Orbital magnetization in dilute ferromagnetic semiconductors

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The relationship between the modern and classical Landau’s approaches to carrier orbital magnetization is theoretically studied within the k · p formalism, taking dilute ferromagnetic (Ga,Mn)As as an example. It is shown that while the evaluation of hole magnetization within the modern theory does not require information on the band structure in a magnetic field, the number of basis wave functions must be much larger than in the Landau approach to achieve the same quantitative accuracy. A numerically efficient method is proposed, which takes advantages of these two theoretical schemes. The computed orbital magnetization is in accord with experimental values obtained by x-ray magnetic circular dichroism in (III,Mn)N compounds. The direct effect of the magnetic field on the hole spectrum is studied too, and employed to interpret a dependence of the Coulomb blockade maxima on the magnetic field in a single electron transistor of (Ga,Mn)As.

The last decade have witnessed the discovery of striking phenomena associated with geometric and topological aspects of the band structure, brought about by the presence of spin-orbit coupling and the breaking of spin rotation symmetry. In the case of ferromagnets the Berry curvature of bands hosting spin-polarized itinerant carriers was found to result in sizable contributions to transport coefficients, such as the anomalous Hall conductance. It has been suggested more recently that the Berry curvature in these systems describes also the orbital part of carrier magnetization coming from delocalized circulation. This recent development is particularly worthwhile, as it has delivered formulae for the total carrier orbital magnetization in the form that can be directly implemented into ab initio methods, allowing to interpret theoretically experimental values of the orbital magnetic moment provided by, for instance, x-ray circular magnetic dichroism (XMCD). The verification of the modern theory in this way is especially meaningful since, comparing to transport coefficients, thermodynamic properties are less sensitive to scattering and localization.

In this paper, we examine quantitatively magnetization of spin-polarized valence band holes in dilute ferromagnetic semiconductors (DFS). A particular versatile method to model semiconductor properties and devices is the Kohn-Luttinger (KL) k · p theory, whose six band version has been exploited to describe various thermodynamic and transport data, including the anomalous Hall effect. Within this scheme, we compare carrier magnetization obtained from the modern approach and determined employing the time-honored Landau theory.

According to the combined KL and Landau’s method, the spin-orbit interaction generates two contributions to orbital magnetization. The kinetic one stems from the orbital Landau’s quantization and is described by the Luttinger’s parameters γ1, γ2, and γ3. The second contribution, parameterized by Luttinger’s κ, is proportional to the orbital angular momentum operator, i.e., it is Zeeman-like. As we demonstrate here, only the first contribution is reproduced by the modern approach but the second term emerges if the set of the basis wave functions in the modern approach is enlarged. Furthermore, we show that both contributions have to be taken into account to describe quantitatively experimental results on XMCD and on the dependence of the chemical potential on the magnetic field in (Ga,Mn)As.

Hole magnetization in (Ga,Mn)As was originally calculated by an explicit algebraic determination of the Landau-levels energies and the corresponding partition function from which the carrier Gibbs free energy Gc and then magnetization M, also in the limit of the vanishing external magnetic field, can be obtained. The six-band k · p Kohn-Luttinger Hamiltonian of holes in a magnetic field and subject to the p-d exchange coupling to localized Mn spins was adopted in the spherical approximation that allowed the description of the valence band in terms of decoupled 6 × 6 blocks. The spherical approximation was relaxed in our previous work at the cost of a huge numerical complexity. Comparing to magnetization of localized Mn spins, the total magnetization of itinerant holes determined for (Ga,Mn)As was found to have an opposite sign and a few times smaller magnitude.

Within the above valence band model the magnetic field acting on the carriers B = ∇ × A is incorporated into the relevant Kohn-Luttinger Hamiltonian via the Peierls substitution, k → −i∇ + cA/ℏ and by adding the free electron Pauli term, g0μBB · s. The resulting Hamiltonian, neglecting the lack of inversion symmetry, consists of the term H_L describing the Landau quantization of the valence band and of the Zeeman-like contribution which, in the basis employed previously, reads

$$H_Z = -μ_B \left[ 3 \left( \kappa + \frac{g_0}{6} \right) \hat{I} - g_0 \hat{s} \right] \cdot B,$$  \hspace{1cm} (1)
operators \( \hat{I} \) and \( \hat{s} \) are given by,

\[
\hat{I} = \left( \begin{array}{cc} \frac{2}{3} & \hat{J} \\ \frac{2}{T} & \frac{2}{3} \hat{\sigma} \end{array} \right), \quad \hat{s} = \left( \begin{array}{cc} \frac{1}{3} \hat{J} & -\hat{U} \\ -\hat{T} & -\frac{1}{3} \hat{\sigma} \end{array} \right).
\]

Following Ref. [21] we denote by \( \hat{\sigma} \) the Pauli matrices, by \( \hat{J} \) the set of spin-3/2 angular-momentum matrices, and by \( \hat{U}, \hat{T} \) the sets of matrices for the cross-space. Besides the term proportional to \( g_0 = 2.002 \) (the ordinary Pauli spin part), there is an orbital term proportional to \( \kappa \equiv (\kappa + g_0)/6 \), where \( \kappa \) is the Luttinger parameter. It represents a contribution to the orbital magnetic moment carried by an admixture of the s-like states to the p-like valence band Bloch wave functions.

