What has been measured by reflection magnetic circular dichroism in $Ga_{1-x}Mn_xAs$/GaAs structures?*

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An explicit expression of reflection magnetic circular dichroism (R_{MCD}) has been derived, taking into account the interference effect that arises from multiple internal reflections in an air/$Ga_{1-x}Mn_xAs$/GaAs dielectric layered system. It unambiguously shows that the R_{MCD} signal is composed by three terms. In addition to the conventional term, which is sufficient in the absence of interference, an oscillatory term is required. Both of them are related to the imaginary part ε''_{xy} of the off-diagonal element of the dielectric tensor. One also becomes aware that in this case R_{MCD} is not actually determined only by the imaginary part ε''_{xy} of the off-diagonal element of the dielectric tensor, as has been widely accepted. In fact, the real part ε'_{xy} of the off-diagonal element will substantially mix into the measured R_{MCD} results by another oscillatory $\cos\theta$ form. It can even reverse the sign of R_{MCD}, when the $Ga_{1-x}Mn_xAs$ layer becomes thicker. The main aspects of these predictions were used to reasonably explain the R_{MCD} results measured in three different types of samples. Our work will bring about a reconsideration of how to correctly explain R_{MCD} results.

Keywords: magneto-optical effects, magnetic semiconductors, optical properties, III–V compounds

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1. Introduction

Magneto-optical spectroscopy has been widely used as a powerful tool to study the electronic structure of $Ga_{1-x}Mn_xAs$, a typical diluted magnetic semiconductor (DMS).[1–3] Especially, magnetic circular dichroism (MCD), due to its clear physical interpretation as the difference of absorption coefficients between right (σ^+) and left (σ^-) circularly polarized light, was believed to give valuable knowledge about spin-dependent electronic states in diluted magnetic semiconductors. However, previous MCD studies of $Ga_{1-x}Mn_xAs$ have given rise to much controversy about the value and sign of the p–d exchange coupling constant (J_{pd}), the position of the Fermi level, the contribution of the optical transitions originating from Mn impurity states, etc.[1,4–9] To clarify the contradictory implication of different experimental observations, many theoretical works on the MCD effect, based on either the spin-polarized valence band model or the impurity band model, have been also performed, mainly by first principles calculations.[7,10–13] Their results also support either model. Because the reported MCD data on $Ga_{1-x}Mn_xAs$ have multitudinous aspects, among which its dependence on the $Ga_{1-x}Mn_xAs$ layer thickness is somewhat strange,[10,14] one has to carefully recheck what are really measured by reflection and transmission MCD. By including the interference effect, induced by multiple reflections within a $Ga_{1-x}Mn_xAs$ layer of thickness d (300 nm) in Fresnel's formulas of reflection coefficients for both circular polarizations, Lang *et al.*[8] calculated both Kerr rotation and MCD as a function of the wavelength at different hole concentrations, and fit the measurement results with them. While they reproduced the oscillatory behavior in the measured spectra of Kerr rotation and MCD, they had to invoke a positive (ferromagnetic) exchange parameter. The oscillatory phenomena were also observed in $Ga_{1-x}Mn_xAs$/GaAs samples, and attributed to the influence of multiple reflections by Ref. [10], where the layer thickness of $Ga_{1-x}Mn_xAs$ is larger than 20 nm.

In the present work, by deriving the explicit expression of R_{MCD} in the case where the interference effect is included due to the multiple reflections in an air/$Ga_{1-x}Mn_xAs$/GaAs dielectric layer system, it is found that there are two additional $\cos\theta$ terms accompanying the conventional non-oscillatory term. One comes from the difference in the refractive indices between right and left circularly polarized lights. The other stems from the difference in the absorption coefficients between right and left circularly polarized light. Eventually, the latter term is even able to reverse the sign of R_{MCD} with increasing the layer thickness of $Ga_{1-x}Mn_xAs$. The main aspects of these predictions are used below to reasonably explain the R_{MCD} results measured in three different types of samples.

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2. The explicit expression of $R_{\rm MCD}$ when the interference is taken into account

We shall first derive an explicit expression for $R_{\rm MCD}$, when the interference effect, caused by the multiple reflections inside a $Ga_{1-x}Mn_xAs$ layer, is taken into account. Let us consider a special case, where the right and left circularly polarized light shines vertically on a dielectric layer structure consisting of air (0), a $Ga_{1-x}Mn_xAs$ layer (1) of thickness d, and a GaAs substrate (2). The GaAs substrate is considered to be infinitely thick with no reflected light beam back to the 1–2 interface. It is easy to strictly prove that after considering the multiple reflections inside the $Ga_{1-x}Mn_xAs$ layer, the sum of the reflection coefficients at the 0–1 interface is given by[8]

$$r^{\pm}(\omega)=\frac{r_{01}^{\pm}+r_{12}^{\pm}\exp(\mathrm{i}2\tilde{n}^{\pm}(\mathrm{GaMnAs})\omega d/c)}{1+r_{01}^{\pm}r_{12}^{\pm}\exp(\mathrm{i}2\tilde{n}^{\pm}(\mathrm{GaMnAs})\omega d/c)}. \quad (1)$$

