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# Effects of Strain and Quantum Confinement in Optically Pumped Nuclear Magnetic Resonance in GaAs: Interpretation Guided by Spin-Dependent Band Structure Calculations

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## Abstract

A combined experimental-theoretical study of optically pumped NMR (OPNMR) has been performed in a GaAs/Al<sub>0.1</sub>Ga<sub>0.9</sub>As quantum well film epoxy bonded to a Si substrate with thermally induced biaxial strain. The photon energy dependence of the Ga OPNMR signal was recorded at magnetic fields of 4.9 and 9.4 T at a temperature of 4.8-5.4 K. The data were compared to the nuclear spin polarization calculated from the differential absorption to spin-up and spin-down states of the electron conduction band using a modified Pidgeon Brown model. Comparison of theory with experiment facilitated the assignment of features in the OPNMR energy dependence to specific interband Landau level transitions. The results provide insight into how effects of strain and quantum confinement are manifested in optical nuclear polarization in semiconductors.

## 1. Introduction

The effects of quantum confinement, magnetic field, and strain on the electronic band structure of III-V semiconductors have been well-studied over the years and are often exploited in device engineering to achieve enhanced or modified operating characteristics [1-9]. Incorporation of these effects into spin-dependent band structure calculations could enable rational design of spintronic devices. Comparison of experimentally measured observables to calculations is critical to the development of advanced theoretical models. Optically pumped NMR (OPNMR) probes the spin dependent band structure because of its sensitivity to the conduction electron spin polarization generated by interband optical absorption.

Here we present an OPNMR study of a 30 nm wide n-doped  $\text{Al}_{0.1}\text{Ga}_{0.9}\text{As}/\text{GaAs}$  quantum well array at high magnetic fields. Optical absorption to spin-up and spin-down conduction electron states is calculated for this structure based on a modified Pidgeon-Brown model [10]. The laser photon energy ( $\hbar\nu_l$ ) dependence of the OPNMR signal for circularly polarized light is compared to theoretical calculations, facilitating assignment of prominent features in the experimental data to specific interband Landau level transitions.

## 2. Background

In the optical nuclear polarization (ONP) effect in GaAs, hyperpolarization of lattice nuclei occurs through spin angular momentum exchange between optically pumped conduction electrons and lattice nuclei. The spin angular momentum exchange is mediated by the Fermi-contact interaction,  $A\hat{\mathbf{I}}\cdot\hat{\mathbf{S}}$ , where  $\hat{\mathbf{I}}$  and  $\hat{\mathbf{S}}$  are the nuclear and electron spin angular momentum operators and  $A$  is the coupling constant. In a high field optically pumped NMR (OPNMR) experiment, the enhanced nuclear spin polarization,  $I_z/I$  (where  $I=3/2$  for  $^{69}\text{Ga}$  and  $^{71}\text{Ga}$ ) generated by ONP is detected by ordinary NMR radio-wave absorption at the Larmor frequency [11-14]. Hyperpolarization by ONP has facilitated OPNMR studies of nuclei in single sub-30 nm wide GaAs quantum wells [11] and arrays of wells [15].

ONP in bulk GaAs has been modeled by a scalar relaxation mechanism in which electron-nuclear spin exchange transitions are mediated by fluctuations in the Fermi contact hyperfine coupling [16, 17]. The initial rate of ONP build-up in this model is proportional to  $S_z - S_{z,eq}$ , i.e. the deviation of the steady-state optically pumped conduction electron spin Zeeman order (which is proportional to the polarization  $S_z/S$ ) from its thermal equilibrium value [16]. Since  $S_z$  is a function of the spin-dependent optical absorption, the photon energy dependence of the

OPNMR signal reflects the spin-dependent band structure in both the valence and conduction bands. The complexity of the band structure at high magnetic fields results in a rich structure in the OPNMR photon energy dependence [11, 15, 18]. Additional complexity is introduced by quantum confinement and strain effects. Here we use computational modeling to compute the conduction band spin dependent absorption coefficients. Based on a literature cross-relaxation model appropriate for ONP in n-doped GaAs [18] we simulate the OPNMR photoexcitation energy dependence and compare it to the experimental data.

OPNMR was previously used to probe the band structure in bulk GaAs.[19, 20] In the work of *Ramaswamy et al.* [20], the photon-energy dependence of OPNMR signals in bulk GaAs at high magnetic field was interpreted by direct comparison of the data to the spin polarization  $S_z / S$  calculated from a modified Pidgeon-Brown model [21]. On this basis, features in the OPNMR energy dependence were assigned to specific interband Landau level transitions. The programming code has since been extended to include effects of both quantum confinement and strain.

Simulation of  $I_z(h\nu_l)$  requires: (i) a calculation of the absorption coefficients  $\alpha^+$  and  $\alpha^-$  for transitions to electron spin-up and spin-down states in the conduction band at a given photon energy and magnetic field, and (ii) an appropriate ONP model relating  $\alpha^+$  and  $\alpha^-$  obtained in the band structure calculations to the nuclear spin polarization,  $I_z / I$ . In the scalar relaxation model for a spin system consisting of one electron coupled to one  $I=3/2$  nucleus [22, 23], ONP at short pumping times (with nuclei initially unpolarized) was modeled by the term

$$I_z \propto \frac{1}{T_1^H} (S_z - S_{z,eq}) \quad (1)$$

where  $1/T_1^H$  is the electron-nuclear cross-relaxation rate constant [16, 24]. ONP in quantum confined GaAs appears consistent with nuclear spin hyperpolarization mediated by Fermi-contact interactions [11, 15, 18, 25, 26].

In the conduction band of a quantum confined semiconductor at high magnetic field,  $T_1^H$  could also vary with the electronic state. While different excited states may be accessed as the photoexcitation energy is varied, ONP occurs in the thermalized state. During thermalization of the electronic states, electron spin relaxation may potentially occur by various mechanisms. The Dyakanov-Perel mechanism, which stems from the lifting of the degeneracy of the conduction

band when  $k \neq 0$  for k-directions other than the principle axes  $\langle 100 \rangle$  and  $\langle 111 \rangle$ , is effectively suppressed at sufficiently high magnetic field [27, 28]. The Elliot-Yaffet mechanism is expected to be very weak since the conduction band states are almost pure spin states [29, 30]. Electron scattering by holes, leading to relaxation by the Bir-Aronov-Pikus mechanism, is not expected to be significant in our n-type GaAs sample since, as discussed below, the photocarrier density is significantly less than the doping density [31]. Therefore, we hypothesize that under our experimental conditions the polarization vector is conserved through the thermalization of the electronic states. The steady-state conduction electron spin polarization depends on the electron spin relaxation time,  $T_{1s}$ , and the excited state lifetime,  $\tau_e$ . If  $T_{1s} \ll \tau_e$ , electron spins thermally equilibrate (i.e.  $S_z \rightarrow S_{z,eq}$ ) and no ONP occurs. The observation of strong OPNMR signals in our GaAs quantum well (QW) sample suggests that  $T_{1s} > \tau_e$ .

We adopt the expression for  $S_z - S_{z,eq}$  derived by Akiba et al. for an n-doped GaAs QW under optical pumping conditions [18]:

$$S_z - S_{z,eq} = \frac{n_l}{n_{eq} + n_l} \frac{S_z^0 - S_{z,eq}}{1 + \tau_e / T_{1s}}. \quad (2)$$

where  $n_l$  is the photoexcited electron density and  $n_{eq}$  is the equilibrium electron density in the absence of optical pumping. The photoexcited electron spin polarization at the instant of optical absorption  $S_z^0$  can be calculated from the energy-dependent spin-up and spin-down absorption coefficients

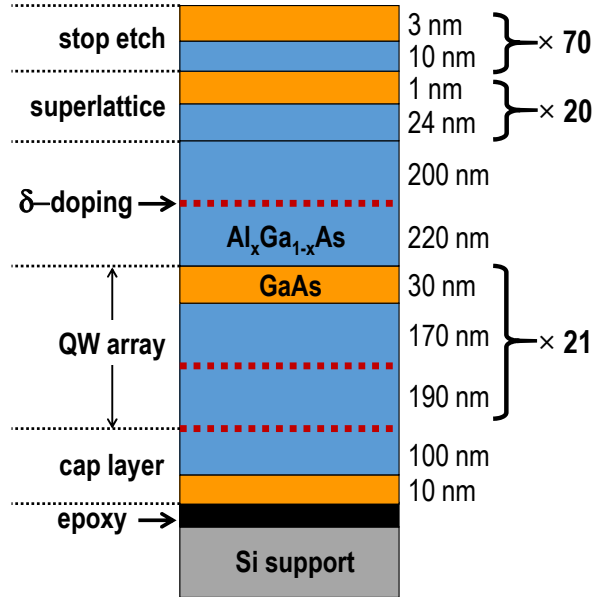
$$S_z^0 = \frac{1}{2} \frac{\alpha^+ - \alpha^-}{\alpha^+ + \alpha^-}. \quad (3)$$

