The electron density in the four 'isoelectronic' alkali-halide crystals (NaF, KCl, RbCl, CsCl) by the method of augmented-spherical waves (ASW) in the self-consistent density-functional theory (DFT). We have verified the quality of the computed electron density by evaluating the heats of formation of the salts from total energy calculation, then computed structure factors for the lower-order 'sum' and 'difference' reflections, the latter being particularly sensitive in ionic deformations. The computed LDA structure factors for the 'sum' reflections agree well with those obtained from Hartree-Fock (HF) free-ion wavefunctions, while those for the 'difference' reflections are smaller than the HF values. This points to a 'contraction' of the anion and a 'dilation' of the cation in passing from free ions to ions in crystals. The contraction of the anion in NaCl has, in fact, been found also from pseudopotential LDA calculations (W. Andreoni, K. Maschke and M. Solimena, Phys. Rev., B26, 2314) and from Lowdin-type HF calculations (F. Gygi, K. Maschke, and W. Andreoni, 1984, Solid State Comm., 56, 437).

For NaF, the fitting of the computed LDA structure factors for the 'sum' reflections to the recent -ray data by H.C. Schmidt, R. Colella and D.R. Yoder-Short (1985, Acta Cryst., A41, 171) with a single Debye-Waller factor, gives also excellent agreement with the only 'difference' reflection measured by Schmidt et al. (the 100 reflection).

For NaCl, the same holds true for several difference reflections from the -ray data by Howard and Jones (1977, Acta Cryst., A33, 796) while an unexplained discrepancy remains with the earlier -ray data by Yoder and Colella (1982, Phys. Rev., B26, 2545). For RbCl and CsCl we have been unable to find accurate experimental data. In 1977, Zunger and Freeman (Phys. Rev., B16, 2901) had already reported structure factors computed in LDA with the self-consistent (non-muffin-tin) IGLO method for LiF and their results have recently been found to be in good agreement with the -ray data by Schmidt et al. (see Böbel et al., 1985, Acta Cryst., A41, 175).

More recently, Jansen and Freeman (1986, Phys. Rev., B33, 8629) have reported structure factors for NaCl computed in LDA with the full-potential linearized augmented-plane-wave method (FLAPW) which includes non-spherical components of the lattice potential which are ignored in our ASW calculations. They find large discrepancies (up to 20%) with (relatively old) experimental -ray structure factors for the high-order 'difference' reflections, using the theoretical Debye-Waller factors given by Sch questi (1957, Z. Naturforsch., 12, 983). We note however that the experimental Debye-Waller factors by Witte and Wolfel (1955, Z. Phys. Chem., 2, 296) or by Renninger (1952, Acta Cryst., 5, 711) eliminate the disagreement for the high-order 'difference' reflections without effecting the agreement for the 'sum' reflections.

Finally we should like to emphasize that Froyen and Cohen (1984, Phys. Rev. B, 30, 3770) have been able to account from first principles for the high pressure phase transition of NaCl to the ceci structure by using the electron density computed by pseudopotential LDA methods which include non-spherical components of the potential for the valence electrons (1-dependent pseudopotentials).

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