Atomic resolution in tunneling induced light emission from GaAs(110)

Maria Reinhardt, Guillaume Schull, Philipp Ebert, and Richard Berndt

Citation: Applied Physics Letters **96**, 152107 (2010); doi: 10.1063/1.3360203
View online: http://dx.doi.org/10.1063/1.3360203
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/96/15?ver=pdfcov
Published by the AIP Publishing

APPLIED PHYSICS LETTERS **96**, 152107 (2010)

Atomic resolution in tunneling induced light emission from GaAs(110)

Maria Reinhardt,[1] Guillaume Schull,[2] Philipp Ebert,[3] and Richard Berndt[1,a)]
[1]*Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, D-24098 Kiel, Germany*
[2]*Institut de Physique et de Chimie de Strasbourg, CNRS UMR 7504, 67034 Strasbourg, France*
[3]*Institut für Festkörperforschung, Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany*

(Received 1 February 2010; accepted 20 February 2010; published online 14 April 2010)

A cryogenic scanning tunneling microscope is used to induce band edge luminescence from GaAs(110). The emission intensity varies within the unit mesh of the surface. This atomic resolution reflects variations of the efficiency of hole injection into states at the valence band maximum. Moreover, the hole injection efficiency is modulated by local potential fluctuations due to dopants. © *2010 American Institute of Physics*. [doi:10.1063/1.3360203]

The tip of a scanning tunneling microscope (STM) may be used to locally inject electrons or holes into a sample and excite the emission of light. Such luminescence has been observed from metals, molecules on metal surfaces and ultrathin insulating films, and semiconductors. In the former cases, the luminescence involves processes at the sample surface, namely, inelastic tunneling or molecular excitations. As the tunneling current is localized to atomic dimensions, atomic scale resolution in maps of the emitted intensity has been achieved.[1–4] In the case of most semiconductors, luminescence is caused by the recombination of minority and majority charge carriers. Minority carriers may be directly injected or created by scattering of injected majority carriers.[5–14] Consequently, the spatial resolution of the luminescence signal is expected to be determined by the diffusion length of thermalized minority carriers inside the sample material. Indeed, STM induced luminescence did not exhibit atomic resolution, in agreement with the expectation based on carrier diffusion. Atomic scale contrast was observed only at atomic steps, and has tentatively been attributed to a modified carrier injection,[6] and for Si(111) surfaces, where it has been linked to spatial variation in localized plasmons.[15]

Here, we present STM induced luminescence data from GaAs(110) surfaces. Atomic resolution is observed for the first time, and interpreted in terms of the efficiency of hole injection at the valence band edge, which varies on the atomic scale.

The experiments were performed with an ultrahigh vacuum (UHV) scanning tunneling microscope operated at low temperature (7.3 K). *n*-type GaAs samples (Si doped, 10^{18} cm^{-3}) were cleaved in UHV to expose a clean (110) surface. Photons emitted from the surface area underneath the apex region of a W tip were collected with a lens mounted inside the vacuum chamber. The collected light was guided via an optical fiber to a grating spectrometer and a liquid nitrogen cooled charge coupled device detector.[16]

In the present experiment, we observed luminescence at both polarities of the applied voltage. At negative sample voltages, hole injection from the STM into the *n*-type GaAs sample leads to photon emission with an estimated quantum efficiency of 5×10^{-5} photons per electron in agreement with earlier work.[17] Spectrally resolved luminescence data (Fig. 1) reveal an intense emission centered at ≈840 nm. This wavelength is consistent with the band gap of 1.49 eV expected for GaAs, doped 10^{18} cm^{-3}, at 7 K.[18] An additional emission is observed around 915 nm. Guo *et al.*[10] tentatively attributed a similar spectral feature to surface defects. Below we suggest that this additional peak may be tip-induced.

At negative sample bias the luminescence is observed to occur above a threshold of $V_S \approx -1.6$ V, which is consistent with the onset of hole injection into the bulk VB (Fig. 2) and previous work.[9,10,17] At positive bias, the emission sets in at a distinctly higher bias voltage of ≈3 V as expected for impact ionization processes.[6] Figure 3(b) shows the spatial distribution of the band edge luminescence recorded simultaneously with the STM topograph in Fig. 3(a), which was measured at $V_S = -1.9$ V. The "photon map" exhibits a 6% intensity modulation with atomic periodicity. We verified that this modulation was not due to a malfunction of the STM feedback. A unit cell indicated in both images reveals a lateral offset of the patterns. Along the $[1\bar{1}0]$ direction, the photon intensity maxima are centered between topographic maxima. Along [001], they are offset by ≈0.09 nm. This is further visualized in Fig. 3(c), where maps of maxima of the light intensity (light gray) and the topograph (dark gray) are superimposed. In addition to the intensity variations on an