Finally, the p-d coupling to the spin-polarized Mn ions is taken into account in the virtual-crystal and molecular-field approximations, leading to additional giant spin-splitting of Landau levels, described by the Hamiltonian

\[
\mathcal{H}_{pd} = (\Delta_\nu / M) \mathbf{M} \cdot \hat{s},
\]

where \( \mathbf{M} \) is Mn magnetization and \( \Delta_\nu \) is the p-d exchange splitting of the valence band top. [1110]

Within Landau’s method [14,16] the carrier magnetization \( \mathbf{M}_c(T, \mathbf{H}) \) is given by the derivative of the Gibbs free energy,

\[
G_c = -\mu_B B k_B T \int_0^\infty \frac{m_0 dk_3}{2(\pi h)^2} \sum_j \ln \{1 + \exp \left(-[E_j(k_3) - \mu]/k_B T\right)\},
\]

with respect to the external magnetic field, \( -\partial G_c / \partial \mathbf{H} \). It is assumed that in the case in question \( \mathbf{B} = \mu_0 \mathbf{H} \); \( E_j(k_3) \) is the \( j \)-th eigenenergy of \( \mathcal{H}_L + \mathcal{H}_Z + \mathcal{H}_{pd} \) for a carrier with the \( \mathbf{k} \) component along the direction of the magnetic field denoted as \( k_3 \), and \( \mu \) is the chemical potential. The values of \( \mathbf{M}_c \) computed in this way for (Ga,Mn)As were reported previously.[20]

Within the modern approach the orbital part of \( \mathbf{M}_c \) at \( H = 0 \) for \( n \) bands is given by,

\[
\mathbf{M}_{orb}^{(mod)} = \mu_B \int \frac{d^3k}{(2\pi)^3} \sum_{n,n'} M_T(E_{n',k}, E_{nk}) \left\{ \text{Im} \left[ m_0 \langle u_{nk} | \bar{\mathbf{v}} | u_{n'k} \rangle \times \langle u_{n'k} | \bar{\mathbf{v}} | u_{nk} \rangle \right] \right\},
\]

where \( u_{nk} \) is the Bloch function corresponding to the eigenenergy \( E_{nk} \) of the Hamiltonian \( \mathcal{H}_k = \mathcal{H}_{KL} + \mathcal{H}_{pd} \), \( \mathbf{h} \mathbf{v} = \partial \mathcal{H}_k / \partial \mathbf{k} \), and

\[
M_T(E_{n',k}, E_{nk}) = M \left[ (E_{n',k} - \mu)/k_B T, (E_{nk} - \mu)/k_B T \right] / k_B T,
\]

where the dimensionless function \( M \) reads,

\[
M(x_n', x_n) = \frac{1}{x_n' - x_n} \left[ \frac{f(x_n') + f(x_n)}{2} + \ln \left[ \frac{1 + \exp(-x_n')}{1 + \exp(-x_n)} \right] \right]
\]

with \( f(x) = [1 + \exp(x)]^{-1} \).

The expression displayed in Eq. (4) unifies the two components of \( M_{orb}^{(mod)} \) and substantiates a picture in which orbital magnetization is described by a sum over pairs of subbands, with significant contributions only from empty--occupied ones. Due to antisymmetrization that has been performed, \( M(x_n', x_n) = -M(x_n, x_n') \), \( M \) vanishes rather than diverges at the band crossings, as required for degenerate bands. As seen in Fig. 1 the function \( M(x_n, x_n') \) beside the antisymmetry, obeys

\[
M(-x_n, -x_n') = M(x_n, x_n') \text{ (electron-hole symmetry)} \quad \text{and} \quad M(x_n, -x_n) = 0.
\]

