Molecular Beam Epitaxy Growth and Magneto Optical Study of Cd1-x Mnx Te Quantum Wells(Cd1-x Mnx Te量子井戸の分子線エピタキシー成長と磁気光学的性質の研究)

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号	2484
発行年	1999
URL	http://hdl.handle.net/10097/7757

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授　与　学　位	博士（工学）
学位授与年月日	平成 12 年 3 月 23 日
学位授与の根拠法規	学位規則第 4 条第 1 項
研究科，専攻の名称	東北大学大学院工学研究科（博士課程）応用物理学専攻
学位論文題目	Molecular Beam Epitaxy Growth and Magneto-Optical Study of $Cd_{1-x}Mn_xTe$ Quantum Wells
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論文内容要旨

Chapter 1: Introduction

Diluted magnetic semiconductors (DMSs) have attracted considerable research interest in the last twenty years. The band gap of these DMSs covers the energy region from near infrared to ultraviolet. Among the II-VI DMSs, $Cd_{1-x}Mn_xTe$ has been investigated with continuous interest, which has been further stimulated by the successful preparation of quantum well (QW) structures by molecular beam epitaxy (MBE) method. The physical properties of $Cd_{1-x}Mn_xTe$ are based on the strong s, p-d exchange interaction between electrons or holes with Mn^{2+} 3d-electrons. The magnetic interaction in this material leads to a giant increase of magneto-optical effects such as exciton Zeeman splittings, Faraday rotation and exciton magnetic polaron (EMP) where exciton is dressed by the local magnetization of magnetic ions within the exciton Bohr radius. A detailed study of the EMP has been discussed in the $Cd_{1-x}Mn_xTe$ epilayers and the QWs. Besides this, the spin relaxation process of excitons in DMSs is still less concerned. Experimentally, the DMSs exhibit a marked long luminescence lifetime of excitons compared to the non-magnetic ($x = 0$) materials. This unusual long lifetime has been explained if the excitons on the ground state correspond to that of the optically forbidden transition (the dark exciton, $J = 2$) and are spin-flipped to the optically active state (the bright exciton, $J = 1$) which is located energetically higher than the dark exciton state due to the electron-hole exchange interaction. Detailed study of the decay properties of the excitons is made in the external magnetic fields and at various temperatures.

Chapter 2: MBE growth of DMS epilayers and QWs

This chapter describes the growth of $Cd_{1-x}Mn_xTe$, $Cd_{1-y}Mg_yTe$ epilayers and QWs by the MBE method. The growth procedures are monitored by the reflection high energy electron diffraction (RHEED) system. QW structures are systematically fabricated in which Mn^{2+} ions are introduced either in the well or in the barrier layer. Single QWs (SQWs), asymmetric double QWs (ADQWs) and multiple QWs (MQWs) are successfully grown for the system of $Cd_{1-x}Mn_xTe/Cd_{1-y}Mg_yTe$ (type (a)) and $CdTe/Cd_{1-x}Mn_xTe$ (type (b)) with the well width (L_w) = 10 ~ 300Å and the barrier width (L_b) = 50 ~ 400Å. The contents of Mn and Mg vary in the range of 15 ~ 30% in the well and the barrier layers. The concentration x, L_w and L_b are determined by the X-ray diffraction measurements. The surface roughness of the samples is 6 nm, which is measured by the atomic force microscopy (AFM).

Chapter 3: Experimental in photoluminescence spectroscopy

Time-resolved photoluminescence (PL) was measured by a mode-locked titanium sapphire laser (repetition rate 76 MHz and pulse width 120 fs). The samples were mounted in a superconducting magnet or a cryogenic refrigeration system with a temperature controller. Magnetic field up to 7 T was applied parallel to the growth axis of the sample (Faraday geometry). Sample temperature was varied from 4.2 to 300 K. A streak camera with 5 ps time resolution took the transient spectra.

