# Spin-flip Raman scattering in $GaAs/Al_xGa_{1-x}As$ multiple quantum wells

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Strong spin-flip-related Raman scattering (SFRS) from $p$-type Be-doped $GaAs/Al_xGa_{1-x}As$ multiple quantum wells (MQW's), as well as from an undoped sample, has been observed. In both cases the SFRS exhibits a strong dependence on the geometry of the experiment. A theory is developed that explains SFRS in $p$-type MQW's as related to a spin flip of a hole bound to an acceptor via exchange interaction with a neighboring exciton. SFRS in the undoped MQW's is explained as a flip of the angular momentum of an exciton localized by interface roughness via interaction with acoustic phonons (doubly resonant scattering by acoustic phonons). The $g$ tensor of the hole bound to the acceptor is determined to be $g_{\parallel}=+2.3$, $g_{\perp}\cong 0$, and that of the localized exciton $(g_h-g_e)_{\parallel}=+1.5$, $(g_h-g_e)_{\perp}\cong 0$ for the narrowest wells measured ($\sim 40$ Å).

## I. INTRODUCTION

Most of the past investigations of spin-flip Raman scattering (SFRS) have been devoted to studies of electron-hole exchange interactions in bulk materials. SFRS has been investigated experimentally and theoretically in bulk CdS (Refs. 1 and 2) and in $p$-type-InSb.[3,4] These investigations gave useful information about exchange interactions and $g$ factors of impurities and free carriers. The SFRS technique has also been successfully used to study exchange interactions in bulk semimagnetic semiconductors,[5–8] and in $CdTe/Cd_{1-x}Mn_xTe$ quantum-well (QW) structures.[9] Thus it seems reasonable to apply SFRS to investigate electron-hole exchange interactions and to determine the $g$-factor tensors of impurity states in $GaAs/Al_xGa_{1-x}As$ QW structures. It is expected that confinement of carriers in QW's will produce a noticeable increase in the SFRS efficiency with respect to the bulk material because such confinement leads to an increase in the exchange interaction[10,11] and oscillator strength of the exciton[12,13] participating in the process. However, up to now only techniques such as magnetophotoluminescence[14] and optically detected magnetic resonances[15] have been used for investigations of electron and hole splittings in a magnetic field and determination of their $g$ factors in quantum wells.

In this paper we report the observation of SFRS in $GaAs/Al_xGa_{1-x}As$ multiple quantum wells (MQW's). Strong SFRS was observed in $p$-type Be-doped as well as in undoped MQW's. In the doped MQW two SFRS lines are seen. One of them is interpreted as due to spin flip of a hole bound to a neutral acceptor via exchange interaction with neighboring excitons. The other is also observed in the undoped MQW's. It is interpreted as the flip of the angular momentum of a localized exciton via interaction with an acoustical phonon (doubly resonant scattering by acoustic phonons). In both cases the SFRS exhibits a cosine dependence of the Raman shift on the angle between the direction of the magnetic field and the [100] growth axis of the MQW. This implies that the $g$ factors of a hole bound to a neutral acceptor and of a localized exciton are strongly anisotropic. The values of the longitudinal $g$ factors are found to be $g_{h\parallel}=2.3$ and $g_{\mathrm{ex}\parallel}=(g_h-g_e)=1.5$ for a hole bound to an acceptor and a localized exciton, respectively, in the narrowest QW's measured, slightly lower for wider QW's. The transverse $g$ factors are equal to zero in both cases within experimental accuracy. The reason for this anisotropy is the reduction of symmetry in (100)-grown MQW's from $T_d$ to $D_{2d}$ which leads to a splitting of the acceptor and exciton states and, as a result, to the strong $g$-factor anisotropy. Recently, an anisotropy of the individual electron and hole $g$ factors has also been reported for type-II GaAs/AlAs quantum wells.[15]

We have found that the SFRS efficiency has a strong temperature dependence similar to that expected for an activated process. This leads to the conclusion that the SFRS process takes place via localized states of the exciton. The effect of exciton localization on the resonant Raman scattering intensity in MQW's has been studied recently.[16] It was shown that resonant Raman scattering is directly related to the exciton dephasing time, which is longer for localized states.

The present work shows that the SFRS technique can be successfully used for the investigation of exchange interaction in quantum wells and possibly also for the determination of the geometrical nature of impurities and localized excitons. It would also be of interest to apply the technique to the study of the electron-hole exchange in systems with dimensionality lower than two.

## II. EXPERIMENTAL

The samples investigated were grown by molecular-beam epitaxy on (100)-oriented undoped semi-insulating

GaAs substrates kept at 580 °C. The growth rates were 1.0 monolayer/s for GaAs and 1.5 monolayer/s for $Al_xGa_{1-x}As$. To confine the Be doping to the GaAs quantum-well region only, two monolayers of GaAs (≅5.6Å) next to every GaAs/$Al_xGa_{1-x}As$ interface were left undoped as "spacer." The sample parameters, listed in Table I, were determined by double-crystal x-ray diffraction using Cu $K\alpha_1$ radiation and by Hall-effect measurements.

The SFRS experiments were carried out in magnetic fields up to 10 T in backscattering Faraday geometry, i.e., the propagation direction of incident and scattered light was normal to the (100) plane of the sample and parallel to the magnetic field. Excitation took place with circularly polarized light while the scattered light was analyzed with circular polarizers. The samples were mounted in an optical exchange-gas cryostat with variable temperature in the range of 4–77 K. For the excitation of SFRS a tunable Ti-sapphire laser pumped by an $Ar^+$-ion laser was used. The scattered light was analyzed by a SPEX 1404 double monochromator equipped with a cooled GaAs photomultiplier and conventional photon counting electronics.

