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Defect-initiated emission of Ga atoms from the GaAs (110) surface induced by pulsed laser irradiation

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Abstract. We have measured the Ga^0 emission yield from GaAs(110) surfaces for laser pulses of several photon energies near the band-gap energy 1.435 eV, ranging from 1.33 eV to 2.53 eV. Similarly to the case for emissions of Si atoms from Si(100) and of Ga atoms from GaP surfaces, we find that the Ga^0 emission yield, under repeated irradiation with laser pulses at fluences smaller than that for ablating the surface, decreases from its initial value rapidly at first and then slowly, while repeated irradiation by laser pulses above the ablation threshold fluence increases the emission yield gradually. It is found that the Ga^0 emission yield for laser pulses below the ablation threshold depends strongly on the photon energy $h\nu$: the emission yield is relatively small for $h\nu < 1.39$ eV (region I); the emission yield is essentially zero for 1.39 eV $< h\nu <$ 1.42 eV (region II); and the yield increases in a stepwise fashion when the photon energy increases across the transition energies involving surface states for $h\nu > 1.42$ eV (region III). It is suggested that the emission for region I is induced by electronic excitation of defects on the surface, while that for region III is induced most dominantly by electronic transitions involving surface states. It is also found that the yield is a power function of the laser fluence with power indices 2–4 for the rapidly decaying component and 4–7 for the slowly decaying component, depending on the photon energy.

1. Introduction

Defects on solid surfaces have been a topic of major importance because of the role played by the defects in surface catalysis [1] and *in* epitaxial growth of superlattices [2]. The invention of scanning tunnelling microscopy (STM) has revealed the existence of several types of defect on so-called cleaned surfaces [3]. It is suggested also that the STM technique can be used for the elimination of atoms on lattice points on surfaces [4] and deposition of atoms on specific sites on surfaces [5]. However, since the observation and control of defects by means of STM is limited to a small surface area, a technique for characterizing and controlling defects over a large area on surfaces has to be developed.

Recently, high-sensitivity measurements, using resonant ionization spectroscopy (RIS), of Ga^0 emission from the GaP($\bar{1}\bar{1}\bar{1}$) [6] and GaP(110) surfaces [7, 8] and of Si^0 emission from the Si(110) surface [9] induced by pulsed laser irradiation, have demonstrated that the particle emission is initiated by defects on the surfaces. The emission yield under repeated irradiation of the same spot on the surfaces, with laser pulses of a fixed fluence below the ablation threshold, is shown to decrease first rapidly from the initial value and then slowly, even after thousands of laser shots. Similar measurements for irradiation with laser pulses above the ablation threshold, detected by low-energy electron diffraction (LEED) have shown that the yield increased as the irradiation was repeated. Several types of defect are

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differentiated by the yield versus shot-number relation; the rapidly decaying component (A component) is ascribed to emission from adatom-type defects, from which elimination of a weakly bonded atom (WBA) leaves a defect-free surface; the slowly decaying component (S component) is ascribed to emission from kink-type defects, from which elimination of a WBA leaves a WBA of nearly the same atomic structure; and the increasing component (D component) is ascribed to emission from vacancy-type defects, from which elimination of a WBA increases the number of WBAs on the surface [7].

One of the characteristics of defect-initiated emission is that the yield depends strongly on the photon energy. In the case of Ga^0 emission from the GaP(110) surface, the emission was observed by laser pulses with photon energies below the indirect band-gap energy, while it was diminished almost completely as the photon energy increased across the band-gap energy [10]. It has been argued that the emission is induced by electronic excitations of the surface states below the conduction-band edge and/or by excitations of the defects on the surface. Further detailed comparison of the excitation spectrum for Ga emission with the surface band structure has not been carried out, partly because the location of the surface valence and conduction bands relative to the bulk bands for GaP is still controversial [11, 12]. The GaAs(110) surface is of most interest for studies of the excitation spectrum, since the surface electronic structures have been studied in detail; occupied and unoccupied surface states on the GaAs(110) surface are swept from the band gap [13, 14], while surface defect states are found within the band gap [15, 16].

