Elimination of local thickness modulation in GaAs/GaAsP strained superlattices for high spin-polarization photocathodes

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Abstract. We successfully developed a transmission-type photocathode, and a high spin-polarization (90%) with a super-high brightness (1.3×10⁷ A cm⁻² sr⁻¹) of electron beam was achieved. In this research, the elimination of thickness modulation of GaAs/GaAsP superlattice by introduction of a GaAs inter-layer on a GaP substrate is the key point to realize the high spin-polarization. The thickness modulation of the superlattice is related with the surface roughness of the buffer layer. The compressive strain introduced in the GaAsP buffer layer on the GaP substrate causes hillock formation, where several degree off-angle surfaces are formed. The GaAs inter-layer deposited on the GaP substrate introduced a tensile strain in the GaAsP buffer layer instead of the compressive strain and relatively smooth GaAsP buffer layer was achieved. The smooth GaAsP buffer layer was attributed to the periodic GaAs/GaAsP superlattice layer growth.

1. Introduction

Highly spin-polarized electron sources are intensively developed for applications in high-energy physics, atomic physics, and studies of thin film and surface magnetism [1]. A strained superlattice structures composed of a GaAs-related semiconductors are known to be most effective as photocathode of the spin-polarized electron beam [2, 3]. In these structures, the heavy hole and light hole mini-bands are largely split due to both the elastic strain and the quantum confinement effect. So the highly spin-polarized electrons are selectively excited by circularly polarized photons only from the heavy hole mini-band to the conduction mini-band, and are then emitted from a negative electron affinity surface. Photocathodes based on a GaAs/GaAsP strained superlattice [4, 5] and an AlInGaAs/AlGaAs strained superlattice [6] provide high spin-polarizations over 90%.

In our previous study [5, 7] we have newly developed a super-high-brightness transmission-type photocathode based on the GaAs/GaAsP strained superlattice. In this novel system, a super-high brightness electron beam was achieved by illuminating a small area on the backside of the photocathode with a pump laser light that was well-focused by a short-focal-length lens. To realize this system, a GaAs/GaAsP strained superlattice was fabricated on a GaP substrate (bandgap energy: 2.26 eV), which is transparent to the pump-laser beam (beam energy: 1.44–1.77 eV), instead of a GaAs (bandgap energy: 1.42 eV) substrate. High spin-polarization (90%) and a super-high brightness (1.3×10⁷ A cm⁻² sr⁻¹) electron beam were reproducibly achieved with this transmission-type photocathode. The key technology to achieve the high spin-polarization in this photocathode is the

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introduction of a 500 nm GaAs inter-layer deposited on the GaP substrate. The spin-polarization of the photocathode without the GaAs inter-layer was approximately 60%.

In this study, the superlattice structure on a GaAs substrate, GaP substrate, and a GaP substrate with the GaAs inter-layer were investigated by cross-sectional transmission electron microscopy (TEM) and atomic force microscopy (AFM). The effect of the GaAs inter-layer on spin-polarization is discussed.

2. Experimental

The photocathodes were grown using a low-pressure metalorganic vapor phase epitaxy system with a vertical cold-wall quartz reactor. Triethylgallium (TEG), tertialybutylphosphine (TBP), and tertialybutylarsine (TBAs) were used as source materials. The substrates were Zn-doped p-type and (001)-oriented GaAs and GaP. All samples were grown at 660°C under a reactor pressure of 76 Torr. The photocathode structures are shown in Figure 1. Following the growth of 1 or 2 µm of the GaAsP buffer layer on the GaAs substrate, the GaP substrate, or the GaP substrate with the GaAs inter-layer, a 12-pair GaAs/GaAsP strained superlattice was grown with a Zn doping to a concentration of $1.5 \times 10^{18}$ cm$^{-3}$. The thickness of the GaAs and GaAsP was 4 nm. Subsequently, each superlattice structure was coated with a highly-doped 5 nm GaAs layer with a Zn dopant concentration of $6 \times 10^{19}$ cm$^{-3}$. The spin-polarizations of the photocathodes on the GaAs substrate and the GaP substrate with the GaAs inter-layer were over 90%, while that on the GaP substrate was only 60%. TEM observation was performed at the electron energy of 200 keV using a Hitachi H-800 electron microscopy. The surface morphology was observed using AFM (NanoScope III).

