第30卷第1期
2011年2月

红外与毫米波学报
J. Infrared Millim. Waves

Vol. 30, No. 1
February, 2011

文章编号:1001 - 9014(2011)01 - 0001 - 05

Influence of magnetocrystalline anisotropy on magneto-optics in GaMnAs/InGaAs epilayer

ZHU Ke, ZHENG Hou-Zhi*, GAN Hua-Dong, LIU Jian, ZHU Hui,
ZHANG Hao, LI Gui-Rong, ZHAO Jian-Hua
(State Key Laboratory of Superlattice and Microstructure, Institute of Semiconductors of CAS, Beijing 100083, China)

Abstract: Abnormal behaviors were observed in magnetic circular dichroism (MCD) in both as-grown and annealed $Ga_{0.95}Mn_{0.05}As$/InGaAs epilayer, as the direction of magnetization vector turns away from or toward the direction of incident beam by applying a magnetic field. Following the mean-field theory of magnetocrystalline anisotropy, it is found that the phenomena are actually the manifestation of the influence of the magnetic anisotropy on interband optical transitions due to the dependence of hole band splitting and warp of E-k dispersion relation on the magnetization orientation.

Key words: (Ga,Mn)As; magneto-optical effect; oscillation; magnetic anisotropic
PACS: 75.50.Pp, 75.30.Gw, 78.20.Ls

磁各向异性对(In,Ga)As衬底(Ga,Mn)As的影响

朱 科, 郑厚植*, 甘华东, 刘 剑, 朱 汇, 章 昊, 李桂荣, 赵建华
(中国科学院半导体研究所 超晶格国家重点实验室,北京 100083)

摘要:在(In,Ga)As缓冲层中生长的$Ga_{0.95}Mn_{0.05}As$薄膜的磁光圆二向色性(MCD)扫描磁场的测量中发现异常现象,这一现象出现在外磁场把样品的磁化矢量扭转到与入射光方向一致或远离入射光方向之时.通过磁各向异性的平均场理论,我们认为这实际上是磁各向异性对带-带跃迁影响的表现,是由于空穴带劈裂和E-k色散关系均与磁化方向相关而引起的.

关 键 词:(Ga,Mn)As;磁光效应;振荡;磁各向异性
中国分类号:O472+.5;O482.55;O541.2 **文献标识码**:A

Introduction

Diluted magnetic semiconductors (DMSs) have been an intensive subject for exploring the prospect of utilizing both charge and spin within a same material in order to create new device functionalities. In this respect, magnetocrystalline anisotropy plays a decisive role in manipulating device's electronic, magnetic and optical properties[1]. Although magneto-optical spectroscopy has been proved to be an effective method for both probing the electronic structures and evaluating the s, p-d exchange interactions in DMSs[2], the influence of magnetocrystalline anisotropy on magneto-optical spectroscopy has not yet been examined in details.

1 Experiment Setup

In this work, by manipulating the direction of magnetization vector in as-grown and annealed $Ga_{0.95}Mn_{0.05}As$/InGaAs epilayer, we report the abnormal behaviors in magnetic circular dichroism (MCD). We provide the evidence that the observed phenomena are the manifestation of the influence from the magnetocrystalline

---

Received date: 2010 - 04 - 20, **revised date**: 2010 - 08 - 15 **收稿日期**:2010 - 04 - 20, **修回日期**:2010 - 08 - 15

Foudantion item: National Basic Research Programs of China (2006CB932801); State Key Project of Fundamental Research of China (2007CB924903); The Special Research Programs of Chinese Academy of Sciences (2007CB924904)

Biography: ZHU Ke (1983 -), Born in Leshan City of Sichuan Province, studied for PH. D in Institute of Semiconductors of CAS. His research field is spintronics and he's currently engaged in magneto-optical properties and device exploration of (Ga,Mn)As. E-mail: zhuke@semi.ac.cn.

* Corresponding Author: E-mail: hzzheng@red.semi.ac.cn.

anisotropy on interband magneto-optical transitions.

