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# Full potential results on the magneto-optical properties of the Heusler compounds $\text{Co}_2\text{FeX}$ ( $X = \text{Al, Ga, Si}$ and $\text{Ge}$ )

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## Abstract

We have calculated the magneto-optical (MO) properties of  $\text{Co}_2\text{FeX}$  ( $X = \text{Al, Ga, Si}$  and  $\text{Ge}$ ) Heusler compounds using the full potential linearized augmented plane wave (FPLAPW) method as implemented in the WIEN2K code using the local spin density approximation (LSDA) and also by using the generalized gradient approximation (GGA) for the electronic exchange and correlation. In all the compounds, Kerr rotation  $\theta_K$  has a strong minimum near 2 eV, the value of  $|\theta_K|$  corresponding to this minimum being almost as large as in pure Co–Fe compounds. The calculated MO spectra help to identify the features of the experimental spectra. A comparison of the results shows that the Kerr spectrum is quite similar from both LSDA and GGA but the latter gives better agreement with experiment. Moreover, we find that inclusion of correlation effects using GGA +  $U$  removes the discrepancy in magnetic moment of  $\text{Co}_2\text{FeX}$  ( $X = \text{Si, Ge}$ ) though it has an insignificant effect on the Kerr spectra.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

Magnetic materials existing in the  $L2_1$  and  $C1_b$  crystallographic phases have been of great interest to both theorists and experimentalists ever since these were first studied by Heusler [1]. His interest focused on the unusual result that some of these materials in these crystallographic phases were strongly ferromagnetic even though they were composed of elements which were considered to be nonmagnetic at that time. Heusler compounds are among the most promising candidates for spintronic applications due to their predicted 100% spin polarization along with a high ferromagnetic transition temperature. In addition, a very important aspect of Heusler compounds is their magneto-optical behavior. The discovery of a giant Kerr rotation for the half-Heusler compound PtMnSb ( $>2^\circ$  at room temperature and  $5^\circ$  at 80 K), which was totally unexpected for a 3d-based material [2], showed the technological relevance of such compounds in the context of magneto-optical reading and recording. Since then, there has

been a great deal of interest in this class of compounds [3] for magneto-optics.

The half-Heusler compounds like NiMnSb and PtMnSb have attracted as much attention as the full Heusler compounds which have been studied extensively due to the existence of diverse magnetic phenomena [4, 5] such as the transition from a ferromagnetic phase to an antiferromagnetic phase on changing the concentration of the carriers [6]. The full Heusler compounds have stoichiometric composition  $X_2YZ$ , where  $X$  is generally a high valence transition or noble metal atom,  $Y$  is a low valence transition metal atom and  $Z$  is often an element with sp-type valence electrons from columns III–VI of the periodic table. These compounds crystallize in the  $L2_1$  structure which consists of four interpenetrating fcc sublattices; the first three displaced by  $(1/4, 1/4, 1/4)$ ,  $(3/4, 3/4, 3/4)$  and  $(1/2, 1/2, 1/2)$  with respect to the  $Z$  species and occupied by  $X$ ,  $X$  and  $Y$ , respectively. Each  $X$  atom has four  $Y$  and four  $Z$  atoms as nearest neighbors and each  $Y$  or  $Z$  atom is surrounded by eight  $X$  atoms.

Co<sub>2</sub>-based Heusler compounds were synthesized and investigated in the 1970s [7]. Buschow and van Engen [8–10] probed many combinations of X, Y and Z for the formation of the L2<sub>1</sub> phase. Their primary motivation was to search for magnetic materials with promising magneto-optical properties for applications in magneto-optical recording devices. They studied the magneto-optical properties of X<sub>2</sub>YAl and X<sub>2</sub>YGa compounds experimentally [8, 10] and showed that the polar Kerr rotation angle  $\theta_K$  for the members of these two groups (Co<sub>2</sub>FeAl and Co<sub>2</sub>FeGa) has a strong minimum near 1.5 eV, the value of the  $|\theta_K|$  corresponding to this minimum being almost as large as in the pure Co–Fe compounds. Kubler *et al* [6] recognized that the minority spin densities at the Fermi energy  $\varepsilon_F$  nearly vanish for Co<sub>2</sub>MnAl and Co<sub>2</sub>MnSn. The authors concluded that this should lead to peculiar transport properties in these Heusler compounds because only the majority electrons contribute to the states at  $\varepsilon_F$ . There also exist studies on the effect of structural defects and disorders on the properties of these compounds. Antonov *et al* [11] have studied the effect of point defects and anti-sites on the magnetic and magneto-optical properties of Co<sub>2</sub>FeAl. Picozzi *et al* [12] have studied the role of structural defects on the half-metallicity of the Co<sub>2</sub>MnX (X = Ge, Si) Heusler alloys whereas Miura *et al* [13] have explored the influence of atomic disorder on the half-metallicity of the Heusler alloys Co<sub>2</sub>(Cr<sub>1-x</sub>Fe<sub>x</sub>)Al. It is quite important to assess the effect of such defects or atomic disorders on the physical properties of half-metallic Heusler alloys, since the spin polarization of these alloys is influenced remarkably by a particular defect/disorder. However, in this paper, we confine ourselves to the pristine Heusler compounds Co<sub>2</sub>FeX (X = Al, Ga, Si and Ge) and have studied the effect of LDA, GGA and GGA + *U*, the various approximations for the electronic exchange and correlations, on the magneto-optical properties of these compounds. These Heusler compounds exhibit the highest magnetic moment per unit cell ( $5.97 \pm 0.05 \mu_B$  for Co<sub>2</sub>FeSi [14]) and the highest Curie temperature ( $T_C = 985$  K for Co<sub>2</sub>MnSi [15]) reported up to now.