Here $\tilde{n}=n+\mathrm{i}\kappa$ is the complex refractive index, having a real part of n and an imaginary part of κ (extinction coefficient). Their superscript "±" stands for the values of right/left circularly polarized light. The exponential factor in Eq. (1) is related to the absorption and phase factor of the light propagation inside the $Ga_{1-x}Mn_xAs$ layer by

$$a^{\pm}=\left|a^{\pm}\right|\exp(\mathrm{i}\theta^{\pm})=\exp(-\alpha^{\pm}d)\exp(\mathrm{i}n^{\pm}2\omega d/c), \quad (2)$$

$$\alpha^{\pm}=2\omega\kappa^{\pm}/c, \quad (3)$$

$$\theta^{\pm}=2\omega n^{\pm}d/c. \quad (4)$$

After a long and tedious algebraic derivation, a final $R_{\rm MCD}$ expression can be approximately obtained as

$$R_{\rm MCD}=\frac{R^{+}-R^{-}}{R^{+}+R^{-}}\approx\frac{4\Delta n}{n^2-1}\left[1\pm(1+\beta)\cos\theta\right], \quad (5)$$

as has been carefully verified in Appendix A. The plus and minus signs in the brackets of Eq. (5) are for the two cases $n>n_0$ and $n<n_0$, and β is the ratio of the second $\cos\theta$ term to the first term in Eq. (A27), which is equal to

$$\beta=2\frac{n_0-n}{n_0+n}\frac{\Delta\alpha d}{\Delta n/n}. \quad (6)$$

Several notable features are worthy of further discussion. It is clearly seen from Eqs. (5) and (A27) that when taking into account the interference effect in the $Ga_{1-x}Mn_xAs$ layer, $R_{\rm MCD}$ is composed by three terms. The first term on the right-hand side is the $R_{\rm MCD}$ without interference, which is contributed by Δn. However, in this case, an oscillatory $\cos\theta$ term is required, which is also related to Δn. For the specific case of $n=3.44$ and $n_0=3.7$,[8,15] the factor of $4nn_0/(n_0+n)^2$ in Eq. (A28) has a value of 0.999. This fact tells us that an oscillatory Δn term is going to make the total Δn-related term vary periodically between zero and two times $4\Delta n/\left(n^2-1\right)$.

Another oscillatory $\cos\theta$ term is related to the absorption difference between right and left circularly polarized light. For the purpose of estimation, we shall adopt the available values for n_0, n, $\Delta\alpha d$, and $\Delta n/n$. We have already used $n_0=3.7$ for GaAs and $n=3.44$ for $Ga_{1-x}Mn_xAs$. The value of $\Delta\alpha d$ is estimated from the relation $\Delta\alpha d=(2\omega/c)d\Delta\kappa=(4\pi d/\lambda)\Delta\kappa$ at $\lambda=805$ nm. The Δn and $\Delta\kappa$ are further related to the real and imaginary parts of the off-diagonal element of the dielectric tensor, ε'_{xy} and ε''_{xy}, through the equations of $\Delta n\approx-\varepsilon''_{xy}/n$ and $\Delta k\approx\varepsilon'_{xy}/n$, respectively.[16] We adopt the calculated values of $\varepsilon'_{xy}\approx0.1$ and $\varepsilon''_{xy}\approx-0.1$ from Ref. [14], and approximately obtain $\Delta\kappa\approx0.03$ and $\Delta n\approx0.03$. From these values it follows $\Delta\alpha d\approx0.045$ for a 100-nm-thick $Ga_{1-x}Mn_xAs$ layer. It is also found that the oscillatory $\cos\theta$ term of $\Delta\alpha d$ is in phase with the oscillatory $\cos\theta$ term of $\Delta n/n$ in the case of $n_0>n$. If n is taken to be 3.44,[8] β becomes 38%, and $1+\beta=1.38$. By increasing the $Ga_{1-x}Mn_xAs$ layer thickness, say, to 200 nm, β becomes as large as 76%, and $1+\beta=1.76$. It is clear that the mixing-up of $\Delta\alpha$, or ε'_{xy}, makes $R_{\rm MCD}$ switch its sign by scanning the wavelength. However, if $n>n_0$, β becomes negative, and the $\cos\theta$ term of $\Delta\alpha d$ has π phase difference from that of $\Delta n/n$. Now, one needs to have more knowledge about the refractive index of $Ga_{1-x}Mn_xAs$. In general, the dielectric constants of GaAs and $Ga_{1-x}Mn_xAs$ are composed of two parts. One is the usual high frequency part, ε_∞, and the other is contributed by interband transitions. Usually, both the real and imaginary parts (ε'_{xx} and ε''_{xx}) of the dielectric constant can be measured by a variable angle spectroscopic ellipsometer (VASE).[17] The refractive index is obtained by taking the square root of the complex dielectric constant. The measured ε'_{xx} in the photon energy range from 1.4 eV to 2.8 eV, seems to decrease with increasing Mn composition (see Fig. 3 of Ref. [17]), so does the refractive index n in the case of $\varepsilon''_{xx}\ll\varepsilon'_{xx}$. On the other hand, by checking the $n=\sqrt{\varepsilon_\infty}$ data of ternary and quaternary III–V compounds,[18] we find that there is also a trend that increasing the composition of light element will reduce n. Since Mn(54.9) < Ga(69.7), n(GaMnAs) may also be smaller than n(GaAs). Therefore, our discussion for the case of $n<n_0$ is reasonable. However, even in the case of $n>n_0$, the β value may still be large enough to reverse the sign of $R_{\rm MCD}$ in thick $Ga_{1-x}Mn_xAs$ layers. As an example, $\beta=2[(n_0-n)/(n_0+n)][\Delta\alpha d/(\Delta n/n)]\approx-2.03$ at $n_0=3.7$, $n=4.3$,[8] $d=200$ nm, and $\lambda=800$ nm (see Eq. (5)). Finally, one may check what happens in the special case of $n_0=n$. As shown by Eqs. (A31)–(A34), $R_{\rm MCD}$ approximately resumes its original form in the absence of the interference effect.

These conclusions drawn from Eq. (5) bring about a significant change in our understanding of $R_{\rm MCD}$ measurements. First, because two oscillatory $\cos\theta$ terms from Δn and $\Delta\alpha$ are in phase in the case of $n_0>n$, their additive contribution is able to easily reverse the sign of $R_{\rm MCD}$. Even for $n>n_0$, the oscillatory $\Delta\alpha$ term is still able to reverse the sign of $R_{\rm MCD}$ as long as the layer is thick enough. Second, recognition of

the interference effect implies that $R_{\rm MCD}$ is not simply related only to the imaginary part of the off-diagonal element of the dielectric tensor, ε''_{xy} (or Δn), but it is also significantly mixed by the contribution from its real part, ε'_{xy} (or $\Delta\alpha$).