When  $n_l \ll n_{eq}$  and  $T_{1s} > \tau_e$ , Eq. (1) and Eq. (2) can be combined to give

$$I_z(h\nu_l) \propto \frac{1}{T_{1l}} \left[ \alpha^+(h\nu_l) + \alpha^-(h\nu_l) \right] \left[ S_z^0(h\nu_l) - S_{z,eq} \right] \quad (4)$$

where we have used  $n_l \propto \alpha^+ + \alpha^-$ . The nuclear spin polarization  $I_z(h\nu_l)/I$  in an OPNMR experiment depends on the laser photon energy  $h\nu_l$  through the photon energy dependence of the absorption coefficients. For 200 mW/cm<sup>2</sup> at 1.52 eV, the maximum possible generation rate

of photoexcited electrons is  $G = 8 \times 10^{17} \text{ s}^{-1} \text{ cm}^{-2}$ . Assuming an electron lifetime of  $\tau_e = 1 \text{ ns}$  [32], the maximum possible steady-state photoexcited electron density is  $G\tau_e = 8 \times 10^8 \text{ cm}^{-2}$ . Thus, the condition  $n_l \ll n_{eq}$  is satisfied for our QW sample which is delta-doped to a level of  $n_{eq} = 7 \times 10^{10} \text{ cm}^{-2}$ . The cross-relaxation rate  $1/T_1^H$  is a function of the conduction electron density. For example, in bulk n-doped Si,  $1/T_1^H$  is linear in  $n_{eq}$  for  $n_{eq} > 10^{17} / \text{cm}^3$  [33]. For our n-doped quantum wells, where the photo-excited electron density is much lower than the doping density  $n_{eq}$ , we will assume  $1/T_1^H$  to be independent of the optical absorption. In the results presented below, OPNMR signals, which are proportional to  $I_z(h\nu_l)$ , are compared to the quantity  $[\alpha^+(\nu_l) + \alpha^-(\nu_l)][S_z^0(\nu_l) - S_{z,eq}]$  obtained from the electronic band structure calculations. The comparison facilitates assignment of prominent features in the  $I_z(h\nu_l)$  data to specific interband transitions, allowing interpretation of the effects of *strain* and quantum confinement in ONP. We find ONP to be extremely sensitive to relatively small changes in the strain or the pump laser photon energy. Interestingly, multiple changes in the sign of  $I_z(h\nu_l)$  are observed over a rather small range of photon energies. These sign changes are related to strain and quantum confinement effects in the electron band structure.



**Figure 1.** Schematic structure of EA-124, which is composed of layers of GaAs (orange),  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  (blue), and Si- $\delta$ -doping (red dashed lines). The Si- $\delta$ -doping level is  $4 \times 10^{11} \text{ cm}^{-2}$  for the bottom dashed line and  $2 \times 10^{11} \text{ cm}^{-2}$  for middle and top dashed line. The Al composition is  $x=0.1$  in the MQW layers,  $x=0.1 \rightarrow 0.55$  in the superlattice layers, and  $x=0.55$  in the stop etch layers. Thicknesses of each layer are specified to the right of the structure along with the repeat multiple. The entire structure is epoxy-bonded (black) to a Si support (gray).

### 3. Experimental Section

The experiments were performed on a  $\text{Al}_{0.1}\text{Ga}_{0.9}\text{As}/\text{GaAs}$  multiple quantum well (MQW) structure (EA-124, see Fig. 1) consisting of 21 30 nm wide GaAs wells separated by 360 nm wide  $\text{Al}_{0.1}\text{Ga}_{0.9}\text{As}$  barriers with Si- $\delta$ -doping to give a 2D electron density of  $7 \times 10^{10} \text{ cm}^{-2}$ . To eliminate background NMR signals due to the GaAs growth substrate, EA-124 was epoxy bonded to a Si single crystal support and the growth substrate was removed by selective chemical etching following the EBASE technique.[34] Upon cooling the sample to low temperature, differential thermal contraction between the sample and substrate resulted in a uniform biaxial tensile strain in the film, as discussed in detail below. Unless otherwise stated, the static magnetic field is applied parallel to both the  $\langle 100 \rangle$  quantum well growth direction and the direction of electronic confinement.

A copper NMR coil was wound in a rectangular shape with dimensions slightly larger than the sample. Coil windings were placed into grooves cut into the face of a sapphire block and were fixed in place using GE varnish. To mitigate sample heating due to laser irradiation, the sample was mounted in physical contact with the sapphire block inside the NMR coil using Apiezon N grease. The sapphire block was mounted into a homebuilt NMR probe and the NMR coil was incorporated into a tank circuit that could be impedance matched to 50 Ohms and tuned to the nuclear Larmor Frequency of the nucleus of interest at low temperature. The Q-factor of the circuit at the experimental temperature was measured to be 70.

Experiments were performed at a series of different magnetic fields ranging from 3.0 to 9.4 T. For experiments above 3.0 T, the probe was lowered into a 0-17 T variable superconducting cold bore magnet. After inserting the probe into the cold bore, a low pressure (  $\sim 5$  mbar) helium exchange gas was introduced into the bore of the magnet, allowing heat exchange with the magnet's liquid helium dewar, to cool the probe and the sample to 4.2 K. The temperature of the sample space was monitored by a Lakeshore Cernox CX-1030 temperature sensor that was mounted approximately 2 cm from the sample.

OPNMR experiments performed at the National High Magnetic Field Laboratory (NHMFL) utilized a Coherent Verdi V5 pumped Coherent Mira 900 Ti:Sapphire laser with a 0.35 meV linewidth operated in continuous wave mode for all experiments. Laser light from the Ti:Sapphire laser was directed through polarization optics, a polarizing beam splitting cube and

a zero order quarter-waveplate centered at 800 nm, and into the bore of the magnet through an optical window. Optical power was measured using an OPHIR thermopile detector (P/N: 7Z02621) and controlled using a variable neutral density filter. Photon energy was measured using a McPherson 0.75 m pathlength monochromator equipped with a 600 grooves/mm grating and a Princeton Instruments CCD (Mo. No. IN/CCD-1024-UV). For an entrance slit width of 20  $\mu\text{m}$ , the energy resolution of the setup is 0.1 meV at 1530.7 meV.

NMR spectra were acquired at the NHMFL using a homebuilt spectrometer. Individual OPNMR spectra were acquired using a standard saturation-recovery pulse sequence with continuous laser irradiation for the entirety of the pulse sequence. A fixed irradiation time of 120 s was used in all experiments. At a given magnetic field, OPNMR spectra were acquired as the photon energy and polarization were varied. This was repeated at several different magnetic field strengths.

Additional experiments were conducted at 3.0 T and 9.4 T using an Oxford Instruments CF-1200 continuous flow cryostat inserted into the bore of the high homogeneity, room-temperature 89 mm bore superconducting magnets. These studies utilized a Coherent Verdi G 15 W pumped Coherent 899 Ti:Sapphire as an energy tunable photon source. The output beam of the Ti:Sapphire laser was focused into a 600  $\mu\text{m}$  core diameter fiber optic which terminated directly above the sample. Optical power was measured using a Coherent Field Master power meter with a Coherent LM10 detector and controlled using a variable neutral density filter. Photon energy was measured using an Ocean Optics USB2000 spectrometer with an 1800 grooves/mm grating and an entrance slit width of 5  $\mu\text{m}$  giving an energy resolution of 0.4 meV at 1530.7 meV. The NMR probe was fitted with a sample rotation stage incorporating a worm/worm gear pair with a 30:1 reduction for variable tilt angle experiments.

## Theory

The model used here incorporates: (i) quantum confinement effects in the EA-124 QW structure; (ii) *strain* that arises from differential thermal contraction between the  $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$  film and the Si support to which the film is bonded; and (iii) the interaction with the applied magnetic field. The calculations are based on an 8-band Pidgeon Brown (PB) model that was modified and extended to include the effects of quantum confinement and strain. A similar model was used previously to study cyclotron resonance in the dilute magnetic semiconductors InMnAs [10] and cyclotron resonance and magneto-absorption in InSb [21, 35, 36] and InMnSb.[37] Since details are explained in reference [10], only the salient features of



this model as it applies to the strained EA-124 QW structure are outlined here. We use the PB model in the *axial* approximation. In this approximation, the Luttinger parameters  $\gamma_2$  and  $\gamma_3$  are set equal to each other. This allows us to neglect the coupling between PB manifolds and solve each one individually which greatly aids in the calculation. The effective mass Hamiltonian is written as

$$H = H_L + H_Z + H_S + H_C \quad (5)$$

where  $H_L$  is the Landau Hamiltonian,[10]  $H_Z$  is the Zeeman Hamiltonian, and  $H_C$  is the confinement Hamiltonian, which results from differences in the band gap between the wells and barriers. The strain Hamiltonian  $H_S$  is given by [38-40]

$$H_S = \begin{bmatrix} S_a & S_c \\ S_c^\dagger & S_b \end{bmatrix} \quad (6)$$

where

$$S_a = \begin{bmatrix} A_\epsilon & 0 & 0 & 0 \\ 0 & -P_\epsilon - Q_\epsilon & -M_\epsilon & i\sqrt{2}M_\epsilon \\ 0 & -M_\epsilon^* & -P_\epsilon + Q_\epsilon & i\sqrt{2}Q_\epsilon \\ 0 & -i\sqrt{2}M_\epsilon^* & -i\sqrt{2}Q_\epsilon & -P_\epsilon \end{bmatrix} \quad (7)$$

$$S_b = \begin{bmatrix} A_\epsilon & 0 & 0 & 0 \\ 0 & -P_\epsilon - Q_\epsilon & -M_\epsilon^* & i\sqrt{2}M_\epsilon^* \\ 0 & -M_\epsilon & -P_\epsilon + Q_\epsilon & i\sqrt{2}Q_\epsilon \\ 0 & -i\sqrt{2}M_\epsilon & -i\sqrt{2}Q_\epsilon & -P_\epsilon \end{bmatrix} \quad (8)$$

