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Growth of electric-field domains in quantum-well structures: Correlation with intersubband Raman scattering

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We report on Raman scattering by intersubband excitations in GaAs-AlAs superlattices which exhibit electric-field domains resulting from sequential resonant tunneling. In regions where two domains coexist, the scattering intensity varies linearly with the applied voltage. This behavior, consistent with the standard domain picture, can be explained by a model relying on the electric-field dependence of the cross section and the domain size.

At sufficiently large carrier densities and for weakly coupled wells, the dominant transport mode of quantum-well (QW) structures is sequential resonant tunneling (SRT).[1–12] As a result of negative conductance associated with SRT the superlattice field distribution can break up into electric-field domains separated by domain boundaries.[1,5–12] The domains are regions where the field strength is determined not by the applied bias, but by the energy separation between the tunneling subbands.[1,5–12] Photoluminescence (PL) spectra have been shown to provide direct evidence for the presence of SRT domains.[8,9] In the voltage ranges for which domains coexist, PL data reveal two peaks at a given bias.[8,9] There, the current exhibits plateaus showing fine structure which bears on the motion of the domain boundary.[1,5–11]

In this paper, we present results of Raman experiments on Si-doped superlattices which are consistent with the domain picture. Earlier work on photoexcited QW structures was inconclusive in this regard.[11] Our studies under SRT conditions show that the intensity for scattering by intersubband transitions of electrons varies linearly with the applied voltage. This behavior can be accounted for by a simple model which considers the field dependence of the cross section and that of the domain size of the applied voltage.

Our n^+-n-n^+ QW sample is from the same wafer as the one studied earlier in Ref. 9. The structure, grown by molecular-beam epitaxy on a (001) n^+-type GaAs substrate, consists of $N = 40$ periods of alternating 90-Å-thick GaAs wells and 40-Å-thick AlAs barriers sandwiched between heavily doped $\mathrm{Al_{0.5}Ga_{0.5}As}$ layers. The wells are doped at $n = 3 \times 10^{17}$ cm^{-3} with Si donors confined to the central 50 Å of the GaAs slabs. Diodes were processed into mesas of area ~ 0.01 mm^2. Current-voltage traces of the devices were obtained with a Keithley 236 source measure unit. Raman and PL measurements were performed at $T = 4$ K using a Dilor-XY multichannel system and an argon laser pumped DCM (4-dicyanomethylene-2-dimethylaminostyril-4H-pyran) dye laser tuned to $\omega_L = 1.8004$ eV. At this energy, Raman scattering by e_1e_2 intersubband transitions exhibits an *outgoing* resonance with a state which manifests itself as a PL band shifted ~ 30 meV to the red of the e_2h_1 exciton (e_i and h_j, with $i, j \geq 1$, denote the ith electron and jth heavy-hole subband, respectively). Based on its position and a comparison with other PL experiments,[13] the resonance is ascribed to $e_2A_C^0$, i.e., the recombination of electrons at e_2 with neutral acceptors A_C^0 (presumably carbon[13]) at the *center* of the wells. Raman spectra were obtained in the $z(x', y')\bar{z}$ and $z(x', x')\bar{z}$ backscattering configurations, with z normal to the layers and x' and y' denoting [110] and [1$\bar{1}$0] directions. These geometries allow, respectively, intersubband scattering by spin- and charge-density excitations.[14] Unless indicated otherwise, the power density was $P = 150 \pm 20$ W cm^{-2}. This value is large enough so as to provide a reasonable rate of scattered photons while sufficiently small so that the ratio between the number of carriers introduced by photoexcitation and doping remains negligible.

Our experiments probe e_1e_2 intersubband scattering as a function of the applied voltage V in the range $3 \leq -V \leq 10$ V (for $V < 0$, the substrate is at a higher voltage than the top layer). At the resonance, the e_1e_2 Raman peak sits on top of the broader $e_2A_C^0$ band which is of a comparable intensity. While the PL is essentially unpolarized, e_1e_2 scatters primarily in $z(x', x')\bar{z}$; its spin-density $z(x', y')\bar{z}$ component is at least an order of magnitude weaker. Because of this, we find it convenient to work with data obtained by subtracting $z(x', y')\bar{z}$ from $z(x', x')\bar{z}$ which effectively removes the

PL contribution. A typical difference spectrum is shown in Fig. 1(a) ($V = -3.5$ V). Our assignment of the line with maximum at ~ 0.137 eV as due to e_1e_2 transitions is supported by the observed separation between e_1h_1 and e_2h_1 PL which varies in the range 0.13–0.15 eV. In addition, the experimental value is in good agreement with effective-mass calculations of the eigenenergies E giving, at zero field, $E(e_2) - E(e_1) = 0.135$ eV. With increasing $|V|$, the e_1e_2 energy exhibits a weak monotonic increase, but no apparent correlation with the domain behavior.[15] In particular, the positions of e_1e_2 at $V = -3$ and -10 V differ by no more than 2%. This is consistent with the calculations predicting a 3% change in the e_1e_2 energy for the corresponding fields.