The purpose of the present paper is threefold. First, we demonstrate that defect-initiated emissions of Ga atoms from the GaAs(110) surface, similar to those from GaP surfaces, indeed take place. Second, we report that the excitation spectra for defect-initiated Ga^0 emission from the GaAs(110) surface for photon energies ranging from 1.33 eV to 2.53 eV, across the bulk band gap $E_g = 1.435$ eV, reflect the surface and bulk band structures. Third, the yield of Ga atoms is found to be a power function of the laser fluence, with power indices of 2–4 for emission from adatom-type defects and 4–7 for that from kink-type defects.

2. Experimental details

A GaAs(110) (n-type, Si-doped) wafer measuring 10 mm × 12 mm × 0.445 mm was mounted on a manipulator placed in an ultra-high-vacuum (UHV) chamber of which the base pressure was 5×10^{-11} Torr. The surface was cleaned by repeated cycles of Ar^+ bombardment (500 eV, 3 μA cm^{-2}) followed by annealing to 800 K for 5 min in UHV. A W wire was attached to a Ta sample holder to heat the specimen, and the temperature of the Ta holder was measured using a chromel–alumel thermocouple. The temperature difference between the specimen and the sample holder was less than 20 K. After the cleaning procedures, the surface exhibited a sharp (1 × 1) LEED pattern, and no residual contaminant on the surface was detected by Auger electron spectroscopy (AES), confirming that the stoichiometry at the surface is preserved.

Laser pulses for exciting the surface ('pump' laser) with several photon energies ranging from 1.33 eV to 2.53 eV were used in this study. Pump laser pulses of 28 ns duration were generated by an excimer-laser-pumped dye laser system. The laser beam was incident at an angle of 45° to the surface and was focused to a spot 0.4 mm in diameter on the surface. The fluence of each laser pulse at the sample surface was evaluated by monitoring the output of a calibrated photodiode onto which a small fraction of the beam is directed by a beam splitter. The fluence was varied from 1 mJ cm^{-2} to several hundreds of mJ cm^{-2} by placing filters in the path of the laser beam.

Emitted Ga^0 atoms induced by laser irradiation were detected using RIS as reported elsewhere [8]: Ga^0 atoms emitted by a pump laser pulse were resonantly ionized by 'probe' laser pulses generated by another excimer-laser-pumped dye laser. The probe laser beam of 28 ns duration consists of fundamental and frequency-doubled beams; the photon energy of the fundamental beam was tuned at 2.157 eV, half of the $^2P_{1/2}$–$^2D_{3/2}$ transition energy of Ga^0 atoms; the frequency-doubled beam was used for excitation and the fundamental beam for ionization of the excited atoms. The probe laser pulse was delayed by 3.0 μs from the pump laser pulse. The beam passed parallel to the surface at a distance of 2.0 mm and was focused in front of the spot irradiated with the pump laser pulse.

Emitted particles including ionized Ga atoms and impurity ions such as K^+ and Na^+ ions [17] were detected by a multi-channel plate placed in a shielded box facing the surface. Signals due to impurity ions were separated from those due to neutral atoms by means of a time-of-flight mass-spectroscopic technique and by measuring the signal without the probe laser beam. The output signals were stored in a microcomputer through an analogue-to-digital converter. The detection limit of Ga^0 atoms by a laser pulse was estimated to be $< 10^{-7}$ monolayers (ML) [8].

In order to obtain the excitation spectra for laser-induced Ga^0 emission, we varied the temperature of the specimen as well as the photon energy $h\nu$ of the pump laser pulses. The temperature dependence of the Ga^0 emission yield at several photon energies was converted to the dependence of the yield on the difference between the photon energy and the band gap energy $E_g(T)$ at the temperature of the specimen. The same method was used by Okano *et al* [10] for Ga^0 emission from GaP(110) surfaces.

3. Results

The Ga^0 emission yield Y as a function of the shot number n of laser pulses with an energy of 1.35 eV, 0.09 eV below E_g, incident on the same spot of the surface at 300 K is shown in figure 1. The laser fluence was fixed at (*a*) 0.21 J cm^{-2} and (*b*) 0.4 J cm^{-2}. Evidently, the Y–n relation for 0.21 J cm^{-2} has a rapidly decaying component (A component) of the emission followed by a nearly constant component (S component), lasting even after 1000 shots. It was found that the LEED pattern at the irradiated spot was unchanged after prolonged laser irradiation. For laser pulses with a laser fluence of 0.4 J cm^{-2}, on the other hand, the yield increases gradually as the shot number increases and then becomes nearly constant (D component). Permanent damage was introduced after repeated irradiation by 0.4 J cm^{-2} laser pulses. Since the increase of the yield as shown in figure 1(*b*) was not observed below 0.4 J cm^{-2}, we determined the ablation threshold Φ_D to be 0.4 J cm^{-2}.

