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Interface measurements of heterojunction band lineups with the Vanderbilt free-electron laser

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We used optical pumping by the Vanderbilt free-electron laser and the technique of internal photoemission to measure with high accuracy the conduction-band discontinuity of semiconductor heterojunction interfaces. The experiment is the first application to our knowledge of a free-electron laser to interface research.

For many years, measuring band discontinuities at the interface between two different semiconductors has been a crucial problem in condensed-matter science.[1,2] On one hand, such discontinuities determine the behavior and performances of an entire class of semiconductor devices, whose technological importance is growing. The fundamental interest of the problem, however, is more important.[1,2] The lineup of the two band structures, after having been mistakenly considered as a simple issue, has revealed itself[1-13] as a very complex conceptual question in condensed-matter physics—a question that touches, for example, the nature of the electronic states, the definition of energies, and the local treatment of many-body effects.

Unfortunately, progress in this field is negatively affected by the limitations of the experimental methods to measure band discontinuities.[1] In most cases, the accuracy of the measurements is not sufficient to discriminate one theory from another; in the few cases in which high accuracy is obtained, one must work with specialized preparation techniques and/or one has questionable reliability.[1]

We present an approach to this problem that reaches high accuracy in a simple and direct way. The approach is based on the use of an ultrabright source of infrared radiation—the recently commissioned[14] Vanderbilt Free-Electron Laser—to optically pump electrons across the conduction-band discontinuity. The photon energy threshold, revealed with a method introduced by Heiblum and co-workers[15] and by Abstreiter *et al.*[16] within the general framework of internal photoemission,[17-19] directly gives the value of the discontinuity.

The importance of this work is determined by the potential applications of the technique and in general of the free-electron laser in interface research. The free-electron laser removes an important limitation of the internal photoemission approach, the low brightness, and in fact our tests have immediately reached the best accuracy of discontinuity measurements, with ample potential for additional improvements. Note that the high brightness makes it possible to work with limited free-electron density, avoiding space-charge problems. Our tests show that the time structure of the source is not a problem in internal photoemission experiments, and it can be used in the future for time analysis of the phenomena. From a more general perspective, to the best of our knowledge this is the first application of a free-electron laser to an interface experiment, and therefore it serves also as a feasibility test for the practical use of this novel source in areas that require high accuracy and reliability.

The Vanderbilt Free-Electron Laser (FEL) is an upgraded version of the Stanford University Mk. III FEL.[14] The electron beam is produced by a 45-MeV radio-frequency accelerator, operating at a frequency of up to 2.856 GHz. The source is tunable over the 2–10-μm wavelength range (first harmonic, down to 1 μm in third harmonic) with high output power and brightness. Pulses with 6-μs duration, 360-mJ energy, and 11-W average power (repetition rate 30 Hz) have been reliably demonstrated in tests conducted at the wavelength of 4.8 μm.

These excellent technical performances, however, are not sufficient to guarantee success in using the source for practical experimental applications. Extensive experience with centralized facilities such as synchrotron radiation sources, has revealed the importance of factors such as the source's reliability, easiness of operation, and flexibility. The present field test on a rather demanding experiment eliminates all questions about the feasibility of advanced materials-science experiments with an FEL.

Figure 1(a) shows a simple scheme of the experimental process to detect the optical pumping of electrons across the conduction-band discontinuity.[15-19] The detection, based on the principle of internal photoemission,[17-19] is obtained by amplifying and measuring the external photocurrent by the optical pumping, and identifying the discontinuity-related threshold in the plot of the current versus the photon energy. Note that the initial-state electrons for the optical pumping across the discontinuity are produced by the forward bias and by the consequent steady-state injection of carriers into the small-gap ma-

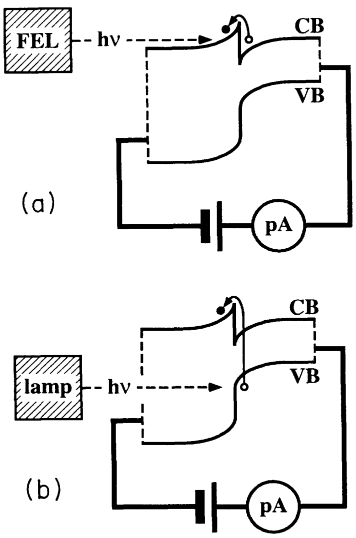

FIG. 1. Simplified schemes of our experiments. (a) Free-electron-laser (FEL) optical pumping experiment, where the pumping across the conduction-band (CB) discontinuity is detected by amplifying and measuring the external current it produces. (b) Similar experiment with a conventional photon source, which gives pumping from the top of the valence band (VB) in one material to the bottom of the CB in the other.

terial. We also conducted experiments with continuous optical pumping (by a visible lamp) from the valence band to the conduction band of the small-gap material, to increase the number of initial-state electrons. The results of this approach are consistent with those obtained with injection alone.

