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Advances in Microcrystalline and Nanocrystalline Semiconductors–1996

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PREFACE

This volume includes the proceedings of Symposium Q 'Advances in Microcrystalline and Nanocrystalline Semiconductors—1996,' held during the 1996 MRS Fall Meeting in Boston, Massachusetts. This is the fourth in a series of MRS fall symposia devoted to research on the theory, preparation, characterization, and application of nanocrystalline, microcrystalline, and polycrystalline semiconductors. The first symposium was conducted in 1989 and yielded the volume entitled 'Materials Issues in Microcrystalline Semiconductors,' edited by P.M. Fauchet, K. Tanaka, and C.C. Tsai (MRS Symposium Proceedings Volume 164), which included about 60 papers. The second symposium was conducted in 1992 and yielded the volume entitiled 'Microcrystalline Semiconductors: Materials Science and Devices,' edited by P.M. Fauchet, C.C. Tsai, L.T. Canham, I. Shimizu, and Y. Aoyagi, (MRS Symposium Proceedings Volume 283), which included about 150 papers. The explosion of interest in nanocrystalline semiconductors, and the consequent expansion in the symposium between 1989 and 1992, was a result of the demonstration in 1990 by L.T. Canham that visible photoluminescence is emitted at room temperature from the nanostructures in porous silicon. Over the last five years, scientific and technological interest has continued undiminished. The third symposium was conducted in 1994 and yielded the volume entitled 'Microcrystalline and Nanocrystalline Semiconductors,' edited by R.W. Collins, C.C. Tsai, M. Hirose, F. Koch, and L. Brus, (MRS Symposium Proceedings Volume 358), which included about 60 papers. In the five-day span of the most recent symposium in 1996, 200 papers were presented in 25 oral and poster sessions, some 80% of which appear in this volume.

Symposium Q of the 1996 MRS Fall Meeting brought together scientists and engineers with diverse backgrounds to share their recent advances in (i) semiconductor nanocrystals and nanostructures, (ii) porous semiconductors, and (iii) nanocrystalline, microcrystalline, and polycrystalline semiconductor films. With the exception of the larger-grained microcrystalline and polycrystalline films, the unifying scientific theme underlying these research areas is the modification of the properties of semiconductors that results from electron confinement in nanometer-sized structures. The challenge in *all* of these fields, however, is to understand (and often to minimize, as in the case of the larger-grained films) the influence of structural features including surfaces, interfaces, grain boundaries, and intergranular phases on the often unique (as in the case of light-emitting Si) semiconducting properties of the grains themselves. A common technological application among semiconductor nanocrystals/nanostructures and porous semiconductors stems from their unique optical properties, specifically their efficient visible photoluminescence that persists at room temperature, owing to the confinement of the photoexcited charge carriers. As a result, electroluminescent devices with a wide variety of designs are being developed and optimized, and their integration with Si microelectronics has now been demonstrated. The challenges are to ensure that the crystallites are optimally-sized for visible light emission, and also that surfaces are optimally-oxidized to avoid nonradiative recombination and instability, while still allowing carrier transport in light-emitting diodes. An important application of the polycrystalline thin films is large-area electronics and photovoltaics on inexpensive glass substrates. In this case, the challenge is one of obtaining large crystallite sizes, while ensuring that grain boundaries are well-passivated. Common themes and challenges have provided a unique opportunity for fruitful interactions among the scientists and engineers active in these fields.

Materials advances presented during the symposium and described in this volume involve structures spanning more than five orders of magnitude in size, ranging from molecular clusters <1 nm in size to single-crystal regions in thin films >10 μm in size. At the smallest end of the scale, detailed descriptions of predicted and observed properties of Group IV molecular clusters are presented here. At the largest scale, novel excimer laser annealing approaches for thin Si films have been described that yield single-crystal grains large enough for fabrication of thin-film transistors within their boundaries. In a plenary session, excellent overviews of the current status of research in (i) microcrystalline and nanocrystalline semiconductor films, (ii) semiconductor nanocrystals, and (iii) porous semiconductors were provided by K. Tanaka (NAIR), L.E. Brus (Columbia University), and L.T. Canham (DRA-Malvern), respectively.

