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Origin of the magnetoelasticity in cubic rare-earth intermetallic compounds

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The importance of the magnetoelastic effects in the cubic rare-earth intermetallic compounds motivates one to look for their microscopic origin. The magnetoelastic coefficients appear to be the strain derivatives of the cubic crystal-field parameters. As the latter ones, they originate from two contributions, the ligands and the conduction electrons. The aim of this paper is to study thoroughly these two contributions to the second-, fourth-, and sixth-order magnetoelastic coefficients associated with the tetragonal and trigonal symmetry-lowering modes. In particular the expressions of the direct and exchange Coulombic contribution of each type of conduction electron (p, d, or f character) are derived. Numerical results are deduced for compounds DyZn and DyCu starting from self-consistent augmented-plane-wave functions. We verify that the magnetoelastic modifications of the cubic fourth- and sixth-order parameters are negligible in these intermetallic compounds and that the second-order magnetoelastic coefficients result from the competition of numerous opposite contributions. Comparisons are made with experimental data in the two isomorphous series of rare-earth compounds with zinc and copper.

I. INTRODUCTION

The magnetoelastic interactions have been extensively investigated over the last few years in rare-earth intermetallic compounds having in particular the cubic symmetry. Their effects are usually revealed by the spontaneous magnetostriction, i.e., the lattice distortion occurring simultaneously with the magnetic ordering. In some cases such as the rare-earth Laves phases with iron the corresponding strains are large enough for suggesting technological applications.[1] Other series have been also particularly investigated such as the rare-earth antimonides (RSb) (Refs. 2 and 3) and the CsCl-type structure compounds with zinc or copper, where tetragonal spontaneous strains of typically 10^{-2} may be observed.[4] In these favorable systems due to the high symmetry of the rare-earth site, the magnetoelastic effects may be studied about the two symmetry-lowering modes: The tetragonal and the trigonal strains which have been systematically measured, for example, throughout the rare-earth zinc (RZn) series. For these studies, three experimental methods have been used: measuring the spontaneous magnetostriction,[5] the elastic constants,[6] and the parastriction,[7] i.e., the anisotropic strain induced by an applied magnetic field in the paramagnetic range. From these studies, it appears that the two-ion magnetoelastic coupling, modulating the exchange interactions between magnetic ions, contributes only weakly to the symmetry-lowering modes in the RZn series.

As a consequence we will restrict ourselves in this paper to the study of the only one-ion magnetoelastic effects lowering the cubic symmetry. They are related to the coupling of the rare-earth ion with its environment due to the large asphericity of the $4f$ shell. However, in these compounds the presence of strong crystalline-electric-field (CEF) effects requires to use a quantum treatment for describing the magnetoelasticity as well as all the magnetic properties.[4] The relevant one-ion magnetoelastic Hamiltonian, linear in strain (harmonic approximation) is usually limited to second-order Stevens operators, the fourth- and sixth-order magnetoelastic coefficients (modification of the fourth- and sixth-order cubic CEF parameters) being generally assumed to be negligible.[3, 7, 8]

As the magnetoelastic coefficients are defined as the strain derivatives of the CEF parameters[9] it is necessary to have a model of how the $4f$ shell of the rare earth is connected to its strained surroundings. The CEF parameters have been extensively studied in the literature; they have been shown to originate mainly from two contributions: the point charges and the conduction-electrons contributions. In metallic rare-earth systems with the cubic symmetry, the effects of the conduction band have been found preponderant,[10, 11] due to the strong anisotropic orbital character of the conduction electrons, as shown for instance by augmented-plane-wave band calculations.[12] It is therefore necessary to study the behavior of the strain derivatives of these two CEF contributions.

Our starting point is the cubic paramagnetic crystal. In the presence of a weak strain we apply a first-order perturbation theory. At first the localized charges are displaced from their previous cubic arrangement, generating a new potential having the new symmetry (tetragonal or trigonal) of the lattice and proportional to the strain; this potential gives the point-charges contribution to the magnetoelastic coefficients. On the other hand and to the first order of perturbation this new potential only shifts the energy of each conduction-electron state. In particular, it lifts their cubic orbital degeneracy according to the Fermi statistics that leads to a redistribution of the conduction electrons in the band which gives rise to a new CEF contribution, i.e., the band contribution to the magnetoelastic coefficients.

The aim of this paper is to evaluate the various contributions to the second-order magnetoelastic coefficients as well as to the modifications of the fourth- and sixth-order cubic CEF parameters. In Sec. II we develop the formalism for calculating all the one-ion harmonic magnetoelastic coefficients in the point charge and the band models. A numerical application of these theoretical results is given in Sec. III, starting from "augmented-plane-wave" band calculations.

II. THEORY

A. Hamiltonian

In the nonstrained lattice the basic Hamiltonian is the usual crystalline-electric-field (CEF) Hamiltonian[13]

$$\mathcal{H}_{\mathrm{CEF}}=\sum_{LM} B_L^M O_L^M = B_4^0(O_4^0+5O_4^4)+B_6^0(O_6^0-21O_6^4)\ , \tag{1}$$

which is written in the fourfold-axes system.

In the present paper we restrict ourselves to the analysis of the one-ion harmonic magnetoelastic coupling lowering the cubic symmetry. The corresponding Hamiltonian consists then of two parts.

(i) The first part includes the second-order CEF operators which were absent in cubic symmetry[4] (where, for instance, $B_2^0=0$):

$$\begin{aligned}\mathcal{H}_{\mathrm{me}}^{(2)} &= -B_1(\epsilon_3 O_2^0+\sqrt{3}\epsilon_2 O_2^2)\\ &\quad -B_2(\epsilon_{xy}P_{xy}+\epsilon_{yz}P_{yz}+\epsilon_{zx}P_{zx})\ ,\end{aligned} \tag{2}$$

with

$$\begin{aligned}&O_2^0=3J_z^2-J(J+1),\quad O_2^2=J_x^2-J_y^2\ ,\\ &P_{xy}=\tfrac{1}{2}(J_xJ_y+J_yJ_x),\ \ldots,\ .\end{aligned} \tag{3}$$

We use here the symmetrized strains

$$\begin{aligned}\epsilon_3&=\frac{1}{\sqrt{6}}(2\epsilon_{zz}-\epsilon_{xx}-\epsilon_{yy})\ ,\\ \epsilon_2&=\frac{1}{\sqrt{2}}(\epsilon_{xx}-\epsilon_{yy})\ ,\end{aligned} \tag{4}$$

the ϵ_{ij}'s being the tensor components of the strain. It appears then two second-order harmonic magnetoelastic coefficients B_1 and B_2, associated, respectively, to a tetragonal and a trigonal strain.

