Strain of GaAs/GaAsP Superlattices Used as Spin-Polarized Electron Photocathodes, Determined by X-Ray Diffraction

Takashi Saka,† Yoichi Ishida, and Masataka Kanda
Department of Electrical and Electronic Engineering, School of Engineering, Daido University, Takiharu-cho 10-3, Minami-ku, Nagoya 457-8530, Japan

Xiuguang Jin, Yuya Maeda, Shingo Fuchi, Toru Ujihara, and Yoshikazu Takeda
Department of Crystalline Materials Science, Graduate School of Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan

Tetsuya Matsuyama and Hiromichi Horinaka
Department of Physics and Electronics, Graduate School of Engineering, Osaka Prefecture University, Gakuen-cho 1-1, Naka-ku, Sakai 599-8531, Japan

Toshihiro Kato
New Business Development Division, Daido Steel Co., Ltd., Daido-cho 2-30, Minami-ku, Nagoya 457-8545, Japan

Naoto Yamamoto
Graduate School of Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan

Atsushi Mano, Yasuhide Nakagawa, Makoto Kuwahara, Shoji Okumi, and Tsutomu Nakanishi
Department of Physics, Graduate School of Science, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8602, Japan

Masahiro Yamamoto
High Energy Accelerator Organization, Oho 1-1, Tsukuba Ibaraki 305-0801, Japan

Takashi Ohshima and Teruo Kohashi
Central Research Laboratory, Hitachi Ltd., Higashi-Koigakubo 1-280, Kokubunji, Tokyo 185-8601, Japan

Masahiko Suzuki, Michihiro Hashimoto, Tsuneo Yasue, and Takanori Koshikawa
Fundamental Electronics Research Institute, Osaka Electro-Communication University, Hatsu-cho 18-8, Neyagawa, Osaka 572-8530, Japan

(Received 5 January 2010; Accepted 20 January 2010; Published 13 March 2010)

The strains in GaAs/GaAsP superlattices used in spin-polarized photocathodes grown on GaAs and GaP (001) substrates were determined by X-ray diffraction. The thicknesses of the GaAs wells and GaAsP barrier layers were also determined. The band structures of the superlattices were calculated on the basis of these experimentally determined strains and layer thicknesses. The thicknesses and band structures were in good agreement with those observed by transmission electron microscopy and photoluminescence, respectively. The strains induced in the GaAs well layers were approximately linearly dependent upon the phosphorous fraction in the GaAsP layer, and the splitting between the heavy hole band and the light hole band of the superlattices grown on GaP substrates was larger than that of superlattices grown on GaAs substrates. In photocathodes grown on GaP substrates, low polarizations were observed, not due to a lack of band splitting, but to depolarization scattering caused by crystal defects, which were different from that induced in superlattices grown on GaAs substrates.

Keywords: Superlattices; Gallium arsenide; X-ray scattering, diffraction, and reflection; Electron emission; Semiconducting films; Spin polarization; Photocathodes

I. INTRODUCTION

Spin-polarized electron photocathodes have primarily been applied in the field of high energy physics [1, 2]. Spin-polarized beams are obtainable by irradiating mate-

rials with a zincblende structure, such as GaAs, with a circularly polarized laser. The electrons are excited from both the heavy hole band and the light hole band, which are degenerate. Since the spin orientations of electrons excited from the two bands are opposite, and the excitation ratio is different, polarized electron beams are obtained. If the degeneracy is removed and electrons are excited from only one band, a high polarization is expected. In order to remove the degeneracy, photocathodes consisting of a strained GaAs layer or superlattice (SL) were developed [3–9]. All of these photocathodes were reflection-type, and the front surface of the photo-
TABLE I: Photocathode structures.

| No. | Number of SL pairs | SL Period (nm) | Substrate |
|-----|--------------------|----------------|-----------|
| #1  | 16                 | 6.0            | GaAs      |
| #2  | 12                 | 6.0            | GaAs      |
| #3  | 12                 | 8.0            | GaAs      |
| #4  | 12                 | 9.0            | GaP       |
| #5  | 12                 | 9.0            | GaP       |
| #6  | 12                 | 9.0            | GaP       |
| #7  | 12                 | 9.0            | GaP       |

cathodes were irradiated by the laser. From the various structures developed, a high polarization and a high efficiency were realized using strained GaAs/GaAsP SLs grown on GaAs substrates [10].

