Qubits em pontos quânticos *

Relatório Final de Atividades Técnico Científico
Processo: FAPEMIG CEX - 523/05

Pesquisador Coordenador: Prof. Dr. Augusto Miguel Alcalde Millia

Instituto de Física
Universidade Federal de Uberlândia

22 de Abril de 2008

*Projeto desenvolvido no Instituto de Física, Universidade Federal de Uberlândia, com auxílio financeiro FAPEMIG
Conteúdo

1 Resumo
   1.1 Palavras chaves ........................................... 1

2 Resumo dos objetivos do projeto ................................ 2
   2.1 Objetivos ...................................................... 2
   2.2 Objetivos específicos ....................................... 3
      2.2.1 Identificação, manipulação e controle de qubits ..... 3
      2.2.2 Decorrência ............................................. 3

3 Atividades ....................................................... 5
   3.1 Pesquisa ..................................................... 5
      3.1.1 Efeitos intrínsecos na relaxação de spins em pontos
             quânticos .............................................. 5
      3.1.2 Controle de propriedades ópticas em pontos quânticos
             DMS .................................................. 5
      3.1.3 Magneto-absorção óptica e número de elétrons de condução
             em pontos quânticos ................................... 6
      3.1.4 Análise de propriedades magnéticas em MnTe e PbMnTe .... 7
      3.1.5 Interacção spin-órbita e fator-g em anéis quânticos ..... 8
      3.1.6 Relaxação de spins via modulação da interacção spin-
             órbita por fôtons ..................................... 11
   3.2 Participação em Eventos Científicos ........................... 12
   3.3 Produção bibliográfica ...................................... 12
      3.3.1 Artigos publicados ...................................... 12
      3.3.2 Artigos submetidos .................................... 13
      3.3.3 Manuscritos em preparação ............................. 13
   3.4 Orientações .................................................. 13
      3.4.1 Orientações concluídas .................................. 13
      3.4.2 Orientações em andamento .............................. 14

4 Conclusões ....................................................... 14

5 Publicações mais relevantes .................................... 16
1 Resumo

O presente projeto se concentra no estudo teórico de diversos processos físicos relacionados com a implementação de dispositivos de processamento quântico de informação baseados em pontos quânticos semicondutores. Serão estudados problemas relacionados com a identificação, manipulação e controle de qubits em pontos quânticos. Abordaremos, diversos aspectos do controle coerente da dinâmica de qubits individuais e acoplados e simularemos numericamente operações lógicas entre qubits. Serão elaboradas estratégias para se contornar ou minimizar os efeitos da perda de coerência através da análise individual de cada canal de decoerência.

1.1 Palavras chaves

- Nanoestruturas semicondutoras
- Pontos quânticos
- Relaxação de spins
- Switch óptico
2 Resumo dos objetivos do projeto

2.1 Objetivos

Os objetivos principais do projeto são:

1. Inicialmente explorar teoricamente diversos problemas físicos relacionados com a implementação tecnológica de sistemas de processamento quântico de informação (PQI) baseados em pontos quânticos (PQs). Abordaremos a identificação, manipulação e controle de qubits; simulação numérica dos efeitos de diversos canais de decoerência sobre a fidelidade do estado quântico criado. Focaremos nosso estudo nas mais importantes propostas baseadas em pontos quânticos:

   • **Propostas eletro-ópticas**: Estados excitônicos [1, 2] ou estados excitônicos polarizados [18] em pontos quânticos são identificados como qubits. O controle coerente de portadores via pulsos laser ultra-rápidos, permite a implementação de portas quânticas e o controle da interação de Coulomb e – h permite a manipulação de qubits.

   • **Spins em pontos quânticos**: Estados da spins da banda de condução em pontos quânticos são usados como qubits. O acoplamento entre qubits, pertencentes a dois pontos quânticos vizinhos, via a interação de troca permite a implementação das portas quânticas [7, 12].


2. Baseados nos resultados obtidos no primeiro item pretendemos sugerir novas propostas de dispositivos de computação quântica (CQ) baseadas em pontos quânticos que verifiquem os requerimentos definidos por DiVincenzo. [5]

3. Contribuir na formação de recursos humanos via a orientação de projetos de pesquisa de graduação e pós-graduação; promover a colaboração entre grupos de pesquisa nacionais e internacionais, e estimular o desenvolvimento de propostas de pesquisa que vinculem aspectos da física de nanoestruturas semicondutoras, ótica quântica e comunicação quântica.
2.2 Objetivos específicos
Os objetivos particulares do presente projeto podem ser organizados da seguinte forma:

2.2.1 Identificação, manipulação e controle de qubits

- Descrever os estados de uma partícula e estados elétron-buraco considerando os efeitos do confinamento espacial e magnético, presença da interação spin-órbita (SO) (contribuições de Dresselhaus e Rashba) e efeitos da geometria do ponto quântico. Este último aspecto tem sido recentemente explorado em nosso grupo, onde mostramos que alterações da forma produzem modificações dramáticas do fator-g eletrônico [15].

- Propor novos canais de modulação do fator-g de elétrons, buracos e excitons. Incluir contribuições da interação SO, morfologia do ponto quântico e presença de campos externos.

- Estudar o impacto da modulação (via mudanças na morfologia, efeitos de campos externos e interação SO) do efeito Zeeman e da interação de troca na implementação de operações entre qubits na proposta de Burkard et al. [3]


- Simular numericamente a execução (óptica e elétrica [8]) das operações quânticas fundamentais em CQ: rotação de qubits individuais e operações entre qubits.

2.2.2 Decoerência

- Estudar, em detalhe, os processos de interação entre qubits (excitons, níveis eletrônicos e spins) e o ambiente externo. Deverão ser abordados diversos processos de decoerência, especialmente os processos que envolvam acoplamento com fôtons. Calcular tempos de relaxação de spins devido à modulação do acoplamento SO por fôtons, ripple mechanism e processos de múltiplos fôtons.

- Estudar a aplicação de outras ferramentas teóricas no estudo de processos de perda de fase de excitons ativados opticamente em pontos quânticos.

- Avaliar a eficiência das propostas eletro-opáticas [11] considerando efeitos intrínsecos em pontos quânticos, como a mistura de bandas de
valência. Elaborar estratégias para controlar, e eventualmente limitar, as principais fontes de decoerência de qubits: decaimento de excitons, efeitos da mistura de bandas e presença de fôtons.
3 Atividades

3.1 Pesquisa

No período correspondente à duração do presente projeto foram desenvolvidas as seguintes atividades de pesquisa:

3.1.1 Efeitos intrínsecos na relaxação de spins em pontos quânticos

Estudamos a relaxação de estados de spin na banda de condução em pontos quânticos de CdTe. A estrutura eletrônica foi calculada usando o modelo k.p 8 x 8 de Kane-Weiler [10, 19]. Este procedimento de cálculo permite incorporar os efeitos de acoplamento inter- e intrabanda, não parabolidade e a aparição de um efeito spin-orbita intrínseco devido fundamentalmente ao acoplamento entre bandas de condução e valência. Considerando o gauge simétrico, um campo magnético externo na direção z é considerado para separar estados de spin oposto via o efeito Zeeman. Os modos de fônon acústicos são descritos como ondas planas. O mecanismo de relaxação considerado foi o potencial macroscópico de deformação (PMD), que esta conectado com a presença de interfaces na nanoestrutura. As vibrações acústicas modificam a simetria da hetero-interface modulando os níveis de energia dos portadores. A necessária mistura de estados de spin na banda de condução é produzida devido ao acoplamento da banda de condução com a banda de valência. Desta forma, analisamos a relaxação de spins associada a efeitos estritamente intrínsecos à nanoestrutura. Encontramos que, para pontos quânticos pequenos (∼ 50Å), a contribuição do PMD é dominante em relação ao associado com o potencial de deformação microscópico. As taxas apresentam um comportamento oscilatório como função do campo magnético devido a mistura de estados de spin. Na figura 1 mostramos a taxa de inversão de spin como função do campo magnético B para um ponto quântico de CdTe para duas transições típicas e considerando que o raio do ponto quântico é pequeno em comparação ao comprimento de Landau.

Detalhes do cálculo, discussão e resultados podem ser vistos no manuscrito submetido a publicação na seção 5.

3.1.2 Controle de propriedades ópticas em pontos quânticos DMS

Estudamos as propriedades ópticas de pontos quânticos baseados em semicondutores magnéticos diluídos do tipo Cd1-xMnxTe. Foi levada em conta a polarização da luz incidente e foi calculado o espectro de absorção óptica para as configurações de Voigt e Faraday. A estrutura eletrônica foi calculada usando o modelo k.p 8 x 8 de Kane-Weiler [10, 19] considerando os efeitos da interação de troca induzida pela acoplagem entre os spins dois ions de Mn e os spins dos portadores. Por exemplo, para o caso dos elétrons, o Zeeman splitting induzido pela interação de troca é dado por:

A. M. Alcalde  Relatório Final Técnico-Científico
\[ \Delta E_{zc} = xN_0 \alpha(S_a(x, B, T)) \], onde \( \alpha \) é a magnetização à temperatura \( T \) em uma amostra com \( N_0 \) células unitárias e concentração \( x \) de íons de Mn, \( N_0 \alpha \) é a constante de troca entre os elétrons \( s \) de Bloch. Nossos resultados mostram que é possível controlar as transições ópticas interbanda através de uma escolha apropriada da concentração \( x \) de íons de Mn e do campo magnético externo \( B \). Definimos um campo magnético crítico \( B_c \) que permite o chaveamento das transições ópticas, como mostra a Fig. 2. Este comportamento é produzido pelo fator-\( g \) eletrônico que, para \( B = B_c \), anula-se como consequência dos efeitos combinados do efeito Zeeman e a interação de troca. Este comportamento das transições ópticas é extremamente importante para propostas de memórias quânticas e controle óptico de qubits excitónicos baseadas em pontos quânticos. Futuramente, este sistema pode ser estendido para incluir dois pontos quânticos acoplados por interação de troca ou acoplamento dipolar, formando assim uma porta quântica que pode ser controlada com muita flexibilidade. Detalhes do cálculo, discussão e resultados podem ser vistos no artigo publicado em *Applied Physics Letters*, 88 052101 (2006), anexo na seção 5.

### 3.1.3 Magneto-absorção óptica e número de elétrons de condução em pontos quânticos

Consideramos os efeitos da população eletrônica na magneto-absorção intrabanda e estabelecemos um método para analisar os espectros de absorção, que permite determinar o número de portadores presentes na banda de condução de pontos quânticos esféricos II-VI. Estudamos detalhadamente as diferentes configurações ópticas e sua conexão com o número de portadores e topologia dos estados orbitais dos pontos quânticos. Este analise inclui um estudo das configurações Voigt e Faraday e das correspondentes regras
de seleção ópticas. A estrutura eletrônica foi calculada usando o modelo k.p 8 x 8 de Kane-Weiler [10, 19] e as propriedades ópticas foram estudadas dentro da aproximação dipolar. Nossos resultados estabelecem os regimes de luz incidente e campos magnéticos que permitem uma clara identificação do estado de ocupação eletrônica como pode ser visto na Fig. 3. Detalhes do cálculo, discussão e resultados podem ser vistos no artigo publicado em *Applied Physics Letters*, 87 231101 (2005), anexo na seção 5.

### 3.1.4 Análise de propriedades magnéticas em MnTe e PbMnTe

Trabalhamos em colaboração com o grupo experimental de nosso instituto na sintetização e caracterização óptica e magnética de materiais semicondutores magnéticos diluídos (DMS). O material teórico e experimental obtido nesta direção da pesquisa foi parte do projeto de mestrado do aluno Daniel Henrique Rodrigues, quem defendeu a dissertação: *Estudo das propriedades ópticas e estruturais de nanocristais de MnTe em vidros óxidos*, sob minha orientação.