$$S_c = \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & -L_\epsilon & -i\sqrt{\frac{1}{2}}L_\epsilon \\ 0 & L_\epsilon & 0 & i\sqrt{\frac{3}{2}}L_\epsilon^* \\ 0 & -i\sqrt{\frac{1}{2}}L_\epsilon & i\sqrt{\frac{3}{2}}L_\epsilon^* & 0 \end{bmatrix} \quad (9)$$

and

$$A_{\mathcal{E}} = a_c (\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}) \quad (10)$$

$$P_{\mathcal{E}} = -a_v (\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}) \quad (11)$$

$$Q_{\mathcal{E}} = -\frac{b}{2} (\varepsilon_{xx} + \varepsilon_{yy} - 2\varepsilon_{zz}) \quad (12)$$

$$M_{\mathcal{E}} = -\frac{\sqrt{3}}{2} b (\varepsilon_{xx} - \varepsilon_{yy}) + i \frac{2\sqrt{3}}{3} d \varepsilon_{xy} \quad (13)$$

and

$$L_{\mathcal{E}} = id (\varepsilon_{xz} - i\varepsilon_{yz}) \quad (14)$$

Here,  $\varepsilon_{ij}$  are the components of the strain tensor and  $a_c$ ,  $a_v$ ,  $b$ , and  $d$  are deformation potentials, which for GaAs can be found in Ref [41]. For the case of biaxial tensile strain,  $\varepsilon_{xx} = \varepsilon_{yy}$  and  $\varepsilon_{ij} = 0$  for  $i \neq j$ , giving  $M_{\mathcal{E}} = L_{\mathcal{E}} = 0$ . Hence,

$$S_a = S_b = \begin{bmatrix} A_{\mathcal{E}} & 0 & 0 & 0 \\ 0 & -P_{\mathcal{E}} - Q_{\mathcal{E}} & 0 & 0 \\ 0 & 0 & -P_{\mathcal{E}} + Q_{\mathcal{E}} & i\sqrt{2}Q_{\mathcal{E}} \\ 0 & 0 & -i\sqrt{2}Q_{\mathcal{E}} & -P_{\mathcal{E}} \end{bmatrix} \quad (15)$$

and

$$S_c = 0. \quad (16)$$

To model quantum confinement effects in the QW structure with an effective mass Hamiltonian, we replace the  $z$ -component of the wavevector  $k_z$  with a differential operator  $k_z \rightarrow -i \nabla_z$  along the confinement direction ( $\hat{z}$ ). We then divide the superlattice unit cell into  $G$  equally spaced grid points so that the  $\nabla_z$  operator can be approximated by finite differences on the grid. In addition, all the material parameters are allowed to vary with position along  $\hat{z}$ . We use a finite difference approach on each PB manifold, allowing us to calculate all the Landau levels for each

quantum confined subband. However, this increases the size of the matrix to be diagonalized from  $8 \times 8$  (size of the effective mass Hamiltonian matrix without grid points) to  $8G \times 8G$ . In the present work, we choose  $G=101$ . The  $808 \times 808$  matrix is diagonalized numerically to obtain Landau subband energies and corresponding wavefunctions. Increasing the number of grid points did not significantly change the calculated band structure.

We solve the eigenvalue equations [10]

$$H_p F_{p,v} = E_{p,v}(k_z) F_{p,v} \quad (17)$$

for the eigenvalues and eigenvectors using the eight Bloch basis states consisting of the spin-up/spin-down conduction band (*CB*), light hole (*LH*), heavy hole (*HH*), and split-off hole (*SO*) states. In Eq. (17),  $p$  is the Pidgeon-Brown manifold index and has allowed values of  $p \geq -1$  while  $v$  labels the eigenstates in the  $p^{\text{th}}$  manifold in ascending order. Solutions are labeled as  $BS_{sb}^p \uparrow$ , where *BS* corresponds to the Bloch basis state (see above) that dominates the character of the eigenvector,  $p$  is the PB manifold index, *sb* is the subband number, and the arrow indicates the “spin state” of the Bloch basis state. Note that the “spin-up” and “spin-down” correspond to the maximum and minimum  $m_j$  values for the type of basis states. For instance,

$$|HH \uparrow\rangle = \left| j = \frac{3}{2}; m_j = \frac{+3}{2} \right\rangle \text{ is the heavy hole up state;}$$

$$|LH \downarrow\rangle = \left| j = \frac{3}{2}; m_j = \frac{-1}{2} \right\rangle \text{ is the light hole down state.}$$

We note that  $|HH \uparrow\rangle$  is a *pure* spin state (i.e eigenstate of  $S_z$ ) while  $|LH \downarrow\rangle$  is not a *pure* spin state. It is important to note that the PB manifold number is *not* the same as the Landau level (LL) number. Within a given PB manifold, the LL number depends on the basis state. For instance, by looking at Eq. (27) of reference [10], we see that in the  $p^{\text{th}}$  PB manifold, the heavy-hole down state is in the  $p+1$  LL harmonic oscillator state, while the conduction band up state is in the  $p-1$  state and the light hole down state is in the  $p$  state, etc. The LL number  $n$  can be calculated from the PB index  $p$  using the relations listed in Table I. We note that, while within a given PB manifold, the sum of the  $n$  and the  $m_j$  quantum number of the Bloch basis state is a constant given by  $n + m_j = p - 1/2$ .

At a magnetic field  $B_z$ , the photon energy dependent absorption coefficients,  $\alpha(h\nu_l)$ , are obtained from the imaginary part of the dielectric function, which is calculated using Fermi's golden rule.[10] The absorption coefficient is given by

$$\alpha(h\nu_l) = \frac{e^3 B_z}{h\nu_l (\hbar c)^2 n_r} \sum_{p,v,p',v'} \int_{-\infty}^{\infty} dk_z \left| \hat{e} \cdot \bar{P}_{p,v}^{p',v'}(k_z) \right|^2 \times (f_{p,v}(k_z) - f_{p',v'}(k_z)) \delta(\Delta E_{p,v}^{p',v'}(k_z) - h\nu_l) \quad (18)$$

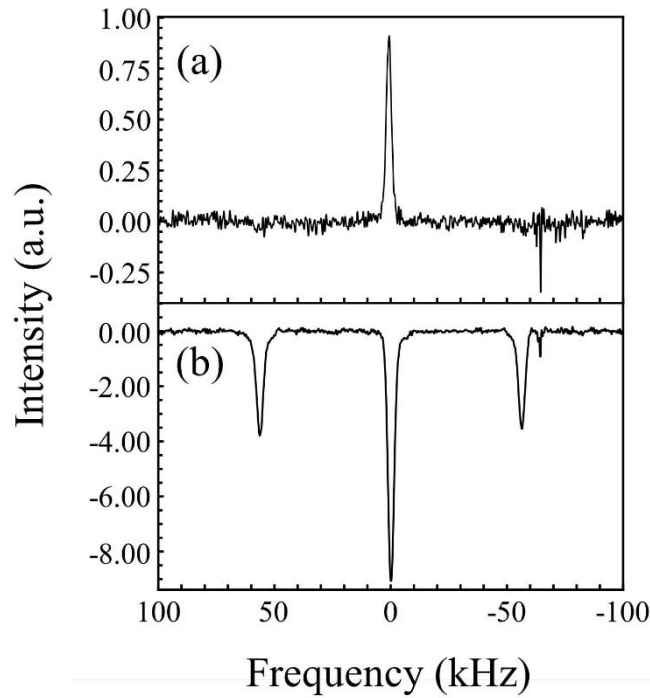
where  $n_r$  is the index of refraction,  $\hat{e} \cdot \bar{P}_{p,v}^{p',v'}$  are the optical matrix elements (see Ref [10]),  $f_{p,v}$  is the Fermi distribution,  $\hat{e}$  is the optical polarization vector and  $\Delta E_{p,v}^{p',v'}$  is the optical transition energy. Separate absorption coefficients were calculated for transitions generating spin-up and spin-down conduction band electrons. This is accomplished by limiting the sum in Eq. 18 to just one of the given conduction band spins. (We note that the conduction band – valence band mixing in GaAs is weak, and as a result, the conduction band Landau levels are over 99% of a given spin type). Using these spin-dependent absorption coefficients, the energy dependence of  $S_z^0 - S_{z,eq}$  can be calculated using Eq. (3) and the energy dependence of  $I_z(h\nu_l)$  can be calculated using Eq. (4). Within the axial approximation, absorption of  $\sigma^+$  circularly polarized light occurs only between PB manifolds which differ by +1, i.e.  $p \rightarrow p+1$  is the selection rule for  $\sigma^+$  polarized light. For  $\sigma^-$  circularly polarized light, the selection rule is  $p \rightarrow p-1$ .

**TABLE I.** Relationship between PB index  $p$  and LL number  $n$  for the given basis states.

Basis State	Relation
$CB \uparrow$	$n = p - 1$
$CB \downarrow$	$n = p$
$LH \uparrow$	$n = p - 1$
$LH \downarrow$	$n = p$
$HH \uparrow$	$n = p - 2$
$HH \downarrow$	$n = p + 1$
$SO \uparrow$	$n = p - 1$
$SO \downarrow$	$n = p$

#### 4. Results and Discussion

Shown in Fig. 2 are the thermally polarized and optically pumped Ga-71 NMR spectra of EA-124 at 4.2 K and 9.4 T. Upon cooling, strain is induced due to differential contraction of the MQW film and the Si support to which it is epoxy bonded. Due to Poisson's law, the biaxial tensile strain in the plane of the sample (the  $xy$ -plane) produces an associated strain along the  $z$ -axis,  $\epsilon_{zz}$ . Strain along the  $z$ -axis produces an electric field gradient along the  $z$ -axis,  $V_{zz}$ , leading to a quadrupole splitting of the nuclear spin states which has been previously reported in the OPNMR spectrum of EA-124 [15].