3. Experimental methods and samples investigated

To check the applicability of the theoretical expressions above, for the sake of generality, three kinds of $Ga_{1-x}Mn_xAs$ samples were used for measuring $R_{\rm MCD}$. Sample A consists of a 100-nm-thick $Ga_{0.95}Mn_{0.05}As$ epilayer, grown by a low-temperature molecular beam epitaxy (LT-MBE) on the top of a semi-insulating $In_{0.19}Ga_{0.81}As$ (1000 nm)/GaAs composite substrate. Sample A showed a Curie temperature $T_{\rm C}$ of 110 K after post-annealing at 250 °C in air for 1 h, and its easy axis was vertical to the surface. Sample B (C) consists of a 200-nm (50-nm)-thick $Ga_{0.936}Mn_{0.064}As$ epilayer grown on a semi-insulating GaAs substrate by LT-MBE, having an in-plane easy axis and a Curie temperature of $T_{\rm C} = 51$ K ($T_{\rm C} = 36$ K).

For MCD measurements, the light beam from a high power supercontinuum source (Fianium SC450-4) first passed through a chopper (at 475 Hz), and was subsequently dispersed by a monochrometer to give a particular wavelength or scanned wavelength at its outlet. Then it became linearly polarized light by passing through a polarizer. Afterward, the light beam was fed through a photo-elastic modulator (PEM), and converted into an alternatively modulated clockwise-polarized and counterclockwise-polarized light at a frequency of 50 kHz. A polarization-insensitive beam splitter (NPBS) then split the light into two beams. One was sent into the first detector, and measured by the first lock-in amplifier at the chopper frequency for monitoring the intensity variation of the incident light. The other beam was shined almost vertically on the sample surface, which was placed inside a superconducting solenoid magnet. The field direction of it was parallel to optical axis. The intensity of its reflected beam was probed by the second detector, whose signal was fed into double lock-in amplifiers, one of which was referenced to the chopper frequency, and the other of which was referenced to the PEM's modulation frequency of 50 kHz. Finally, $R_{\rm MCD}$ was deduced by the output ratio of the last two lock-in amplifiers.

4. Experimental results and their comparison with $R_{\rm MCD}$ theoretical expression

Figure 1 first shows the magnetic field dependence of $R_{\rm MCD}$ for Sample A at different wavelengths. The nearly rectangular shapes of their hysteresis loops indicate that its easy axis is vertical to the sample surface. Although the saturated amplitudes at different wavelengths vary slightly, their coercive fields are exactly the same even beyond the long wavelength limit of the interband absorption (around 850 nm, as explained later). The polarity of the hysteresis loop at 750 nm is reversed as compared to that at either 880 nm or 920 nm. To show the possible interference effect, the $R_{\rm MCD}$ of Sample A was measured at 4 K by sweeping the wavelength from 980 nm to 600 nm under saturated magnetic fields of ± 0.08 Tesla applied perpendicularly to the surface. As seen in Fig. 2, when the direction of the applied magnetic field is reversed, the spectrum of $R_{\rm MCD}$, as a whole, is reversed as well. Their symmetry about the zero line indicates the high quality of our data. Two zero-cross points show up, one at the wavelength of 780 nm and the other at 633 nm, while a peak appears at the wavelength of 729 nm. However, none of them can be simply related to the interband transitions at critical points. For example, the transition from the split-off hole band to the conduction band is expected to occur at an energy of $E_\Gamma(T)+\Delta$ (Δ is the spin-orbit split-off at Γ point in the valence band), which is about 1.8 eV, corresponding to a wavelength of 690 nm, at 4 K.[8,19] More important is that the wavelength positions of all these zero-cross points and peaks change dramatically when the thickness of the $Ga_{1-x}Mn_xAs$ layer varies, as becomes clear later. In the vicinity of the energy gap, the $R_{\rm MCD}$ signal rises sharply at $\lambda \approx 850$ nm first, which is a feature commonly observed and attributed to the onset of an interband transition in $Ga_{1-x}Mn_xAs$.[9] Later, the $R_{\rm MCD}$ signal drops very quickly, and then sweeps its sign. After passing a peak of the reversed sign, it crosses the horizontal axis again. The above behavior can be reasonably explained by the interference effect described in Eq. (5). By recalling the relations of ε'_{xy} (or $\Delta\alpha$) and ε''_{xy} (or Δn) with the interband transitions,[8,10] one knows that whenever a new channel of spin polarized interband transition opens up, both of them will vary. The sharp rise at $\lambda \approx 850$ nm comes from the contribution from both $\Delta\alpha$ and Δn. Afterward, the oscillatory terms, $(1+\beta)\cos\theta$, become dominant. However, among them, the contribution from $\Delta\alpha$ is the only term able to reverse the sign of the total $R_{\rm MCD}$ signal. All of that can explain the phenomena observed in Fig. 2.

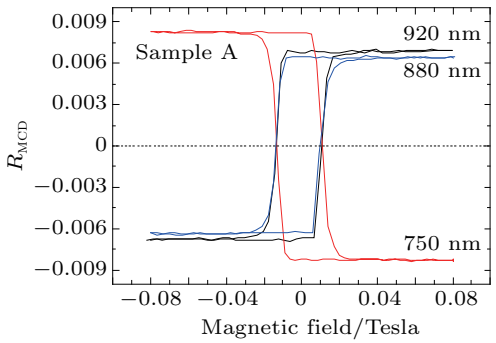


Fig. 1. (color online) $R_{\rm MCD}$ of Sample A measured at 4 K in a sweep of magnetic field between ± 0.8 Tesla for three different wavelengths.