A 100nm-thick $Ga_{0.95}Mn_{0.05}As$ epilayer was grown by low temperature molecular beam epitaxy (LT-MBE) on an InGaAs buffer with a semi-insulating GaAs substrate. On-site RHEED and X-ray diffraction showed that the sample in study had excellent crystalline quality. The Curie temperature of the sample was 50K, tested by superconductivity quantum interference device (SQUID) measurement. The experimental setup for measuring reflectance MCD is schematically described in Fig. 1. The light source in use was provided by a super-continuous white light generator (Fianium SC-450). After passing through a chopper (at 475 Hz), the white light was dispersed by a monochrometer to have either a particular wavelength or scanned wavelength at its outlet. The linearly polarized light, coming out of a polarizer, was fed through a photo-elastic modulator (PEM), and converted to alternatively modulated clockwise-polarized (σ^+) and counterclockwise-polarized (σ^-) light at a frequency of 50kHz. Then, a polarization-insensitive beam splitter (NPBS) split it into two beams. One was sent into the first detector, the photo-electric signal of which is detected by the first lock-in amplifier at the chopper frequency in order to monitor the intensity variation of the incident light. The other shined on the sample surface, and the intensity of its reflected beam was measured by the second detector and lock-in amplifier at the modulation frequency of 50 kHz. The final result is the ratio of the output of the second lock-in amplifier to that of the first lock-in amplifier. The MCD data were expressed in degrees by multiplying it with a factor of $90°/\pi$, as adopted in literatures[3]. Therefore, it is the intensity difference of the reflected beams between clockwise (σ^+) and counterclockwise (σ^-) polarized lights that MCD measures. Our sample was mounted in a separated cryostat from a superconducting magnet. The former could be fitted into the clear bore of the latter, but its temperature could vary independently from the magnet in a range from 3K to 300K. Two configurations were used in experiments. In the $B_\perp$ Sample configuration, sample lay on the top mounting plate of the cryostat, and the incident light was vertical to the sample surface in line with the magnetic field, as seen in the main panel of Fig. 1. Its inset shows the $B_{//}$ Sample configuration, where the sample was mounted vertically to the cryostat's mounting plate. The incident and reflected lights were turned by 90° at a polarization-conserved triangle prism, and made themselves still vertically to the sample's plane.

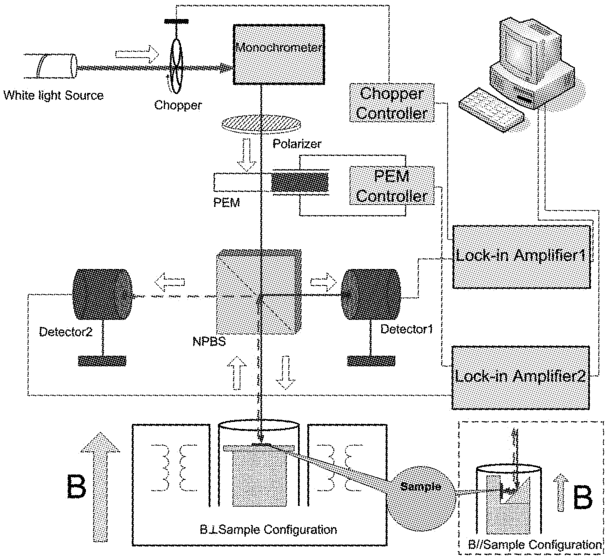

Fig. 1 Schematic experimental setup for measuring reflectance magnetic circular dichroism (MCD). Sample was mounted in $B_\perp$ Sample configuration in the main panel, and in $B_{//}$ Sample configuration in the inset

