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First-principles calculations of optical and magneto-optical properties of $Ga_{1-x}Mn_xAs$ and MnAs

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ABSTRACT

We present a first-principles density-functional investigation of the optical and magneto-optical properties of $Mn_xGa_{1-x}As$ (with $x = 0, 0.0625$, and 1) systems. Our calculated dielectric function, magneto-optical Kerr effect, and magnetic circular dichroism spectra agree reasonably well with existing experimental results. A comparison of the optical and magneto-optical spectra of MnAs in the naturally occurring hexagonal phase and in the cubic zinc-blende structure is made. The differences in the spectral properties of these two phases could aid detection of MnAs in the zinc-blende structure.

1. Introduction

Transition metal doped semiconductors are considered to be very interesting due to their possible applications in semiconductor spintronic devices [1]. The quest for a suitable, doped semiconducting material showing room temperature ferromagnetism is continuing. Mn-doped GaAs [2] is perhaps the most studied system among the whole class of diluted magnetic semiconductors (DMS) exhibiting ferromagnetism. Despite its low Curie temperature of around 170 K, it has been investigated in quite some detail regarding its electronic structure and magnetism. Besides III–V DMS, there are also numerous reports of room-temperature ferromagnetism in II–VI DMS, e.g., in Co doped ZnO [3], but there is no unanimous consensus regarding the ferromagnetism due to the occurrence of defects and secondary crystal phases in the samples. Mn-doped GaAs is a much better understood system since the effects of prominent defects, e.g., As antisites and interstitial Mn, have already been thoroughly studied (Ref. [2] and references therein).

Some of the ferromagnetic DMS have been reported to show interesting magneto-optical behavior [4–11]. From an application point of view, materials with large magneto-optical effects are very appealing. Such materials can, for example, be used as optical isolators, which are important components for optical fiber networks. Also, for a theoretical understanding of the electronic structure and the magnetic interactions, optical and magneto-optical experiments are important. Magneto-optical measurements on Mn-doped GaAs have shown promising results [8–11]. These experiments have been done not only for DMS, but also for their counterpart at full doping, viz., half-metallic ferromagnets in zinc-blende (ZB) structure [12,13]. These compounds are very interesting as they show 100% spin polarization due to their half-metallic character.

Ab initio calculations of magneto-optical effects of DMS are not abundant in the literature due to the challenging, high-precision, numerical computations. The challenges include, for instance, relativistic effects in band structure calculations and precise integration in the Brillouin zone over a large number of k-points. Recently, two works were reported [14,15] on studies of Mn-doped GaAs by density-functional full-potential linearized augmented plane wave calculations. In the paper by Stroppa et al. [14], a relatively large concentration of Mn was considered, in order to analyze the relationship between the band structure and the magneto-optical Kerr effect (MOKE). Picozzi et al. [15] studied the magneto-optics for both substitutional and interstitial Mn in GaAs. According to the latter calculations, the Kerr spectra was in better agreement with experiments in presence of interstitial Mn.

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It is worthwhile mentioning that the presence of interstitial Mn affects not only the hole concentration, but also decreases the effective ferromagnetic coupling as the interstitial Mn atoms couple antiferromagnetically to the substitutional Mn atoms. In order to model a realistic situation, clusters containing substitutional and interstitial Mn atoms [16] should be taken into account in the calculations of magneto-optical properties. But this would be an enormously computer intensive calculation and may be even beyond the scope of available computational facilities.

A systematic evolution of optical and magneto-optical properties of DMS systems due to doping is not available in the literature. The present work is motivated by this and starting from pure GaAs semiconductor, we investigate the change in properties due to Mn doping for a dilute concentration as well as for full doping. The paper is organized in the following way: first, we describe the computational methods. In the results section, we first show the optical properties of pure GaAs and, subsequently, present the optical and magneto-optical properties of GaAs with 6.25% Mn doping. After that we present electronic structures of the MnAs system in both the cubic zinc-blende and hexagonal structures and investigate the magneto-optical properties of MnAs in these two phases in detail.

2. Computational methods

Magnetic spectroscopy, with its variety of different experimental setups, is a sensitive tool to investigate the electronic structure of magnetic materials [17]. In this work we concentrate on two widely studied magneto-optical effects: the intrinsic polar magneto-optical Kerr effect (P-MOKE) and the magnetic circular dichroism (MCD). Both effects, being linear in the magnetization, are to some extent complementary as P-MOKE is a polarization analysis measured in reflection, whereas MCD is an intensity measurement in transition. The Kohn–Sham density-functional theory (DFT) provides a theoretical framework for an accurate calculation of ground state properties. *A priori*, excited state properties such as optical and magneto-optical spectra are not described by the DFT. The appropriate starting point for excitation spectra is the Dyson equation for quasi-particle excitations, which contains the non-local, non-hermitian self-energy operator $\Sigma(\boldsymbol{r},\boldsymbol{r}',\varepsilon)$. If the DFT is used to compute excitation spectra, the quasi-particle excitation energies are consequently approximated by the Kohn–Sham single-particle energies, which implies $\Sigma(\boldsymbol{r},\boldsymbol{r}',\varepsilon)\approx\delta(\boldsymbol{r}-\boldsymbol{r}')v_{\mathrm{xc}}(\boldsymbol{r})$, i.e., the self-energy is replaced by the Kohn–Sham exchange correlation potential v_{xc}. Numerical investigations showed that this approximation is surprisingly accurate, particularly for energies ε close to the Fermi energy [18]. Moreover, the Fermi level itself is correctly given by the DFT [19]. Consequently, the Kohn–Sham spectrum provides a good approximation for the quasi-particle spectrum of weakly correlated materials.