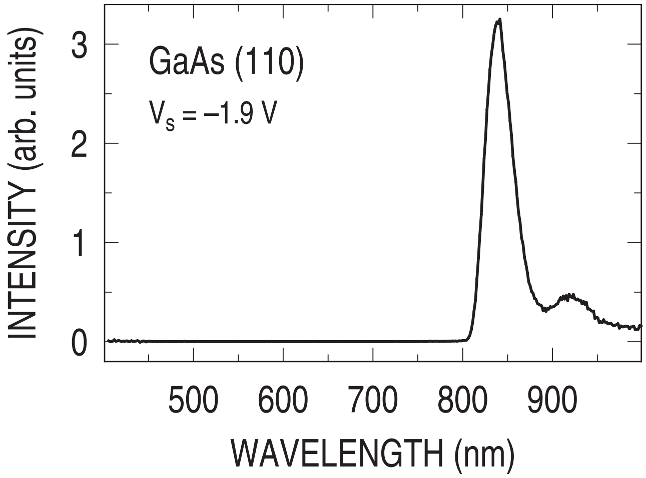

FIG. 1. Luminescence spectrum recorded at a sample voltage $V_S = -1.9$ V and tunneling current $I = 1$ nA over an exposure time of 1 s. The spectrum is corrected for detector response.

[a)] Electronic mail: berndt@physik.uni-kiel.de.

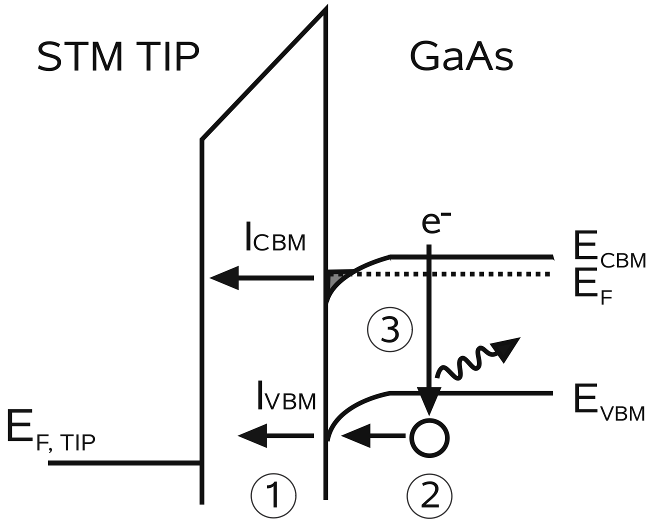

FIG. 2. Schematic energy diagram of the tunneling junction with negative bias applied to the *n*-type GaAs sample. Most of the tunneling current is due to tunneling from the CB minimum (I_{CBM}), which determines the topographic image. A small current component (I_{VBM}) (1) originates from states near the VB maximum and (2) generates holes at the VB maximum which (3) recombine with conduction band electrons.

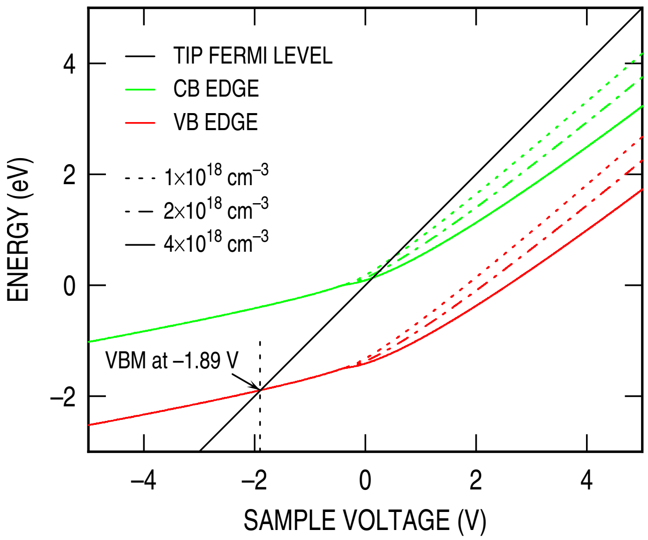

FIG. 4. (Color online) Calculated energies of CB minimum and VB maximum along with Fermi energy of the tip vs sample voltage. Energies are measured with respect to the Fermi energy of the sample. Dotted, dotted-dashed, and solid lines correspond to doping levels 1×10^{18}, 2×10^{18}, and 4×10^{18} cm^{-3}, respectively. Further parameters: tip work function 4.5 eV, tip-sample distance 0.9 nm, and donor level 5.8 meV.

atomic scale, an additional modulation is observed on a larger length scale of ≈3 nm. A related slow variation in the apparent topographic height may be discerned in the topograph.