We first compare hole orbital magnetization \( M_{orb}^{(mod)} \) determined within the \( \mathbf{k} \cdot \mathbf{p} \) formalism from the modern approach (Eq. (4)) and from the Landau theory (Eq. (3)) in the limit \( H \to 0 \) employing the six band Hamiltonian \( \mathcal{H}_L + \mathcal{H}_{pd} \), i.e., neglecting \( \mathcal{H}_Z \). For Ga\textsubscript{1-x}Mn\textsubscript{x}As, in the explored parameter space (\( T = 10 \text{ K}, 5 \times 10^{19} \leq p \leq 10^{21} \text{ cm}^{-3} \), and \( \Delta_\nu = -180 \text{ meV}, \ i.e., \ x \simeq 0.05 \), the relative difference between the data obtained by these two methods is within our numerical uncertainty of \( 10^{-6} \). This finding highlights a major progress provided by the modern approach that allows one to circumvent the computational load associated with the determination of Landau level energies for complex band structures. Furthermore, this new formalism makes it possible to evaluate separately contributions coming from the local circulation (LC) and itinerant circulation (IC), the latter proportional to the Berry curvature.[22,23] As shown in Fig. 2(a), similarly to ferromagnetic metals (Fe, Co, Ni),[24] these two contributions have opposite signs and the term corresponding to local circulation dominates.

However, orbital effects contribute also to the Zeeman Hamiltonian displayed in Eq. (1). The magnitude of corresponding magnetization \( M_k \) can be evaluated from the Gibbs free energy (Eq. (3)) with eigenenergies \( E_{nk} \) of the Hamiltonian \( \mathcal{H}_k + \mathcal{H}_Z \) in the limit \( H \to 0 \) for \( g_0 = 0 \) and \( \kappa = 1.534 \). According to results presented in Fig. 2(b), \( M_k \) compensates largely \( M_{orb}^{(mod)} \) provided by the modern approach. For comparison we also show in Fig. 2(c) the total hole magnetization \( M_c \) that is seen to be dominated by the spin part \( M_{spin} \), obtained by putting \( \kappa = 0 \)
and $g_0 = 2.002$ in $\mathcal{H}_Z$. Similar results for (In,Ga)As are shown in Fig. 3.

In order to find out how to determine the missing part $M_\kappa$ within the modern approach, we have computed orbital magnetization within the eight-band model \(^{23}\) employing either the modern approach or within the Landau theory disregarding the Zeeman term (solid line), as decomposed into the local circulation (LC) and the itinerant circulation (IC) contributions (dashed and dotted lines, respectively); (b) total orbital magnetization ($M_{\text{orb}}$, solid line) from Landau’s method, as decomposed into $M_{\text{orb}}^{\text{(mod)}}$ [solid line in (a)] and the part $M_\kappa$ coming from the Zeeman Hamiltonian $\mathcal{H}_Z$ with $\kappa_I = 1.534$ and $g_0 = 0$ (dashed and dotted lines, respectively); (c) total hole magnetization $M_\zeta$ (solid line) decomposed into $M_{\text{orb}}$ [solid line in (b)] and the spin part $M_{\text{spin}}$ (dashed and dotted lines, respectively).

Actually, as shown in Fig. 4, $M_{\text{orb}}^{\text{(mod)}}$ has much larger magnitude and the opposite sign to $M_{\text{orb}}$. This divergence means that it is not enough to enlarge the basis of the $u_n$ functions by those corresponding to the conduction band to obtain a reasonable value of $M_{\text{orb}}$ within the modern approach. Indeed, according to results presented in Fig. 4, the six- and eight-band models give similar values of $M_{\text{orb}}$ if the modern theory is supplemented by $M_\kappa$ corresponding to the coupling to bands disregarded within the eight-band model, i.e., calculated from the relevant Zeeman Hamiltonian for $g_0 = 0$ and $\kappa_I' = \kappa_I - m_0 P^2/3h^2 E_g$, where for GaAs the momentum matrix element $P = 1.0493 \text{ eV nm}$, the band gap $E_g = 1.519 \text{ eV}$, and hence $\kappa_I' = -1.637$.

Altogether these findings imply that (i) the modern approach provides the magnitude of $M_{\text{orb}}$ without referring to the hole spectrum in an external magnetic field, but

