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Alloy Fluctuations in $Si_{1-x}Ge_x$/Si Quantum Wells

YANG-FANG CHEN*, LI-HENG CHU*, SAN-CHANG PAN*, YUAN-SING YUANG*,
I-MING CHANG*, DING-CHANG CHANG**, AND CHING-YUANG CHANG**

**Department of Physics*
National Taiwan University
Taipei, Taiwan, R.O.C.
***Department of Electronics Engineering and Institute of Electronics*
National Chiao Tung University
Hsinchu, Taiwan, R.O.C.

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ABSTRACT

We report several interesting observations in the study of the optical properties of strained $Si_{1-x}Ge_x$/Si quantum wells. Both the photoluminescence energy and intensity showed anomalous temperature behavior: The energy first increased, and then decreased with increasing temperature; the intensity showed a temperature dependence similar to that of amorphous semiconductors and disordered superlattices; and blue shifting of the photoluminescence energy with increasing excitation intensity was observed. Persistent photoconductivity was detected, and the kinetics of its decay and transient buildup followed a stretched-exponential function and a parabolic time dependence, respectively. The first-order longitudinal-optical Si-like phonon mode showed asymmetric broadening behavior. All these observations can be explained by alloy potential fluctuations in a consistent way. Thus, our results firmly establish the existence of compositional fluctuations in $Si_{1-x}Ge_x$ epilayers and provide evidence showing that alloy potential fluctuations can greatly influence the properties of materials.

Key Words: alloy potential fluctuations, $Si_{1-x}Ge_x$/Si quantum wells

I. Introduction

One of the most important aspects of substitutional semiconductor alloys is the nature of the alloy potential fluctuations which arise from local fluctuations in composition and, hence, in the potential energy of carriers. The alloy potential fluctuations (APF) can greatly change the electrical and optical properties of a material, such as it's influence on carrier mobility (Blood and Grassie, 1984), the profiles of photoluminescence (Singh and Bajaj, 1986) and Raman scattering (Parayanthal and Pollak, 1984), and localization of exciton (Lai and Klein, 1980). APF also provides a model system for the study of carrier transport in percolative solids (Jiang and Lin, 1990) as well as a simple disordered system for the study of vibrational properties (de Gironcoli and Baroni, 1992).

$Si_{1-x}Ge_x$ is one of the substitutional semiconductor alloys. It has received increasing attention recently because of its potential application in silicon-based high-speed electronic circuits as well as in new optoelectronic devices. The occurrence of compositional fluctuations in $Si_{1-x}Ge_x$ alloys has been recognized for a long time (Feldman *et al.*, 1966). However, the study of the effects of the APF on the electronic and optical properties of $Si_{1-x}Ge_x$ epilayered structures has been rather limited. One of the reasons for this may be the fact that the Si/SiGe interface is smeared by Ge surface segregation during growth. Thus, the well resolved band edge luminescence and the shift of emission with a varying well width in $Si_{1-x}Ge_x$/Si quantum wells were reported only very recently (Sturm *et al.*, 1991; Fukatsu *et al.*, 1992; Xiao *et al.*, 1992; Vescan *et al.*, 1992). In this paper, we report several peculiar behaviors of optical properties observed in $Si_{1-x}Ge_x$/Si quantum wells, which include photoluminescence, persistent photoconductivity, and Raman scattering. All the anomalous behaviors can be explained by the existence of APF in a unified way. Our results firmly establish the existence of compositional fluctuations in $Si_{1-x}Ge_x$ epilayers and provide evidence showing that alloy potential fluctuations can significantly change the properties of materials.