Foram crescidos pontos quânticos de MnTe e PbMnTe de simetria esférica em matrices vítreas através do método de fusão. Este procedimento é relativamente simples e de baixo custo operacional. De acordo com nossa pesquisa bibliográfica, acreditamos estar em posse da primeira evidência bem-sucedida de crescimento de nanocristais semimagnéticos em matrices vítreas. Para verificar a formação de nanocristais, foram realizadas medidas de absorção e fotoluminiscência para diversas temperaturas. Cálculos baseados na aproximação de massa efetiva foram implementados para analisar a dependência das propriedades ópticas com o tamanho dos nanocristais.
Figura 3: Comparação entre espectros de magneto-absorção óptica para dois e três elétrons ocupando o mesmo ponto quântico, considerando um campo magnético $B=3$T. Foram consideradas a configuração de Faraday $\sigma_-$ e a configuração de Voigt $\pi^+$. 

Os resultados destes cálculos foram verificados através de técnicas de AFM. Por outro lado, medidas de espectroscopia Raman permitiram comprovar a composição e presença do composto MnTe nas matrizes vitreas.

Foram realizadas medidas de ressonância paramagnética eletrônica (EPR) nas amostras de MnTe e PbMnTe para verificar a presença do Mn nos nanocristais. Cálculos de EPR foram também desenvolvidos para analisar o comportamento das propriedades magnéticas como função de alguns parâmetros de crescimento, como tempo de tratamento térmico, velocidade de esfriamento, etc. Nossos resultados mais importantes estão resumidos nas Figs. 4 e 5.


3.1.5 Interação spin-órbita e fator-$g$ em anéis quânticos

O material desta parte de nosso projeto faz parte do projeto de mestrado do aluno Gineton Souza Diniz, quem defendeu a dissertação: *Efeitos da interação spin-órbita em anéis quânticos semicondutores*, sob minha orientação.

Utilizando a técnica da diagonalização numérica, tendo como base a
Hamiltoniana não-perturbada de uma partícula simples, em presença de um campo magnético na direção z, com um potencial de confinamento ajustável, estudamos os efeitos do acoplamento SO, levando em consideração as contribuições de Rashba e Dresselhaus. Os efeitos devido ao acoplamento SO, mostraram ser extremamente importantes na estrutura eletrônica, provocando fortes acoplamientos entre estados, com aparecimento de anticrossings nas energias em campos magnéticos críticos $B_c$, com $minigaps$ bem definidos, proporcionais aos parâmetros de acoplamento SO. Analisamos o comportamento do espectro de energia para duas geometrias: ponto quântico e anel quântico. Para um mesmo regime de parâmetros de Rashba $(dV/dz)$ e Dresselhaus $(\beta)$, comprovamos que a geometria tipo anel quântico fornece uma estrutura mais rica de mistura de estados de spin, permitindo identificar e manipular regiões onde uma determinada orientação do spin seja dominante. Mostramos que a mistura de estados afeta significativamente o comportamento dos estados de menor energia produzindo $minigaps$ e oscilações em função do campo magnético. Considerando os efeitos da interação SO, analisamos o comportamento do fator-g, usando a definição $g \propto \Delta E$, onde $\Delta E$ é a Zeeman spin splitting. Vimos que, quando são considerados os efeitos de SO, devido ao forte acoplamento entre os estados eletrônicos, dados pela
regra de seleção $\Delta m = m - m' = \pm 1$, os estados tem seu estado quântico de spin alterado, sendo assim, o fator-$g$, que é proporcional ao spin splitting do estado fundamental, apresentará um comportamento do tipo função degrau. A altura dos degraus estão associadas as formações de minigaps nos anticrossings das energias do estado fundamental. Também, propomos um esquema experimental para medir o fator-$g$ em nanoestruturas em presença da interação SO, através de absorção óptica intrabanda, em presença de campo magnético externo aplicado na nanoestrutura. A incidência de luz com polarizações bem definidas, tornaria possível acompanhar o spin splitting, já que as transições ópticas seguem regras de seleção bem definidas. Encontramos desta forma, em nosso trabalho, regimes característicos de campo magnético, confinamento lateral e geometria para manipular as misturas de estados eletrônicos e o fator-$g$. Uma correta compreensão do comportamento deste parâmetro é de vital importância nos processos de decoerência e manipulação de estados de spins em nanoestruturas semicondutores, para implementação em dispositivos spintrônicos e de informação quântica.

Um dos nossos mais importantes resultados está representado na Fig. 6, onde mostramos os efeitos da interação spin-orbita na estrutura eletrônica de um anel quântico de InAs. A complexa mistura de níveis modifica significativamente as propriedades ópticas e o fator-$g$ eletrônico; este aspecto será discutido em um manuscrito que se encontra em fase de preparação.

Figura 6: Espectro de energia para um elétron na banda de condução de um anel quânticos de InAs com raio efetivo $r_0=60$ Å. Foram consideradas as contribuições de Rashba e Dresselhaus na interação spin-orbita. No panel inferior é mostrado o valor esperado de $S_z$ que mostra a mistura de estados de spin como consequência da interação spin-orbita.
3.1.6 Relaxação de spins via modulação da interação spin-órbita por fôtons

Calculamos taxas de relaxação de estados de spin, considerando a modulação devido a fôtons acústicos da interação spin-órbita. Para isto, usamos a hamiltoniana spin-fônon proposta por Pavlov and Firsov [13, 14]. Neste modelo, a hamiltoniana que descreve as transições com inversão de spin, devido ao espalhamento de elétrons com fôtons, pode ser escrita em forma geral como: \( H_{ph} = V_{ph} + \beta [\sigma \times \nabla V_{ph}] \cdot (p + e/cA) \), onde \( V_{ph} \) é o operador associado ao potencial dos fôtons, \( \sigma \) o operador de spin, \( p \) é o momento do elétron e \( A \) é o potencial vetorial magnético associado com um campo magnético externo \( B \) na direção \( z \). O modelo pode ser facilmente adaptado a vários mecanismos de interação elétron fônon como: potencial de deformação, acoplamento piezoeletrico e acoplamento Fröhlich. Na Fig. 7, mostramos o comportamento oscilatório das taxas de transição com inversão de spin como função do campo magnético \( B \) para um ponto quântico parabólico de InAs. Identificamos claramente regiões de coerência robusta dos estados de spin e colocamos em evidência os efeitos competitivos entre o confinamento magnético e espacial. As oscilações das taxas estão diretamente relacionadas com a presença da interação spin-órbita.


**Figura 7:** Taxas de relaxação de spin como função do campo magnético \( B \) e do tamanho lateral de um ponto quântico de InAs. Os efeitos da contribuição de Dresselhaus foram levados em conta.
3.2 Participação em Eventos Científicos


3.3 Produção bibliográfica

3.3.1 Artigos publicados


### 3.3.2 Artigos submetidos

• *Phonon modulation of the spin-orbit interaction as a spin relaxation mechanism in quantum dots*. A. M. Alcalde, C. L. Romano, L. Sanz and G. E. Marques

• *Phonon mediated spin relaxation due to intrinsic level admixture in quantum dots*. A. M. Alcalde; S. J. Prado; L. Sanz and G. E. Marques

### 3.3.3 Manuscritos em preparação


• *Optical properties of PbMnTe quantum dots*. A. M. Alcalde, S. J. Prado and G. E. Marques

### 3.4 Orientações

#### 3.4.1 Orientações concluídas

• Iniciação Científica: *Manipulação de estados de spin em nanoestruturas.*
  - Aluna: Tatiana Vieira de Sousa
  - Ano de finalização: 2006
  - Bolsa: Programa PIBIC-CNPq-UFU.
4 CONCLUSÕES

- Dissertação de mestrado: Estudo das propriedades ópticas e estruturais de nanocristais de MnTe em vidros óxidos.
  Aluno: Danilo Henrique Rodrigues
  Data da defesa: 11 agosto de 2006
  Banca Examinadora: Prof. Dr. Augusto Miguel Alcalde Milla (orientador- INFIS-UFU); Prof. Dr. José Candido Xavier (INFIS-UFU); Prof. Dra. Yara Galvão Gobato (DF-UFSCAR).

- Dissertação de mestrado: Efeitos da interação spin-órbita em anéis quânticos semicondutores.
  Aluno: Ginetom Souza Diniz
  Data da defesa: 22 junho de 2007
  Banca Examinadora: Prof. Dr. Augusto Miguel Alcalde Milla (orientador- INFIS-UFU); Prof. Dr. Gilmar Eugenio Marques (DF-UFSCAR); Prof. Dra. Ragnh Augusta da Silva Zadra Armond (INFIS-UFU)

3.4.2 Orientações em andamento

- Iniciação científica: Controle coerente em nanoestruturas semicondutores.
  Aluna: Halyne Silva Borges
  Data de início: junho 2007

4 Conclusões

Durante o período de vigência do presente projeto, trabalhamos em problemas relacionados com identificação e controle óptico e magnético de estados quânticos em pontos e anéis quânticos semicondutores. Estabelecemos regimes de controle e chaveamento de excitações ópticas em pontos quânticos semimagnéticos. Este aspecto é de grande interesse, já que permite o estudo de sistemas mais complexos como memórias e portas quânticas. Estudamos o problema da perda de coerência de estados de spin através da interação elétron-fônon considerando as diversas contribuições da interação spin-órbita em nanoestruturas. Identificamos diversos regimes onde a coerência de qubit (estados de spin) é robusta, e analisamos o impacto da modulação da interação spin-órbita por fôtons acústicos. Baseados em resultados experimentais obtidos em nosso Instituto, analisamos diversas propriedades ópticas e magnéticas em nanocristais de MnTe e CdMnTe, compostos de grande interesse em spintrônica e CQ. Como mostra nossa lista de publicações, trabalhamos também em temas não diretamente relacionados com os objetivos do projeto. Analisamos o impacto da forma dos pontos quânticos nos fôtons de superfície e comparamos nossos resultados com medidas experimentais de espectroscopia Raman.
Acreditamos, que os objetivos do projeto foram atingidos de forma satisfatória. Estudos sobre dinâmica de qubits e controle coerente de excitons e spins estão sendo desenvolvidos. Nos últimos meses, temos trabalhado em problemas de controle coerente de processos de tunelamento em pontos quânticos duplos, pretendendo incluir efeitos da interação spin-órbita e canais de dissipação. Ainda, estamos implementando esquemas de cálculo de propriedades dinâmicas baseada em métodos da óptica quântica e teoria de Floquet. Estes estudos estão sendo desenvolvidos em colaboração com o Prof. Dr. Gilmar E. Marques e Prof. Dr. V. López-Richard do Departamento de Física da Universidade Federal de São Carlos, Dra. Carla Romano da Universidad de Buenos Aires e Dra. Liliana Sanz, pós-doutoranda em nosso grupo de pesquisa no INFIS-UFU.

É importante mencionar que, dentro do contexto de nossa instituição, contribuímos significativamente na formação de recursos humanos, já que no prazo aproximado de três anos conseguimos orientar duas dissertações de mestrado e um trabalho de iniciação científica, todos com bolsas de instituições de fomento. Atualmente, os alunos orientados dentro do presente projeto se encontram engajados em programas de doutorado em instituições brasileiras (DF-UFSCar) e internacionais (Ohio University-USA).