FIG. 2. (Color online) Contributions to hole magnetization at 10K computed within the six-band Kohn-Luttinger model for parameters of the GaAs valence band ($\gamma_1 = 6.85$, $\gamma_2 = 2.1$, $\gamma_3 = 2.9$, $\kappa = 1.2$, $\Delta_{SO} = 0.341 \text{ eV}$, $\mathbf{M} \parallel (100)$), and the parameter of valence band exchange splitting $\Delta_L = -180 \text{ meV}$, corresponding to the magnitude of saturation magnetization in Ga$_{0.95}$Mn$_{0.05}$As: (a) orbital magnetization $M_{\text{orb}}^{\text{(mod)}}$ obtained by either the modern approach or from the Landau theory disregarding the Zeeman term (solid line), as decomposed into the local circulation (LC) and the itinerant circulation (IC) contributions (dashed and dotted lines, respectively); (b) total orbital magnetization ($M_{\text{orb}}$, solid line) from Landau’s method, as decomposed into $M_{\text{orb}}^{\text{(mod)}}$ [solid line in (a)] and the part $M_\kappa$ coming from the Zeeman Hamiltonian $\mathcal{H}_Z$ with $\kappa_I = 1.534$ and $g_0 = 0$ (dashed and dotted lines, respectively); (c) total hole magnetization $M_\zeta$ (solid line) decomposed into $M_{\text{orb}}$ [solid line in (b)] and the spin part $M_{\text{spin}}$ (dashed and dotted lines, respectively).

FIG. 3. (Color online) Carrier magnetization in (In,Mn)As computed within the eight-band model \(^{23}\) using the relevant Zeeman Hamiltonian $\mathcal{H}_Z$ with $\kappa_I = 1.534$ and $g_0 = 0$ (dashed and dotted lines, respectively) for (Ga,Mn)As with splitting of the valence and conduction bands, $\Delta_L = -180 \text{ meV}$ and $\Delta_\kappa = 30 \text{ meV}$, respectively. The dotted and dash-dotted lines show $M_{\text{orb}}^{\text{(mod)}}$ computed for the same parameters from the modern approach within the eight-band model (the latter does not include a constant offset which we disregard in the Landau approach).
the set of the basis wave functions $u_n$ must be much larger than needed within the Landau approach to achieve the same quantitative accuracy, and (ii) the most efficient numerical method is to compute orbital magnetization at $H = 0$ as $M_{\text{orb}} = M_{\text{orb}}^{(\text{mod})} + M_b$, which exploits advantages of these two equivalent theoretical schemes. Below, we apply this procedure for comparing experimental data to the eight-band $\mathbf{k} \cdot \mathbf{p}$ theory for (Ga,Mn)As.

Figure 5 presents the orbital moment of As $4p$ states determined by XMCD for (Ga,Mn)As and (In,Ga,Mn)As films with different saturation magnetizations $M_{\text{Sat}}$ and Curie temperatures $T_C$. Since orbital moment of cations appear to be much smaller than needed within the Landau approach to achieve the same quantitative accuracy, and (ii) the most efficient numerical method is to compute orbital magnetization at $H = 0$ as $M_{\text{orb}} = M_{\text{orb}}^{(\text{mod})} + M_b$, which exploits advantages of these two equivalent theoretical schemes. Below, we apply this procedure for comparing experimental data to the eight-band $\mathbf{k} \cdot \mathbf{p}$ theory for (Ga,Mn)As.

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![Experiment vs. Theory](image)

**Figure 5.** (Color online) Orbital magnetization of As $4p$ states determined experimentally at $\sim 10$ K and in 2 T by Wadley et al.\cite{Wadley2004} as a function of saturation magnetization $M_{\text{Sat}}$ for (Ga,Mn)As (open circles) and (In,Ga,Mn)As (open squares) compared to theoretical values of orbital magnetization $M_{\text{orb}}$ computed within the eight-band model for (Ga,Mn)As directly (empty diamonds) and including a possible contribution $\Delta M_{\text{orb}} = a \Delta \nu + b$, where $a$ and $b$ are fitting parameters (full squares).

In summary, as emphasized in this work, the Landau approach to orbital magnetization requires information on eigenenergies in the magnetic field. Within the Kohn-Luttinger method, the relevant energy spectrum can be determined employing a finite set of basis wave functions $u_n$, and by evaluating corrections originating from $\mathbf{k} \cdot \mathbf{p}$ coupling to remaining bands by the second order perturbation theory. In contrast, the accurate form of wave functions in the absence of a magnetic field is needed to compute orbital magnetization within the modern method. This means that in order to obtain a similar quantitative accuracy, a larger set of $u_n$ has to be employed than in the case of Landau’s theory. We have proposed a numerically efficient method that combines advantages of these two theoretical schemes. The computed hole magnetization within the formalism developed here explains the magnitude of orbital and spin magnetizations implied by experimental studies of XMCD and the Coulomb blockade in (Ga,Mn)As. An interesting question arises about implications of our findings to the theory of anomalous and spin Hall effects in semiconductors. We thank B. L. Gallagher, K. Edmonds, and P. Wadley for instructive discussions on XMCD results. This work was supported by the European Research Council through the FunDMS Advanced Grant (No. 227690) within the “Ideas” 7th Framework Programme of the EC.

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