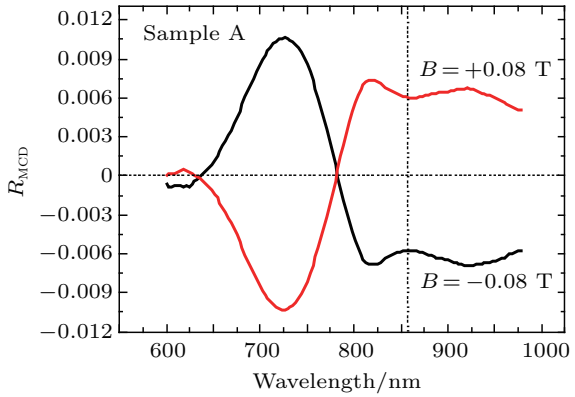


Fig. 2. (color online) Wavelength dependence of R_{MCD} measured at 4 K for Sample A under ±0.08 Tesla.

Sample B is of a different type, consisting of a 200-nm-thick $Ga_{0.936}Mn_{0.064}As$ epilayer grown on a semi-insulating GaAs substrate with an in-plane easy axis. Its wavelength dependence of R_{MCD} was measured at 4 K in Fig. 3 under several perpendicular magnetic fields (0, ±0.06, ±0.12, and ±0.25 Tesla). Not only does oscillatory behavior persist, but the patterns can be varied dramatically by changing the magnetic field. That is why the appearance of those zero-cross points and peaks cannot simply be assigned to particular interband transitions. Increasing the perpendicular magnetic field turns the sample's easy axis gradually from the in-plane direction to the vertical direction. The observed behaviors during this process must be related to the possible variation in the interband transition that can be caused by rotating the easy axis. Following the mean field theories for magnetic anisotropy in diluted magnetic semiconductors (DMS),[20,21] it is known that the single-particle energy $\varepsilon(\boldsymbol{k})$ for the hole in a spin coherent state, induced by its antiferromagnetic exchange interaction with localized spins, will warp differently by changing the direction angle of $(\hat{\boldsymbol{M}}, \hat{\boldsymbol{k}})$. In other words, the hole's effective mass becomes a spin-orientation-dependent one, as given by $m^* = m/\gamma(\hat{\boldsymbol{M}}, \hat{\boldsymbol{k}})$, and so does the density of states $\rho = m^* k_{exc}/\pi^2\hbar^2$ with k_{exc} determined by the photon excitation energy $\hbar\omega$. The Luttinger parameter $\gamma(\hat{\boldsymbol{M}}, \hat{\boldsymbol{k}})$, after being averaged over the reciprocal space direction $\hat{\boldsymbol{k}}$, changes with the cubic crystalline anisotropy. Both the diagonal and non-diagonal elements, ε_{xx} and ε_{xy}, of dielectric function[10] will be affected by rotating the magnetization vector $\hat{\boldsymbol{M}}$, because the interband transition is determined by the joint density of states of conduction and valence bands at the contour, defined by photon excitation energy $\hbar\omega$. Now, the instructive idea from the mean field theory helps us to understand the measured results in Fig. 3. First, the period of the oscillation, determined by the phase angle $\theta = 2\omega nd/c$, will vary by rotating the easy axis due to the change in $n = \sqrt{\varepsilon_{xx}}$. Second, the variations in ε'_{xy} (or $\Delta\alpha$), ε''_{xy} (or Δn), and ε_{xx} (or n^2), induced by the interband transitions when the easy axis is oriented differently, will unavoidably affect the wavelength dependence of R_{MCD}. This is exactly what is seen in Fig. 3. Figure 4 plots the magnetic field dependence of R_{MCD} at three different wavelengths (700 nm, 760 nm, and 880 nm). At first glance, the large fluctuation of R_{MCD} during the sweep of a magnetic field is somewhat strange. It is well known that the magnetic gyrotropy of $Ga_{1-x}Mn_xAs$ is completely negligible because the optical frequency is higher than the ferromagnetic resonance frequency (9–35 GHz) by more than four orders of magnitude.[22,23] As a result, instead, if we look at the variation in R_{MCD} at a particular wavelength in Fig. 3, we can trace the fluctuation in Fig. 4. Therefore, the huge fluctuation in Fig. 4 has the same origin as the oscillations seen in Fig. 3. Both of them stem from the influence of the magnetization direction $(\hat{\boldsymbol{M}}, \hat{\boldsymbol{k}})$ on the interband permittivity tensor.

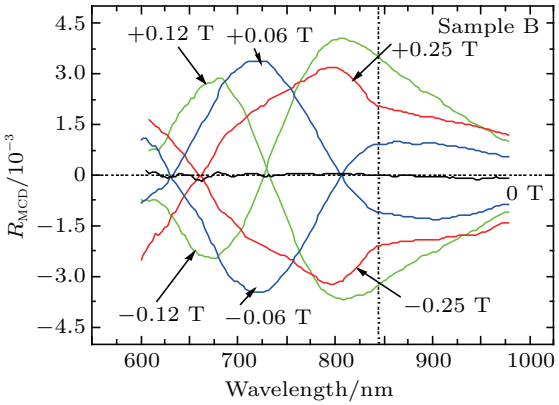


Fig. 3. (color online) Wavelength dependence of R_{MCD} measured at 4 K for Sample B under ±0.06, ±0.12, and ±0.25 Tesla.

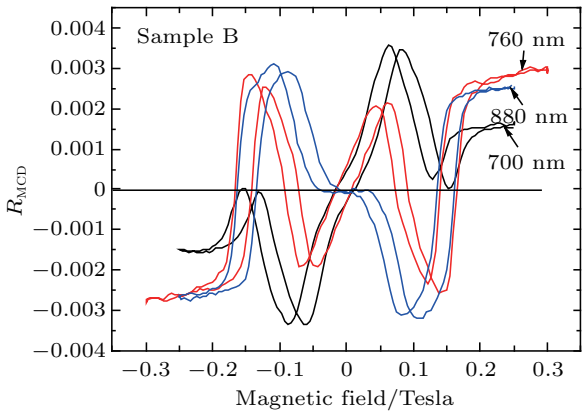


Fig. 4. (color online) R_{MCD} of Sample B measured at 4 K in a sweep of magnetic field between ±0.3 Tesla for three different wavelengths.

