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Magnetotunneling Spectroscopy of Dilute Ga(AsN) Quantum Wells

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We use magnetotunneling spectroscopy to explore the admixing of the extended GaAs conduction band states with the localized N-impurity states in dilute GaAs$_{1-y}$N$_y$ quantum wells. In our resonant tunneling diodes, electrons can tunnel into the N-induced $E_-$ and $E_+$ subbands in a GaAs$_{1-y}$N$_y$ quantum well layer, leading to resonant peaks in the current-voltage characteristics. By varying the magnetic field applied perpendicular to the current direction, we can tune an electron to tunnel into a given $k$ state of the well; since the applied voltage tunes the energy, we can map out the form of the energy-momentum dispersion curves of $E_-$ and $E_+$. The data reveal that for a small N content ($\sim 0.1\%$) the $E_-$ and $E_+$ subbands are highly nonparabolic and that the heavy effective mass $E_+$ states have a significant $\Gamma$-conduction band character even at $k = 0$.

The incorporation of low concentrations of N ($y = 0\% - 5\%$) in GaAs leads to a number of unusual physical properties [1–3]. In the ultradilute regime ($y < 0.01\%$), N introduces a single-impurity level at an energy of $\sim 0.2$ eV above the conduction band minimum of GaAs [4]. A further increase of N causes a large band-gap bowing with a band-gap reduction of about 0.1 eV per atomic percent of N content [1–3,5,6]. This has raised the interesting possibility of using N-containing alloys grown in GaAs substrates for long-wavelength optoelectronic applications [7]. At present, research on GaAs$_{1-y}$N$_y$ represents one of the most active topics in semiconductor physics. However, despite years of extensive studies, there is not yet direct experimental evidence that the conduction band of GaAs$_{1-y}$N$_y$ has the hybridized character and highly nonparabolic energy–wave-vector dispersion that has been predicted by theory [5,6,8–11]. According to band anticrossing models, the interaction of the extended $\Gamma$-conduction band states of GaAs with the localized N energy level causes a splitting of the conduction band into two nonparabolic subbands $E_-$ and $E_+$, as well as to additional states. The coexistence of different local environments in the alloy also complicates the electronic properties of GaAs$_{1-y}$N$_y$: Single N impurities form perturbed delocalized states in the host lattice whereas N aggregates, such as impurity N-N pairs and higher-order clusters, form strongly localized states; these two different types of states can admix to form a so-called “amalgamated” conduction band, thus leading to a duality of bandlike and localization behaviors [3,8,9].

In this Letter, we explore the electronic states of GaN$_y$As$_{1-y}$ quantum well (QW) layers using magnetotunneling spectroscopy (MTS). The MTS technique has been previously used to study the electronic properties of different semiconductor heterostructures [12–14] including valence band QWs [12] and quantum dots [14]. Here we show that the MTS can provide us with a powerful means of exploring the admixing of the extended conduction band states of GaAs with the localized N-impurity states. In our resonant tunneling diodes (RTDs), electrons can tunnel into the N-induced $E_-$ and $E_+$ subbands in a GaN$_y$As$_{1-y}$QW layer, thus leading to resonant peaks in the current-voltage characteristics, $I(V)$. Varying a magnetic field applied perpendicular to the current direction allows us to tune an electron to tunnel into a given $k$ state of the well; the voltage tunes the energy so that we can map out the energy–wave-vector dispersion $e(k)$ curves of the $E_-$ and $E_+$ subbands. The data reveal that, for a small N content ($\sim 0.1\%$), the $E_-$ and $E_+$ subbands are highly nonparabolic and that the heavy effective mass $E_+$ states have a significant $\Gamma$-conduction band character even at $k = 0$.

In this study, we used a series of five $n-i-n$ GaAs/Al$_{0.4}$Ga$_{0.6}$As/GaAs$_{1-y}$N$_y$ RTDs grown by molecular beam epitaxy (MBE) on (100)-oriented $n$-type (Si: $1 \times 10^{18}$ cm$^{-3}$) GaAs substrates. In our samples, an 8 nm thick GaAs$_{1-y}$N$_y$ $(y = 0.118, 0.43, 0.93$, and 1.55%) layer is embedded between two 6 nm thick Al$_{0.4}$Ga$_{0.6}$As tunnel barriers; undoped GaAs spacer layers of width 50 nm separate the Al$_{0.4}$Ga$_{0.6}$As barriers from $n$-doped GaAs layers in which the dopant concentration is increased from $2 \times 10^{17}$ cm$^{-3}$, close to the barrier, to $2 \times 10^{18}$ cm$^{-3}$. The samples were processed into mesas with diameters ranging between 25 and 400 $\mu$m, with a ring-shaped metallic top contact layer to provide optical access for $I(V)$ measurements under illumination with above-band gap laser light (633 nm).