Now many Heusler compounds are known to be half-metallic ferromagnets (HMF) and are seen to be potential candidates for spintronics [16] where electronics makes use of the electron's spin instead of its charge. The concept of half-metallic ferromagnetism was first presented by de Groot [17], predicting it to appear in half-Heusler compounds. The model suggests that the density of states exhibits a gap for minority electrons. Thus these materials are supposed to be 100% spin polarized at  $\varepsilon_F$ . For both scientific and technological reasons, it is useful to define the electron spin polarization of a material at the Fermi energy, although it is difficult to measure [18]. The spin polarization at  $\varepsilon_F$  is given by

$$P = \frac{\rho_{\uparrow}(\varepsilon_F) - \rho_{\downarrow}(\varepsilon_F)}{\rho_{\uparrow}(\varepsilon_F) + \rho_{\downarrow}(\varepsilon_F)} \quad (1)$$

where  $\rho_{\uparrow}(\varepsilon_F)$  and  $\rho_{\downarrow}(\varepsilon_F)$  are the spin-dependent density of states at  $\varepsilon_F$  for the majority and minority spins, respectively. *P* vanishes for paramagnetic and antiferromagnetic materials even below the magnetic transition temperature. However, it has a finite value in ferromagnetic or ferrimagnetic materials below the Curie temperature. The electrons at  $\varepsilon_F$  are fully

spin polarized ( $P = 1$ ) when either  $\rho_{\uparrow}(\varepsilon_F)$  or  $\rho_{\downarrow}(\varepsilon_F)$  equals zero. This exceptional property would make HMFs the ideal candidates for spin injection devices to be used in spin electronics.

In the present paper, we first investigate the magnetic properties of Co<sub>2</sub>FeX (X = Al, Ga, Si and Ge) by density functional calculations to confirm half-metallic ferromagnetism in these compounds. Moreover, despite the existence of ample data on the magnetic and MO properties of these compounds, not much attention has been paid to the same theoretically. A balance between the experiments and theory is desirable for developing a good understanding of these compounds, particularly of their MO properties. To the best of our knowledge, the only previous calculation is from van Ek *et al* [19] who calculated the Kerr rotation of Co<sub>2</sub>FeX (X = Al and Ga) using the linear muffin-tin orbital (LMTO) method within the atomic sphere approximation (ASA). Their calculations of polar Kerr rotation and Kerr ellipticity showed good agreement with the experimental data in the energy range 2–4 eV. However, for lower energies (<2 eV), the agreement is rather poor. To bridge the gap between experiments and theory and with the aim to improve the agreement with experiments, we report calculations on magneto-optical properties of Co<sub>2</sub>FeX (X = Al, Ga, Si and Ge). Further, the aim of this paper is to investigate the effect of substitution of different p-block elements in Co<sub>2</sub>FeX on the electronic, magnetic and magneto-optical properties by using X = Al (3s<sup>2</sup>, 3p<sup>1</sup>), Ga (4s<sup>2</sup>, 4p<sup>1</sup>), Si (3s<sup>2</sup>, 3p<sup>2</sup>) and Ge (4s<sup>2</sup>, 4p<sup>2</sup>) individually. We performed full potential calculations using two different types of exchange correlation potentials, namely local spin density approximation (LSDA) and generalized gradient approximation (GGA), and also by using GGA + *U* for Co<sub>2</sub>FeSi and Co<sub>2</sub>FeGe for reasons cited in the text.

This paper is organized as follows. In section 2, we focus on the description of the theoretical approach to the magneto-optical properties. The computational and structural details of these Heusler compounds are presented in section 3. The comparison of theoretical results with available experimental data for Kerr rotation and ellipticity is presented in section 4. Finally in section 5, we draw the conclusions.

## 2. Theory

The computational framework for the evaluation of the MO Kerr effect has been described previously in several papers [20]. We recall that in the polar geometry, where the *z* axis is chosen to be perpendicular to the surface of the sample and parallel to the magnetization direction, the Kerr effect is given by [21]

$$\theta_K(\omega) + i\varepsilon_K(\omega) = \frac{-\sigma_{xy}(\omega)}{\sigma_{xx}(\omega)\sqrt{1 + \frac{4\pi i}{\omega}\sigma_{xx}(\omega)}} \quad (2)$$

with  $\theta_K$  as the Kerr rotation and  $\varepsilon_K$  as the so-called Kerr ellipticity. Here  $\omega$  is the frequency of the electromagnetic waves. This geometry is the one most studied experimentally.

The optical conductivity tensor  $\sigma_{\alpha\beta}(\alpha, \beta \equiv x, y, z)$  appearing in equation (2) is related to the dielectric tensor  $\varepsilon_{\alpha\beta}$  through

$$\varepsilon_{\alpha\beta}(\omega) = \delta_{\alpha\beta} + \frac{4\pi i}{\omega} \sigma_{\alpha\beta}(\omega) \quad (3)$$

where  $\delta_{\alpha\beta}$  is the delta function.