**Figure 2.** (a) Thermally polarized  $^{71}\text{Ga}$  NMR spectrum of EA-124 at 4.2 K and 9.4 T acquired immediately after cooling the sample. (b)  $^{71}\text{Ga}$  OPNMR spectrum of EA-124 at 4.2 K, acquired using  $\tau_{\text{ONP}} = 50$  s.

The *thermally* polarized NMR spectrum exhibits a single line because it is dominated by the NMR signal from nuclei located in the  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  regions of the structure;  $>90\%$  of Ga nuclei are in  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  regions of EA-124. The single resonance in the thermally polarized spectrum is assigned to the central transitions of  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ . Satellite transitions from the  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  barriers are not observed due to first-order quadrupole broadening resulting from the alloy disorder in the  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  material. Similar observations are reported for  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  for  $x = 0.1$

[42] and  $x = 0.5$ . [43] Because optical absorption occurs only within the GaAs quantum wells, ONP hyperpolarizes nuclear spins *in the quantum well region*. The nuclear spin hyperpolarization diffuses from the quantum well nuclei into the  $\text{Al}_{0.1}\text{Ga}_{0.9}\text{As}$  barriers of EA-124. Thus, our OPNMR spectrum contains signal contributions from both the GaAs quantum wells and the  $\text{Al}_{0.1}\text{Ga}_{0.9}\text{As}$  barriers. As previously reported, *nuclei in the quantum well region* of EA-124 exhibit a strain-induced nuclear quadrupole splitting, which is observable because (1) ONP produces large NMR signal enhancements and (2) there is no alloy disorder within the well. [15] The central and satellite transitions in the OPNMR spectrum in Fig. 2b are in phase. The linear phase correction applied to obtain the spectrum in Fig. 2b was validated by setting the spectrometer frequency on resonance with each of the three transitions and then applying the same zero order phase correction as was applied to the thermally polarized signal.

Using the quadrupole moment of the nucleus and the observed quadrupole splitting in the OPNMR spectrum, we obtain  $V_{ZZ} = 1.45 \times 10^{13} \text{ erg} \cdot \text{statC}^{-1} \cdot \text{cm}^{-2}$  and  $\epsilon_{zz} = -7.7 \times 10^{-4}$  from the Ga-71 OPNMR spectrum (see Appendix 1). Similarly,  $V_{ZZ} = 1.25 \times 10^{13} \text{ erg} \cdot \text{statC}^{-1} \cdot \text{cm}^{-2}$  and  $\epsilon_{zz} = -6.6 \times 10^{-4}$  is obtained from the Ga-69 OPNMR spectrum. To validate our calculation of  $\epsilon_{zz}$ , we compare the strain in EA-124 to that of a 1.1  $\mu\text{m}$  thick GaAs epitaxial liftoff (ELO) sample bonded to Si [44]. Photoluminescence measurements from the ELO sample reveal a valence band splitting (between the light and heavy holes) of 4.2 meV. To relate the valence band splitting to strain, we use equations for the transition energy, to first order, of the light hole-to-conduction band,  $\Delta E_{lh}$ , and heavy hole-to-conduction band,  $\Delta E_{hh}$ , under biaxial strain,  $\epsilon = \epsilon_{xx} = \epsilon_{yy}$ , which are [45, 46]

$$\Delta E_{lh} = \left( -2a \frac{C_{11} - C_{12}}{C_{11}} + b \frac{C_{11} + 2C_{12}}{C_{11}} \right) \epsilon \quad (19)$$

and

$$\Delta E_{hh} = \left( -2a \frac{C_{11} - C_{12}}{C_{11}} - b \frac{C_{11} + 2C_{12}}{C_{11}} \right) \epsilon \quad (20)$$

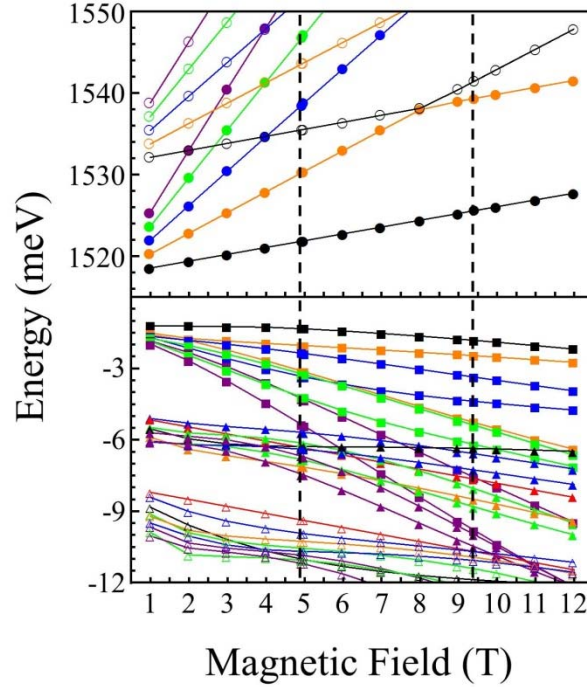
Thus, the relationship between the valence band splitting,  $\Delta E_{lh} - \Delta E_{hh}$ , and  $\epsilon$  is

$$\Delta E_{lh} - \Delta E_{hh} = 2b \frac{C_{11} + 2C_{12}}{C_{11}} \epsilon \quad (21)$$

Using values of  $12.1 \times 10^{12}$  dynes cm<sup>-2</sup> [47],  $5.48 \times 10^{12}$  dynes cm<sup>-2</sup> [47], and -2.0 eV [48] for  $C_{11}$ ,  $C_{12}$ , and  $b$ , respectively, we find that a strain of  $\epsilon = 5.5 \times 10^{-4}$  would produce a 4.2 meV valence band splitting. Finally,  $\epsilon_{zz}$  is given by [45]

$$\epsilon_{zz} = -\frac{2C_{12}}{C_{11}}\epsilon \quad (22)$$

Using Eq. (22), we calculate  $\epsilon_{zz} = -5.0 \times 10^{-4}$  in the ELO sample. The slightly smaller value of  $\epsilon_{zz}$  in the ELO sample is expected because it was bonded to Si at room temperature, while EA-124 was bonded to Si at a temperature of 100 °C. The slightly higher bonding temperature results in a larger strain upon cooling the sample.



**Figure 3.** Calculated magnetic field dependence of conduction band (top) energies for  $CB_1$  (●),  $CB_2$  (○) and the valence band (bottom) energies for  $LH_1$  (■),  $HH_1$  (▲), and  $HH_2$  (▲) in EA-124. The dotted vertical lines are 4.9 T and 9.4 T. Colors correspond to PB indices of -1 (red), 0 (black), 1 (orange), 2 (blue), 3 (green), and 4 (purple).

The strain estimated from the quadrupole splitting ( $\epsilon_{zz} = -7 \times 10^{-4}$ ) was used as input into the calculations of the magnetic-field dependence of the band structure from 1 to 12 T (see Fig. 3). Only the  $CB_1$ ,  $CB_2$ ,  $LH_1$ ,  $HH_1$ , and  $HH_2$  subbands are displayed because they contribute most

significantly to the optically pumped electron spin polarization. First, we note that the gap between the valence and conduction bands is 1519.8 meV at 1T. There are 3 different contributions which shift the gap in different directions. Tensile strain narrows the gap. By comparing calculations with and without strain, we estimate this contribution to be about 5 meV in our sample. Conversely, both confinement and magnetic field increase the gap. Because the gap at 1 T is only slightly larger than that of bulk GaAs, the combination of confinement and magnetic field effects is estimated to be slightly larger in magnitude than that of strain.

Next, we note that as expected in the case of tensile strain, the  $LH_1$  states are higher in energy than the  $HH_1$  states. At 1 T, the valence band splitting is about 3.9 meV. This value is smaller than would be expected due to strain alone and is likely due to different quantum confinement shifts for the  $LH_1$  states as compared to the  $HH_1$  states. At 1 T, we also observe a 13.6 meV splitting between  $CB_1^0$  and  $CB_2^0$  and a 3.0 meV splitting between  $HH_1^{-1}$  and  $HH_1^{-2}$ , which are predominantly due to quantum confinement. The different splittings at 1, 4.9, and 9.4 T are summarized in Table II.

Note that the Coulomb interaction was not included in the band structure calculations. While a thorough treatment of Coulomb effects in strained and doped quantum wells in a magnetic field is beyond the scope of the present work, we briefly discuss some expected effects of the Coulomb interaction on a qualitative level. The Coulomb interaction between the photo-excited electrons and holes leads to exciton formation. For excitons in bulk GaAs in a magnetic field  $B_z$ , Coulombic interactions produce a shift in the absorption edge that depends on the ratio of the cyclotron and Coulomb binding energies,  $2\beta$ , with[49]

$$\beta = \frac{\varepsilon^2 \hbar^2 B_z}{\mu^2 e^3 c} \quad (23)$$

where, in Eq. (23),  $\varepsilon$  is the absolute permittivity,  $\hbar$  is the reduced Planck's constant,  $\mu$  is the reduced mass of the exciton,  $e$  is the electron charge, and  $c$  is the speed of light. Though EA-124 is quite different than bulk GaAs, we expect that at 4.9 T, we are in a regime of  $\beta > 1$  where magnetic effects begin to exceed Coulomb effects. Typical exciton binding energies in bulk GaAs are about 5 meV. In a quantum confined system at high magnetic field, the exciton binding energies can be larger. In our current system, strain changes the ordering of the heavy and light hole energies, further complicating calculation of exciton binding energy. More work is required to quantitatively evaluate  $\beta$  and solve for the shift in the absorption edge in EA-



124. To account for Coulombic shifts of the absorption edge at a given magnetic field, the entire energy scale of the absorption coefficient calculations was shifted by a fixed amount. We have neglected any dependence of Coulombic effects on Landau level number, though in a real system this can be important. Coulombic effects are expected to be strongest near the absorption edge and become weaker at higher photon energies.

