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## Revised Dielectric Parameters of Alkali and Halide Ions

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IN a previous publication[1] a classical model of a polarizable ion was suggested which was consistent with the experimental data for dielectric constants, indices of refraction, and infrared absorption frequencies of all the alkali-halide crystals. The ionic model was defined by three linear stiffness coefficients or "springs" between the nucleus, the electron cloud, and the external boundary. An alternative set of parameters to describe the same model was comprised of an effective nuclear charge $Ae$, a nuclear compliance or polarizability $B$, and an electronic compliance $C$.

The dielectric polarizability $\alpha_D$ of a single ion in a crystal was written $A^2B+C$ and the optical polarizability $\alpha_g$ of an isolated "gaseous" ion was $(A-1)^2B+C$ for alkali ions or $(A+1)^2B+C$ for halides. The polarizability per ion pair at optical frequencies in a crystal was somewhat less, namely:

$$\alpha_e=[(A_1+A_2)^2/(1/B_1)+(1/B_2)]+C_1+C_2\cong C_1+C_2, \tag{1}$$

where the subscripts 1 and 2 refer to the alkali and halide ions, respectively.

Tentative values of the parameters $A$, $B$, and $C$ were given in which $B$ and $C$ for lithium had been arbitrarily set equal to zero. It is now possible to choose more definite values for the parameters of lithium which lead to a new set of values for the other ions as shown in Table I. The dielectric constants, etc., calculated from the new ionic parameters are identical with those calculated from the original values. The table also gives values of the dielectric polarizability $\alpha_D$ and the electronic polarizability of free ions $\alpha_g$.

The new values are chosen so as to fulfill the following conditions for lithium: (1) $\alpha_g=0.4\text{A}^3$, in accord with the calculations of Pauling;[2] (2) $\alpha_D=11.7$, in agreement with the author's previous results;[3] (3) the ratios of the respective stiffness coefficients of lithium are to compare systematically to those of the other alkali ions.

In spite of the vagueness of the last condition, the resulting values are quite well defined. It turns out that the stiffness coefficient "$b$" between the nucleus and the boundary is in every case negative. This coefficient will indeed have to be negative if the actual forces between the nucleus and the neighboring ions are predominantly electrostatic, since it is axiomatic in classical mechanics and electrostatics that a charged particle cannot be supported at rest in stable equilibrium by purely electrostatic forces.

TABLE I. Revised ionic parameters (all but $A$ are in cubic angstroms).

| | $A$ | $B$ | $C$ | $\alpha_D$ | $\alpha_g$ |
|---|---|---|---|---|---|
| Li | 1.108 | 9.3 | 0.28 | 11.7 | 0.4 |
| Na | 1.234 | 10.7 | 3.40 | 19.7 | 4.0 |
| K | 1.291 | 17.4 | 12.39 | 41.4 | 13.9 |
| Rb | 1.449 | 19.2 | 17.00 | 57.3 | 20.9 |
| Cs | 1.215 | 25.3 | 34.14 | 71.5 | 35.3 |
| F | −0.780 | 25.9 | 10.04 | 25.8 | 11.3 |
| Cl | −0.645 | 66.3 | 35.22 | 62.8 | 43.6 |
| Br | −0.627 | 78.1 | 50.32 | 81.0 | 61.2 |
| I | −0.568 | 99.9 | 77.14 | 109.4 | 95.8 |

As an extreme illustration, suppose that we take away all the electrons and consider the motion of a lone nucleus of charge $pe$ inside a spherical conducting boundary of radius $R$. The electrostatic force on such a particle displaced a distance $X$ from the center is

$$F_n=+p^2e^2X/4\pi\epsilon_0R^3=-bX$$

or

$$b=-p^2e^2/4\pi\epsilon_0R^3, \tag{2}$$

where $\epsilon_0$ is the permittivity of free space. Equation (2) gives values of $b$ which are of the right order of magnitude if $R$ is chosen to be about three times the usual ionic radius. This would correspond to a reasonable average distance between the nucleus and other particles with which it has a strong electrostatic interaction.

The electronic polarizabilities of free ions are given roughly by the formula $\alpha_g=2.9R^{4.7}$, where $R$ and $\alpha_g$ are given in angstrom units. The polarizabilities of noble gas atoms satisfy the same equation if Pauling's "univalent" radii are substituted for $R$.

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## $2^{L-1}$-Magnetic $2^L$-Electric Interference Terms in γ-γ-Angular Correlations

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IT is the purpose of this note to point out that the relative phases of the reduced (magnetic quantum number independent) matrix elements for the simultaneous emission of a magnetic $2^{L-1}$-pole quantum and an electric $2^L$-pole quantum in a transition between eigenstates of angular momentum are determined almost completely by the time-reversal properties[1] of the emitting system. In the notation of Ling and Falkoff,[2] the quantities $\alpha$ and $\beta$ can both be made real by choice of nuclear phases, so that the phase angle $\delta$ can have only the value 0° or 180°. Which of these two applies would have to be determined from a more detailed model of the nucleus.