As discussed in detail below, the QW structure at $V = -3.5$ V divides into two electric-field domains of comparable lengths. The presence of two values of the field is reflected in the splitting of e_1h_1 shown in Fig. 1(b) for $P \approx 5$ W cm^{-2}. Unlike e_1e_2 scattering, the e_1h_1 line shape depends strongly on P and the doublet becomes poorly resolved at higher power densities. It is important to emphasize that our data show but a single Raman line irrespective of P (or V). However, this is not inconsistent with the PL results. The absence of e_1e_2 splitting reflects primarily the quadratic dependence of the cross section on the electric field (see below) together with the fact that the width of the Raman peak, ~ 5 meV, is considerably larger than the expected doublet separation. Here, we notice that the splitting of e_1h_1 results from the strong field dependence of $E(h_1)$.

The e_1e_2 intensity as a function of applied voltage is depicted in Fig. 2. The figure reproduces also the V dependence of the current obtained under identical illumination conditions; the trace displays features that are typical of superlattices for which the dominant form of transport is SRT.[1,5–10] In particular, the plateaulike regions $0.75 \leq -V \leq 5.5$ V and $-V > 7.5$ V signal the presence of coexisting domains.[5–10] Specific domains can be identified by comparing values of the resonant fields F, related to the voltage *per superlattice period* at the plateau boundaries, with the QW intersubband energies (derived from calculations or, say, PL and Raman experiments).[8,9,11] SRT requires that $Fdq \approx |E(e_m) - E(e_n)|$; q is the magnitude of the electron charge and d is the superlattice period. For our structure, the high- and low-field domains at the first plateau result from neighboring-well alignment of e_1 with, respectively, e_2 and e_1. In the following, these domains will be referred to as $\{e_1 - e_2\}$ and $\{e_1 - e_1\}$; the latter bears on the so-called regime of miniband conduction.[1,5] At the $-V > 7.5$ V plateau, the domains are $\{e_1 - e_2\}$ and the one relying on the alignment of e_2 and e_3, $\{e_2 - e_3\}$.[9] In both plateaus, the regularly spaced discontinuities in the current follow the growth of the high-field domain as it incorporates additional superlattice periods; this is shown by the remarkable fact that the number of current jumps is comparable to that of wells in the sample.[1,5–9]

The results in Fig. 2 show that the Raman intensity at the plateaus depends linearly on the voltage and, furthermore, that it is nearly constant in the range where there is a single domain $\{e_1 - e_2\}$, i.e., $5.5 \leq -V \leq 7.5$ V.[16] This behavior can be explained as follows. Let σ_H (F_H) and σ_L (F_L) be the Raman cross section (the magnitude of the electric field) corresponding to the high- and low-field domain containing, respectively, N_H and N_L wells. Ignoring weak absorption effects (the light penetration length is $\approx 60d$), the intensity in the plateaus is given by $I \propto (N_H\sigma_H + N_L\sigma_L)$ with $(N_H + N_L) = N$. Since $|V|/d \approx (F_HN_H + F_LN_L)$ (this ignores voltage drops at the contacts), the expression for I can be written as

$$I \propto \left(\frac{|V|}{d} \frac{(\sigma_H - \sigma_L)}{(F_H - F_L)} + N \frac{(\sigma_L F_H - \sigma_H F_L)}{(F_H - F_L)} \right), \qquad (1)$$

which is obviously consistent with the experimental findings. That $\partial I/\partial|V| > 0$ follows from the fact that the cross section increases with field, i.e., $\partial\sigma/\partial F > 0$, for the Raman process is parity forbidden at $F = 0$.[17] Equation