To apply spin-polarized electron beams to the field of materials science, it is necessary to realize a high brightness. Therefore, electrons should be excited by a finely focused laser beam. For this purpose, a transmission-type photocathode with a strained GaAs/GaAsP SL was developed, in which the cathode is irradiated from the rear. In a transmission-type photocathode, a transparent substrate for the excitation laser is required. Photocathodes with the same structure as that grown on the GaAs substrates were grown on GaP substrates. Photocathode growth on GaAs substrates resulted in high polarization, with a maximum value as high as 92% [10]. In contrast, the maximum polarization was as low as 64% when using GaP substrates, for reasons discussed in another paper [11]. The polarization was improved by inserting a 500-nm-thick GaAs layer between the GaP substrate and GaAsP layer [12, 13]. A high polarization was realized, but no precise structural analysis of the SLs grown on GaAs and GaP substrates was performed. In the present paper, strains in the SLs were determined using X-ray diffraction, and the band structures were calculated on the basis of the experimentally determined parameters. The correlation between strain and polarization was examined.

II. EXPERIMENTAL

A. Specimens

Seven spin-polarized electron photocathode specimens were prepared. These photocathodes were fabricated by metal organic vapor-phase epitaxy on GaAs and GaP substrates. Both the GaAs and GaP substrates were p-type and (001)-oriented. Specimens #1-3 were grown on GaAs substrates, and the others were grown on GaP substrates. The photocathodes consisted of stacks of a 1- to 2-µm-thick GaAsP layer (called the buffer layer) and 12 or 16 pairs of GaAs-GaAsP strained SL. In specimen #5, an intermediate layer, with a phosphorous fraction changed stepwise from one to that of the GaAsP buffer layer, was inserted between the GaP substrate and the buffer layer. For specimens #6 and #7, a 500-nm-thick GaAs layer was inserted (referred to as the inserted layer) between the GaP substrate and the GaAsP layer. Finally, the SL was coated with a highly doped 5-nm GaAs layer. The important parameters are tabulated in Table I. Specimen #1 was grown at Daido Steel Co., and all other specimens were grown at Nagoya University. References [9, 10] detail the respective source materials and growth conditions.

B. Measurements

To enable strain analysis, the lattice constants of the respective layers should first be determined. For this purpose, reciprocal maps were measured using 004 symmetric reflections, as well as asymmetric 113 reflections. Asymmetric 113 reflection measurements were performed in which the glancing angle was either smaller or larger than the Bragg angle. These conditions are specified as 113S and 113L reflections, respectively. Furthermore, for some specimens, asymmetric 224 reflections were also measured, but only with a glancing angle smaller than the Bragg angle. All reciprocal maps were measured using the “ω step and radial (2θ − ω) scan” method [14]. The measurements were repeated by rotating the specimens stepwise in 90° increments around the [001] direction. This rotation is referred to as φ rotation, in which the zero position of φ was defined as the position at which the incident beam was perpendicular to the [011] direction, and X-rays impinged upon the specimens from the (110) surface in the direction of the flat orientation cut on the (110) surface. The φ rotation was performed counter-clockwise. An X-ray diffractometer, ATX-G (RIGAKU), was used. The accelerating voltage was 50 kV, the beam current was 300 mA, and the target was Cu. A triple axis diffractometer was used, and the incident X-rays were monochromated and collimated by two channel-cut monochromators employing the Ge 220 reflection. The diffraacted X-rays were analyzed by a channel-cut monochromator, using the Ge 220 reflection. To compare the X-ray diffraction results, the band gap energy was observed by a photoluminescence method, and the thicknesses of the SL layers were observed by transmission electron microscopy.