As predicted by Eqs. (5) and (6), if the layer thickness d is reduced, the β factor decreases in proportion, and so does the contribution from $\Delta\alpha$. At the same time, the wavelength separation between two adjacent minima becomes wider, as approximately given by $(\lambda_m - \lambda_{m+1})/\lambda_m\lambda_{m+1} = 1/(2nd)$. This prediction is verified by measuring the wavelength dependence of R_{MCD} for Sample C, as shown in Fig. 5. Now, the only one zero-cross point moves to $\lambda = 618$ nm. Beyond it

the sign of $R_{\rm MCD}$ seems still reversed, but its amplitudes are already very small. In contrast to Fig. 3, the shapes of the wavelength dependence of $R_{\rm MCD}$ at different magnetic fields are quite similar, and they differ only in their amplitudes, indicating $\Delta\alpha$-related oscillatory term diminished rather fast. Since the nominal composite ratio of Mn in Sample C is the same as Sample B, obviously, it is the reduced $Ga_{1-x}Mn_xAs$ layer thickness that gives rise to the significantly different behaviors between Figs. 3 and 5, as predicted by Eq. (5).

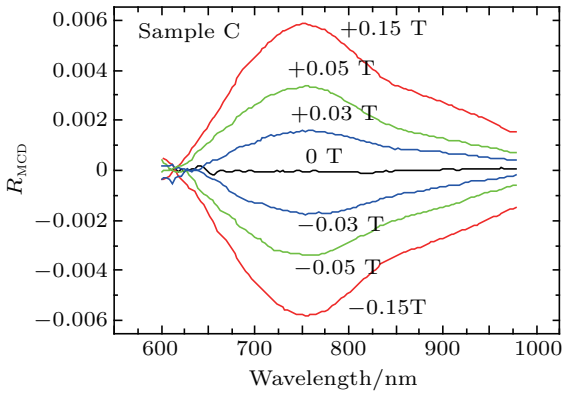


Fig. 5. (color online) Wavelength dependence of $R_{\rm MCD}$ measured at 4 K for Sample C under ±0.03, ±0.05, and ±0.15 Tesla.

5. Conclusion

An explicit expression of $R_{\rm MCD}$ has been derived when the interference effect, arising from multiple internal reflections, is considered in an air/$Ga_{1-x}Mn_xAs$/GaAs dielectric layered system. It unambiguously shows that the $R_{\rm MCD}$ signal is composed of three terms. In addition to the conventional term, which is sufficient in the absence of interference, an oscillatory term is required. Both of them are related to the imaginary part ε''_{xy} of the off-diagonal element of the dielectric tensor. One is also aware that in this case, $R_{\rm MCD}$ is not simply determined only by the imaginary part ε''_{xy} of the off-diagonal element of the dielectric tensor through Δn, as has been widely accepted. In fact, the real part ε'_{xy}, due to the presence of $\Delta\alpha$, will substantially mix into the measured $R_{\rm MCD}$ results in terms of an oscillatory $\cos\theta$ form when the layer thickness of $Ga_{1-x}Mn_xAs$ is larger than, say, 50 nm. Moreover, this $\Delta\alpha$-related term can reverse the sign of the whole $R_{\rm MCD}$. As the $Ga_{1-x}Mn_xAs$ layer becomes thinner than 50 nm, $R_{\rm MCD}$ resumes being dominated by ε''_{xy}. The wavelength separation between two adjacent minima becomes wider as well. The main aspects of these predictions were used to reasonably explain the $R_{\rm MCD}$ results measured from three different samples (A, B, C). Our work will bring about a reconsideration of how to correctly explain the measured $R_{\rm MCD}$ results.

Appendix A: Explicit expression of $R_{\rm MCD}$ when the interference effect in $Ga_{1-x}Mn_xAs$ is taken into account

Let us consider a special case, where the right (+)/left (−) circularly polarized light shines vertically on a dielectric layer structure, composed of air (0), a $Ga_{1-x}Mn_xAs$ layer (1) of thickness d, and a GaAs substrate (2). The GaAs substrate is considered to be infinitely thick with no light reflected back to the 1–2 interface. Using Fresnel's formulas for circular polarizations,[24] the reflection coefficients $r^{\pm}_{01}$ and transmission coefficients $t^{\pm}_{01}$ at the first air (0)/$Ga_{1-x}Mn_xAs$ (1) interface, and the reflection coefficients $r^{\pm}_{12}$ and transmission coefficients $t^{\pm}_{12}$ at the second $Ga_{1-x}Mn_xAs$ (1)/GaAs (2) interface can be directly written respectively as follows:

$$r^{\pm}_{01}=\frac{\tilde{n}^{\pm}(\mathrm{GaMnAs})-n(\mathrm{air})}{\tilde{n}^{\pm}(\mathrm{GaMnAs})+n(\mathrm{air})}=\frac{n^{\pm}+\mathrm{i}\kappa^{\pm}-1}{n^{\pm}+\mathrm{i}\kappa^{\pm}+1}, \tag{A1}$$

$$t^{\pm}_{01}=\frac{2n(\mathrm{air})}{n(\mathrm{air})+\tilde{n}^{\pm}(\mathrm{GaMnAs})}=\frac{2}{n^{\pm}+\mathrm{i}\kappa^{\pm}+1}, \tag{A2}$$

$$r^{\pm}_{12}=\frac{\tilde{n}_0(\mathrm{GaAs})-\tilde{n}^{\pm}(\mathrm{GaMnAs})}{\tilde{n}_0(\mathrm{GaAs})+\tilde{n}^{\pm}(\mathrm{GaMnAs})}=\frac{n_0-n^{\pm}+\mathrm{i}(\kappa_0-\kappa^{\pm})}{n_0+n^{\pm}+\mathrm{i}(\kappa_0+\kappa^{\pm})}, \tag{A3}$$

$$t^{\pm}_{12}=\frac{2\tilde{n}^{\pm}(\mathrm{GaMnAs})}{\tilde{n}_0(\mathrm{GaAs})+\tilde{n}^{\pm}(\mathrm{GaMnAs})}=\frac{2(n^{\pm}+\mathrm{i}\kappa^{\pm})}{n_0+n^{\pm}+\mathrm{i}(\kappa_0+\kappa^{\pm})}. \tag{A4}$$