The temperature dependences of the S-component yield measured by repeated irradiation on the same spot of the GaAs(110) surface with 0.21 J cm^{-2} laser pulses for photon energies 1.33, 1.35 and 1.36 eV, are shown in figure 2. Each yield-versus-temperature curve consists of three regions, I, II and III, bordered by two critical temperatures, T_L and T_H: emissions are observed in region I below T_L and in region III above T_H, while no emission is observed in region II, between T_L and T_H.

In figure 3, the photon energy dependences of T_H and T_L are compared with that for the temperature T_C at which the photon energy coincides with E_g, obtained by Panish and Casey [18]: $E_g(T) = 1.522 - 5.8 \times 10^{-4} T^2/(T+300)$. It is clear that T_H is almost the same as T_C and varies in parallel to T_C, while T_L is significantly smaller than T_C and shows a photon energy dependence different from T_H and T_C. In view of these experimental results, we ascribe the temperature dependence of the Ga^0 emission yield to the temperature-induced variation of the band gap.

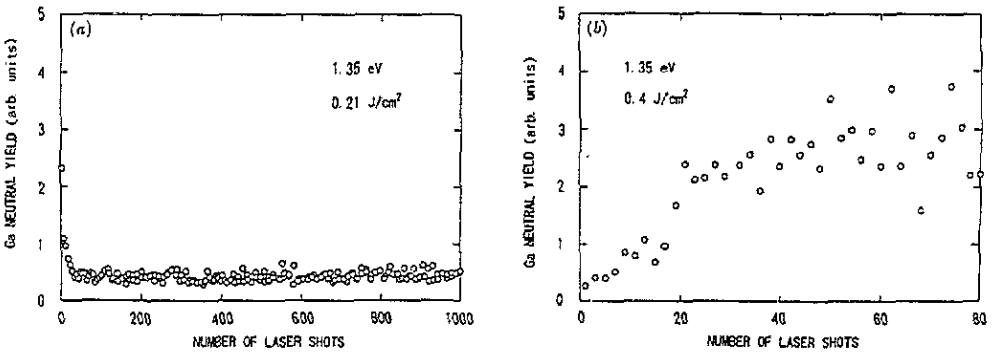

Figure 1. The emission yield of Ga^0 as a function of the shot number of 1.35 eV laser pulses, incident repeatedly on the same spot of a cleaned GaAs(110) surface at 300 K, at fluences of (*a*) 0.21 J cm^{-2} and (*b*) 0.4 J cm^{-2}. For 0.21 J cm^{-2} laser pulses, the yield decays rapidly from its initial value (A component) and becomes almost constant (S component), while the yield increases on repeated irradiation by 0.4 J cm^{-2} laser pulses.

