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Time-Resolved Optically-Detected Magnetic Resonance of Luminescence from $Si/Si_{1-x}Ge_x$ Superlattices

E.R. GLASER, T.A. KENNEDY, P.B. KLEIN, D.J. GODBEY, and P.E. THOMPSON

Naval Research Laboratory
Washington, D.C. 20375-5347

Time-resolved optically-detected magnetic resonance (ODMR) experiments have been performed on $Si/Si_{1-x}Ge_x$ superlattices grown by molecular beam epitaxy (MBE) on (001) Si. These structures exhibit a single, broad (FWHM ~ 70 meV) photoluminescence (PL) band with peak energy ~ 100 meV below the expected bandgap. Fast ($\tau \le 5$ μs) and slow ($\tau \cong 15$ μs) processes have been separated with the ODMR. The slow resonances are new and provide more detailed information on the nature of the recombination. These features are tentatively assigned to Ge-rich regions in the SiGe layers based on the resonance parameters and the higher spin-lattice relaxation.

There has been much interest recently in the development and fabrication of SiGe-based heterostructures and superlattices for opto-electronic applications. The effort has produced much progress in the controlled growth of $Si/Si_{1-x}Ge_x$ single-well and superlattice structures by both chemical vapor deposition (CVD) and molecular beam epitaxy (MBE) techniques. The optical properties as revealed by photoluminescence spectroscopy now serve as a benchmark of the material quality for many research groups. There have been reports of both extrinsic and intrinsic recombination processes from this material system.[1-7] Also, multiple processes have been found to exist for some samples.[1] However, in some cases the underlying mechanisms are not well understood.

We have studied the character of the emission processes in $Si/Si_{1-x}Ge_x$ superlattices by a combination of photoluminescence (PL), optically-detected magnetic resonance (ODMR) performed under continuous light (CW) excitation, and PL decay measurements.[7] The ODMR technique is particularly powerful since it provides information on the spin properties of the recombining electron and hole. However, we have found that it is sometimes difficult to perform CW-ODMR studies of these samples due to strong background signals produced by microwave electric fields.[8] These signals require free carriers and occur on a fast time scale (<< 1 μs). In the present work, the scheme of the CW light and on/off modulated microwaves has been modified to perform time-resolved ODMR studies of luminescence from $Si/Si_{1-x}Ge_x$ superlattices. We have been able to separate fast ($\tau \le 5$ μs) and slow ($\tau \cong 15$ μs) processes with this technique. The new resonances provide more detailed information on the nature of the optical processes.

This paper focuses on a 140 Å/70 Å $Si/Si_{0.7}Ge_{0.3}$ superlattice (25 periods) grown on an (001) Si substrate. This structure exhibits the now well-documented single, broad (FWHM ~ 70 meV) PL band with peak energy ~ 100 meV below the expected bandgap.[3,7] Similar results were also detected on the emission from a 120 Å/40 Å $Si/Si_{0.65}Ge_{0.35}$ superlattice sample provided by K.L. Wang and C.H. Chern at UCLA. Detailed PL and CW-ODMR studies of these samples have been reported.[7] In addition, PL decay measurements of these bands have been carried out with a Q-switched Nd:YAG laser.

We have proposed donor-acceptor pair recombination as the origin of the broad emission bands.[7] This assignment was based on the band-edge character of the hole and electron as established by the CW-ODMR studies. The binding energies of the donor and acceptor explain the shift of the peak energy of the PL. The linewidth derives from the variation in the binding energy of the acceptor with its position in the SiGe layers. An alternate mechanism associated with excitons bound to isoelectronic centers (related to Ge complexes) in the alloy layers has been suggested.[3]

The pulsed ODMR method employed in this study follows work by other groups.[9] An energy level diagram and the pulse sequence are shown in Fig. 1. The technique takes advantage of the different lifetimes of the Zeeman-split excited states. The microwave radiation transfers population from the slow state to the fast (emitting) state. The light pulse is generated by switching the output of an Ar^+ laser at 488 nm with an acousto-optic modulator. After a variable delay (5 μs $\le t_d \le$ 1 ms), the 35 GHz microwave pulse was applied at half the repetition rate of the light pulse. A lock-in amplifier subtracts the responses with and without microwaves. The maximum response is realized when the delay time (t_d) is approximately equal to the radiative lifetime (τ_r) of the fast emitting state.[9] The emission was detected with a Ge photodiode. The samples were studied under pumped helium conditions (T ~ 1.6 K) in the tail section of an Oxford Instruments 7-T magneto-optical cryostat.