For the light propagating along a reversed direction, the reflection and transmission coefficients, $r^{\pm}_{10}$ and $t^{\pm}_{10}$, at the first $Ga_{1-x}Mn_xAs$ (1)/ air (0) interface are given by

$$r^{\pm}_{10}=\frac{n(\mathrm{air})-\tilde{n}^{\pm}(\mathrm{GaMnAs})}{n(\mathrm{air})+\tilde{n}^{\pm}(\mathrm{GaMnAs})}=\frac{1-n^{\pm}-\mathrm{i}\kappa^{\pm}}{1+n^{\pm}+\mathrm{i}\kappa^{\pm}}=-r^{\pm}_{01}, \tag{A5}$$

$$t^{\pm}_{10}=\frac{2\tilde{n}^{\pm}(\mathrm{GaMnAs})}{n(\mathrm{air})+\tilde{n}^{\pm}(\mathrm{GaMnAs})}=\frac{2(n^{\pm}+\mathrm{i}\kappa^{\pm})}{n^{\pm}+\mathrm{i}\kappa^{\pm}+1}. \tag{A6}$$

Here $\tilde{n}=n+\mathrm{i}\kappa$ is the complex refractive index, having a real part of n and an imaginary part of κ (extinction coefficient). Their superscripts "±" stand for the values of right/left circularly polarized light. It is easy to strictly prove that after including the interference effect due to the multiple reflections inside the $Ga_{1-x}Mn_xAs$ layer, the summed reflection coefficient at the 0–1 interface is given by[8]

$$r^{\pm}(\omega)=\frac{r^{\pm}_{01}+r^{\pm}_{12}\exp(\mathrm{i}2\tilde{n}^{\pm}(\mathrm{GaMnAs})\omega d/c)}{1+r^{\pm}_{01}r^{\pm}_{12}\exp(\mathrm{i}2\tilde{n}^{\pm}(\mathrm{GaMnAs})\omega d/c)}. \tag{A7}$$

The exponential factor in Eq. (A7) is related to the absorption and phase factor of the light propagation inside the $Ga_{1-x}Mn_xAs$ layer through

$$a^{\pm}=|a^{\pm}|\exp(\mathrm{i}\theta^{\pm})=\exp(-\alpha^{\pm}d)\exp(\mathrm{i}n^{\pm}2\omega d/c), \tag{A8}$$

$$\alpha^{\pm}=2\omega\kappa^{\pm}/c, \tag{A9}$$

$$\theta^{\pm}=2\omega n^{\pm}d/c. \tag{A10}$$

The corresponding intensity reflection coefficients for $\sigma^{\pm}$ polarized light are defined as

$$R^{\pm} = r^{\pm}(\omega)^{*} \times r^{\pm}(\omega) = \frac{\left|r_{01}^{\pm}\right|^{2} + \left|r_{12}^{\pm} a\right|^{2} + 2\left|r_{01}^{\pm} r_{12}^{\pm} a^{\pm}\right| \cos(\theta_{12}^{\pm} + \theta^{\pm} - \theta_{01}^{\pm})}{1 + \left|r_{01}^{\pm} r_{12}^{\pm} a\right|^{2} + 2\left|r_{01}^{\pm} r_{12}^{\pm} a^{\pm}\right| \cos(\theta_{12}^{\pm} + \theta^{\pm} + \theta_{01}^{\pm})}, \tag{A11}$$

where $r_{01}^{\pm} = \left|r_{01}^{\pm}\right| \exp(\mathrm{i}\theta_{01}^{\pm})$ and $r_{12}^{\pm} = \left|r_{12}^{\pm}\right| \exp(\mathrm{i}\theta_{12}^{\pm})$. Under the usual condition of $n^{\pm} \gg \kappa^{\pm}$, $\theta_{01}^{\pm} \approx 0$, and $\theta_{12}^{\pm} \approx 0$.

Before we continue deriving for the reflection MCD, let us first estimate the relative magnitudes of $\left|r_{01}^{\pm}\right|$ and $\left|r_{12}^{\pm}\right|$. In general, the refractive indices of GaAs and $Ga_{1-x}Mn_xAs$ vary with the photon energy, carrier concentration, temperature, composition ratio of Mn, etc. For the frequency dependence of the GaAs refractive index, we adopt the experiential formula[15] $n_0^2 = 7.10 + 3.78/[1 - 0.18(\hbar\omega)^2]$ ($\hbar\omega$ in eV), and obtain $n_0 = 3.7$ at $\hbar\omega = 1.54$ eV (or $\lambda = 805$ nm); $n_0 = 3.97$ at $\hbar\omega = 1.77$ eV (or $\lambda = 700$ nm). Since the MCD spectrum near the band gap (~824 nm) is most interesting, we shall adopt $n = 3.7$ for GaAs. For the refractive index of $Ga_{1-x}Mn_xAs$, we take its value of $n = \sqrt{\varepsilon_{\infty}} \sim 3.44$ from Ref. [8], which is assumed to have a photon energy dependence similar to that of GaAs. Then, it is found that

$$|r_{01}| = \frac{n-1}{n+1} \approx 0.55, \quad |r_{01}|^2 = \left(\frac{n-1}{n+1}\right)^2 \approx 0.30,$$
$$|r_{12}| = \frac{n_0 - n}{n_0 + n} \approx 0.036, \quad |r_{12}|^2 = \left(\frac{n_0 - n}{n_0 + n}\right)^2 \approx 0.0013,$$

