Study of Phonon Effects by Resonant "Forbidden" Reflections

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Introduction

In this report, we present a survey of vibration effects on the anisotropy of x-ray resonant scattering and discuss thermal-motion-induced (TMI) and point-defect-induced (PDI) "forbidden" reflections. In view of the very different x-ray and phonon energies it is rather surprising that phonons can drastically affect both the intensity and the spectrum of resonant x-ray scattering in crystals [1-5]. The reason is that resonant scattering is extremely sensitive even to very small atomic displacements (owing to distortions of electronic states by the displacements). Moreover, atomic displacements can change the symmetry of an atomic site, and thus induce an additional anisotropy of the atomic scattering factor near an absorption edge and therefore give rise to extra Bragg reflections, otherwise forbidden.

Methods and Materials

Strongly temperature dependent TMI reflections were recently observed in Ge (the 002 and 006 reflections) [3,4] and in ZnO (the 113 and 115 reflections) [5]. Drastic changes of the diffraction spectra were found in ZnO (fig. 1), contrary to the rather small changes observed for Ge.

Phenomenological consideration of the phenomenon, explains satisfactory the thermal growth of the intensity. Mainly optical phonon modes contribute to the effect, but for best agreement with the experimental results in Ge the different frequencies must be attribute to the atomic vibrations parallel and perpendicular to the chemical bonds [4]. The phenomenological description was also carried out for ZnO taking into account different phonon modes for TMI mechanism. Apart from the TMI mechanism, the dipole-quadrupole resonant contribution must be taken into account, especially at low temperatures. Owing to interference of TMI contribution with the temperature-independent contribution, the intensities can increase or decrease with temperature.

To describe the experimental spectra we present *ab initio* simulations of the temperature dependence and of diffraction spectra for Ge and ZnO. Different computer codes were applied: FDMNES [6] and PARATEC [7] and XKDQ [8].

Results

While the phenomenological approach gives a rather good qualitative description of thermal dependence of forbidden reflections in Ge and ZnO, the *ab initio* calculations are in satisfactory quantitative agreement with the experimental spectra. The cooperative character of atomic displacements are essential for calculations, for example, in ZnO, the E_{2l} mode prevails.

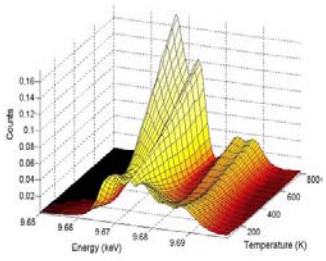

Fig.1. Temperature-dependent energy spectrum of the 115 reflection in ZnO

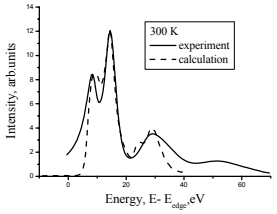

Fig.2. Numerical simulation of the 115 reflection in ZnO at room temperature

Discussion

Owing to their resonant character, the TMI and PDI reflections allow for separate studies of both impurity and host atoms of different types. The considered phenomena can provide a very sensitive tool to access point defects because only the atoms, "distorted" by defects, produce contributions to the PDI reflections. Also the numerical simulation of the forbidden reflections energy spectra allows determining of those phonon modes, which prevail at different temperatures.

Acknowledgments

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