图1 反射式磁光圆二向色性(MCD)实验配置原理图,其中主图为磁场垂直于样品平面的配置,右下方小图为磁场平行于样品平面的配置

2 Experiment Results and Discussion

Fig. 2 shows the reflectance MCD spectra of as-grown $Ga_{0.95}Mn_{0.05}As/InGaAs$ sample, measured within a wavelength range from 530nm to 950nm at 3.3K for positive and negative saturated magnetic fields of 0.3 Tesla. The MCD data points at the wavelength shorter than 550nm are not reliable, since the intensity of the light source was too weak. The MCD spectra were very symmetric about the zero base line measured under the positive and negative saturated magnetic field, applied perpendicularly to sample surface. No spurious signal exists in the measurements. A noticeable feature is that a zero-cross point appeared at a wavelength of about 765nm, which will be discussed later. MCD signals of as-grown sample were also measured (shown in Fig. 3) by scanning magnetic field from −0.3T to +0.3T and vice versa for different

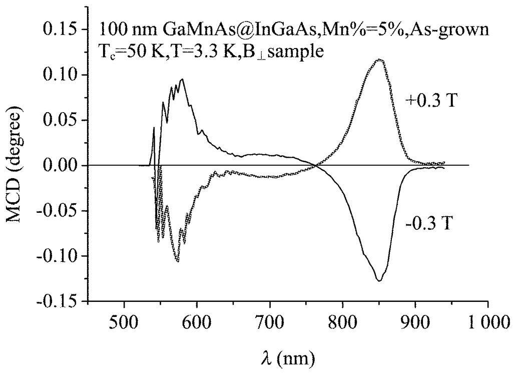

Fig. 2 MCD spectra in a wavelength sweep measured at 3.3K under ± 0.3T perpendicular magnetic fields for as-grown $Ga_{0.95}Mn_{0.05}As$ /InGaAs epilayer

图2 InGaAs 缓冲层上生长的原生 $Ga_{0.95}Mn_{0.05}As$ 薄膜在 T = 3.3K, B = ± 0.3T 且垂直于样品平面的配置下, MCD 扫描波长的光谱图

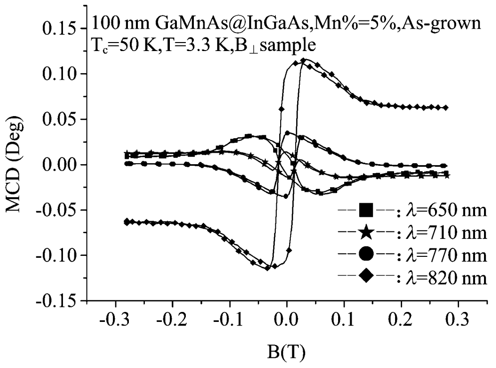

Fig. 3 MCD vs. B loops measured at 3.3K for wavelengths λ = 820, 770, 710 and 650nm of as-grown $Ga_{0.95}Mn_{0.05}As$ /InGaAs epilayer

图3 InGaAs 缓冲层上生长的原生 $Ga_{0.95}Mn_{0.05}As$ 薄膜在 T = 3.3K, 磁场垂直于样品平面, 且探测光波长分别为 820nm, 770nm, 710nm, 650nm 的配置下, MCD 磁滞回线图

wavelengths, λ = 820, 770, 710 and 650nm. The polarity of saturated MCD was reversed after the wavelength passes 765nm. All of the MCD vs. B loops showed hysteresis-type characteristics, but did not look like the standard rectangular hysteresis loops. Such abnormal change was more pronounced on the long wavelength side. The other feature is that MCD signal was almost depressed near λ = 765nm. Across this wavelength the sign of MCD is reversed.

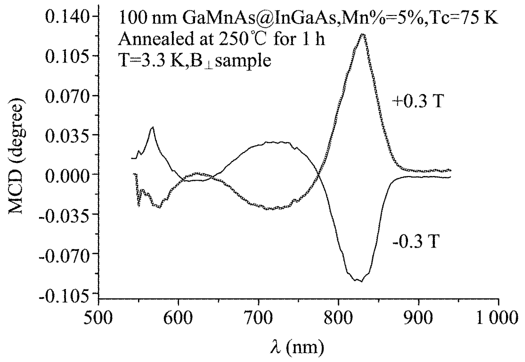

Fig. 4 MCD spectra in a wavelength sweep measured at 3.3K under ± 0.3T perpendicular magnetic fields for annealed $Ga_{0.95}Mn_{0.05}As$ /InGaAs epilayer

