Electron-Energy-Loss Scattering near a Single Misfit Dislocation at the GaAs/GaInAs Interface

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Spatially resolved electron-energy-loss scattering has been used to study changes in the inelastic scattering near the bulk band-gap energy for locations near the GaAs-$Ga_{0.85}In_{0.15}As$ interface. We observe the expected bulk band gap on either side of the interface. At a single interface-misfit dislocation we observe scattering which is consistent with an excitation of transitions between a localized state near the dislocation and the crystal conduction band. Within this interpretation, the energy of the state is estimated to be 0.7 ± 0.05 eV above the GaAs valence-band maximum.

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In a recent Letter,[1] current-voltage measurements were used to demonstrate an association between Schottky-barrier height and misfit dislocation density at n^+-GaInAs/GaAs heterojunctions. The proposed mechanism for this behavior involved Fermi-level pinning by defect states associated with these dislocations. However, the presence of defect states could not be directly confirmed because of the general lack of an appropriate analytical technique.

In this Letter, we report the first direct observation of electron-energy-loss scattering (EELS) associated with a single misfit dislocation at the GaAs/GaInAs interface, using a new technique, spatially resolved electron-energy-loss scattering (SREELS). We believe that this scattering is related to a filled electronic state, or set of states, which occurs in association with the misfit dislocation in this system. We measure the energy of this proposed state to be 0.7 ± 0.05 eV above the GaAs valence-band maximum, within the range required to produce Fermi-level pinning as suggested in the prior work.[1]

The SREELS technique utilizes the VG Microscope, Ltd., scanning transmission electron microscope (STEM) to form a 0.5–0.1-nm-diam beam of 100-keV electrons. A Wien-filter electron spectrometer[2] provides a 70-meV energy resolution with a collection semiangle at the specimen of 8 mrad, corresponding to a total transverse wave-vector resolution of 2.8 $Å^{-1}$, comparable to the limit imposed by the uncertainty principle for a 0.5-nm-diam electron probe. The accuracy and stability of the energy-loss axis is ± 20 meV in this low-energy range.

Heterojunctions were prepared by molecular-beam epitaxy in the following way. A 1000-nm film of GaAs was epitaxially deposited onto a (100 GaAs n-type substrate. This was followed by a 1000-nm layer of $Ga_{0.85}In_{0.15}As$. All layers and the substrate were doped with 2×10^{18} Si atoms cm^{-3} to guarantee that defect electronic states existing within the gap will be occupied. [100] planar and [110] cross-section TEM samples were thinned by mechanical polishing and ion milling (Ar^+ 4 keV). Transmission electron microscope images of the interface in planar view are similar to those shown previously.[1] In Fig. 1 we show a bright-field image of the GaInAs/GaAs interface obtained in cross-section view with the STEM. The interface lies in a plane perpendicular to the photograph running from bottom to top. GaAs lies to the right, while GaInAs lies to the left. A single misfit dislocation is indicated by the arrow. The characteristic asymmetric intensity due to crystal strain is visible. Carbon contamination buildup during the 10–20-min energy-loss acquisitions produced the 2-nm dark spots on the dislocation and to the right in the GaAs. We do not expect this amount of contamination to affect these measurements materially.

In these experiments, energy-loss intensity in the region of the interband absorption edge was acquired for positions on or near various structures in the planar-view and cross-section–view samples. The expected onset positions for this absorption are 1.42 eV for GaAs and 1.26 eV for the GaInAs material used here. The intensity data are obscured mainly by the energy distribution of

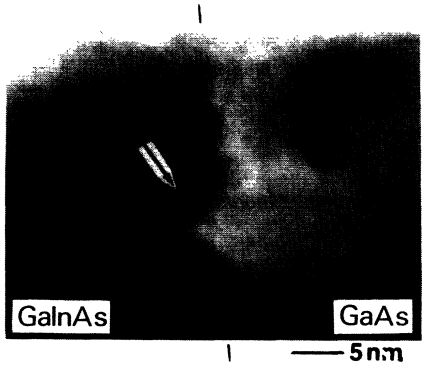

FIG. 1. Bright-field image of the GaAs-GaInAs interface in the cross-section view specimen. A misfit dislocation is indicated by the arrow.