(ii) The second part is the modification of the fourth- and sixth-order cubic CEF terms [Eq. (1)] by the strain. It is written in the case of a tetragonal strain:

$$\mathcal{H}_{\mathrm{me}}^{(4)}=-B_1^{(4)}[\epsilon_3(O_4^0-7O_4^4)-4\sqrt{3}\epsilon_2 O_4^2] \tag{5}$$

and

$$\begin{aligned}\mathcal{H}_{\mathrm{me}}^{(6)}&=-B_1^{(6)}[\epsilon_3(O_6^0+3O_6^4)\\ &\quad+\tfrac{5}{2}\sqrt{3}\epsilon_2(O_6^2+\tfrac{11}{5}O_6^6)]\ ,\end{aligned} \tag{6}$$

the expressions being more complex for a trigonal strain in this fourfold axes system.

Another expression of the magnetoelastic Hamiltonian consists in starting from the Stevens notation for the cubic CEF Hamiltonian[13]:

$$\mathcal{H}_{\mathrm{CEF}}=\sum_{LM} A_L^M\langle r^L\rangle \Theta_L O_L^M\ , \tag{7}$$

where $\langle r^L\rangle$ is the average value of r^L for the $4f$ shell and where Θ_L are the Stevens's coefficients ($\Theta_2=\alpha_J$, $\Theta_4=\beta_J$, $\Theta_6=\gamma_J$). Comparing with Eqs. (2), (5), and (6), we obtain for the magnetoelastic coefficients associated to the tetragonal strain mode

$$\frac{B_1}{\alpha_J}=-\frac{1}{\alpha_J}\frac{\partial}{\partial\epsilon_3}B_2^0=-\frac{\partial}{\partial\epsilon_3}(A_2^0\langle r^2\rangle)\ , \tag{8}$$

$$\frac{B_1^{(4)}}{\beta_J}=-\frac{1}{\beta_J}\frac{\partial}{\partial\epsilon_3}B_4^0=-\frac{\partial}{\partial\epsilon_3}(A_4^0\langle r^4\rangle)\ , \tag{9}$$

$$\frac{B_1^{(6)}}{\gamma_J}=-\frac{1}{\gamma_J}\frac{\partial}{\partial\epsilon_3}B_6^0=-\frac{\partial}{\partial\epsilon_3}(A_6^0\langle r^6\rangle)\ . \tag{10}$$

In the case of a trigonal deformation, it is more convenient to consider the threefold-axes system, where the [111] direction is the new z axis. The cubic CEF Hamiltonian is written in this new system[13]

$$\begin{aligned}\mathcal{H}'_{\mathrm{CEF}}&=\sum_{LM} B_L'^M O_L^M = B_4'^0(O_4^0-20\sqrt{2}O_4^3)\\ &\quad+B_6'^0(O_6^0+\tfrac{35}{4}\sqrt{2}O_6^3+\tfrac{77}{8}O_6^6)\\ &=\sum_{LM} A_L'^M\langle r^L\rangle\Theta_L O_L^M\ ,\end{aligned} \tag{11}$$

with $B_4'^0=-\frac{2}{3}B_4^0$ and $B_6'^0=\frac{16}{9}B_6^0$. The second-order magnetoelastic Hamiltonian [Eq. (2)] may be written in this new system, taking into account a pure trigonal strain ($\epsilon_3=\epsilon_2=0$; $\epsilon_{xy}=\epsilon_{yz}=\epsilon_{zx}=\epsilon_{ij}$):

$$\mathcal{H}_{\mathrm{me}}'^{(2)}=-\tfrac{1}{2}B_2\epsilon_{ij}O_2^0\ . \tag{12}$$

That leads to the following expression for the second-order magnetoelastic coefficient associated with the trigonal strain mode

$$\frac{B_2}{\alpha_J} = -\frac{2}{\alpha_J}\frac{\partial}{\partial\epsilon_{ij}}B_2^{\prime 0} = -2\frac{\partial}{\partial\epsilon_{ij}}(A_2^{\prime 0}\langle r^2\rangle) \tag{13}$$

and by extension for the fourth- and sixth-order coefficients

$$\frac{B_2^{(4)}}{\beta_J} = -\frac{1}{\beta_J}\frac{\partial}{\partial\epsilon_{ij}}B_4^{\prime 0} = -\frac{\partial}{\partial\epsilon_{ij}}(A_4^{\prime 0}\langle r^4\rangle) \ , \tag{14}$$

$$\frac{B_2^{(6)}}{\gamma_J} = -\frac{1}{\gamma_J}\frac{\partial}{\partial\epsilon_{ij}}B_6^{\prime 0} = -\frac{\partial}{\partial\epsilon_{ij}}(A_6^{\prime 0}\langle r^6\rangle) \ . \tag{15}$$

The evaluation of all these harmonic magnetoelastic coefficients requires therefore to calculate the derivatives of the cubic CEF parameters $A_L^0\langle r^L\rangle$ successively with regard to a tetragonal and a trigonal strain mode, and within the point charge (PC) and the band model.

B. Tetragonal strain mode

First we consider a pure tetragonal deformation, where the c axis remains a fourfold axis. The only nonzero strain parameter is then

$$\epsilon_3 = (\tfrac{2}{3})^{1/2}\left(\frac{c}{a} - 1\right) \ . \tag{16}$$

1. *Point-charge model*

The ligands charges of the strained lattice generate an electric potential $V(\vec{r})$ which is written as

$$V(\vec{r}) = \sum_{LM} r^L \left(V_L^M + \frac{\partial V_L^M}{\partial\epsilon_3}\epsilon_3 \right) Y_L^M(r) \ , \tag{17}$$

where the coefficient in parentheses includes the summation over all the neighbors j (charge q_j, position $\vec{r}_j$):

$$V_L^M + \frac{\partial V_L^M}{\partial\epsilon_3}\epsilon_3 = \frac{4\pi}{2L+1}\sum_j q_j \left[\frac{Y_L^{M^*}(\hat{r}_j)}{r_j^{L+1}} + \frac{\partial}{\partial\epsilon_3}\left(\frac{Y_L^{M^*}(\hat{r}_j)}{r_j^{L+1}} \right)\epsilon_3 \right] \ . \tag{18}$$

TABLE I. Some values of the sums $S_1^{p_1 p_2 p_3}$ [Eq. (20)] for the simple cubic (sc) and body centered cubic (bcc) lattices.