图4 InGaAs 缓冲层上生长的 $Ga_{0.95}Mn_{0.05}As$ 薄膜经过退火后在 T = 3.3K, B = ± 0.3T 且垂直于样品平面的配置下, MCD 扫描波长的光谱图

Next, we examined how these magneto-optical spectroscopic features changed after $Ga_{0.95}Mn_{0.05}As$/InGaAs sample was annealed at 250℃ for one hour. The overall aspect of the reflectance MCD spectra (MCD vs. λ) in Fig. 4, measured in appealed $Ga_{0.95}Mn_{0.05}As$ /InGaAs sample is similar to that in Fig. 2. However, by a close look, it was noticed that the first zero-crossing point on the long wavelength side was shifted from 765nm to 775nm. Fig. 5 gives the hysteresis loops of MCD in magnetic sweeps, measured at 3.3K for wavelengths of 800nm and 660nm. Also, MCD reverses its sign as the wavelength varies from 800nm to 660nm. The novel feature is that both of them displayed rectangular hysteresis loops in sharp contrast to that seen in Fig. 4.

It is well established that (Ga, Mn) As epilayers grown with tensile strain on (In, Ga) As buffers are in favor of their easy axes perpendicularly to sample surfaces at a temperature higher than half Curie temperature[4,5]. In the temperature range below the half Curie temperature, the magnetization orientates away from the sample's normal since the in-plane cubic magnetic anisotropy plays a role as well. The recovery of the standard rectangular hysteresis loops in Fig. 4 indicated, on one hand, that the easy axis of the annealed sample lies in the perpendicular direction even at a low temperature like 3.3K. Since no magnetization rotation is expected when the magnetic vector is in line with the magnetic field, thus, there is no abnormal variation seen in MCD. On the other hand, the magnetization vector in our as-grown sample must not point exactly in the sample's normal at 3.3K. Applying a perpendicular magnetic field is going to bring its magnetization

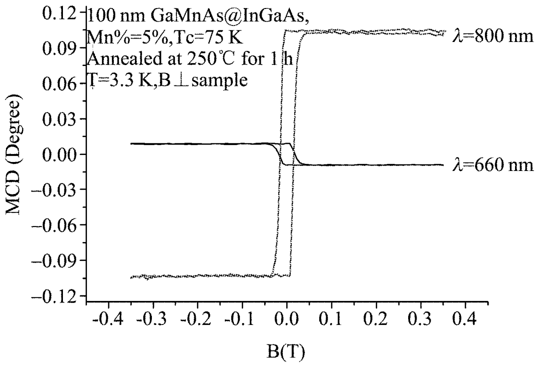

Fig. 5 MCD vs. B loops measured at 3.3K for wavelengths λ = 800, 660 nm in annealed $Ga_{0.95}Mn_{0.05}As$ /InGaAs epilayer, with B field perpendicular to sample

图5 InGaAs 缓冲层上生长的 $Ga_{0.95}Mn_{0.05}As$ 薄膜经过退火后在 T = 3.3K,磁场垂直于样品平面,且探测光波长分别为 800nm,660nm 的配置下,MCD 磁滞回线图

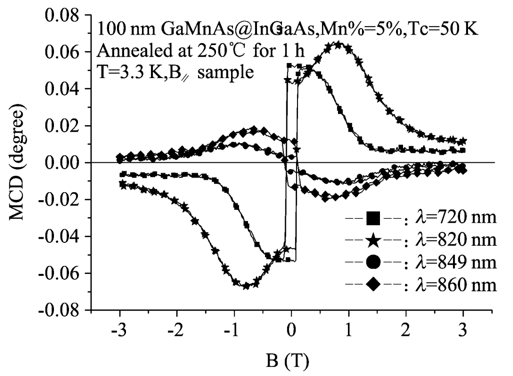

Fig. 6 MCD vs. B loops measured at 3.3K for wavelengths λ = 860, 849, 820 and 720nm in annealed $Ga_{0.95}Mn_{0.05}As$ /InGaAs epilayer with B field parallel to sample

