Observation of Strain-Enhanced Electron-Spin Polarization in Photoemission from InGaAs

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Electron-spin polarization in excess of 70% has been observed in photoemission from a 0.1-μm-thick epitaxial layer of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ with $x \approx 0.13$ grown on a GaAs substrate. Under these conditions, the epitaxial layer is expected to be highly strained by the 0.9% lattice mismatch. The electron polarization and the quantum efficiency have been measured as a function of the excitation photon energy from 1.25 to 2.0 eV. A significant enhancement of the electron polarization occurs in the vicinity of 1.33 eV where the expected strain-induced level splitting permits optical excitation of a single-band transition.

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Polarized electron sources have wide applications in many branches of physics.[1] The use of polarized electron sources with linear electron accelerators is a mature field generally requiring high-intensity sources. For example, the SLAC Stanford Linear Collider requires peak currents of about 16 A in a 2.5-nsec pulse at 120 Hz. These requirements can be met using photoemission from negative-electron-affinity (NEA) GaAs, and this is the technique adopted for linear electron accelerators. The symmetry of GaAs is such that the maximum polarization is limited to 50% due to the valence-band degeneracy of the heavy- and light-hole bands at the Γ point. Much effort has been devoted to achieving higher polarization. The basic approaches involve removing the degeneracy of the valence bands and selectively exciting a single transition for the GaAs-type materials or using crystals such as the ternary chalcopyrites which already have the appropriate band structure.[2] The structures which have been investigated for GaAs include GaAs-AlGaAs superlattices and strained GaAs produced by epitaxial growth of thin GaAs layers on a Si substrate. No significant enhancement of emitted electron-spin polarization has yet been achieved.[3]

This Letter reports the first significant enhancement of electron-spin polarization above 50% from a NEA photocathode. The sample for this experiment was a single heterojunction of InGaAs epitaxially grown on a GaAs substrate under conditions to create pseudomorphic strain by lattice mismatch. Strain-induced changes of band structure in crystals have been extensively studied theoretically as well as experimentally.[4] A suitably thin epitaxial layer of InGaAs grown on a GaAs substrate incorporates a biaxial compressive strain in the plane of the interface and a tensile strain along the growth direction. Full strain is realized when the epilayer is matched to the lattice constant of the substrate without appreciable production of dislocations. The strain alters the band structure of the InGaAs such that the strain-dependent energy difference of the heavy-hole and light-hole bands relative to the conduction band is given by[5]

$$E_0^{C,\mathrm{HH}} = E_0(\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}) + \delta E_H - \delta E_S\,,$$
$$E_0^{C,\mathrm{LH}} = E_0(\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}) + \delta E_H + \delta E_S - (\delta E_S)^2/2\Delta_0 + \cdots\,,$$

where $E_0(\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As})$ is the direct band gap of fully relaxed InGaAs and Δ_0 is the spin-orbit splitting. The quantities δE_H and δE_S represent the hydrostatic shift of the center of gravity of the $P_{3/2}$ multiplet and the linear splitting of the $P_{3/2}$ multiplet, respectively, and are given in terms of the biaxial strain ϵ parallel to the interface by

$$\delta E_H = 2a[(C_{11} - C_{12})/C_{11}]\epsilon\,,$$
$$\delta E_S = b[(C_{11} + 2C_{12})/C_{11}]\epsilon\,,$$

where the parameters a and b are the interband hydrostatic pressure and uniaxial deformation potentials, respectively, and the C_{ij} are the elastic-stiffness constants appropriate to the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ crystal structure. Since the biaxial strain ϵ is compressive for the present structure, the effect of strain is to increase the band-gap energy of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ and remove the degeneracy of the heavy-hole and light-hole levels such that the heavy-hole band moves up in energy and the light-hole band moves down relative to the unstrained case. Using deformation potentials and elastic constants for $x = 0.13$ and assuming full strain, one obtains a valence-band energy splitting $E_0^{C,\mathrm{HH}} - E_0^{C,\mathrm{LH}} = 61$ meV for a biaxial stress parallel to [100] and [010].[5] The corresponding heavy-hole and light-hole to conduction-band energy differences are 1.29 and 1.35 eV, respectively, for $T=293$ K. This energy splitting makes it possible to preferentially excite the heavy-hole band by tuning the excitation photon energy. An electron-spin polarization of higher than 50% is then in principle possible for photoexcited electrons which have thermalized to the bottom of the conduction band. The heavy-hole band to conduction-band transition would result in 100% electron-spin polarization in the absence of depolarizing effects. A tensile strain in the plane of the interface, on the other hand, has the effect of the light-hole band lying higher in energy and the resulting polarization becomes -100%, opposite in sign from the 50% polarization obtained when the heavy-hole and light-hole bands are degenerate.