Acreditamos finalmente que, o apoio da FAPEMIG, no desenvolvimento e execução do presente projeto de pesquisa foi fator fundamental no logro das nossas metas.
Phonon mediated spin relaxation due to intrinsic level admixture quantum dots

A. M. Alcalde*  
Instituto de Física, Universidade Federal de Uberlândia, 38400-902, Uberlândia, MG, Brasil

S. J. Prado and G. B. Marques  
Departamento de Física, Universidade Federal de São Carlos, 13565-905 São Carlos SP, Brasil  
(Dated: July 10, 2007)

We calculate spin-flip relaxation rates for electrons in QDs spherical quantum dots due to the electron interaction with acoustical phonons via macroscopic deformation potential, also referred as the ripple mechanism. The relaxation between spin states is possible since the applied magnetic field and the coupling between valence and conduction bands produce and intrinsic spin admixture of the electron spin states. We show that the mixing produces an appreciable transition probability between coupled spin states. The electronic states are calculated within the k·p Kane-Warner model and the phonon modes are described by plane waves. We have studied the dependence of the spin-flip rates with the magnetic field and with the dot size. Our results show fair compatibility with the available experimental results.

PACS numbers: PACS 73.20.Da, 63.20.Mb, 71.30.+i

The ability to manipulate and to control processes that involve transitions between spin states is, at the moment, of great importance due to the recent applications involving electronic transmission and processing of information in areas that include quantum computation and quantum communication. Quantum dots (QD's) of different geometries are candidates for the implementation of semiconductor quantum communication devices because their electronic, and spin properties can be designed during growth controlled nano-fabrication techniques or by using the action of external fields. While for bulk [9] and for two-dimensional systems the spin relaxation processes have been studied to some extent [3, 4], the problem for fully quantized states in QD's still requires deeper discussions. Many processes that lead to spin relaxation in semiconductor systems have been identified and studied. One important aspect, for low dimensional systems, resides on the determination of the dominant process leading to spin-flip relaxation. In general, the identification of the processes through direct comparison with the experiments can become a formidable task. This problem is even more critical in QD's, since few experimental results exist and the theoretical discussion of spin-flip mechanisms for the system is still an open subject. Khaneja and Nazarev [6, 7] and de Souza [8] have studied several processes leading to spin-flip relaxation in GaAs and InAs QDs. In general, it was shown that the quantum confinement produces strong reduction on the relaxation rates with the magnitude ranging from $10^5$ to $10^6$ s$^{-1}$. It has also been verified that those relaxation processes that include spin admixture are dominant if compared to those "pure" processes based on the direct spin-phonon interaction.

It has been reported [9] that novel mechanisms, like the ripple mechanism (RM) in small QD's, dominates over the usual microscopic deformation potential (MDP) form of electron-phonon interaction. [10] The RM is inherent to any system containing interface and has its origin on the modulation generated by the vibrational modes localized at the interface region. Woods et al. [11] have made calculations of spin relaxation rates due to RM mechanism in GaAs QD's, where the spin admixture is provided by Donnathau spin-orbit interaction and the obtained scattering rates are small ($10^5$ s$^{-1}$) and strongly dependent on size and magnetic field strength.

In this work, we have studied spin-flip transition rates between electron spin-split states, as induced by the RM processes, in II-VI SQD's under the $I_d > R$ regime with $I_d$ and $R$ being the cyclotron and dot radii, respectively. We will consider small dot sizes where the RM displays significant contribution and the system shows interesting magnetic dependence of the relaxation processes.

Our starting point considers a realistic description of the electronic structure for a QD's in presence of magnetic field, within the k·p Kane-Warner Hamiltonian model (see Ref. [12] and references therein). The approach includes conduction-valence band coupling which generates an indirect spin-orbit coupling and the interaction produces conduction band electronic states with appreciable spin admixture. Following the effective mass approximation, we describe a carrier, in the presence of a perpendicular magnetic field $B = (0, 0, B)$ and a confinement potential $V$, as an eight component spinor $\psi$ that can be shortly summarised [12] as $\psi$. The symmetry inherent in the k·p Hamiltonian allows the separation of the Hilbert space into two orthogonal subspaces. Each spinor component, in the subspaces, is written in terms of exact solutions for each carrier type at $B = 0$.
for each subspace I and II are given by

\[ |\psi_{\text{M}}^{(\text{f})}\rangle = \sum_{\text{M}} \sum_{L=2(M)}^{L=\text{M}} |\phi_{\text{M}}^{(\text{L})}\rangle \]

where \( l \) represents the respective carrier periodic \( 1 \) function at the \( \Gamma \)-point and the coefficients \( c_{\text{M}}^{(\text{L})} \) are of quantum numbers \( n, L \) and \( M \). The boundary condition \( \phi_{\text{M}}^{(\text{L})}(R) = 0 \) determines the \( n^{\text{th}} \) zero, \( \mu^{(L)} \), for \( \Gamma \) function, \( j_{1}(\mu^{(L)}/R) \).

Figure 1 shows the magnetic field dependence of energy level separation for CeTe (13) SQD with \( R = 30 \, \text{Å} \) (part a) and \( R = 50 \, \text{Å} \) (part b). For \( L \) value of magnetic field, the levels are labeled by the \( \epsilon \) quantum number \( M \) by the energy ordering number \( \mu \), by the spin number \( s = |\mu| \) (spin-up) or \( s = -|\mu| \) (spin-down). This notation is reached after selecting the lowest solution value at \( B = 0 \), of the coefficients in the \( l \) combination (see Eq. 1) forming each component of carriers I and II. Thus, electron levels are labeled \( M^{(L)}(H) \) with \( H \) indicating which Hilbert subspace belongs to. The solid (dashed lines) in the \( l \) represent spin-flip (spin-preserving) allowed optical transition energies where the initial state displays significant spin admixture. For instance, for \( R = 30 \, \text{Å} \) and \( B \) the state \( M^{(L)} \) (II) is a spin-up state with the largest gain. This characteristic is charged as the magnetic field increases and, above \( B = 2T \), the system acquires spin-down dominant character because the strength of coefficient becomes larger.

These optical transitions are not zero since there is an induced indirect electron spin-orbit coupling inside the \( k \)-\( p \) Hamiltonian due to the conduction-valence coupling between Bloch states \( |\phi^{(1)}\rangle \) (for the \( M = 0 \) level in the space I) and \( |\phi^{(1)}\rangle \) (for the \( M = -1 \) level in the space I). They are connected, in the framework of the \( k \)-\( p \) Hamiltonian, through the operators \( \sigma^{+} \) and \( \sigma^{-} \), respectively (see columns \( H_{\text{C}} \) and \( H_{\text{V}} \) in Appendix of Ref. [13]). These indirect couplings between spin-up and spin-down electron states determine the parity of conduction and valence state components, as can be observed in Eq. 1. The overall contribution is different from zero, because the operators \( \sigma^{+} \sim (\partial/\partial \theta|x_{i}|(\partial/\partial \theta|x_{i}|) \) changes the parity of the function on which they act. These processes are very sensitive to the QD shape and material (strength of coupling) and may produce a Zeeman-split ground-state level inversion, as reported in Ref. [14].

We use the procedures described in Ref. [9] to treat the RM electron-phonon interaction. It can be shown that the important terms in RM interaction are related with the spatial dependence of the conduction potential \( V(x) \) and the effective mass \( m^{*}(x) \). The matrix elements for the overall dipole interaction between initial (I) and final (F) electron states can be written as [9]

\[ \langle \psi_{\text{F}} | H_{\text{int}} | \psi_{\text{I}} \rangle = \frac{e^{2}}{2m} \int d^{3}q \frac{\partial^{2} \phi_{\text{F}}^{*}}{\partial \theta^{2}} A_{\alpha} | A_{\beta} \rangle | A_{\alpha} \rangle \phi_{\text{I}} \]

(2)

where \( A(x) \sim \exp(\pm i \alpha) \) is normalized LA-phonon displacement field, \( \hat{a} \) is an unitary vector pointing outwardly.
and the normal derivatives are evaluated inside the Q. Since RM interaction is independent of spin variable, its mechanism can only guarantee energy conservation as the only possible way to obtain nonzero spin transition matrix elements for electronic states displaying spin admixture. We can calculate transition rates \( \Gamma_{i\rightarrow f} \) between initial (i) and final (f) spin states, for an electron scattered after the emission of one acoustic phonon frequency \( \omega = \nu_k \), as

\[
\Gamma_{i\rightarrow f} = \frac{2\pi}{\hbar} \sum_{q} |\langle \psi_f | H_{\text{ac}} | \psi_i \rangle|^2 \delta(E_f - E_i - \hbar \nu_k).
\]

The angular integration is simplified by expanding the wave function in spherical harmonics and the zero vector value for the angular integration establishes selection rules for the phonon emission between states i and f. Within RM mechanism, the emitted phonons in SQUID have mostly the \( \nu_k = 0 \) character. Small deviations from sphericity induce transitions with higher angular momentum. The phonon-mediated transitions occur between an initial state displaying spin admixture and the ground-state characterized by negligible spin mixture. To comply with these RM mechanism selection rules, \( \Delta L = \pm 1 \), initial states must have \( L = 0 \) dominant component since the final state has \( L = 0 \) character predominantly.

In the Fig. 2 we are showing the allowed spin-flop, \( \phi^A_{\parallel}(\parallel) \rightarrow \phi^A_{\perp}(\parallel) \) (solid lines) and spin-preserving relaxation rates, \( \phi^A_{\parallel}(\parallel) \rightarrow \phi^A_{\perp}(\parallel) \) (dashed lines) for \( \hbar = 30 \) A CdTe SQUID. As field increases, we observe oscillations for the spin-flop and a smooth variation in the phonon-induced relaxation rates. The reason why these transitions exist for \( \langle \psi_f | H_{\text{ac}} | \psi_i \rangle \neq 0 \) is identical to the optical transition case, after replacing the linear momentum by the coordinate operator. Omitting some constants coming from phonon normalization and normal derivatives of the envelope wave functions \( J_{\text{s},\text{s}} \), the RM matrix elements have the form

\[
\langle \psi_f | H_{\text{ac}} | \psi_i \rangle \propto \sum_{\Delta k_\lambda} C_{\mathrm{gs}}(\lambda) C_{\mathrm{gs}}(\lambda') \langle \psi_i | \text{ac} | \psi_f \rangle.
\]

Here, \( |i\rangle \) and \( |f\rangle \) are sets of quantum numbers corresponding to initial and final states connected by RM selection rules. Thus, the origin of the oscillation in the spin-flop transitions comes from the harmonic-like term \( J_{\text{s},\text{s}}(\Delta k, \Delta k') \), where \( \Delta k = (E_f - E_i)/\hbar \nu_k \) and \( \Delta k \) is the spin-flip energy difference.

The observed singular fact on the relaxation rates has not been reported previously and opens new possibilities towards manipulation and control of spin properties in QD's. In general, the conduction-valence band coupling between spin states change when quantum dot shape deviates from its spherical form. We observe an increase of the former "restrict" selection rules and transitions rate softening for increasing magnetic "squeezing" of spin states. As observed in Fig. 2, these spin-flop transition rates vary between \( 10^8 \text{ s}^{-1} \) to \( 10^{10} \text{ s}^{-1} \), values that are significantly smaller than those reported for the change relaxation with phonon emission via RM scattering [12] in CdS SQUID (\( 10^9 \cdot 10^{10} \text{ s}^{-1} \)). A very long spin-flip re-
Publication relevant

Location time leads to longer spin coherence, and may be of great importance for quantum computing application, for example.

In the Fig. 3, we are showing the phonon-induced relaxation rates for $R = 50 \AA$ CdTe SQD. In panel (a) we plotted the spin-flip transitions $\sigma^{+}(I) \rightarrow \sigma^{-}(II)$ (solid line) and spin-preserving transitions $\sigma^{x}(I) \rightarrow \sigma^{x}(II)$ (dashed lines) for that ground-states. In panel (b) we show the transitions between excited and ground-states for $\sigma^{+}-\sigma^{-}$ case $\sigma^{+}(I) \rightarrow \sigma^{-}(II)$ (solid lines) and for the spin-preserving case, $\sigma^{x}(I) \rightarrow \sigma^{x}(II)$ (dashed lines).

