F-Tensor Coefficients in Electric Field Gradient Calculations for Non-Coulombic Potentials *

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The *F*-tensor coefficients are useful in evaluating the stress-induced electric field gradients in cubic crystals. These coefficients are derived for the non-Coulombic inter-atomic potentials Gr^{-n} and Ge^{-br}. Interesting differences are pointed out between these results and those for Coulombic potentials.

Key words: Nuclear Quadrupole Resonance, Electric Field Gradient.

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Introduction

A nucleus with spin $I \geq 1$ has a non-zero quadrupole moment Q. When such a nucleus is placed in a non-uniform electric field whose non-uniformity is described by the electric field gradient (EFG) q, the quadrupole interaction energy depends on the product qQ, which may be studied through nuclear quadrupole resonance. In a pure cubic metal like Al or Cu, the EFG vanishes at any nucleus due to symmetry considerations [1]. Elastic strain and/or imperfections can remove the cubic symmetry and lead to a finite EFG at any nuclear site.

Due to the presence of internal strains from crystal imperfections, Watkins and Pound [2] have shown that quadrupole interactions were strong in their alkali halide samples. In the semi-conductors InSb and GaSb, Shulman et al. [3] have reported quadrupole interaction due to applied stress. They introduced a relation connecting the EFG to the applied stress through a fourth order "gradient-elastic" tensor. A similar relation between the EFG and the strain, through a fourth order *F*-tensor, was utilized by Cohen and Reif [4]. In a cubic crystal, using Voigt notation, there are only three non-vanishing coefficients F_{11} and $F_{12} = F_{44}$ of this tensor. Expressions for these coefficients were obtained by Faulkner [5] for monovalent Cu, using a Coulomb inter-atomic potential.

In analyzing the impurity-induced EFG in Cu, Sagalyn et al. [6] labelled the strain-related EFG as the size EFG q^s, while the EFG due to the electric charge perturbation was labelled the valence EFG q^v. Now, imperfections produce inhomogeneous strain. The simplest approximation in this case is to assume that the EFG is determined by the local value of the strain. Within these limits, the above authors [6] evaluated q^s in Cu alloys. A modified version was given by Sagalyn and Alexander [7]. These two groups, and many others [8, 9], have used Faulkner's results for the *F*-tensor coefficients in Cu.

Efforts have also been made to evaluate q^s without using the *F*-tensor coefficients. Taking the discrete nature of the lattice into account and using the available lattice displacements, q^s has been evaluated (a) due to interstitial muons in Al and Cu, by Schmidt et al. [10]; (b) due to monovacancies in Al and Cu, by Hafizuddin and Mohapatra [11]; and (c) in V alloys, by Singh et al. [12]. Both q^v and q^s are combined in a unified approach by Dederichs et al. [13], using a multiple scattering KKR Green's function method. Schwarz and Blaha [14] have developed a new scheme for the evaluation of the EFG's in HCP metals, molecular crystals and high T_c superconductors. Their scheme is based on self-consistent energy band-structure calculations by the full potential linearized augmented plane wave method.

A general result for the *F*-tensor coefficients in any cubic system was obtained for Coulomb inter-atomic potentials in [15]. The present paper presents the details on these coefficients for some non-Coulombic potentials.

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1. Theory

Let us consider a reference atom at the origin O in a cubic host crystal. It will then be surrounded by shells of first *nn*'s, second *nn*'s etc. Let us focus on one *nn* at $\boldsymbol{r}(x, y, z)$. Due to this, at the site of the reference atom, let the potential be V' and its 2nd derivatives V'_{ij}. In the presence of stress, let our *nn* undergo a displacement $\boldsymbol{u}(\Delta x, \Delta y, \Delta z)$. The stress-induced change V_{ij} can be expressed in terms of V'_{ij} as

$$V_{ij} = \frac{\partial V'_{ij}}{\partial x}\Delta x + \frac{\partial V'_{ij}}{\partial y}\Delta y + \frac{\partial V'_{ij}}{\partial z}\Delta z\,. \qquad (1)$$

This is the stress-induced EFG at the origin. However, the reference atom itself would have shifted away from the origin because of the stress. The stress-induced EFG at the displaced position of the reference atom can be expressed by a Taylor series – the leading term being its value at the origin as in (1). The first correction term would involve the product of the displacement-components of the reference atom and the *nn* under consideration. Hence, it can be neglected or incorporated into a correction factor.

When the components of the displacement of the ion at $\boldsymbol{r}$ are expressed in terms of the strain components ε_{ij}, one obtains a complicated result. However, symmetry considerations lead to simplifications, so that one gets

$$V_{xx} = \left(x\,\frac{\partial V'_{xx}}{\partial x}\right)\varepsilon_{xx} + \left(y\,\frac{\partial V'_{xx}}{\partial y}\right)\varepsilon_{yy} + \left(z\,\frac{\partial V'_{xx}}{\partial z}\right)\varepsilon_{zz}\,,$$

$$V_{xy} = \left(y\,\frac{\partial V'_{xy}}{\partial x}\right)2\,\varepsilon_{xy}\,. \qquad (2)$$

Comparing this with the standard relation for a cubic crystal [6]

$$V_{ij} = \delta_{ij}\big[(F_{11} - F_{12})\,\varepsilon_{ii} + F_{12}\sum_k \varepsilon_{kk}\big] + (1 - \delta_{ij})\,2F_{44}\,\varepsilon_{ij}\,, \qquad (3)$$

one obtains

$$F_{11} = \sum\left(x\,\frac{\partial V'_{xx}}{\partial x}\right), \qquad F_{12} = \sum\left(y\,\frac{\partial V'_{xx}}{\partial y}\right), \qquad (4)$$

where the sum is over all the *nn*'s and $F_{44} = F_{12}$ due to the cubic symmetry.