II. Experiment

$Si_{1-x}Ge_x$/Si strained-layer multiple quantum wells were grown on 3-inch diameter (001) Si substrates at 550 °C using a home-made hot-wall multi-wafer ultrahigh vacuum chemical vapor deposition system. The construction and operation of our system is similar to that which has been reported elsewhere (Meyerson, 1986). Prior to growth, the substrate was subjected to H_2SO_4:H_2O_2=3:1 clean and a 10% HF dip. The gas sources for Si and Ge were SiH_4 and GeH_4, respectively. The base pressure of the system was maintained at about 2×18^{-8} torr in the growth chamber. During growth, the system was operated at about 1.0 mTorr. The growth was initiated by depositing a thin Si film 10 nm in thickness. The multiple quantum wells consisted of a total of 20 periods of Si/$Si_{1-x}Ge_x$ layers and a top Si cap layer about 96 nm in thickness. A ten second growth interruption was employed after each $Si_{1-x}Ge_x$ layer growth while no interruption was used after each Si layer growth. The reason for employing the growth interruption is that the growth rate of $Si_{1-x}Ge_x$ is much higher than that of Si. For Ge atomic mole fraction x of $Si_{1-x}Ge_x$ in Si/SiGe strained multiple quantum wells at about 0.16, the growth rates of Si layers and $Si_{1-x}Ge_x$ layers were about 0.8 nm/min and 5 nm/min, respectively.

High-resolution double-crystal X-ray diffraction was used to determine the structural quality of these multiple quantum wells. X-ray rocking curves were obtained by using a Philips DCD-3 double crystal diffractometer. The system was operated in the parallel (+,−) diffraction geometry using CuK_α radiation. A GaAs single crystal with (004) symmetric reflection was used for the first reflection. A dynamical X-ray simulation program was used to aid the determination of the perfection, interfacial abruptness, layer thickness and Ge composition of these multiple quantum wells. Cross-sectional transmission electron microscopy was performed to examine the crystal quality and interfacial abruptness. The crystalline quality of these samples was further checked by means of high resolution transmission electron microscopy. Details of the growth technique and results of structural characterization have been published separately (Chang *et al.*, 1994).

For the photoluminescence measurements, the sample was placed in a Janis cryostat, in which the temperature can be varied from 10 to 300 K. An Argon ion laser with 2.54 eV was used as the pumping source. The luminescence signal was analyzed using a 0.75-m double monochromator and detected using a North Coast Ge detector connected to a lock-in amplifier. For measurement of Raman scattering, the experimental system was the same as that used for photoluminescence measurements except that the detecting system was replaced by a charge coupled device (CCD) using a Si diode array.

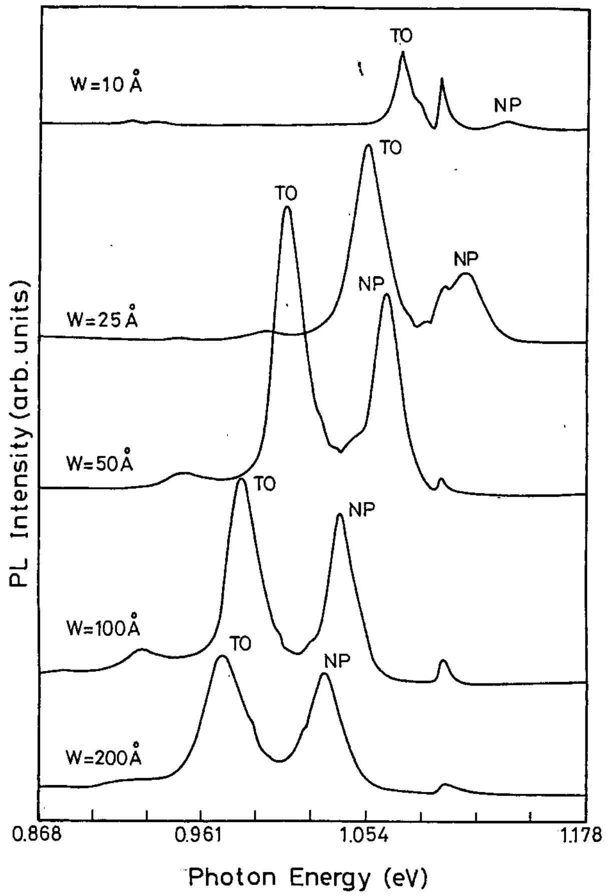

Fig. 1. Photoluminescence spectra at 10 K of five $Si_{0.84}Ge_{0.16}$/Si quantum well samples with well widths of 10, 25, 50, 100, and 200 Å.