$S_1^{p_1 p_2 p_3}$	sc	bcc
$S_7^{400} - 3S_7^{220}$	1.556	−0.960
$5S_{11}^{600} - 47S_{11}^{420} + 66S_{11}^{222}$	7.353	22.298
$S_{11}^{600} - 22S_{11}^{420} + 51S_{11}^{222}$	0.7577	19.007
S_{15}^{800}	2.083	2.473
S_{15}^{620}	0.0275	0.3128
S_{15}^{440}	0.0259	0.3013
S_{15}^{422}	0.0026	0.2752

The first part leads to the usual expression of the cubic CEF parameters $A_L^M\langle r^L\rangle$ while the second one allows to obtain their strain derivatives[13]

$$\frac{\partial}{\partial\epsilon_3}(A_L^M\langle r^L\rangle) = -|e|\epsilon_L^M\langle r^L\rangle\frac{\partial V_L^M}{\partial\epsilon_3} = \frac{4\pi|e|}{2L+1}\epsilon_L^M\langle r^L\rangle\sum_j q_j\frac{\partial}{\partial\epsilon_3}\left(\frac{Y_L^{M^*}(\hat{r}_j)}{r_j^{L+1}} \right) \ , \tag{19}$$

where ϵ_L^M is the numerical coefficient of Y_L^M [$\epsilon_2^0 = \frac{1}{4}(5/\pi)^{1/2}$; $\epsilon_4^0 = (\frac{2}{35})^{1/2}\epsilon_4^{\pm 4} = 3/16\sqrt{\pi}$; $\epsilon_6^0 = \frac{1}{3}(\frac{2}{7})^{1/2}\epsilon_6^{\pm 4} = \frac{1}{32}(13/\pi)^{1/2}$]. The summations over j can be expressed for each type of neighbor $\{j\}$ as a function of the following elementary sums which are independent of the cubic lattice parameter[14] a:

$$S_1^{p_1 p_2 p_3}(\{j\}) = a^{1-p_1-p_2-p_3}\sum_{\{j\}}\frac{x_j^{p_1}y_j^{p_2}z_j^{p_3}}{r_j^1} \ . \tag{20}$$

Table I collects some values of these sums for neighbors belonging to simple cubic (sc) or body centered cubic (bcc) lattices. In the cubic CsCl-type structure the summations will be written, respectively, for the rare-earth (R) and the alloyed metal (M) neighbors as

$$S_1^{p_1 p_2 p_3}(R) = S_1^{p_1 p_2 p_3}(\mathrm{sc}) \ , \qquad S_1^{p_1 p_2 p_3}(M) = S_1^{p_1 p_2 p_3}(\mathrm{bcc}) - S_1^{p_1 p_2 p_3}(\mathrm{sc}) \ . \tag{21}$$

In the present case we need the summations in Eq. (19) for $(L,M) = (2,0)$, $(4,0)$, and $(6,0)$ (see Table II). The final expressions for the PC contribution to the magnetoelastic coefficients are then

$$\left.\frac{B_1}{\alpha_J}\right|_{\mathrm{PC}} = -\frac{3\sqrt{6}}{4}\frac{e^2\langle r^2\rangle}{a^3}\sum_{\{j\}} q_{\{j\}}(S_7^{400} - 3S_7^{220})_{\{j\}} \ , \tag{22}$$

$$\left.\frac{B_1^{(4)}}{J}\right|_{\mathrm{PC}} = -\frac{5\sqrt{6}}{192}\frac{e^2\langle r^4\rangle}{a^5}\sum_{\{j\}} q_{\{j\}}(5S_{11}^{600} - 47S_{11}^{420} + 66S_{11}^{222})_{\{j\}} \ , \tag{23}$$

$$\left.\frac{B_1^{(6)}}{\gamma_J}\right|_{\mathrm{PC}} = -\frac{7\sqrt{6}}{256}\frac{e^2\langle r^6\rangle}{a^7}\sum_{\{j\}} q_{\{j\}}(7S_{15}^{800} - 190S_{15}^{620} + 225S_{15}^{440} + 30S_{15}^{422})_{\{j\}} \ , \tag{24}$$

TABLE II. Expression of some strain derivatives of CEF summations [see Eq. (19)].

$\frac{a^3}{\epsilon_2^0}\sum_{\{j\}}\frac{\partial}{\partial\epsilon_3}\frac{Y_2^0(\hat{r}_j)}{r_j^3}$	$-3\sqrt{6}(S_7^{400}-3S_7^{220})$
$\frac{a^5}{\epsilon_4^0}\sum_{\{j\}}\frac{\partial}{\partial\epsilon_3}\frac{Y_4^0(\hat{r}_j)}{r_j^5}$	$-\frac{5}{3}\sqrt{6}(5S_{11}^{600}-47S_{11}^{420}+66S_{11}^{222})$
$\frac{a^7}{\epsilon_6^0}\sum_{\{j\}}\frac{\partial}{\partial\epsilon_3}\frac{Y_6^0(\hat{r}_j)}{r_j^7}$	$-7\sqrt{6}(7S_{15}^{800}-190S_{15}^{620}+225S_{15}^{440}+30S_{15}^{422})$
$\frac{a^3}{\epsilon_2^0}\sum_{\{j\}}\frac{\partial}{\partial\epsilon_{ij}}\frac{Y_2^0(\hat{r}_j)}{r_j^3}$	$12(S_7^{400}-3S_7^{220})$
$\frac{a^5}{\epsilon_4^0}\sum_{\{j\}}\frac{\partial}{\partial\epsilon_{ij}}\frac{Y_4^0(\hat{r}_j)}{r_j^5}$	$-\frac{80}{3}(S_{11}^{600}-22S_{11}^{420}+51S_{11}^{222})$

where $q_{\{j\}}$ is the effective charge of the rare earth or the alloyed metal in $|e|$ unit. In insulators these effective charges are the ionic ones; in metallic compounds they have to be evaluated taking into account the conduction-electron density, as explained in the following (Sec. III A).

2. *Band model*

Band-structure calculations have been previously carried out in some rare-earth intermetallic compounds having the CsCl-type structure, using the self-consistent augmented-plane-wave (APW) method.[12] In this APW method the space is divided into contiguous spheres centered on each ion. The calculations have evidenced a strong anisotropic orbital character for the conduction electrons, essentially of p and d type, with a small amount of f type, in particular, inside the rare-earth APW sphere. Starting from these results crystal-field calculations have showed that the d and f type orbital character of the conduction electrons around the rare earth has a predominant influence on the cubic CEF parameters.[10,11] In particular, the Coulombic exchange contributions have been evaluated: if they cancel partially the direct ones for d electrons, they have been found to be preponderant for $5f$ conduction electrons, explaining the experimental order of magnitude of the sixth-order CEF parameters.

In presence of a given deformation, the problem is to know how the electronic states of the conduction band are modified by the strain. Our hypothesis will remain very simple: The orbital degeneracy of the conduction electrons is lifted by the only potential $V_2(\vec{r})$ originating in the shifted ligands charges and which is written [Eq. (17) with $V_2^0=0$] as

$$V_2(\vec{r})=r^2\frac{\partial V_2^0}{\partial\epsilon_3}\epsilon_3 Y_2^0(\hat{r}) \ , \tag{25}$$

$\partial V_2^0/\partial\epsilon_3$ being given by Eq. (19).