Chapter 4: Energy levels of electrons in QWs and exchange interaction in DMSs

This chapter describes the theoretical description of the energy levels of electrons in QWs and the exchange interaction in DMSs. The energy levels of the square well are calculated by the one-dimensional Kronig-Penney model of the crystal. The resulting Schrödinger equation has been solved analytically for a single square well potential. This is solved by expanding the particle wavefunction in terms of a set of basis function and then applying the relevant boundary conditions which include the continuity of the wavefunction and its derivative at each interface. The exchange interaction between the s and p band electrons of the DMS and the 3d electrons of the Mn^{2+} ions have been described by the Heisenberg exchange interaction. The s, p-d exchange interaction affects optical and transport properties of DMSs such as the interband and intraband magneto-optical effects, Faraday rotations, the magnetic polaron effects, and the giant negative magnetoresistance.

Chapter 5: Magneto-optical study of $Cd_{1-x}Mn_xTe$ and $Cd_{1-y}Mg_yTe$ epilayers

Magneto-optical study of the $Cd_{1-x}Mn_xTe$ and $Cd_{1-y}Mg_yTe$ epilayers has been described in this chapter. Magnetic field induces a large Zeeman shift (~30 meV) of the exciton PL at 7 T in the $Cd_{0.79}Mn_{0.21}Te$ epilayer, which is due to the spin splitting of the exciton states by the s, p-d exchange interaction. On the other hand, the exciton peak energy in the $Cd_{0.77}Mg_{0.23}Te$ epilayer only shifts by 0.2 meV for the variation of the external field.

The dynamics of the EMP of the $Cd_{1-x}Mn_xTe$ epilayers with x = 0.11 and 0.21 have been studied. The EMP binding energy is deduced from the value at 0 T as 6.4 and 8.22 meV for x = 0.11 and x = 0.21, respectively. The increase of the EMP binding energy with increasing x is affected by the increased number of Mn^{2+} ions which are participating to form the EMP states by the exchange interaction. The magnetic field of 2.5 T and 4.5 T completely suppresses the EMP formation for x = 0.11 and x = 0.21, respectively. The EMP suppression behavior is governed by the spin alignment in the system of Mn^{2+} ions. When the Mn concentration increases, a larger magnetic field is needed to cause a Mn^{2+} spin alignment in the crystal, which reduces the portion of the spin alignment by the polaron effect. The EMP formation time at 0 T decreases from 110 to 80 ps with increasing the Mn content from x = 0.11 to 0.21. This result indicates the stronger localization condition for Mn contents due to the stronger magnetic and non-magnetic fluctuations.