Since the modulator has an on/off ratio of about 200, some tests were performed to ascertain which parts of the response were time-resolved (see Fig. 2). The feature labeled H in the CW spectrum with $2W/cm^2$ (Fig. 2a) has been assigned to holes derived from the strain-split $M_J = \pm 3/2$ valence band in the $Si_{0.7}Ge_{0.3}$ layers.[7] The line at 1.2 T was assigned to the recombining electron associated with the valley-degenerate $Si/Si_{0.7}Ge_{0.3}$ conduction band. The positive resonance at twice the resonance position of the $M_J = \pm 3/2$ hole is a multiple quantum transition. These features are described in more detail elsewhere.[7] The CW spectrum with $20\ mW/cm^2$ was taken to approximate the light level in the off-state of the pulsed experiment. Feature H was also observed by the D-ODMR experiment (Fig. 2c, $t_d = 10\ \mu s$) with intensity equal to that found in the CW case with light power reduced by two orders of magnitude (Fig. 2b). This result suggests that the resonance H is associated with a spin-dependent process which occurs on a time scale less than 10 μs.

Two new resonances, not observed in the CW studies, are found in the D-ODMR experiment (arrows in Fig. 2c). The lifetime associated with these lines was found to be ~ 15 μs from D-ODMR spectra obtained as a function of the delay time (t_d). This lifetime is similar to the decay time determined from low-temperature time-resolved PL measurements. Similar PL lifetimes (~30 μs) have been reported by other workers for MBE-grown $Si/Si_{1-x}Ge_x$ superlattices which exhibit the broad emission.[1]

The field positions of the feature labeled H and the two new resonances are plotted in Fig. 3 as a function of the angle (θ) between **B** and the [001] axis. The resonance H can be described by a highly anisotropic g-tensor with [001] axial symmetry: $g_{||} = 3.73 \pm 0.05$ and $g_{\perp} = 0$. As discussed in the earlier ODMR studies,[7] the shift to higher field following $B_{res} \propto 1/\cos\theta$ is a "fingerprint" of a hole state derived from the $M_J = \pm 3/2$ valence band in the SiGe layers.

The new resonances appear to have a derivative- or dispersion-like lineshape with a luminescence-decreasing component and a luminescence-increasing signal at higher field. The angular rotation studies reveal that these features split apart, shift to higher field, and broaden as the field is rotated towards the [110] axis. The last two characteristics are similar to the behavior observed for the hole resonance (H). In addition, the g-value of these features determined from the average resonance field is higher than the g-value of the hole resonance H for a particular magnetic field orientation in the $(1\bar{1}0)$ plane.

The signs of the resonances can be understood by considering thermalization within the manifold of excited states. An energy level diagram which describes the four excited states composed of a hole with spin $M_J = \pm 3/2$ and an electron with spin $m_s = \pm 1/2$ in a magnetic field is shown in Fig. 4. The hole spin-flip transitions (i.e. $-3/2 \rightarrow +3/2$) occur at different fields because of an exchange interaction term: $c\mathbf{S}_e \bullet \mathbf{S}_h$, where c is the spin-spin exchange interaction.[10] With the spin relaxation rates greater than the recombination rates of the strongly emitting states, the lower-lying states become more populated than the states higher in energy. This is referred to as the spin-thermalized case. The microwave-induced hole spin transitions will result in an emptying of the $|-3/2,+1/2\rangle$ state and a build-up of the $|+3/2,-1/2\rangle$ state. From electric-dipole selection rules, the two inner states can recombine with the emission of a circularly polarized photon. Thus, for this spin-thermalized case, the σ^- emission will decrease and the σ^+ emission will increase at the respective resonance field positions. This corresponds to what is found in the D-ODMR experiments.

Another important aspect of the new features is the higher g-value compared to the g-value of the hole resonance H derived from the $M_J = \pm 3/2$ VB in the $Si_{0.7}Ge_{0.3}$ layers. Regions in the SiGe layers of higher Ge concentration can account for the higher g-values since the Luttinger parameter (κ) for the valence band from which the hole g-values are determined increases from -0.42 for Si to 3.41 for Ge.[11] Furthermore, the higher spin-lattice relaxation rate associated with these resonances as deduced from the signs of the signals is also consistent with this model since the greater Ge concentration leads to an increase in spin-orbit coupling and, thus, faster spin-lattice relaxation times. These new resonances may be associated with the Ge interstitial platelets seen in recent Transmission Electron Microscopy (TEM) experiments.[3]

In summary, time-resolved ODMR experiments have been performed on the broad emission bands from $Si/Si_{1-x}Ge_x$ superlattices. The temporal studies reveal an exchange-split resonance spectrum derived from states with a lifetime of ~ 15 μs, similar to the lifetime of the emission bands determined from PL decay measurements. In addition, the lifetime of the hole resonance H derived from the strain-split $M_J = \pm 3/2$ valence band was found to be much faster, ≤ 5 μs. More work needs to be done to determine if these times refer to different parts of the same optical cycle (e.g. fast capture followed by a slow recombination) or two separate recombination processes, one fast and one slow.