III. RESULTS AND DISCUSSION

As typical examples, the reciprocal maps for 113S reflections from specimens #1, #2, #4, and #7 at φ = 0° are shown in Figs. 1(a), (b), (c), and (d), respectively. In Figs. 1(a)-(d), the main satellite peak and peaks due to the GaAs or GaP substrate and the GaAsP buffer layer are observed, while in Figs. 1(c) and (d), two sub-peaks of the SL of the 1st order are observed in addition to the main peak. The main satellite peak of the SL is denoted as ST0 and the sub-peaks are denoted as ST±1. In Fig. 1(d), a peak due to the inserted GaAs layer is also observed. These maps were plotted on a logarithmic scale. In the present paper, the results of the reciprocal maps are presented using scales of the ω scan (abscissa) and the 2θ − ω scan (ordinate), instead of the conventional reciprocal scales. Because the angular width of 2θ for the 2θ − ω scan was set to equal twice the value of the ω step, the lines parallel to the diagonal line from the...
FIG. 1: Reciprocal map of (a) specimen #1, (b) specimen #2, (c) specimen #4, and (d) specimen #7 from the (113) plane at $\phi = 0^\circ$. The glancing angle is smaller than the Bragg angle. The abscissa corresponds to the $\omega$ step and the ordinate corresponds to the $2\theta - \omega$ scan. Values of $2\theta$ and $\omega$ for the respective peaks were determined by the method of least squares, using the numerical data from the maps. Then, the coordinates of the peaks in reciprocal space were calculated. In this process, we assumed that the GaAs or GaP substrates were located at their perfect crystal positions. The observed positions of the substrates deviated from those of the perfect crystals, due to uncertainty of the zero-positions of $2\theta$ and $\omega$ in specimen setup, so that the calculated positions of all peaks deviated by the same amount from the observed positions, and the deviations were modified.

The lattice constants of the GaAsP buffer layers were determined for all specimens. For specimens #6 and 7, the lattice constants of the inserted GaAs layers were also determined. The lattice constant $c$ along the [001] direction was determined from the 004 position, and the lattice constants $a$, which was parallel to the [100] and [010] directions, and $c$ were determined from the 113 positions. It was assumed that the lattice constants along [100] and [010] were identical. The results for specimens #4 and #7 are shown in Figs. 2(a) and (b), respectively. It should be noted that the lattice constants $c$ determined from the 004 reflection and from the asymmetric reflections were almost identical for all specimens, supporting the reliability of the present analysis. Therefore, the following analysis was performed using only 113S reflections, in which the clearest peaks were observed among the asymmetric reflections.

FIG. 2: Lattice constants of the GaAsP buffer layers of (a) specimen #4 and (b) specimen #7 determined using various reflections.

TABLE II: Phosphorous fractions ($x$), and strains of GaAs well layers ($\varepsilon_{xx,\text{GaAs}}$) and GaAsP barrier layers ($\varepsilon_{xx,\text{GaAsP}}$) of the superlattices.

| No. | $x$  | $\varepsilon_{xx,\text{GaAs}}$ | $\varepsilon_{xx,\text{GaAsP}}$ |
|-----|------|-------------------------------|-------------------------------|
| 1   | 0.20 | $-6.2 \times 10^{-3}$          | $1.1 \times 10^{-3}$          |
| 2   | 0.27 | $-7.1 \times 10^{-3}$          | $2.4 \times 10^{-3}$          |
| 3   | 0.34 | $-11.3 \times 10^{-3}$         | $0.8 \times 10^{-3}$          |
| 4   | 0.38 | $-11.9 \times 10^{-3}$         | $1.7 \times 10^{-3}$          |
| 5   | 0.37 | $-12.8 \times 10^{-3}$         | $0.4 \times 10^{-3}$          |
| 6   | 0.23 | $-7.1 \times 10^{-3}$          | $1.3 \times 10^{-3}$          |
| 7   | 0.30 | $-8.8 \times 10^{-3}$          | $1.9 \times 10^{-3}$          |