Equation (2) shows that the optical conductivity tensor, or equivalently the dielectric tensor, is the important spectral quantity needed for the evaluation of the Kerr effect [22]. The optical conductivity can be computed from the energy band structure by means of the linear-response expression [23] based on Kubo's approach [24]:

$$\sigma_{\alpha\beta}(\omega) = \frac{-ie^2}{m^2 \hbar V_{uc}} \sum_{\mathbf{K}} \sum_{nn'} \frac{f(\varepsilon_{nk}) - f(\varepsilon_{n'k})}{\omega_{nn'}(k)} \times \frac{\prod_{nn'}^{\alpha}(k) \prod_{nn'}^{\beta}(k)}{\omega - \omega_{nn'}(k) + i/\tau} \quad (4)$$

with  $f(\varepsilon_{nk})$  as the Fermi function and  $\hbar\omega_{nn'}(k) = \varepsilon_{nk} - \varepsilon_{n'k}$  as the difference in the Kohn–Sham energies  $\varepsilon_{nk}$  and  $\varepsilon_{n'k}$  for the two bands  $n$  and  $n'$  at the point  $\mathbf{k}$ .  $\gamma \equiv \tau^{-1}$  is the lifetime parameter which is included to simulate the finite lifetime of the excited Bloch electron states. The physical effect of  $\tau$  is that each infinitely ‘sharp’ optical transition is convoluted with a Lorentzian width  $\gamma$ . This lifetime parameter was originally introduced on phenomenological grounds, but its approximateness for describing the conductivity is beyond doubt [20, 23].  $\prod_{nn'}^{\alpha}(k)$  are the dipolar optical transition matrix elements which, in a fully relativistic description, are given by

$$\prod_{nn'}^{\alpha}(k) = m \langle \psi_{nk} | c\alpha | \psi_{n'k} \rangle \quad (5)$$

with  $\psi_{nk}$  as the four-component Bloch electron wavefunction.

Expression (4) for the conductivity contains a double sum over all energy bands, which naturally separates into the so-called interband contribution, i.e.  $n \neq n'$ , and the intraband contribution i.e.  $n = n'$ . The intraband contribution to the diagonal components of  $\sigma$  may be rewritten for zero temperature as

$$\sigma_{\alpha\alpha}(\omega) = \frac{(\omega_{p,\alpha})^2}{4\pi} \frac{i}{\omega + i\gamma_D} \quad (6)$$

with  $\omega_{p,\alpha}$  as the components of plasma frequency, which are given by

$$(\omega_{p,\alpha})^2 \equiv \frac{4\pi e^2}{m^2 V_{uc}} \sum_{nk} \delta(\varepsilon_{nk} - \varepsilon_F) \left| \prod_{nn'}^{\alpha} \right|^2. \quad (7)$$

Here  $\varepsilon_F$  is the Fermi energy and  $V_{uc}$  is the unit cell volume. For cubic symmetry, we further have  $\omega_p^2 = \omega_{p,x}^2 = \omega_{p,y}^2 = \omega_{p,z}^2$ , as in the case for the present study.

### 3. Method of calculation

The present Heusler compounds have a cubic  $L2_1$  structure with the space group  $Fm\bar{3}m$  such that there are 16 atoms or, in other words, 4 fu in the unit cell. This is a close-packed complex fcc structure with Co atoms at (1/4 1/4 1/4)

and (3/4 3/4 3/4), Fe at (1/2 1/2 1/2) and X at (000). First-principles band structure calculations for these compounds have been performed using the WIEN2K [25] implementation of the full potential linearized augmented plane wave (FPLAPW) method [26] based on the density functional theory (DFT) [27, 28]. In order to calculate the MO properties, we performed calculations including a fully self-consistent treatment with the spin–orbit interaction. The exchange correlation potential is treated with (i) LSDA [29] using the von Barth and Hedin parameterization [30] and (ii) GGA according to the Perdew–Burke–Ernzerhof parameterization [31]. It has been observed [30–32] that LSDA has a tendency to reduce the charge asphericity, while GGA, including nonlocal contributions, can enhance such effects, also resulting in a more accurate description of the energy bands. It is therefore, particularly important to use GGA for these compounds. These calculations have been carried out at lattice constants of 5.73 Å, 5.737 Å, 5.647 Å and 5.738 Å for  $\text{Co}_2\text{FeX}$  with  $X = \text{Al, Ga, Si}$  and  $\text{Ge}$ , respectively, as taken from experimental data [15].

In the full potential method, the potential and the charge in the crystal are treated with no shape approximation. The radii for muffin-tin spheres were chosen as  $R_{\text{MT}}^{\text{Co}} = R_{\text{MT}}^{\text{Fe}} = 2.1$  au,  $R_{\text{MT}}^{\text{Al}} = R_{\text{MT}}^{\text{Si}} = 2.3$  au and  $R_{\text{MT}}^{\text{Ga}} = R_{\text{MT}}^{\text{Ge}} = 2.4$  au. The cutoff parameters criterion is  $R_{\text{MT}} K_{\text{Max}} = 9$  for the plane waves and  $G_{\text{Max}} = 14$  au<sup>-1</sup> in the Fourier expansion of the potential in the interstitial region. The  $\mathbf{k}$ -space integration has been carried out using the modified tetrahedron method [33] with 405  $\mathbf{k}$ -points in the irreducible Brillouin zone (IBZ) to obtain self-consistency. The calculations for optical and magneto-optical properties have been carried out using a denser mesh of 726  $\mathbf{k}$ -points in the IBZ. In order to consider the effect of finite lifetimes as well as of the experimental resolution, a Lorentzian broadening of 0.45 eV is applied for both intraband and interband contributions.