The single heterostructures were grown at the Electronics Research Laboratory of the University of California at Berkeley (UCB). The samples grown were a thin strained InGaAs epilayer (thin sample) and a thick relaxed InGaAs layer (thick sample). The substrate material used was (100) n-type GaAs (Si doped to 5×10^{18} cm^{-3}). Since heavy p-type doping is necessary to achieve a NEA surface, GaAs buffer layers were grown at 600 °C to change the carrier type: a 0.6-μm-thick n-type GaAs (Si doped to 6×10^{18} cm^{-3}) followed by a 0.2-μm-thick p-type GaAs (Be doped to 6×10^{18} cm^{-3}). The substrate temperature was then reduced to 550 °C for the growth of the InGaAs layer. The thin sample had a 0.1-μm-thick p-type $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ layer (Be doped to 2×10^{18} cm^{-3} for the first 600 Å and to 4×10^{18} cm^{-3} for the final 400 Å), and the thick sample a 1.14-μm-thick p-type $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ layer (Be doped to 2×10^{18} cm^{-3} for the first 11 000 Å and to 4×10^{18} cm^{-3} for the final 400 Å). The indium concentration was nominally $x=0.13$, and the agreement between the indium concentration of the two samples was estimated to be $\delta x=\pm0.01$. After the molecular-beam-epitaxy growth, the sample was cooled to room temperature and was exposed to an As_4 beam for about 10 min to deposit a 200–500-Å arsenic protection layer.

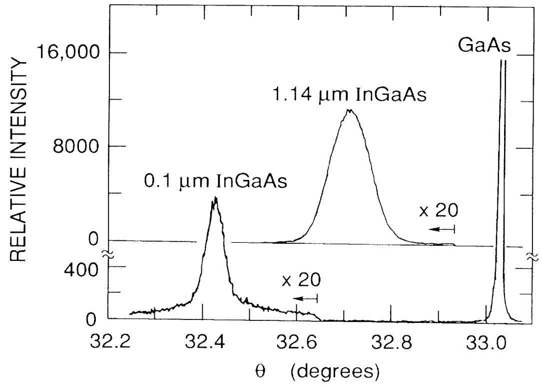

FIG. 1. X-ray rocking curves for the (004) reflection from the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ heterostructures.

In order to estimate the crystal strain in the InGaAs layer, the samples were analyzed with a double-crystal x-ray diffractometer in the Department of Materials Science and Engineering at the University of Wisconsin. Figure 1 shows the x-ray rocking curves around the (004) reflection using the Cu $K\alpha$ line. From these rocking curves, the lattice spacing along the growth direction of the epilayers can be measured precisely. Since the 1.14-μm-thick sample is sufficiently thick to be fully relaxed,[6] the observed lattice spacing of this sample ($a_{\mathrm{InGaAs}}=5.7023$ Å) was used to determine the In concentration giving $x=0.123$. As a result of the biaxial strain in the heterostructure interface, the InGaAs peak of the 0.10-μm-thick sample shifts towards a smaller Bragg angle reflecting a lattice expansion along the [001] direction. The observed lattice spacing $a^{\perp}=5.7470$ Å measures the perpendicular strain to the heterostructure interface $\epsilon^{\perp}=(a^{\perp}-a_{\mathrm{InGaAs}})/a_{\mathrm{InGaAs}}$. The biaxial strain $\epsilon^{\parallel}$ in the plane of the interface is related to the perpendicular strain $\epsilon^{\perp}$ by the expression[7]

$$\epsilon^{\parallel}=-(C_{11}/2C_{12})\epsilon^{\perp}\,.$$

The biaxial strain thus measured for the 0.10-μm-thick sample was $\epsilon=\epsilon^{\parallel}=-0.008\,59$. Given the uncertainty of the indium concentration of the 0.10-μm-thick sample, this strain corresponds to between 80% and 100% of the fully allowed strain.

