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Half-metallic ferromagnets and their magneto-optical properties (invited)

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We have calculated the electronic structure of PtMnSb in order to explain the very high magneto-optical Kerr effect (over 2.5° at 720 nm at room-temperature) of this compound. It is shown that this behavior is related to the unusual electronic properties of PtMnSb: it is a half-metallic ferromagnet like NiMnSb. The extreme asymmetry in the electronic structure of these compounds—metallic behavior for one spin direction and at the same time semiconducting behavior for the other spin direction—is responsible for the unusual magneto-optical properties.

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I. INTRODUCTION

The magneto-optical Kerr effect (MOK) has been studied extensively over the past several years. Much of this interest has centered on the use of this technique to investigate the electronic structure of magnetic systems. Also, part of the interest stems from the possibility of using the MOK for erasable optical recording. PtMnSb has the highest MOK at room temperature of all known metallic systems.¹ Recently we reported on the electronic structure of the isostructural isoelectronic compound NiMnSb.² This ferromagnetic compound showed a very unusual electronic structure: it is a metal for the majority-spin electrons while at the same time it is a semiconductor for the minority spin electrons. This situation was named half-metallic ferromagnetism. In this paper the electronic structure of PtMnSb is described. It will be shown that the electronic structure resembles that of NiMnSb very closely. However, there is one important difference: the position of the Fermi level with respect to the top of the valence band of the minority-spin direction. This feature has important consequences when spin-orbit interaction is included. As will be shown by perturbation theory, the top of the spin-orbit-split valence band for the minority-spin direction is just above the Fermi level. Usually the MOK is caused by incomplete cancellations of left and right polarized electronic excitations. In PtMnSb we have the situation that one type of excitation is absent resulting in a large contribution to the MOK.

This paper is organized as follows: In Sec. II we describe the electronic structure of PtMnSb as obtained from a self-consistent scalar-relativistic spin-polarized augmented spherical wave (ASW) calculation. The effect of spin-orbit interaction in magnetic systems is discussed in Sec. III. Section IV briefly summarizes the MOK. Section V is devoted to the calculation of oscillator strengths. The results from the preceding sections will be combined in Sec. VI.

II. THE ELECTRONIC STRUCTURE OF PtMnSb

PtMnSb crystallizes in the Heusler C1b structure, sometimes referred to as the MgAgAs structure. This structure is face-centered cubic, with atomic positions as follows: Pt has the positions 0,0,0;fcc, Mn occupies 0.25,0.25,0.25;fcc, while Sb resides at 0.75,0.75,0.75;fcc. This structure can be thought of as the zinc-sulphide structure with the difference that the empty 0.75, 0.75, 0.75 position in the zinc-sulphide structure is occupied now by the third constituting element of the compound. The 0.5,0.5,0.5 position is still empty, in contrast with the Heusler L21 structure where this crystallographic position is also filled. Thus the manganese and the antimony atoms are coordinated by tetrahedra of platinum atoms, while the platinum is coordinated by two interpenetrating tetrahedra, one composed of manganese atoms, the second composed of antimony atoms.

The electronic band structure calculations were performed using the self-consistent augmented spherical wave (ASW) method by Williams, Kuebler, and Gelatt.³ Scalar relativistic effects were included as described by Methfessel and Kuebler.⁴ The empty 0.5,0.5,0.5 positions were treated as atoms with nuclear charge 0. The basis functions were composed of *s*, *p*, and *d* functions on all sites. Self-consistency was achieved in 13 iterations to a precision of $1 : 10^{-5}$. The experimental lattice constant was used (11.73501 a.u.). Since this lattice constant resulted only in a small pressure, no attempts were undertaken to minimize the total energy as a function of lattice constant. Figure 1 shows the band structure for the majority and minority spin direction along the high symmetry lines. This band structure shows a close resemblance to the bandstructure of NiMnSb.² In this calculation PtMnSb is also a half-metallic ferromagnet, i.e., the majority electrons show metallic behavior, while the minority electrons have an electronic structure of a semiconductor. It