For the persistent photoconductivity measurements, the sample was attached to a copper sample holder and placed inside a closed-cycle He refrigerator with care taken to ensure good thermal contact yet electrical isolation. The data at each temperature were obtained in such a way that the system was always warmed up to room temperature after a measurement, allowed to relax to equilibrium, and then cooled down in darkness to the desired temperature of measurements. This was to ensure that the data for each temperature were obtained under the same initial conditions. A HeNe laser was used as the pumping source. The conductivity was measured using a Keithley 236 source measure unit.

III. Results and Discussions

1. Photoluminescence Measurements

Figure 1 shows the photoluminescence spectra at 10 K of several of $Si_{0.84}Ge_{0.16}$/Si quantum well samples with well widths ranging from 1 to 20 nm. To identify the origin of the emission peaks, we followed the assignment made by Weber and Alonso (1989). The emission at 1.098 eV, which did not change with the variation of the qnantum well width, was due to free exciton assisted by transverse optical phonons in Si crystals. Intense luminescence lines labeled as no phonon (NP) and transverse phonon (TO), having an energy difference of 59 meV, which matches the TO phonon energy of Si, are no-phonon and transverse optical-phonon-assisted emissions, respectively, originating from SiGe quantum wells. Strong NP emission is a manifestation of the broken translational symmetry of the SiGe layers and is effected through momentum-conserving scattering due to compositional disorder in SiGe, as pointed out previously (Weber and Alonso, 1989). Phonon-assisted emissions due to Si-Ge and Ge-Ge TO phonons and transverse acoustic (TA) phonons were also observed in between the NP and TO emission lines. The peak positions of NP, TO, and other phonon-assisted emissions were obviously shifted to higher energies with decreasing SiGe well width, which is an indication of the effect of the quantum confinement. The lowest energy component shown in Fig. 1, which has not been identified before, can be attributed to the two-phonon replica since the energy difference between this peak and the NP line is equal to two times the TO phonon energy of Si.

For a more quantitative comparison with theory, we calculated the energy gap using the standard square potential model of Fukatsu *et al.* (1992). The band alignment for this alloy composition, 0.16, has conduction and valence band offsets of 10 and 136 meV, respectively, and the unconfined strained SiGe energy gap is 1.042 eV (Van de Walle and Martin, 1986). Electron and heavy-hole effective mass is assumed to be 0.19 m_0 and 0.28 m_0, respectively, where m_0 is the mass of the free electron. The effective mass of the electron in Si was used because it has little effect on the result due to the very small conduction band offset, and because the mass value in strained SiGe has not been clearly established. The calculated result as shown in Fig. 2 is represented by the solid curve. The solid points in Fig. 2 are the energy positions of the NP peaks taken from Fig. 1. We can see that the calculated and experimental values are in good agreement. Thus, the result shown here provides evidence that there was a quantum confinement shift in SiGe/Si multilayers grown using the ultrahigh vacuum chemical vapor deposition technique at low temperature, and that the samples studied here have good quality. The result also clearly shows the unambiguity of the identification of the band edge luminescences.

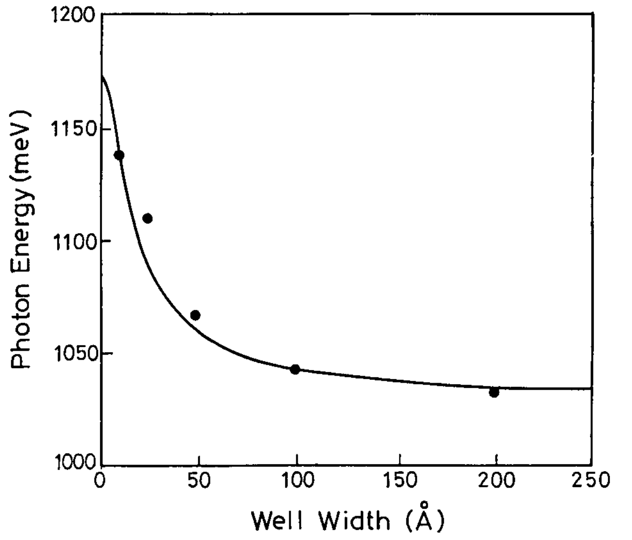

Fig. 2. Emission energy as a function of well width, showing the quantum confinement shift of the fundamental band gap. The solid line is the calculated result, and the solid dots are the experimental data.