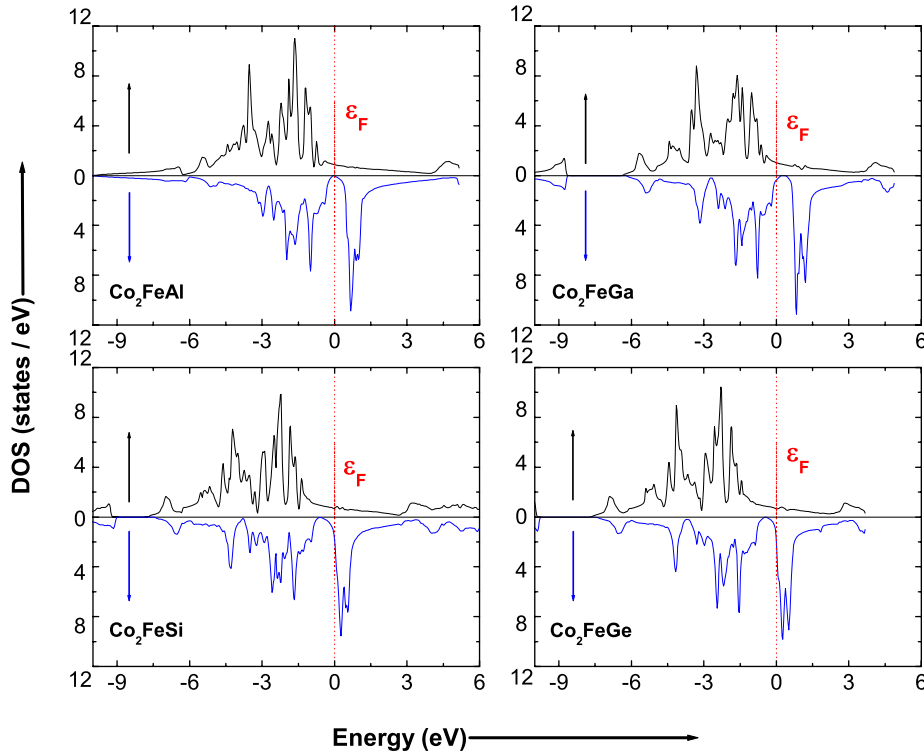
### 4. Results and discussions

In the present work, the classification scheme for half-metals as proposed by Coey *et al* is used [16]. They have classified half-metals into four types, depending upon spin polarization and the localized or itinerant nature of the electrons. In types I and II half-metals, the spin polarization  $P$  is always 100%, whereas lower values appear in types III and IV. It is well known that half-metallic Heusler compounds based on Co follow the Slater–Pauling rule [34, 35] for predicting their total spin magnetic moment. Thus the saturation magnetization scales with the number of valence electrons [36–38]. The following Slater–Pauling rule with

$$m_{\text{HMF}} = n_V - 6 \quad (8)$$

is strictly fulfilled for the mean magnetic moment per atom ( $m_{\text{HMF}}$ ). Here  $n_V$  is the mean number of valence electrons per atom found by averaging over all the atoms. Thus, in the case of 4 atoms per unit cell (full Heusler compounds), one has to subtract 24 from the accumulated number of valence electrons in the unit cell  $N_V$  to find  $m_H$ , the magnetic moment per unit cell:

$$m_H = N_V - 24. \quad (9)$$



**Figure 1.** Calculated spin resolved density of states (DOS) for  $\text{Co}_2\text{FeX}$  ( $X = \text{Al, Ga, Si and Ge}$ ). The DOS values above (below) 0 represent the majority (minority) states.  $\epsilon_F$  represents the Fermi level.

We first investigate the half-metallic ferromagnetism in the present Co-based Heusler compounds. In the present Heusler compounds, the energy bandgap stays, with few exceptions, in the minority spin state but  $\epsilon_F$  does not intersect the gap in the minority spin channel. Our calculations show nearly half-metallic behavior for  $\text{Co}_2\text{FeAl}$  with GGA formalism as the density of states (DOS) for the minority spin becomes approximately zero at  $\epsilon_F$  (table 1) and the bandgap is present in the vicinity of the Fermi level as shown in the DOS plot (figure 1). The spin polarization in  $\text{Co}_2\text{FeAl}$  becomes 93% (maximum among the present compounds using GGA) as governed by equation (1). Thus  $\text{Co}_2\text{FeAl}$  is very nearly a type I half-metallic compound. According to the Slater–Pauling rule,  $\text{Co}_2\text{FeAl}$  ( $N_V = 29$ ) is expected to show the magnetic moment per unit cell as  $m_H = 5 \mu_B$  in accordance with equation (9). This predicted value of  $m_H$  is confirmed from our DFT calculations as we have obtained  $m_H = 5 \mu_B$  for  $\text{Co}_2\text{FeAl}$ . For other compounds, the degree of polarization is small as we observe finite DOS at  $\epsilon_F$  for both the spin states and the gap is present away from  $\epsilon_F$  so that the compounds do not obey Slater–Pauling behavior for half-metallic compounds. Hence these compounds can be described as type III half-metals. This is consistent with the LDA +  $U$  calculations performed by Kandpal *et al* [39] showing the half-metallic ferromagnetism of type III for all these  $\text{Co}_2\text{FeX}$  compounds. In that calculation, the inclusion of electron correlation in the form of LDA +  $U$  on top of the LSDA and GGA does not induce or destroy the half-metallic gap in these compounds. Its effect is only to enlarge the gap if it is already present and to shift the minority bands with respect to the majority

bands or the Fermi energy. Moreover in DOS plots, the valence band has contributions from both spins but in the conduction band, only the minority spin state dominates. All the DOS plots are almost similar, predicting the existence of similar types of magnetic and magneto-optical properties for all these compounds. On investigation, we find that the main contributions to the structures in DOS come from Co 3d and Fe 3d states, with a smaller contribution from X p states.