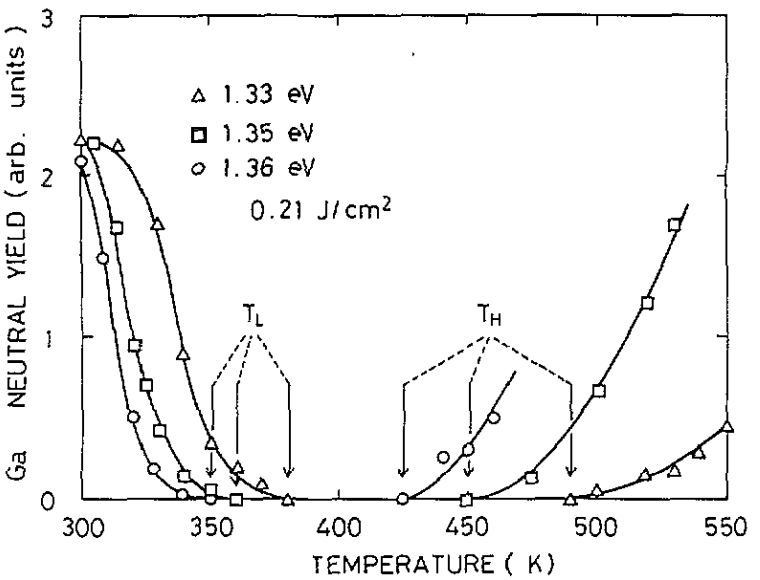

Figure 2. The temperature dependences of the S-component yield of the Ga^0 emission from the same spot of the cleaned GaAs(110) surface repeatedly irradiated by 0.21 J cm^{-2} laser pulses for sub-gap photon energies, 1.33, 1.35, and 1.36 eV. The yield becomes effectively zero at a temperature denoted by T_L and starts increasing again at a temperature denoted by T_H.

We measured the temperature dependences of the S-component yield in the temperature range from 300 K to 550 K for several photon energies $h\nu$ between 1.35 eV and 2.53 eV. The yield is plotted as a function of $h\nu - E_g(T)$ in figure 4. Evidently, the yield is scaled by $h\nu - E_g(T)$ except below the band gap, for which only the result obtained at 1.35 eV is shown. The figure includes the variation of the bulk optical absorption coefficient of GaAs with photon energy [19]. The yield increases as $h\nu - E_g(T)$ increases from zero, and

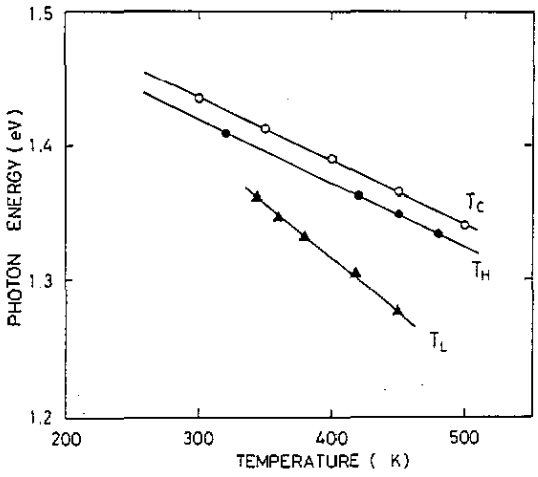

Figure 3. The photon energy dependences of the temperatures T_L and T_H, shown in figure 2. Also shown is the photon energy dependence of the temperature T_C at which the photon energy coincides with the band-gap energy E_g of GaAs.

becomes constant at around 100 meV above E_g. It is enhanced further when $h\nu - E_g(T)$ increases across 0.42 eV and 1.10 eV. Apart from the step-wise increases, the dependence of the yield on $h\nu - E_g(T)$ is extremely small. Evidently the variation in the yield as a function of $h\nu - E_g(T)$ does not follow the variation in the optical absorption coefficient, shown by a broken curve.

So far, we have described the results of the spectroscopic studies for the S component of the emission. In order to see whether the temperature dependence of the A component is similar to that of the S component, we obtained the Y–n relations for laser pulses of 1.35 eV at sample temperatures of 450 K (region II, $h\nu - E_g(T) = -0.015$ eV) and of 550 K (region III, $h\nu - E_g(T) = 0.034$ eV). Figure 5(*a*) and (*b*) shows typical results obtained by laser pulses of a fluence of 0.21 J cm^{-2} at 450 K and at 550 K, respectively. Comparing these results with that of figure 1(*a*) for region I, it is clear that the yields of the A component are almost the same for regions I and III, while the yield for region II is much smaller. (Note that the scale of the abscissa is expanded for figure 5(*a*) and (*b*) compared with figure 1(*a*).) Similar results were obtained for several spots on the surface. It follows that the electronic transitions for region II are ineffective for both the A and S components of the emission.