Even though the above result can be used to demonstrate the quantum confinement shift in SiGe/Si multilayers, it does not reveal the exact nature of the photoluminescence (PL) processes. In order to explore this property, we measured the dependence of the PL spectra on temperature and excitation intensity. Figure 3 shows the temperature dependence of the PL intensities of the $Si_{0.84}Ge_{0.16}/Si$ quantum wells with a well width of 50 Å. It appears that the data can not be fitted by an Arrhenius plot. Instead, the temperature dependence is similar to the relationship used for amorphous semiconductors and disordered superlattices (Street *et al.*, 1974):

$$I_{PL} = \frac{I_0}{1 + A\exp(T/T_0)}, \tag{1}$$

where I_{PL} is the PL intensity, T is the measured temperature, T_0 is the characteristic temperature corresponding to the energy depth of localized states, A is a tunneling factor, and I_0 is the PL intensity at the low-temperature limit. Equation (1) is valid for amorphous semiconductors and disordered superlattices, rather than for a typical Arrhenius-type relationship, because of the existence of localized states (Van de Walle and Martin, 1986; Street *et al.*, 1974). Therefore, it seems reasonable to infer that the localization of carriers may occur in SiGe/Si quantum wells. The localized states could be due to regions of higher than average Ge content that arise simply from the random nature of the alloy. Similar localized exciton luminescence peaks

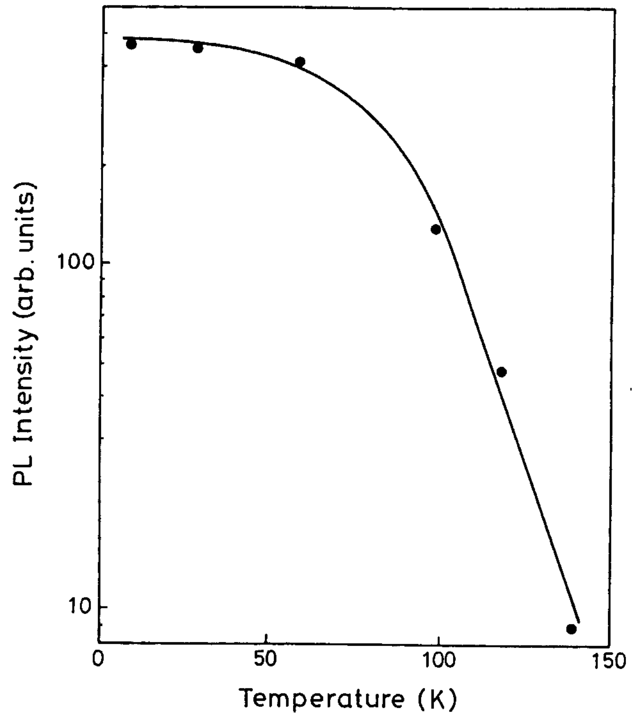

Fig. 3. Temperature dependence of the photoluminescence intensity for the $Si_{0.84}Ge_{0.16}/Si$ quantum well with a well width of 50 Å.