For the present  $\text{Co}_2\text{FeX}$  Heusler compounds, the site resolved moments, total spin moments and other ground state properties from this and other theoretical works [39, 40] and from experiments [14, 15, 41–43] are summarized in table 1. The plasma frequency, as obtained from our FPLAPW calculations and used to evaluate the Kerr rotation, is also mentioned in the last column of the table for all the compounds. In all these Heusler compounds, the Fe atom carries the largest moment of about 2.8–2.9  $\mu_B$  while Co has its value in the range 1.2–1.4  $\mu_B$  in the GGA formalism. The X atoms carry a negligible magnetic moment which does not contribute significantly to the overall moment. In  $\text{Co}_2\text{FeAl}$  and  $\text{Co}_2\text{FeGa}$ , the magnetic moment at X is aligned antiparallel to Co and Fe moments whereas for  $\text{Co}_2\text{FeSi}$  and  $\text{Co}_2\text{FeGe}$ , it is almost zero. The total magnetic moment is of the order of 5  $\mu_B$  in  $\text{Co}_2\text{FeX}$  ( $X = \text{Al and Ga}$ ) compounds whereas for  $\text{Co}_2\text{FeSi}$  and  $\text{Co}_2\text{FeGe}$ , it is found to be 5.58  $\mu_B$  and 5.72  $\mu_B$ , respectively. With an increase in the number of valence electrons and of atomic number of p-block elements (X), the magnetic moment also increases. These GGA values for the magnetic moments are in good agreement with the experimental values except for  $\text{Co}_2\text{FeSi}$  and  $\text{Co}_2\text{FeGe}$ . For  $\text{Co}_2\text{FeSi}$  ( $N_V = 30$ ), the calculated



**Table 1.** Calculated magnetic moments (in Bohr magnetons  $\mu_B$ ) within the muffin-tin spheres at their respective equilibrium, compared with available experiments, calculated DOS for the majority and minority spin (i.e.  $\rho_{\uparrow}(\varepsilon_F)$  and  $\rho_{\downarrow}(\varepsilon_F)$ ), spin polarization ( $P$ ) at  $\varepsilon_F$  along with plasma energy for  $\text{Co}_2\text{FeX}$  ( $X = \text{Al, Ga, Si}$  and  $\text{Ge}$ ) compounds.

Compound		$\mu_{\text{Co}}$	$\mu_{\text{Fe}}$	$\mu_X$	$\mu_{\text{tot}}$	$\rho_{\uparrow}(\varepsilon_F)$	$\rho_{\downarrow}(\varepsilon_F)$	$P$ (%)	$\omega_P$
$\text{Co}_2\text{FeAl}$	This work								
	GGA	1.23	2.79	-0.07	5.0	0.86	0.03	93.2	5.17
	Others <sup>a</sup>	—	—	—	5.193	—	—	—	—
	<sup>b</sup>	1.22	2.97	—	5.0	—	—	—	—
	Expt <sup>c</sup>	—	—	—	4.96	—	—	—	—
$\text{Co}_2\text{FeGa}$	This work								
	GGA	1.18	2.91	-0.08	5.03	1.02	0.33	51.1	4.84
	Others <sup>a</sup>	—	—	—	5.27	—	—	—	—
	Expt <sup>c</sup>	—	—	—	5.13	—	—	—	—
	<sup>d</sup>	—	—	—	5.15	—	—	—	—
$\text{Co}_2\text{FeSi}$	This work								
	GGA	1.39	2.84	0.0	5.58	0.69	1.58	39.2	5.69
	GGA + $U$	1.52	3.12	-0.04	6.0	0.582	0.006	98.0	5.72
	Others <sup>a</sup>	—	—	—	5.8	—	—	—	—
	<sup>b</sup>	1.50	3.14	—	6.0	—	—	—	—
	Expt <sup>c</sup>	—	—	—	5.18	—	—	—	—
	<sup>e</sup>	—	—	—	5.7	—	—	—	—
<sup>f</sup>	—	—	—	$5.97 \pm 0.05$	—	—	—	—	
$\text{Co}_2\text{FeGe}$	This work								
	GGA	1.43	2.88	0.01	5.72	0.66	1.57	40.8	5.20
	GGA + $U$	1.53	3.12	-0.03	6.02	0.615	0.002	99.3	5.32
	Others <sup>b</sup>	1.42	2.92	—	5.70	—	—	—	—
	Expt <sup>a</sup>	—	—	—	5.54	—	—	—	—
	<sup>g</sup>	—	—	—	5.90	—	—	—	—

<sup>a</sup> Reference [40]; <sup>b</sup> Reference [39]; <sup>c</sup> Reference [15]; <sup>d</sup> Reference [41]; <sup>e</sup> Reference [42];

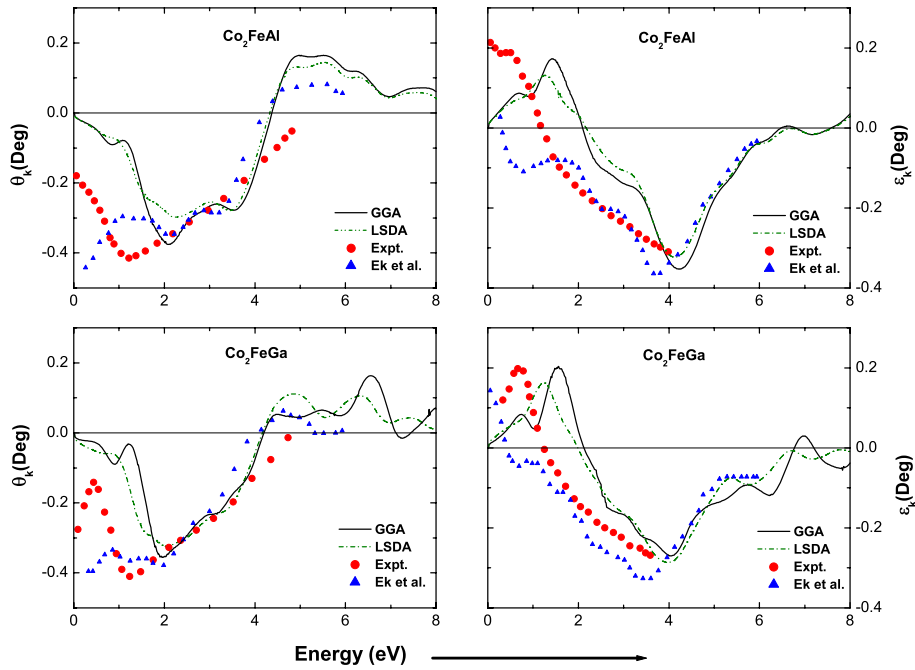
<sup>f</sup> Reference [14]; and <sup>g</sup> Reference [43].