The first tests were performed on a GaAs-$Ga_{1-x}Al_xAs$ interface, obtained by molecular-beam epitaxy (MBE) growth of 3 μm of Si-doped ($n = 3\times10^{16}$ cm^{-3}) GaAs on a Si-doped ($n = 3\times10^{18}$ cm^{-3}) GaAs substrate, followed by MBE growth of 1200 Å of nominally undoped $Ga_{1-x}Al_xAs$, with 30 at. % Al content. The structure was capped by 300 Å of highly doped $Ga_{1-x}Al_xAs$ with the same Al content and the electrical contacts were made of deposited Au.

Figure 2 shows the result of the experiment, performed at a temperature of 77 K on a GaAs-$Ga_{1-x}Al_xAs$ interface: the photon energy threshold E'' is immediately visible, and it directly corresponds to the conduction-band discontinuity. Using a square-root line shape as predicted by Fowler for Schottky barriers and by Kane and Williams for heterostructures,[17] a least-square best fit gives a discontinuity of 0.218 eV.

We estimated the accuracy of this result by taking into account the uncertainty in the photon energy (≈ 5 meV), the $\approx 10\%$ fluctuation in the photocurrent, and the limitations of the method to extract the threshold from the data points. These last two factors were analyzed by the standard least-square procedures and also by changing the fitting line shape: a linear line shape gives a discontinuity of 0.22 eV. Overall, the uncertainty analysis gives an estimate of ± 0.010 eV. Possible space-charge effects were analyzed and ruled out by performing experiments at different intensities of the photon source (obtained with a neutral filter). We also performed experiments with different bias levels without observing changes in the results. Note that we have an undoped GaAs layer and therefore no quantum states in the interface potential well; for all bias levels the Fermi level was always in the gap, contrary to the situation of Ref. 16.

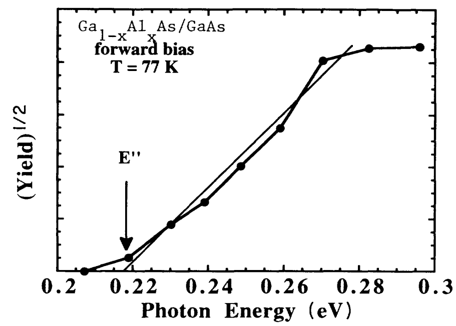

FIG. 2. Results of the experiment of Fig. 1(a) for the system GaAs-$Ga_{1-x}Al_xAs$.

The validity of this result was corroborated by parallel measurements with the alternate technique illustrated in Fig. 1(b).[17–19] In this case, optical pumping is performed with a conventional photon source at larger photon energies. The threshold E' observed at 1.70 ± 0.04 eV in the corresponding 77-K data of Fig. 3 is produced by transitions from the top of the valence band in the small-gap material (GaAs) to the bottom of the conduction band in the large-gap material ($Ga_{1-x}Al_xAs$). This means that the difference between the two thresholds E' and E'' should be equal to the smaller gap; and in fact, $E'-E''=1.48(2)\pm0.05$ eV, which is consistent with the 77-K gap width of GaAs, 1.51 eV.

Using the revised empirical rule of Refs. 20 and 21 that relates the conduction-band discontinuity of this type of interfaces to the Al concentration, the measured conduction-band discontinuity of 0.21–0.23 eV would correspond to an Al concentration in $Ga_{1-x}Al_xAs$ of 25–27%, close to the bulk value of approximately 30% that we derived from photoluminescence measurements. Note, however, that from the data of Fig. 3, we can obtain a rough estimate of the *interface* gap width of $Ga_{1-x}Al_xAs$, 1.78–1.80 eV, that is 0.27–0.29 eV larger than the GaAs gap at 77 K; this approximately corre-

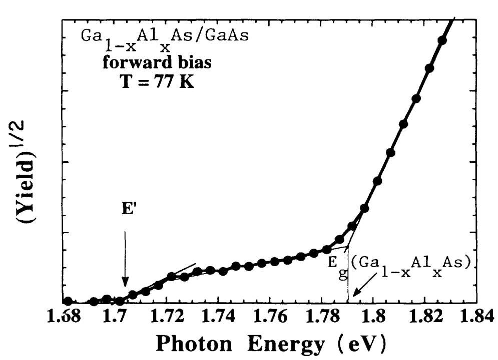

FIG. 3. Results of the experiment of Fig. 1(b) for the same system GaAs-$Ga_{1-x}Al_xAs$ as in Fig. 2.

sponds[21] to an Al concentration of 20%, and thus indicates changes in the stoichiometry on going from the bulk to the region immediately close to the interface. We speculate that such changes could be due to interdiffusion during the deposition; note that this phenomenon cannot be detected by ordinary optical measurements of the gap whereas our method measures the gap at the interface.

We note that the accuracy of ± 10 meV in measuring conduction-band discontinuities is much better than any approach based on photoemission or inverse photoemission.[1] In principle, this accuracy could be affected by the fact that transitions across the discontinuities involve quantized initial states in the quantum well at the conduction-band discontinuity. This would lead to an underestimate of the discontinuity; such a possibility, however, can be ruled out in our case because of the large notch width, as confirmed by comparing tests at different bias values.