The group theory forecasts how an orbitally degenerated state of a conduction electron, belonging to a given cubic representation Γ, is split by a tetragonal deformation (see Table III, with the notations of Ref. 15). If we define the wave function of a conduction electron having the orbital character λ and symmetry Γ

$$\psi_{\lambda,\Gamma}^{\varrho}(\vec{r})=R_\lambda(r)\varphi_{\lambda,\Gamma}^{\varrho}(\hat{r}) \ , \tag{26}$$

TABLE III. Reduction of the cubic representations (of dimension m_Γ) in a tetragonal and trigonal symmetry.

m_Γ	1	1	2	3	3	Symmetry group
Cubic representations	a_1	a_2	e	t_1	t_2	O
Tetragonal representations	a_1	b_1	a_1+b_1	a_2+e	b_2+e	D_4
Trigonal representations	a_1	a_2	e	a_2+e	a_1+e	D_3

p varying from 1 to the degeneracy m_Γ of the Γ representation ($m_\Gamma = 2$ or 3), the shift in energy $\Delta^p_{\lambda,\Gamma}$ of the corresponding electronic state will be

$$\Delta^p_{\lambda,\Gamma} = \langle \psi^p_{\lambda,\Gamma} | -|e| V_2(\vec{r}) | \psi^p_{\lambda,\Gamma} \rangle$$

$$= -|e| \langle r^2 \rangle_\lambda \frac{\partial V_2^0}{\partial \epsilon_3} \epsilon_3 \langle \varphi^p_{\lambda,\Gamma} | Y_2^0 | \varphi^p_{\lambda,\Gamma} \rangle \quad , \tag{27}$$

with

$$\langle r^2 \rangle_\lambda = \int r^2 [R_\lambda(r)]^2 r^2 dr \tag{28}$$

and on condition that the $V_2(\vec{r})$ perturbation is diagonal in the $\varphi^p_{\lambda,\Gamma}$ basis ($\langle \varphi^p_{\lambda,\Gamma} | Y_2^0 | \varphi^{p'}_{\lambda,\Gamma} \rangle = 0$ if $p \neq p'$).

The tetragonal strain modifies thus the population $n^p_{\lambda,\Gamma}$ of each resulting "subband"

$$n^p_{\lambda,\Gamma} = \frac{1}{m_\Gamma} \int^{E_F - \Delta^p_{\lambda,\Gamma}} N_{\lambda,\Gamma}(E) dE \quad , \tag{29}$$

E_F being the Fermi energy of the cubic lattice and $N_{\lambda,\Gamma}(E)$ the partial density of states associated to the character λ and the cubic symmetry Γ.

For a twofold degeneracy ($m_\Gamma = 2$), the representation is split in two singlets (see Table III) with an opposite energy shift ($\Delta^1_{\lambda,\Gamma} = -\Delta^2_{\lambda,\Gamma}$) and the difference of population between both "subbands" may be written as

$$\Delta n_{\lambda,\Gamma} = n^1_{\lambda,\Gamma} - n^2_{\lambda,\Gamma} = -\frac{1}{m_\Gamma - 1} \Delta^1_{\lambda,\Gamma} N_{\lambda,\Gamma}(E_F) \quad . \tag{30}$$

For a threefold degeneracy ($m_\Gamma = 3$) the representation is split into a singlet ($p = 1$) and a doublet ($p = 2, 3$) with $\Delta^1_{\lambda,\Gamma} = -2\Delta^2_{\lambda,\Gamma} = -2\Delta^3_{\lambda,\Gamma}$ and the difference $\Delta n_{\lambda,\Gamma}$ which will be needed in the following is written here:

$$\Delta n_{\lambda,\Gamma} = n^1_{\lambda,\Gamma} - \tfrac{1}{2}(n^2_{\lambda,\Gamma} + n^3_{\lambda,\Gamma})$$

$$= -\frac{1}{m_\Gamma - 1} \Delta^1_{\lambda,\Gamma} N_{\lambda,\Gamma}(E_F) \quad . \tag{31}$$

We note that only the partial density of states at the Fermi level occurs in Eqs. (30) and (31).

We have now to calculate the contribution of each subband to the CEF parameters. Previous work showed that these contributions, reduced to one conduction electron of character λ and symmetry Γ' in the strained state, may be written as[11]

$$A_L^0 \langle r^L \rangle \Big|_{\lambda,\Gamma'} = K^L_{\lambda,\Gamma'} F^L_{\lambda\lambda} + \sum_l J^{L,l}_{\lambda,\Gamma'} G^l_{\lambda\lambda} \quad , \tag{32}$$

where $K^L_{\lambda,\Gamma'}$ and $J^{L,l}_{\lambda,\Gamma'}$ are numerical coefficients depending only on the angular part of the wave functions, and $F^L_{\lambda\lambda}$ and $G^l_{\lambda\lambda}$ the direct and exchange Slater's integrals depending only on their radial part.

Symmetry rules (see Ref. 10) imply some relations between the angular coefficients corresponding to the different representations issued from a cubic representation Γ ($\Gamma \to \Gamma'_1 + \Gamma'_2$ where Γ'_1 is a singlet and Γ'_2 a singlet or a doublet), leading to

$$A_L^0 \langle r^L \rangle |_{\lambda,\Gamma'_2} = \sigma^L_{\lambda,\Gamma} A_L^0 \langle r^L \rangle |_{\lambda,\Gamma'_1} \quad . \tag{33}$$

The resulting CEF parameters will be obtained by summing the contributions of each subband:

$$A_L^0 \langle r^L \rangle \Big|_{\substack{\lambda,\Gamma \\ \text{tot}}} = \sum_{\Gamma'} A_L^0 \langle r^L \rangle \Big|_{\lambda,\Gamma'} n_{\lambda,\Gamma'} = A_L^0 \langle r^L \rangle |_{\lambda,\Gamma'_1} \left[\frac{1 + (m_\Gamma - 1)\sigma^L_{\lambda,\Gamma}}{m_\Gamma} \left(\sum_{\Gamma'} n_{\lambda,\Gamma'} \right) + \frac{m_\Gamma - 1}{m_\Gamma} (1 - \sigma^L_{\lambda,\Gamma}) \Delta n_{\lambda,\Gamma} \right] \quad , \tag{34}$$

where $n_{\lambda,\Gamma'}$ is the population of the subband Γ' and $m_\Gamma = 2$ or 3. The first part of this expression gives the contribution of the conduction electrons of symmetry (λ, Γ) to the cubic CEF parameters while the second part proportional to $\Delta n_{\lambda,\Gamma}$ then to ϵ_3, gives their contribution to the magnetoelastic coefficients. Using Eqs. (27), (30), and (31), we obtain finally

$$-\frac{\partial}{\partial \epsilon_3} (A_L^0 \langle r^L \rangle |_{\lambda,\Gamma}) = -\frac{|e|}{m_\Gamma} (1 - \sigma^L_{\lambda,\Gamma}) \langle r^2 \rangle \frac{\partial V_2^0}{\partial \epsilon_3} \langle \varphi^1_{\lambda,\Gamma} | Y_2^0 | \varphi^1_{\lambda,\Gamma} \rangle N_{\lambda,\Gamma}(E_F) (A_L^0 \langle r^L \rangle |_{\lambda,\Gamma'_1}) \quad . \tag{35}$$

The Tables IV and V summarize the different coefficients involved in the Eqs. (32) and (35), for each type of conduction electron. In particular, starting from Eq. (34) we can verify that the total contribution of each type of cubic orbital (λ, Γ) to $A_2^0 \langle r^2 \rangle$ is zero, whilst their fourth- and sixth-order contributions previously obtained[10,11] are found again. The angular coefficients $K^L_{\lambda,\Gamma'}$ and $J^{L,l}_{\lambda,\Gamma'}$ were calculated as in Ref. 11 by a numerical integration of the electrostatic (direct plus exchange) interaction between the wave functions of one conduction electron and of the whole $4f$ shell. They are listed here in the form of the nearest fraction with a relative accuracy better than 10^{-8}.