for the case of $n_0 > n$. As a result, we shall neglect the square term of $|r_{12}|$ in Eq. (A11)

$$R^{\pm} \approx \frac{\left|r_{01}^{\pm}\right|^{2} + 2\left|r_{01}^{\pm} r_{12}^{\pm} a^{\pm}\right| \cos(\theta^{\pm})}{1 + 2\left|r_{01}^{\pm} r_{12}^{\pm} a^{\pm}\right| \cos(\theta^{\pm})} = \frac{\left|r_{01}^{\pm}\right|^{2} + A^{\pm}}{1 + A^{\pm}} \approx \left|r_{01}^{\pm}\right|^{2} + A^{\pm}\left(1 - \left|r_{01}^{\pm}\right|^{2}\right). \tag{A12}$$

The last equation above is based on the condition of $A^{\pm} \ll 1$. Indeed, the maximum value of A at $n \neq n_0$ is about 0.04 (see Eq. (A17)).

Then, the R_{MCD} is determined by its definition as

$$\frac{R^{+} - R^{-}}{R^{+} + R^{-}} = \frac{\left|r_{01}^{+}\right|^{2} + A^{+}\left(1 - \left|r_{01}^{+}\right|^{2}\right) - \left|r_{01}^{-}\right|^{2} - A^{-}\left(1 - \left|r_{01}^{-}\right|^{2}\right)}{\left|r_{01}^{+}\right|^{2} + A^{+}\left(1 - \left|r_{01}^{+}\right|^{2}\right) + \left|r_{01}^{-}\right|^{2} + A^{-}\left(1 - \left|r_{01}^{-}\right|^{2}\right)}. \tag{A13}$$

As a conventional assumption, due to $n^{\pm} \gg \kappa^{\pm}$, all the $\kappa^{\pm}$ terms in the expressions (A1)–(A6) can be ignored. The refractive indices for right (+) and left (−) circularly polarized light are redefined to be $n^{\pm} = n \pm \Delta n$ with $n = (n^{+} + n^{-})/2$ and $\Delta n = (n^{+} - n^{-})/2$. The corresponding absorption coefficients become $\alpha^{\pm} = \alpha \pm \Delta\alpha$ with $\alpha = (\alpha^{+} + \alpha^{-})/2$ and $\Delta\alpha = (\alpha^{+} - \alpha^{-})/2$.

By using progression expansion and neglecting higher-order terms in Δn, $\Delta\alpha d$, $(n_0 - n)/(n_0 + n)$, and their products, in what follows, we simply write a series of the expressions for the case of $n_0 > n$ and $n_0 - n > \Delta n$ as

$$\left|r_{01}^{\pm}\right| = \frac{n - 1 \pm \Delta n}{n + 1 \pm \Delta n} \approx \frac{n-1}{n+1} \pm \frac{2\Delta n}{(n+1)^2}, \tag{A14}$$

$$\left|r_{01}^{\pm}\right|^2 = \left(\frac{n - 1 \pm \Delta n}{n + 1 \pm \Delta n}\right)^2 \approx \frac{(n^2-1)^2 \pm 4(n^2-1)\Delta n}{(n+1)^4}, \tag{A15}$$

$$\left|r_{12}^{\pm}\right| = \left|\frac{n_0 - n \mp \Delta n}{n_0 + n \pm \Delta n}\right| \approx \frac{n_0 - n}{n_0 + n} \mp \frac{2n_0\Delta n}{(n_0+n)^2}, \tag{A16}$$

$$A^{\pm} = 2\left|r_{01}^{\pm} r_{12}^{\pm} a^{\pm}\right| \cos\theta \approx 2\left(\frac{n-1}{n+1}\frac{n_0-n}{n_0+n} - \frac{n-1}{n+1}\frac{n_0-n}{n_0+n}\alpha^{\pm} d \pm \frac{n_0^2 - n^2 - n^2 n_0 + n_0}{(n_0+n)^2}\frac{2\Delta n}{(n+1)^2}\right)\cos\theta, \tag{A17}$$

$$\left|r_{01}^{+}\right|^2 - \left|r_{01}^{-}\right|^2 \approx \frac{8(n-1)\Delta n}{(n+1)^3}, \tag{A18}$$

$$A^{+} - A^{-} \approx 4\left(-\frac{n-1}{n+1}\frac{n_0-n}{n_0+n}\Delta\alpha d + \frac{n_0^2 - n^2 - n^2 n_0 + n_0}{(n_0+n)^2} \times \frac{2\Delta n}{(n+1)^2}\right)\cos\theta, \tag{A19}$$

$$-A^{+}\left|r_{01}^{+}\right|^2 \approx -2\left(\frac{n-1}{n+1}\right)^2\left(\frac{n-1}{n+1}\frac{n_0-n}{n_0+n} - \frac{n-1}{n+1}\frac{n_0-n}{n_0+n}\alpha^{+} d + \frac{3n_0^2 - 3n^2 - n^2 n_0 + n_0}{(n_0+n)^2(n+1)^2}2\Delta n\right)\cos\theta, \tag{A20}$$

$$+A^{-}\left|r_{01}^{-}\right|^2 \approx 2\left(\frac{n-1}{n+1}\right)^2\left(\frac{n-1}{n+1}\frac{n_0-n}{n_0+n} - \frac{n-1}{n+1}\frac{n_0-n}{n_0+n}\alpha^{-} d - \frac{3n_0^2 - 3n^2 - n^2 n_0 + n_0}{(n_0+n)^2(n+1)^2}2\Delta n\right)\cos\theta, \tag{A21}$$