We should emphasize here that the initial states are not the same for the $R = 30 \AA$ SQD of Fig. 2. This is because for $R = 50 \AA$ SQD the state $\sigma^{x}(I)$ does not present relevant spin admixture. This behavior is a consequence of the level anti-crossing and consequent exchange of spin character that occurs at $R \simeq 40 \AA$. For a detailed discussion of the size and SQD morphology effects on the spin-states we refer to Refs. [12] and [14].

For easy comparison, the allowed optical transitions are shown in Fig. 1. Both optical and phonon-induced relaxation rates between these states are strictly related to the effects of level coupling caused by the spatial confinement and by magnetic exchange inside the Hamiltonian model. These new oscillations observed in the spin-flip relaxation rate occur because the magnetic confinement increases the indirect spin-orbit interaction between states. At the critical magnetic field where the relaxation rate reaches a minimum, is observed a stronger resonance with interband phonon energies of the RH modes.

For low $B$ values, the rates do not present any important variation with the dot size. For both dot radii, the phonon mediated rates are approximately $10^{10}$ s$^{-1}$. This range of calculated values agrees with some experimental observations by Gupta et al. [15], where spin-flip relaxation times are reported as approximately constant dot radius in the range 20 Å to 80 Å.

In conclusion, we have suggested the alternative electron-phonon RM for small QD's that present oscillatory spin-flip relaxation rates for SQD's. Our work provides a better understanding of the spin-flip relaxation processes, especially the rate dependence with the size and the indirect spin-orbit interaction produced by the conduction-valence band coupling. The realistic band model is essential to the description of the dot electronic structure. Oscillatory variations of the rates with increasing magnetic field, for given dot size, occur as a consequence of the competition between spatial and magnetic confinement.

These oscillations in the spin-flip rates were also found by Destefani et al. [16] that used full spin-orbit, including Rashba and Dresselhaus terms, to study the spin-flip relaxation of electrons in cylindrical QD with nanohole structure, more specifically GaAs and InAs. For small magnetic field, we have obtained relaxation rates of the order of $10^{11}$ s$^{-1}$ which is in fair agreement with few reported experimental results. Finally, it is important to mention that our results have allowed the identification of regimes of magnetic field where the spin-split states present robust coherence. This fact would help the tuning of necessary input conditions in order to achieve efficient form of precessing quantum information, for example.

This work has been supported by Fundação de Amparo a Pesquisa do Estado de São Paulo (FAPESP) and Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq).

[13] The Off's material parameters are: the energy band gap $E_{g} = 1.6009$ eV, the spin orbit energy $E_{SO} = 0.053$ eV, the Luttinger parameter $\gamma_{L} = 5.37$, $\gamma_{R} = 1.97$, $\gamma_{T} = 1.96$, the electron effective mass $m_{e} = 0.051m_{0}$, the non-parabolicity conduction band parameter $1 = 1.34$, the Kane parameter $E_{g} = 1.79$ eV.
Manipulation of g-factor in diluted magnetic semiconductors quantum dots: Optical switching control

V. López-Richard
Faculdade de Física, Universidade de São Paulo, 05508-090, São Paulo-SP, Brazil

S. J. Prado, G. E. Marques, and C. Trajano-Güeme
Departamento de Física, Universidade Federal de São Carlos, 13565-905, São Carlos-SP, Brazil

A. M. Alcalde
Faculdade de Física, Universidade Federal de Uberlândia, 38400-002, Uberlândia-MG, Brazil

(Received 11 May 2005; accepted 6 December 2005; published online 30 January 2006)

We propose an optical switch based on a spin-tuning mechanism in diluted magnetic semiconductor quantum dots. At certain critical magnetic fields, Bc, the Zeeman spin-splitting energies can cross leading to a zero value of the effective electron g-factor and the Fermi level undergoes a spin-flip transition. Magneto-optical switching is obtained for magnetic fields below and above Bc. Correlations between Bc, confinement shapes, dot sizes, and how material compositions have been established within well-defined temperature and magnetic impurity composition ranges. The generality of the presented theoretical framework allows for its application to magnetic field controlled quantum dot arrays, and spin-injection among others. © 2006 American Institute of Physics. [DOI: 10.1063/1.2168499]

We present a theoretical description of a fundamental effect in diluted-magnetic semiconductor quantum dots (DMSQDs), based on a cooperative magnonic phenomenon. It takes into account the complex orbital contribution to the magnetic moment of conduction band electrons due to spatial confinement and interband coupling, along with the interaction with magnetic ions. In order to characterize the system response, the theoretical analysis will be based on the optical properties. The exchange-enhanced Zeeman spin-split levels cross (see Fig. 1(a)) at B=Bc, leading to a null electron Landé factor at this crossing point (see Fig. 1(b)). For one-electron DMSQD systems, the Fermi level undergoes a spin-flip transition at B=Bc. According to the Fermi-level position, the system can relativistically filter or emit, if properly excited, light with two different and well-defined resonant frequencies, ωo and ωp, corresponding to the electron-hole pair ground state excitation energies (with orbital momentum ℓ=0) and different spin orientation.

We have used three different light-beam sources: Two with circular polarizations in Faraday configuration, σ±, and one with linear polarization in Voigt configuration, σ∥, as shown in Fig. 2. The values characterizing the operational regime of the magnetic switch are: ωo, ωp, and the critical field, Bc, as shown in Fig. 2. Given a critical magnetic field, Bc, the switching mechanism here proposed can be activated within two regimes: B<Bc and B>Bc. At B=Bc, the values of ωo and ωp for different optical polarization depends on the energy difference between the valence-band levels involved in the electronic transition to the conduction-band spin-split ground state. Along with the frequency ωo, this provokes a switch in oscillatory character. Following the selection rules for the electron-hole pair excitation, we obtain, at B=Bc, A(ωo−ωp)=E(ωo)−E(ωp) for σ± and A(ωo−ωp)=E(ωo)−E(ωp) for σ∥. Here, E(ωo) and E(ωp) are the energies of the heavy and light-hole spin-split states, respectively. This property can be tuned by the QD size and Mn content, xM, as depicted in Fig. 2(b).

The adjustment of DMSQD properties can be analyzed with a suitable method of calculating the electronic structure. We have used the k·p Hamiltonian for a Cd1−xMnxTe quantum dot (QD), including the effects of exchange interaction within the same theoretical framework by adopting virtual crystal and mean field approximations, the cooperative magnetic system response can easily be described. The antiferromagnetic interaction between the spin of 3d electrons of Mn2+ ions with spins of conduction-band free carriers has the form $\mathcal{H} = -\frac{1}{2} \sum_{j,k} (\mathbf{S}_j \cdot \mathbf{S}_k + \mathbf{S}_j \cdot (\mathbf{S}_j - \mathbf{S}_k)) \mathbf{S}_i \mathbf{S}_o$, where $\mathcal{S}_i (\mathbf{B}, T, \mathbf{s})$ is the magnetization, at temperature T, in a sample with Mn unit cells and Mn content, x. $\mathcal{S}_i \mathbf{S}_o$ is a uniform exchange constant between $\mathbf{S}_i$ conduction band Bloch states (for details, see Refs. 3 and 4). Furthermore, we shall work within the strong confinement regime, where $\hbar^2 \kappa^2 >> 1$.

![Figure 1](image-url)
Publicações mais relevantes

The system, which helps to clarify the correlation between different parameters and their effects on $B_s$. The energy contributions from exchange-induced and normal Zeeman terms can be treated separately:

$$\Delta E_{\text{ex}} = \Delta E_Z + \Delta E_{\text{ex}}$$

(1)

where $\Delta E_Z = g^2\mu_B B$, calculated with the effective Landé factor, $g^2$, in the absence of exchange. The exchange-induced Zeeman term is $\Delta E_{\text{ex}} = \frac{1}{2} \sigma \cdot (\mathbf{S}_a \times \mathbf{B})$, or twice the semi-magnetic offset for electrons. The values of $B_s$ can be obtained by solving for the zeroes in Eq. (1), i.e., $\Delta E_{\text{ex}} = 0$. However, above the saturation, we may expect that significant alignment of magnetic ions will occur. Under this condition, the magnetization does not depend on the field and we can give an analytical estimate of $B_s$ for a given sample, as

$$B_s(\lambda) = \frac{N_{\lambda}}{H_{\lambda}} \lim_{\lambda \to 0} \left( \sin \left( \frac{\pi}{2} \lambda + \frac{\pi}{2} \right) \right).$$

(2)

This approximate, valid for low temperatures near saturation, shows the functional dependence of the critical field on $\lambda$ and on the system parameters: $N_{\lambda}$ and $g^2$. By defining $\Delta E_{\text{ex}} = \frac{1}{2} \sigma \cdot (\mathbf{S}_a \times \mathbf{B})$, we have the sign $f(x) = 1.750 - 0.012(x + 0.015)$, for $C_{2v}$. The small and for large values of $x$, Eq. (2) shows slightly different slopes for $B_s(\lambda)$, analogous to the result shown in Fig. 3.

For a parabolic conduction-band model, the effect of confinement shape is not spin related, since it equally affects both spin-split levels. Thus, in such approximation, $\Delta E_{\text{ex}}$ will not be affected by the confinement geometry or by its size. However, as we have shown in Fig. 3, the critical field values are, in fact, sensitive to confinement geometry. The whole effect can be explained by the transformation of the effective Landé factor, because, in previous calculations, we have shown a dependence, $g_{\text{eff}}(\lambda) = 1/\lambda$, for $C_{2v}$ QDs. Introducing this size-dependent effective Landé factor into the Zeeman splitting estimate, we can give the functional dependence of $B_s(\lambda)$ on the geometry. This justifies the increase of the $B_s(\lambda)$ slope with size $\lambda$, but was obtained numerically and shown in Fig. 3. The difference between the critical field values for $S$ and $S'$-sp geometries can be deduced easily from the discussion, presented in Ref. 5, on the slope dependence of the effective Landé factor.

As explained above, the switching process is a result of the correlation between $\Delta E_Z$ and $\Delta E_{\text{ex}}$. It is only effective if the condition $|\Delta E_Z| < |\Delta E_{\text{ex}}|$ is fulfilled at any finite value of $\lambda$. Thus opens a question whether the switching mechanism is only effective below a certain temperature. The critical field dependence on temperature has been calculated and depicted in the inset (b) of Fig. 3. Observe that $B_s$ decreases with increasing temperature until it becomes zero at a certain $T_c$. The critical temperature, $T_c$, for $S$-sp geometries can be obtained from a multiband calculation, by analyzing the limit $B_s(\lambda) \to 0$, as depicted in the inset (b) of Fig. 3. We can also propose an accurate analytical expression for $T_c$.

The spin-split level crossing, seen in Fig. 1, can be suppressed when the slope of $|\Delta E_{\text{ex}}(B)|$ equals the slope of $|\Delta E_Z(\lambda)|$ at $B = 0$ (see the inset (d) of Fig. 3).
5 PUBLICAÇÕES MAIS RELEVANTES

Lopes-Richard et al.

\[ \frac{2}{dH} \left( \frac{\partial^2 H}{\partial s^2} \left|_{s=0} \right. \right) \cdot \frac{\partial^2 H}{\partial s^2} \left( x(B,T) \right) \left|_{s=0} \right. \cdot \frac{\partial^2 H}{\partial s^2} \left( x(B,T) \right) = \mu_0 \chi(T) \left[ \frac{1}{2} n \right] . \] (3)

Thus, we can obtain the critical temperature as

\[ T_c(x) = \frac{\pi^2}{3} \left( \frac{1/2 + 1}{k_B T_m(x)} \right)^{1/2} \left[ \frac{1}{2} n \right] . \] (4)

This expression is not restricted to the saturation condition of sample magnetization; therefore, it is expected to be a good estimate of real critical-temperature values. Low fields ($B \to 0$) and high temperatures (important from the application point of view) come within the validity frame of the theoretical description presented, highlighting the desired applicability framework of the described critical phenomenon.