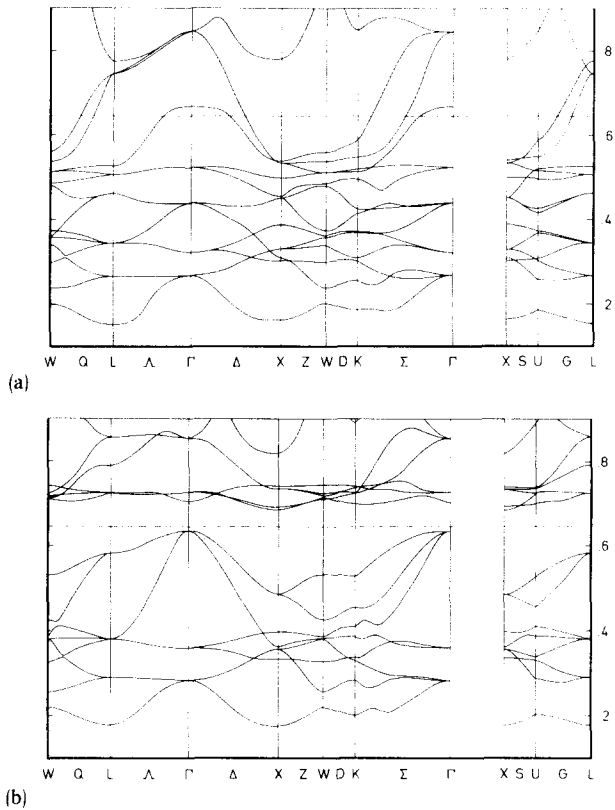


FIG. 1. Band structure of PtMnSb for (a) the majority spin direction and (b) the minority-spin direction. A band at low energies of primarily Sb *s* character has been omitted for visual clarity.

must be realized that this situation is very different from that in other ferromagnets as for example Ni. It is also true in Ni that there is no density of states of *d* electrons at the Fermi energy in the majority-spin direction. But there is an *s* band still intersecting the Fermi energy in both spin directions leading to normal metallic conduction in the minority as well as in the majority band. As a matter of fact, the majority spin direction band structure of Ni resembles quite closely that of Cu, a good metal.⁵ Here the opposite is true. In half-metallic ferromagnets there is no density of states at all at the Fermi level for one spin direction (in this case for the minority spin direction). Semiconducting behavior for this spin direction occurs, similar to the III-V semiconductors, while the opposite spin direction has metallic properties. To illustrate this point more clearly we display in Fig. 2 the total density of states for the two spin-directions. The arrows indicate the position of the Fermi energy. This figure also shows the largest difference between NiMnSb and PtMnSb. In NiMnSb the Fermi level is positioned well into the gap in the minority-spin direction, somewhat closer to the conduction band than to the valence band. In PtMnSb the Fermi level is lower: just above the valence band. (This is due to the difference in electronegativity of Pt and Ni.) The inclusion of spin-orbit interaction will cause the top of the minority-spin direction valence band to cross the Fermi energy as will be shown in the next section.