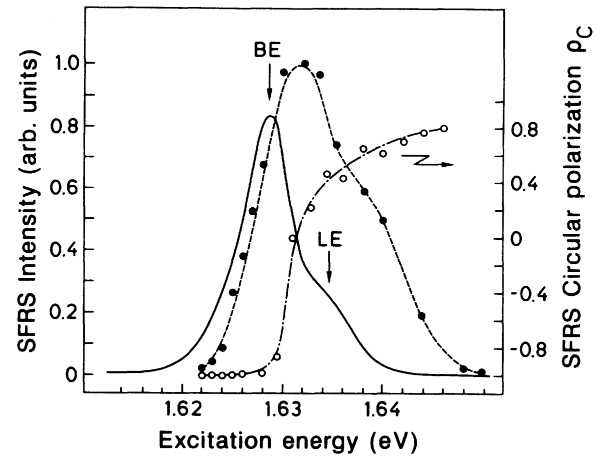


FIG. 1. Resonance profile of the SFRS efficiency for the $z(\sigma^-,\sigma^+ + \sigma^-)\bar{z}$ configuration in a magnetic field $B=10$ T (dashed line, full dots). Photoluminescence spectrum of the MQW 46/110 also for $B=10$ T and excitation at $\hbar\omega=1.7$ eV (solid line). The circles and dashed-dotted line show the dependence of the circular polarization $\rho_c$ on excitation energy ($\rho_c=[I(\sigma^-,\sigma^-)-I(\sigma^-,\sigma^+)]/[I(\sigma^-,\sigma^-)+I(\sigma^-,\sigma^+)]$, where $I(\sigma^-,\sigma^-)$ and $I(\sigma^-,\sigma^+)$ are the intensities of the Raman lines measured in $(\sigma^-,\sigma^-)$ and $(\sigma^-,\sigma^+)$ configurations).

## III. EXPERIMENTAL RESULTS

The SFRS experiments discussed in this paper were performed by exciting in resonance with the luminescence band of excitons bound to neutral acceptors in Be-doped QW's or with the luminescence band of excitons localized to interface roughness in the undoped QW's.[17] The observed SFRS spectra exhibited circular polarization characteristics. To describe this polarization we use the notation $z(\sigma^\eta,\sigma^\lambda)\bar{z}$, where $z$ and $\bar{z}$ correspond to the direction of the exciting and scattered light, respectively. $(\sigma^\eta,\sigma^\lambda)$, with $\eta,\lambda=\pm$, denotes the circular polarization of the exciting $\sigma^\eta$ and scattered $\sigma^\lambda$ light. Here the sign $\eta$ or $\lambda$ is determined by the sign of the projection of the angular momentum of the photons on the propagation direction of the *exciting* light ($z$ direction).

### A. Doped quantum wells

Figure 1 shows the photoluminescence (PL) spectrum of a Be-doped GaAs/$Al_xGa_{1-x}As$ MQW 46/110 (where 46 is the thickness of the wells and 110 that of the barriers in Å; we use this notation throughout) excited with a photon energy larger than the QW band gap. Only the PL spectra of the exciton bound to acceptor (BE) and localized on roughness (LE) are presented. A very weak (100 times smaller than the BE line) band of free-electron to acceptor recombination was also observed at 1.606 eV (this line is not displayed in Fig. 1). For excitation in the region of this PL band we observed in a magnetic field two Stokes and two anti-Stokes narrow lines (see Fig. 2). Further investigations of doped and undoped QW's showed that the line with smaller Raman shift (LE line) is also typical for the relatively narrow ($L_z \leq 100$ Å) undoped QW's. In this section we focus on the line with the larger Raman shift (*H* line), which was observed only in Be-doped QW's and thus is assigned to spin flip of holes bound to the acceptors. The Raman shifts of the Stokes and anti-Stokes lines are equal and depend linearly on the magnetic field (see Fig. 3, curve 1). The width of SFRS lines in all Be-doped MQW's is $\Delta \cong 0.6$ cm$^{-1}$, and should be determined mainly by inhomogeneous broadening of the acceptor states due to their interaction with the walls of the QW (the spacer in the Be-doped QW's was smaller than the Bohr radius of the acceptor). The Raman shift (at $B=10$ T) reveals a weak dependence on the QW width. It increases from 9.5 cm$^{-1}$ for $L_z=100$ Å to 11

TABLE I. Parameters of the GaAs/$Al_xGa_{1-x}As$ MQW samples and the measured $g$ factors of excitons and holes.

| Sample | Well-barrier width (Å) | $x$ | Doping $\times 10^{16}$ cm$^{-3}$ | Periods | Exciton $g$ factor | Hole $g$ factor |
|---|---|---|---|---|---|---|
| 46/110 | 46/110 | 0.33 | 7 | 100 | 1.1(1) | 2.3(1) |
| 72/110 | 72/110 | 0.33 | 5 | 100 | | 2.1(1) |
| 102/110 | 102/110 | 0.33 | 5 | 100 | | 2.0(1) |
| 29/101 | 29/101 | 0.34 | | 150 | 1.5(1) | |
| 71/104 | 71/104 | 0.33 | | 70 | 1.0(1) | |
| 98/103 | 98/103 | 0.35 | | 40 | 0.8(1) | |
| 198/103 | 198/103 | 0.35 | | 25 | | |