图6 InGaAs 缓冲层上生长的 $Ga_{0.95}Mn_{0.05}As$ 薄膜经过退火后在 T = 3.3K,磁场平行于样品平面,且探测光波长分别为 860nm,849nm,820nm 和 720nm 的配置下,MCD 磁滞回线图

vector back to the normal. The abnormal MCD behaviors observed in as-grown sample (see Fig. 3) are naturally related to the rotation of its magnetization vector driven by the applied perpendicular magnetic field.

Next, by mounting the sample vertically to the tope plate of crystat's cooled head ($B_{//}$ Sample configuration as shown in the inset to Fig. 1), we made the magnetic field lie in the sample plane. Meanwhile, the incident and reflected lights were turned by 90° at a polarization conserved triangle prism, and make themselves still vertically to the sample's plane. In such measurement configuration we were able to rotate the magnetization vector of the annealed $Ga_{0.95}Mn_{0.05}As$/InGaAs sample from the normal to in-plane direction by applying in-plane magnetic field. Fig. 6 shows the results of MCD vs. B loops under four different wavelengths. First, one finds that the rectangular hysteresis loops existed still in a field range about 0.1 Tesla, which may indicate the stiffness of the initial domain with its magnetization pointing vertically. With increasing the in-plane field, the previously observed abnormal variation in MCD came back again as compared to Fig. 4. It appeared first as an enhanced bump at most of wavelengths, and then eventually decays to zero. The MCD vs. B loops reversed its polarity after passing λ = 775nm, as it did in Fig. 4. Combining the results in Fig. 3, 5 and 6, it is concluded that the magnetization vector and its rotation have significant influences on the magneto-optical spectra of both as-grown and annealed GaMnAs/InGaAs sample.

At this point, we turn to discuss the physical mechanism responsible for the above observations. Recalling the mean field theory for magnetocrystalline anisotropy in DMS[6,7], one knows in general that the giant Zeeman splitting, induced by p-d exchange interaction in valence bands, appears only in the direction of the magnetization vector. Since MCD signal is detected as the intensity difference of the reflected lights from (Ga, Mn) As surface between clockwise (σ^+) and counterclockwise (σ^-) polarized lights. That can be directly written as: $MCD = (r_+^2 - r_-^2)/(r_+^2 + r_-^2)$.

With r_+^2 and r_-^2 being the reflectivities for clockwise (σ^+) and counterclockwise (σ^-) polarized lights respectively, a rotation of the magnetization with respect to the optical axis is unavoidable to dramatically change the hole band structure along the optical quantizing axis. In other word, the p-d exchange interaction makes hole bands in (Ga, Mn) As warp dramatically, as seen from the spin-orientation dependent mass, expressed by $m^* = m/\bar{\gamma}(\hat{M})$ [7]. Here, $m^*(M)$ is effective mass depending on the magnetization direction, and $\bar{\gamma}(M)$ is evaluated as a function of $\hat{M}$ in a cubic harmonic expansion, which was used commonly for explaining magnetocrystalline anisotropy in the literature [7]:

$$\bar{\gamma}(\hat{M}) = \bar{\gamma}(<100>) + \gamma_1^{ca}(\hat{M}_x^2\hat{M}_y^2 + \hat{M}_y^2\hat{M}_z^2 + \hat{M}_x^2\hat{M}_z^2) + \gamma_2^{ca}\hat{M}_x^2\hat{M}_y^2\hat{M}_z^2 \quad .$$