had been observed earlier in other semiconductor alloys (Lai and Klein, 1984; Fried *et al.*, 1989; Delong *et al.*, 1991; Olsthoorn *et al.*, 1993). Another piece of evidence to support the argument here is the temperature dependence of the PL energy as shown in Fig. 4. In the figure, the PL energy of the quantum wells first increases and then decreases with increasing temperature. This is different from the temperature dependence of the band gap of a semiconductor. In Fig. 4, we also show the PL spectra due to the Si substrate. It behaves normally; i.e. the PL energy decreases with increasing temperature. The anomalous temperature dependence of the PL energy can be explained easily by the localized excitons. At low temperature, carriers are located in the sites of minimum potential energies. As the temperature increases, carriers can absorb enough thermal energy and be excited to higher energy states; thus, an increase in emission energy is observed. As the temperature increases further, most of the carriers are located in the extended states, and the temperature dependence of the emission energy is determined by the band gap of the semiconductor, which decreases with increasing temperature. Additional evidence of the existence of the localized states due to potential fluctuations of the random nature of the alloy is shown by the measurement of the excitation intensity dependence of the PL spectra. Similar to the result obtained by Lenchyshyn *et al.* (1993), we observed a blue shift of the PL energy with increasing excitation intensity. This result can be understood by considering the fact that the localized states of band-edge fluctuations are limited in number and are easily filled by the photoexcited carriers. Increasing excitation intensity will increase the state filling; thus the emission energy is increased.

According to the above results, we suggest that localization of carriers in SiGe/Si quantum wells does occur. If this is true, we expect that at low temperature and low excitation intensity, the photoexcited electrons and holes will be spatially separated due to potential fluctuations, and that the PL will be caused by spatially indirect recombinations. The photoexcited carriers thus have a long radiative lifetime. With increasing excitation intensity, the photoexcited carriers will occupy the less localized states; hence, the radiative lifetime will decrease. This prediction is consistent with the recent measurement obtained by Lenchyshyn *et al.* (1993). Thus, all the PL measurements mentioned here can be explained by APF in a consistent way, which implies the existence of compositional fluctuations in $Si_{1-x}Ge_x$ alloys.

2. Persistent Photoconductivity Measurements

According to the above prediction, photoexcited electrons and holes can be spatially separated due to APF. We can then expect that light-induced conductivity will persist for a very long period of time after

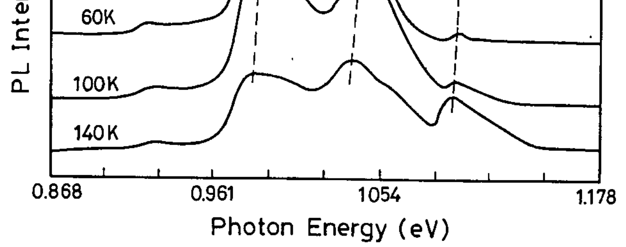

Fig. 4. Temperature dependence of the photoluminescence energy for the $Si_{0.84}Ge_{0.16}/Si$ quantum well with a well width of 100 Å. The PL energy of the quantum well first increases, and then decreases with increasing temperature. The PL energy of the Si substrate decreases with increasing temperature.

illumination. This phenomenon is usually called as persistent photoconductivity (PPC). In agreement with this expectation, we observed the existence of PPC for temperatures below 250 K in the studied samples. Figure 5 shows a typical plot of PPC decay at T=40 K. It can be seen that the photoconductivity persisted for several thousand seconds after termination of illumination. In order to analyze the kinetics of the PPC decay, we have tried to obtain the function of the time dependence for the decay profile. It is found that the PPC relaxation shows a stretched-exponential behavior, which can be expressed as

$$I_{ppc}(t)=I_{ppc}(0)\exp[-(t/\tau)^{\beta}], \quad (2)$$

where $I_{ppc}(0)$ is the conductivity level immediately after termination of the light source, $I(t)$ the conductivity at the decay time t, τ the decay-time constant, and β the decay exponent. Figure 6 shows the representative plot of $\ln[\ln I_{ppc}(0)-\ln I_{ppc}(t)]$ v.s. $\ln(t)$. The good linear behavior of the plot demonstrates that the PPC decay is well described by Eq. (2). Stretched-exponential relaxation has been commonly observed in disordered systems (Kakalios *et al.*, 1987; Chen *et al.*, 1991). The origin of the stretched-exponential relaxation can be understood by considering the fact the rate constant ν in the decay-rate equation is time dependent, that is,

$$d\Delta/dt=-\nu(t)\Delta. \quad (3)$$

Equation 3 describes the rate of decay for small departures Δ from equilibrium. A single-valued rate constant ν will yield the conventional Debye relaxation. Many previous investigations (Campos *et al.*, 1979; Jackson, 1988; Crandall, 1991) showed that if