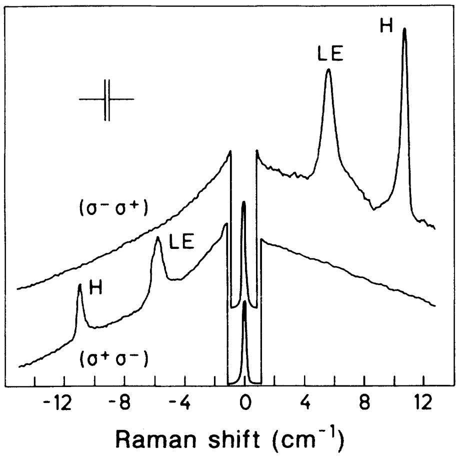


FIG. 2. Raman spectra measured in $(\sigma^-,\sigma^+)$ (upper spectrum) and $(\sigma^+,\sigma^-)$ (lower) configurations in a magnetic field $B=10$ T and for excitation energies $\hbar\omega=1.628$ eV. $H$ labels the hole spin-flip Raman line, LE the localized exciton angular momentum flip line. Sample 46/110.

cm$^{-1}$ for $L_z=46$ Å. The intensity of the $H$ line depends linearly on the excitation intensity in the region of 0.1–3 W/cm$^2$. At higher power densities it first saturates and then decreases.

The highest Raman scattering efficiency was observed in all Be-doped MQW's for excitation frequencies slightly above the maximum of the PL band. The Raman resonance profile was very narrow in the relatively wide QW's 72/110 and 102/110, with a half width of $\cong 3$ meV (half width of PL band $\cong 7$ meV). The Stokes and anti-Stokes lines were strongly circularly polarized, i.e., the Stokes line was observed in $z(\sigma^-,\sigma^+)\bar{z}$ configuration while the anti-Stokes one was observed only for $z(\sigma^+,z^-)\bar{z}$. Both lines were absent in the $(\sigma^+,\sigma^+)$ or $(\sigma^-,\sigma^-)$ configurations. In the case of the relatively narrow QW's in sample 46/110 the resonance profile was wider (see Fig. 1, dashed line). The $H$ line was observed for excitation with an energy very close to the band gap of the QW. Moreover, the polarization of the Stokes and anti-Stokes lines was found to depend on the excitation energy (see Fig. 1, dotted-dashed curve). In the low-energy region Stokes and anti-Stokes lines were observed in the $z(\sigma^-,\sigma^+)\bar{z}$ and $z(\sigma^+,\sigma^-)\bar{z}$ configurations, respectively, as in the samples 72/110 and 102/110, whereas for high-energy excitation both Raman lines were observed predominantly in $z(\sigma^+,\sigma^+)\bar{z}$ or $z(\sigma^-,\sigma^-)\bar{z}$ configuration, both in the Stokes and anti-Stokes region.

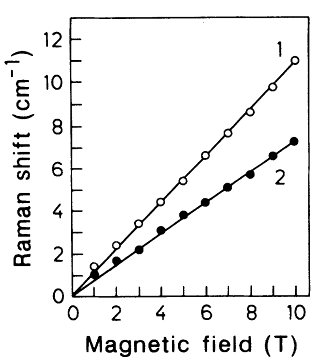


FIG. 3. Dependence of Raman shift on magnetic field strength: (1), SFRS from Be-doped MQW 46/110 (spin flip of hole bound on acceptor). (2), SFRS from undoped MQW 29/101 (angular momenta flip of localized exciton).

We conjecture that we are dealing here with two different limits of the same process of light scattering. Case $A$, where Stokes and anti-Stokes lines are seen only in crossed circular polarizations [$z(\sigma^-,\sigma^+)\bar{z}$ for Stokes and $z(\sigma^+,\sigma^-)\bar{z}$ for anti-Stokes], was observed in samples 72/110, 102/110, and 46/110, only for excitation in the low-energy tail of the PL band. Case $B$, where Stokes and anti-Stokes lines are seen predominantly in $z(\sigma^+,\sigma^+)\bar{z}$ or $z(\sigma^+,\sigma^-)\bar{z}$ configurations, was observed for excitation in the high-energy tail of the PL band. The ratio of intensities of Stokes and anti-Stokes lines in case $B$ (excitation well above resonance) can be represented by $I_{\mathrm{St}}/I_{\mathrm{aSt}}=\exp(\Delta E/kT)$, where $\Delta E$ is the Raman shift and $T$ is the lattice temperature. It will be shown in Sec. IV that the strong Raman scattering observed in both limiting cases $A$ and $B$ originates from the magnetically split ground state of the neutral acceptor and produces an angular-momentum flip (transitions between magnetic quantum numbers $+\frac{3}{2}\rightarrow-\frac{3}{2}$ or $-\frac{3}{2}\rightarrow+\frac{3}{2}$) of the bound hole coupled via exchange interaction to the photoexcited neighboring exciton.