The reduced matrix elements in question are:

$$\beta_l=(j\|e_l\|j')=\Sigma_{mm'n}(-1)^{j-m+n}(jj'-mm'|jj'l-n)\times(jm|e_l{}^n|j'm') \tag{1a}$$

for electric $2^l$-pole transitions;

$$\alpha_l=(j\|m_l\|j')=\Sigma_{mm'n}(-1)^{j-m+n}(jj'-mm'|jj'l-n)\times(jm|m_l{}^n|j'm') \tag{1b}$$

for magnetic $2^l$-pole transitions between levels $j'\rightarrow j$. The matrix elements for the individual components of $j'\rightarrow j$ are[3]

$$(jm|e_l{}^n|j'm')=-ik^{-1}[l(l+1)]^{-\frac{1}{2}}\int\mathbf{j}(\mathbf{r})\cdot\nabla\mathbf{x}(\mathbf{r}\mathbf{x}\nabla)f_l(kr)Y_l{}^n(\mathbf{r})d\mathbf{r} \tag{2a}$$

$$(jm|m_l{}^n|j'm')=-[l(l+1)]^{-\frac{1}{2}}\int\mathbf{j}(\mathbf{r})\cdot\mathbf{r}\mathbf{x}\nabla f_l(kr)Y_l{}^n(\mathbf{r})d\mathbf{r}, \tag{2b}$$

where $f_l(x)=(8\pi^3/x)^{\frac{1}{2}}J_{l+\frac{1}{2}}(x)$, $Y_l{}^n(\mathbf{r})$ is the Condon and Shortley[4] spherical harmonic, and

$$\mathbf{j}(\mathbf{r})=\Sigma_i\int\Psi_j{}^m(X)\dagger\mathbf{j}_i(\mathbf{r})\Psi_{j'}{}^{m'}(X)d_iX. \tag{3}$$

In Eq. (3), $X$ is the set of nuclear coordinates; $\Psi_{j'}{}^{m'}(X)$, $\Psi_j{}^m(X)$ are the initial and final nuclear state functions; and the integral is over all nuclear coordinates except the coordinate $\mathbf{r}$ of the $i$th nuclear particle. The dimensionless current operator $\mathbf{j}_i(\mathbf{r})$ has the form $\mathbf{j}_i(\mathbf{r})=(\mathbf{v}_i/c)$ for a positively charged elementary particle. The normalization is such that $R$ quanta/sec of frequency

$\nu=(ck/2\pi)$ are emitted in the transition, where

$$R=\nu(e^2/hc)\sum_{l=1}^{\infty}[|(j\|e_l\|j')|^2+|(j\|m_l\|j')|^2]/[(2l+1)(2j'+1)].$$

For any wave function $\Psi$, the "time-reversed" wave function[1] is $K\Psi=U\Psi^*$, where $U$ is a hermitian unitary operator ($U=1$ for spinless nucleons, $U=\sigma_y$ for spin$=\frac{1}{2}$ nucleon). $K$ is not a linear operator. If the nuclear level $j$ has no other degeneracies than that of the magnetic quantum number, it can be shown that

$$K\Psi_j{}^m(X)=U\Psi_j{}^m(X)^*=\kappa_j(-1)^{j-m}\Psi_j{}^{-m}(X),\tag{4}$$

where $\kappa_j$ is a phase factor of modulus unity which depends on the choice of nuclear phase. The property $K\mathbf{j}\Psi=-\mathbf{j}K\Psi$, (all $\Psi$), which can be written in the form

$$U\mathbf{j}_i(\mathbf{r})^*U=-\mathbf{j}_i(\mathbf{r}),\tag{5}$$

expresses the fact that the current changes sign under reversal of the direction of time.

Combination of Eqs. (1) to (5) and use of the following identity for the vector-addition coefficients,

$$(jj'-mm'|jj'l-n)=(-1)^{j+j'-l}(jj'm-m'|jj'ln),$$

obtained from Wigner's formula,[5] shows that

$$\beta_l{}^*=\kappa_j{}^*\kappa_{j'}(-1)^l\beta_l\tag{6a}$$

$$\alpha_l{}^*=\kappa_j{}^*\kappa_{j'}(-1)^{l+1}\alpha_l.\tag{6b}$$

Assuming that the nuclear levels have definite parities, the nonvanishing $\beta_l$ will be those for which $l$ is even (odd) and the nonvanishing $\alpha_l$ will be those for which $l$ is odd (even), so that the phase factors in (6a) and (6b) are the same; by proper choice of phase for the levels $j'$ and $j$ the nonvanishing $\alpha_l$ and $\beta_l$ may be made real for all $l$. Only the signs and magnitudes of $\alpha_l$ and $\beta_l$ are left undetermined by these considerations. The relative phases cannot be used as arbitrary parameters to fit the data.

I wish to thank Professor John Blatt for stimulating discussions on time-reversal and on the multipole radiation moments.

