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Temperature dependence of hole mobility in $GaAs_{1-x}Bi_x$ alloys

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The Hall mobility of holes has been measured in GaAs grown at low temperatures and in $GaAs_{1-x}Bi_x$ alloys for Bi concentrations x ranging from 0.94% to 5.5%. The hole mobility is found to decrease with increasing Bi content. The temperature dependence of the mobility in the 25 to 300 K range is fit with a combination of phonon scattering, ionized impurity scattering, and Bi related scattering. The hole scattering cross-section for an isolated Bi impurity is estimated to be 0.2 nm^2. The temperature independent mobility at the highest Bi concentration (x=5.5%), is interpreted as being limited by scattering from Bi clusters. © *2010 American Institute of Physics*.
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I. INTRODUCTION

Gallium arsenide bismide ($GaAs_{1-x}Bi_x$) is an emerging III-V semiconductor alloy with potential applications in high efficiency solar cells, heterojunction bipolar transistors and long wavelength light emitters. The transport properties of the $GaAs_{1-x}Bi_x$ alloys are important for device applications but so far there are no published measurements of the hole mobility and no transport measurements for Bi concentrations $x>2.5\%$.[1,2] The hole mobility is of particular interest as Bi alloying introduces nearly localized Bi $6p$ orbitals that are similar in energy to the valence band maximum (VBM), which can be expected to strongly affect hole transport. The same Bi orbitals are responsible for the large reduction in band gap with Bi incorporation. In this paper, measurements of the temperature dependence of the Hall mobility of holes in $GaAs_{1-x}Bi_x$ alloys as a function of temperature, in the range 25 to 300 K, for Bi concentrations as high as 5.5% are presented.

The growth of $GaAs_{1-x}Bi_x$ requires rather different conditions than those normally used for GaAs growth, namely, low temperature and nearly stoichiometric As:Ga ratios.[3] Under conventional GaAs growth conditions Bi surface segregates or evaporates and does not incorporate. Growth of GaAs at low temperatures without Bi normally leads to films with poor electronic properties.[4] Nevertheless the bismide alloys grown under these unconventional conditions show strong electro- and photoluminescence.[5,6] In the dilute nitride alloys GaN_xAs_{1-x}, a small amount of incorporated N greatly reduces the electron mobility[7,8] Analogously, Bi is expected to effect the hole mobility.[9]

II. EXPERIMENT

The GaAs and $GaAs_{1-x}Bi_x$ samples were grown on semi-insulating GaAs (001) substrates in a molecular beam epitaxy system equipped with Knudsen cells for Ga and Bi, along with a valved two-zone cracker source for As_2. Optical band gap thermometry is used to measure the sample temperature. After removing the surface oxide, a thick (>300 nm) GaAs buffer layer was grown under conventional GaAs growth conditions at a growth rate of 1.0 μm/h. p-type GaAs epilayers were also grown under these conditions without Bi, as reference samples. In the case of the $GaAs_{1-x}Bi_x$ samples, the Ga flux was reduced to achieve a growth rate of 0.1 μm/h and the growth was interrupted for 10 min following the buffer layer. During the interruption the growth temperature was reduced to 350 C, the As flux was reduced to nearly stoichiometric levels and the Bi flux initiated just before the Ga shutter reopened. The interruption reduces the amount of low temperature GaAs that is deposited. The $GaAs_{1-x}Bi_x$ layers were grown to thicknesses of 250 to 400 nm with Bi concentrations as high as 5.5%. The Bi concentration was controlled by adjusting the flux of Bi atoms and As_2 overpressure. The samples were doped p-type with a CBr_4 gas source controlled by a mass flow controller. $GaAs_{1-x}Bi_x$ samples were doped between 5.1×10^{17} and 2.5×10^{18} cm^{-3} (depletion widths are 45 nm and 20 nm, respectively). At these doping levels the bismide epilayers were significantly thicker than the depletion width, assuming that the Fermi energy is pinned at midgap at the surface.