The Raman scattering efficiency in both $A$ and $B$ cases was found to be sensitive to the lattice temperature. Figure 4 shows the dependence of the Stokes intensity on this temperature. In both cases the temperature dependence of the Raman intensity corresponds to that of a temperature-activated process. In case $A$ the activation process does not occur till $T\cong 10$ K. The slope of the temperature dependence in the activation region ($T\cong 10$–30 K) yields an activation energy $\Delta\epsilon\cong 5$ meV. This value is very close to that estimated for the binding energy (4.5 meV) of excitons bound to Be in a 70-Å

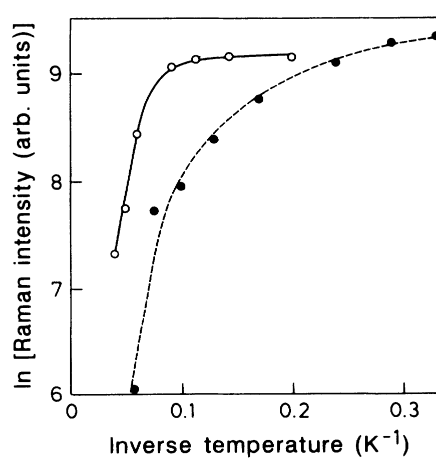


FIG. 4. Temperature dependence of Stokes line intensity at excitation in the low-energy region of the PL band ($\hbar\omega=1.628$ eV) (see Fig. 1, solid line) and in the region of the high-energy tail of the PL band ($\hbar\omega=1.648$ eV) (dashed line) and at $B=10$ T. Sample 46/110.

QW.[18] In case $B$ the activation process starts at rather low temperatures ($T \cong 3$ K) and the temperature dependence of the intensity is more gradual. This means that states with widespread energies of localization, smaller in the average than in case $A$, participate in process $B$.

We also found a significant enhancement of the Raman intensity in a magnetic field in both cases (see Fig. 5 for case $A$). The influence of the magnetic field on the Raman efficiency manifests itself in two different ways, an "additional" localization of the excitons and an increase of their oscillator strength. However, the role of the latter is probably not very important for our magnetic fields, giving only a factor $\cong 1.5$ for $B \cong 10$ T.[19] Hence, the main reason for the increase in Raman efficiency above $H \cong 4$ T must lie in the additional localization of the excitons at the interface roughness or impurities due to the shrinkage of the exciton Bohr radius. We expect the role of this effect to become noticeable when the cyclotron energy is comparable with the Coulomb energy. Both the temperature and magnetic-field dependences of the Raman efficiency support our conjecture that the SFRS takes place via bound or localized states of the exciton. The effect of temperature and magnetic field is in our case very similar to that observed recently by Zucker *et al.*[16] These authors showed that there is a close relation between the Raman intensity and the homogeneous exciton linewidth which is determined mainly by exciton localization at low temperatures and in a strong magnetic field.

The Raman shift shows in both cases *A and B* a strong dependence on the angle between the direction of the magnetic field and the growth axis ($z$) of the MQW (see Fig. 6, solid line). It is maximum when the magnetic field is parallel to the $z$ direction and vanishes when it is normal to $z$. The angular dependence can be well represented by a cosine function (solid line in Fig. 6). The tilting of the magnetic field also leads to a significant increase in the Raman intensity in case $A$ and to a breakdown of the polarization selection rules in both cases $A$ and $B$. Thus even for the small angle $\phi \simeq 15°$ a noticeable Raman line was observed in case $A$ for all polarization configurations with the intensity ratios

$$I(\sigma^-,\sigma^+):I(\sigma^-,\sigma^-):I(\sigma^+,\sigma^+):I(\sigma^+,\sigma^-) = 1:0.17:0.22:0.03 \ .$$

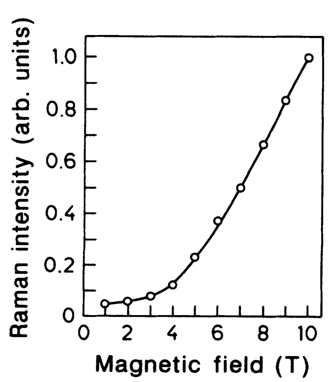


FIG. 5. Dependence of the SFRS efficiency on magnetic field at $T=4$ K (46/110, spin flip of the hole bound to the acceptor).

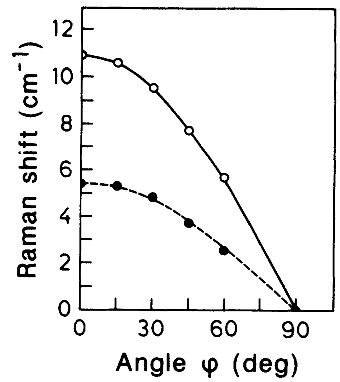


FIG. 6. Dependence of the Raman shift on the angle between the direction of the magnetic field ($B=10$ T) and the growth axis of the MQW. Open circles: spin flip of a hole bound to an acceptor; full circles: angular-momentum flip of the localized exciton. The solid and dashed lines represent cosine functions (sample 46/110).

In case $B$ the Raman line changed polarization so as to be observed mainly in $z(\sigma^-,\sigma^+)\bar{z}$ (Stokes) and $z(\sigma^+,\sigma^-)\bar{z}$ (anti-Stokes) configurations, i.e., the same polarization features as for line $A$.

The observed angular dependence of the Raman shift suggests that the $g$ factor of a hole bound to an acceptor is strongly anisotropic. The fact that this shift follows rather accurately a cosine dependence means that only the component of holes $g_{h\parallel}$ factor parallel to $z$ differs from zero, the other two transverse components $g_x, g_y = g_{h\perp}$ being zero within the experimental accuracy. Analysis of the experimental results shows that the sign and value of the parallel component of the $g$ factor is $g_{h\parallel} = +2.3$ for the 46/110 sample. Slightly lower $g$ factors are found with increasing well width (Table I).

