Comments

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Comment on "Piezoreflectance study of short-period strained Si-Ge superlattices grown on (001) Ge"

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Yin *et al.* have reported the use of piezoreflectance to characterize Ge-Si superlattices. Their results show only signals that can be attributed to extended and confined states in bulk Ge, but the method is apparently insensitive to the optical properties of the Ge-Si superlattice itself. In this paper we examine the method used and experimental interpretation proposed by Yin *et al.* From the data available, we conclude that the piezoreflectance experiment of Yin *et al.* may be less sensitive than electroreflectance for these superlattice materials, to the degree that it conveys little useful information on the optical properties of Ge-Si structures.

I. INTRODUCTION

The physical properties of crystalline solids are determined by two features: the chemistry of the atoms involved and the symmetry of the unit cell in which they crystallize. Carbon is the best-known example of an atom that solidifies in several different crystalline configurations: diamond, graphite, and fullerene. However, each of these changes in crystalline symmetry also involves a change in the valence chemistry of the carbon atom, obscuring the effect of symmetry on the properties of the material. We have been working to isolate the fundamental effect of symmetry on properties of crystalline solids by studying Ge-Si strained-layer superlattices where it is possible to impose significant changes in the fundamental symmetry of the unit cell[1] without altering the basic tetrahedral valence chemistry of the atoms involved.[2]

In one of our studies in this area, we reported measurements of the energy-band structure of Ge-Si superlattices having a period of ten atomic monolayers in the ⟨001⟩ direction, and the usual two atomic monolayers in the ⟨100⟩ and ⟨010⟩ directions.[3] In these measurements, recently called into question by Yin *et al.*,[4] we found that new interband optical transitions at about 0.75, 0.94, and 1.24 eV are expected because of the change of the fundamental symmetry of the crystalline unit cell. We used electroreflectance spectroscopy for this measurement because of (a) the proven sensitivity of this method for Ge-Si superlattices having less than 1% of the thickness of the materials used in these measurements;[5] and (b) because photoluminescence could not be measured in these samples.

In the years following the publication of our paper,[3] there have been several studies that have examined the influence of interference effects arising from the multilayer structure of the superlattice and also from Franz-Keldysh oscillations from the electric-field modulation. These studies showed that such interference effects can modify the amplitude of modulated reflectance signals, making it difficult to associate a transition matrix element with a measured reflectance peak in this kind of multilayer structure.[6–8] A common conclusion from these studies is that difficulties in interpretation of modulation spectra taken on multilayer structures mean that these techniques may best be used to support results obtained by the measurements of choice such as absorption or photoluminescence spectroscopy.

Fortunately, during the same time, improvements in crystal-growth procedure have led to samples in which photoluminescence and absorption measurements can now be made routinely on samples of superlattice periods similar to those investigated by us in Ref. 3. In a recent meeting of the European Materials Research Society, the principal conclusions of Ref. 3 have been confirmed and extended by more accurate and conclusive measurements on Ge-Si superlattices, a variety of periodic structures of improved design and quality, so that both photoluminescence and absorption can be obtained easily on samples of more than 2000-Å continuous extent.[9–16]

In a recent paper,[4] Yin *et al.* reported having obtained some Ge-Si samples, previously measured by the author of this comment (TPP) and co-workers. In their paper they compare the results of piezoreflectance measurements made on these samples with some related structures more recently fabricated at AT&T Bell Laboratories. The principal conclusion of this paper is that optical properties of Ge-Si superlattices are not measureable by piezoreflectance, and that the results reported by us

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must not reflect properties of the Ge-Si superlattice structure but rather those of the underlying Ge substrate. In this paper, we wish to respond to some of the issues raised by Yin *et al.*