For the electrical measurements, small Ti/Pt/Au Ohmic contacts[10] were deposited by e-beam evaporation onto the corners of cleaved 7 mm squares and then annealed at 450 C for 30 s to decrease their resistivity. The samples were wet etched with HCl prior to contact deposition to remove surface oxide. Sheet resistance, carrier concentration and mobility were measured using the van der Pauw method; volume carrier density is based on epilayer effective thickness, where the depletion width has been subtracted. All Hall effect measurements were carried out in a magnetic field of 0.265 T. The temperature dependence of the hole mobility was measured in a closed cycle helium cryostat, in the temperature range from 300 to 25 K. Measurements of an undoped GaAs epilayer showed the GaAs buffer layers in the device structure to be highly resistive.

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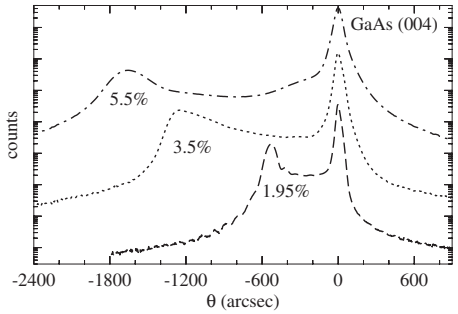

FIG. 1. High resolution x-ray rocking curves of the GaAs (004) peak showing a split off peak for various Bi compositions. Data is vertically offset for clarity.

III. RESULTS

The Bi content of the films was measured by high resolution x-ray diffraction (XRD). Figure 1 shows (004) rocking curves for three $GaAs_{1-x}Bi_x$ samples. Sample composition is determined from the separation of the film peak from the GaAs (004) peak.[3] Off-axis (115) scans of a 270 nm thick 3.5% GaAsBi sample (not shown) indicate that this film is pseudomorphically strained to match the GaAs lattice constant in the plane of the film. The Matthews–Blakeslee critical thickness for this composition is 140 nm.[11] It is believed that all our samples are pseudomorphically strained to match the substrate. Photoluminescence measurements show all samples to be strong light emitters, similar to Lu *et al.*[5] This result is consistent with our interpretation that the films are pseudomorphically strained and free of misfit dislocations.

Several GaAs reference samples were measured to quantify the effect of growth conditions on the hole mobility, as shown in Fig. 2. These samples were grown under the following conditions: (1) conventional GaAs growth conditions, T_{subs}=580 C and As:Ga ratio ≃8 (□); (2) low temperature, T_{subs}=350 C and As:Ga ratio ≃8 with no Bi flux (◇); (3) low temperature, low As:Ga, T_{subs}=350 C, and As:Ga ratio 1–2 (◆); and (4) low temperature, low As:Ga with Bi flux but with a sufficiently high As_2 flux that there is no detectable Bi incorporation detected by XRD (●). GaAs samples grown under conventional conditions have hole mobilities similar to literature values for their doping levels,[12] while GaAs samples grown at low temperatures and without Bi show a reduced hole mobility. A decrease in mobility for

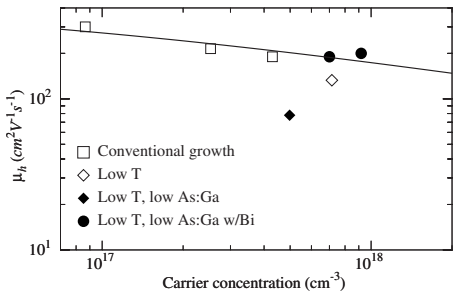

FIG. 2. GaAs hole mobilities for samples grown under various growth conditions. The solid line is the GaAs hole mobility reported in Adachi (Ref. 12). Hole mobility is reduced for growth at low temperatures and low As:Ga. Samples grown under these conditions, with Bi present as a surfactant, show no reduction in mobility.

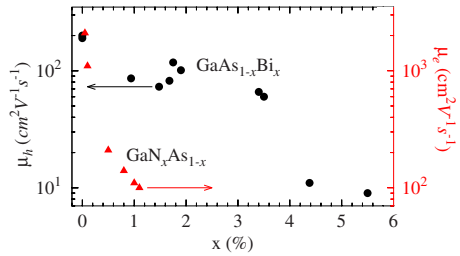

FIG. 3. (Color online) $GaAs_{1-x}Bi_x$ hole mobility and GaN_xAs_{1-x} electron mobility (Ref. 8) for various concentrations, x.