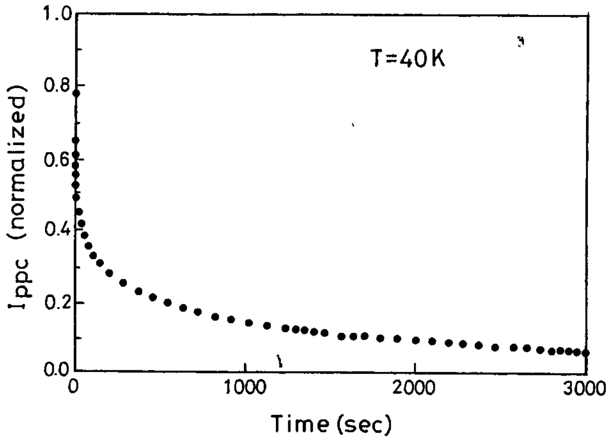

Fig. 5. The kinetics of the PPC decay curve for the $Si_{0.84}Ge_{0.16}/Si$ quantum well with a well width of 200 Å at 40 K.

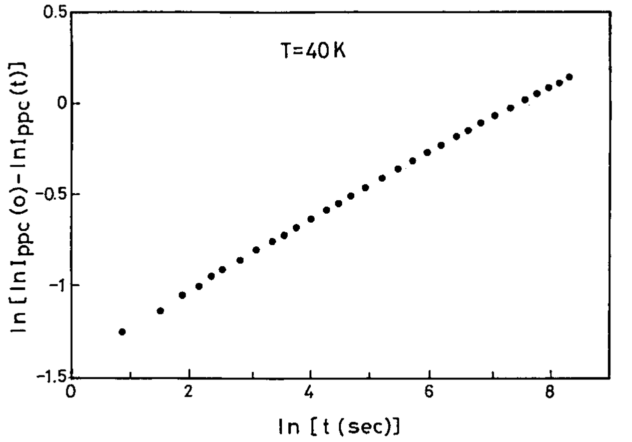

Fig. 6. Plot of $\ln[\ln I_{ppc}(0)-\ln I_{ppc}(t)]$ v.s. $\ln(t)$ for $Si_{0.84}Ge_{0.16}/Si$ quantum wells with a well width of 200 Å at 40 K. The linear curve indicates that the PPC decays according to the stretched exponential, $I_{ppc}(t)=I_{ppc}(0)\exp[-(t/\tau)^{\beta}]$.

the density of states is exponentially distributed in terms of energy, such as the band tail states in disordered semiconductors, the rate constant ν will exhibit a power-law time dependence. Inserting a power-law time dependence for ν into Eq. (3) and intergrating immediately yields a stretched-exponential function. Thus, the stretched-exponential decay reveals similarities of the present system with disordered systems and implies that the microscopic random-potential fluctuations are the origin of the observed PPC effect.

Further evidence showing that the observed PPC results from the existence of APF in the $Si_{1-x}Ge_x$ alloys can be obtained from the measurements of the PPC buildup. Figure 7 shows a typical plot of the initial transients of the PPC buildup. One cannot observe this behavior in materials exhibiting only conventional photoconductivity, which has a typical transient response time on the order of 10^{-6}s (Jiang and Lin, 1990). In Fig. 7, the striking feature is that the initial transients show a parabolic dependence on illumination time. The PPC buildup kinetics can be easily formulated using

$$dn/dt=g-\alpha n, \quad (4)$$

where n is the photogenerated carriers in the conduction band, g is the carrier generation rate, and α is the carrier decay rate. From Eq. (4), we obtain

$$n(t)=g/\alpha(1-e^{-\alpha t}). \quad (5)$$

If we assume that the carrier mobility does not depend on the carrier concentration, n, we then have