the field-emission electron source, which closely follows the prediction of the Fowler-Nordheim model.[3] In Fig. 2, we show raw data for the field-emission distribution (line) compared with the distribution measured in transmission through GaAs (dots). Between 1.3 and 2.5 eV we show the same data expanded by 50× to show the GaAs interband absorption (upper dots), the field-emission background (lower dots), and an exponential extrapolation from the region between 0.7 and 0.9 eV (line). The energy resolution for the experiment was thus limited by the width of the field emission distribution to about 0.35–0.40 eV. Also, the interband signal must be separated from the exponentially decaying background caused by tunneling of electrons from well below the Fermi energy in the electron source. In Figs. 3 and 4, this background was determined by extrapolation from regions below the interband absorption edge. Figure 2 confirms that this gives a fairly accurate approximation to the actual measured intensity. We have subtracted the extrapolated background from the GaAs intensity in Fig. 2 and show the interband scattering (inset, dots) labeled for cross section.

We estimate the band gap by comparing the data with a calculated joint density of states (JDOS) for interband transitions. For two parabolic bands with a minimum separation E_g, and characterized by effective masses m_h and m_e (giving measures of the DOS at the band extrema), the JDOS per scattering center has the simple form,

$$N(E)=[m_h m_e/(m_h+m_e)]^{3/2}(E-E_g)^{1/2}. \quad (1)$$

For comparison with the experimental data, we have broadened the model with a 0.35-eV-wide Gaussian distribution. In Fig. 2, we have plotted this prediction (inset, line) using a band gap of 1.42 ± 0.05, as is expected for GaAs. Clearly, the shape and energy-loss position of these data are consistent with the model.

In Fig. 3, we show the interband absorption for the cross-section specimen. The results for GaAs and GaInAs were obtained about 50 nm from the interface. The result from the interface was obtained near the misfit dislocation shown in Fig. 1. Although inaccuracies in the background subtraction mentioned above make absolute intensity comparisons difficult, we note that the scattering is similar in the two bulk materials. This is expected, in consideration of Eq. (1), because the electron and hole effective masses are similar for GaAs[4] and InAs.[5] The solid curves in Figs. 3 and 4 are results of the model with $E_g=1.42$ eV for GaAs, and 1.26 eV for the GaInAs areas. Agreement with the experimental data is quite good, further confirming the sensitivity of the energy-loss scattering to interband transitions. At this point it is obvious that the scattering is different at the interface, and indicates a shift of the absorption edge to 1.08 eV.

In Fig. 4, for the planar-view specimen, we show the results for a beam position in the middle of a 50-nm-wide coherent area bounded by misfit dislocations, and for a position on a misfit dislocation. We have included the model results from Fig. 3 without changing E_g for comparison. When the beam does not intersect a dislo-

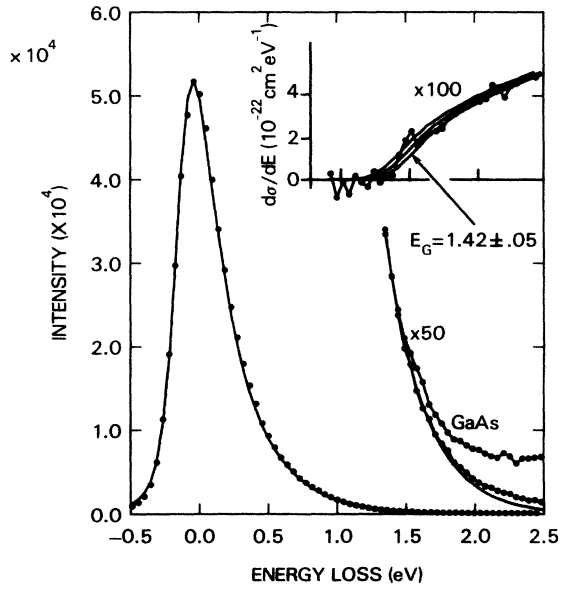

FIG. 2. Measured data for the field emission distribution (line) compared with the GaAs scattering (dots). The extrapolated background (line×50) differs from the measured background (upper dots×50) only above 1.8-eV energy loss. The model result (inset) is explained in the text.