An empirical method of determining both, the existence of the magnetic switching regime and the accurate value of the critical magnetic field, can be achieved by using resonant spin-flip Raman scattering, in Faraday configurations, with complementary circular and linear polarizations; $\sigma_\alpha^L, n_\alpha^L$.

The calculated Raman shifts for these complementary configurations are shown in Fig. 4. Using this procedure, implemented experimentally in Ref. 6, we can obtain a very precise determination of $T_c(x)$.

In conclusion, we have theoretically described a critical behavior found in zero-dimensional nanosystems built on dilated-magnetic semiconductors. The correlation between $B_c$, $T_m$, and QD parameters has been evaluated in a compact and comprehensive form. In the general properties discussed, the external field acts as a switch control, changing between two regimes: $B < B_c$ and $B > B_c$. We have presented accurate analytical estimates of $B_c(x)$ and $T_c(x)$, be-

FIG. 4. Calculated Raman shifts, as a function of field, for the complimentary scattering configurations.

low which the switching operation always exists. The validity frame of this phenomenon, that includes low fields and high temperatures, lies within an unexplainably model range, which should encourage new experimental and theoretical endeavors.

This work was supported by Brazilian agencies FAPESP and CNPq.

Intraband magnetoabsorption as a probing tool for the quantum dot charge

V. López-Richard
Faculdade de Filosofia, Ciências e Letras de Ribeirão Preto, Departamento de Física e Matemática, Universidade de São Paulo, 14060-901, Ribeirão Preto, São Paulo, Brazil

A. M. Alcalde
Universidade Federal de Uberlândia, Caixa Postal 953, 38400-901, Uberlândia MG, Brazil

S. J. Praco and G. E. Manques
Departamento de Física, Universidade Federal de São Carlos, 13565-905, São Carlos São Paulo, Brazil

C. Trallero-Giner
Department of Theoretical Physics, Havana University, Vedado 10400, Cuba

(Received 11 May 2005; accepted 5 October 2005; published online 28 November 2005)

A method of characterizing the quantum dot charge buildup in a magnetic field is proposed based on the far-infrared magneto-optical response properties. The inherent topological symmetry of the nanostructure and several optical configurations are analyzed as key factors determining the appropriate use of intraband transitions as a probing tool for the quantum dot charge buildup.


Recently, semiconductor quantum dots (QDs) have been proposed for new and promising applications as fundamental building blocks in quantum information systems\(^1\) and new electro-optical devices.\(^2\) In this context, physical properties of the conduction band, such as spin (charge) relaxation and optical excitations,\(^3\) have come into the focus of current research. The present study demonstrates that the precise control and identification of optical intraband transitions can be useful for the design and for the characterization of these novel systems. Two critical problems concerning intraband transitions are the dependence of absorption on the polarization and the efficiency of the electron population of the states.\(^4\)

Zhang\(^5\) and Aslam\(^6\) have demonstrated experimentally the effects of polarization on photoresponse spectra as well as the appearance of new features in the far-infrared (FIR) photoresponse that were clearly identified as due to the varying numbers of electrons inside the dot. Precise control of the QD charge plays a critical role in the identification of spin-dephasing mechanisms, in the performance of IR detectors, and in the analysis of the magnetic properties of diluted magnetic semiconductor QDs.\(^7\) Also, significant progress has been made towards the use of intraband absorption measurements as a tool for the determination of QD charge buildup.

In this letter, we consider the effects of the electron population of QDs on their intraband magneto-optical spectrum and an accurate method of analyzing these spectra, so as to characterize the charge buildup, is proposed. The advantages and disadvantages of such an approach are analyzed in terms of factors such as: response resolution, magnetic field values, and energy ranges. We present a detailed discussion of the configuration symmetry of optical measurements and their relationship to the orbital topologies associated with the electronic levels involved in the optical transitions. This study affects a general view of the system response to different optical configurations. Thus, we have highlighted the importance of an adequate choice of factors that control the transition selection rules and which, in turn, may enhance or reduce the response sensitivity to the charge buildup on the QD. According to the incident light polarization, two main geometries have been analyzed: the Faraday (incidence perpendicular to the magnetic field) and the Voig (incidence transverse to the magnetic field) configurations.

We have derived the intraband magneto-optical selection rules and calculated the corresponding oscillator strengths and magnetoabsorption spectra for both optical configurations with circular and linear polarizations, thus providing a compact and general analysis of the system response that can be put into a comprehensive and tractable form.

The Weiler-Kane k·p method (see Ref. 5, and references therein) has been used to describe the electronic structure of a QD with a spherical confining potential in the presence of a magnetic field. B=0,0,0. B is the mixture of the spherical (spatial) and cylindrical (z) field components on the eight-component spinors β and on the eigenvalues (E) are studied by solving H_{αβ}β=Eβ, where the magnetic field is included in the symmetric space through the usual canonical operator \(g(\vec{r})/α\). We restrict our discussion to the weak magnetic field regime \(\vec{B} \cdot \vec{B} \gg 1\), \(\vec{B} \cdot \vec{B} \gg 1\), where the magnetic length and \(\vec{R}\) is the QD radius. Within the k·p Hamiltonian the states with different symmetries are coupled even at \(B=0\) and the angular momentum \(L\) is not a good quantum number. Nevertheless, by neglecting only the warping term, proportional to \(μ(\gamma_2-\gamma_3)/2\), the solution space of the Kane-Winter Hamiltonian can be separated into two independent Hilbert subspaces (\(H=1,1\)). Following the symmetry requirements for B1z axis, the calculated spinor states, \(β_{11z}\), can be labeled according to the following set of quantum numbers: \(L_z=\pm M_\text{z}\) (i) The z component of the angular momentum \(L_z=\pm M_\text{z}\). (ii) The well-defined total angular projection \(F_\text{z}=L_z+J_z\) \(F_\text{z}=\pm 1/2\), \(\pm 3/2\), ...), where \(J\) is the quantum number of the spherical harmonics of the \(G_\text{z}\) and \(G_\text{z}\) Bloch functions at the \(\Gamma\) point. (iii) The orbital index \(N\) for increasing energy.

The intraband optical selection rules allow only transitions between states belonging to different Hilbert subspaces (\(1\rightarrow 2\) or \(2\rightarrow 1\)) and can be summarized as: (i) In the Faraday geometrical configuration, the transition selection rules are given by: (ii) In the Voig geometrical configuration, the transition selection rules are given by:
day geometry (xEBE) with circular polarizations, \( \Phi^\lambda \), the transitions connect states with \( \Delta F, m = 1 \) (or equivalently \( \Delta M = 1 \)); (ii) in Voigt geometry (xEBE) with linear polarization in the z direction, \( \Phi^\lambda \), the corresponding transition connect states with \( \Delta F, m = 0 \) (or equivalently \( \Delta M = 0 \)). Here \( \mathbf{x} \) is the light wave vector.

Within the electric-dipole approximation, we can write the absorption coefficient as

\[
a_{\Phi}^\lambda = \alpha_{\text{em}} \sum_{\nu, \nu'} \frac{|\langle \Phi^\lambda | \mathbf{E}_{\nu} | \Phi^\lambda \rangle |^2}{(\Delta \nu)^2 + 1} \delta \left( \nu - \nu' \right),
\]

(1)

where, \( \Delta \nu = \nu - \nu' \), being \( \nu' \) being the linear frequencies of the space containing the \( \nu \) and \( \nu' \) is the resonant effective mass of the carrier. In Eq. (1), \( \Delta \nu = \nu' - \nu' \) is the photon frequency, \( \mathbf{E}_{\nu} \) is the probability of finding an electron in the state \( \nu \) with energy \( E_{\nu} \), \( T \) is the homogeneous broadening, \( \alpha_{\text{em}} = 1.2 \pi \mu_0\mu_0/(1 + \omega_0)^2 \), and \( \eta \) is the refractive index.

We study the effects of a change in the QD population, through the term \( \Delta \nu = \nu - \nu' \), on the absorption coefficients for Faraday (xF) and Voigt (xV) configurations in the electric dipole approximation. \( \Phi^\lambda \) being the occupation function. These statistical terms determine the probabilities of finding a filled state \( \nu \) with energy \( E_{\nu} \) in the initial \( \nu \) and an empty final \( \nu' \) state which are connected according to the optical selection rules summarized above.

We choose CdTe, with well-established band parameters, as the proper material for the calculation of the QD electronic structure. The discussion will be focused on general qualitative features of the optical response. In Fig. 1 we show the calculated magnetoabsorption spectra, for Faraday (xF) and Voigt (xV) configurations, for a QD with \( R = 30 \) Å, at various magnetic fields.

The panels (a), (b), and (c) of Fig. 1 show the magnetooptical response in the singly occupied spin-polarized \( \nu = 1 \) QD. The spin-polarized double-occupied case \( \nu = 2 \) is displayed in panels (d), (e), and (f). First we observe, for \( B = 0 \) T, an identical shape of the absorption spectra for all these configurations, a direct consequence of the degenerate spin states in a QD without field. The charged QD may become spin polarized by the magnetic field and this will subsequently affect the optical response, which depends strongly on the light polarization. The choice of optical configuration plays a decisive role in these. If these spectra are to be used to characterize the QD charge and the change in the energy position with increasing magnetic field is reflected in the variation of the resonant energies. Thus, important qualitative differences are obtained between \( B = 0 \) and \( B \neq 0 \) and between spin-polarized and spin-unpolarized systems.

The observed differences between the optical configurations analyzed in Fig. 1, for a given \( \nu = n \), is due to selection rules and electronic structure dependence on external fields. We must point out that the selection rules for Faraday and Voigt geometries allow only those transitions where \( \Delta F = 1 \) or \( 0 \), respectively. Thus, the rich spectral structure displayed depends, essentially, on the interlevel coupling effects and on the subsequent variations of the magnetic fields with the field, for different levels (see Ref. 8). Therefore, a relaxation in the selection rules must be considered an undesirable effect. Even when this sometimes inevitable effect occurs, we observe in Fig. 1 that for certain configurations the well-defined qualitative modifications can be observed in the system response when changing from \( n = 1 \) to \( n = 2 \). As the occupation number increases, the absorption spectra reflect the statistical level distribution of electrons in the QD. In the right-hand panels of Fig. 1 we have assumed a two-electron occupation where one electron is in the ground state \( \nu = 1 \) and the other in the first excited state \( \nu = 2 \). When the second electron is injected into the QD and the temperature of the system is such that \( k_B T < E_{\nu}^{1,2} \), the incident light provides a new set of electron transitions marked with arrows in the right panels of Fig. 1. These new peaks can be independently studied by selecting the best optical configurations. For the range of frequency considered, the calculation showed that \( \nu = 1 \) has the better resolution for the characterization of a QD with \( n = 1 \) or \( n = 2 \) occupation numbers. Clearly, these characteristics can be ascribed to the particular energy structure of a given system. Although this assertion is correct, we have shown that a better and well-resolved procedure for system characterization depends on the proper choice of the light configuration. For the energy range considered, the absorptivity decreases as the magnetic field increases.