The inset of Fig. 1 shows schematically the conduction band profile of our RTDs. At zero bias, equilibrium is established by electrons diffusing from the doped GaAs layers into the N-induced localized states in the...
GaAs$_{1-y}$N$_y$ layer, e.g., N-cluster states and N-related defects with energies below the GaAs conduction band edge [15]. Negative charge in the well gives rise to two depletion layers in the regions beyond the Al$_{0.4}$Ga$_{0.6}$As barriers and a corresponding band bending. When a voltage is applied, resonant tunneling through a particular subband state in the GaAs$_{1-y}$N$_y$ QW layer gives rise to a peak in $I(V)$, whenever this state is resonant with an adjacent filled state in the negatively biased electron emitter layer. As shown in Fig. 1, when $y$ is increased from 0 to 0.08%, the resonance $E_0$ due to electrons tunneling through the lowest bound state of the QW splits into two main features, $E_{0-}$ and $E_{0+}$. These can be seen more clearly in the $I(V)$ measured under illumination (see dashed line in Fig. 1). In contrast, as shown in Fig. 2, further increase of $y$ smears out the resonances (weak features are revealed in the differential conductance, $dI/dV$, plot), quenches the current, and shifts to higher biases the threshold voltage, $V_{th}$, at which the current increases rapidly.

The monotonic shift of $V_{th}$ to higher biases with increasing $y$ is consistent with an increasing charge in the well due to formation of denser low-energy N states that act as traps of electrons. The negative charge in the well and the resulting bending of the conduction band become more pronounced with increasing $y$ and tend to inhibit the current flow at low voltages. The smearing of the resonances at large $y$ is likely to be due to an increasing disorder in the system. In the absence of disorder, both the energy and the in-plane momentum of the electron are conserved in the tunneling process; these are the conditions required to observe a peak in the $I(V)$ curve. At low $y$ ($y = 0.08\%$), the disorder in the well is weak and tunneling of electrons through the QW subbands leads to the peaks $E_{0-}$ and $E_{0+}$ in $I(V)$. A further increase of $y$ increases the probability of alloy fluctuations and formation of N clusters with strongly localized levels. The destruction of translational symmetry due to disorder tends to break the momentum conservation condition and smears out the resonances, as is observed even for $y = 0.08\%$. The resonances observed for this sample can be enhanced by optical excitation. This enhancement is probably due to the effect of screening of the disorder in the QW by the photogenerated holes and to the effect on the current of hole recombination with majority electrons through the QW by the photogenerated holes and to the effect on the current and in-plane momentum of the electron are conserved in the tunneling process; these are the conditions required to observe a peak in the $I(V)$ curve. At low $y$ ($y = 0.08\%$), the disorder in the well is weak and tunneling of electrons through the QW subbands leads to the peaks $E_{0-}$ and $E_{0+}$ in $I(V)$. A further increase of $y$ increases the probability of alloy fluctuations and formation of N clusters with strongly localized levels. The destruction of translational symmetry due to disorder tends to break the momentum conservation condition and smears out the resonances, as is observed even for $y = 0.08\%$. The resonances observed for this sample can be enhanced by optical excitation. This enhancement is probably due to the effect of screening of the disorder in the QW by the photogenerated holes and to the effect on the current of hole recombination with majority electrons tunneling in the N-related states [16].