$$I(t)=I_0(1-e^{-\alpha t}), \quad (6)$$

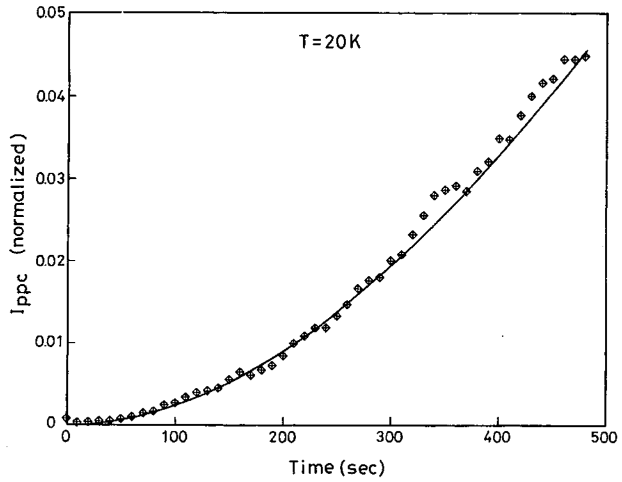

Fig. 7. The kinetics of the initial transients of PPC buildup for the $Si_{0.84}Ge_{0.16}/Si$ quantum well with a well width of 200 Å at 20 K. The solid curve is the plot of the equation $I(t)=I_0(1-e^{-\alpha t})^2$.

where I_0 represents the saturation level of PPC. For short illumination time, this equation leads to the result that the initial PPC level increases linearly with increasing illumination time. However, our experimental results shown in Fig. 7 obviously deviate from linear time dependence. The discrepancy can be attributed to the existence of APF in the $Si_{1-x}Ge_x$ alloys, which can cause tail states below the band edge and localize charge carriers at low temperatures. Thus, the carrier mobility is no longer independent of the carrier concentration as in the above assumption. According to the Kubo-Greenwood formula, if we assume that the distribution of the band tail states is exponential in terms of energy, we can obtain (Jiang *et al.*, 1992)

$$I(t)=I_0(1-e^{-\alpha t})^2. \tag{7}$$

For short illumination time, in Eq. (7), $I(t)$ has a parabolic time dependence which is consistent with our experimental results. Therefore, the PPC buildup result also tends to support the existence of APF in $Si_{1-x}Ge_x$ epilayer structures.

It is interesting to note that Eq. (7) can be obtained only if an exponential distribution of the tail states is used (Jiang *et al.*, 1992). Thus, the existence of an exponential tail of the states in $Si_{1-x}Ge_x$ is confirmed. Together with previous results obtained for doped semiconductors and amorphous materials, this indicates that the exponential tail of states is independent of the origin of the disorder, which is consistent with the theoretical argument (Soukoulis *et al.*, 1984).

Previously, PPC has been observed in layered structures of III-V compound semiconductors (Queisser, 1985; Schubert *et al.*, 1985). The spatial separation of photo-generated electrons and holes by a build-in electric field from a macroscopic barrier due to band bending at surfaces or interfaces was used to explain the observed PPC. It is possible that our measured PPC may also have the same origin. However, this model predicts a logarithmic PPC decay in time which is inconsistent with our results. Furthermore, we have set the wavelength of the pumping source such that its photon energy is between the energy gaps of Si barrier layer and $Si_{1-x}Ge_x$ well layer. For this wavelength, the photoexcited electrons and holes can only be created in the well region. Under this condition, PPC can still be observed. Thus, spatial separation of photoexcited electrons and holes due to a macroscopic barrier can be ruled out in the interpretation of our observed PPC.