The Kohn–Sham optical spectrum can be evaluated by means of the Kubo linear response theory [20], which provides an expression for the complex optical conductivity tensor $\sigma_{ij}(\omega)$ in terms of single-particle band energies and matrix elements of momentum operators. Relaxation processes can be taken into account by including a (phenomenological) relaxation time $\tau=\delta^{-1}$, which was treated as state and energy independent in our approach [17,21].

A crucial point of a MOKE and MCD calculation is an accurate evaluation of the transition matrix elements and the k-integration of the Brillouin zone (BZ). The latter can be achieved by the so-called analytical tetrahedron technique. An implementation of this *ab initio* scheme, which we applied to compute the necessary components of σ, has been described in Ref. [21]. Once the complex optical conductivity tensor elements $\sigma_{ij}(\omega)$ are calculated, the dielectric function $\varepsilon(\omega)$ can, for cubic materials, be obtained from

$$\mathrm{Re}[\varepsilon]=1-\frac{4\pi}{\omega}\mathrm{Im}[\sigma_{xx}]\quad\text{and}\quad\mathrm{Im}[\varepsilon]=\frac{4\pi}{\omega}\mathrm{Re}[\sigma_{xx}]. \tag{1}$$

The P-MOKE spectrum is given by the following expression [17]:

$$\theta_K+i\eta_K\approx\frac{-\sigma_{xy}}{\sigma_{xx}\left(1+i\frac{4\pi}{\omega}\sigma_{xx}\right)^{1/2}}, \tag{2}$$

where the angle θ_K is the Kerr rotation, η_K is the Kerr ellipticity, and ω the circular frequency of the incident light.

Furthermore, the MCD spectrum, in transmission, can be calculated by

$$A_{MCD}\approx\frac{d\omega}{2c}\mathrm{Im}[n_+-n_-]\approx\frac{2\pi d}{c}\mathrm{Im}\left[\frac{-\sigma_{xy}}{\left(1+i\frac{4\pi}{\omega}\sigma_{xx}\right)^{1/2}}\right], \tag{3}$$

where d is the thickness of the sample, c the speed of light, and $n_\pm=(\varepsilon_{xx}\pm\varepsilon_{xy})^{1/2}$ is the refractive index for right (left, respectively) circularly polarized light.

Two sets of electronic structure calculations were performed using density-functional theory. In one case, the electronic structure of DMS $Ga_{15}Mn_1As_{16}$ (6.25% Mn) was calculated in zinc-blende structure by means of a full-potential linear muffin-tin orbital code (FP-LMTO) [22] adopting the local spin-density approximation (LSDA). Self-consistency was obtained for 3456 k-points in the irreducible BZ for the experimental lattice constant 5.65 A of pure GaAs.

In another set of calculations, we used relativistic DFT within the LSDA to study the properties of GaAs and MnAs. In these calculations, the augmented spherical wave (ASW) basis functions were used in the spherical potential approximation [23]. Two empty atomic spheres have been introduced in the ZB structure, in order to obtain a good description of the potential. Self-consistency was obtained with 6552 k-points in the irreducible BZ.

3. Results and discussion

3.1. GaAs

We are interested to study the gradual change in optical and magneto-optical properties of pure GaAs to GaAs doped with a dilute concentration of Mn and finally to MnAs with 100% Mn doping. Let us first start with the optical properties of GaAs.

It is well known [24] that the band-gap of semiconductors, computed in the LDA, is systematically underestimated compared to the experimental values. In fact, this problem persists even in the (unknown) exact Kohn–Sham DFT [25,26]. It has been shown for semiconducting Si that an evaluation of the quasi-particle spectrum on the basis of an approximated Dyson equation leads to a better band-gap value [27]. However, it has also been found that the shape of the optical spectrum is better given by Kohn–Sham DFT calculations, except for the value of the band-gap [27]. A practicable and commonly used way of treating the underestimated band-gap is to apply a rigid energy shift of to the computed Kohn–Sham spectra (the so-called scissors operation). In case of GaAs, the experimental band-gap is 1.5 eV, whereas LDA calculations give about half this value. This underestimation will be treated by a rigid shift of the computed spectra.

In Fig. 1 we show the computed and experimental optical spectra of GaAs. For the experimental GaAs lattice constant of