A magnetic field, $B$, applied parallel to the QW plane $(X,Y)$ is used to probe in detail the $I(V)$ curves. For $y > 0.08\%$, the $I(V)$ are almost unaffected by the magnetic field up to the available field of 12 T, thus suggesting that the current is due to tunneling of electrons through localized N-related states, as was also observed for similar RTDs with $y \sim 2\%$ [17]. In contrast, resonances $E_{0-}$ and $E_{0+}$ in the $I(V)$ plot of the RTD with $y = 0.08$ are strongly affected by $B$. Figures 3(a) and 3(b) show the $B$ dependence of the $I(V)$ plots under illumination for $y = 0.08\%$. An identical $B$ dependence of the resonant peaks in the $I(V)$ curves was also measured in dark conditions. The data show a shift to higher bias of $E_{0-}$ and $E_{0+}$ and a general increase of current with increasing $B$. However, the relative weight of $E_{0-}$ and $E_{0+}$ changes significantly with $B$. The $B$ dependence of the amplitude of the $E_{0-}$ and $E_{0+}$ peaks in $I(V)$ has the characteristic form of a quantum mechanical admixing effect, i.e., with increasing $B$, the $E_{0-}$ feature tends to become weaker and...
FIG. 3. (a) $I(V)$ curves under illumination at $T = 4.2$ K and various $B$ for a RTD with $y = 0.08\%$. $B$ is increased from 0 to 11 T by steps of 0.5 T. For clarity, the curves are displaced along the vertical axis. The double step decrease of $I(V)$ marked by asterisks on one of the high-field curves arises from the instability in the current due to the strong negative differential conductance. The left inset is a sketch of the magnetotunneling experiment. (b) $I(V)$ curves with $B$ in the range 0 to 7.5 T. Dotted lines are guides to the eye and show resonances $E_{0-}$, $E_{0+}$, and $E'$. (c) Differential conductance, $dI/dV$, plot for $B = 0$ T.

disappear, whereas the $E_{0+}$ peak increases significantly in amplitude. Also, the $I(V)$ curves show a further weak feature ($E'$), which shifts to higher bias with increasing $B$ and disappears for $B > 6$ T. This feature is more clearly revealed in the differential conductance plot shown in Fig. 3(c). By carrying out a series of measurements for different orientations of $B$ in the (100) growth plane, we find that both the intensity and bias position of the resonances are isotropic.

We can understand the $B$ dependence of the resonances in terms of the effect of $B$ on an electron tunneling into the states of the GaAs$_{1-y}$N$_y$ QW. Let $\alpha$, $\beta$, and $Z$ indicate, respectively, the direction of $B$, the direction normal to $B$ in the growth plane, and the normal to the tunnel barrier, respectively. Because of the action of the Lorentz force, when an electron tunnels from the emitter into the well, it acquires an additional in-plane momentum given by $k_\parallel = eB s/\hbar$, where $s$ is the tunneling distance from the emitter to the QW. The structure of our device indicates that $s$ is approximately equal to 30 nm. The sum of the barrier width plus the half-width of the GaAs$_{1-y}$N$_y$ well is 10 nm, and we also need to take into account the spread of the electron wave function in the emitter region adjacent to the Al$_{0.1}$Ga$_{0.6}$As barrier (~ 20 nm). Varying $B$ allows us to tune an electron to tunnel into a given $k_\parallel$ state of the well; the voltage tunes the energy so that by measuring the voltage position of the peaks in $I(V)$ as a function of $B$, we can map out the energy–wave-vector dispersion curve $\varepsilon(k_\parallel)$ of the GaAs$_{1-y}$N$_y$ QW layer.

Figure 4(a) shows the $B$ and $k$ dependence of the voltage position of the current features $E_{0-}$, $E_{0+}$, and $E'$. The $V(k)$ plots for $E_{0-}$ and $E_{0+}$ resemble the dispersion curves $\varepsilon(k)$ of the $E_-$ and $E_+$ subbands calculated by using a two-band anticrossing model of bulk GaAs$_{1-y}$N$_y$ [see Fig. 4(b)] [5]. This suggests that $E_{0-}$ and $E_{0+}$ arise, respectively, from electron tunneling into the N-induced $E_-$ and $E_+$ subbands of the GaAs$_{1-y}$N$_y$ QW layer. However, as shown in Fig. 4(c), the QW confinement has the effect of modifying the detailed form of the $\varepsilon(k)$ curves associated with $E_-$ and $E_+$ [18]: The hybridization between the first QW subband states $qw_0$ and the N-impurity level occurs at $k$ values smaller than those for bulk GaAs, as also observed experimentally, and leads to subbands $E_{0-}$ and $E_{0+}$ with smaller energy separation than for the bulk case; the QW confinement effect also gives rise to additional subbands, $E_{1-}$ and $E_{1+}$, arising from the hybridization of the higher energy QW subband states $qw_1$ with the N-impurity level. The latter effect may explain the presence of the $E'$ resonance and the form of its associated $\varepsilon(k)$ dispersion curve. We attribute the $E'$ resonance to electron tunneling into the $E_{1-}$ states. As shown in Fig. 4(c), the energy minimum of the $qw_1$ subband is very close to the N level, which gives rise to