magnetic moment is lower than both the recent experimental value ( $5.97 \pm 0.05 \mu_B$ ) [14] and the predicted value of  $6 \mu_B$  as governed by the Slater–Pauling rule (equation (8)). We find that the discrepancy in magnetic moment with respect to the experimental value gets removed by performing band structure calculations using the Coulomb-corrected GGA +  $U$  approach, as also observed by Kandpal *et al* [39] on using the LDA +  $U$  approach. We have used  $U_{\text{eff}}$  values of 1.80 and 1.92 eV for Fe and Co, respectively, in GGA +  $U$  calculations, as also used by Kandpal *et al* [39]. Our GGA +  $U$  results give the total magnetic moment for  $\text{Co}_2\text{FeSi}$  as  $6.0 \mu_B$  (same as the experimental value) and for  $\text{Co}_2\text{FeGe}$  as  $6.02 \mu_B$  (experimental value  $5.9 \mu_B$ ). Also the spin polarization  $P$  for the two compounds is enhanced on use of GGA +  $U$ , approaching almost unity. Due to high magnetic moment values, these compounds are expected to have a large Kerr rotation, which we have explored next.

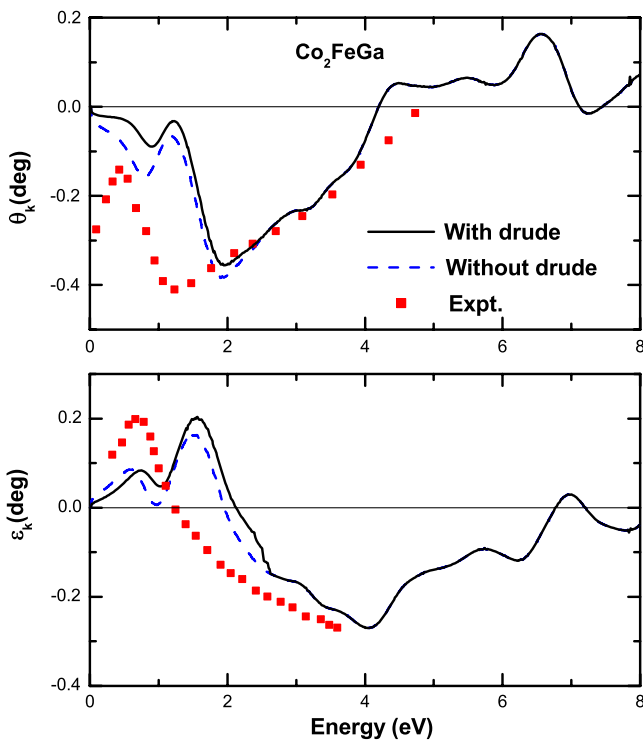
Figure 2 shows our calculated spectra of Kerr angle (left panel) and Kerr ellipticity (right panel) for  $\text{Co}_2\text{FeAl}$  and  $\text{Co}_2\text{FeGa}$  with the results of LMTO calculations by van Ek *et al* [19]. The experimental data by Buschow *et al* [10] has also been presented for the sake of comparison. The maximum Kerr rotation of  $-0.38^\circ$  and  $-0.35^\circ$  at  $\sim 2$  eV is obtained for  $\text{Co}_2\text{FeAl}$  and  $\text{Co}_2\text{FeGa}$ , respectively. There exists apparently a rather good agreement between the experimental Kerr spectra (for both Kerr rotation and Kerr ellipticity) and the present calculations as compared to the results from van Ek *et al*. The qualitative trends in the calculated results within the LSDA and GGA formalism are very similar but the Kerr minima near 2 eV

are more pronounced with GGA. Moreover, GGA defines the experimental peaks very sharply. This indicates that, to get a better comparison of results for these compounds, GGA should be preferred over LSDA. We obtain a maximum at  $E \sim 1.5$  eV for  $\text{Co}_2\text{FeGa}$  in the Kerr ellipticity spectrum, as is also present in the experimental data, which was not there in the previous calculation. The ellipticity spectra of both the compounds ( $X = \text{Al, Ga}$ ) have fairly similar shapes. A comparison with experimental results [10] shows that the theoretical Kerr rotation and ellipticity spectra have more structures at high photon energies  $E > 5$  eV, but unfortunately the experimental data is not available for  $E > 5$  eV to compare with. Moreover the calculated structures are shifted upwards in energies by about 0.8 eV as compared to the experimental ones.

We have tried to analyze the effect of the Drude term by plotting the Kerr spectra of  $\text{Co}_2\text{FeGa}$  with the Drude (intraband + interband) and without the Drude (interband only) term which is shown in figure 3. It is clear that the two spectra have almost similar shapes. We know that the MO effects arise generally from both interband and intraband transitions. In the low energy regime, the optical transitions can be ascribed to both interband and intraband transitions. However, in the higher energy regime, the contribution from intraband transitions is normally neglected due to its small amplitude or nonexistence. Due to the high intraband contribution at lower energy, the value of diagonal conductivity  $\sigma_{xx}(\omega)$  increases abruptly as compared to its value when the Drude term is not taken into account. Thus, according to equation (2), the Kerr rotation decreases at low energies if the Drude contribution



**Figure 2.** Polar Kerr rotation  $\theta_K$  and ellipticity  $\epsilon_K$  of  $\text{Co}_2\text{FeX}$  ( $X = \text{Al}$  and  $\text{Ga}$ ) in left and right panels, respectively, along with experimental data [10] and other calculation [19].

