¹¹. At the same time the corresponding value $V_E = 2.40 \times 10^{11} \text{ cm}^{-1}/\text{cm}$ is, within the limits of the experimental error of the EPR measurements, discussed in section 4.

We have thus obtained a set of parameters which are consistent between themselves and with the existing experimental data. In view of the arguments considered above, we shall adopt the values $k_0 = 0.96 \pm 0.01$, $k_1 = 0.92 \pm 0.01$ and $V_E = (2.4 \pm 0.1) \times 10^{11} \text{ cm}^{-1}/\text{cm}$. These values have been employed in the calculation of the contribution of the JT effect to the electronic Raman spectra of the $\text{MgO}:\text{Co}^{2+}$ (Ref. 12), and the results seem to indicate that they are essentially correct within the stated precisions. We then conclude that the JT mechanism considered here provides an accurate explanation of the properties of the system $\text{MgO}:\text{Co}^{2+}$, and that its effects are comparable to those of covalency.

The reduction factors associated to the parameters adopted above are

$$k_s = 0.945 \quad k_l = 0.965 \quad k_A = 0.986 \quad k_B = 0.998$$

and their departure from unity is due only to the JT interaction. Note that our definition of the reduction factors is different from that given by Ham^{1,2}, which gives values different from 1 even in the absence of the JT effect.

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Resumo

O efeito da interação Jahn-Teller sobre o fator giromagnético g é calculado para o estado fundamental de um ion Co^{2+} substitucional em MgO cristalino. No modelo usado, somente é considerada a interação entre o Co^{2+} e os seis O^{2-} mais próximos, e seus deslocamentos da posição de equilíbrio são expressos como combinações lineares de todos os modos fonônicos do cristal. O espectro de fonons utilizado é uma aproximação analítica dos resultados do cálculo detalhado de um outro autor. Os efeitos da covalência são levados em conta no modelo.