Further improvements in the accuracy appear possible because of the superior brightness of the photon source. However, the ± 10-meV accuracy appears already sufficient to discriminate from one another the different theoretical models of band lineups.[1-13] Such a discrimination will be based on a systematic study of different interfaces that has been initiated. The method is in fact easily applicable to other systems, and we have already obtained preliminary results for a chemical-beam-epitaxy-grown $Ga_{0.47}In_{0.53}As$-InP interface. We obtained a conduction-band discontinuity of approximately 0.24 eV, in agreement with the values reported in the literature.[1]

In summary, we have shown that direct measurements of buried conduction-band discontinuities can be performed by optical pumping with a free-electron laser. The accuracy can significantly contribute to the understanding of semiconductor band lineups, a basic conceptual problem in condensed matter physics. Further tests and experiments are in progress at the Vanderbilt Free Electron Center.

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[1]See, for example, *Heterojunction Band Discontinuities: Physics and Device Applications,* edited by F. Capasso and G. Margaritondo (North-Holland, Amsterdam, 1987); *Electronic Structure of Semiconductor Heterojunctions,* edited by G. Margaritondo (Kluwer, Dordrecht, 1988); Proceedings of the 3rd Internaltional Conference on the Formation of Semiconductor Interfaces, edited by P. Perfetti, G. Margaritondo, and O. Bisi [Appl. Surf. Sci. **56-58** (1992)].

[2]G. Margaritondo, J. T. McKinley, D. Rioux, and D. W. Niles, Appl. Surf. Sci. **56-58**, 713 (1992).

[3]F. Gozzo, C. Coluzza, G. Margaritondo, and F. Flores, Solid State Commun. **81**, 553 (1992).

[4]C. Tejedor, F. Flores, and E. Louis, J. Phys. C **10**, 2163 (1977).

[5]F. Flores, A. Muñoz, and J. C. Durán, Appl. Surf. Sci. **41/42**, 144 (1989); F. Flores and J. Ortega, *ibid.* **57-58**, 301 (1992).

[6]J. Tersoff, Phys. Rev. B **30**, 4874 (1984); Surf. Sci. **168**, 275 (1986).

[7]M. Cardona and N. E. Christensen, Phys. Rev. B **35**, 6182 (1987).

[8]M. Schluter, Phys. Rev. B **17**, 5044 (1978); J. Tersoff, *ibid.* **32**, 6968 (1985).

[9]A. R. Miedema, R. Boom, and F. R. De Boer, J. Less-Common Met. **41**, 238 (1975); P. E. Schmid, Helv. Phys. Acta **58**, 371 (1985).

[10]J. L. Freeouf and J. M. Woodall, Appl. Phys. Lett. **39**, 727 (1981).

[11]A. D. Katnani and G. Margaritondo, Phys. Rev. B **28**, 1944 (1983).

[12]S. Baroni, R. Resta, A. Baldereschi, and M. Peressi, in *Spectroscopy of Semiconductor Heterojunctions,* Vol. 206 of *NATO Advanced Study Institute, Series B: Physics,* edited by G. Gasol, A. Fasolino, and P. Lugli (Plenum, New York, 1989), p. 251.

[13]S. Baroni, R. Resta, and A. Baldereschi, in *Proceedings of the Nineteenth International Conference of the Physics of Semiconductors,* edited by W. Zawadzki (Institute of Physics, Polish Academy of Sciences, Wroclaw, 1988).

[14]N. H. Tolk, C. A. Brau, G. S. Edwards, G. Margaritondo, and J. T. McKinley, Proc. SPIE (to be published).

[15]M. Heiblum, M. I. Nathan, and M. Eizenberg, Appl. Phys. Lett. **47**, 503 (1985); Surf. Sci. **174**, 318 (1986); **174**, 312 (1986); M. Eizenberg, M. Heiblum, M. I. Nathan, N. Braslau, and P. M. Mooney, J. Appl. Phys. **61**, 1516 (1987).

[16]G. Abstreiter, U. Prechtel, G. Weimann, and W. Schlapp, Physica **134B**, 433 (1985).

[17]R. H. Fowler, Phys. Rev. **38**, 45 (1931); R. Williams, *ibid.* **140**, A569 (1965); E. Kane, *ibid.* **147**, 335 (1966).

[18]M. Haase, M. A. Emanuel, S. C. Smith, J. J. Coleman, and G. E. Stillman, Appl. Phys. Lett. **50**, 404 (1987).

[19]C. Coluzza, G. Margaritondo, A. Neglia, and R. Carluccio, J. Vac. Sci. Technol. (to be published).

[20]G. Margaritondo and P. Perfetti, in *Heterojunction Band Discontinuities: Physics and Device Applications* (Ref. 1), p. 72.

[21]C. Bosio, J.-L.Staehli, M. Guzzi, G. Burri, and R. A. Logan, Phys. Rev. B **38**, 3263 (1988).