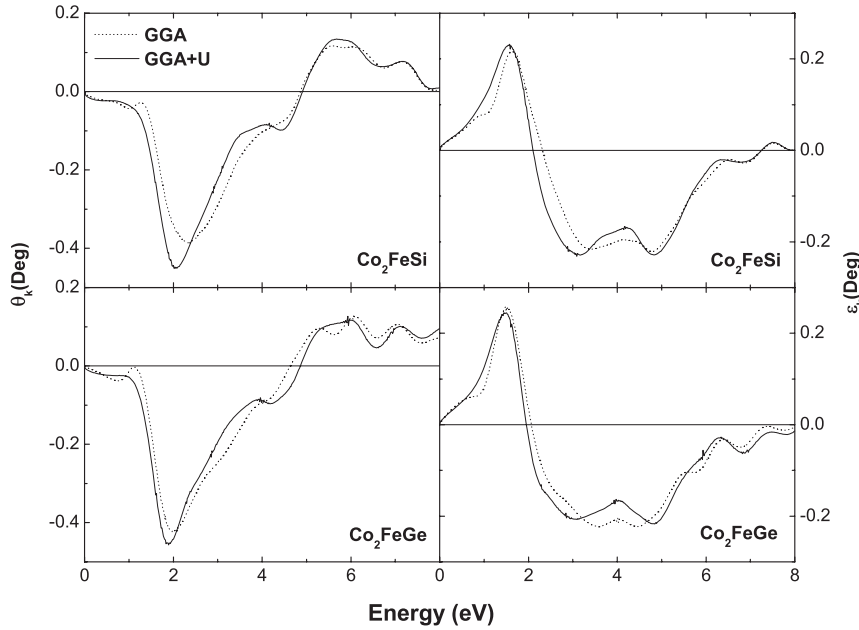


**Figure 3.** Calculated polar Kerr rotation  $\theta_K$  and ellipticity  $\epsilon_K$  of  $\text{Co}_2\text{FeGa}$  using GGA with and without the Drude term along with the experimental data.

is included. This impact is well conveyed by our results in figure 3. In the present case, the effect of the Drude term is dominant for energies up to  $\sim 2$  eV. However, the inclusion of the Drude term does not bring any drastic change in the Kerr spectra for the low energy region.

Having confirmed better trends for the Kerr spectra of  $\text{Co}_2\text{FeX}$  ( $X = \text{Al}$  and  $\text{Ga}$ ) with GGA as compared to the LSDA formulism, we have presented our results for  $\text{Co}_2\text{FeX}$  ( $X = \text{Si}$  and  $\text{Ge}$ ) with GGA in figure 4. There are no previous calculations or experimental results to compare with for these two compounds. We find that the Kerr spectra for both the compounds are very similar, which shows that the effect of the change of the p-block element ( $X$ ) on Kerr spectra is very small. A maximum Kerr rotation of the order of  $-0.39^\circ$  and  $-0.42^\circ$  at  $\sim 2$  eV is obtained for  $\text{Co}_2\text{FeX}$  ( $X = \text{Si}$  and  $\text{Ge}$ ), respectively. This value is slightly larger than that obtained for  $\text{Co}_2\text{FeAl}$  and  $\text{Co}_2\text{FeGa}$  ( $-0.38^\circ$  and  $-0.35^\circ$ , respectively). This is the largest Kerr angle for  $L_{21}$  full Heusler compounds ( $X_2YZ$ ) with  $Y = \text{Fe}$ ,  $\text{Co}$  and  $\text{Ni}$ , which correlates with the large magnetic moment found in these compounds. The Kerr minimum for  $\text{Co}_2\text{FeSi}$  is blueshifted by 0.3 eV as compared to that for  $\text{Co}_2\text{FeGe}$ . Kerr ellipticity is maximum for both these compounds at  $\sim 1.5$  eV as is also observed in the case of  $\text{Co}_2\text{FeAl}$  and  $\text{Co}_2\text{FeGa}$ . As GGA +  $U$  gives a better representation of the band structure and magnetic properties of  $\text{Co}_2\text{FeSi}$  and  $\text{Co}_2\text{FeGe}$ , we have also compared the Kerr spectra obtained from the GGA and GGA +  $U$  approaches for these two compounds in figure 4. The Kerr spectra from both approaches are qualitatively the same. A slight difference (overestimation by about  $0.05^\circ$ ) is observed in the magnitude of Kerr rotation in both  $\text{Co}_2\text{FeSi}$  and  $\text{Co}_2\text{FeGe}$  at  $\sim 2$  eV.

Since the Kerr spectra depend on the conductivity spectra in an entangled way, so it becomes difficult to assign the features in the Kerr spectra to particular band transitions. Therefore, to identify the electronic interband transitions responsible for the MO Kerr effect, the analysis of the spectral dependence of the optical conductivity tensor should be performed. The frequency-dependent absorptive parts of



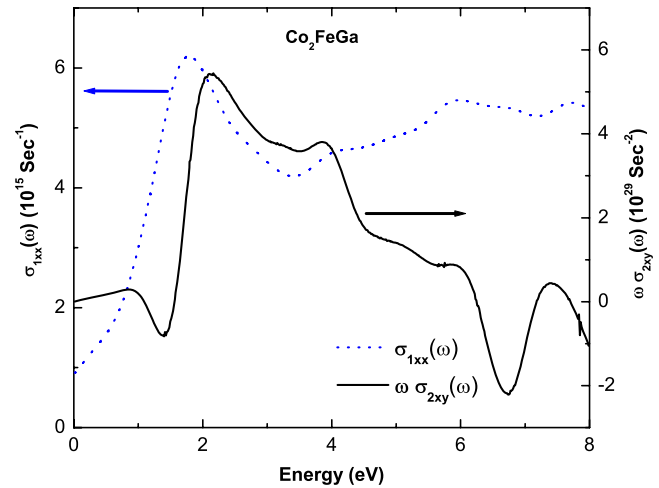
**Figure 4.** Calculated polar Kerr rotation  $\theta_K$  (left panel) and ellipticity  $\epsilon_K$  of  $\text{Co}_2\text{FeSi}$  and  $\text{Co}_2\text{FeGe}$  using GGA and GGA +  $U$  approaches.

the diagonal optical conductivity tensor element,  $\sigma_{1xx}$ , and of the off-diagonal part,  $\sigma_{2xy}$ , are connected via equation (4) to the microscopic interband optical transitions and thus provide more physical insight.