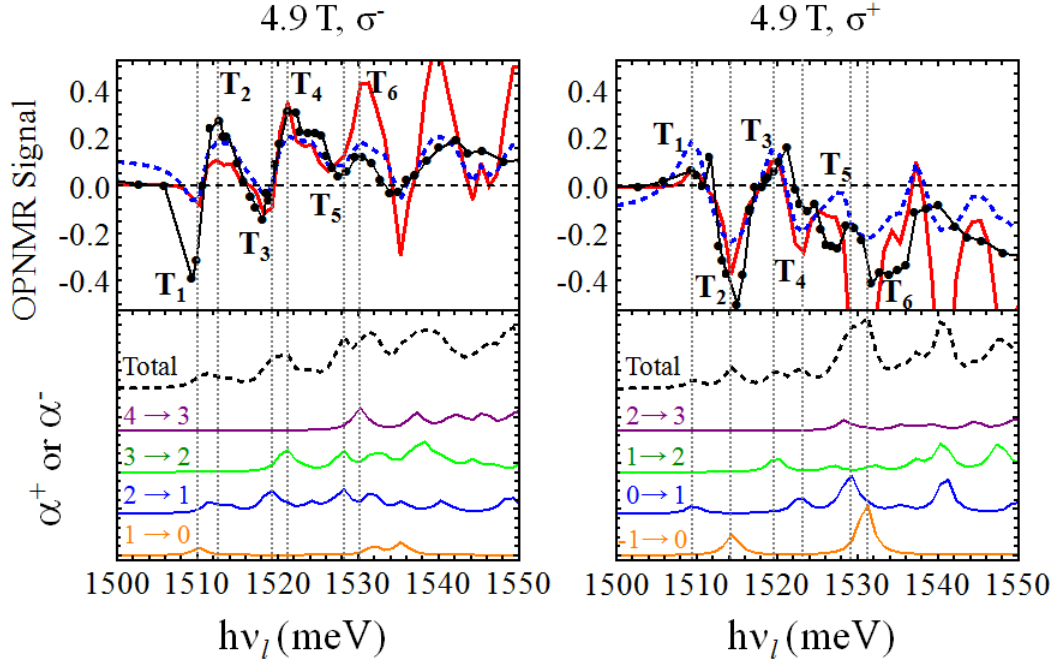
**TABLE II.** Summary of relevant energy splittings determined from the calculated band structure of EA-124 for fields of 1, 4.9 and 9.4 T. Energies are given in meV.

	1 T	4.9 T	9.4 T
	Gap		
$LH_1^0 \rightarrow CB_1^0$	1519.8	1523.3	1527.4
	Confinement		
$CB_1^0 \rightarrow CB_2^0$	13.6	13.7	15.9
$HH_1^{-1} \rightarrow HH_2^{-1}$	3.7	3.7	4.1
	Strain		
$LH_1^0 \rightarrow HH_1^{-1}$	3.9	4.3	4.8

The dependence of the OPNMR signal (the peak area of the central transition) on photon energy for  $\sigma^-$  and  $\sigma^+$  light at 4.9 T is plotted in Fig. 4. In the top panels,  $I_z(h\nu_{laser})$  obtained using Eq. (4) and  $S_z^0$  are superimposed on the experimental OPNMR data. The bottom panels present the PB manifold-resolved (i.e. separated by transitions from  $p$  to  $p'$ ) absorption coefficients, also at 4.9 T. This aids us in identifying the transitions. The energy scale for the calculations at 4.9 T is shifted by 13.7 meV. Prominent features are labeled according to the transition that dominates at that energy. The transitions are summarized in Table III, along with the subbands involved in each transition and the composition of the states in terms of the eight Bloch basis states (i.e. the band mixing). For  $\mathbf{T}_6$  with  $\sigma^-$  polarization, the spin label is omitted because the transition results from the  $HH\uparrow$  character of the initial state even though it is predominantly a  $LH\downarrow$  state.

The calculated absorption coefficients vary rapidly with photon energy in a strained QW at high field, which explains the rapid oscillations in  $I_z(h\nu_l)$ . Reasonably good agreement is obtained between theory and experiment, particularly in the predicted positions of the maxima and minima. However, not all features in the data are captured by the theory. With increasing photon energy, the amplitude of the oscillations in the calculated  $I_z(h\nu_l)$  steadily increases

while in the data these oscillations are damped. At the higher photon energies, the data show a closer match to  $S_z^0$ . This might be due to one or more of the following factors: penetration depth effects, non-linear dependence of the OPNMR signal with absorption at some photon energies, and effects of sample heating (such as reduction of  $T_{1s}$  and/or  $\tau_e$ ).



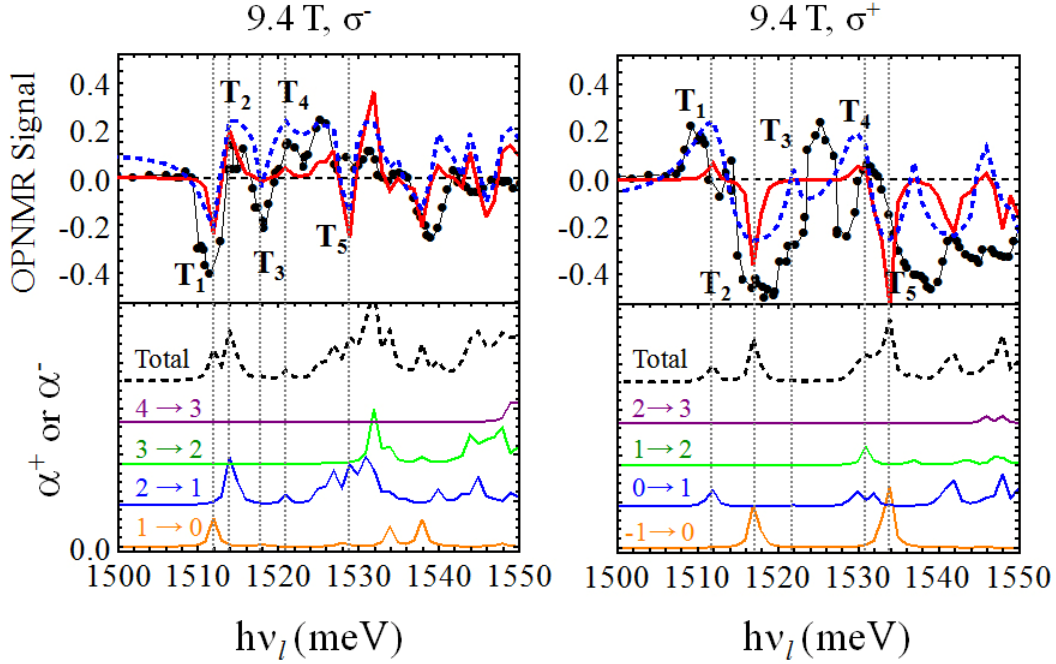
**Figure 4.** Upper panels: Photon energy dependence of the experimental OPNMR data  $T=5.1 \pm 0.3$  K (black points, arbitrary units),  $I_z(h\nu_l)$  calculated using Eq. (4) (red curve, arbitrary units), and  $S_z^0$  (dashed blue curve) and at 4.9 T for  $\sigma^-$  and  $\sigma^+$  light. Lower panels: Decomposition of the total optical absorption coefficients according to PB manifold, as indicated. Grey dotted vertical lines mark prominent features in the data, which are labeled according to transition,  $T_i$ .

The OPNMR photon energy dependence data and corresponding calculations at 9.4 T are shown in Fig. 5. The energy scale for all calculated quantities has been shifted by 16.1 meV. The increase in the shift, compared to 4.9 T, reflects larger Coulombic effects at higher magnetic field. The dominant transitions at 9.4 T are listed in Table IV along with the subbands involved in the transitions and their decomposition in terms of probability densities of the basis states. Similar to the 4.9 T data, there are several transitions ( $T_2$  and  $T_3$  for  $\sigma^-$  and  $T_3$  for  $\sigma^+$ ) where the spin label has been omitted from the valence band subband descriptor because the

transition involves the eigenstate with a spin state that differs from that of the dominant eigenstate in the linear combination.