II. CHARACTERIZATION OF MBE-GROWN SAMPLES

The growth of Ge-Si superlattices is an art involving careful control of strain, epitaxial layer thickness to a few monolayers, substrate preparation, and above all characterization of grown epitaxial structures.[17] The samples whose properties we reported on had been characterized by transmission electron microscopy (TEM) and high-resolution x-ray diffraction (HRXRD). Although not specifically mentioned, the films were also examined by Rutherford backscattering spectroscopy (RBS).[18] These three techniques are needed to measure the existence of zone-folded superlattice spots (TEM), extended superlattice layering (TEM), chemical composition and dislocation level (RBS), and superlattice buffer-layer spacing and spacing and strain (HRXRD). This characterization is not routine, and the interpretation of the HRXRD results alone took several months. In these several samples, the nominal layer composition of the superlattice can be determined from x-ray data, while TEM studies have shown that the interfaces are atomically flat to within about ± 2 atomic monolayers, even though the long-range layering is well controlled, as we have shown in the micrographs of Ref. 17. As a result, the first Fourier component of the superlattice periodicity was well enough defined to yield the appropriate superlattice spot in TEM, establishing that folding of the Brillouin zone had occurred.

Two samples are reported by Yin *et al.* One of them has not been characterized again to confirm its structure, and the other has been examined only by HRXRD. It is important to note that specular HRXRD alone cannot be used to confirm the superlattice nature of the Ge-Si region. It can only give the average composition and thickness. Since Yin *et al.* used only this technique, their characterization analysis cannot distinguish between growth of a strained alloy and growth of a ten-monolayer-period superlattice.

III. MODULATION SPECTROSCOPY

Electroreflectance spectroscopy has been used successfully to measure direct-band-gap transitions in single quantum wells as thin as 50 Å with good signal to noise.[19] It is well established that different forms of modulation spectroscopy, such as photoreflectance and electroreflectance, give different results because the modulation takes place in different locations in the sample. In a heterogeneous sample such as a Ge-Si superlattice, this difference may determine whether or not a signal is seen at all. For example, photoreflectance has been used to study interface states because the modulation occurs preferentially at interfaces between the superlattice and the buffer region.[19]

Piezoreflectance is based on a stress modulation of the band structure using a uniaxial stress field containing both hydrostatic and shear components. Strain-induced modulation of the heterojunction band offset of only 1 meV would create an electric field ten times larger than the field introduced electrically (10 kV/cm). Therefore one might expect to see similarities between piezoreflectance spectra and those obtained in electroreflectance for an "identical" sample. However, this appears not to be the case. While Yin *et al.* may observe features in their spectra due to the Ge buffer and substrate regions in their superlattice samples, they resolve no features at any wavelength that are attributable to the Ge-Si superlattice region whose total thickness exceeds 1000 Å. TEM results make it clear that the superlattice region exists. Therefore it should have optical properties that can be measured. The observation by Yin *et al.* that piezoreflectance does not reveal optical properties of the superlattice region suggests that this technique may be too insensitive to be used for characterization of Ge-Si superlattices. Another possibility, that the optical properties of the superlattice are themselves inherently too weak to be measured, will be discussed presently.

IV. OTHER EXPERIMENTAL RESULTS

The growth and characterization of Ge-Si short-period superlattices, such as those discussed by Yin *et al.*, is a rapidly developing area. For several years, thick (>1000 Å) continuous superlattice films have been grown by the symmetrically strained method on Si substrates.[20] This method eliminates entirely the Ge buffer layers discussed by Yin *et al.* Samples grown in this way show easily measurable band-edge absorption and band-edge photoluminescence.[9,21] The extended nature of the superlattice region gives TEM diffraction with all Fourier components of the superlattice present, in contrast to our samples in which only the first component of the ten-atom period could be detected.[10] Experimental results from photoluminescence,[11,12] absorption,[9,13] ellipsometry,[14] and photocapacitance[15,16] all have identified superlattice band-structure properties in Ge-Si short-period superlattices using optical probes. The photoluminescence results place the low-temperature band gap of the ten-monolayer superlattice near 0.75 eV. Results from Menczigar *et al.* include electroluminescence from diode structures at the same energy.[12] Absorption measurements by Olajos *et al.* identify the band gap for a ten-monolayer-period superlattice at 0.76 eV, with additional absorption at 0.94 eV.[9] These results[9–16] provide indisputable evidence that Ge-Si superlattices have optical properties that can be measured over the energy range of 0.7 to 2.5 eV. In addition, Asami *et al.* have studied such short-period superlattices grown on Si substrates by electroreflectance and photoreflectance spectroscopy.[22] Their results, obtained independently from and concurrently with those reported in Ref. 3, agree closely with our own. The absence of measured piezoreflectance signals in this energy range suggests that the technique may be inappropriate for characterization of Ge-Si short-period superlattices.