III. SPIN-ORBIT INTERACTION IN MAGNETIC SYSTEMS

In nonmagnetic systems, the effect of the spin-orbit operator,

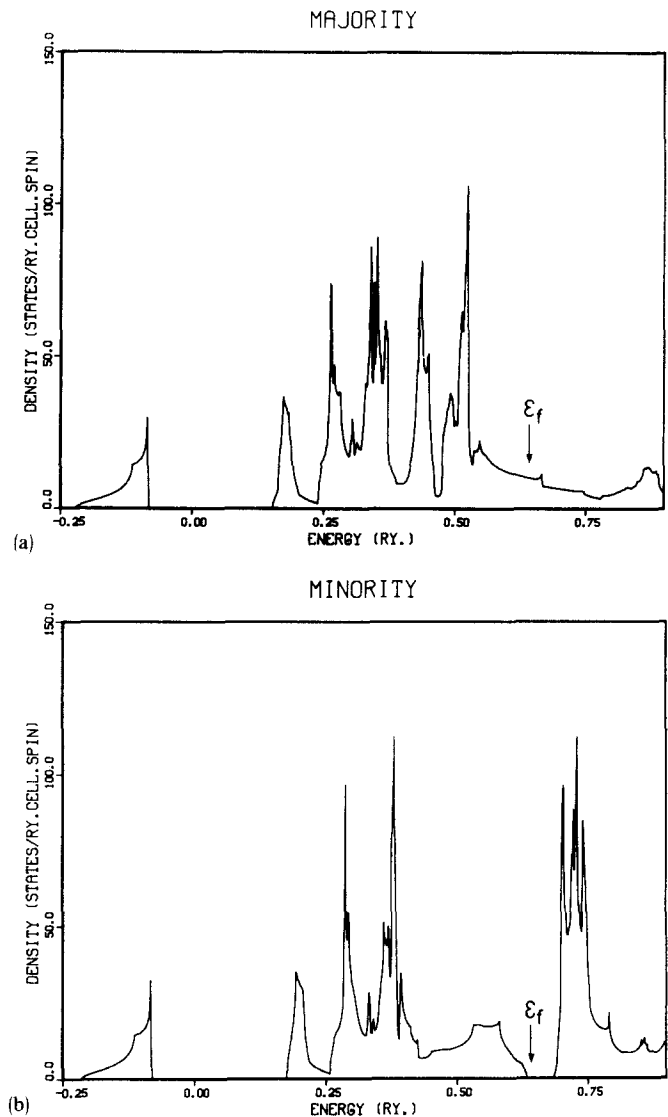


FIG. 2. Total density of states of PtMnSb for the majority and minority spin directions.

$$l s = \zeta [l_z s_z + 1/2(l^+ s^- + l^- s^+)], \quad (1)$$

is straightforward. In magnetic systems the situation is more complicated because the nondiagonal parts of the spin-orbit operator compete with the exchange interaction Δ . Since we are interested in the highest valence band states for the minority-spin direction and since these states are primarily composed of Sb *p* states the spin-orbit interaction of six isolated (atomic) *p* states will be considered first. Figure 3 shows the energy levels for this case as a function of the spin-orbit parameter 2ζ . Two cases are considered: the nonmagnetic case $\Delta = 0$ and the case where the *p* states are split into 3 spin-up and 3 spin-down states by an exchange splitting 2Δ in the absence of spin-orbit interaction. For small values of the spin-orbit interaction these triplets split into equidistant singlets: The $s = 1/2$ state into $m = -1, 0, +1$ states and the $s = -1/2$ state into $m = +1, 0, -1$ states, respectively. The equidistant splittings for small spin-orbit interaction are obtained if the nondiagonal parts of the spin-orbit interaction are negligible compared with the exchange interaction. For very large values of the spin-orbit interaction the

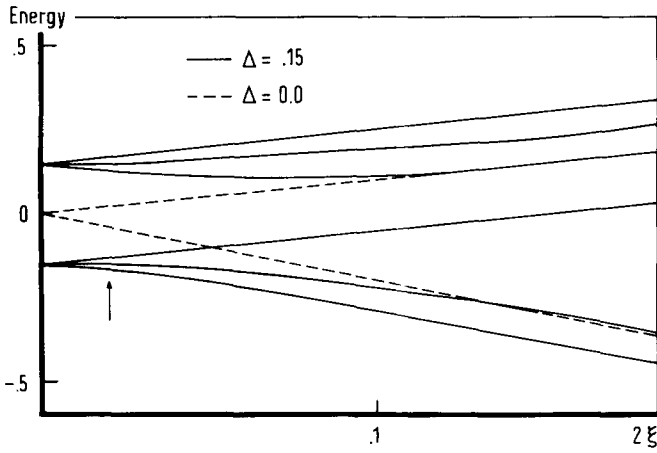


FIG. 3. Energy levels of a p state as a function of spin-orbit interaction strength. Two cases are plotted: The nonmagnetic case (broken lines) and the case with an exchange splitting of 0.15 for the nonrelativistic case. The arrow indicates the actual ratio of the parameters for PtMnSb.

levels form two groups: one group of two singlets and one group of four singlets. The situation becomes comparable with the $\Delta = 0$ case in this limit, but the levels never become degenerate as in the nonmagnetic case. The arrow in Fig. 3 represents the actual situation of the top of the valence band in the minority-spin direction of PtMnSb. The value of the exchange splitting was derived from the self-consistent band structure calculation, the spin-orbit parameter from spectroscopic data of Sb.⁶ The conclusion is that in PtMnSb we have a situation where the exchange interaction dominates the spin-orbit interaction.