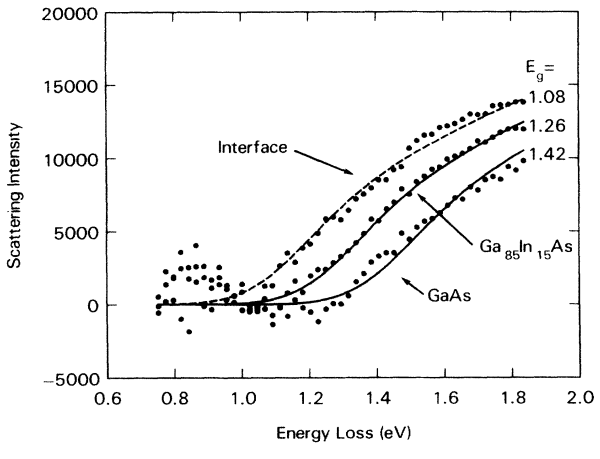

FIG. 3. Inelastic scattering after removal of the background intensity for GaAs, GaInAs, and the end on dislocation shown in Fig. 1. Model calculations for different band gaps are included for comparison. Scatter in the data below 0.8 eV results from inaccurate subtraction of the large field-emission background.

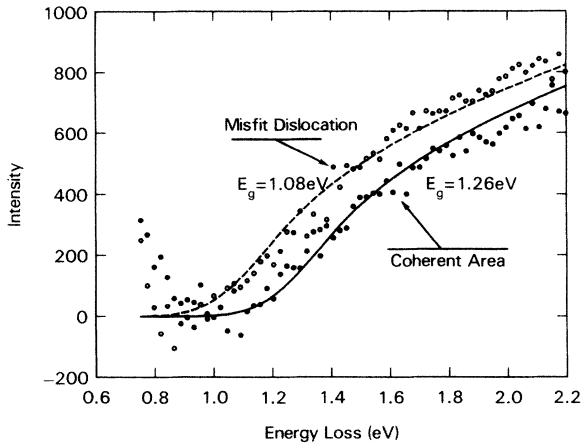

FIG. 4. Inelastic scattering for the planar-view specimen. The models calculated for Fig. 2 are included for comparison.

cation, we have a result which is very similar to that obtained in GaInAs above. We expect in this situation to observe two overlapping absorption edges, but an evaluation of the model above for two overlapping edges shows that the experimental resolution (0.35 eV) is not good enough to allow detection of the higher-energy edge. When the beam intersects a single misfit dislocation at the boundary of the square, the absorption edge clearly shifts downward to a position consistent with that measured in the cross-section experiment. Therefore we believe that the measured inelastic scattering can be associated with the misfit dislocation rather than with the coherent area of the interface.

If this scattering indicates the presence of a localized electronic state within the gap, we can estimate its energy by noticing first that a highly localized state can be characterized by an essentially flat band ($m_h = \infty$) extending throughout the Brillouin zone. Vertical transitions to the conduction band are thus possible throughout momentum space. Other conduction-band minima occur[4] at L_6 (1.18 eV, $m_e = 0.55 m_0$) and at X_6 (2.03 eV, $m_e = 0.85 m_0$). InAs is very similar to GaAs for these indirect minima, varying by a maximum of 0.25 eV,[6] leading to 0.04-eV variation for this GaInAs alloy. Also, very little misalignment of the indirect band minima occurs across the heterojunction.[7] Because of these large effective masses (or, alternatively, large density of states), the localized scattering will be dominated by transitions to these indirect minima. This disparity in density of states between the bands at the indirect minima relative to that at Γ is peculiar to III-V compounds and has been noted earlier by Tersoff.[7] Evaluation of Eq. (1) shows that the JDOS for localized scattering to these indirect minima should be as much as 80 times larger than the JDOS for localized scattering to Γ. Therefore we can locate the hypothetical electronic state at 1.08 eV below the average indirect conduction-band minima at 1.76–1.81 eV or about 0.7 ± 0.05 eV above the GaAs valence-band maximum.