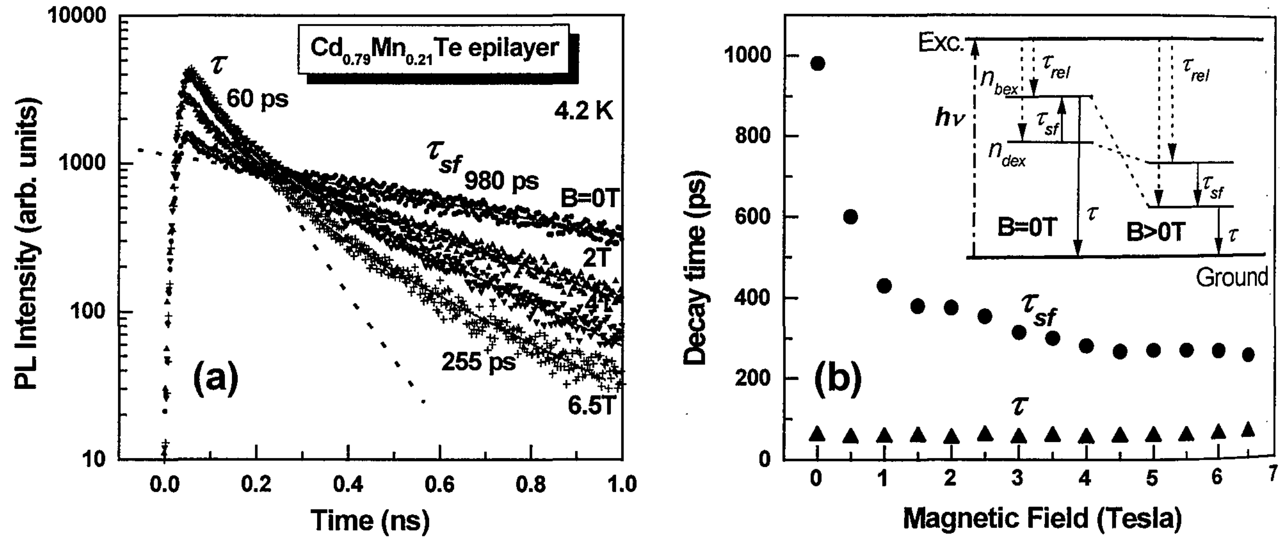

Fig. 1 (a) Transient PL intensity in the $Cd_{0.79}Mn_{0.21}Te$ epilayer for B = 0 ~ 6.5 T. (b) The spin-flip time of the dark exciton (τ_{sf}) and the lifetime of the bright exciton (τ) as a function of the magnetic field. The dynamics of the photoexcited bright and dark excitons with and without magnetic fields are shown in the inset.

The decay properties of the excitons have been studied in the $Cd_{1-x}Mn_xTe$ epilayers. Fig.1(a) shows the magnetic field dependence of the PL decay curves of the $Cd_{0.79}Mn_{0.21}Te$ epilayer. The decay of the exciton is found considerably long at zero magnetic field and is reduced drastically in the external field. The decay curves are well fitted by a double-exponential decay function at 0 and 6.5 T as plotted by the solid lines. From the fitting, the fast decay time τ is found to remain unchanged (~ 60 ps) with and without magnetic field, whereas the slow decay time τ_{sf} is decreased from 980 to 225 ps by the magnetic field. The short lifetime τ, which is nearly field independent, is assigned to the lifetime of the bright exciton while the longer one τ_{sf}, which decreases drastically in magnetic fields, is assigned to the spin-flip time of the dark exciton, as shown in the Fig.1(b). This result is understood in the model taken into account by the spin-flip process of the dark excitons to the bright excitons with and without magnetic fields, as shown in the inset of the Fig.1(b). After the laser excitation, both the bright and the dark exciton states are populated within few hundred femto seconds. The energy relaxation of the photoexcited carriers contributes to the rise time of the PL intensities. In zero magnetic field, excitons are populated at the dark exciton state which is energetically lower than the bright exciton state due to the electron-hole exchange interaction. If a spin-flip process does not exist, then the dark excitons will live very long until they are quenched by the non-radiative decay. However, these dark excitons are converted to the bright excitons by the spin scattering of the electrons, or of the holes with τ_{sf}. The converted excitons decay with the lifetime τ. The population in the dark exciton state prolongs the decay time of the excitonic emission effectively. In magnetic field, however, due to the giant Zeeman splitting, the bright exciton lowers the energy below that of the dark exciton. Then the initial occupation of the dark exciton state n_{dex} is drastically reduced and the dark excitons easily flip to the lower energetic bright excitons. Therefore, the τ_{sf} is also reduced in magnetic fields. Both the reduced population and the increased spin-flip rate decrease the effective PL decay time.

Chapter 6: Magneto-optical study in the CdTe/$Cd_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe/Cd_{1-y}Mg_yTe$ SQWs