Very clearly from these expressions, as long as the magnetization vector turns, it brings about a drastic change in the hole bands, especially the warp of $E(\hat{k})$ dispersion in addition to the hole band splitting. It is well established that Fermi contours, obtained by intersecting six hole bands with $k_z = 0$ plane, play an important role when the magnetocrystalline anisotropy is concerned in transport measurements. Both the perimeter size and shape of Fermi contours in (k_x, k_y) plane are sensitively affected by the in-plane magnetization orientation, leading to abnormal magneto-transport phenomena[8]. In a similar way, we may define an excitation contour for an inter-band transition. We may first plot out the dispersion relation for each of interband transitions from the different spin-polarized hole bands: ±3/2 heavy holes, ±1/2 light holes and ±1/2 split-off holes, to ±1/2 conduction bands, which are allowed by energy and momentum conservation rules. Then the intersection of a constant photon energy plane with them gives the excitation contours, which counts the all contributions to the interband transition at a particular wavelength. Due to the same physical origin (change in the hole band splitting and warping of $E(\hat{k})$ induced by rotating the magnetization), both the perimeter size and shape of the excitation contours in (k_x, k_y) plane change as the magnetization vector turns with respect to the optical quantization axis. In this respect, the magnetocrystalline anisotropy of DMS should also find their footprint in magneto-optical spectra, as we reported here. The zero-crossing point in a wavelength sweep of MCD (see Fig. 2 and 7) should correspond to the wavelength at which the interband transitions, probed by this particular photon energy, gives equally summed oscillatory strength for clockwise (σ^+) and counterclockwise (σ^-) polarized excitations and a zero MCD signal. It has nothing to do with the sign change in p-d exchange interaction.

3 Summary and Forecast

In summary, we have studied in detail how the magneto-optical properties of both as-grown and annealed $Ga_{0.95}Mn_{0.05}As$/InGaAs epilayer vary as the direction of magnetization vector turns with respect to the optical axis by applying a magnetic field or changing the temperature. The observations are actually the manifestation of the influence of the magnetocrystalline anisotropy on the interband optical transitions in similar to that previously observed in abnormal magneto-transport induced by in-plane magnetic anisotropy.

4 Acknowledgement

This work was in part supported by National Basic Research Program of China under the contract No. 2006CB932801; No 2007CB924903 and No 2007CB924904; by Special Research Programs of Chinese Academy of Sciences, the Knowledge Innovation Program Project of Chinese Academy of Sciences under the contract No. KJCX. YW. W09; also by National Natural Science Foundation of China under the contract No. 60836002, 10674130 and 60521001.

REFERENCES

[1] Jungwirth T, Sinova J, Ma ek J, *et al.* Theory of ferromagnetic (III, Mn) V semiconductors [J]. *Review of Modern Physics*, 2006, **78**: 809.

[2] Ando K. Seeking room-temperature ferromagnetic semiconductors [J]. *Science*, 2006, **312**: 1883.

[3] Sugano S, Kojima N. *Magneto-Optics* [M]. Springer-Verlag: Berlin, 2000, 137—177.

[4] Thevenard L, Largeau L. Exchange constant and domain wall width in (Ga, Mn) (As, P) films with self-organization of magnetic domains [J]. *Phys. Rev. B*, 2006, **73**: 195331.

[5] Liu X, Sasaki Y, Furdyna J K. Ferromagnetic resonance in $Ga_{1-x}Mn_xAs$: Effects of magnetic anisotropy [J]. *Phys. Rev. B*, 2003, **67**: 205—206.

[6] Dietl H, Ohno F, Matsukura J, *et al.* Model description of ferromagnetism in zinc-blende magnetic semiconductors [J]. *Science*, 2000, **287**: 1019.

[7] Abolfath M, Jungwirth T, Brum J, *et al.* Theory of magnetic anisotropy in (III, Mn) V ferromagnets [J]. *Phys. Rev. B*, 2001, **63**: 054418.

[8] Matsukura F, Sawicki M, Dietl T, *et al.* Tunneling magnetoresistance in (Ga, Mn) As-based heterostructures with a GaAs barrier [J]. *Physica E (Amsterdam)*, 2004, **21**: 1032.