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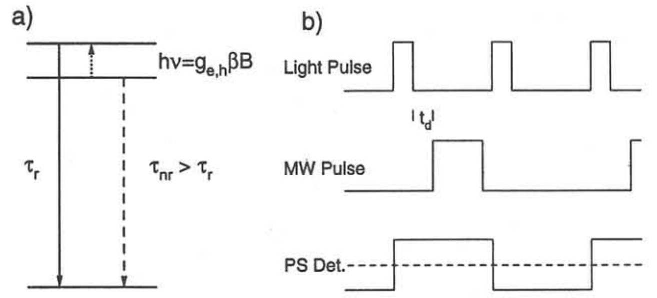

Fig. 1 a) Energy diagram for Zeeman-split excited states and the ground state. Microwave radiation (dotted arrow) transfers population from the slow state to the fast (emitting) state. b) Pulse sequence employed in the time-resolved ODMR experiments.

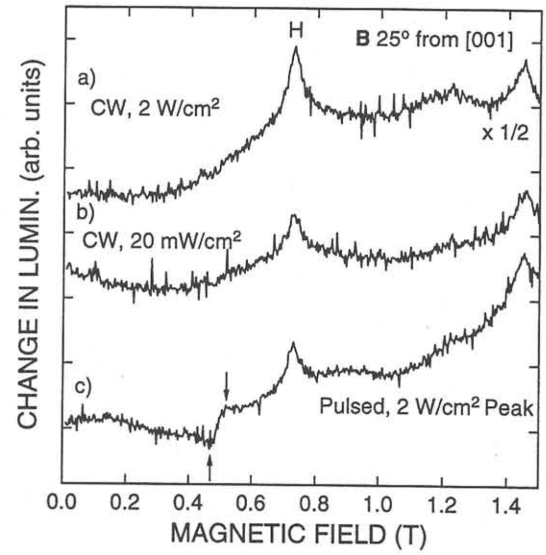

Fig. 2 ODMR spectra under three excitation conditions. Spectrum a) was taken with 2 W/cm^2 CW excitation and b) with 20 mW/cm^2 CW. Spectrum c) was taken with 10 μs light pulses with 2 W/cm^2 peak power and a delay of 10 μs between the light and microwave pulses.

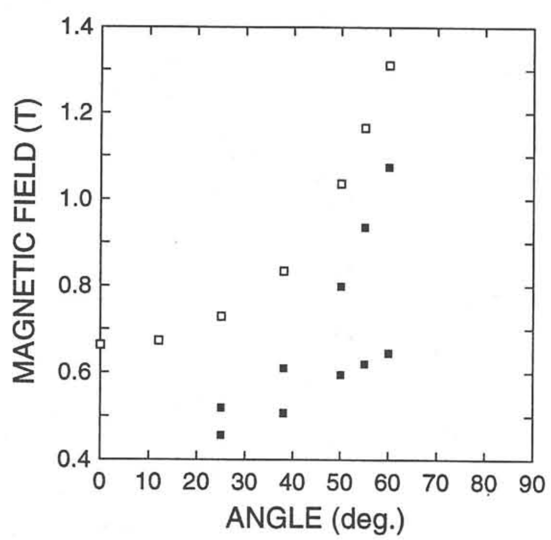

Fig. 3 The fields for resonance as a function of the angle between **B** and the [001] axis. Open symbols denote the hole resonance (H) derived from the strain-split $M_J = \pm 3/2$ valence band in the SiGe layers. Closed symbols denote new resonances revealed by the D-ODMR experiments.

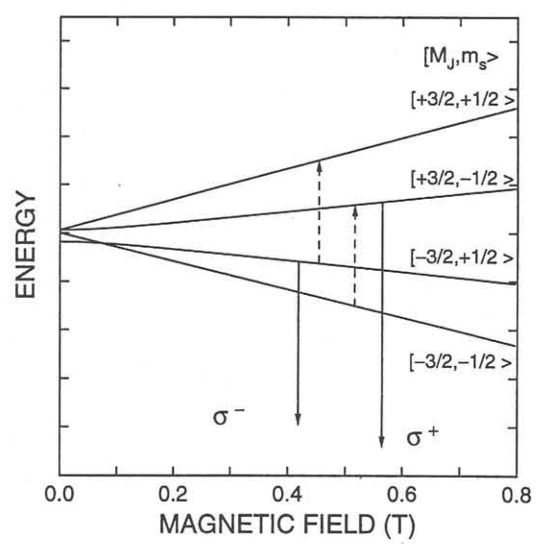

Fig. 4 The energies for the four excited states in a magnetic field, including an exchange interaction term. Microwave-induced hole spin-flip transitions (dashed arrows) for the thermalized case and allowed circularly polarized optical transitions (solid arrows) are shown.