FIG. 1. Intraband magnetoabsorption for Faraday (xF) and Voigt (xV) geometries of a QD of 30 Å. CdTe charged QD at various fields. The left and right panels show the spectra for one- and two-electron occupation. The arrows show the new transitions induced by population effects.
In conclusion, we have shown that magnetoabsorption techniques can be employed as a valuable probe of the occupation number of free electrons in a QD. The appropriate selection of light sources and external field configurations is a key factor for the enhancement of the required resolution of the QD charge characterization. Intermixed admixture of states, leading to the relaxation of the selection rules, if properly treated, may help in the study of characteristics associated with the electronic population of the QD states. Our results are the first step toward a complete and rigorous discussion of other magneto-optical properties and the spin-polarized injection of carriers into semiconductor QDs, explored through the Faraday and Voigt geometries.

This work has been supported by the Brazilian agencies Fapemig and Fapesp.

A. M. Alcalde Relatório Final Técnico-Científico

Downloaded 28 Nov 2007 to 203.136.245.26. Redistribution subject to AIP license or copyright, see http://ajp.aip.org/ajp/copyright.jsp
Synthesis and characterization of MnTe nanocrystals in glass

D.H. Rodrigues a,b, A.M. Alcalde a,**, N.O. Dantas b

a Grupo de Processamento de Materiais com Laser (GPML), Instituto de Física, Universidade Federal de Uberlândia,
CP 530, CEP 38400-902, Uberlândia-MG, Brazil
b Laboratório de Nossos Materiais Compoundos e Semicondutores (LNMS), Insumo de Física, Universidade Federal de Uberlândia,
CP 530, CEP 38400-902, Uberlândia-MG, Brazil

Available online 1 August 2006

Abstract

The first evidence of the growth of MnTe nanocrystals successfully synthesized in a glass matrix (SiO₂–Na₂CO₃–Al₂O₃–B₂O₃) using the fusion method is reported. Measurements of optical absorption, photoluminescence, electronic paramagnetic resonance and atomic force microscopy were carried out in order to characterize the produced nanocrystals.

© 2006 Elsevier B.V. All rights reserved.

Keywords: Nanocrystals; Band structure; Magnetic properties; Nanoparticles; Absorption

1. Introduction

Nanostructures based in diluted magnetic semiconductors (DMS) – also referred to as semiconductor magnetic or spintronic materials – are a recent trend of research in the scientific community due to their unique magnetic and electronic properties [1-5,12]. The main characteristic of these compounds is the presence of an exchange interaction between the electronic subsystem and the electrons originating in the partially filled d or f layers of the magnetic ions constituting the DMS. This exchange interaction enables the control of the electrical and optical properties of the material by applying external magnetic fields, which are of great interest in spintronics [6,7].

The nanostructures, based on semiconductor compounds of the CdTe₆₋ₓMnₓTe type, are of great interest. For a more accurate analysis of the CdTe₆₋ₓMnₓTe system, an analysis of the magnetic and magneto-optical properties of CdTe and MnTe is essential. The CdTe is a non-magnetic binary system whose properties have been thoroughly analyzed and discussed. On the other hand, studies on the properties of the MnTe compound and its nanostructures are scarce or not satisfactorily detailed. The lack of information is particularly critical in the case of nanostructures based in MnTe, owing to difficulties in applying the growth techniques [10].

Among the growth techniques used in the synthesis of MnTe nanostructures, such as quantum wells and quantum dots, the most successful is the molecular beam epitaxy (MBE) [11]. The high operational cost of this procedure has motivated this group to search for alternative processes of synthesis that have been explored in magnetic and semiconductor materials. In this paper, the first evidence of successful growth of MnTe nanocrystals synthesized in a glass matrix using the fusion method is reported.

2. Experimental results

Manganese-telluride (MnTe) nanocrystals were synthesized in the SnAs glass matrix (SiO₂–Na₂CO₃–Al₂O₃–B₂O₃) and subsequently treated with thermal treatments. The glass matrix used in this study was prepared using
SIO₂ as the glass former and Na₂CO₃ to reduce the melting point. The SNAkB glass matrix, initially doped only with Mn, was melted at 1300 °C for 60 min. After fusion, the resulting melt was submitted to a rapid cooling, bringing it down to room temperature, thus forming a super-cooled liquid (glass). The next step was to pulverize it, followed by a subsequent melting, with the addition of Te as the dopant. The melting points of the Mₐ and Te crystals, respectively, 1245 °C and 449.5 °C. This procedure of melting and recrystallization in order to retain and melt efficiently dopants, with the second fusion carried out at 1000 °C for 30 min. This process is carried out under an atmosphere rich in carbon in order to avoid dopant oxidation and optimize the formation process of binary MₐTe.

The Te was added in excess in comparison to the Mₐ in order to prevent its evaporation. The resulting melt was then quickly cooled down to a temperature close to 0 °C in order to avoid formation of Mn and Te crystals, which are precursors of the MₐTe composition. Finally, due to the glass transition temperature of the SNAkB matrix (doped and undoped) is approximately 550 °C, the glass matrix was submitted to a thermal treatment at 550 °C at different time intervals. This allows for the diffusion of the Mn⁺ and Te⁺⁺ ions, which were created in the fusion process. As a result of the thermal treatment, MnₐTe nanocrystals were formed in the glass matrix. A series of samples was thus produced. In this paper, the analysis was carried out on the following samples, treated at 550 °C for 9 h: SNAkB glass matrix (SNAB), SNAkB doped with Te (SNAkB + Te), SNAkB doped with Mn (SNAkB + Mn), and SNAkB doped with MnₐTe (SNAkB + MnₐTe). The Optical Absorption (OA) spectra were obtained at room temperature using a Varian-500 spectrophotometer operating between 175 and 3300 nm. Low temperature photoluminescence (PL) measurements were carried out. The samples were mounted in a Janis closed cycle cryostat. The spectra were recorded by a Spec 500m single spectrometer. A Coherent: Ar ion laser was used as an excitation source (line 457.9 nm) and the PL signal was detected by a photoacoustic system connected to a thermoelectrically cooled Hamamatsu photo-multiplier. Measurements of electronic paramagnetic resonance (EPR) at room temperature were carried out at 9.5 GHz (microwave frequency). The atomic force microscopy (AFM) images were obtained with a Veeco NanoScope IV MultiMode SPM using the tapping mode.

3. Discussion

The energy gap at room-temperature for the zincblende MnₐTe semimagnetic semiconductor structure is ~2.8 eV (436 nm). In Fig. 1, the OA spectrum of the analyzed samples in the 300–700 nm range can be seen. The spectral lines of the glass matrix without dopants (SNAkB) and doped with Te (SNAkB + Te), did not show an optical absorption band, allowing the assumption that they did not form nano-crystals. On the other hand, OA measurements carried out with SNAkB + Mn and SNAkB + MnₐTe exhibit an absorption band centered at 483 and 485 nm, respectively.

Materials that contain Mn show a well-known emission band in the orange region (580 nm), which is due to the Mn d–d (T₁g – T₂g) transitions [13]. Hence, only the region between 500–600 nm is of interest. Studying the PL at 10 K it is verified that the appearance of an emission band in the region of ~540 nm on the investigated SNAkB + MnₐTe sample can be seen (see Fig. 2), confirming the presence of MnₐTe nano-crystals in this system. The representative lines of PL of the SNAkB and SNAkB + Te samples reveal emission bands very similar to each other, in accordance with the OA measurements.

Room-temperature EPR measurements with the SNAkB + MnₐTe sample reinforce that the state of oxidation of the Mn is 2+ (g-factor = 2.005). In this valence, the Mn is paramagnetic. Although not showed here, the EPR measurements were carried out with the wide set of samples. However, we only detect EPR signals in the Mn-doped samples. This shows that the observed EPR signal is due strictly to the presence of Mn, and not to another compound present in the glass matrix.

![Fig. 1. Room-temperature optical absorption spectra for samples treated at 550 °C for 9 h.](image1)

![Fig. 2. Photoluminescence spectra at 10 K for MnₐTe nanocrystal samples treated at 550 °C for 9 h.](image2)
The formation process of the MnTe nanocrystals is favored when the Mn is in the 2+ valence, since the potential responsible for the nucleation of the MnTe is of the Coulombian type. In II-VI diluted magnetic semiconductors, according to Hund rules, the ground state of the Mn^{2+} ion is six-times degenerated. This leads to the presence of six lines in EPR spectra of nanostructures containing Mn^{2+}. Nevertheless, when Mn ions are too close to one another, their interactions destroy such lines [14]. This is exactly what took place in the EPR measurements for the SNAB + MnTe sample (see Fig. 3). This is due to the high concentration of Mn^{2+} ions in the system, in contrast to the concentration of MnTe nanocrystals.

Atomic force microscopy (AFM) measurements confirmed the formation of MnTe nanocrystals in the SNAB + MnTe sample. The nanocrystals that are present...
in the surface of this material are \( h = 4.0 \) nm in height and have a diameter of approximately \( D = 20.0 \pm 5.0 \) nm. As can be seen in Fig. 4(c), these MnTe nanocrystals grown by the fusion method on the surface of the SnAB + MnTe sample are of a conical shape, just as the MnTe nanocrystals grown by MBE [11].

4. Conclusions

MnTe nanocrystals have been successfully grown on the surface of SnAB (\( \text{SiO}_2 = \text{Na}_2\text{CO}_3 = \text{Al}_2\text{O}_3 = \text{B}_2\text{O}_3 \)) glass matrix through the fusion method. The room-temperature OA and PL measurements showed the electron-hole pair band of MnTe nanocrystals close to the theoretically predicted region. EPR measurements provided evidence that the oxidation state of the Mn ions is \( 2+ \), which favors the synthesis of MnTe nanocrystals. Finally, AFM images confirmed the presence of MnTe nanocrystals of approximately \( D = 20.0 \pm 5.0 \) nm in diameter and \( h = 4.0 \) nm in height, and also confirmed their conical form.

Acknowledgements

We thank Alessandra A. Ribeiro for critical reading of the manuscript and Elisangela Pinho for AFM images. This work was supported by the agencies CNPq and FAPEMIG.

References

Synthesis process controlled magnetic properties of Pb$_{1-x}$Mn$_x$S nanocrystals

R. S. Silva and P. C. Morais
Universidade de Brasília, Instituto de Física, Núcleo de Física Aplicada, Brasília DF 70910-919, Brazil

Fanyao Qu, A. M. Alcalde, and N. O. Santos
Universidade Federal de Uberlândia, Instituto de Física, Uberlândia MG 38400-902, Brazil

H. S. L. Sulliva
Universidade de São Paulo, Instituto de Física, São Paulo SP 05508-090, Brazil and Faculdade de
Tecnologia de São Paulo (UNITEC-SP), Praca do Forno de Fábrica, 56, São Paulo SP 01246-060, Brazil

(Received 27 March 2007; accepted 10 May 2007; published online 22 June 2007)

Mn-doped PbS nanocrystals (NCs) in an oxide glass matrix have been synthesized by the fusion method. Two kinds of Mn$^{2+}$ sites, located inside and on the surface of NCs, are observed by electron paramagnetic resonance (EPR) spectroscopy in the X band and at room temperature. The proportion of their contribution to the hyperfine structure in the EPR spectrum depends strongly on thermal annealing time. The authors illustrate how thermal annealing process manifests itself in engineering the magnetic properties of NCs. © 2007 American Institute of Physics. [DOI: 10.1063/1.2744076]

With the development of magnetic nanostructures, such as diluted magnetic semiconductor (DMS) quantum dots, the control of spin-related phenomena on a nanoscale becomes possible. The ability to incorporate a few magnetic Mn$^{2+}$ ions into a controlled environment, such as nanocrystallites (NCs), would make an important breakthrough in spintronics devices because it allows one to control, manipulate, and detect individual spins, which plays a crucial role in spintronics and quantum information processing. As it is well known, many of the physical properties of nanometer-sized DMS crystallites differ from those of the bulk crystals due to the surface effects and quantum confinement of the electronic states. For instance, the magnetic properties of DMS NCs are markedly enhanced compared to those observed in the bulk phase. Because of the fascinating properties of DMS NCs, they demonstrate a variety of potential applications. Most applications require wide control of magneto-optical properties, which demand precise engineering of the structural and chemical properties of the NCs. Unfortunately, the control of growth of DMS NCs is a formidable task, even for the most sophisticated techniques such as molecular beam epitaxy. Therefore, developing an alternative technique which allows one to synthesize DMS NCs in a controlled way is in great demand. In this letter, we demonstrate the possibility of tailoring magnetic properties of DMS NCs embedded in glass matrix using thermal annealing.