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# Nonindependent Observational Equations in the Theory of Least Squares

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In recent years several very precise measurements have been performed which yield information on the values of the fundamental physical constants $e$, $m$, $h$, $N$, respectively the charge and mass of the electron, Planck's constant, and Avogadro's number. The recent measurements, such as those by (1) Hipple, Sommer, and Thomas,[1] (2) Thomas, Driscoll and Hipple,[2] (3) Gardner and Purcell,[3] (4) Taub and Kusch,[4] and (5) Nafe and Nelson[5] are unfortunately not as direct determinations of functions of $e$, $m$, $h$, $N$ as one is accustomed to in direct measurements of $h/e$ or $e/m$. For example, item (4) can be combined with item (5) to obtain $e^2/h$, with item (1) to obtain $Nm$, and with item (2) to obtain $e/m$. This clearly implies that the errors in the numerical values obtained for $e^2/h$, $Nm$, and $e/m$ are not independent; and, therefore, the usual least squares analysis to determine an unbiased set of "best values" of the atomic constants must be modified so that it properly takes into consideration the nonindependence of the data.

In the usual application of the theory of least squares the implicit assumption is made that if $P_i(\epsilon_i)$ is the probability of obtaining an error $\epsilon_i$ in the measurement of the $i$th quantity, then the probability $P(\epsilon_1, \epsilon_2, \epsilon_3, \cdots \epsilon_n)$ of obtaining simultaneously the errors $\epsilon_1, \epsilon_2, \cdots \epsilon_n$ in the least squares fitted set of quantities is just equal to $P_1(\epsilon_1)\cdot P_2(\epsilon_2)\cdots P_n(\epsilon_n)$. Such a situation is possible only if the errors are unrelated. The observational equations are usually written in a linearized form as

$$a_ix+b_iy+\cdots+f_iw=s_i+\epsilon_i,\tag{1}$$

where $x, y, \cdots w$ are the variables to be fitted, $a_i, b_i, \cdots f_i$ are known coefficients, $s_i$ is the observed quantity, and $\epsilon_i$ its (unknown) error. The method of least squares is based on the criterion of finding values of the variables which have the highest probability of being correct. When these errors are independent, this leads to the conditions that

$$\Sigma_i p_i\epsilon_i{}^2=\Sigma_i p_i(a_ix+b_iy+\cdots+f_iw-s_i)^2=\text{minimum}.\tag{2}$$

The "weight" $p_i$ of each equation is given as $1/\sigma_i{}^2$, the reciprocal of the mean square deviation of $s_i$.

In many cases (such as the one that prompts this note) it may be inconvenient to cast the observational equations into the above independent form. If such is the case, let the equations be written instead as

$$A_ix+B_iy+C_iz+\cdots+F_iw=S_i+E_i,\tag{3}$$

in which the quantities $S_i$ are not the results of single observations, but are each compounded from several observed quantities which may be involved in more than one $S_i$. (The case in which $S_i$ is compounded from several observed quantities, none of which, however, appears in any of the other $S_j$, is considered in Whittaker and Robinson;[6] but the more general case seems to have been neglected.) If we write $S_i$ as functions of the independent variables $s_j$, we have $dS_i=\Sigma_j\alpha_i{}^jds_j$ (i.e., $E_i=\Sigma_j\alpha_i{}^j\epsilon_j$). If the "weights" associated with the $s_j$ are $p_j=\sigma_j{}^{-2}$, we can define a weight matrix $\pi_{ik}$ which is the inverse of the error matrix $\rho_{ik}=\Sigma_j\alpha_i{}^j\alpha_k{}^j\sigma_j{}^2$. Equation (2) is then replaced by the quadratic form

$$\Sigma_{ik}\pi_{ik}(A_ix+B_iy+C_iz+\cdots+F_iw-S_i)\times(A_kx+B_ky+\cdots+F_kw-S_k),\tag{4}$$

which leads to "normal" equations with coefficients of a generalized form. A typical coefficient would be $\Sigma_{ik}\pi_{ik}A_iB_k$ in place of the coefficient $\Sigma_ip_ia_ib_i$ associated with the quadratic form of Eq. (2).

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# On the Angular Correlation Theorem

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In the theory of the angular correlation between successive particles emitted by a system undergoing a double transition, among states of definite total angular momentum, the following theorem appears. If the $z$ axis of quantization is along the direction of emission of one of the particles, the probability of the double transition is given by the product of the probabilities of each of the two successive transitions, summed over those intermediate states that are degenerate in the magnetic quantum number. Interference terms are absent.

This property of the double transition, first employed by Hamilton[1] for $\gamma-\gamma$-emission, was verified for a number of other particular cases by Falkoff and Uhlenbeck.[2] Recently, Lloyd[3] has given a general proof for arbitrary decay particles by applying group-theoretical arguments to second-order perturbation theory.

According to the physical interpretation of the quantum-mechanical formalism, however, the probability of a transition from an initial to a final state, via a set of intermediate states, contains interference terms only when it is not possible to measure the system in the intermediate state; if the experiment is inherently capable of specifying the particular intermediate state the