In the solid the effect of orbital quenching needs also to be considered. At Γ the three p states are degenerate so no orbital quenching occurs. The maximum spin-orbit interaction is expected here. There are other parts in the zone where orbital quenching is not complete but excitations from these levels show up in the MOK spectrum at wavelengths completely different from the ones we are interested in here. Therefore we will concentrate on Γ . Figure 4 shows the situation at Γ of the minority-spin direction around the Fermi energy. As stated before, the top of the valence band Γ_4 state is primarily composed of Sb p states. The singlet Γ_1 level is of mixed Pt and Mn s character and shows no spin-orbit splitting. The Γ_3 level is almost exclusively made out of Mn d states: its spin-orbit interaction is negligible compared with the spin-orbit interaction of the Sb p states. The Fermi energy is indicated by ϵ_f . We see an important consequence here: the $m = +1$ top of the valence band has crossed the Fermi energy and is empty now. The consequences of this situation for the MOK will be discussed after a small excursion into the theoretical background of the MOK.

IV. THE MAGNETO-OPTICAL KERR EFFECT

The magneto-optical Kerr effect has been discussed extensively in general⁷ and in connection with experiments on Ni.^{8,9} Since it is well documented some crucial steps will be repeated here only; a more thorough discussion can be found in the literature. The following discussion concentrates on cubic systems with the axis of magnetization along the z direction. In first order the MOK is determined by the off-

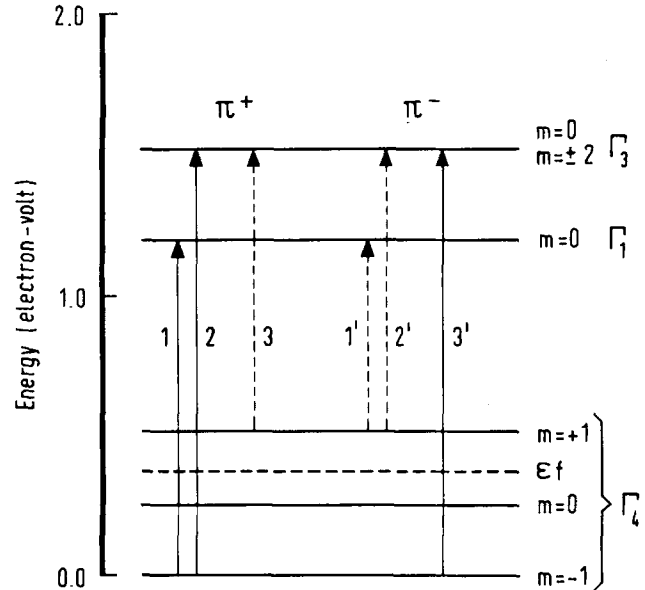


FIG. 4. Energy levels around the Fermi energy for the minority-spin direction. The arrows indicate the various allowed optical transitions. The excitations which are eliminated by the crossing of the $m = +1$ level and the Fermi level have been drawn with broken lines.

diagonal element $i\epsilon_{xy}$ of the dielectric tensor ϵ . It can be shown that the absorptive part of this off-diagonal matrix element is given by^{7,10}

$$W^2 \text{Im}(E_{xy}) = C \sum_{\alpha\beta} \{ |\langle \beta | \Pi^- | \alpha \rangle|^2 - |\langle \beta | \Pi^+ | \alpha \rangle|^2 \} \delta(W_{\alpha\beta} - W), \quad (2)$$

where $|\alpha\rangle$ and $|\beta\rangle$ are occupied and empty states, respectively, and Π^\pm is defined by

$$\Pi^\pm = \Pi_x \pm i\Pi_y,$$

and

$$\Pi = P + (e/c)A + (1/2mc^2)\sigma \times \nabla,$$

consists of the nonrelativistic momentum operator p and terms introduced by the spin-orbit interaction. The MOK originates from an incompleteness of the cancellation of the differences between the squares of the transition probabilities in Eq. (2). This difference vanishes in the absence of spin-orbit interaction or magnetization and external magnetic fields. Usually three different contributions to the MOK are distinguished.¹⁰

(1) The spin-orbit part to the Π operator alone can contribute. This contribution is usually considered to be small.⁹

(2) The spin-orbit operator can modify the functions $|\alpha\rangle$ and $|\beta\rangle$ in a different way, so that the terms in Eq. (1) no longer cancel. This is considered to be the dominant effect in the case of not too short wavelengths and not too large spin-orbit interactions; in other words, in cases where the bandwidth exceeds the spin-orbit interaction.

