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Relationship between Dielectric Responses and Constituent Atoms in Crystal Materials

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Introduction. Material characterization for optical device purposes relies on the knowledge of material compliances related to the dielectric response. These include both linear (static and high-frequency dielectric constants) and various orders of nonlinear response (Kerr and Pockels effects, second harmonic generation, light scattering, etc.). Most of the properties of materials depend sensitively on the nature of the chemical bonding structure between the constituent atoms or ions. Therefore, there have been some interesting theoretical attempts to understand the relationship between the dielectric response of solids and constituent atoms or ions from various viewpoints [1, 2, 3, 4]. Yet up to date, there is no universal method for the determination of dielectric responses. In this note we will discuss the correlation between dielectric responses and composition variations in crystal materials.

Theory. Previous works [3, 5] have shown that the dielectric constants of solid state compounds are correlated to the average atomic number of the constituent atoms Z_{av} . This is based on the fact that the dielectric constant of a solid, ϵ , may be expressed in terms of the polarizabilities of its constituent atoms, α_i :

$$\epsilon = 1 + \frac{\sum N_i \alpha_i / \epsilon_0}{1 - \sum \gamma N_i \alpha_i / \epsilon_0}, \quad (1)$$

where N_i is the number of atoms of species i per unit volume, ϵ_0 is the free-space permittivity, and γ is the Lorentz factor. Since α_i may be considered to be linearly related with the atomic number Z , the dielectric constant, ϵ , may be expressed as [3]

$$\epsilon = (a_1 - b_1 Z_{av})^{-1} \quad \text{or} \quad \epsilon = a'_1 - b'_1 Z_{av}, \quad (2)$$

where a_1 and b_1 or a'_1 and b'_1 , respectively, are constants for each group of solids, Z_{av} is defined as the mean of the atomic numbers of the constituent atoms. For the case of e. g. an $A_{1-x}B_xC$ type compound, $Z_{av} = [(1-x)Z_A + xZ_B + Z_C]/2$.

On the basis of the optical properties of all chemical bonds, the NLO tensor coefficient d_{ij} of a crystal material can be expressed as [6, 7]

$$d_{ij} = \sum_{\mu} \frac{G_{ij}^{\mu}}{d^{\mu} q^{\mu}} \left\{ N_b^{\mu} (0.5) \frac{(Z_A^{\mu})^* + n(Z_B^{\mu})^*}{(Z_A^{\mu})^* - n(Z_B^{\mu})^*} f_i^{\mu} (\chi_b^{\mu})^2 + \frac{N_b^{\mu} s (2s-1) (r_0^{\mu})^2}{(r_0^{\mu} - r_c^{\mu})^2} f_c^{\mu} (\chi_b^{\mu})^2 \rho^{\mu} \right\}, \quad (3)$$

where ρ^{μ} is the difference in the atomic sizes, r_c^{μ} is the core radius, q^{μ} is the bond charge of the μ -th bond, and G_{ij}^{μ} is the geometrical contribution of chemical bonds of type μ . All of the above parameters can be deduced from the detailed chemical bonding structures of all constituent atoms [6, 7]. For a certain group of solids with a similar crystal structure, the parameters in Eq. (3) only change with different constituent atoms, the dependence on the mean of the atomic numbers can be approximated – again linearly – by $d_{ij} = a_2 - b_2 Z_{av}$ where a_2 and b_2 are constants for each group of solids.

Examples. The theoretical results are illustrated by two example materials, CdS_xSe_{1-x} , a typical semiconductor, and $Li_{1-x}Nb_{1+x/5}O_3$, an important electrooptic material. In Fig. 1 typical linear and nonlinear responses are plotted as a function of the average atomic number for a wide range of the composition parameter x . For CdS_xSe_{1-x} , $x = 0 \dots 1$ (data taken from [8] and [9]); for $Li_{1-x}Nb_{1+x/5}O_3$, $x = 0 \dots 0.092$ (for details see [10]). The linear dependencies are quit apparent.

Conclusion. From chemical bonding structure consideration it could be derived that in closely related crystal materials not only the linear [3, 5] but also the nonlinear dielectric responsivities

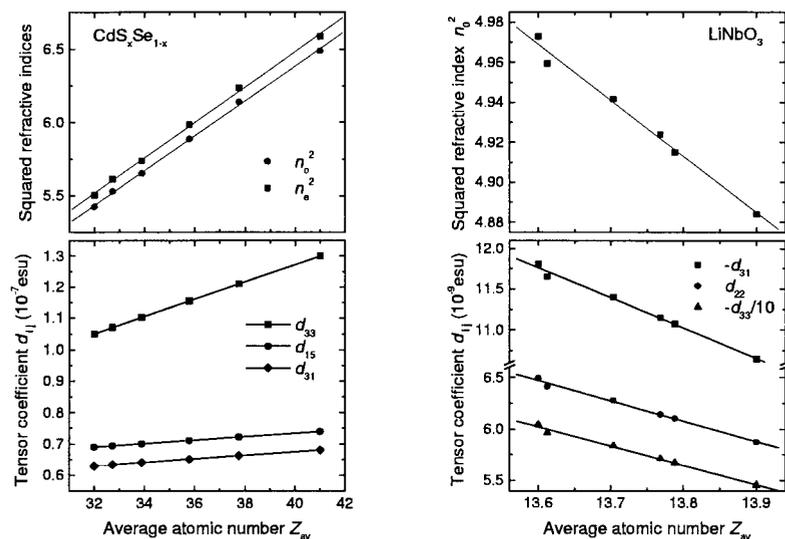


Fig. 1. Composition dependence of the linear (top) and nonlinear (bottom) dielectric responses for $\text{CdS}_x\text{Se}_{1-x}$ (left) and $\text{Li}_{1-x}\text{Nb}_{1+x/5}\text{O}_3$ (right). Experimental data are denoted by dots for compositions of $x = 1.00, 0.92, 0.79, 0.58, 0.36, 0.00$ in the case of $\text{CdS}_x\text{Se}_{1-x}$ and $x = 0.000, 0.012, 0.032, 0.052, 0.058, 0.092$ in the case of $\text{Li}_{1-x}\text{Nb}_{1+x/5}\text{O}_3$ (left to right)

are linearly dependent on the average atomic number of constituent atoms Z_{av} . Thus for calculations in many cases simple interpolation schemes can be applied.

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