http://www.sssj.org/ejssnt (J-Stage: http://www.jstage.jst.go.jp/browse/ejssnt/)
For specimen #4, the lattice constants $a$ and $c$ were nearly equal, indicating that the layer was unstrained. However, for specimen #7, the $c$-values were smaller than the $a$-values, indicating that the layer was under tensile strain along the crystal surface. The implicit lattice constants of the barrier layers, $a_{0, \text{GaAsP}}$, which would exist when the layer was strain-free, were determined by assuming that the layers were distorted into a tetragonal form. The implicit lattice constants are given by,

$$a_{0, \text{GaAsP}} = \frac{C_{11}}{C_{11} + 2C_{12}}a + \frac{2C_{12}}{C_{11} + 2C_{12}}c,$$

where $C_{11}$ and $C_{12}$ are elastic constants, and are given by [15]

$$C_{11} = 11.88 + 2.24x(N/m^2) \quad \text{and} \quad C_{12} = 5.38 + 0.873x(N/m^2),$$

where $x$ is the P fraction of the GaAsP buffer layer. Although the elastic constants depend on the P fraction, both $\frac{C_{11}}{C_{11} + 2C_{12}}$ and $\frac{2C_{12}}{C_{11} + 2C_{12}}$ are almost independent of this fraction. Therefore, $a_{0, \text{GaAsP}}$ could be determined, even though the exact fraction was unknown. The P fraction was determined from the obtained implicit lattice constants using the following equation,

$$a_{0, \text{GaAsP}}(\text{nm}) = 0.5654 - 0.0203x,$$

and the results are tabulated in Table II.

Secondly, the strains of the GaAs well layers and GaAsP barrier layers of the SLs are obtainable if we assume that the layers are tetragonally distorted, and further assume that the P fraction of the GaAsP barrier layer is identical to that of the buffer layer. Furthermore, it was also assumed that the lattice constants along the [100] and [010] directions were the same for both layers, and were determined by the reciprocal positions of the main satellite peaks, denoted by $a_{\text{ST}(0)}$. The strains of the barrier layers, $\varepsilon_{xx, \text{GaAsP}}$, along the specimen surface are thus given by

$$\varepsilon_{xx, \text{GaAsP}} = \frac{a_{\text{ST}(0)} - a_{0, \text{GaAsP}}}{a_{0, \text{GaAsP}}} - \varepsilon_{xx, \text{GaAs}},$$

where $a_{0, \text{GaAs}}$ is the lattice constant of perfect GaAs, 0.5654 nm. Here, the average lattice constant of the SL along the [001] direction was also determined from the reciprocal position. The results are shown in Table II, and are compared with the P fraction. The relationship between $x$ and $\varepsilon_{0, \text{GaAs}}$ was approximately linear with respect to the phosphorous fraction of the GaAsP layer, as shown in Fig. 3.

Finally, the thicknesses of the layers were determined. The strains along the [001] direction are given by $\varepsilon_{zz} = -2\varepsilon_{11}x$, where $C_{11}$ and $C_{12}$ are the elastic constants for the respective layers, and $\varepsilon_{xx}$ are the strains of the respective layers. As the strains of both layers and the P fraction of the barrier layer were obtained, the lattice constants for the layers along the [001] direction were determined, as shown in the Appendix. The periods of the SLs were experimentally determined from the satellite peaks for specimens from which these peaks were observable, and the design values were used for specimens from which only the main satellite peak was observable. The results are

**TABLE III: Thickness of the GaAs well layers ($t_{\text{GaAs}}$) and GaAsP barrier layers ($t_{\text{GaAsP}}$) of the superlattices.**

| No. | $t_{\text{GaAs}}$ (nm) | $t_{\text{GaAsP}}$ (nm) |
|-----|------------------------|------------------------|
| #1  | 2.86                   | 3.14                   |
| #2  | 1.76                   | 4.24                   |
| #3  | 2.55                   | 5.45                   |
| #4  | 3.60                   | 5.50                   |
| #5  | 3.95                   | 5.47                   |
| #6  | 2.90                   | 6.20                   |
| #7  | 3.78                   | 5.17                   |

**TABLE IV: Band gap energy ($E_{\text{th}}$) and the splitting between the heavy hole band and the light hole band ($\Delta_s$).**

| No. | $E_{\text{th}}$ (eV) | $\Delta_s$ (meV) |
|-----|----------------------|------------------|
| #1  | 1.52                 | 46.8             |
| #2  | 1.59                 | 55.0             |
| #3  | 1.60                 | 108              |
| #4  | 1.60                 | 114              |
| #5  | 1.59                 | 117              |
| #6  | 1.54                 | 67.8             |
| #7  | 1.55                 | 86.4             |
FIG. 5: Relationship between superlattice luminescence energy ($E_{ph}$) and calculated band gap energy ($E_{th}$).