**TABLE III.** Transitions, labels, subband labels, and wavefunction probabilities for 4.9 T.

Transition	Label	Subband Label	Wavefunction Probability
$\sigma^-$	$T_1$	$LH_1^1 \uparrow$	89% $LH \uparrow$ + 8% $HH \downarrow$
		$CB_1^0 \downarrow$	99.6% $CB \downarrow$
	$T_2$	$HH_1^2 \uparrow$	33% $HH \uparrow$ + 15% $HH \downarrow$ + 13% $LH \downarrow$ + 38% $LH \uparrow$
		$CB_1^1 \uparrow$	99.7% $CB \uparrow$
	$T_3$	$LH_1^2 \uparrow$	77% $LH \uparrow$ + 13.3% $HH \uparrow$
		$CB_1^1 \downarrow$	99% $CB \downarrow$
$\sigma^-$	$T_4$	$HH_1^3 \uparrow$	35% $HH \uparrow$ + 57% $LH \downarrow$
		$CB_1^2 \uparrow$	99% $CB \uparrow$
	$T_5$	$HH_1^2 \uparrow$	33% $HH \uparrow$ + 15% $HH \downarrow$ + 13% $LH \downarrow$ + 38% $LH \uparrow$
		$CB_2^1 \uparrow$	99% $CB \uparrow$
		$LH_1^3 \uparrow$	74% $LH \uparrow$ + 15% $HH \uparrow$
		$CB_1^2 \downarrow$	98.8% $CB \downarrow$
$\sigma^+$	$T_1$	$LH_1^4 \rightarrow$	53.5% $LH \downarrow$ + 36.7% $HH \uparrow$
		$CB_1^3 \uparrow$	99% $CB \uparrow$
	$T_2$	$LH_1^0 \downarrow$	92% $LH \downarrow$
		$CB_1^1 \uparrow$	99% $CB \uparrow$
	$T_3$	$HH_1^{-1} \downarrow$	100% $HH \downarrow$
		$CB_1^0 \downarrow$	99.6% $CB \downarrow$
$\sigma^+$	$T_4$	$LH_1^1 \downarrow$	85% $LH \downarrow$ + 14% $HH \downarrow$
		$CB_1^2 \uparrow$	99.3% $CB \uparrow$
	$T_5$	$HH_1^0 \downarrow$	45% $HH \downarrow$ + 54% $LH \downarrow$
		$CB_1^1 \downarrow$	99% $CB \downarrow$
		$HH_2^0 \downarrow$	71% $HH \downarrow$ + 28% $LH \downarrow$
		$CB_1^1 \downarrow$	99% $CB \downarrow$
$\sigma^+$	$T_6$	$LH_1^2 \downarrow$	58.5% $LH \downarrow$ + 34.5% $HH \uparrow$
		$CB_1^3 \uparrow$	99% $CB \uparrow$
	$T_6$	$HH_2^{-1} \downarrow$	100% $HH \downarrow$
		$CB_2^0 \downarrow$	99% $CB \downarrow$



**Figure 5.** Upper panels: Photon energy dependence of experimental OPNMR data at  $T=5.1 \pm 0.3$  K (black points, arbitrary units),  $I_z(h\nu_l)$  calculated using Eq. (4) (red curve, arbitrary units), and  $S_z^0$  (blue dashed curve) at 9.4 T for  $\sigma^-$  and  $\sigma^+$  light. Lower panels: Decomposition of the total optical absorption coefficients according to PB manifold, as indicated. Grey dotted vertical lines mark prominent features in the data, which are labeled according to transition,  $T_i$ .

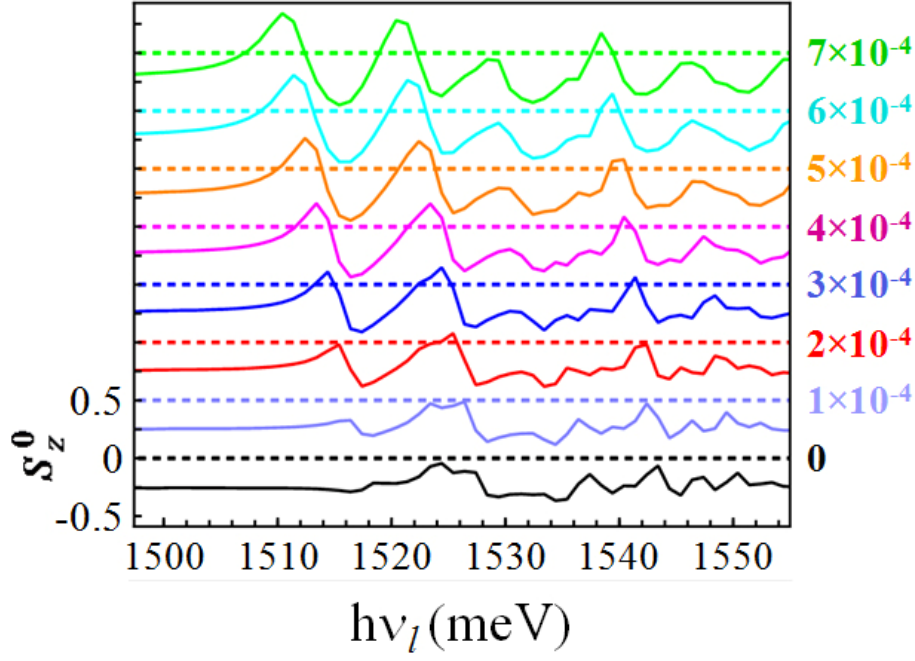
Our results show that the OPNMR photon energy dependence is highly sensitive to both confinement and strain effects. For example, the  $T_2$  and  $T_6$  for  $\sigma^+$  and  $T_2$  and  $T_5$  for  $\sigma^-$  excitation involve transitions to different confinement subbands of the conduction band. An obvious effect of strain is that *the lowest energy transition for both polarizations of light is from a LH initial state*. In the absence of strain, the lowest energy transition would be expected to originate from a HH initial state.

Strain effects are seen to be significant in EA-124, producing a LH/HH splitting of  $\sim 4$  meV. This is noteworthy because, near the band edge in unstrained bulk GaAs in zero magnetic field, the LH/HH degeneracy limits  $S_z$  to  $S_z = \pm 1/4$ , [14] which in turn limits the maximum achievable nuclear spin polarization. In the case of EA-124, our calculations show that it is possible to selectively excite a LH or HH to CB transition, resulting in fully spin polarized optically pumped electrons. Furthermore, these calculations reveal that the optically pumped

$S_z$  can be switched from  $S_z^0 = +1/2$  to  $S_z^0 = -1/2$  with a change in photon energy of less than 6 meV at 9.4 T. To demonstrate the effect of strain on the photon energy dependence of electron  $S_z$ , Fig. 6 presents a plot of  $S_z$  versus photon energy for seven different values of strain for EA-124 at 4.9 T. The calculations show that increasing the tensile strain shifts  $S_z(h\nu_l)$  to lower energies and results in more pronounced features and larger oscillations in  $S_z$  with photon energy.

**TABLE IV.** Transitions, labels, subband labels, and wavefunction probabilities for 9.4 T.

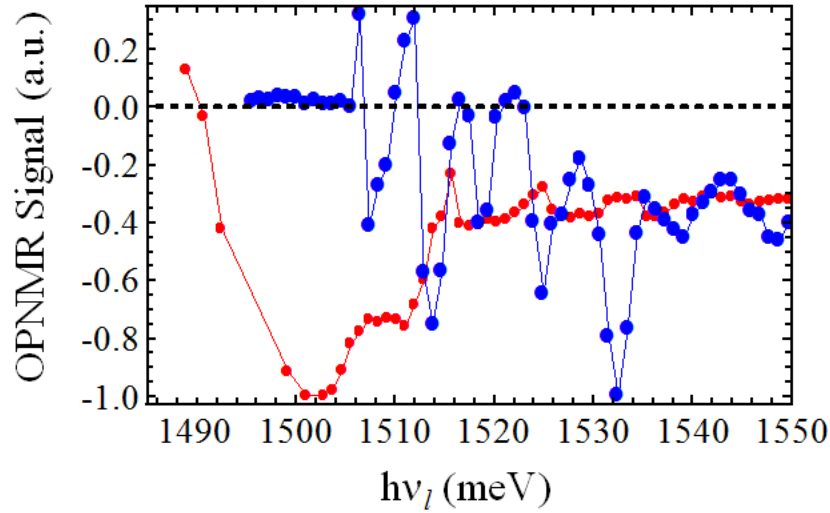
Transition	Label	Subband Label	Wavefunction Probability
$\sigma^-$	$T_1$	$LH_1^1 \uparrow$	86% $LH \uparrow$ + 10% $HH \downarrow$
		$CB_1^0 \downarrow$	99.4% $CB \downarrow$
	$T_2$	$LH_1^2 \uparrow$	58% $HH \uparrow$ + 28% $LH \downarrow$ + 14% $LH \uparrow$
		$CB_1^1 \uparrow$	99.8% $CB \uparrow$
	$T_3$	$HH_1^1 \uparrow$	60% $LH \downarrow$ + 30% $HH \downarrow$ + 9% $LH \uparrow$
$\sigma^+$	$T_3$	$CB_1^0 \downarrow$	99.4% $CB \downarrow$
		$HH_2^2 \uparrow$	48% $HH \uparrow$ + 30% $LH \downarrow$ + 20% $HH \downarrow$
	$T_4$	$CB_1^1 \uparrow$	99.5% $CB \uparrow$
	$T_5$	$LH_1^1 \uparrow$	69% $LH \uparrow$ + 23% $HH \uparrow$ + 7.5% $CB \downarrow$
		$CB_1^1 \downarrow$	98.7% $CB \downarrow$
	$T_1$	$LH_1^0 \downarrow$	92% $LH \downarrow$ + 8% $HH \downarrow$
		$CB_1^1 \uparrow$	99.5% $CB \uparrow$
	$T_2$	$HH_1^{-1} \downarrow$	100% $HH \downarrow$
		$CB_1^0 \downarrow$	99.4% $CB \downarrow$
	$T_3$	$HH_2^0 \uparrow$	44% $LH \downarrow$ + 56% $HH \downarrow$
		$CB_1^1 \uparrow$	98.8% $CB \uparrow$
$\sigma^+$	$T_4$	$LH_1^1 \downarrow$	74% $LH \downarrow$ + 17% $HH \downarrow$
		$CB_1^2 \uparrow$	98.8% $CB \downarrow$
	$T_5$	$HH_2^{-1} \downarrow$	100% $HH \downarrow$
		$CB_2^0 \downarrow$	99% $CB \downarrow$