The relations between the yield Y and the fluence Φ for the GaAs(110) surface are found to be non-linear, similar to the GaP [8] and Si [9] surfaces. Figure 6(*a*) and (*b*) shows the Y–Φ relations for the A and S components obtained with 1.35 eV and 2.53 eV photons, respectively, at 300 K. The Y–Φ relation for the A component was obtained by repeated irradiation of a previously unirradiated spot on the surface by increasing laser fluence gradually, while the Y–Φ relation for the S component was obtained for the surface from which the A component is eliminated. Each curve exhibits a superlinear dependence of the yield on the laser fluence: there is an apparent threshold laser fluence above which the yield becomes appreciable. The threshold laser fluences of the A and S components are 60 mJ cm^{-2} and 110 mJ cm^{-2}, respectively, for 1.35 eV photons, and 6 mJ cm^{-2} and 28 mJ cm^{-2}, respectively, for 2.53 eV photons, at room temperature. Similar Y–Φ relations were obtained on spots of surfaces with and without pre-irradiation to eliminate the A component. The power indices were in the range of 2–4 for the A component and 4–7 for the S component. The ablation threshold laser fluence for laser pulses at 2.53 eV was found to be 60 mJ cm^{-2}, much smaller than the value of 400 mJ cm^{-2} for 1.35 eV laser pulses.