GaAs grown under these conditions is consistent with previous work by Melloch *et al.*[4] GaAs samples grown at low temperature, low As:Ga flux ratio and with Bi flux but having no Bi incorporation show similar hole mobility to the GaAs samples grown using conventional conditions. In earlier experiments, the presence of a Bi surfactant was found to reduce surface roughness and increase photoluminescence intensity.[13] From these results it is concluded that the reduction in hole mobility in the $GaAs_{1-x}Bi_x$ samples is solely due to the incorporation of Bi and not to changes in the structure or stoichiometry of GaAs associated with the nonstandard growth conditions.

Figure 3 shows the hole mobilities of a series of $GaAs_{1-x}Bi_x$ samples, which are doped in the range 1.2×10^{17} to 2.5×10^{18} cm^{-3}. GaAs samples grown using bismuth as a surfactant from Fig. 2 are shown at x=0. Also shown is electron mobility data for GaN_xAs_{1-x} (Ref. 8) samples. The hole mobility is found to decrease with Bi concentration, however, the decrease in hole mobility is less dramatic than in the case of electron mobility in GaN_xAs_{1-x}.

The temperature dependence of the hole mobility is shown in Fig. 4 for $GaAs_{1-x}Bi_x$ samples with 0.94%, 1.95%, 3.5%, and 5.5% Bi content, along with a GaAs sample grown at 350 °C with Bi surfactant. Two factors limit the hole mobility of the GaAs: phonon scattering and ionized impurity scattering. An additional temperature independent scattering mechanism related to Bi incorporation is consid-

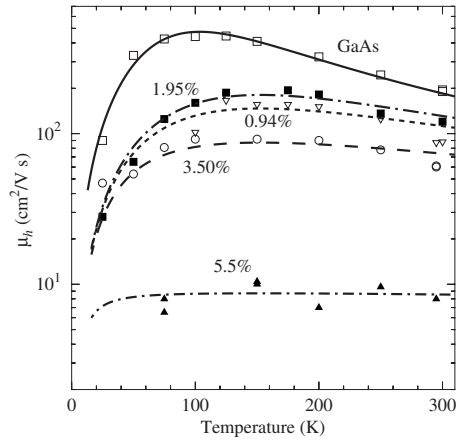

FIG. 4. Temperature dependence of the hole mobility for p-type GaAs and four $GaAs_{1-x}Bi_x$ samples. The GaAs sample was grown at 350 C with an As:Ga ratio of 1–2 and bismuth as a surfactant. The mobility is dominated by phonon and Bi related scattering at high temperatures and by ionized impurity scattering at low temperatures.

TABLE I. Fit parameters for the temperature dependence of the mobility for various Bi concentrations. Units for the coefficients can be inferred from Eq. (1). Thicknesses are based on growth rate calibrations and fringes in XRD spectra. Blank boxes in the table indicate that the factor is not included in the fit. C_{Bi} is used as the fitting parameter for the Bi containing samples.

[Bi] (%)	0%	0.94%	1.95%	3.5%	5.5%
Thickness (nm)	260	260	250	270	300
Doping (cm^{-3})	7.8×10^{17}	5.1×10^{17}	8.2×10^{17}	1.2×10^{18}	5.1×10^{17}
μ_{RT} ($\mathrm{cm}^2/\mathrm{V\ s}$) $\pm5\%$	200	88	100	60	8
C_{ph}	1.0×10^{6}	1.0×10^{6}	1.0×10^{6}	1.0×10^{6}	⋯
C_I	0.9	0.28	0.28	0.28	0.28
C_{Bi}	⋯	330	570	130	9

ered for the $GaAs_{1-x}Bi_x$ samples. Assuming that each of these mechanisms act independently, we can add the terms as follows:

$$\frac{1}{\mu}=\frac{T^{1.5}}{C_{ph}}+\frac{1}{C_I T^{1.5}}+\frac{1}{C_{\mathrm{Bi}}}. \tag{1}$$

In this equation the temperature dependence of the various scattering mechanisms is shown explicitly and the subscripts in the denominators *ph*, *I*, and Bi indicate phonon, ionized impurity, and Bi related scattering, respectively. The temperature dependences of the phonon scattering and ionized impurity scattering are taken from Adachi.[12] The Bi related scattering is taken to be independent of temperature based on the data for the 5.5% sample. A weak temperature dependence is consistent with the theoretical results of Fahy *et al.*[14] Fit parameters for the various Bi contents are given in Table I. The fits are not very sensitive to the ionized impurity scattering, and, therefore, they do not provide an accurate determination of the coefficient for ionized impurity scattering.