3. Raman Scattering Measurements

Figure 8 shows the Raman spectra of the Si-like longitudinal-optic (LO) phonon mode from the Si substrate as well as the $Si_{1-x}Ge_x/Si$ epilayer. The spectrum of the Si substrate shows a symmetric line shape. For the $Si_{1-x}Ge_x$ epilayer, the spectrum has an asymmetrical line shape in which the lower half-width Γ_e on the lower-energy side is larger than the higher half-width Γ_h on the higher-energy side. The line shape of the Si-like mode becomes more and more asymmetric with the increase of the Ge molar fraction x. Similar results indicating broadening and asymmetry of first-order LO phonon Raman spectra have been observed in several alloy semiconductors (Parayanthal and Pollak, 1984; Ksendzov *et al.*, 1987; Cohen *et al.*, 1986; Olege *et al.*, 1986; Shen and Chen, 1994). These behaviors have been attributed to the effect of APF (Parayanthal

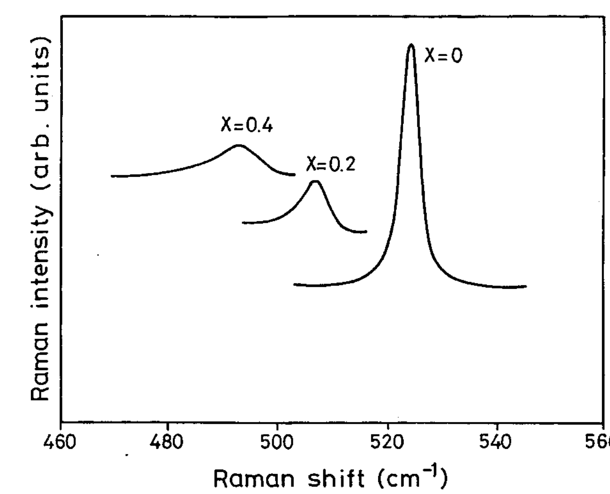

Fig. 8. Raman spectra of the Si-like LO phonon mode from $Si_{1-x}Ge_x/Si$ at 300 K for x=0.0, 0.2, 0.4.

and Pollak, 1984). The APF destroy translational invariance, an effect that manifests itself as a breakdown of the usual q=0 Raman selection rule, thus leading to broadening and asymmetry of the Raman line shape. Thus, the broadened and asymmetrical line shape observed here may imply that APF do exist in $Si_{1-x}Ge_x$ epilayered structures.

IV. Conclusions

In summary, well defined band edge exciton luminescence has been observed in as-grown $Si_{1-x}Ge_x$/Si strained-layer quantum wells, grown using ultrahigh vacuum chemical vapor deposition at low temperature (550 °C). The agreement between the theoretical and experimental results for the effect of quantum confinement on the photoluminescence spectra indicates the good quality of the samples produced using the technique and unambiguity in the identification of emission peaks. Several striking features of the optical properties have been observed, which include the temperature dependence of the PL intensity and peak position, the existence of PPC and its kinetics, and the line profiles of the Raman spectra. All these phenomena can be interpreted in a consistent way by taking into account the alloy potential fluctuations. Thus, our results firmly establish that compositional fluctuations do exist in $Si_{1-x}Ge_x$ epilayers. We have also shown that the fluctuations can greatly change the electrical as well as optical properties of materials, which are important in fundamental physics and device applications.

Acknowledgment

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矽化鍺與矽量子井之合金位能起伏現象

陳永芳*　朱立寰*　潘善中*　剡永聖*　張毅敏*　張鼎張**　張俊彥**

*國立台灣大學物理學系
**國立交通大學電子工程學系

摘　要

本研究報告一些在矽化鍺與矽量子井內的有趣現象。光螢光能量與強度表現出不尋常的溫度變化。當溫度增加時，能量先增加後減小；光強度展出類非晶半導體之特性；同時隨激發光強度之增加，光螢光能量會有藍位移現象。持續性光電導可以明顯的觀測到，光電導之衰退符合扭曲指數函數之描述，而光電導之建立遵循一拋物函數。第一階的縱向聲子頻譜呈現不對稱變寬之行爲。所有這些觀測到的現象，都能用合金成分的起伏來解釋。因此我們的結果建立了矽化鍺薄膜存在有合金成分的起伏，同時提供合金位能起伏如何影響了材料特性的證據。