FIG. 6: Relationship between band splitting, $\Delta s$, and the maximum polarization.

tabulated in Table III. The obtained thicknesses differed from their design values for some specimens. Therefore, the thicknesses of the SL layers were measured by transmission electron microscopy, and the results for specimen #3 are shown in Fig. 4. The image indicates that the ratio of the thickness of the well layer to that of the barrier layer was almost 1:2, in agreement with the present X-ray diffraction results. These results support the reliability of the present analysis.

Once the strains and thicknesses of the well and barrier layers were known, the band structures of the SLs were calculated on the basis of model solid theory [16]. In Table IV, the band gap energy ($E_{th}$) and the splitting between the heavy hole band and the light hole band ($\Delta s$) are tabulated. Figure 5 shows a comparison of $E_{th}$ with the peak energies of photoluminescence ($E_{ph}$), demonstrating good agreement. The splitting amounts were important to realizing high polarization, and the relation between the band splitting ($\Delta s$) and the maximum polarization were compared, with the results shown in Fig. 6. Figure 6 indicates that the polarization from photocathodes grown directly on GaP substrates was not due to a lack of band splitting. One possible explanation is that before polarized photoelectrons are extracted into vacuum, they become depolarized due to scattering by lattice defects induced by compressive strain, which are different from those induced by tensile strain. The present results support previous results, in which the insertion of a GaAs layer between the GaP substrate and the superlattice effectively yielded 90% polarization [13].

IV. CONCLUSIONS

The strains and thicknesses of GaAs/GaAsP superlattices used in spin-polarized photocathodes grown on GaAs and GaP (001) substrates were determined by X-ray diffraction, and the band structures were calculated. The splitting between the heavy hole band and the light hole band of superlattices grown on GaP substrates was large compared to that of superlattices grown on GaAs substrates. The reason for the low polarization of photocathodes with a GaP substrate was not the lack of band splitting, but depolarization scattering caused by crystal defects, which were different from those present in the superlattices grown on GaAs substrates.

Appendix

The lattice constants of the GaAsP barrier and the GaAs well layers along the [001] direction are given by

\[
c_B = a_0_{GaAsP} (1 + \varepsilon_{zz,GaAsP}) \quad \text{and} \quad (5)
\]

\[
c_W = a_0_{GaAs} (1 + \varepsilon_{zz,GaAs}) \quad \text{and} \quad (6)
\]

respectively. Denoting the layer numbers for the barrier and well layers as $n_B$ and $n_W$, respectively, and denoting thicknesses of these layers as $t_B$ and $t_W$, respectively, the following equations are obtained:

\[
c_{ST(0)} = n_B c_B + n_W c_W, \quad \text{and} \quad (7)
\]

\[
n_Bc_B + n_Wc_W = T, \quad \text{and} \quad (8)
\]

where $c_{ST(0)}$ is the average lattice constant along the [001] direction of the SL, and $T$ is the period of the SL. From the above equations, the layer numbers for the barrier and well layers are given by

\[
n_B = \frac{c_{ST(0)} - c_W}{c_B c_{ST(0)} - c_W T}, \quad \text{and} \quad (9)
\]

\[
n_W = \frac{c_B - c_{ST(0)}}{c_{ST(0)} - c_B T}, \quad \text{and} \quad (10)
\]
and the thicknesses of the barrier and well layers, \( t_B \) and \( t_W \), are given by

\[
\begin{align*}
\quad \quad \quad \quad t_B &= n_B c_B, \quad \quad (11) \\
\quad \quad \quad \quad t_W &= n_W c_W. \quad \quad (12)
\end{align*}
\]