Pb$_{1-x}$Mn$_x$S NCs embedded in an oxide glass matrix were synthesized by the fusion method. The synthesis process proceeds as follows. First, the Mn-doped SiO$_2$-Na$_2$O-Al$_2$O$_3$-PbO-B$_2$O$_3$-S (w) powder was melted in an alumina crucible at 1200 °C for 30 min. Then, it was cooled down to room temperature. After that, thermal annealing treatment proceeded at 500 °C. Finally, spherically shaped Pb$_{1-x}$Mn$_x$S NCs were formed in the glass matrix. In order to study the effects of the synthesis process on the magnetic properties of DMS NCs, four Pb$_{1-x}$Mn$_x$S samples with $x$=0.06, 0.16, 0.26, and 0.36, have been synthesized under different thermal treatments, with annealing times of 2, 4, 8, and 10 h, respectively. Atomic force microscopy was used to analyze the shape morphology and the size dispersion of the Pb$_{1-x}$Mn$_x$S NCs. We found spherical NCs with average diameters (size dispersion of about 6%) of 4.6, 4.7, 4.8, and 4.9 nm in samples Mn$_2$, Mn$_4$, Mn$_6$, and Mn$_8$, respectively. The magnetic states and local structures of Mn$^{2+}$ ions have been examined by EPR spectroscopy in the 9.5 GHz X band and at room temperature.

Thermal treatment process changes the magnetic properties of DMS NCs. Figure 1 shows the EPR spectra of Mn$_2$, Mn$_4$, Mn$_6$, and Mn$_8$. It is noted that each EPR spectrum is composed of two components: the first component located on the higher magnetic field side exhibits two sets of sextet signals superimposed on a broad background, the sextets being attributed to hyperfine interaction between d electrons and the Mn$^{2+}$ ions located at different sites of NCs. The first well resolved sextet is originated from Mn$^{2+}$ ions predominantly present on the NC surface at sites of lower crystal...
field symmetry, whereas the second sextet is stemmed from Mn$^{2+}$ ions in the host NC lattice. The broad component of the EPR spectrum, located on the lower magnetic field side, is attributed to Mn-$\text{Mn}^3$ interactions.

Figure 2 illustrates the evolution of the hyperfine structure as a function of the thermal annealing time. It is noted that the longer the annealing time, the better resolved the hyperfine structure appears. Moreover, for the sample under the lower thermal treatment, such as Mn$_2$O$_3$, the six EPR lines that stemmed from the Mn$^{2+}$ ions located on the NC surface dominate the EPR signal. We also found that with increasing annealing time, the overall background becomes stronger while the EPR intensity change (A) is enhanced, as shown in the inset of Fig. 2. In addition, for the sample Mn$_2$O$_3$, the EPR broad peak turns out to be very intense while dominating the spectrum.

To understand the underlying physics we have performed EPR spectral simulation using anisotropic perturbation theory. The energy levels of a DMS NC with incorporated Mn$^{2+}$ ion ($I=5/2$ and $S=5/2$) are determined by spin Hamiltonian $H_{\text{spin}}=H_{\text{A}}+H_{\text{B}}$. Here $H_{\text{A}}=\sum_{i} S_i^{(A)} \cdot \sum_{i} S_i^{(A)}$ and $H_{\text{B}}=\sum_{i} S_i^{(B)} \cdot \sum_{i} S_i^{(B)}$. The spin Hamiltonian $H_{\text{A}}$ describes all field-independent terms responsible for the zero-field splitting. The first and second terms in $H_{\text{A}}$ stand for the spin-spin interaction between electrons, whereas the third and fourth terms describe nuclear-nuclear spin interaction. For Mn$_2$O$_3$ NCs the constants $P$ and $Q$ are quite small, which can be safely neglected. The last term represents the hyperfine interaction between electron ($e$) and nuclear ($n$) spins, where $A$ is the interaction constant. $H_{\text{B}}$ describes the Zeeman interactions of the electron and nuclear spins with the external magnetic field $B$. Time-dependent perturbation Hamiltonian $H_{\text{B}}$ is a linear function of the microwave field $B_{\text{MW}}$ such that $H_{\text{B}} = H_{\text{B}}\mu_B\mathbf{B}_{\text{MW}}/\hbar$. Here $\mathbf{B}$ is the unitary vector along the orientation of $B_{\text{MW}}$ defined by the Euler angles $\phi$ and $\theta$ as $\mathbf{B} = \sin\theta \cos\phi \mathbf{a} + \sin\theta \sin\phi \mathbf{b} + \cos\theta \mathbf{c}$. The spin magnetic moment of $S_i^{(e)}$ is given by $\mu_S^{(e)} = \mu_B S_i^{(e)}$. The transition rates $\gamma_{ij}$ from spin states $i$ to $j$ depend on the strength and orientation of the microwave field $B_{\text{MW}}$. In standard EPR experiments, $B_{\text{MW}}$ is perpendicular to the static field $B$, i.e., parallel to the $z$ axis of the laboratory frame. Then $\gamma_{ij} = \frac{1}{\hbar} (\mathbf{S}_i \cdot \mathbf{B}_{\text{MW}}) (\mathbf{S}_j \cdot \mathbf{B}_{\text{MW}})$ and the EPR spectra $I_{\text{sp}}$ are governed by $I_{\text{sp}} = I_{\text{sp}}^0 e^{-\gamma_{ij} t}$, where $\gamma_{ij}$ is the frequency-field conversion factor and $I_{\text{sp}}^0$ is the polarization factor, which is proportional to the population difference between two involved states. After some algebra one finds that the allowed transitions obey the following selection rule: $\Delta m_e = \pm 1$, $\Delta m_n = 0$ or $\Delta m_n = \pm 1$, $\Delta m_n = 0$, $\pm 1$, where $m_e$ ($m_n$) stands for the projection of the spin $S_i^{(e)}$ and $S_i^{(n)}$ represents differences of $m_i$ between two transition involved states, $i=S$ or $i$. For most systems, under experimental conditions, only a small fraction of all allowed transitions are observable in an EPR spectrum. For a non-centrosymmetry with long manganese concentration, for example, only transitions associated with $\Delta m_n = \pm 1$ and $\Delta m_n = 0$ are visible, as shown in the inset of Fig. 1. In addition, the interaction constants $A$, $D$, and $E$ depend strongly on the characteristics of the crystal field. For instance, when a Mn$^{2+}$ ion is located close to or on the NC surface, a large structural difference between the NC and the glass matrix results in a larger hyperfine constant $A$ and larger $D$ and $E$ values. Hence the EPR spectrum varies when the local structure of Mn$^{2+}$ ion in the NC changes.

As the thermal annealing time extends the six lines in the hyperfine EPR pattern turn out to be more and more spread out and their intensities increase. It indicates an enhancement in hyperfine interaction. The underlying physics can be understood in the following ways. Firstly, with increasing thermal treatment time, the NCs grow while becoming more and more uniformly distributed in the glass matrix. In addition, the density of NCs increases, accompanied with a reduction of the NC-size dispersion. Hence the effective hyperfine interaction constant $A$ is enhanced. Secondly, with increasing NC size, more Mn$^{2+}$ ions are added into one NC and brought closer together, resulting in diffusion to the surface. An increased proportion of Mn$^{2+}$ ions on the NC surface results in a further enhancement of hyperfine interaction constant $A$.

This analysis is strongly supported by a good agreement between the experimental EPR spectrum and the calculated one, as shown in Fig. 3, which shows the EPR spectra of Mn$_2$O$_3$ measured in the X band and at room temperature (red solid line) and the computed EPR spectra (blue solid line). The calculated spectrum was obtained by a summation of the spectra of Mn$_2$O$_3$ measured in the X band and at room temperature (red solid line) and the computed EPR spectra (blue solid line).
two spectra with \( A=(8.12, 8.12, 9.38) \) (mT) and \( (9.22, 9.22, 9.68) \) (mT), corresponding to Mn\(^{2+}\) sites inside the NC (labeled as \( S_1 \)) and on the NC surface (labeled as \( S_2 \)) for a system with \( S=3/2, I=5/2, D=25 \) mT, \( K=1.1 \) mT, and \( g=(2.055, 2.005, 2.006) \). On the other hand, as the annealing time increases, the probability of magnetic ions inside NCs to occupy neighboring lattice sites and the number of spin-correlated antiferromagnetic clusters increase. It enhances the dipolar interaction and increases the distortion in the Mn\(^{2+}\) sites. Furthermore, accumulation of Mn\(^{2+}\) ions on the NC surface also strengthens Mn-Mn interactions. Consequently, the intensity of the broad background peak increases.

In conclusion, Mn-doped PCS nanocrystals in an oxide glass matrix have been synthesized by the fusion method. Two distinct Mn\(^{2+}\) sites, which are located inside and on the NC surface, are distinguished by EPR spectroscopy in the \( X \) band and at room temperature. The contribution of their proportion to the EPR depends strongly on the thermal treatment process. Increasing annealing time favors diffusion of Mn\(^{2+}\) ions from internal NC sites to the NC surface, because of larger lattice distortion and larger zero-field splitting constant on the surface, the hyperfine interaction, the nuclear quadrupole interaction, as well as the exchange interactions between electron spins are strongly enhanced. Hence the magnetic properties of NCs can be engineered by thermal treatment. We also present how the annealing time manifests itself in the spectral simulation.

The authors gratefully acknowledge the financial support of the Brazilian Agencies CNPq, FAPERJ, and FINEP.

Phonon modulation of the spin-orbit interaction as a spin relaxation mechanism in quantum dots

A. M. Alcalde¹, C. L. Romano², L. Sanz¹ and G. E. Marques³

¹ Instituto de Física, Universidade Federal de Uberlândia, 38400-907 Uberlândia MG, Brazil
² Departamento de Física, Universidade de Buenos Aires, C1428HA, Buenos Aires, Argentina
³ Departamento de Física, Universidade Federal de São Carlos, 13565-905 São Carlos SP, Brazil

E-mail: alcald@fafei.ufscar.br

Abstract. We calculate the spin relaxation rates in a parabolic InAs quantum dots due to the spin interaction with acoustic phonons. We considered the deformation potential mechanism as the dominant electron-phonon coupling in the Pavlov-Firsov spin-phonon Hamiltonian. By studying suitable choices of magnetic field and lateral dot size, we determine regions where the spin relaxation rates can be practically suppressed. We analyze the behavior of the spin relaxation rates as a function of an external magnetic field and mean quantum dot radius. Effects of the spin admixture due to Dresselhaus contribution to spin-orbit interaction are also discussed.