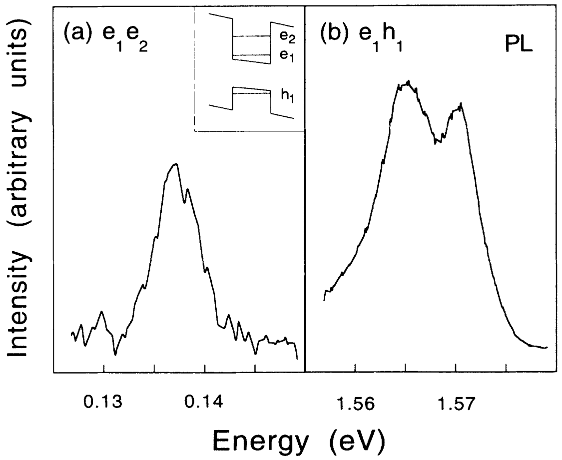

FIG. 1. (a) Intersubband (charge-density) Raman and (b) e_1h_1 PL spectrum of the QW at $V = -3.5$ V. Measurements were performed at $T = 4$ K and $\omega_L = 1.8004$ eV. Power densities were (a) $P \approx 150$ W cm^{-2} and (b) $P \approx 5$ W cm^{-2}. The two PL peaks correspond to domains $\{e_1 - e_1\}$ and $\{e_1 - e_2\}$. A schematic QW energy diagram is shown in the inset (not to scale).

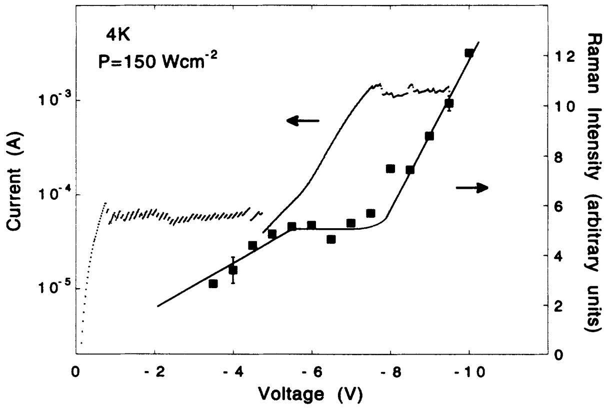

FIG. 2. Current (•) and intersubband scattering intensity (■) vs applied voltage V at $\omega_L = 1.8004$ eV. Domains $\{e_1 - e_1\}$ and $\{e_1 - e_2\}$ coexist for $-V < 5$ V. For $-V > 7.5$ V, the domains in the sample are $\{e_1 - e_2\}$ and $\{e_2 - e_3\}$. The solid line is a guide to the eye.

(1) defines a relationship between the slope at a given plateau and the F-dependent cross section. To leading order in F, $\sigma \propto F^2$,[18] and, thus,

$$I \propto \left(\frac{|V|}{d}(F_H + F_L) - NF_H F_L \right) . \qquad (2)$$

Assuming that $F \approx 0$ for domain $\{e_1 - e_1\}$, this expression predicts 2.78 for the ratio between the first and second plateau slopes. Considering experimental errors, this value is in very good agreement with the measured ratio of $\approx$ 2.9–3.2.

In deriving Eq. (2) it was implicitly assumed that the energy separation between $e_2 A_C^0$ and the scattered photon energy ω_S [$\equiv \omega_L - E(e_2) + E(e_1)$], bearing on the resonant enhancement, depends weakly on F. This is consistent with the experiments as we measure a shift of $e_2 A_C^0$ to the red and also an increase of the $e_1 e_2$ energy from $V = -3$ to -10 V of $\approx 2\%$. Irrespective of the dependence of the difference $(\omega_S - e_2 A_C^0)$ on F, it is important to emphasize that our model predicts a linear dependence of I on V. This is because σ_H and σ_L are determined by the resonant field and do *not* vary with V. Changes in $(\omega_S - e_2 A_C^0)$ are expected to modify only the ratio between plateau slopes.

The origin of the characteristic rapid rise of the current in the intermediate region is not well understood.[6–9] Our results support PL experiments indicating that, in this range, all the wells belong to a single domain.[9] The fact that the Raman intensity does not vary much with V indicates that whatever causes the current increase leads to minor changes in the magnitude of the field.

In summary, we have found a correlation between Raman scattering by intersubband excitations and domain behavior which manifests itself in a linear dependence of the scattering intensity on the voltage in the domain regime. The correlation, consistent with the standard domain picture, can be accounted for by a simple model which relies on the field dependence of the cross section and the domain size.

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[15] This is in contrast with the behavior of a photoexcited structure consisting of much wider wells (130 Å thick) for which the intersubband energy varies appreciably in the range of domain coexistence (Ref. 11).

[16] The observation in Ref. 11 that the intersubband intensity is roughly proportional to the photocurrent (undoped QW) bears on the strong V dependence of the photoelectron density. This is unlike the present case where $I(V)$ reflects primarily the field dependence of the cross section.

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[18] Symmetry arguments indicate that the expansion of σ in powers of F cannot contain odd terms. Alternatively, in the presence of an electric field, the leading-order correction to the envelope functions is linear in the field and, hence, $\sigma \propto F^2$.