V. INTERPRETATION OF RESULTS

One theoretical viewpoint expressed by Yin *et al.* is that the interband matrix element in the superlattice is small because the band-edge states are derived from those of bulk Si and Ge. Two theoretical contributions[23,24] presented at this symposium confirm the notion that direct-band-gap structures can be formed from short-period Ge-Si superlattices, with the calculated optical matrix element several orders of magnitude below that for bulk direct transitions. Both of these papers show that the presence of perturbations such as interface effects and external modulation can have a strong effect on calculated transition probabilities, increasing the amplitude of forbidden transitions by orders of magnitude at the expense of the amplitude of allowed transitions which are decreased to a smaller extent. Under these conditions, it does not appear possible to relate directly the amplitude of features seen in modulation spectroscopy to calculated transition matrix elements.

In preparing this paper, we have reviewed in some detail the conditions under which our original data were taken. This review includes a repetition of the original experiments in which we have reproduced our published results. In this review, we have also paid attention to our effect of possible heating of the samples by the optical beam and the applied bias voltage. The maximum power dissipated in our samples under all modulation conditions was less than 100 mW. This power is insufficient to raise the temperature of the sample more than a degree above the heat sink temperature of 40 K. Evaluation of the standard model for thermal conductivity[25] gives a temperature rise of less than 10^{-2} K after 1 h with a continuous dissipation of 100 mW. It will be recognized by many readers that low-temperature photoluminescence using this level of excitation power also does not lead to a significant temperature rise of the sample.

VI. CONCLUSIONS

The study of Si-Ge superlattices is helping to develop a framework for understanding the relationship between chemistry and symmetry in the electronic structure of crystalline materials. There is controversy surrounding both experimental and theoretical results in this field. Nonetheless most scientists would agree that Ge-Si superlattices do possess optical properties, and that appropriate experimental techniques to use consist of those which are sensitive enough to measure those properties. Because they were able only to measure properties of bulk Ge, the results of Yin *et al.* contribute no new understanding of Ge-Si superlattice structures.

The interpretation of experimental results involving new techniques and materials is a synthesis that may combine guesswork with measured results. The conclusions of Yin *et al.* are based in part on this kind of interpretation regarding (a) the characterization of the sample, (b) the interpretation of the negative piezoreflectance results, and (c) the assertion that sample heating is distorting results. We have shown that the interpretation of Yin *et al.* in each of these areas needs modification. Their experimental results do not support or justify their principal conclusion that superlattice optical properties cannot be measured by modulation spectroscopy.

Finally, we make the hypothesis that new experiments, free from the defects in the work by Yin *et al.*, may show that none of the features that we reported in Ref. 3 can be attributed to optical transitions from the superlattice region of these Ge-Si samples. Such a result would apply only to the samples reported in Ref. 3, and is thus, we think, of historical rather than scientific interest. The progress in the growth of Ge-Si short-period superlattices has produced higher-quality samples whose structure is much better suited to the measurement of the fundamental energy-band structure. The measurements now being performed on these short-period samples are better, more precise experiments that are largely free of the difficulties we encountered in trying to measure the properties of samples grown on a Ge substrate using modulation spectroscopy techniques. We would gladly defer to the results of these measurements. These samples show photoluminescence that peaks near 0.75 eV, the band-gap energy identified in Ref. 3, a result that is closely confirmed in absorption measurements. Additional structure in the absorption spectrum has been identified at 0.94 eV, which is close to the second transition energy of 0.94 eV identified in our original work.

[1] M. I. Alonso, M. Cardona, and G. Kanellis, Solid State Commun. **69**, 479 (1989).

[2] T. P. Pearsall, CRC Crit. Rev. Solid State Mater. Sci. **15**, 551 (1989).