### B. Undoped quantum wells

We come back to Fig. 2, which shows the Raman spectrum of the relatively narrow Be-doped QW 46/110. Besides the line with the large Raman shift discussed in the previous section (line $H$) one can see a line with a smaller shift (denoted as LE). We assign this line to Raman scattering by flipping the angular momentum of a localized exciton through interaction with acoustical phonons. Our further investigations have shown that this line is the only spin-flip feature also observed in undoped QW's. It is strongly circularly polarized, the Stokes component being seen in $z(\sigma^-,\sigma^+)\bar{z}$ and the anti-Stokes in the $z(\sigma^+,\sigma^-)\bar{z}$ configuration (like case $A$ of the $H$ line). The Raman shift of the LE line also follows rather closely a cosine dependence on $\phi$ (see Fig. 6, dashed line) and its polarization does not change when tilting the magnetic field, contrary to the behavior of the $H$ line. The angular dependence of the Raman shift also reveals an anisotropic $g$ factor. Only its component parallel to $z$ differs from zero $[(g_{\text{ex}\parallel} = (g_h - g_e)_\parallel = +1.5)]$ for sample 29/101, while the perpendicular components $g_{\text{ex}\perp} = g_x, g_y$ are zero within the experimental accuracy.

The intensity of this LE line depends on the width of the undoped QW. It is maximum for sample 29/101, smaller for 98/103, and becomes unobservable for 198/103. This behavior of the SFRS efficiency can be attributed to the increase of the exciton oscillator strength at the 3D to 2D transition,[12,13] plus the increase of the electron-hole exchange interaction in the QW.[10,11] We believe the effect of exciton localization on SFRS efficiency is very important (the wider the QW the smaller the potential for localization due to interface roughness). It should be noted, however, that the Raman shift at a fixed magnetic field reveals a weak dependence on the QW width, increasing slowly when $L_z$ decreases from 100 to 50 Å and more rapidly (from 5 to 7 $\text{cm}^{-1}$ at $B=10$ T) when $L_z$ decreases from 50 to 30 Å.

## IV. THEORY AND DISCUSSION

The process of optical excitation (or recombination) of electron-hole pairs or excitons in quantum wells is governed by the following selections rules. Excitation (recombination) into the $|-\frac{1}{2},+\frac{3}{2}\rangle$ state takes place under absorption (emission) of $\sigma^+$-polarized light and into the $|\frac{1}{2},-\frac{3}{2}\rangle$ state by $\sigma^-$-polarized light. Optical excitation into $|\frac{1}{2},-\frac{3}{2}\rangle$ and $|-\frac{1}{2},-\frac{3}{2}\rangle$ states is forbidden in the dipole approximation. In $|s,m\rangle$ the notation $s$ denotes the projection of the electron spin while $m$ denotes that of the hole angular momentum on the $z$ axis. The latter is governed by Luttinger's Hamiltonian for the $\Gamma_8$ band of bulk GaAs.

For a (001)-grown MQW the symmetry is reduced from $T_d$ to $D_{2d}$. Hence the fourfold degeneracy of the hole bound to the acceptor splits into two, $|\pm\frac{1}{2}\rangle$ and $|\pm\frac{3}{2}\rangle$, the ground state being $|\pm\frac{3}{2}\rangle_A$. We shall assume that the transverse $g$ factor of holes is equal to zero (it has been shown by symmetry consideration that this $g$ factor should be very small).[20] We determine the longitudinal $g$ factor of the hole bound to the acceptor ($A$) or in an exciton ($h$) as follows: the Zeeman energy of states $\pm\frac{3}{2}$ in the magnetic field **B** is equal to $\pm g_{A,h}\mu_0 B_z/2$, where $\mu_0$ is the Bohr magneton. We now consider the experimentally observed processes of SFRS in MQW's.

### A. SFRS in *p*-doped (Be) MQW's

The Raman lines observed only in $p$-type-doped QW's were interpreted as light scattering by a flip of the angular momentum of a hole bound to a neutral acceptor ($+\frac{3}{2}\to-\frac{3}{2}$ or $-\frac{3}{2}\to+\frac{3}{2}$). The process of light scattering can then be viewed as a three-step process.

(i) Absorption of light with creation of an exciton (we apply the term exciton to a localized $e$-$h$ pair, the state of which is modified by Coulomb interaction of the hole with the electron).

(ii) Spin flip of the hole bound to an acceptor due to pair-exchange interaction in the complex including three particles: hole bound to the acceptor, a spin-polarized electron, and the corresponding hole of a photoexcited exciton. The following transitions contribute to the light scattering process:

$$A:\ |\pm\tfrac{3}{2}\rangle_A|\pm\tfrac{1}{2},\mp\tfrac{3}{2}\rangle\to|\mp\tfrac{3}{2}\rangle_A|\mp\tfrac{1}{2},\pm\tfrac{3}{2}\rangle$$

and

$$B:\ |\pm\tfrac{3}{2}\rangle_A|s,m\rangle\to|\mp\tfrac{3}{2}\rangle_A|s,m\rangle\ .$$

(iii) Annihilation of the exciton with the emission of a photon.

Let us consider the processes $A$ *and* $B$ occurring after step (i), e.g., after absorption of a photon with $\sigma^-$ polarization, followed by the creation of an exciton in the state $|\frac{1}{2},-\frac{3}{2}\rangle$. In case $A$ only a hole in the state $|+\frac{3}{2}\rangle_A$ will participate in the exchange interaction after $\sigma^-$ absorption. The spins of the three particles flip and Stokes emission occurs only in the $(\sigma^-,\sigma^+)$ configuration if $g_A B<0$, while anti-Stokes emission takes place for $g_A B>0$. The opposite occurs for $(\sigma^+,\sigma^-)$ scattering.