[1] D. T. Pierce and F. Meier, Phys. Rev. B 13, 5484 (1976).
[2] D. T. Pierce, R. J. Celotta, G.-C. Wang, W. N. Unertl, A. Galejs, C. E. Kuyatt, and S. R. Mielczarek, Rev. Sci. Instrum. 51, 478 (1980).
[3] T. Maruyama, E. L. Garwin, R. Prepost, G. H. Zapalac, J. S. Smith, and J. D. Walker, Phys. Rev. Lett. 66, 2376 (1991).
[4] T. Nakanishi, H. Aoyagi, H. Horinaka, Y. Kamiya, T. Kato, S. Nakamura, T. Saka, and M. Tsubata, Phys. Lett. A 158, 345 (1991).
[5] H. Aoyagi, H. Horinaka, Y. Kamiya, T. Kato, T. Kosugoh, S. Nakamura, T. Nakanishi, S. Okumi, T. Saka, M. Tawada, and M. Tsubata, Phys. Lett. A 167, 415 (1992).
[6] T. Maruyama, E. L. Garwin, R. Prepost, and G. H. Zapalac, Phys. Rev. B 46, 4261 (1992).
[7] T. Omori, T. Nakanishi, H. Aoyagi, T. Baba, T. Furuya, K. Itoga, M. Mizuta, S. Nakamura, Y. Takeuchi, M. Tsubata, and M. Yoshioka, Phys. Rev. Lett. 67, 3294 (1991).
[8] T. Omori, T. Kurihara, Y. Takeuchi, M. Yoshioka, T. Nakanishi, S. Okumi, M. Tsubata, M. Tawada, K. Togawa, Y. Tanimoto, C. Takahashi, T. Baba, and M. Mizuta, Jpn. J. Appl. Phys. 33, 5676 (1994).
[9] T. Saka, T. Kato, T. Nakanishi, S. Okumi, K. Togawa, H. Horinaka, T. Matsuyama, and T. Baba, Surf. Sci. 454-456, 1042 (2000).
[10] T. Nishitani, T. Nakanishi, M. Yamamoto, S. Okumi, F. Furuta, M. Miyamoto, M. Kuwahara, N. Yamamoto, K. Naniwa, O. Watanabe, Y. Takeda, H. Kobayakawa, Y. Takashima, H. Horinaka, T. Matsuyama, K. Togawa, T. Saka, M. Tawada, T. Omori, Y. Kurirane, T. Tsubata, T. Kato, and T. Baba, J. Appl. Phys. 97, 094907 (2005).
[11] X.G. Jin, Y. Maeda, T. Saka, M. Tanioku, S. Fuchi, T. Ujihara, Y. Takeda, N. Yamamoto, Y. Nakagawa, A. Mano, S. Okumi, M. Yamamoto, T. Nakanishi, H. Horinaka, T. Kato, T. Yasue, and T. Koshikawa, J. Crystal Growth 310, 5039 (2008).
[12] N. Yamamoto, T. Nakanishi, A. Mano, Y. Nakagawa, S. Okumi, M. Yamamoto, T. Konomi, X.G. Jin, T. Ujihara, Y. Takeda, T. Oshima, T. Saka, T. Kato, H. Horinaka, T. Yasue, T. Koshikawa and M. Kuwahara, J. Appl. Phys. 103, 064905 (2008).
[13] X.G. Jin, N. Yamamoto, Y. Nakagawa, A. Mano, T. Kato, M. Tanioku, T. Ujihara, Y. Takeda, S. Okumi, M. Yamamoto, T. Nakanishi, S. Saka, H. Horinaka, T. Kato, T. Yasue, and T. Koshikawa, Appl. Phys. Express 1, 045002 (2008).
[14] M. Iseki, T. Saka, T. Kato, H. Horinaka, and T. Matsuyama, Jpn. J. Appl. Phys. 46, 502 (2007).
[15] S. Adachi, J. Appl. Phys. 53, 8775 (1982).
[16] C. G. van de Walls, Phys. Rev. B 39, 1871 (1989).