L_w dependence of the EMP binding energy and the decay properties of exciton of the two types of SQWs at external magnetic field have been discussed in this chapter. Fig. 2 shows the L_w dependence of the EMP binding energy of the type (a) and the type (b) SQWs. In the type (a) SQWs, the EMP binding energy gradually increases with increasing L_w and remains almost unchanged ($\approx$ 6 meV) for $L_w \geq$ 100Å, whereas in the type (b) SQWs, the EMP binding energy is 6 meV for a thin L_w of 10Å, which monotonically decreases for thicker L_w and vanishes in L_w of 30Å. In the type (a) SQWs, only the part of the exciton wave function situated in the $Cd_{1-x}Mn_xTe$ well contributes to the EMP formation. For a thin well of the type (a) SQWs, the most part of the exciton wave function penetrates into the non-magnetic $Cd_{1-y}Mg_yTe$ barrier layer which causes of the EMP reduction. When L_w increases, the exciton wave function confined in the wide well and penetration of the exciton wave function into the non-magnetic barrier is obviously small. Therefore, the EMP binding energy is increased and becomes constant. On the other hand, in the type (b) SQWs, EMP are formed from exciton confined in the non-magnetic CdTe well layer due to the penetration of the exciton wave function into the $Cd_{1-x}Mn_xTe$ barrier layer. For a thin well of the type (b) SQWs, the exciton wave function can easily penetrate into the $Cd_{1-x}Mn_xTe$ barrier layer where a large number of Mn^{2+} spins can participate for the exchange interaction. Therefore, the EMP binding energy is large in thin QWs. In thicker QWs, the penetration of the exciton wave function is less into the $Cd_{1-x}Mn_xTe$ barrier and smaller number of Mn^{2+} spins are included to form the EMP. As a result, the EMP effect decreases and finally disappears for thick QWs.

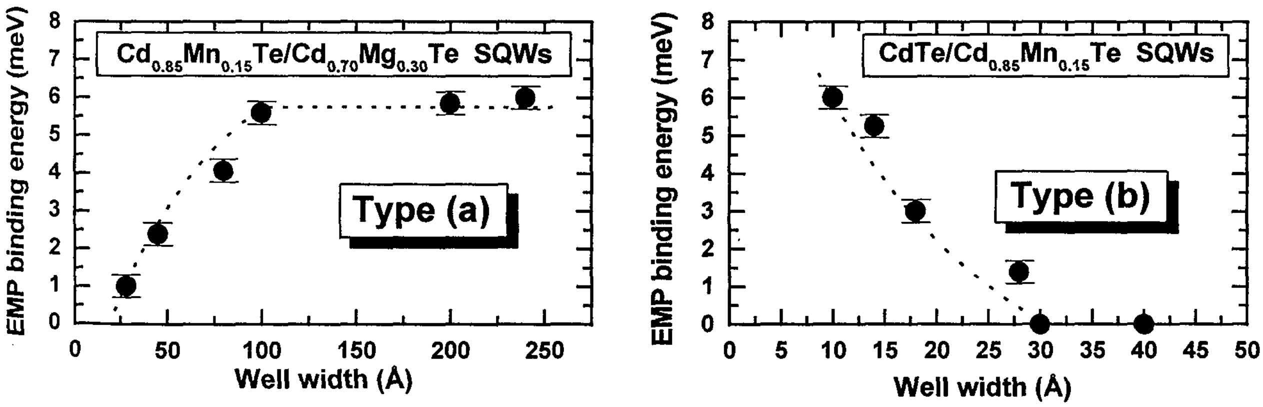

Fig. 2 The L_w dependence of EMP binding energy in the type (a) and the type (b) SQWs.

The reduction of the τ_{sf} has been observed in the type (a) SQWs in magnetic fields. The variation of τ_{sf} in external field have been seen much slower in the SQW with L_w = 28Å compared to the SQW with L_w = 200Å according to the amount of their Zeeman splittings. The giant Zeeman splitting lowers the energy position of the bright exciton in high magnetic field than that of the dark exciton. Therefore, rapid spin-flipping occurs in the high magnetic field.