To understand the effect of interband transitions on the MO properties, experimentalists often display  $\omega\sigma_{2xy}$  instead of  $\sigma_{2xy}$  alone. In view of this, we compare the Kerr rotation spectra with the features of the  $\sigma_{1xx}$  and  $\omega\sigma_{2xy}$  spectra shown in figure 5. We expect the Kerr effect in all these compounds to have the same origin. Hence, we have displayed the  $\sigma_{1xx}$  and  $\omega\sigma_{2xy}$  spectra only for  $\text{Co}_2\text{FeGa}$ . The dominant peak present in both the Kerr spectra and  $\omega\sigma_{2xy}$  spectra is at almost the same photon energy, 2 eV, suggesting that the main contribution to the Kerr spectra is from the off-diagonal optical conductivity in accordance with equation (2). The magnitude of  $\omega\sigma_{2xy}$  is smaller for energy  $E < 1.5$  eV whereas  $\sigma_{1xx}$  increases almost linearly in magnitude up to energy 2 eV. Due to having a large magnitude of  $\omega\sigma_{2xy}$  near 2 eV, we get the maximum magnitude of Kerr rotation  $|\theta_K|$  near this energy. In the energy range 2–6 eV,  $\omega\sigma_{2xy}$  starts decreasing while  $\sigma_{1xx}$  shows much less variation beyond 2 eV. The combined effect is to reduce the magnitude of Kerr rotation. In test calculations, without spin-orbit coupling, we obtained  $\sigma_{xy}(\omega) = 0.0$  and therefore Kerr rotation  $\theta_K = 0$ . Thus, spin-orbit coupling plays a crucial role by introducing asymmetry for the RCP and LCP electromagnetic waves in producing large Kerr rotation.

## 5. Conclusions

In summary, we have presented a detailed first-principles characterization of the magnetic and MO Kerr effect in  $\text{Co}_2\text{FeX}$  ( $X = \text{Al, Ga, Si}$  and  $\text{Ge}$ ) using LSDA and GGA. The magnetic moments for the present compounds are found to be very large ( $\geq 5 \mu_B$ ). The main contribution to the magnetic moments comes from Co and Fe atoms. There is a very



**Figure 5.** Absorptive parts of diagonal and off-diagonal optical conductivity of  $\text{Co}_2\text{FeGa}$  using GGA (units:  $\omega\sigma_{xy}^{(2)}(\omega)$  is in  $10^{29} \text{ s}^{-2}$ ,  $\sigma_{xx}^{(1)}(\omega)$  is in  $10^{15} \text{ s}^{-1}$ ).

good agreement of our GGA values with the experimental values for the magnetic moment of  $\text{Co}_2\text{FeGa}$  and  $\text{Co}_2\text{FeAl}$ , but not that good for  $\text{Co}_2\text{FeSi}$  and  $\text{Co}_2\text{FeGe}$ . We find that the agreement gets significantly improved on performing the GGA+ $U$  calculations. We note that all the present compounds are governed by the half-metallic ferromagnetism of type III. Using GGA,  $\text{Co}_2\text{FeAl}$  has the maximum spin polarization (93%) among the present compounds, showing its strong tendency to be a type I half-metal to a good extent, whereas with GGA +  $U$ , the maximum spin polarization ( $P$ ) becomes almost unity for  $\text{Co}_2\text{FeSi}$  and  $\text{Co}_2\text{FeGe}$ .

We have observed that our results on MO spectra show most of the features of the experimental data, though both Kerr rotation and ellipticity spectra are shifted towards higher



energy by about 0.8 eV for  $\text{Co}_2\text{FeX}$  ( $X = \text{Al}$  and  $\text{Ga}$ ). In the present Co-based Heusler compounds, a p-block element ( $X$ ) of the same group has an almost negligible influence on Kerr spectra, i.e. substitution of Al with Ga or Si with Ge hardly affects the calculated Kerr spectra. We have observed that GGA is essential to give an overall improvement of Kerr spectra with experiments and to describe more accurately the electronic and magnetic properties for the present compounds, whereas with LSDA the agreement is relatively poor. Our calculations suggest that the low energy discrepancy in the previous theoretical calculation [19] for  $\text{Co}_2\text{FeX}$  ( $X = \text{Al}$  and  $\text{Ga}$ ) from experimental trends is removed by the use of the full potential and the effect of the Drude term is rather limited for all compounds in optical conductivity due to the observation of a small difference in Kerr spectra with and without inclusion of the Drude (intraband) contribution. We observe that, though the GGA +  $U$  calculations give significant improvement over the GGA results for the magnetic moment of  $\text{Co}_2\text{FeX}$  ( $X = \text{Si}$  and  $\text{Ge}$ ), the Kerr spectra for these compounds have similar characteristics with these two approaches.

From detailed analysis, we have found that the optical transitions in Kerr spectra are governed by the absorptive parts of optical conductivity. The main peak in the Kerr rotation is due to the maximum in  $\omega\sigma_{2xy}$  at 2 eV. The observation of the peaks in MOKE spectra at high energies (corresponding to smaller wavelengths) is very promising for the possible technological applications for high density MO recording.

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