Kinetic theory can be used to express the contribution to the scattering by Bi in terms of the scattering cross-section, σ_{Bi}

$$\mu_{\mathrm{Bi}}=C_{\mathrm{Bi}}=\frac{e}{m^* v_{th}\sigma_{\mathrm{Bi}}n_{\mathrm{Bi}}}. \tag{2}$$

In this expression m^* is the average effective mass of holes, v_{th} is the thermal velocity of the holes, and n_{Bi} is the number density of Bi atoms. For the most dilute concentration sample, [Bi]=0.94%, Bi related scattering is likely related to isolated Bi atoms rather than clusters of bismuth atoms. Based on the fit to the temperature dependence, the value of C_{Bi} for this samples is used to infer a value for σ_{Bi}. Substituting $m^*=0.51m_e$, $v_T=1.7\times10^7$ cm/s and $n_{\mathrm{Bi}}=4.1\times10^{20}$ cm^{-3} into Eq. (2), we find $\sigma_{\mathrm{Bi}}=0.2$ nm^2.

Fahy *et al.*[14] provide an expression for the scattering cross-section for individual N atoms in dilute GaN_xAs_{1-x}, as follows

$$\sigma_{\mathrm{Bi}}=\frac{1}{16\pi}\left(\frac{m^*}{\hbar^2}\right)^2\left(\frac{dE_c}{dx}\right)^2 a_o^6, \tag{3}$$

where a_o is the GaAs lattice constant and dE_c/dx is the change in band gap with increasing nitrogen concentration. This expression is used to estimate the scattering cross-section from individual Bi atoms. In order to apply this expression to Bi atom scattering in $GaAs_{1-x}Bi_x$, dE_c/dx is replaced by dE_g/dx, where E_g is the band gap of the bismide alloy.[15] It is assumed that the change in bandgap is entirely due to a shift in the valence band. Using $(dE_g/dx)=8.8$ eV we find $\sigma_{\mathrm{Bi}}=2.0$ nm^2, which is 10× larger than the value for isolated Bi atoms found above of 0.2 nm^2.

As shown in Fig. 3 the electron mobility in the dilute nitride alloy GaN_xAs_{1-x} drops rapidly with increasing N content. In Ga(In)NAs alloys, N related states are resonant with the conduction band minimum and are believed to be responsible for the sharp drop in the electron mobility. Although the hole mobility in $GaAs_{1-x}Bi_x$ also decreases with increasing Bi content, the reduction is not as strong. For the Bi concentration $x=0.94\%$, the probability that a particular Bi atom will have a Bi atom next nearest neighbor (Bi dimer) is 10%, taking into account the 12 next nearest neighbor sites and assuming that the Bi atoms are randomly distributed on the As lattice. For [Bi]=5.5%, the probability that a Bi atom has at least one Bi next nearest neighbor approaches 50%, and the density of trimers and other larger clusters increases dramatically. Due to the proximity of the Bi orbitals to the VBM, it is likely that Bi clusters will have states that are resonant with the VBM. These states are likely to have large scattering cross sections for holes. Applying Eq. (2) to the 5.5% sample, a per atom scattering cross-section of 2.0 nm^2 is found. This is an order of magnitude larger than the value for the low concentration sample. This is an indication that there is scattering from Bi clusters, and/or an increase in hole effective mass.[16] Therefore, we suggest that the temperature independent mobility for the sample with [Bi]=5.5% is controlled by scattering from Bi cluster states.

IV. CONCLUSION

The growth of GaAs under conditions necessary for Bi incorporation, low temperatures and nearly stoichiometric As:Ga flux ratios, result in a reduction in hole mobility. This reduction is not observed in GaAs samples grown under the same conditions when Bi is used as a surfactant. It was found that incorporation of Bi in $GaAs_{1-x}Bi_x$ resulted in a reduced hole mobility. The scattering cross-section of an isolated bismuth in GaAs was estimated to be $\sigma_{\mathrm{Bi}}=0.2$ nm^2 in an $x=0.94\%$ concentration sample. The modest degradation in transport properties in the ternary bismide alloys relative to the binary parent compound, when compared with the dilute nitrides alloys, is a promising indicator for future device applications of the bismide alloys.

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