To interpret the observation of atomic resolution in the photon emission, we first discuss the tunneling conditions. The proximity of the biased tip to the unpinned GaAs(110) surface induces a band bending in the semiconductor, due to its limited carrier concentration. Calculations of this band bending, taking into account the effects of carrier dynamics,[19] yield the valence and conduction band edge positions shown in Fig. 4 as a function of the voltage for different doping levels. Tunneling out of the VB at the surface is possible once the VB edge has an energy higher than the Fermi energy of the tip. Figure 4 shows that the onset for tunneling from the VB occurs thus at $V_S=-1.89$ V. In addition, at negative voltages, the CB edge is pulled below the Fermi energy of the GaAs sample (located at 0 eV) and electrons accumulate at the surface in the CB. At the voltage used to measure the topograph in Fig. 3(a), i.e., 1.9 V, the current is thus predominantly due to tunneling out of the electron accumulation zone in the CB and a minor fraction is due to hole injection into the VB. Thus, the topographic maxima reflect the positions of empty C_3 dangling bond states localized above the Ga atoms and the image shows the Ga sublattice.[19]

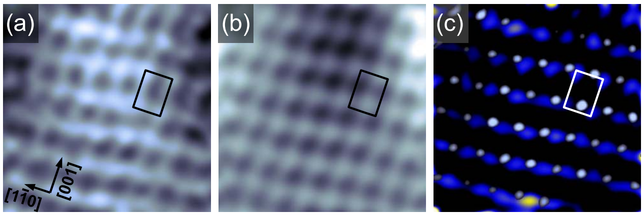

FIG. 3. (Color online) (a) STM topograph ($I=1$ nA; $V=-1.9$ V_S; and 3×3 nm^2) of a terrace of GaAs(110) and (b) map of the photon intensity with wavelengths 791 nm$\leq\lambda\leq$950 nm acquired simultaneously. (c) Superposition of maxima in topographic (dark gray) and intensity (light gray) maps. A unit cell at identical lattice positions is indicated in each image.

Combining the analysis of the topograph with the experimentally observed offset between topographic and luminescence maxima, we find that the luminescence is most intense when the tip is located above As atom sites, i.e., where the maxima of the occupied surface density of states at the VB maximum are localized.[20] We interpret the atomic-scale intensity variations as maps of the effective efficiency of the hole injection for radiative recombination. The enhanced photon intensity is related to the enhanced occupied density of states arising from the particular electronic structure of the GaAs(110) surface. Filled states near the Fermi energy, which are suitable for hole injection, are present only close to the As atom sites. This is where most intense luminescence is excited in the experiments. Whereas the topographic image is determined by the current from the CB (I_{CBM}) the luminescence is particularly sensitive to the smaller current component I_{VBM} which originates from states near the VB maximum.

The energy of the emitted photons suggests that electron transitions occur from the GaAs bulk CB minimum to the VB maximum, since the surface band gap is larger.[21] In addition, the surface gap is indirect. Therefore, we suggest that the holes which give rise to light emission are injected into the tails of bulk VB states near the surface and recombine with electrons in the tails of the bulk CB states near the surface.

An alternative mechanism that might lead to atomic resolution involves the vertical motion of the tip, which is induced by the constant-current mode of operation of the STM, as observed from metals.[22] However, in the present case, enhancement of the electromagnetic field by local modes is not known to play a significant role. Moreover, the intensity maxima do not coincide with topographic minima as would be expected for such a scenario. We discard this possibility.

At this stage we address the second emission peak at 915 nm, corresponding to a photon energy of 1.35 eV, i.e., 120 meV smaller than the bulk band gap. Since there are no surface states in the fundamental band gap on GaAs(110) surfaces and no defects were present, a lower transition energy cannot be attributed to the GaAs(110) material alone. Similarly a transition from a Si donor state can be excluded as an origin of the emission, because its binding energy is too small even when taking into account the increase due to a modified surface dielectric constant $\epsilon_{\text{surf}} = (\epsilon_{r,\text{GaAs}} + \epsilon_{r,\text{vacuum}})/2$.[23] We tentatively propose that the sharpness of the tip induces a *local* band bending and forms a potential well, which confines quantum states with energies within the band gap and leads to an emission with a lower photon energy.[24]

Finally, the long-range variation in the topographic height can be attributed to the presence of a subsurface Si donor.[25] The donor is positively charged and thus surrounded by a screened Coulomb potential. At the surface the screened Coulomb potential induces a downward band bending of the VB by up to ≈80 meV depending on the distance to the charge center.[26] This leads to a reduction in the VB density of states available for hole injection and, hence, a lower intensity of the luminescence. In analogy fluctuations of the local potential induced by fluctuations in the dopant distribution[27] will also induce fluctuations on the luminescence intensity.