$$R^{+} - R^{-} = \left|r_{01}^{+}\right|^2 + A^{+}\left(1 - \left|r_{01}^{+}\right|^2\right) - \left|r_{01}^{-}\right|^2 - A^{-}\left(1 - \left|r_{01}^{-}\right|^2\right) \approx \frac{8(n-1)\Delta n}{(n+1)^3} + 4\frac{4(n^2 - 4n + 1)(n^2 - n_0^2) - 8nn_0(n^2-1)}{(n_0+n)^2(n+1)^4} \times \Delta n\cos\theta - 16n\frac{n-1}{(n+1)^3}\frac{n_0-n}{n_0+n}\Delta\alpha d\cos\theta, \tag{A22}$$

$$\left|r_{01}^{+}\right|^2 + \left|r_{01}^{-}\right|^2 \approx 2\left(\frac{n-1}{n+1}\right)^2, \tag{A23}$$

$$A^+ + A^- \approx 4\left(\frac{n-1}{n+1}\frac{n_0-n}{n_0+n}(1-\alpha d)\right)\cos\theta, \quad \text{(A24)}$$

$$-A^+\left|r_{01}^+\right|^2 - A^-\left|r_{01}^-\right|^2 \approx -4\left(\left(\frac{n-1}{n+1}\right)^3\frac{n_0-n}{n_0+n}(1-\alpha d)\right)\cos\theta, \quad \text{(A25)}$$

$$\begin{aligned} &R^+ + R^- \\ &= \left|r_{01}^+\right|^2 + A^+\left(1-\left|r_{01}^+\right|^2\right) + \left|r_{01}^-\right|^2 \\ &\quad + A^-\left(1-\left|r_{01}^-\right|^2\right) \approx 2\left(\frac{n-1}{n+1}\right)^2 \\ &\quad + \frac{16n}{(n+1)^2}\left(\frac{n-1}{n+1}\frac{n_0-n}{n_0+n}(1-\alpha d)\right)\cos\theta. \end{aligned} \quad \text{(A26)}$$

Finally, an explicit expression of R_{MCD} is derived to be

$$\begin{aligned} &\frac{R^+ - R^-}{R^+ + R^-} \\ &\approx \frac{4\Delta n}{n^2-1} + \frac{8\left(n^2+1\right)\left(n^2-n_0^2\right) - 16nn_0\left(n^2-1\right)}{\left(n_0+n\right)^2\left(n^2-1\right)^2}\Delta n\cos\theta \\ &\quad - \frac{8n}{n^2-1}\frac{n_0-n}{n_0+n}\Delta\alpha d\cos\theta. \end{aligned} \quad \text{(A27)}$$

Because $8(n^2+1)(n^2-n_0^2) \approx -190$ and $16nn_0(n^2-1) \approx 2206$, evaluated at $n = 3.44$ and $n_0 = 3.7$, equation (A27) may be further simplified into a compact form as

$$\begin{aligned} R_{\mathrm{MCD}} &\approx \frac{4\Delta n}{n^2-1}\left[1 - \frac{4nn_0}{(n_0+n)^2}\cos\theta\right] \\ &\quad - \frac{8n}{n^2-1}\frac{n_0-n}{n_0+n}\Delta\alpha d\cos\theta \\ &\approx \frac{4\Delta n}{n^2-1}\left[1-(1+\beta)\cos\theta\right], \end{aligned} \quad \text{(A28)}$$

$$\beta = 2\frac{n_0-n}{n_0+n}\frac{\Delta\alpha d}{\Delta n/n}, \quad \text{(A29)}$$

where β is the ratio of the second oscillatory term to the first term in Eq. (A27).

In the case of $n > n_0$, it can be easily proved that

$$R_{\mathrm{MCD}} \approx \frac{4\Delta n}{n^2-1}\left[1+(1+\beta)\cos\theta\right]. \quad \text{(A30)}$$

For a particular case of $n_0 = n$, one can no longer obtain the result directly from Eq. (A28), because equation (A16) does not hold. Instead, one has to start with the following equations:

$$\left|r_{12}^{\pm}\right| = \left|\frac{n_0-n\mp\Delta n}{n_0+n\pm\Delta n}\right| \approx \frac{\Delta n}{n_0+n}, \quad \text{(A31)}$$

$$\begin{aligned} A^{\pm} &= 2\left|r_{01}^{\pm}r_{12}^{\pm}a^{\pm}\right|\cos\theta \\ &\approx 2\frac{(n-1)\Delta n}{(n+1)(n_0+n)}\mathrm{e}^{-\alpha d}\cos\theta \equiv A, \end{aligned} \quad \text{(A32)}$$

$$R^{\pm} \approx \frac{\left|r_{01}^{\pm}\right|^2 + A}{1+A} \approx (1-A)\left|r_{01}^{\pm}\right|^2 + A, \quad \text{(A33)}$$

$$\begin{aligned} \frac{R^+ - R^-}{R^+ + R^-} &\simeq \frac{(1-A)\left|r_{01}^+\right|^2 + A - (1-A)\left|r_{01}^-\right|^2 - A}{(1-A)\left|r_{01}^+\right|^2 + A + (1-A)\left|r_{01}^-\right|^2 + A} \\ &\approx \frac{\left|r_{01}^+\right|^2 - \left|r_{01}^-\right|^2}{\left|r_{01}^+\right|^2 + \left|r_{01}^-\right|^2}\left(1 - \frac{A}{(1-A)\left|r_{01}\right|^2}\right), \end{aligned} \quad \text{(A34)}$$

where $|r_{01}|^2 = \left(\left|r_{01}^+\right|^2 + \left|r_{01}^-\right|^2\right)/2 \approx (n-1)^2/(n+1)^2$.

From Eq. (A32), $A \approx 0.0044$, as $\Delta n \sim 0.03$ (see text) and $n = n_0$. The following term in Eq. (A34)

$$\frac{A}{(1-A)\left|r_{01}\right|^2} \approx \frac{2\Delta n(n+1)}{(n_0+n)(n-1)},$$

which is of the order of 0.013, can be neglected, so that R_{MCD} resumes its form in the absence of interference.

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