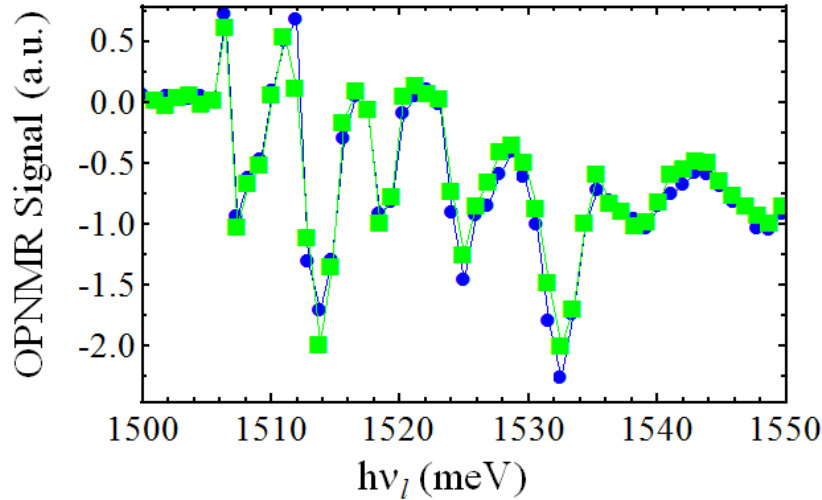
**Figure 6.** Calculated photon energy dependence of  $S_z^0(h\nu_l)$  at 4.9 T for  $\sigma^+$  light value for the values  $\epsilon_{zz}$  given to the right of the plot.

In Fig. 7, we compare the photon energy dependence of OPNMR signals from EA-124 to that of a bulk GaAs sample. Comparison of the photon energy dependence of these two samples demonstrates the dramatic difference between the band structure of a strained and quantum confined semiconductor and that of a bulk semiconductor. Notably, no inversions of the signal in the bulk material are observed, indicating that in the case of bulk GaAs the optically pumped electron spin polarization is always greater than the thermal equilibrium electron spin polarization. This is in contrast to the data acquired from EA-124, where multiple signal inversions are observed, indicating that the sign of  $S_z^0(h\nu_l) - S_{z,eq}$  can be reversed. The difference between the bulk (unconfined) semiconductor and the quantum confined semiconductor can be attributed to fundamental difference in the density of states of the 2D and 3D system in high magnetic field. In the 3D system,  $k_z$  is a good quantum number. As a result, in 3D systems, the heavy and light hole density of states overlap due to the tail extending to the continuum in  $k_z$ . On the other hand, the density of states in the 2D system is fully discrete, which allows the pumping of transitions from the heavy and light hole bands to be resolved, resulting in an inversion in the sign of the electron spin polarization.

Finally, in Fig. 8 we present the photon energy dependence of the OPNMR signal at 3.0 T for 2 different orientations of the sample with respect to the magnetic field. The OPNMR profile of EA-124 is shown to be *insensitive* to tilt angle for small tilt angles, indicating that Landau level filling factor effects do not contribute significantly under the present experimental conditions, presumably because of the high ambient temperature ( $\sim 4.2$  K).



**Figure 7.** OPNMR photon energy dependence of EA-124 (blue circles) and bulk GaAs (red circles) at 3.0 T using unpolarized light. Data has been normalized to the largest OPNMR signal for each data set.



**Figure 8.** OPNMR photon energy dependence of EA-124 at 3.0 T using unpolarized light at oriented angles of  $0^\circ$  (blue circles) and  $6^\circ$  (green squares) with respect to the magnetic field. Angles refer to the orientation of the growth direction with respect to the magnetic field.

## 5. Conclusions:

To summarize, the strain in a MQW was estimated from the quadrupole splitting observed in the OPNMR spectrum and used as an input parameter in calculations of the MQW's band structure at 4.9 and 9.4 T. From the band structure, photon energy dependent absorption coefficients to the spin-up and spin-down states of the conduction band are calculated, yielding the electron spin polarization. Using the spin-dependent absorption coefficients, two different quantities are calculated and compared to the experimental OPNMR photon energy dependence. A shift of the energy scale had to be included in the calculations to account for excitonic effects. We find that for an n-doped quantum well, Eq. (4) provides a reasonable match to the OPNMR data. Deviations could be due to one or more of the following: penetration depth effects, where the optical intensity is not homogeneous across the quantum well array, non-linear dependence of the OPNMR signal on absorption at some photon energies, energy dependent spin relaxation, which have been assumed to be unimportant under our experimental conditions, and effects of sample heating (such as reduction of  $T_{1s}$  and/or  $\tau_e$ ). Further work is obviously necessary to reach a quantitative match between theory and experiment.

With the aid of theory, we are able to assign specific features in the OPNMR photon energy dependence to specific transitions in the band structure. Comparing data acquired from EA-124 to that of bulk GaAs shows that the OPNMR photon energy dependence is strongly influenced not only by quantum confinement effects, but also by strain. Experiments in a tilted field suggest that at these temperatures, the data is insensitive to filling factor effects. Finally, we find that the strained quantum well exhibits a heavy hole/light hole splitting of  $\sim 4$  meV. This separates the light hole band from the heavy hole bands and as a result, can substantially change the conduction band spin polarization and hence the nuclear polarization. Calculations show this splitting can be used to increase the maximum possible optically-induced electron spin polarization. Application of strain to remove degeneracies in the band structure could be crucial in potential applications such as in spintronics or in quantum computation which demand initialization of a pure state.

## Acknowledgements

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## 6. Appendix

At sufficiently high magnetic field, the frequencies of the single quantum transitions  $m \rightarrow m-1$  of a quadrupolar nucleus are given by

$$\Delta \nu_{m \rightarrow m-1} - \nu_0 = \frac{3(2m-1)eQ}{8I(2I-1)h} V_{zz} (3\cos^2 \theta - 1) \quad (\text{A-24})$$

where  $\nu_0$  is the nuclear Larmor frequency,  $Q$  is the quadrupole moment of the nucleus,  $h$  is Planck's constant,  $V_{zz}$  is the component of the EFG along the  $z$ -axis in the principal axis frame of reference, and  $\theta$  is the angle between the  $z$ -axis of the principal axis frame and the magnetic field. For  $I = 3/2$ , the three single quantum transition frequencies, to first order, are

$$\Delta \nu_{+3/2 \rightarrow +1/2} - \nu_0 = \frac{eQ}{4h} V_{zz} (3\cos^2 \theta - 1), \quad (\text{A-25})$$

$$\Delta \nu_{+1/2 \rightarrow -1/2} - \nu_0 = 0, \quad (\text{A-26})$$

$$\Delta \nu_{-1/2 \rightarrow -3/2} - \nu_0 = -\frac{eQ}{4h} V_{zz} (3\cos^2 \theta - 1). \quad (\text{A-27})$$

The quadrupole splitting  $\Delta \nu_Q$  is defined as

$$\Delta \nu_Q = \Delta \nu_{+3/2 \rightarrow +1/2} - \Delta \nu_{+1/2 \rightarrow -1/2} = \Delta \nu_{+1/2 \rightarrow -1/2} - \Delta \nu_{-1/2 \rightarrow -3/2} \quad (\text{A-28})$$

Hence, we can infer the component of the EFG from the measured quadrupole splitting:

$$V_{zz} = \frac{4h}{eQ} \Delta \nu_Q (3\cos^2 \theta - 1)^{-1}. \quad (\text{A-29})$$

The differential thermal contraction between the Si substrate and the GaAs films is assumed to produce a biaxial tensile strain. In this case, the  $z$ -principal axis of the EFG tensor will be parallel to both the growth direction and the applied magnetic field (i.e. perpendicular to the samples). Hence, the field gradient tensor elements in the principal axis system,  $V_{ij}$ , are equivalent to those of the lab frame,  $V_{ij}$ , and  $\theta = 0$ , so Eq. (A-29) reduces to:

$$V_{zz} = \frac{2h}{eQ} \Delta v_Q. \quad (\text{A-30})$$

The elements  $V_{ij}$  are related to the elastic strains,  $\epsilon_{ij}$ , by the gradient-elastic tensor  $S'_{ijkl}$ , [50]

$$V_{IJ} = V_{ij} = \sum_{k,l} S'_{ijkl} \epsilon_{kl}. \quad (\text{A-31})$$

Neglecting  $\epsilon_{kl}$  for  $k \neq l$  strains, this reduces to

$$\begin{bmatrix} V_{xx} \\ V_{yy} \\ V_{zz} \end{bmatrix} = \begin{bmatrix} S'_{11} & S'_{12} & S'_{12} \\ S'_{12} & S'_{11} & S'_{12} \\ S'_{12} & S'_{12} & S'_{11} \end{bmatrix} \begin{bmatrix} \epsilon_{xx} \\ \epsilon_{yy} \\ \epsilon_{zz} \end{bmatrix} \quad (\text{A-32})$$

where we have used  $S'_{11} = S'_{22} = S'_{33}$  and  $S'_{12} = S'_{13} = S'_{23} = S'_{21} = S'_{31} = S'_{32}$  for GaAs. [50]  
From Eq. (A-32),

$$V_{zz} = V_{zz} = S'_{12} \epsilon_{xx} + S'_{12} \epsilon_{yy} + S'_{11} \epsilon_{zz}. \quad (\text{A-33})$$