In the type-$B$ process of exchange interaction the electron and hole spins in the exciton do not change their orientations. This is the reason why light scattering takes place predominantly in either the $(\sigma^+,\sigma^+)$ or $(\sigma^-,\sigma^-)$ configurations. Since in this case the processes of spin flip $|+\frac{3}{2}\rangle_A\to|-\frac{3}{2}\rangle_A$ as well as $|-\frac{3}{2}\rangle\to|+\frac{3}{2}\rangle$ are possible one sees in the Raman spectrum Stokes and anti-Stokes lines simultaneously. The process of spin flip is described by the following Hamiltonian, involving an acceptor and an exciton localized at the interface roughness:

$$H_1=\Delta_1\sigma_z^h(\sigma_x^A O_y+\sigma_y^A O_x)\ . \qquad (1)$$

Here the matrices $\sigma_j^h$ and $\sigma_j^A$ are the analogs of the Pauli matrices for the hole in the exciton ($h$) or bound to an acceptor ($A$) in the basis $[\beta(X-iY);\ -\alpha(X+iY)]$ which transforms as the double group representation $\Gamma_7$ of $D_{2d}$. $O$ is the unit two-dimensional vector in the $\rho_h-\rho_A$ direction, where $\rho_h$ and $\rho_A$ represent the center of the hole ($h$) and acceptor ($A$) localization in the $x,y$ plane (the dependence on $|\rho_h-\rho_A|$ is included in the coefficient $\Delta_1$). In the derivation of the $H_1$ invariant we take into account the fact that the components $O_x$ and $O_y$ of $O$ and those of the pseudovectors $\sigma_x$ and $\sigma_y$ transform according to the same $E$ representation of the $D_{2d}$ group, but $\sigma_z$ transforms according to the $A_2$ representation. If we do not consider the effects of the absence of inversion symmetry in the QW, its symmetry can be characterized by the point group $D_{4h}$ instead of $D_{2d}$. In this case the coefficient of Eq. (1) would be nonzero only when the wave function of the hole bound to the acceptor (and/or hole in the exciton) is not invariant upon reflection with respect to the plane which bisects the QW. This will happen when the acceptor is not located on the bisector plane. It should be noted that the coefficient $\Delta_1$ in Eq. (1) would then be an odd function of the acceptor position $z_i$ with respect to that plane. Obviously $H_1$ flips the acceptor spin while leaving $s$ and $m$ invariant.

In a tilted magnetic field the Raman shift in SFRS depends on the angle between $B$ and the $z$ direction of the QW as follows:

$$\hbar\Delta\omega=g_A\mu_0 B\cos\phi\ . \qquad (2)$$

Besides this, the deviation of the magnetic field from the $z$ direction leads to changes in the relative contributions of the two processes $A$ *and* $B$. The electron spin in a tilted magnetic field can change its orientation due to Zeeman interaction with the field. Therefore for process $A$ to take place in a tilted $B$ only hole-hole exchange interaction with their mutual spin flip is needed. This process is described by the Hamiltonian

$$H_2=\Delta_2(\sigma_x^h\sigma_x^A+\sigma_y^h\sigma_y^A) . \quad (3)$$

To explain a process of type $A$ in the exact $\mathbf{B}\|\mathbf{z}$ geometry besides the exchange interaction (3) we have to take into account exchange interaction between the electron and hole in the exciton

$$H_{e\text{-}h}=\Delta_\|\sigma_z^e\sigma_z^h+\Delta_\perp(\sigma_x^e\sigma_x^h+\sigma_y^e\sigma_y^h) , \quad (4)$$

and the anisotropic interaction, involving an acceptor and a neighboring exciton,

$$H_3=\Delta_3\sigma_z^A(\sigma_x^hO_y+\sigma_y^hO_x) . \quad (5)$$

The coefficients $\Delta_1$ in (1) and $\Delta_3$ in (5) should be small compared with $\Delta_2,\Delta_\|,\Delta_\perp$ since the binding of the exciton to the acceptor is weak. The efficiency and detailed mechanism of light scattering with spin flip of a hole bound to an acceptor is determined by the values of these coefficients.

In the frame of this model it is easy to explain the SFRS polarization dependence presented in Fig. 1. The SFRS efficiency profile shown in this figure is a sum of two processes $A$ and $B$. The high-energy tail of this profile corresponds to a $B$-type process (spin flip of a hole bound to acceptor via interaction with localized on roughness excitons) and it is seen predominantly in the $z(\sigma^-,\sigma^-)\bar{z}$ configuration. The low-energy tail corresponds to process $A$ (spin flip of a hole bound to an acceptor via its interaction with neighboring exciton), and it is seen only in the $z(\sigma^-,\sigma^+)\bar{z}$ configuration. In the region where the intensities of both process are comparable the polarization $\rho$ of the SFRS is zero. It should be mentioned that the $A$-type process could be also connected with the resonant intermediate state "exciton bound to neutral acceptor" if the spin of the polarized electron in the complex changes its direction. Possible mechanisms for this electron spin flip can be the hyperfine coupling with nuclear spin or spin-lattice relaxation. No likely alternative mechanisms for the type-$B$ process have occurred to us at this time.