[3] T. P. Pearsall, J. M. Vandenberg, R. Hull, and J. M. Bonar, Phys. Rev. Lett. **63**, 2104 (1989).

[4] Y. Yin, D. Yan, F. H. Pollak, M. S. Hybertsen, J. M. Vandenberg, and J. C. Bean, Phys. Rev. B **44**, 5955 (1991).

[5] T. P. Pearsall, J. Bevk, J. C. Bean, J. M. Bonar, J. P. Mannaerts, and A. Ourmadz, Phys. Rev. B **39**, 3741 (1989).

[6] C. Pickering, D. C. Houghton, and J. M. Baribeau, Proc. SPIE **1286**, 165 (1990).

[7] A. C. Churchill, P. C. Klipstein, C. J. Gibbings, M. A. Gell, M. E. Jones, and C. G. Tuppen, Semicond. Sci. Technol. **6**, 18 (1991).

[8] R. A. Batchelor, Ph.D. thesis, Merton College, Oxford, 1991, pp. 142–188.

[9] Janos Olajos, Jesper Engvall, Hermann Grimmeiss, Horst Kibble, Erich Kasper, and Hartmut Presting, Thin Solid Films **222**, 243 (1992).

[10] W. Jäger, D. Steinkamp, P. Ehrhart, W. Sybertz, K. Leifer, H. Kibbel, H. Presting, and E. Kasper, Thin Solid Films **222**, 221 (1992).

[11] K. Dettmer and J. Weber, Thin Solid Films **222**, 234 (1992).

[12] U. Menczigar, J. Brunner, E. Friess, M. Gail, G. Abstreiter, H. Kibbel, H. Presting, and E. Kasper, Thin Solid Films **222**, 227 (1992).

[13] T. P. Pearsall, C. C. M. Bitz, L. B. Sorenson, H. Presting, and E. Kasper, Thin Solid Films **222**, 254 (1992).

[14] U. Schmid, J. Humlicek, F. Lukes, M. Cardona, H. Presting,

H. Kibbel, E. Kasper, K. Eberl, W. Wegscheider, and G. Abstreiter, Thin Solid Films **222**, 246 (1992).

[15]H. G. Grimmeiss, J. Engvall, V. Nagesh, J. Olajos, H. Presting, H. Kibbel, and E. Kasper, Thin Solid Films **222**, 237 (1992).

[16]V. Nagesh, H. G. Grimmeiss, H. Presting, H. Kibbel, and E. Kasper, Thin Solid Films **222**, 265 (1992).

[17]T. P. Pearsall, R. Hull, J. C. Bean, and J. M. Bonar, Thin Solid Films **183**, 9 (1989).

[18]J. Bevk, L. C. Feldman, T. P. Pearsall, G. P. Schwartz, and A. Ourmazd, Mater. Sci. Eng. B **6**, 159 (1990).

[19]Fred H. Pollak and O. J. Glembocki, Proc. SPIE **946**, 2 (1988).

[20]E. Kasper, H. Kibbel, H. Jorke, H. Brugger, E. Friess, and G. Abstreiter, Phys. Rev. B **38**, 3599 (1988).

Abstreiter, Phys. Rev. B **38**, 3599 (1988).

[21]R. Zachai, K. Eberl, G. Abstreiter, E. Kasper, and H. Kibbel, Phys. Rev. Lett. **64**, 1055 (1990).

[22]K. Asami, K. Miki, K. Sakamoto, T. Sakamoto, and S. Gonda, Jpn. J. Appl. Phys. **29**, L381 (1990).

[23]M. Jaros, A. W. Beavis, P. J. Hagon, R. J. Turton, A. Miloszewski, and K. B. Wong, Thin Solid Films **222**, 205 (1992).

[24]G. Theodorou, C. Tserbak, and H. M. Polatoglou, Thin Solid Films **222**, 209 (1992).

[25]E. R. G. Deckert and R. M. Drake, Jr., *Analysis of Heat and Mass Transfer* (McGraw-Hill, New York, 1972), pp. 168 and 169.