TABLE IV. Value of the coefficients $\sigma^L_{\lambda,\Gamma}$ and $\sigma'^L_{\lambda,\Gamma}$ [see Eq. (33)] corresponding to the tetragonal and trigonal strain modes.

		Tetragonal strain mode			Trigonal strain mode	
(λ,Γ)	$\sigma^2_{\lambda,\Gamma}$	$\sigma^4_{\lambda,\Gamma}$	$\sigma^6_{\lambda,\Gamma}$	$\sigma'^2_{\lambda,\Gamma}$	$\sigma'^4_{\lambda,\Gamma}$	$\sigma'^6_{\lambda,\Gamma}$
$(1,t_{1u})$	$-\frac{1}{2}$	...	...	$-\frac{1}{2}$	...	...
$(2,e_g)$	-1	$\frac{1}{6}$	...		No splitting	
$(2,t_{2g})$	$-\frac{1}{2}$	-4		$-\frac{1}{2}$	$-\frac{1}{9}$	...
$(3,t_{1u})$	$-\frac{1}{2}$	$\frac{3}{8}$	$-\frac{5}{16}$	$-\frac{1}{2}$	$-\frac{17}{13}$	$\frac{3}{10}$
$(3,t_{2u})$		No splitting		$-\frac{1}{2}$	$-\frac{1}{9}$	$\frac{23}{2}$

TABLE V. Expression of $A^0_L\langle r^L\rangle$ for one conduction electron of character λ, wave function $\varphi^1_{\lambda,\Gamma}$, and tetragonal symmetry Γ'_1 issued from the cubic m_Γ-fold degenerated representation Γ [see Eq. (32)]; the corresponding matrix element of Y^0_2 is also given.

λ	Γ	m_Γ	Γ'_1	$\varphi^1_{\lambda,\Gamma}$	$\langle\varphi^1_{\lambda,\Gamma}\vert Y^0_2\vert\varphi^1_{\lambda,\Gamma}\rangle$	L	$A^0_L\langle r^L\rangle\vert_{\lambda,\Gamma'_1}$
1	t_{1u}	3	a_{2u}	Y^0_1	$\frac{1}{\sqrt{5\pi}}$	2	$\frac{1}{5}F^2-\frac{9}{140}G^2-\frac{5}{252}G^4$
2	e_g	2	a_{1g}	Y^0_2	$\frac{1}{7}\left(\frac{5}{\pi}\right)^{1/2}$	2	$\frac{1}{7}F^2-\frac{9}{98}G^1+\frac{11}{588}G^3-\frac{125}{6468}G^5$
						4	$\frac{1}{28}F^4-\frac{99}{3920}G^1-\frac{11}{980}G^3-\frac{5}{8624}G^5$
2	t_{2g}	3	b_{2g}	$\frac{1}{\sqrt{2}}(Y^2_2-Y^{-2}_2)$	$-\frac{1}{7}\left(\frac{5}{\pi}\right)^{1/2}$	2	$-\frac{1}{7}F^2+\frac{9}{98}G^1-\frac{11}{588}G^3+\frac{125}{6468}G^5$
						4	$\frac{1}{168}F^4-\frac{33}{7840}G^1-\frac{11}{5880}G^3-\frac{5}{51\,744}G^5$
3	t_{1u}	3	a_{2u}	Y^0_3	$\frac{2}{3\sqrt{5\pi}}$	2	$\frac{2}{15}F^2-\frac{5}{28}G^0-\frac{19}{1260}G^2+\frac{5}{308}G^4-\frac{625}{36\,036}G^6$
						4	$\frac{1}{44}F^4-\frac{9}{112}G^0+\frac{3}{280}G^2-\frac{97}{13\,552}G^4-\frac{75}{88\,088}G^6$
						6	$\frac{25}{1716}F^6-\frac{13}{224}G^0-\frac{13}{2016}G^2-\frac{13}{27\,104}G^4-\frac{25}{3\,171\,168}G^6$
3	t_{2u}	3	b_{2u}	$\frac{1}{\sqrt{2}}(Y^2_3+Y^{-2}_3)$	0		No contribution to the magnetoelastic coefficients

C. Trigonal strain mode

In this section we consider a pure trigonal deformation where the only nonzero strain parameters are

$$\epsilon_{xy}=\epsilon_{yz}=\epsilon_{zx}=\epsilon_{ij}=\frac{\omega}{\sqrt{2}} \quad , \tag{36}$$

ω being the angular displacement of each fourfold axis. As seen above it is more convenient to use the threefold axes system where the [111] direction is the new z axis.

1. *Point-charge model*

The ligands charges of the strained lattice generates an electric potential which may be written, in the new axes system, as

$$V'(\vec{r})=\sum_{LM} r^L \left(V_L'^M + \frac{\partial V_L'^M}{\partial \epsilon_{ij}} \epsilon_{ij} \right) Y_L^M(\hat{r}) \quad . \tag{37}$$

As in Sec. II B *1* the PC contribution of the trigonal magnetoelastic coefficients can then be derived using the expressions of Table II, for example [see Eqs. (13) and (14)]:

$$\left.\frac{B_2}{\alpha_J}\right|_{\mathrm{PC}} = 6\frac{e^2\langle r^2\rangle}{a^3} \sum_{\{j\}} q_{\{j\}} (S_7^{400} - 3S_7^{220})_{\{j\}} \quad , \tag{38}$$

$$\left.\frac{B_2^{(4)}}{\beta_J}\right|_{\mathrm{PC}} = -\frac{5}{12}\frac{e^2\langle r^4\rangle}{a^5} \sum_{\{j\}} q_{\{j\}} (S_{11}^{600} - 22S_{11}^{420} + 51S_{11}^{222})_{\{j\}} \quad . \tag{39}$$

2. *Band model*

As in Sec. II B *2* we suppose that the orbital degeneracy of the conduction electrons is lifted by the only potential $V_2'(\vec{r})$ originating in the shifted ligand charges

$$V_2'(\vec{r}) = r^2 \frac{\partial V_2'^0}{\partial \epsilon_{ij}} \epsilon_{ij} Y_2^0(\hat{r}) \quad . \tag{40}$$

The same formalism is then applicable for evaluating the contribution of the conduction band to $A_L'^0\langle r^2\rangle$. The degenerate cubic representations Γ are split by the trigonal symmetry (see Table III) and the perturbation potential $V_2'(\vec{r})$ shifts the energy of the

TABLE VI. Expression of $A_L'^0\langle r^L\rangle$ for one conduction electron of character λ, wave function $\varphi_{\lambda,\Gamma}'^1$ and trigonal symmetry Γ_1' issued from the cubic m_Γ-fold degenerated representation Γ [see Eq. (32)]; the corresponding matrix element of Y_2^0 is also given.