We can estimate the expected bulk-scattering intensity by evaluating the scattering cross section for fast electrons in the Born approximation for a uniform medium of dielectric constant ε and atomic density n. We find[8]

$$\frac{\partial \sigma}{\partial E} = \frac{2}{\pi n a_0 E_0} \ln\left(\frac{\theta_c}{\theta_E}\right) \mathrm{Im}\left(\frac{-1}{\varepsilon(E)}\right), \tag{2}$$

where a_0 is the Bohr radius, E_0 is the incident electron energy, θ_c is the maximum scattering angle, and θ_E is related to the energy loss.[8] For the bulk interband absorption, we have estimated ε from the optical-absorption data,[9,10] finding the real part $\varepsilon_1 = 11.2$ and the imaginary part $\varepsilon_2 = 0.6$ at $E = 1.5$ eV. We estimate θ_c from the combined momentum width of the valence and conduction bands contributing to the near-edge absorption. Equation (2) gives $\partial\sigma/\partial E = 0.84 \times 10^{-22}$ cm^2 eV^{-1}. Referring to Fig. 2, we observe $\partial\sigma/\partial E = 1.4 \times 10^{-22}$ cm^2 eV^{-1}. The main uncertainty in the measured cross section derives from the thickness, which was not measured, but which we estimate to lie within the range 200–400 nm for that measurement. Thus the bulk scattering is easily observable and agrees with the expected result.

We can estimate the expected defect scattering intensity by considering an atomic core-to-continuum excitation characterized by a binding energy E, and a generalized oscillator strength (GOS) $\partial f/\partial E$, embedded within a medium with dielectric constant ε. The "golden rule" cross section is given by[11]

$$\frac{\partial \sigma}{\partial E} = \frac{4\pi e^4}{|\varepsilon|^2 E_0} \ln\left(\frac{\theta_c}{\theta_E}\right) \frac{1}{E} \frac{\partial f}{\partial E}. \tag{3}$$

We model the localized state as a single atom embedded in GaAs and having a core binding energy of 1 eV. We estimate the GOS to be equal to that for ionization of hydrogen,[11] noting that this number is between 0.01 and 0.2 eV^{-1} for most ionization processes and is not strongly atom dependent. Then Eq. (3) gives $\partial\sigma/\partial E = 0.6 \times 10^{-20}$ cm^2 eV^{-1} for the defect scattering, almost 100 times larger than the bulk interband cross section for GaAs. For a 0.5-nm probe incident on a 50-nm-thick sample, 430 atoms are illuminated. With this cross section, as few as five localized states within the probed volume would produce as much scattering as the bulk.

We wish to emphasize that this experiment, while establishing the presence of misfit-dislocation scattering, does not identify the nature of the scattering center. From our arguments above, it seems to us plausible that this scattering is the result of transitions from a filled dislocation-related state, or states, to the crystal conduction band. These states can be related to the structure of the dislocation. They could also be associated with impurity segregation at the dislocation, perhaps by In or the Si dopant. Other explanations are possible. However, the arguments above relating to cross section should

be sufficiently general to be applicable to most possibilities. Therefore, this work has directly established that energy-loss scattering occurs at single dislocations in this system; it has suggested that the most plausible cause is the presence of localized electronic states at or near the dislocation; and it has demonstrated that this explanation is consistent with both experimental and theoretical findings. It thus offers support for the dislocation-pinning mechanism for generation of a Schottky barrier in the presence of a high dislocation density. The work also establishes spatially resolved electron energy-loss scattering as a viable method for measurement of local electronic structure with a near atomic spatial resolution.

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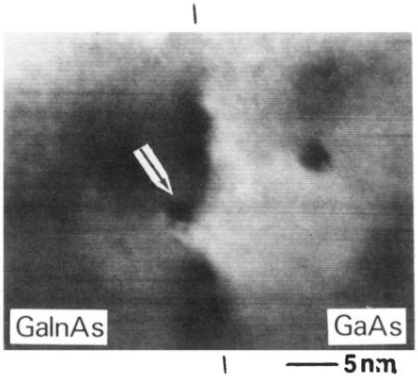

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