Chapter 7: Magneto-optical study in $Cd_{1-x}Mn_xTe/Cd_{1-y}Mg_yTe$ ADQWs

ADQW structures are usually used to study the exciton transfer and the carrier tunneling in semiconductor heterostructures. $Cd_{0.85}Mn_{0.15}Te/Cd_{0.70}Mg_{0.30}Te$ ADQWs of wide well (WW) with a thickness L_{ww} = 200Å and a narrow well (NW) with a thickness L_{nw} = 40Å which are separated by an inner barrier width with thickness L_b = 50 ~ 400Å, have been used to investigate the tunneling mechanism. The exciton decays of the WW and NW of ADQW with L_b = 250Å show longer decay time (≈ 1000 ps) while only the WW of L_b = 100Å shows this trend at B = 0 T. The long decay component has been explained by the spin-flip time of the dark exciton in the SQWs. The absence of the long decay time in NW of L_b = 100Å thus demonstrate the possibility of the tunneling of the dark and the bright excitons from NW to WW. To clarify this result, a model is developed by using the rate equations for the tunneling of the dark and bright excitons and then the calculation is fitted with the experimental results. The calculated results show that the tunneling time of the dark and bright excitons is more than 1000 ps in 0 and 7 T for the ADQW with L_b = 250Å. This means that the tunneling effect in this sample is negligible. On the other hand, in the ADQW with L_b = 100Å, the tunneling time of the bright and dark excitons has been determined as 75 ps and 350 ps at 0 T, respectively, while these tunneling times are 75 ps and 10 ps at 7 T.

Chapter 8: Exciton dynamics in $Cd_{1-x}Mn_xTe/Cd_{1-y}Mg_yTe$ MQWs

MQW structures have been investigated as the sources of intense luminescence in the excitonic region. A set of $Cd_{0.85}Mn_{0.15}Te/Cd_{0.75}Mg_{0.25}Te$ MQWs of 50 periods with L_w = 10 ~ 200Å and L_b = 100Å have been used to investigate the temperature dependence of the exciton relaxations. By increasing temperature, the depopulation of the dark exciton states and the increase of the bright exciton population are expected. These populations are controlled by the thermal activation energy. Such behavior of the exciton decay has been observed with a slow decay component of τ_{sf}, which varies from 1050 ps (4.2 K) to 60 ps (150 K), while the variation of the fast decay component τ is less obvious (≈100 ps). The spin-flip process from the dark to bright excitons by the thermal activation effect could explain this behavior. At higher temperatures, the spin-flip process from the dark to bright excitons becomes faster. Fig. 3(a) shows the temperature dependence of the τ_{sf} and τ in the MQWs with L_w = 150Å and L_b = 100Å. The τ_{sf} below 100 K can be fitted by the dependence of $1/\tau_{sf} \propto T^{1/2}$ as shown by the solid line in the Fig. 3(a). This result is well interpreted in theory by Bir, Aronov and Pikus (BAP process), where the spin relaxation due to the exchange interaction between electrons and holes takes place over the thermalization process. The spin-flip process below 100 K is proved as the thermal activation effect by evaluating the thermal activation energy (E_A = 3.6 meV) as shown by the solid line to fit the experimental data in Fig. 3(b). Fig. 3(b) also shows that the PL intensity drops drastically above 100 K which is consistent with the decay time at temperatures above 100 K as shown by a dependence of $1/\tau_{sf} \propto T^{5/2}$ in Fig. 3(a). The result indicates that the non-radiative recombination process in the MQWs dominates the exciton recombination above 100 K.