The electron-spin polarization was measured by Mott scattering at 65 keV. The electron-gun and Mott-scattering apparatus have been described elsewhere.[8] A nitrogen-ion pumped dye laser, using various dyes to obtain the desired wavelength range, was used as the photoexcitation source. The dye laser output from the various dyes varied from 0.5 to 20 μJ/pulse in a 5-nsec pulse with a linewidth of ≈0.1 nm. Prior to installation in the source, the sample was degreased sequentially in boiling solutions of trichloroethylene, acetone, and methanol. After the sample was installed and the chamber evacuated, the gun was baked at 220 °C for about 80 h and at 150 °C for 24 h to achieve the necessary ultrahigh vacuum. During the bake, the sample was maintained at about 270 °C by a resistive heater at the back of the cathode support structure. The final pressure during the subsequent polarization measurements was less than 10^{-10} Torr. The cathode was activated to obtain a NEA surface using cesium and nitrogen trifluoride. Prior to activation, the cathode was heat treated for 1–2 h at 400–425 °C. The lower than usual temperature (typically 600 °C for GaAs) was used to protect the InGaAs layer and the heterostructure. The arsenic cap layer was removed during the first heat treatment. All the present measurements were made with the cathode at room temperature.

The photoelectric quantum efficiencies were measured using two methods. A 2.2-mW HeNe laser was used to measure the absolute quantum efficiency at a photon wavelength of 632.8 nm. A halogen lamp and a monochromator, whose output was monitored by a photodiode, were used to measure the relative quantum efficiency as a function of photon energy, and these measurements were then normalized to the HeNe laser measurement at 632.8 nm.

Figure 2 shows the cathode quantum efficiency as a function of excitation photon energy for the two samples. The band-gap energies of GaAs and relaxed InGaAs are also indicated in the figure. In the energy region greater than about 1.41 eV, both GaAs and InGaAs layers contribute to the photoemission. But the contribution of the InGaAs layer remains at about 10% of the total photoemission for both samples. In the energy region smaller than 1.41 eV, the photoemission from GaAs is observed to diminish sharply as the excitation photon energy approaches the GaAs band gap, and where the major contribution to the photoemission can come only from the InGaAs layer. Photoemission was observed down to 1.21 eV indicating that a NEA surface was successfully achieved.

Figures 3(a) and 3(b) show the measured electron-spin polarization as a function of excitation photon energy for the 0.1- and 1.14-μm-thick samples, respectively. The experimental uncertainty shown in the figure includes the statistical error only. In the energy region greater than 1.41 eV, the observed electron polarization is consistent with the previous measurements on thin-GaAs samples.[9] Although electrons produced in the InGaAs layer can also contribute in this region with a lower polarization, their contribution to the net spin polarization is negligible because of the much larger photoemission from the GaAs layer. However, in the energy region between 1.34 and 1.40 eV where the photoemission from GaAs is diminished, the spin polarization is lower due to lower spin polarization in photoemission from the InGaAs layer. In the energy region smaller than 1.34 eV, the spin polarizations of the two samples show a significant difference. The polarization of the 0.10-μm-thick sample is observed to increase sharply at about 1.34 eV reaching 71% at about 1.26 eV. The sharp enhancement at about 1.34 eV corresponds to the expected gap energy between the light-hole band and the conduction band for an indium concentration between 0.121 and 0.133, values consistent with the uncertainty in the indium concentration and the thin-sample x-ray analysis. The corresponding energy range is indicated by the shaded region in Fig. 3(a). The sample still has significant photoemission and high polarization at 1.25 eV which is beyond the expected heavy-hole to conduction-band energy difference of 1.30 eV. This can be explained by a partial strain relaxation in the epilayer causing the heavy- and light-hole bands to converge towards the relaxed InGaAs valence band located at 1.25 eV. The partial strain relaxation may also explain why the maximum polarization did not reach 100%. On the other hand, the polarization of the 1.14-μm-thick sample remains at 40% and does not show any enhancement. The observed 40% polarization is consistent with the values measured for other bulk III-V compounds. The strained sample was repeatedly heat cleaned, cathode activated, and the spin polarization remeasured. The high polarization observed was reproducible after many heat-treatment cycles, indicating that the heterostructure and the layer strain were preserved under the 400 °C activation temperature environment. After the polarization measurements the sample was reanalyzed with x-ray diffraction. The InGaAs peak showed a 40-arcsec shift from the original location and had a peak broadening, indicating some strain relaxation due to the thermal treatments.