(3) At excitation energies so large that core-like initial states become involved, another contribution to the MOK enters the picture. The effect of the spin-orbit interaction on these energy levels is so large (compared with the bandwidth) that the Π^+ transition will take place at a quite different position in the spectrum than the Π^- transition and a MOK

results even in the case of equal oscillator strengths. This is the case for example in Ni with the M_{23} absorption edge.¹⁰

In PtMnSb, due to the special positions of the energy levels a new contribution to the MOK is possible, different from the three previously described ones. In Fig. 4 we have indicated the various Π^+ and Π^- excitations possible between the levels of interest. If the Fermi energy had fallen into the gap, transition 1 would have (partially) cancelled transition 1', transition 2 would have cancelled 2', and transition 3 would have cancelled transition 3'. Since the $m = +1$ singlet of the Γ_4 state is empty now, it cannot serve as an initial state anymore. In particular the transitions 3, 1', and 2' are no longer possible, leaving transitions 1, 2, and 3' completely uncompensated. One might be tempted to assume that transitions 2 and 3' will cancel and to attribute the observed MOK to transition 1. (Ironically, this will be our final conclusion, although based on a different argument.) Up to now our picture is only qualitatively correct and there is absolutely no reason to assume that excitations 2 and 3' cancel, not even in an atomic case. What needs to be done is to calculate the actual oscillator strengths from the band structure results with this qualitative picture in mind.

V. CALCULATION OF OSCILLATOR STRENGTHS

The evaluation of the oscillator strength can be done straightforwardly by direct integration. However, especially when one is only interested in the matrix elements of the momentum operator in a part of the Brillouin zone around a high symmetry point, an alternative approach is useful. In $k \cdot p$ perturbation theory one can describe the band structure locally to a very high accuracy by free electron terms and dot products of the k vector and the momentum operator matrix elements. A fit of the $k \cdot p$ Hamiltonian to the bands of interest yields all the momentum operator matrix elements directly. We have used this approach here, neglecting the perturbation of the initial wave functions by the spin-orbit operator. Transitions 1, 2, and 3 in Fig. 4 show oscillator strengths of 30.8, 3.3, and 8.8, respectively. The numbers refer to the squares of the matrix elements in units of electron volts. It is clear that transition 1 is the strongest transition. Transitions 2 and 3' cancel only partially but are inherently weaker than transition 1.

VI. CONCLUDING REMARKS

From the experimental Kerr rotation and ellipticity together with the diagonal optical constants obtained from ellipsometry measurements the absorptive part of the off-diagonal element of the dielectric tensor [Eq. (1)] can be

calculated. This experimental curve does not show much structure apart from a rather broad strong peak at 2 eV. It has been demonstrated that there is a strong magneto-optical absorption at Γ at an energy of 1.2 eV. The joint density of states (JDOS) has to be considered here also. The JDOS shows an onset caused by transitions at Γ . It increases rapidly with energy because excitations away from Γ become important [see Fig. 1(b)] and the volume in k space over which the excitations take place increases with energy. Thus there are two competing factors here: at Γ (or close to it) the contributions to the MOK is very high but the JDOS is small, while further away from Γ the spin orbit diminishes because of orbital quenching. This last effect together with the fact that the valence bands bend down away from Γ causes the upper $m = +1$ level of the valence band to cross the Fermi level with the consequence of the vanishing of the unusual contribution to the Kerr effect. Thus theoretically a peak in the expression of Eq. (1) is expected with an onset of 1.2 eV, in excellent agreement with experiment. To calculate quantitatively the JDOS requires a full relativistic magnetic calculation on PtMnSb. Such a calculation is nearly completed and preliminary results support the semiquantitative picture outlined in this paper.

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