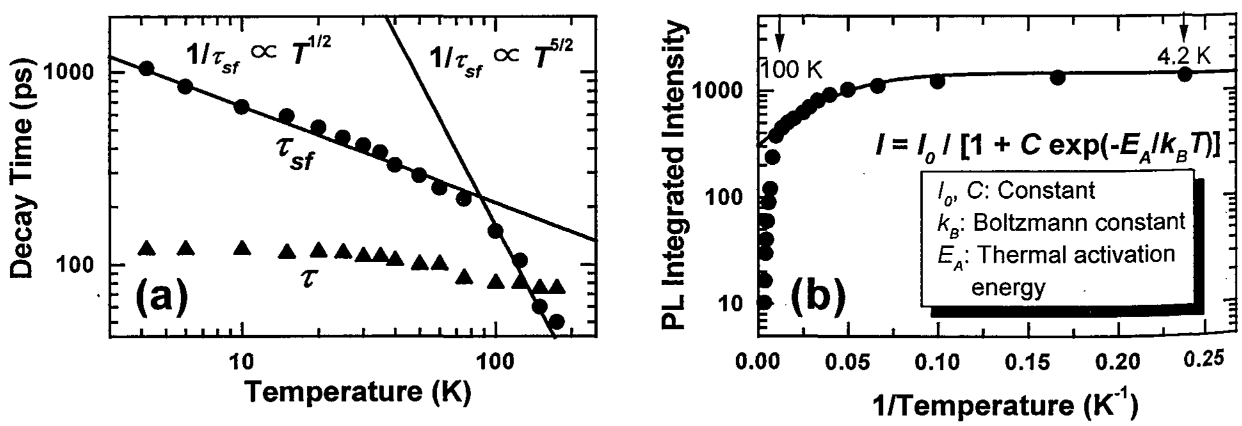

Fig. 3 (a) The temperature dependence of the τ_{sf} and τ in the $Cd_{0.85}Mn_{0.15}Te/Cd_{0.75}Mg_{0.25}Te$ MQWs with L_w = 150 Å and L_b = 100 Å. (b) The PL integrated intensity as a function of inverse temperature. The solid lines represent the best fit by the equation shown in the figure.

Chapter 9: Conclusions

This chapter summarizes the results discussed above.

審査結果の要旨

磁性半導体は、磁気光学的特性が極めて大きく現れ、応用が期待される物質である。著者は、分子線エピタキシー法による磁性半導体の成長技術を確立し、種々の量子井戸構造を作製した。また、これらの物質の磁気光学的特性とそのダイナミクスを、超高速レーザー分光により明らかにした。本論文は、その研究成果をまとめたもので、全文9章からなる。

第1章は、序論であり、本論文の背景と目的を述べている。

第2章では、分子線エピタキシー法による磁性半導体の薄膜と量子井戸の成長に関して記述している。分子線エピタキシー装置を用いた(CdMn)Te、(CdMg)Teの薄膜成長法の確立と、この2つの薄膜を交互に重ねた量子井戸の作製について述べている。また、これらの作製した試料について、高速反射電子回折、原子間力顕微鏡、X線回折により評価した結果を示している。

第3章では、発光分光を行うためのレーザーと分光測定系について、説明している。

第4章では、磁性半導体量子井戸における電子状態の基礎理論を記述している。とくに量子井戸中のエネルギー準位と、電子・正孔と磁性イオンの交換相互作用について詳述している。

第5章では、(CdMn)Te、(CdMg)Teの薄膜の磁気光学的特性の測定結果を示している。磁場下の発光の時間分解分光を行い、励起子磁気ポーラロン形成のダイナミクスや、明るい励起子、暗い励起子の存在を明らかにしている。

第6章では、(CdMn)Teを用いた単一量子井戸の磁気光学的性質について述べている。磁性イオンを井戸層に含むタイプと障壁層に含むタイプで、異なった磁気光学特性が生じることを示しており、本研究の有用な成果の一つとなっている。また、これらの量子井戸における励起子ダイナミクスを明らかにしている。

第7章では、(CdMn)Teの非対称2重量子井戸における励起子発光過程を調べており、2つの井戸間で起きる励起子のトンネル過程を議論している。

第8章では、(CdMn)Te多重量子井戸における励起子状態の温度による影響を調べた結果を述べており、この物質の応用に関連する重要な成果である。

第9章は、結論である。

以上要するに本論文は、磁性半導体を素材として、高品質の量子井戸構造を設計・作製し、これらの磁気光学的性質を調べ、その応用性を明らかにしたもので、応用物理学の発展に寄与するところが少なくない。

よって、本論文は博士（工学）の学位論文として合格と認める。