![Image](61x508 to 288x745)

FIG. 4. (a) Voltage position of current peaks $E_{0-}$, $E_{0+}$, and $E'$ in $I(V)$ as a function of $B$ (top axis) and of $k = esB/\hbar$ (bottom axis). (b) Energy–wave-vector dispersion curves for bulk GaAs$_{1-y}$N$_y$. (c) Energy–wave-vector dispersion curves for a GaAs$_{1-y}$N$_y$ QW (continuous lines) and for a GaAs QW (dotted lines). The energies are measured relative to the minimum of the GaAs conduction band and the N content is equal to 0.08\%.
if we assume a value of the energy separation \( \Delta \) is known precisely. However, if we assume a value of \( f \sim 2 \), as expected within a simple electrostatic model of our device, we find that the energy separation \( \Delta \) between the \( E_\pm \) and \( E_\pm \) subbands is smaller (\( \sim 0.16 \) eV) that predicted by theory for bulk GaAs \( N \) layer with respect to the bulk case.

Finally, the \( B \) dependence of the amplitude of the \( E_0 \) and \( E_0 \) features in \( I(V) \) reveals interesting details about the nature of the \( E_\pm \) and \( E_\pm \) subbands. The magnitude of the tunnel current is proportional to the modulus squared of the tunneling matrix element between the in-plane components of the emitter and QW states. At \( B = 0 \), the conservation of the in-plane wave vector implies that the QW states into which an electron can tunnel must have at least a partial \( \Gamma \)-like character, since the states of the GaAs emitter are almost pure \( \Gamma \) states. Since \( E_0 \) is observed at \( B = 0 \), we infer that the very weakly dispersed \( E_\pm \) states have a significant \( \Gamma \)-conduction character even at \( k = 0 \). The strong enhancement of the \( E_0 \) resonance at large \( B \) and the corresponding increase of the momentum-energy dispersion at large \( k \) (\( k \sim B \)) indicate that, with increasing \( k \), the \( E_\pm \) subband states become more localized in real space. In contrast, the disappearance of the \( E_0 \) resonance at large \( B \) indicates that the \( E_\pm \) subband states become strongly localized at large \( k \) values. The \( E^* \) feature in \( I(V) \) has a similar dependence as \( E_0 \), which confirms our assignment of this resonance to tunneling into states of the \( E_0 \) subband.

In conclusion, magnetotunneling spectroscopy can provide us with a powerful means of probing the N-induced states in a dilute GaAs \( N \), QW. The data reveal that, for a small \( N \) content (\( \sim 0.1 \% \)), the N-induced \( E_\pm \) and \( E_\pm \) subbands have a very well-defined character and that the heavy effective mass \( E_\pm \) states have a significant \( \Gamma \)-conduction band character even at \( k = 0 \). Our results suggest that the highly nonparabolic dispersion of \( E_\pm \) and \( E_\pm \) could be tailored by the QW confinement and exploited to design novel band-structure engineered devices with negative differential velocity characteristics, similar to those occurring in semiconductor superlattices or Gunn diodes [19,20].

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[3] For a review article on dilute nitrides, see C. Skierbiszewski et al., Rev. Mod. Phys. 65, 035207 (2001), and references therein.


[18] For bulk GaAs \( N \), we calculate the energy–wave-vector dispersion curves of the \( E_\pm \) and \( E_\pm \) subbands by using the relation \( E_\pm = \sqrt{\frac{1}{2} [E_M(k) + E_N] \pm \sqrt{(E_M(k) - E_N)^2 + 4yC_{MN}^2}} \), where \( E_M(k) \) is the energy–wave-vector dispersion curve of GaAs, \( E_N \) is the energy position of the N-related level (\( E_N = 1.725 \) eV at 4.2 K), and \( C_{MN} \) is the hybridization matrix element (\( C_{MN} = 2.7 \) eV). For the case of the GaAs \( N \)/Al\(_{x}\)Ga\(_{1-x}\)As QW, we use the same relation but with \( E_M(k) \) equal to the energy–wave-vector dispersion curve for each subband of the GaAs/Al\(_{x}\)Ga\(_{1-x}\)As QW.