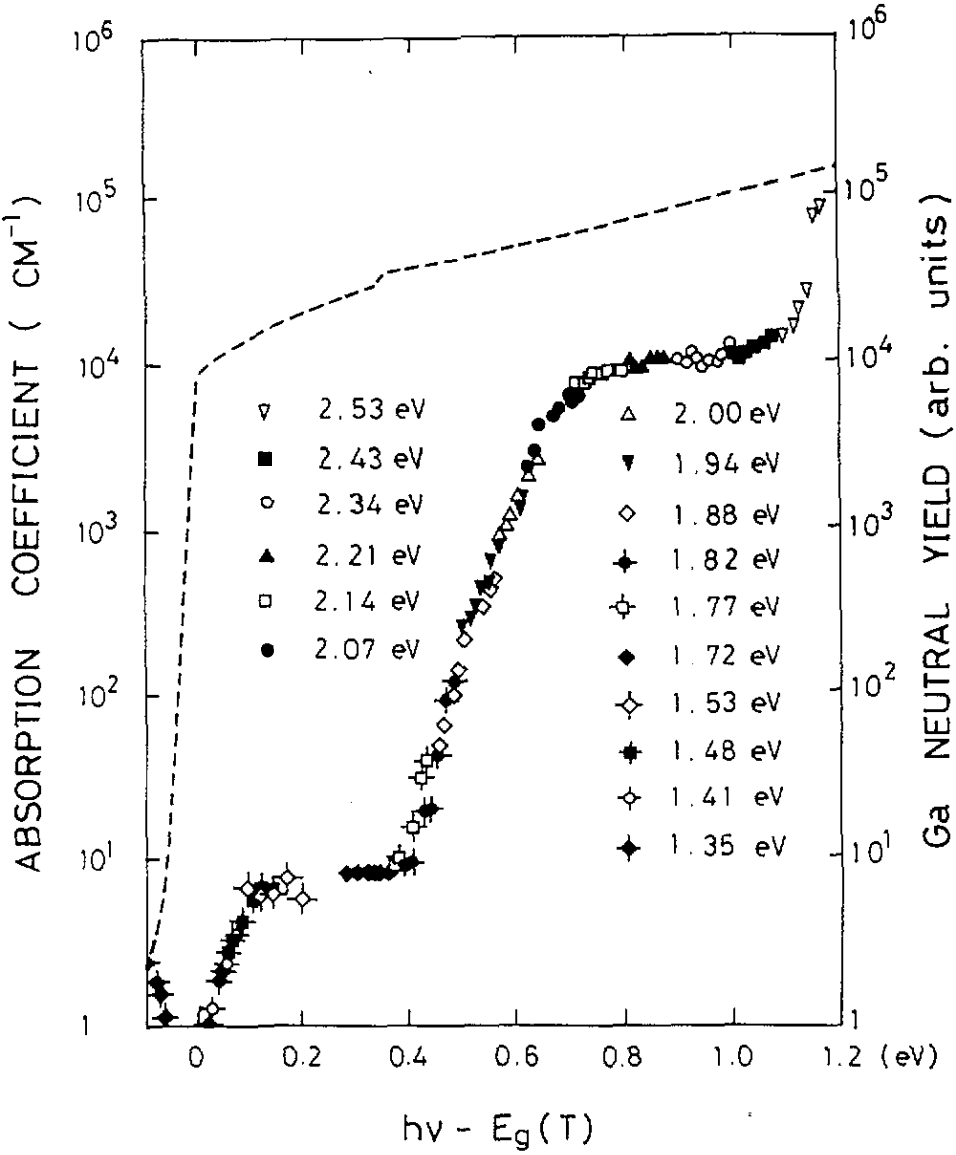

Figure 4. The emission yield of Ga^0 atoms as a function of $h\nu - E_g(T)$, where $h\nu$ is the photon energy and $E_g(T)$ is the band-gap energy at temperature T. The data plots are compiled from the results of the measurements of the temperature dependences obtained by laser pulses of several photon energies between 1.35 and 2.53 eV. A multiplication factor was used so that the yields at the same value of $h\nu - E_g(T)$ obtained at two different photon energies are identical. The broken curve shows the optical absorption coefficient of GaAs obtained by Sturge [19].

4. Discussion

The present results for the laser-induced Ga^0 emission from the GaAs(110) surface exhibit several features typical of defect-initiated emission obtained for the Ga^0 emissions from GaP($\bar{1}\bar{1}\bar{1}$) [6] and (110) [7, 8] surfaces and for the Si^0 emissions from Si(100) surfaces [9]. First, the emission yield is a power function of laser fluence with power indices of 2–7.

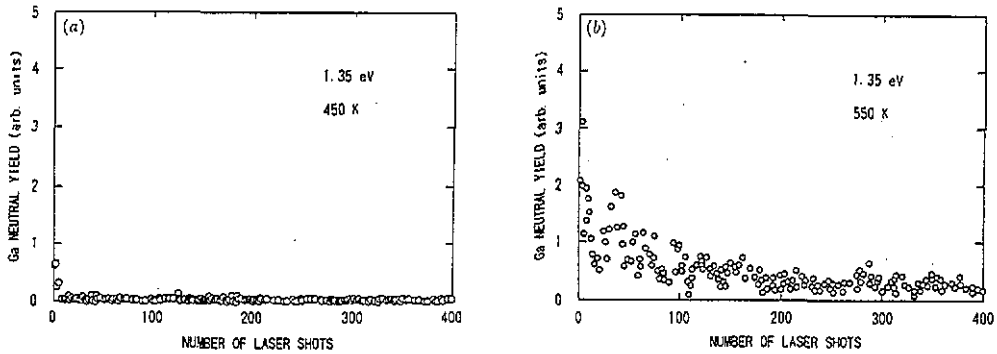

Figure 5. The emission yield of Ga^0 as a function of the shot number of 1.35 eV laser pulses of a fluence of 0.21 J cm^{-2} at sample temperatures of (*a*) 450 K (region II) and (*b*) 550 K (region III).