### B. SFRS in undoped MQW's

The SFRS observed in undoped MQW's (see Sec. III B, also line LE in Fig. 2) can be explained as resonance scattering of light by acoustic phonons. In this process the light in $\sigma^\mp$ polarization excites an exciton in the state $|\pm\frac{1}{2},\mp\frac{3}{2}\rangle$. Due to interaction with an acoustic phonon the exciton is scattered to the state $|\mp\frac{1}{2},\pm\frac{3}{2}\rangle$ and annihilates with emission of $\sigma^\pm$ light. If the product of $(g_h-g_e)\,B_z>0$ ($g_e$,$g_h$ electron and hole $g$ factors), the scattering takes place in $(\sigma^+,\sigma^-)$ configuration in the Stokes region and $(\sigma^-,\sigma^+)$ configuration in the anti-Stokes region. If $(g_h-g_e)B_z<0$ the Stokes and anti-Stokes spectra reverse their polarizations. A comparison of the polarizations in the two cases of Be-doped QW (spin-flip hole bound on neutral acceptor) and undoped QW shows that the signs of $g_A$ and $(g_h-g_e)$ coincide.

A possible contribution to the exciton angular momentum flip is piezoinduced exchange interaction between electron and hole in the exciton:

$$\delta H=\Delta' e_{xy}(\sigma_x^e\sigma_y^h+\sigma_y^e\sigma_x^h) + \Delta''(e_{xx}-e_{yy})(\sigma_x^e\sigma_x^h-\sigma_y^e\sigma_y^h) . \quad (6)$$

Here $e_{\alpha\beta}$ is the strain tensor of the phonons involved. Under the assumption of the same electron-phonon coupling constants as in the bulk material $\Delta'$ would be zero while $\Delta''$ would correspond to deformation-potential interaction with LA phonons. For a single QW $\Delta'$ and $\Delta''$, and thus the Raman intensity, should depend monotonically on the Raman shift, i.e., on the magnetic field. For MQW's, however, oscillations in the Raman intensity versus $B$ should result depending on whether the period is a multiple of the wavelength of the phonon required for the scattering or a half multiple, a fact which could be used to investigate the type of phonons involved. The exciton-scattering process under consideration can also take place with emission or absorption of two phonons using $|\frac{3}{2},\pm\frac{1}{2}\rangle$ states as intermediate states. A detailed investigation of the dependence of the scattered intensity on magnetic field is needed to unravel the possible mechanisms.

We assume that the exciton $g$ factor equals $g_h-g_e$, where $g_e$ and $g_h$ are the $g$ factors of the corresponding conduction- and valence-band edges. It is thus possible to estimate $g_{h\|}$ from the measured values of $g_{h\|}-g_e\simeq+1.5$. In bulk GaAs $g_e\simeq-0.4$,[21] a value which should be renormalized in order to take into account the increase of the gap in our quantum well ($g_e\cong2\{(1-E_p\Delta_0/[3(E_0+\Delta_0)E_0]\}$). With this renormalization we find for our QW's $g_e\simeq0$ and thus $g_{h\|}\simeq+1.5$. This value is remarkably small since for the $(\frac{3}{2},\pm\frac{3}{2})$ band edge we estimate from the $kp$ theory $g$ factor

$$g_{h\|}\simeq6k_L=7 ,$$

where $k_L$ is the well-known "Luttinger" parameter ($k_L\simeq1.2\pm0.25$).[22]

The fact that the excitonic factors $g_{h\|}-g_e$ in quantum wells are much smaller than those expected from bulk parameters has already been pointed out in Refs. 15 and 23 ($g_h=1.4$ was obtained for a well width of 79 Å and $g_h=2.6$ for a width of 25 Å). It has been suggested that the drastic difference between the band edge $g_{h\|}$ (+7.2) and the measured one (+1.5), is due to admixture of the $(\frac{3}{2},\pm\frac{3}{2})$ 1$s$ exciton envelope function with the 3D functions of the light-hole exciton $(\frac{3}{2},\pm\frac{1}{2})$.[24] Calculations show indeed that the "$g$ factor" of the latter is negative and drastically reduces through admixture that of the 1$s$ exciton, being even able to produce a sign reversal in $g_{h\|}$. Nonlinearities in the spin splitting, leading to sign reversal appear, however, at higher $B$'s (the field at which the crossing occurs is very sensitive to the choice of parameters). Although we have seen a superlinear behavior of the splitting versus $B$ for the QW with $L_z>70$ Å, no zero crossing was observed up to 10 T.

We point out that all such calculations available so far do not include linear terms in $k$ in the valence bands. Such linear terms should also tend to quench spin splitting in magnetic fields and may be essential to explain quantitatively the observations described here and in other works.

## V. CONCLUSIONS

Strong Raman scattering was observed from $p$-type Be-doped $GaAs/Al_xGa_{1-x}As$ MQW's as well as from undoped samples in the presence of a magnetic field. This scattering was interpreted as spin-flip Raman scattering involving magnetically split states of neutral acceptors (in Be-doped MQW's) or localized excitons (in Be-doped MQW's and also in undoped MQW's).

Theoretical models have been developed to explain SFRS in doped and undoped MQW's. In the frame of these models the SFRS specific of $p$-type-doped QW's is related to angular-momentum flip ($\pm\frac{3}{2}\rightarrow\mp\frac{3}{2}$) of a hole bound to a neutral acceptor via exchange interaction with a neighboring exciton. The SFRS observed in undoped QW's is interpreted as the angular-momentum flip of a localized exciton via its interaction with one or more acoustical phonons (doubly resonant scattering by acoustical phonons). The interaction with one phonon may be considered as piezoinduced exchange interaction between the hole and the electron bound in an exciton.