λ	Γ	m_Γ	Γ_1'	$\varphi_{\lambda,\Gamma}'^1$	$\langle\varphi_{\lambda,\Gamma}'^1\|Y_2^0\|\varphi_{\lambda,\Gamma}'^1\rangle$	L	$A_L'^0\langle r^L\rangle\|_{\lambda,\Gamma_1'}$
1	t_{1u}	3	a_{2u}	Y_1^0	$\frac{1}{\sqrt{5\pi}}$	2	$\frac{1}{5}F^2-\frac{9}{140}G^2-\frac{5}{252}G^4$
2	e_g	2	e_g	$\frac{Y_2^2+Y_2^{-2}}{\sqrt{6}}$ $+\frac{Y_2^1-Y_2^{-1}}{\sqrt{3}}$	0		No contribution to the magnetoelastic coefficients
2	t_{2g}	3	a_{1g}	Y_2^0	$\frac{1}{7}\left(\frac{5}{\pi}\right)^{1/2}$	2	$\frac{1}{7}F^2-\frac{9}{98}G^1+\frac{11}{588}G^3-\frac{125}{6468}G^5$
						4	$\frac{1}{28}F^4-\frac{99}{3920}G^1-\frac{11}{980}G^3-\frac{5}{8624}G^5$
3	t_{1u}	3	a_{2u}	$\frac{2}{3}Y_3^0$ $-\frac{1}{3}\left(\frac{5}{2}\right)^{1/2}(Y_3^3-Y_3^{-3})$	$-\frac{1}{6\sqrt{5\pi}}$	2	$-\frac{1}{30}F^2+\frac{5}{112}G^0+\frac{19}{5040}G^2-\frac{5}{1232}G^4+\frac{625}{144\,144}G^6$
						4	$\frac{13}{792}F^4-\frac{13}{224}G^0+\frac{13}{1680}G^2-\frac{1261}{243\,936}G^4-\frac{25}{40\,656}G^6$
						6	$\frac{125}{20\,592}F^6-\frac{65}{2688}G^0-\frac{65}{24\,192}G^2-\frac{65}{325\,248}G^4-\frac{125}{38\,054\,016}G^6$
3	t_{2u}	3	a_{1u}	$\frac{1}{\sqrt{2}}(Y_3^3+Y_3^{-3})$	$-\frac{\sqrt{5}}{6\sqrt{\pi}}$	2	$-\frac{1}{6}F^2+\frac{25}{112}G^0+\frac{19}{1008}G^2-\frac{25}{1232}G^4+\frac{3125}{144\,144}G^6$
						4	$\frac{1}{88}F^4-\frac{9}{224}G^0+\frac{3}{560}G^2-\frac{97}{27\,104}G^4-\frac{75}{176\,176}G^6$
						6	$-\frac{5}{6864}F^6+\frac{13}{4480}G^0+\frac{13}{40\,320}G^2+\frac{13}{542\,080}G^4+\frac{5}{12\,684\,672}G^6$

corresponding electronic state. The only differences are that: (i) the $V_2'(\vec{r})$ perturbation must be diagonal for each representation Γ in the threefold-axes system, that gives new wave functions $\varphi'^{p}_{\lambda,\Gamma}$; and (ii) the values of $\sigma'^{L}_{\lambda,\Gamma}$ are therefore different (see Table IV), as also the values of the angular CEF coefficients $K^{L}_{\lambda,\Gamma'}$ and $J^{L,l}_{\lambda,\Gamma'}$ [Eq. (32)]. Except for these modifications the parameters $A_L'^0\langle r^L\rangle$ have the same expression in the threefold-axes system as the parameters $A_L^0\langle r^L\rangle$ [Eq. (34)] in the fourfold axes one. We can deduce the contribution of the conduction electrons of symmetry (λ,Γ) to the trigonal magnetoelastic coefficients

$$-\frac{\partial}{\partial\epsilon_{ij}}(A_L'^0\langle r^L\rangle|_{\lambda,\Gamma})=-\frac{|e|}{m_\Gamma}(1-\sigma'^{L}_{\lambda,\Gamma})\langle r^2\rangle_\lambda\frac{\partial V_2'^0}{\partial\epsilon_{ij}}\langle\varphi'^{1}_{\lambda,\Gamma}|Y_2^0|\varphi'^{1}_{\lambda,\Gamma}\rangle N_{\lambda,\Gamma}(E_F)(A_L'^0\langle r^L\rangle|_{\lambda,\Gamma_1'}) \ . \tag{41}$$

Table VI summarizes the values of all the coefficients involved for this trigonal strain mode and for each type of conduction electron. Particularly we note that the expression of the wave functions $\varphi'^{1}_{\lambda,\Gamma}$ is different from the expression of Table V due to the different axes system. We can also verify that the total contribution of the conduction electrons of a given type (λ,Γ) to the cubic CEF parameters $A_L'^0\langle r^L\rangle$ [Eq. (11)] leads to the same conclusion as in Sec. II B *2*, after returning in the fourfold-axes system.

TABLE VII. Average value $\langle r^2\rangle_\lambda$, partial density of states $N_{\lambda,\Gamma}(E_F)$, and direct F^L and exchange G^1 Slater's integrals at the Fermi level, for conduction electrons of character λ and cubic symmetry Γ, in the compounds DyCu and DyZn. The cubic lattice parameters a and the effective charges q_j are also given (R, rare earth; M, alloyed metal).

	λ	1	2	3
	$\langle r^2\rangle_\lambda$ (Å^2)	1.70	1.36	2.07
	$N_{\lambda,\Gamma}(E_F)$ (10^{-5} K^{-1})	0.63 (t_{1u})	2.15 (e_g)	0.007 (t_{1u})
			2.93 (t_{2g})	0.113 (t_{2u})
	F^2 (K)	41 100	36 300	24 400
DyCu	F^4 (K)		16 500	11 200
($a = 3.455$ Å	F^6 (K)			7 360
$q_R = 3.44$	G^0 or G^1		14 800	19 500
$q_M = 1.49$)	G^2 or G^3	14 800	12 000	13 300
	G^4 or G^5	13 000	9 120	8 960
	G^6			6 610
	$N_{\lambda,\Gamma}(E_F)$ (10^{-5} K^{-1})	0.86 (t_{1u})	4.39 (e_g)	0.009 (t_{1u})
			1.90 (t_{2g})	0.080 (t_{2u})
	F^2 (K)	40 600	37 100	23 800
DyZn	F^4 (K)		16 500	10 800
($a = 3.565$ Å	F^6 (K)			7 000
$q_R = 2.98$	G^0 or G^1		15 200	18 300
$q_M = 1.60$)	G^2 or G^3	14 600	12 350	12 600
	G^4 or G^5	12 800	9 400	8 500
	G^6			6 300

III. APPLICATION TO CsCl COMPOUNDS

A. Numerical results

We present now a numerical application of the theoretical expressions derived in the preceding section using the results of self-consistent APW band calculations carried out for the two CsCl-type structure compounds DyCu (Ref. 16) and DyZn.[12] These compounds represent two peculiar cases of band structure: in the former case the copper $3d$ shell is located in energy within the conduction band and is therefore hybridized with it; in the latter case the zinc $3d$ shell is oppositely located below the conduction band. The main consequence is a strengthening of the d-t_{2g} character of the conduction electrons in DyCu (Ref. 10) to the detriment of the e_g character.