2. Results and Discussion

A general inter-atomic potential is of the form $\Sigma_k\, a_k\, r^{-k}$, where $k > 0$, a_k is a constant and r is the inter-atomic distance. Let us consider the term $k = n$, i.e.

$$V' = G/r^n\,, \qquad n > 0\,. \qquad (5)$$

Using this in (4) and summing over the 1st *nn*'s at the distance r_1, we get

$$F_{11}\,(\mathrm{FCC}) = 2(2-n)\,n(n+2)\,G/r_1^{n+2}\,, \qquad (6)$$

$$F_{12}\,(\mathrm{FCC}) = -\,n\,n(n+2)\,G/r_1^{n+2}\,, \qquad (7)$$

$$F_{11}\,(\mathrm{BCC}) = (5-n)\,8\,n(n+2)\,G/9\,r_1^{n+2}\,, \qquad (8)$$

$$F_{12}\,(\mathrm{BCC}) = -\,(n+1)\,8\,n(n+2)\,G/9\,r_1^{n+2}\,. \qquad (9)$$

Using these in (3), the strain induced EFG q^{s} may be obtained if the strain components ε_{ij} are known by analytic means or otherwise. Further, we note the following: (i) If we put $n = 1$ (Coulomb potential) and $G = e$ in (6) and (7), we get Faulkner's results [5] for monovalent Cu. (ii) The results for FCC and BCC are similar but not identical. (iii) When G is negative, as in an attractive potential, since $n > 0$, (a) F_{12} is always positive, irrespective of the value of n, for both FCC and BCC. (b) F_{11} (FCC) is negative only when $n = 1$ (Coulomb potential), zero when $n = 2$ and positive for $n > 2$. F_{11} (BCC) is negative for $n < 5$, zero when $n = 5$ and positive when $n > 5$. (c) The above shows that F_{11} (and hence q^{s}) varies dramatically with the value of n. (d) When $F_{11} = 0$ ($n = 2$ for FCC and $n = 5$ for BCC), there is only one independent F-tensor coefficient $F_{12} = F_{44}$ so that great simplification occurs in the evaluation of q^{s}.

(iv) when $F_{11} - F_{12} = 2F_{44}$, it has been shown [6] that q^{s} is axially symmteric, so that the asymmetry parameter $\eta = 0$. The equations (6)–(9) show that this happens for both FCC and BCC only when $n = -4$, which is physically unacceptable. Thus, we see that any potential of the form G/r^n, $n > 0$ will yield a q^{s} with a non-zero η. (v) It has been shown [7, 9] that when $F_{11} - F_{12} = -3F_{44}$, great simplification occurs in the diagonalization of the q^{s}_{ij} matrix. Equations (6)–(9) show that the above condition holds only when $n = 1$, i.e. for the Coulomb potential.

When the inter-atomic potential

$$V' = G\exp(-\,b\,r) \qquad (10)$$

is substituted in (4) and the result summed over the 1st *nn*'s at a distance r_1, we get

$$F_{11}(\mathrm{FCC}) = 2bG\exp(-br_1)(3+3br_1-b^2r_1^2)/r_1\,, \quad (11)$$

$$F_{12}(\mathrm{FCC}) = bG\exp(-br_1)(1+br_1-b^2r_1^2)/r_1\,, \quad (12)$$

$$F_{11}(\mathrm{BCC}) = 8bG\exp(-br_1)(6+6br_1-b^2r_1^2)/9r_1\,, \quad (13)$$

$$F_{12}(\mathrm{BCC}) = 8bG\exp(-br_1)(-b^2r_1^2)/9r_1\,. \quad (14)$$

Using this in (3), the strain-induced EFG q^s may be obtained if the strain components ε_{ij} are known. Further, the following observations deserve special mention: (i) The results are similar excepting for F_{12} (BCC). (ii) When G is negative, as in an attractive potential, F_{12} (BCC) is always positive, while the other three may be positive, zero or negative-depending on the ratio $b^2r_1^2/(1+br_1)$. (iii) As mentioned earlier, when $F_{11}-F_{12}=2F_{44}$, q^s is axially symmetric so that the asymmetry parameter $\eta=0$. The equations (11)–(14) show that this can happen for both FCC and BCC only when $br_1=-1.5\pm i\sqrt{0.75}$, i.e. when the potential is complex! Thus, we see that a (real) exponential potential leads to an axially asymmetric q^s. (iv) Earlier it was mentioned that great simplification occurs in the evaluation of q^s when $F_{11}-F_{12}=-3F_{44}$. Equations (11)–(14) show that this happens for both FCC and BCC only when $br_1=1+\sqrt{3}$.

3. Summary

F-tensor coefficients are useful in the evaluation of the strain-induced EFG. In this paper, these coefficients are evaluated for the non-coulombic interatomic potentials $\Sigma_k a_k r^{-k}$ and $\mathrm{G}e^{-br}$ in cubic systems. The results are different for FCC and BCC. Further, the above potentials lead to axially asymmetric EFGs. They also show interesting differences from those obtained for Coulombic potentials. The relation $F_{11}-F_{12}=-3F_{44}$, which simplifies the evaluation of q^s, is used by many authors [7–9]. It is not valid for the above non-Coulombic potentials.

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