Experimental verification of electron redistribution caused by an external electric field

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In the recent years a lot of theoretical work was done to understand the mechanism of dielectric screening in crystals, especially in tetrahedrally coordinated insulators and semiconductors. On the one hand, the distorsion of the entire unit cell and the displacement of atoms within the unit cell under external perturbations (*e.g.* Piezo-effect and internal strain) were subject of interest [1]. On the other hand the redistribution of the electron density (with fixed ions) [2] caused by external perturbations was treated theoretically.
The comparison of the theoretical predictions and experimental results was performed on the basis of rather macroscopic quantities, e.g. the coefficients of the strain or dielectrical tensors. This is due to the fact that only a few experiments have been performed probing the effects on the microscopic scale. Fujimoto [3] and later Mahlberg [4] measured the variation of the integral intensity of some weak Bragg-reflections of GaAs caused by an externally applied electric field. These experiments showed up the order of magnitude of the effect, but they suffered from the low intensity and the fixed wavelength of conventional X-ray sources.
To verify these first findings the experiment was repeated in the last two years at HASYLAB, beamline D3, and extended sets of reflections for GaAs and ZnSe were measured at several wavelengths close to the absorption edge of the anion. A description of the experiment and first results are given in [5] and an extended version in [6]. As reported earlier [5], an almost linear dependence of the intensity variation on the field-strength was found for low order reflections, but a still not understood nonlinear behaviour for high order reflections of GaAs. In ZnSe the non-linearity is much weaker.
In the recent experiments the dependence of the effect on the wavelength of the synchrotron radiation was determined. A pronounced increase of the intensity variation is found close to the absorption edge of the anions: Two orderes of magnitude for low order reflections and a factor of 2 for high order reflections. This can be understood with the following model: the electron density is subdivided into the spherical (atomic) contributions centered at the atomic sites and the 'rest' represented by the bond charge. This leads to a structure factor for the weak reflections ($h+k+l=4n+2$) where the phases of the anionic contributions are -1 (including anomalous dispersion)

$$F = \left(f_{\mathrm{Ga}} + f'_{\mathrm{Ga}} - f_{\mathrm{As}} - f'_{\mathrm{As}} + \sum \Re[f_{\mathrm{BC}} \exp(\mathrm{i}\mathbf{kr})]\right) + \mathrm{i}\left(f''_{\mathrm{Ga}} - f''_{\mathrm{As}} + \sum \Im[f_{\mathrm{BC}} \exp(\mathrm{i}\mathbf{kr})]\right)$$

If $f_{\mathrm{Ga}} + f'_{\mathrm{Ga}} - f_{\mathrm{As}} - f'_{\mathrm{As}} \approx 0$ (which is the case close below the As-absorption edge), the integral intensity R is proportional to

$$|F|^2 \approx \left(f''_{\mathrm{Ga}} - f''_{\mathrm{As}} + \sum \Im[f_{\mathrm{BC}} \exp(\mathrm{i}\mathbf{kr})]\right)^2$$

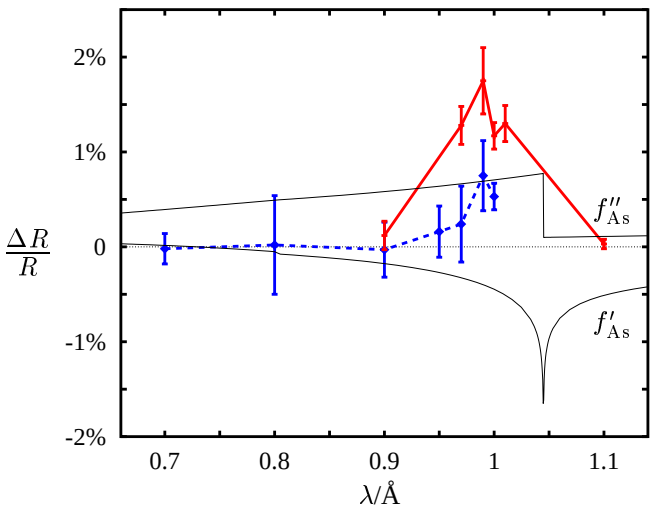
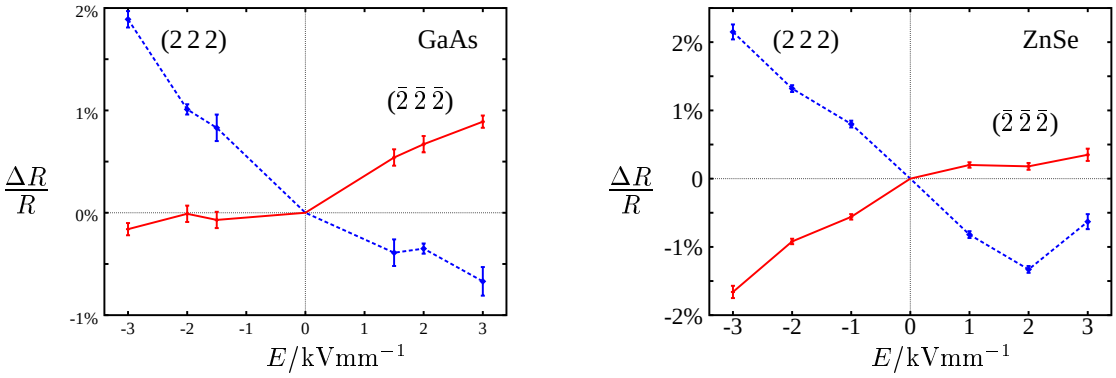


Figure 1: Relative intensity variation $\Delta R/R$ for the GaAs reflection (2 2 2) as function of the wavelength. The dashed (blue) line connects data obtained with a field-strength of $E = -1465\,\mathrm{Vmm}^{-1}$, the solid (red) line such with $E = -2935\,\mathrm{Vmm}^{-1}$. The thin solid lines are the coefficients of the anomalous dispersion of As f' and f'' (Cromer and Liebermann), scaled by a factor of 0.2.

Figure 2: Relative intensity variation $\Delta R/R$ for the reflections (2 2 2) and $(\bar{2}\,\bar{2}\,\bar{2})$ of GaAs (left) and ZnSe (right) as functions of the field-strength. The wavelength is 0.99 Å for GaAs and 0.96 Å for ZnSe. The field is parallel [1 1 1].



and therefore the intensity variation is dominated by the term $f''_{\mathrm{As}} \sum \Im[f_{\mathrm{BC}}]$. $|F|^2$ is connected to the experimentally determined relative intensity variation by

$$\frac{\Delta R}{R} \approx \frac{|F_E|^2 - |F_0|^2}{|F_0|^2}$$

From this dependence we can conclude that the measured effect of the low indexed reflections is mainly caused by the aspheric part of the charge density. This may be a redistribution of the charge in the bond-region or the formation of a dipole at the cores (though the latter one should mainly affect high order reflections).
As simulations with a bond-charge model show, the influence of the Piezo-effect is negligible. In contrast, the relative shift of the sublattices (internal strain) has to be considered. A combination of internal strain and bond-charge redistributon could 'describe' the findings, except for the pronounced non-linearity. Theoretical calculations based on DFT are in work in order to find a better qualitative interpretation of the experiments.

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