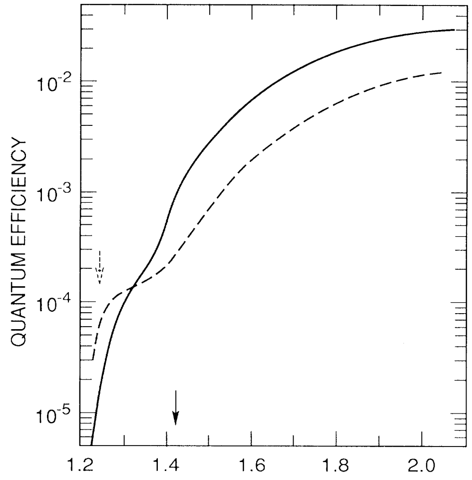

FIG. 2. Cathode quantum efficiency as a function of excitation photon energy. The solid curve is for the 0.1-μm-thick sample, and the dashed curve for the 1.14-μm-thick sample. The band-gap energies of GaAs (solid arrow) and relaxed InGaAs (dashed arrow) are indicated.

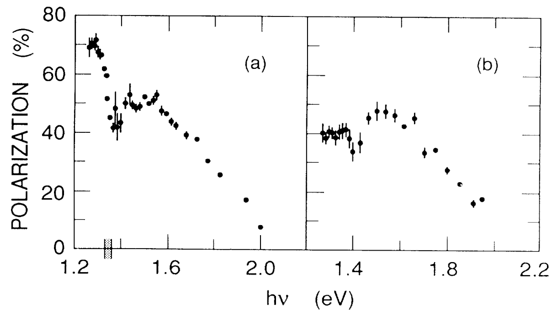

FIG. 3. Electron-spin polarization as a function of excitation photon energy (a) for the 0.1-μm-thick sample and (b) for the 1.14-μm-thick sample. The shaded region in (a) shows the expected light-hole to conduction-band energy difference compatible with the indium concentration uncertainty and the thin-sample x-ray analysis.

There is a systematic uncertainty in the absolute polarization measured by Mott scattering. The present polarized electron gun and Mott polarimeter were previously used for the SLAC inelastic-electron-scattering experiment which observed a parity-violating asymmetry.[10] During that experiment, the electron polarization was measured regularly by high-energy elastic electron-electron scattering (Møller scattering) and compared to the polarization measurements using the present Mott apparatus. From this comparison, the average ratio of Møller to Mott polarization was found to be 0.979 ±0.011.[8,11] Electron-polarization measurements have been made frequently with the Mott apparatus since the parity-violation experiment and no indications of calibration drift have been found. The systematic uncertainty of the present measurements is thus estimated to be $\delta P_{e^-}/P_{e^-} = \pm 0.05$, which is the estimated systematic uncertainty of the Møller polarimeter.

In conclusion, spin polarization has been measured for photoemitted electrons from strained and unstrained InGaAs layers. Polarization in excess of 70% was observed for the 0.1-μm-thick strained InGaAs sample. Polarization for the 1.14-μm-thick unstrained sample was 40% and consistent with the values of bulk III-V compounds. It is concluded that the observed polarization enhancement of the strained sample is due to the strain-induced energy-level splitting of the valence band. This measurement is the first observation of strain-enhanced electron-spin polarization for photoemitted electrons.

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