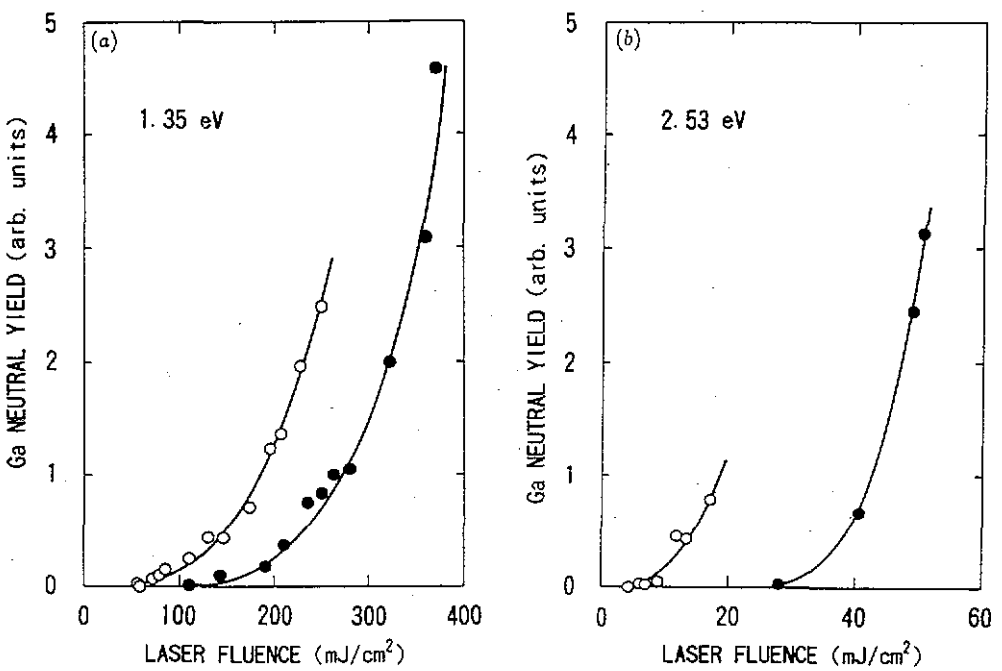

Figure 6. The laser fluence dependences of the A (open circles) and S (filled circles) components of the emission from the GaAs(110) surface induced by (*a*) 1.35 eV and (*b*) 2.53 eV laser pulses at 300 K. The threshold laser fluences above which the emission is appreciable are (*a*) 60 mJ cm^{-2} and (*b*) 6 mJ cm^{-2} for the A component and (*a*) 110 mJ cm^{-2} and (*b*) 28 mJ cm^{-2} for the S component.

Second, the emission yield is reduced by repeated irradiation with laser pulses first rapidly (A component) and then slowly (S component) below the ablation threshold fluence, while it increases above the ablation threshold (D component). Thus it appears that defect-initiated emissions of constituent atoms are generally observed in semiconductors. The A and S components have been ascribed to the emissions of WBAs from adatom-type and kink-type

defects on surfaces, respectively, while the D component has been ascribed to emissions from vacancy-type defects on surfaces. The decrease of the yield reflects the removal of defects and the increment reflects the growth of vacancy clusters.

A mechanism for explaining the superlinear relation between Y and Φ has been suggested by Hattori *et al* [8]; the bond breaking leading to the defect-initiated emission occurs due to cascade excitations of defects on the surface: a metastable state induced after each excitation is excited successively within a laser pulse and emission occurs if an anti-bonding-type excited state emerges. Khoo *et al* [20, 21] argued that the anti-bonding state emerges by multiple excitation, since the (multi-)holes are transferred into inner defect orbitals. The value of the power index in the Y–Φ relation is indicative of the number of cascade excitations until the anti-bonding excited state emerges. The present results indicate that 2–4 cascade excitations are needed to break a bond of WBAs at adatom-type defects and 4–7 cascade excitations are needed for kink-type defects. We presume that the value of the power index is larger for defects for which the surrounding atoms are bonded more strongly [22].

As shown in the preceding section, the defect-initiated Ga^0 emissions from the GaAs(110) surface are observed for laser pulses in regions I and III. In view of theoretical and experimental studies of surface electronic structures of GaAs(110) surfaces [13, 14], both occupied and unoccupied surface bands are swept from the bulk band-gap region. Thus we suggest that the emissions by photons in region I are caused by electronic excitations of the surface defects. We presume that the metastable states produced by the defect excitation as well as the initial defect state possess substantial optical absorption coefficients for photons slightly below the band-gap energy, and hence the superlinear dependence of the yield on fluence is conceivable. In fact, a very weak photon energy dependence has been observed in a wide photon energy range below the band-gap energy for GaP [23].

In order to assign surface optical transitions associated with the enhancements of the emission at $h\nu - E_g(T) = 0.42$ eV and 1.10 eV, we compared these energies with the transition energies involving surface states. Figure 7 shows a schematic band structure of the GaAs(110) surface including possible optical transitions at the $\bar{\Gamma}$ point: T_1, the transition from the occupied surface band to the unoccupied surface band; T_2, from the occupied surface band to the bulk conduction band; and T_3, from the bulk valence band to the unoccupied surface band. In view of normal and inverse photoemission experiments [24, 25], the differences $T_2 - E_g(T)$ and $T_3 - E_g(T)$ are estimated to be (0.5 ± 0.2) eV and (0.56 ± 0.1) eV, respectively. It follows that $T_1 - E_g = T_2 + T_3 - 2E_g = (1.06 \pm 0.3)$ eV. Thus the stepwise rise in the yield at 0.42 eV is close to the T_2 and T_3 transition energies and that at 1.10 eV is close to the T_1 transition energy.