1. Introduction

The ability to manipulate and control processes that involve transitions between spin states is at the moment of extreme importance due to the recent applications in polarized spin electronics and quantum computation. Spin dephasing is the most critical aspect that should be considered in the elaboration of proposals of quantum computation based in single spin states as qubits in quantum dots (QDs) [1]. While for bulk and for 2D systems the spin relaxation processes have been studied in some detail, the problem for QD's still require deeper and further discussions. Several processes that can induce spin relaxation in semiconductors have been identified and were studied. At the moment remains in discussion which, between these processes, is dominant in zero-dimensional systems. Some experimental results have shown good agreement with the theoretical predictions for 2D systems [2] but, in general, the identification of the processes through direct comparison with the experimental results may become a formidable task. This problem is more critical for QDs, since few experimental results exist and the theoretical discussion of the spin relaxation mechanisms is still an open subject. Extensive theoretical works in QD systems have studied the main phonon mediated spin-flip mechanisms, including admixture processes due to spin-orbit coupling [3] and phonon coupling due to interface motion (ripple mechanism) [4]. Spin relaxation rates strongly dependent on the dot size, magnetic field strength, and temperature, as reported by several authors [3, 5]. It was shown that the quantum confinement produces, in general, a strong reduction of the QD relaxation rates.

In this work, we calculate the spin-flip transition rates, considering the phonon modulation by the spin-orbit interaction. For this purpose we will use the spin-phonon interaction Hamiltonian proposed by Pavlov and Firsov [6, 7]. In this model, the Hamiltonian describing the transitions
with spin reversal, due to the scattering of electrons by phonons, can be written in a general form, \( H_{ph} = V_{ph} + \beta \Delta \times \nabla V_{ph} \cdot \sigma (p + eA) \), where \( V_{ph} \) is the phonon operator, \( \sigma \) is the spin operator, \( p \) is the linear momentum operator and \( A \) is the vectorial potential related with the external magnetic field \( \mathbf{B} \). This model has the advantage of being easily adapted to the study of other interaction mechanisms with phonons.

2. Theory

Based on the effective mass theory applied to the problem of the interaction of an electron with lattice vibrations, including the spin-orbit interaction and in presence of an external magnetic field, Pwkov and Frawe [6, 7] have obtained the spin-phonon Hamiltonian that describes the transitions with spin reversal of the conduction band electrons due to scattering with longitudinal lattice vibrations as

\[
H_{ph} = \sum_{\mathbf{k}} \left( \frac{\hbar}{\rho_{ph} V} \right)^{1/2} \left\{ \alpha \sigma_{0} \left[ \begin{array}{cc} 0 & \mathbf{t}_{x} \times \mathbf{t}_{y} \\ \mathbf{t}_{x} \times \mathbf{t}_{y} & 0 \end{array} \right] \left( \frac{\mathbf{p}}{\hbar} \frac{\epsilon_{A}}{\rho_{ph} V} + \alpha \right) + \text{h.c.} \right\},
\]

where, \( \alpha, \beta_{0} \) are annihilation (creation) phonon operators, the magnetic vector potential \( A \) is obtained in the symmetric gauge considering an external magnetic field \( \mathbf{B} \) oriented along the z axis, \( \mathbf{t}_{x} = \hat{x} \times \hat{y} \), where \( \hat{x}, \hat{y} \) are unitary vectors along the \( x \) and \( y \) axis, \( \theta_{0} \) is a unit vectors in the direction of the phonon polarization, \( q \) is the phonon wave vector, \( p \) is the momentum operator, \( V \) is the average sound velocity, \( \rho_{ph} \) is the mass density, \( V \) is the system volume and \( d(q) \) is a coupling constant that depends on the electron-phonon coupling mechanism. Detailed expressions for the parameter \( d(q) \) can be found in Ref. [7].

It has been assumed that the confinement along the \( z \) axis is much stronger than the lateral confinement. Thus, the lateral motion is decoupled from the one along \( z \) and the envelope functions separate \( \psi(r) = f(x, y)\phi(z) \). The \( z \)-dependent part of \( \psi(r) \) is an eigenfunction of a symmetric quantum well of width \( L \). In weakly-coupled two-dimensional systems, the bound states of both electrons and valence-band holes can be understood by assuming a lateral spatial confinement modeled by a parabolic potential with rotational symmetry in the \( x-y \) plane [8], \( V(r) = \frac{1}{2} m \omega_{0} r^{2} \), where \( \omega_{0} \) is the characteristic confinement energy, and \( r \) is the radial coordinate. By using the one-band effective mass approximation and considering an external magnetic field \( B \) applied normal to plane of the QD, the electron lateral wave function can be written as

\[
f_{l}(x, y, \sigma) = C_{n, l, \sigma} \left( \frac{\hbar}{\rho_{ph} V} \right)^{1/2} e^{-\frac{\hbar^{2} \omega_{0}}{2 \rho_{ph} V}} N_{l}^{\sigma}(\rho^{2}/\alpha^{2}) \chi_{\sigma}(x).
\]

where \( C_{n, l, \sigma} = \sqrt{n!/[\sigma!(\sigma + l)!]} \), \( N_{l}^{\sigma} \) is the Laguerre polynomial, \( n \) (\( l \)) is the principal (angular) quantum number, and \( \chi_{\sigma}(x) \) is the spin wave function for the spin variable \( \sigma \). The corresponding eigenenergies are \( E_{n, l, \sigma} = (2\alpha^{2} + 1) \hbar^{2} + \sigma^{2} / m_{\perp} B \), where \( B \) is the magnetic field, \( m_{\perp} \) is the effective length and \( \hbar = \epsilon B / m \). In our model, we also consider the effects of the Dresselhaus contribution that provides additional admixture between spin states. For 2D systems, the linear Dresselhaus Hamiltonian can be written as

\[
H_{D} = \frac{\hbar}{\sqrt{2}} (\sigma_{x} p_{y} - \sigma_{y} p_{y}),
\]

where \( p_{y} = -i \hbar \nabla_{y} + (\alpha/c) A_{y} \) and \( \beta \) is the Dresselhaus coupling parameter for this contribution. If the confinement potential in the \( z \)-direction is considered highly symmetrical, then \( V_{z} \approx 0 \) and the Rashba contribution can be safely ignored.

The spin relaxation rates \( W \) between the electronic states: \( (n, l, \sigma|1) \rightarrow (n', l', \sigma'|1) \), with emission of one acoustic phonon, are calculated from the Fermi golden rule. In the
Hamiltonian (1), we only consider the deformation potential (DP) electron-phonon coupling, this is due to the large $g$-factor in narrow gap InSb ($|g| \sim 51$), the dominant electron-phonon coupling for spin relaxation is the DP mechanism [9]. The piezoelectric (PE) coupling governs the spin relaxation processes in wide or intermediate gap semiconductors. In the transition matrix elements calculation, we do not consider the linear term $\mathbf{R} \cdot \mathbf{r}$ in the expansion of $\exp(i \mathbf{q} \cdot \mathbf{r})$ [3], but the integral representation of Bessel function is used in the evaluation of electron-phonon overlap integrals. The linear approximation of $\exp(i \mathbf{q} \cdot \mathbf{r})$ may be valid for spin inversion transitions in the spin polarized ground-states of GaAs based QDs where, due to the small value of the electron $g$-factor, only long wavelength phonons are involved.

3. Results and discussion
The calculations were performed for a parabolic InSb QD at $T \sim 6$ K. The material parameters for the InSb system are listed in Ref. [10]. We only have considered electron transitions between ground state electron Zeeman levels $(0,0,0) \rightarrow (0,0,1)$ and $(0,1,1) \rightarrow (0,1,0)$. The temperature dependence for one-phonon emission rate is determined from $W = W_0 \Gamma_{ph}(1 + \Gamma_{ph})$, where $\Gamma_{ph}$ is the Bose-Einstein distribution function and $W_0$ is the rate at $T = 0$ K. In the temperature regime $T < 10$ K, we obtain $\Gamma_{ph} \approx 1$ and $W \approx W_0$. For temperatures larger than few Kelvin degrees, two-phonon processes should be considered as the dominant spin relaxation mechanism. These types of processes have not been considered in the present calculation.

![Figure 1](image)

**Figure 1.** Spin relaxation rates, $W$, for a parabolic InSb QD considering the DP coupling mechanism. Panel a) shows $W$ as a function of the magnetic field $B$, for two different electronic transitions and several lateral dot radius $r_0 = 50$, 75, 100, 125, 150, 175, and 200 Å (same $r_0$ ordering for both transitions). b) Contour plot of the spin relaxation rate as a function of $B$ and $r_0$.

In the Figs. 1a) and b) we show the spin relaxation rates due to DP electron-phonon mechanism, as a function of the external magnetic field $B$ and considering some typical values for the effective lateral QD size, $r_0 = \sqrt{\hbar/m_0}$. Some interesting facts about these results should be pointed out: i) The rates show a strong dependence with the magnetic field. This fact can be explained from the dependence of the rates with the transition energy $\Delta E$. In general, we obtain that $W \sim \left| g^* \mu_B B \right|^n \sim (\Delta E)^n$, $n$ being an integer number that depends on the electron-phonon coupling process and $g^*$ the effective $g$-factor. As can be seen in Fig. 1 a), when the magnetic...
field increases, the rates also increase until reaching a maximum near \( B \sim 0.5 \, T \). The position of this maximum is defined from the transition energy conservation: \( E_{\text{opt}} - E_{\text{opt}} = h v \). ii) The oscillatory behavior of the rate, observed for \( B > 0.7 \, T \), are mainly produced by the Dresselhaus spin splitting, which modifies the effective Lande \( g^* \)-factor. As is shown in Fig. 1 a), the \( g^* \)-factor effects are particularly important for the ground-state Zeeman transition. For small magnetic fields, \( g^* \rightarrow g_{\text{Nat}} \), and we may neglect the spin admixture effects. Therefore, the spin relaxation shows no oscillations and becomes almost independent of \( B \). This small \( QD \) size dependence is in agreement with the experimental observations of Gupta and Kikkawa [11]. iii) The rates dependence with the lateral \( QD \) size is related to the interference effects between the spatial and magnetic configurations. This competing effects are contained in the electron-phonon overlap integral, \( f \propto \int \rho \, f_{\text{el},\rho}^{\text{el}}(\rho) \exp(iq \cdot \rho)f_{\text{ph},\rho}(\rho) d\rho \). For large fields, the magnetic confinement causes a gradual decrease in the overlap integral as the \( B \) increases. For small magnetic fields, the spatial confinement is dominant. Thus, when \( B \) diminishes the wavefunctions become more localized and the overlap integral should increase. This effects explain the behavior of the spin transition \((0, 1, 1) \rightarrow (0, 1, 7)\) showed in Fig. 1 a) (red line). The Zeeman ground-state rates (black lines) are strongly dependent on \( \Delta \) and, for small \( B \), the rates are weakly dependent on \( L \). iv) The same rates calculated for GaAs (not showed here), are in general, one order of magnitude smaller than InSb rates. As we expected, the relaxation via PE coupling is more efficient than via the DP phonon processes.

In Fig. 1 b) we have plotted the spin relaxation rates for the ground-state Zeeman transition as a function of \( \Gamma_0 \) and \( B \). We clearly identify a region of strong spin coherence, defined by \( B > 1 \, T \) and \( \Gamma_0 > 100 \, \text{K} \). In this regime, the relaxation times are in the ns order and this is an important feature for spin qubit engineering. In the \( B < 0.1 \, T \) regime, the relaxation times are approximately of few \( \mu \)s. This spin frozen region are not robust against the temperature and will disappear whenever the thermal energy is larger than the spin transition energy.

Acknowledgments
This work has been supported by Fundação de Amparo à Pesquisa do Estado de Minas Gerais (FAPEMIG) and by Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq).

References

A. M. Alcalde
Relatório Final Técnico-Científico
Referências