Starting from the band-structure results the evaluation of the effective charges q_j inside the ligand APW sphere j is made as following, in order to take into account partially the conduction electrons which modulate the charge Z_j of the ion ($Z_R = +3$, $Z_{\text{Cu}} = +1$, $Z_{\text{Zn}} = +2$),

$$q_j = Z_j - n_j + V_j \rho_{\text{ext}} , \qquad (42)$$

where n_j is the total number of conduction electrons within the APW sphere j of volume V_j and ρ_{ext} the mean electronic density outside the APW spheres. The used values are given in Table VII for both compounds.

Table VII provides also the numerical values of the partial densities of states at the Fermi level for each type of orbital character (from Ref. 10), as well as the average values $\langle r^2 \rangle_\lambda$ and the Slater's integrals F^L and G^1 obtained starting from APW radial wave functions. The main difference between DyCu and DyZn is the inversion of the type of d orbital at the Fermi level, the predominant one being the t_{2g} type for DyCu and the e_g type for DyZn.

Tables VIII and IX summarize, respectively, the numerical estimation of the tetragonal and trigonal magnetoelastic coefficients for both compounds. The PC estimation was made using the values given in the Tables I and VII.

About the conduction band contributions the Coulombic exchange and direct parts are always opposite to each other. However, the larger the L and λ values, the more important the exchange contribution relatively to the direct one: For example, the ratio exchange over direct contributions reaches -15% for $L = 2$ and p electrons ($\lambda = 1$), -25% for $L = 2$ and d electrons ($\lambda = 2$), -88% for $L = 4$ and $\lambda = 2$, and -1200% for $L = 6$ and $5f$ electrons ($\lambda = 3$). We find thus again the ratio previously evaluated for the cubic CEF parameters,[11,17] that is correlated with the expressions of the Tables V and VI in terms of Slater's integrals and angular coefficients [see Eq. (32)].

If we compare now the conduction band contribu-

TABLE VIII. Calculated tetragonal magnetoelastic coefficients arising from the point charges (PC) and from the conduction electrons of symmetry (λ, Γ) for the compounds DyCu and DyZn (the total represents the sum of the conduction-electron contributions).

		B_1/α_J (K)		$B_1^{(4)}/\beta_J$ (K)		$B_1^{(6)}/\gamma_J$ (K)	
	Origin	Direct	Exchange	Direct	Exchange	Direct	Exchange
	PC	−2610	⋯	−122	⋯	−20.3	⋯
	(1, t_{1u})	420	−62	⋯	⋯	⋯	⋯
DyCu	(2, e_g)	1040	−261	49	−43	⋯	⋯
	(2, t_{2g})	705	−178	−44	38	⋯	⋯
	(3, t_{1u})	1.5	−1.7	0.05	−0.29	0.04	−0.49
	Total [all (λ, Γ)]	2166	−503	5	−5.3	0.04	−0.49
	PC	−907	⋯	−100	⋯	−15.3	⋯
	(1, t_{1u})	197	−29	⋯	⋯	⋯	⋯
DyZn	(2, e_g)	749	−191	35	−31	⋯	⋯
	(2, t_{2g})	162	−41	−10	9	⋯	⋯
	(3, t_{1u})	0.65	−0.71	0.02	−0.12	0.02	−0.21
	Total [all (λ, Γ)]	1109	−262	25	−22	0.02	−0.21

TABLE IX. Calculated trigonal magnetoelastic coefficients arising from the point charges (PC) and from the conduction electrons of symmetry (λ, Γ) for the compounds DyCu and DyZn (nc not calculated; the total represents the sum of the conduction-electron contributions).

		B_2/α_J (K)		$B_2^{(4)}/\beta_J$ (K)		$B_2^{(6)}/\gamma_J$ (K)	
	Origin	Direct	Exchange	Direct	Exchange	Direct	Exchange
DyCu	PC	8540	...	−497	...	nc	...
	$(1, t_{1u})$	−1370	202	...	...	...	...
	$(2, t_{2g})$	−2300	581	−96.8	84.5	...	...
	$(3, t_{1u})$	−0.31	0.34	0.05	−0.31	0.004	−0.045
	$(3, t_{2u})$	−123	139	1.44	−8.39	0.57	−6.49
	Total [all (λ, Γ)]	−3793	922	−95	76	0.57	−6.53
DyZn	PC	2960	...	−449	...	nc	...
	$(1, t_{1u})$	−642	94	...	...	...	...
	$(2, t_{2g})$	−529	135	−21.8	19.5	...	...
	$(3, t_{1u})$	−0.13	0.14	0.02	−0.13	0.002	−0.019
	$(3, t_{2u})$	−29.6	32.0	0.34	−1.94	0.13	−1.50
	Total [all (λ, Γ)]	−1201	261	−21	17	0.13	−1.52

tion to the PC contribution the following remarks can be made.

(i) The $5f$ conduction electrons contribute only weakly to the modification of the sixth-order CEF parameter, due to the small amount of such electrons present at the Fermi level.

(ii) The PC contribution to the fourth-order magnetoelastic coefficients is also predominant: That results here essentially from the cancellation of the direct and exchange part of the band contribution. We have to recall that the situation is opposite to the case of the cubic fourth- and sixth-order CEF parameters which originate mainly in the conduction band.[11] That has to be related to the mechanism by which the conduction band is split for generating the magnetoelastic coefficients, that implies only the electronic states near the Fermi level are involved while the whole conduction band contributes to the cubic CEF parameters.

(iii) The predominance of the PC contribution vanishes in the case of the second-order magnetoelastic coefficients B_1 and B_2 for which the two contributions (PC and band) have the same order of magnitude but are however opposite in sign. It can also be noticed that the p type conduction electrons contribute here contrarily to the situation in cubic symmetry.

The comparison between DyCu and DyZn reveals only small differences which are mainly due to different values for the effective charges and the partial densities of states. In particular the antagonism between the e_g and t_{2g} contributions (see the beginning of this section and Ref. 10), which produces an inversion of the sign of $A_4^0 \langle r^4 \rangle$, has a much less pronounced effect on the magnetoelastic coefficients: a similar behavior in the vicinity of the Fermi level explains finally similar results for both compounds.