We thank J. Aizpurua, DIPC San Sebastian, for discussions and F. Rinaldi and P. Unger, University of Ulm, and M. Wenderoth, University of Göttingen, for providing GaAs wafers. Financial support by the Deutsche Forschungsgemeinschaft (Grant No. GR 1409/5) is acknowledged.

[1] R. Berndt, R. Gaisch, W. D. Schneider, J. K. Gimzewski, B. Reihl, R. R. Schlittler, and M. Tschudy, Phys. Rev. Lett. **74**, 102 (1995).
[2] X. H. Qiu, G. Nazin, and W. Ho, Science **299**, 542 (2003).
[3] G. Hoffmann, T. Maroutian, and R. Berndt, Phys. Rev. Lett. **93**, 076102 (2004).
[4] G. Schull, M. Becker, and R. Berndt, Phys. Rev. Lett. **101**, 136801 (2008).
[5] D. L. Abraham, A. Veider, C. Schönenberger, H. P. Meier, D. J. Arent, and S. F. Alvarado, Appl. Phys. Lett. **56**, 1564 (1990).
[6] R. Berndt and J. K. Gimzewski, Phys. Rev. B **45**, 14095 (1992).
[7] T. Tsuruoka, Y. Ohizumi, S. Ushioda, Y. Ohno, and H. Ohno, Appl. Phys. Lett. **73**, 1544 (1998).
[8] P. Dumas, V. Derycke, I. V. Makarenko, R. Houdré, P. Guaino, A. Downes, and F. Salvan, Appl. Phys. Lett. **77**, 3992 (2000).
[9] T. Yokoyama and Y. Takiguchi, Surf. Sci. **482-485**, 1163 (2001).
[10] X. L. Guo, D. Fujita, N. Niori, K. Sagisaka, and K. Onishi, Nanotechnology **18**, 195201 (2007).
[11] S. E. J. Jacobs, M. Kemerink, P. M. Koenraad, M. Hopkinson, H. W. M. Salemink, and J. H. Wolter, Appl. Phys. Lett. **83**, 290 (2003).
[12] P. Schmidt, R. Berndt, and M. I. Vexler, Phys. Rev. Lett. **99**, 246103 (2007).
[13] K. Watanabe, Y. Nakamura, and M. Ichikawa, J. Vac. Sci. Technol. B **26**, 195 (2008).
[14] U. Håkanson, M. K.-J. Johansson, M. Holm, C. Pryor, L. Samuelson, W. Seifert, and M.-E. Pistol, Appl. Phys. Lett. **81**, 4443 (2002).
[15] A. Downes and M. E. Welland, Phys. Rev. Lett. **81**, 1857 (1998).
[16] G. Hoffmann, J. Kröger, and R. Berndt, Rev. Sci. Instrum. **73**, 305 (2002).
[17] X. Guo, D. Fujita, N. Niori, K. Sagisaka, and K. Onishi, Surf. Sci. **601**, 5280 (2007).
[18] S. M. Sze, *Physics of Semiconductor Devices* (Wiley, New York, 1981).
[19] N. D. Jäger, E. R. Weber, K. Urban, and P. Ebert, Phys. Rev. B **67**, 165327 (2003).
[20] B. Engels, P. Richard, K. Schroeder, S. Blügel, P. Ebert, and K. Urban, Phys. Rev. B **58**, 7799 (1998).
[21] H. Carstensen, R. Claessen, R. Manzke, and M. Skibowski, Phys. Rev. B **41**, 9880 (1990).
[22] J. Aizpurua, G. Hoffmann, S. P. Apell, and R. Berndt, Phys. Rev. Lett. **89**, 156803 (2002).
[23] G. Mahieu, B. Grandidier, D. Deresmes, J. P. Nys, D. Stièvenard, and P. Ebert, Phys. Rev. Lett. **94**, 026407 (2005).
[24] M. D. Croitoru, V. N. Gladilin, V. M. Fomin, J. T. Devreese, M. Kemerink, P. M. Koenraad, K. Sauthoff, and J. H. Wolter, Phys. Rev. B **68**, 195307 (2003).
[25] P. Ebert, Surf. Sci. Rep. **33**, 121 (1999), and references therein.
[26] A. Laubsch, K. Urban, and P. Ebert, Phys. Rev. B **80**, 245314 (2009).
[27] S. Landrock, Y. Jiang, K. H. Wu, E. G. Wang, K. Urban, and P. Ebert, Appl. Phys. Lett. **95**, 072107 (2009).