According to theoretical calculations by Manghi *et al* [26], the dipole moments of the T_1 and T_2 transitions are strongly oriented along the $[1\bar{1}0]$ direction, the direction of the Ga–As chains on the surface, while that of T_3 is almost isotropic. In order to reveal the anisotropy for the optical transitions leading to the stepwise increase at $h\nu - E_g(T) = 0.42$ eV and 1.10 eV, the present authors have measured the temperature dependences of the Ga^0 emission yield for laser pulses of photon energies of 1.88 eV and 2.53 eV, polarized along the $[1\bar{1}0]$ and [001] directions [27]. It has been shown that the steps are observed only for laser beams polarized along the $[1\bar{1}0]$ direction. The results indicate clearly that the step at $h\nu - E_g(T) = 0.42$ eV is due to the T_2 transition and that at 1.10 eV is due to the T_1 transition.

The stepwise increases at T_1 and T_2 indicate that the two-dimensional (2D) electron–hole (e–h) pairs generated in the surface states play an important role in the emission of Ga atoms from the surface. In view of the superlinear dependence of the yield on fluence,

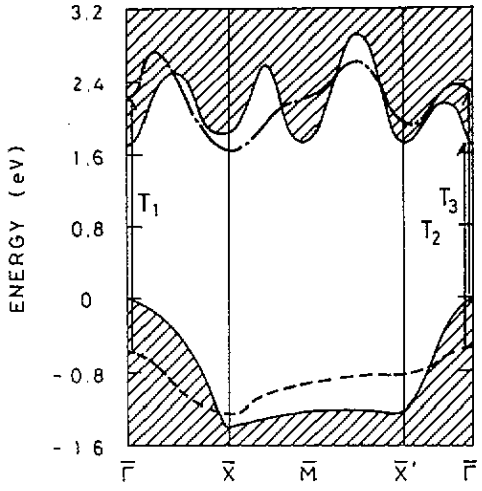

Figure 7. A schematic band structure of the GaAs(110) (1 × 1) surface. The broken curve shows the top of the highest occupied surface band obtained from a photoemission measurement, the chain curve indicates the theoretical results for the bottom of the lowest unoccupied surface band, and full curves show the bulk band structures. T_1 and T_2 are the electronic transitions at the $\bar{\Gamma}$ point from the occupied surface band to the unoccupied surface band and to the bulk conduction band, respectively, and T_3 is that from the bulk valence band to the unoccupied surface band [14].

the following two mechanisms of the emission initiated from 2D e–h pairs are conceivable. The first possibility is that a metastable state is generated by trapping a 2D e–h pair by a defect, and the resulting metastable state is further excited by incoming photons. In this case the photon energy dependence is indicative of that of the number of 2D e–h pairs. An alternative mechanism is that the metastable state interacts with e–h pairs, leading to formation of the anti-bonding state. In this case, the photon energy dependence is indicative of the mth power of the photon energy dependence of the number of 2D e–h pairs, where m is the power index for the $Y-\Phi$ relation. It is not yet possible to decide which mechanism is effective. Both models assume that the onset of the metastable state formation starts by the interaction of the 2D e–h pairs with defects on surfaces and the anti-bonding state is induced, in one case as a result of multiple excitation of the metastable state, and in the other as a result of the interaction of the metastable state with 2D e–h pairs.

We note that the bulk optical absorption coefficient of GaAs increases steeply around the boundary between region I and II where the A and S components are reduced. Thus an absence of emissions is observed in the photon energy range where the bulk optical absorption coefficient is considerably large. As suggested by Okano *et al* [10], we ascribe the reduction of the emissions to the transfer of the excitation energy into the bulk, owing to resonance interaction between defect excitation and the bulk excitation. It appears that phonons are involved in the resonance interaction, since the separation between E_g and the critical photon energy T_L at which the yield becomes zero increases as the sample temperature increases.

It has been suggested that the laser-induced defect-initiated atomic emissions from semiconductor surfaces can be utilized for eliminating adatom- and kink-type defects from surfaces [22]. The present experimental results demonstrate that the electronic excitation involving the surface valence band is particularly effective for elimination of these defects. Furthermore, it has been shown by Kanasaki *et al* [28, 29] that the interaction of the kink-type defects with reactive adsorbates such as Br [28] and O [29] enhances the emissions from the kink-type defects. Thus use of lasers with photon energies capable of excitation from the surface valence band under Br_2 and O_2 atmosphere eliminates steps on the surface with considerably high efficiency.

Acknowledgments

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