We note that the stresses, and therefore the strains, are equal along the  $x$ - and  $y$ -axes, giving:

$$V_{zz} = V_{zz} = 2S'_{12} \epsilon_{xx} + S'_{11} \epsilon_{zz} = -S'_{11} \epsilon_{xx} + S'_{11} \epsilon_{zz}. \quad (\text{A-34})$$

We have used the fact that  $2S'_{12} = -S'_{11}$ . [50] The strains along the  $x$ - and  $z$ -axes can be related using the compliance tensor,  $S_{ij}$ . For an isotropic material where  $\epsilon_{kl} = 0$  for  $k \neq l$ ,

$$\begin{bmatrix} \epsilon_{xx} \\ \epsilon_{yy} \\ \epsilon_{zz} \end{bmatrix} = \begin{bmatrix} S_{11} & S_{12} & S_{12} \\ S_{12} & S_{11} & S_{12} \\ S_{12} & S_{12} & S_{11} \end{bmatrix} \begin{bmatrix} \sigma_{xx} \\ \sigma_{yy} \\ \sigma_{zz} \end{bmatrix}. \quad (\text{A-35})$$

In the case of equal stress  $\sigma$  along the  $x$ - and  $y$ -axes (and no stress along the  $z$ -axis), the strains are related to the stress by

$$\epsilon_{xx} = \epsilon_{yy} = S_{11} \sigma + S_{12} \sigma \quad (\text{A-36})$$

and

$$\epsilon_{zz} = 2S_{12}\sigma . \quad (\text{A-37})$$

From (A-36) and (A-37),

$$\sigma = \frac{\epsilon_{xx}}{S_{11} + S_{12}} = \frac{\epsilon_{zz}}{2S_{12}} \quad (\text{A-38})$$

This gives the following relation between  $\epsilon_{xx}$  and  $\epsilon_{zz}$

$$\epsilon_{xx} = \frac{S_{11} + S_{12}}{2S_{12}} \epsilon_{zz} \quad (\text{A-39})$$

Finally, substituting Eq. (A-34) and (A-39) into (A-30) and rearranging:

$$\epsilon_{zz} = \frac{2h}{eQ} \left( 1 - \frac{S_{11} + S_{12}}{2S_{12}} \right)^{-1} \frac{1}{S'_{11}} \Delta \nu_Q. \quad (\text{A-40})$$

Numerical values for the constants, quadrupole splitting, and calculated strain using Eq. (A-40) are given in Table A-1.

**TABLE A.1.** Values used to calculate  $\varepsilon_{zz}$  from the Ga-71 and Ga-69 nuclear quadrupole splitting for EA-124.

Parameter	Value	Units	Reference
$S_{11}$	$1.17^b$	$10^{-12} \text{ cm}^2 \text{ dyn}^{-1}$	[51]
$S_{12}$	$-0.37^b$	$10^{-12} \text{ cm}^2 \text{ dyn}^{-1}$	[51]
$S'_{11}$	$-9.1^c$	$10^{15} \text{ statC cm}^{-3}$	[52]
<b>Ga-71</b>			
$Q$	$1.07^a$	$10^{-25} \text{ cm}^2$	[53]
$\Delta\nu_Q$	56	$10^3 \text{ Hz}$	
$\varepsilon_{zz}$	-7.6	$10^{-4}$	
<b>Ga-69</b>			
$Q$	$1.99^a$	$10^{-25} \text{ cm}^2$	[53]
$\Delta\nu_Q$	90	$10^3 \text{ Hz}$	
$\varepsilon_{zz}$	-6.6	$10^{-4}$	

## 7. References

- [1] C. Gmachl *et al.*, Reports on Progress in Physics **64**, 1533 (2001).
- [2] T. Chakraborty, and V. M. Apalkov, Advances in Physics **52**, 455 (2003).
- [3] B. S. Williams, Nature Photonics **1**, 517 (2007).
- [4] A. Wade *et al.*, Nature Photonics **3**, 41 (2009).
- [5] G. Scalari *et al.*, Physical Review Letters **93**, 4 (2004).
- [6] G. Scalari *et al.*, Physical Review B **76**, 7 (2007).
- [7] D. Smirnov *et al.*, Physical Review B **66**, 4 (2002).
- [8] Y. Sun, S. Thompson, and T. Nishida, Journal of Applied Physics **101** (2007).
- [9] H. Morkoc, B. Sverdlov, and G. B. Gao, Proceedings of the Ieee **81**, 493 (1993).
- [10] G. D. Sanders *et al.*, Physical Review B **68**, 19 (2003).
- [11] S. E. Barrett *et al.*, Physical Review Letters **72**, 1368 (1994).
- [12] S. E. Hayes, S. Mui, and K. Ramaswamy, Journal of Chemical Physics **128**, 16 (2008).
- [13] J. A. Reimer, Solid State Nuclear Magnetic Resonance **37**, 3 (2010).
- [14] F. Meier, and B. Zakharchenya, *Optical Orientation* (Elsevier, Amsterdam, 1984).
- [15] S. A. Vitkalov *et al.*, Physical Review B **61**, 5447 (2000).
- [16] P. L. Kuhns *et al.*, Physical Review B **55**, 7824 (1997).
- [17] C. R. Bowers, Solid State Nuclear Magnetic Resonance **11**, 11 (1998).
- [18] K. Akiba *et al.*, Physical Review B **87**, 7 (2013).
- [19] S. Mui *et al.*, Physical Chemistry Chemical Physics **11**, 7031 (2009).
- [20] K. Ramaswamy *et al.*, Physical Review B **82** (2010).
- [21] M. B. Santos *et al.*, Physics Procedia **3**, 1201 (2010).
- [22] A. Abragam, *Principles of Nuclear Magnetism* (Oxford University Press, 1961).
- [23] D. Paget, Physical Review B **25**, 4444 (1982).
- [24] A. Patel *et al.*, Physical Review B **60**, R5105 (1999).
- [25] R. Tycko *et al.*, Science **268**, 1460 (1995).
- [26] S. E. Barrett *et al.*, Physical Review Letters **74**, 5112 (1995).
- [27] M. I. Dyakonov, and V. I. Perel, Soviet Physics Jetp-Ussr **33**, 1053 (1971).
- [28] M. I. Dyakonov, and V. I. Perel, Soviet Physics Solid State,Ussr **13**, 3023 (1972).
- [29] R. J. Elliott, Physical Review **96**, 266 (1954).
- [30] Y. Yafet, Solid State Physics-Advances in Research and Applications **14**, 1 (1963).
- [31] G. L. Bir, A. G. Aronov, and G. E. Pikus, Zhurnal Eksperimentalnoi I Teoreticheskoi Fiziki **69**, 1382 (1975).

- [32] H. Okamura *et al.*, Physical Review B **58**, 15985 (1998).
- [33] R. G. Shulman, and B. J. Wyluda, Physical Review **103**, 1127 (1956).
- [34] M. V. Weckwerth *et al.*, Superlattices and Microstructures **20**, 561 (1996).
- [35] X. Pan *et al.*, 2012), pp. 84611P.
- [36] W. Gempel *et al.*, in *Narrow Gap Semiconductors 2007*, edited by B. Murdin, and S. Clowes (Springer Netherlands, 2008), pp. 213.
- [37] G. A. Khodaparast *et al.*, Physical Review B **88**, 11 (2013).
- [38] G. E. Pikus, and G. L. Bir, Soviet Physics-Solid State **1**, 1502 (1960).
- [39] G. L. Bir, and G. E. Pikus, *Symmetry and Strain-Induced Effects in Semiconductors* (Wiley, New York, 1974).
- [40] C. CHAO, and S. CHUANG, Physical Review B **46**, 4110 (1992).
- [41] I. Vurgaftman, J. Meyer, and L. Ram-Mohan, Journal of Applied Physics **89**, 5815 (2001).
- [42] V. L. Berkovits, and V. I. Safarov, Fizika Tverdogo Tela **20**, 2536 (1978).
- [43] C. Degen *et al.*, Physical Review B **69** (2004).
- [44] C. M. N. Mateo *et al.*, Journal of Applied Physics **101** (2007).
- [45] H. Asai, and K. Oe, Journal of Applied Physics **54**, 2052 (1983).
- [46] F. H. Pollak, and M. Cardona, Physical Review **172**, 816 (1968).
- [47] R. I. Cottam, and G. A. Saunders, Journal of Physics C-Solid State Physics **6**, 2105 (1973).
- [48] W. Stolz, F. E. G. Guimaraes, and K. Ploog, Journal of Applied Physics **63**, 492 (1988).
- [49] R. J. Elliott, and R. Loudon, Journal of Physics and Chemistry of Solids **15**, 196 (1960).
- [50] E. F. Taylor, and N. Bloembergen, Physical Review **113**, 431 (1959).
- [51] S. Adachi, Journal of Applied Physics **58**, R1 (1985).
- [52] R. K. Sundfors, Physical Review B **10**, 4244 (1974).
- [53] P. Pyykko, Molecular Physics **99**, 1617 (2001).