

X-ray magnetic dichroism in the (Zn,Co)O diluted magnetic semiconductors from *ab initio* calculations

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Diluted magnetic semiconductors (DMSs), obtained by doping magnetic element as impurity into the host semiconductor, can be used for spintronic devices. As a wide-band-gap II-VI semiconductor, ZnO has recently been considered as a base material for realizing transparent DMSs.

Using the fully relativistic Dirac linear muffin-tin orbital (LMTO) band structure method, we have investigated theoretically from first principles the electronic structure of (Zn_{1-x}Co_x)ODMSs, the x-ray absorption spectra (XAS), x-ray magnetic circular dichroism (XMCD), and x-ray linear dichroism (XLD) spectra at the Co, Zn, and O *K* and Zn, Co *L*_{2,3} edges.

Since the local density approximation fails to describe correctly strongly localized *d* states in ZnO we treated these states within the LSDA+*U* approach. The effective on-site Coulomb repulsion *U* was considered as an adjustable parameter. The application of LSDA+*U* leads to a shift of the Zn *d* states downward in energy and makes the bandgap larger (2.12 eV within the LSDA+*U* versus 0.8 eV within the LSDA in pure ZnO wurtzite). The same effect can be achieved treating the Zn *d* states as core states. In our calculations we tried both approaches and did not find any significant difference in the XAS and XMCD spectra. The Co *d* states were treated within the LSDA as well as the LSDA+*U* approach. We tried various *U* values for the description of the XAS and XMCD spectra and found that the agreement between the theoretically calculated and experimental spectra does not significantly change in comparison with the LSDA method.

The theory reproduced the shape and energy positions of major fine structures of the Co XAS, XLD, and XMCD spectra at the *K* and *L*_{2,3} edges reasonably well. We found that oxygen deficiency is responsible for some fine structures of the Co *L*₃ XAS and XMCD spectra and has minor influence on the shape of the O and Zn *K* x-ray absorption spectra.

We found that a small peak at about 528 eV at the O *K* edge XAS evolves with Co doping, and its intensity increases with Co concentration suggesting a strong hybridization of O 2*p* states with Co 3*d* states. This feature originates in the Co doped ZnO alloys, and it is ascribed to dipole transitions from O 1*s* to O 2*p* states that are hybridized with the unoccupied 3*d* states of Co. Thus, the intensity of this peak is a fingerprint of the Co 3*d* density of states. A continuous increase of this peak with Co doping indicates more unoccupied states at the Co 3*d* levels.