Both types of SFRS show a strong dependence of the Raman shift on the angle between the magnetic field and the MQW growth axis. The longitudinal components of the $g$ factor for the hole bound to the acceptor are determined to be $g_{h\parallel}=2.3$, and for the exciton $(g_h-g_e)_\parallel=1.5$ (for the narrowest wells measured), which is much smaller than that of the corresponding band edge. Possible reasons for this effect have been discussed. The transverse $g$'s, $g_{h\perp}$ and $(g_h-g_e)_\perp$, amount to less than 0.01. The dependence of the $g$ factor on the well width has been investigated. The $g$ factor of the hole bound to the acceptor and of the exciton increase slightly (by about 10%) when the well width decreases from 100 to 50 Å, while that of the exciton increases 1.4 times when the QW width decreases from 50 to 30 Å.

The SFRS efficiency of excitons reveals a strong dependence on well width, disappearing for $L_z\geq 200$ Å. We believe that this effect is due to the dependence of the oscillator strength, the electron-hole exchange interaction, and the potential of exciton localization on the width of the QW. It is reasonable to assume that the SFRS technique will be useful for the investigation of the dependence of the hole $g$ factor on QW width, i.e., the mixing of heavy- and light-hole wave functions in the acceptor states, and also for studying the dependence of the $g$ factor of electrons bound to donors on well width. We believe that the SFRS technique can also be used to study impurities and exciton states in systems with dimensionality smaller than two.

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[1]G. D. Thomas and J. J. Hopfield, Phys. Rev. **175**, 1021 (1968).

[2]S. Geschwind and R. Romestain, in *Light Scattering in Solids*, edited by M. Cardona and G. Güntherodt, Topics in Applied Physics Vol. 54 (Springer, Berlin, 1984).

[3]R. Ebert, H. Pascher, G. Appold, and H. G. Hafele, J. Appl. Phys. **14**, 155 (1977).

[4]J. F. Scott, Phys. Rep. **194**, 379 (1990).

[5]M. Nawrocki, R. Planel, G. Fishman, and R. Galazka, Phys. Rev. Lett. **46**, 735 (1981).

[6]D. L. Alov, S. I. Gubarev, and V. B. Timofeev, Zh. Eksp. Teor. Fiz. **86**, 1124 (1984) [Sov. Phys. JETP **59**, 658 (1984)].

[7]D. L. Peterson, D. U. Bartholomew, U. Debska, A. K. Ramdas, and S. Rodriguez, Phys. Rev. B **32**, 323 (1985).

[8]S. I. Gubarev, T. Ruf, and M. Cardona, Phys. Rev. B **43**, 14 564 (1991).

[9]R. Meyer, M. Hirsch, G. Schaack, A. Waag, and R.-N. Bicknell-Tassius, Superlatt. Microstruct. **9**, 165 (1991).

[10]R. Bauer, D. Bimberg, J. Christen, D. Oertel, D. Mars, J. N. Miller, T. Fukunaka, and H. Nakashima, in *Proceedings of the 18th International Conference on Semiconductor Physics, Stockholm, 1986*, edited by O. Engström (World Scientific, Singapore, 1987), p. 525.

[11]Y. Chen, B. Gil, P. Lefebre, and H. Mathieu, Phys. Rev. B **37**, 6429 (1988).

[12]W. T. Masselnik, P. J. Pearah, J. Klem, C. K. Peng, H. Morkoç, G. D. Sanders, and Yia-Chung Chang, Phys. Rev. B **32**, 8027 (1985).

[13]Y. Masumoto, M. Matsuura, S. Tarucha, and M. Okamoto, Surf. Sci. **170**, 635 (1986).

[14]W. Ossau, B. Jakel, E. Bangert, and G. Weimann, in *The Basic Properties of Impurity States in Superlattice Semiconductors*, edited by C. Y. Fong (Plenum, New York, 1988).

[15]H. W. van Kesteren, E. C. Cosman, W. A. J. A. van der Poel, and C. T. Foxon, Phys. Rev. B **41**, 5283 (1990).

[16]J. E. Zucker, E. Isaacs, D. Heiman, A. Pinczuk, and D. S. Chemla, Surf. Sci. **196**, 563 (1988).

[17]M. Kohl, D. Heitmann, S. Tarucha, K. Leo, and K. Ploog, Phys. Rev. B **39**, 7736 (1989).

[18]P. O. Holtz, M. Sundaram, R. Simes, J. L. Merz, A. G. Gossard, and J. N. English, Phys. Rev. B **39**, 13 293 (1989).

[19]G. E. W. Bauer and T. Ando, Phys. Rev. B **38**, 6015 (1988).

[20]G. L. Bir and G. E. Pikus, *Symmetry and Strain-Induced Effects in Semiconductors* (Halsted, Jerusalem, 1974).

[21]*Tables of Numerical Data and Functional Relationships in Science and Technology*, edited by O. Madelung, Landolt-Börnstein, New Series, Group III, Vol. 17, Pt. a (Springer-Verlag, Berlin, 1982).

[22]M. Reine, R. L. Aggarwal, B. Lax, and C. M. Wolf, Phys. Rev. B **2**, 458 (1970).

[23]B. Lefebvre, B. Gil, J. P. Lascaray, H. Mathieu, D. Bimberg, T. Fukunaga, and H. Nakashima, Phys. Rev. B **37**, 4171 (1980).

[24]G. E. W. Bauer, in *High Magnetic Fields in Semiconductors Physics II*, edited by G. Landwehr (Springer-Verlag, Berlin, 1989).