3. Results and discussion

3.1 TEM image of GaAsP buffer layers and GaAs/GaAsP superlattice layers

Figure 2 shows the cross-sectional TEM images of the GaAsP buffer layers and superlattice layers on the various substrates. A stacking fault extended from the GaAsP buffer layer into the GaAs/GaAsP superlattice layer, as shown in figure 2 (a). In other areas, the period of the superlattice layers are well controlled. Figure 2 (b) shows the TEM image of the superlattice and the GaAsP buffer layer on the GaP substrate. The remarkable feature in this sample is the bump structures. The bump structures take place on the vicinal GaAsP buffer layer in which the surface inclined by about 6° from (001) plane. In the bump structure, the layer thickness in the superlattice is modulated. The thickness modulation increases with the increasing number of the layer pairs (gray/black) and is especially enhanced after the fifth pair. A thicker GaAsP barrier-layer (gray) is grown on a thinner GaAs well-layer (black), while a thin GaAsP barrier-layer is grown on a thicker GaAs well-layer. On the other hand, the periodic superlattice layers are grown on the flat GaAsP buffer layer. The TEM images of the superlattice and the GaAsP buffer layer on the GaP substrate with the GaAs inter-layer are shown in Figure 2 (c). The surface of the superlattice is smooth with no bump structure. The image is similar
to that of the superlattice on the GaAs substrate, i.e., the superlattice layers are periodic and stacking faults penetrate from the buffer layer into the superlattice.

Figure 2. Cross-sectional TEM images of GaAs/GaAsP strained superlattice layer and GaAsP buffer layer on (a) GaAs, (b) GaP, and (c) a GaP substrate with GaAs inter-layer. In (b), bump structures observed in the superlattice layer.

3.2 Surface morphology of GaAsP buffer layer on various substrates

The surface morphology of GaAsP buffer layers were evaluated using AFM and are shown in Figure 3 (a)–(c). In the GaAsP layer on the GaAs substrate, some cracks along [1 1 0] and [1-10] directions were observed as shown in Figure 3 (a). Except the cracks areas, the surface of GaAsP layer shows relatively flat. The surface of the GaAsP layer on the GaP substrate has some anisotropic hillocks as shown in Figure 3 (b). The size of the hillocks structure is approximately 100 nm height and 2 μm wide. The surrounding of the hillocks show vicinal surface and the vicinal surfaces inclined by about 5° from (001) plane. Figure 3 (c) shows the surface of the GaAsP layer on the GaP substrate with GaAs inter-layer. The surface roughness was much better controlled by introduction of the GaAs inter-layer on the GaP substrate.

3.3 Discussion

In the superlattice on the GaAs substrate and the GaP substrate with the GaAs inter-layer, the stacking fault is the main defect and the density is in the range of 10−15 per 10μm. In spintronics research, spin-flip by defect has been reported. In spin-polarized light-emitting diodes (spin-LED)[8], spin-
polarized electrons are injected from ZnMnSe into a GaAs/AlGaAs quantum well (QW), where radiative recombination of the carriers results in the emission of a circularly polarized light. The spin-

polarization of the injected electron correlates inversely with the stacking fault density at the ZnMnSe/AlGaAs interface. According to the experiments, spin-polarization of ~90% can be realized even with the stacking fault density of 10–15 per 10µm. The value of the spin-polarization is almost the same as those in the present study. Therefore, we believe that the effect of the stacking fault (10–15 per 10 µm) on the spin-polarization is not serious.

In the strained superlattice, the heavy hole mini-band and the light hole mini-band are split due to both the quantum confinement and the elastic strain effect. Adjusting the excitation photon energy to the energy gap between the heavy hole mini-band and the conduction mini-band, the spin-polarization of electron beam reaches a maximum value. The energy band gap greatly depends on the thicknesses of the superlattice layer. According to the calculation of the band structure, we found that the energy gap in the thickness modulation area is smaller than that in the periodic area. The difference of the energy gap in both areas is larger than the valence band split. Therefore, it is possible that electrons of both up-spin and down-spin are excited from the heavy hole mini-band in the periodic areas and from the light hole mini-band in the thickness modulated areas by the same energy of the exited photons. We consider the spin-polarization degradation in the superlattice on the GaP substrate is related with the thickness modulation.

In the crystal growth, the thickness modulation of the superlattice on the vicinal surface was reported by several groups [9-11] and it’s mechanism was attributed to the elastic relaxation [10]. The lattice constant of GaAsP is larger than that of GaP, and smaller than that of GaAs. Therefore, a compressive
strain introduces in the GaAsP buffer layer grown on the GaP substrate. The compressive strain usually causes hillock formation, where several degree off-angle surfaces are formed. As shown in Figure 3 (d), the bump structures were formed on the surrounding of the hillocks. The GaAs inter-layer deposited on the GaP substrate introduced a tensile strain in the GaAsP buffer layer instead of the compressive strain and relatively smooth GaAsP buffer layer was achieved. The smooth GaAsP buffer layer was attributed to the periodic GaAs/GaAsP superlattice layer growth. As a result, high spin-polarization of 90 % was achieved by introduction of the GaAs inter-layer on the GaP substrate.

4. Conclusion
Superlattice structure on a GaAs substrate, GaP substrate, and a GaP substrate with a GaAs inter-layer, as spin-polarized photocathodes, were investigated by TEM and AFM. In the superlattice on the GaP substrate, the thickness modulation of the superlattice is the main reason to reduce the spin-polarization. The thickness modulation of the superlattice related with the roughness of the buffer layer. The introduction of the GaAs inter-layer on the GaP substrate controlled the surface roughness of the GaAsP buffer layer and was attributed to the periodic superlattice growth.

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