B. Comparison with experiment

1. Fourth- and sixth-order magnetoelastic coefficients

We want first to estimate the amplitude of a possible shift of the cubic CEF parameters $A_4^0 \langle r^4 \rangle$ and $A_6^0 \langle r^6 \rangle$, due to the magnetoelastic coefficients $B_i^{(4)}$ and $B_i^{(6)} (i=1,2)$. This shift is equal to $B_1^{(L)} \epsilon_3 (L=4,6)$ in the case of a tetragonal distortion, and to $B_2^{(L)} \epsilon_{ij}$ in the case of a trigonal one. In the rare-earth zinc and copper series we find the possible typical values[5] $\epsilon_3 \sim 10^{-2}$ and $\epsilon_{ij} \sim 10^{-3}$. That gives a shift of the order of 1 K for $A_4^0 \langle r^4 \rangle$ and less than 1 K for $A_6^0 \langle r^6 \rangle$ in the case of a tetragonal distortion, the situation being analogous for a trigonal distortion. Comparing with experimental typical values[18] ($A_4^0\langle r^4\rangle \sim -40$ K, $A_6^0\langle r^6\rangle \sim -20$ K) leads to neglect their variation due to magnetoelastic effects. That confirms *a posteriori* the hypothesis usually assumed to consider the only second-order magnetoelastic coupling in these intermetallic systems.

2. *Second-order magnetoelastic coefficients: R*Zn *series*

In previous papers[5,7] we reported experimental values for the second-order magnetoelastic coefficients B_1 and B_2 from spontaneous magnetostriction and parastriction measurements performed on the heavy rare-earth zinc series and the conclusions were: (i) the ratio B_1/α_J remains of the same order of magnitude, varying from ~ -1000 K/at. in TbZn to ~ -3000 K/at. in TmZn; and (ii) the variation of B_2/α_J is more erratic ($\sim +2000$ K/at. in Ho- and TmZn, but ~ 0 in Dy- and ErZn and ~ -1500 in TbZn).

Comparing with the present theoretical predictions for the only compound DyZn, we find a rather bad agreement with the experiment. Indeed, if the various contributions are of the same order of magnitude as the experimental values, it is not the case for the total. This is reminiscent of the evaluation of the cubic CEF parameters[11] which led to the same conclusion: a possible explanation was the failing of the APW model for describing quantitatively with accuracy these parameters. The theoretical results for DyZn have then to be taken as a starting point, more qualitative than quantitative, for estimating the possible order of magnitude of the magnetoelastic coefficients throughout the RZn series.

Concerning the variation of the magnetoelastic coefficients in the series, the following remarks can be made.

(i) The PC contribution is an important term; however obtaining a reliable theoretical value for each compound remains delicate: taking for example $q_R = 3.10$ and $q_M = 1.50$ for the effective charges in DyZn instead of 2.98 and 1.60 leads to strengthen the PC contribution by more than 50%.

(ii) The band contribution is far from negligible; in addition to the above source of variation [via $\partial V_2^0/\partial\epsilon_3$ or $\partial V_2'^0/\partial\epsilon_{ij}$ in Eqs. (35) and (41)], it may vary through another way. Indeed the contribution of each type of conduction electrons, proportional to the corresponding partial density of states at the Fermi level, is susceptible to alter drastically from a compound to another, in particular when the Fermi level lies near a peak of the density of states, that is the case in DyZn or in isomorphous compounds.[12]

(iii) The last uncertainty arises from the oversimple starting model for the conduction-electrons contribution, in which no self-consistency of the effect of the distortion on the band structure itself was taken into account. Such a modification of the band via the self-consistency may lead to a noticeable diminution or reinforcement of the magnetoelastic contribution of each type of conduction electrons.

3. *Second-order magnetoelastic coefficient: R*Cu *series*

Few experimental informations are available unfortunately for this series. The only known value is $B_1/\alpha_J \sim -3000$ K/at. for TmCu (Refs. 9 and 19); in addition the spontaneous tetragonal distortion observed in TbCu ($c/a - 1 = 1.14\%$ at $T = 4.2$ K),[20] a little larger than in TbZn, would lead to a B_1/α_J value of the order of -2000 K/at. The magnetoelastic coefficients B_1 seem thus to be of the same order of magnitude in both series, while no information is available for the coefficient B_2.

The main remark concerning the theoretical predictions in the rare-earth copper series is that the expected values for B_1 and B_2 are larger than in the zinc series: this is related principally to a larger PC contribution due to different values of the effective charges. That confirms the possibility of noticeable variations for the magnetoelastic coefficients from a compound to another one.

C. Conclusion

In this paper we studied thoroughly the origin of the one-ion harmonic magnetoelastic coefficients in cubic rare-earth intermetallic compounds. They may be considered as the strain derivatives of the cubic CEF parameters. It appears that they originate in numerous antagonistic contributions which may be of the order of magnitude of the experimental values and which arise from the surrounding localized charges as well as from the conduction electrons having an anisotropic orbital character around the $4f$ shell. In particular the p type electronic character of the band contributes to the magnetoelastic coefficients in addition to its d and f type character, that was not the case for the cubic CEF parameter.

On the other hand, contrarily to the case of the cubic CEF parameters, the PC contribution is here dominant, especially for the fourth- and sixth-order magnetoelastic coefficients. However these latter have a weak influence on the magnetic properties, a conclusion asserted previously from pure PC estimates[3]; the main effect of the magnetoelastic interactions remains here the appearance of a second-order CEF term in the Hamiltonian, which is zero for the nonstrained lattice.

Because of the various contributions, the magnetoelastic coefficients are liable to vary drastically from a compound to another. This variation may occur via a shift of the values of the effective charges for the PC contribution; for the conduction band contribution the variation may be related to the modification of the Slater's integrals as well as of the partial densities of states at the Fermi level. This latter source of variation was not present for the cubic CEF parameters which originate in the whole conduction band[11]; it is all the more important since the Fermi level lies near a peak of the density of states. This complex behavior explains the absence of scale law for the B_2/α_J variation through the RZn series for example, the smooth B_1/α_J variation being probably purely accidental.

This drastic dependence on the behavior of the conduction band at the Fermi level is reminiscent of the magnetic exchange integrals which have been found to clearly vary between two isomorphous compounds without any scale law[21,22]; that may also be related to the change of magnetic structure within a given rare-earth series, for example, in the RZn series[23] from a $(\pi 00)$-type antiferromagnetic structure (Ce-, Pr-, NdZn) to a ferromagnetic one (Sm-, Gd-, . . . , TmZn).

It can be noticed that in series where the metallic behavior is less obvious, as in rare-earth antimonides, we can expect a better agreement of the magnetoelastic coefficient with the PC predictions, as for the cubic CEF parameters, that is effectively the case.[3,24,25]

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