In the area of thin films, advanced techniques for characterizing the nucleation, growth, and structure of nanocrystalline Si films were reported, based on *in situ* scanning tunneling microscopy (K. Ikuta et al., NAIR). The motivation for better understanding and optimizing the growth of such films was provided through the demonstration of a 10.7%-efficient tandem solar cell made with a stable, all-microcrystalline Si bottom cell (H. Keppner et al., University of Neuchatel). In the area of nanocrystals, phase transition kinetics as a function of size are providing information on the intrinsic nucleation of solid-solid phase transformations, e.g., from four-coordinate wurtzite to six-coodinate rock-salt in CdSe (C.-C. Chen et al., University of California - Berkeley). In the future, this research may lead to approaches for chemical synthesis of a wide variey of nanocrystals in metastable phases. Other studies of the optical properties of CdSe nanocrystals clearly reveal the atomic-like nature of the light-emitting state. By eliminating spectral inhomogeneities, a factor of 50 reduction in the photoluminescence linewidth (to <120 μeV at 10 K – resolution limited) in comparison with previous work was reported (S. Empedocles et al., MIT). In the area of porous silicon, materials advances reported during the symposium focused on improving control over the structure and enhancing the stability of room-temperature light-emitting material. The fact that such advances are meeting with success is demonstrated with the report of device integration of Si-based LEDs (Hirschman et al., University of Rochester). In addition, new applications of porous silicon continue to emerge in biotechnology (L.T. Canham et al.) and in passive optics such as Bragg reflectors, Fabry-Perot filters, rugate filters, and gratings (M. Thönissen et al., KFA-Jülich; G. Lérondel et al., Université de Joseph Fourier).

This is just a sampling of the wide-ranging, but interconnected topics covered by the microcrystalline/nanocrystalline semiconductor symposium and included in this volume. Owing to the challenging nature of the problems, the rapid advance of the field, and the importance of microcrystalline and nanocrystalline semiconductors in electronic and optoelectronic device applications, future symposia on this subject are expected to continue generating great interest. For this reason, we have adopted the title "Advances in Microcrystalline and Nanocrystalline Semiconductors - 1996," to indicate that the symposium and volume provide a snapshot in time of rapidly advancing research and applications. The volume is divided into twelve parts. In many cases, assigning a paper to a given part has been difficult, owing to the many interconnections of the three materials fields listed above. The plenary papers are collected in Part I, entitled *Current Status and Future Prospects of Research in Microcrystalline and Nanocrystalline Semiconductors*, and include the excellent overviews by K. Tanaka, L. Brus, and L.T. Canham.

The theme of the papers in Part II is the theory of semiconductor molecular clusters and nanocrystals confined in two or three dimensions (wires or dots, respectively), with an emphasis on calculating electronic band structure, density of electronic states, and optical properties. Studies on this topic are essential in order to develop a better understanding of many of the experimental observations presented in the proceeding parts of the volume. The themes of the papers in Part III concern luminescent Group IV clusters/nanocrystals and quantum wells, semiconductor systems confined in three and one dimensions, respectively. The papers in this part are sequenced primarily by the preparation technique, starting with an invited article by J. Budai and coworkers (Oak Ridge National Laboratory) on nanocrystals prepared by the ion implantation technique. After several articles of similar topic, the articles that follow describe nanocrystal and nanophase materials prepared by magnetron sputtering, pulsed laser ablation, spark processing, inert gas evaporation, and chemical synthesis. The later papers in Part III describe studies of Si quantum wells, including an invited article by D.J. Lockwood and coworkers (National Research Council of Canada), in which the wells are prepared by a variety of vapor deposition techniques, as well as studies of crystallites in thin films prepared by chemical vapor deposition. The themes of the papers in Part IV are similar to those of Part III, but the primary interest is in the electronic and vibrational states, self-assembly mechanisms, and thermoelectric properties of the Group IV nanostructures, rather than their photoluminescence characteristics. The papers in this part are sequenced by material, starting with an invited article by G.P. Lopinski and coworkers (Penn State University) on graphitic carbon nanocrystals, and progressing through Si, Ge, and $Si_{1-x}Ge_x$ alloy systems. The papers in Part V are concerned with the Group III-V, Group II-VI, and metal sulfide, iodide, and oxide nanocrystals, and are sequenced in that order. In the research described in the first several papers on III-V and II-VI nanostructures, the preparation method is vapor deposition including molecular beam epitaxy and chemical vapor deposition, whereas in most of the remaining papers, chemical or electrochemical deposition methods from solutions are employed.

Porous silicon provides the focus of Parts VI and VII. Part VI starts with papers that concentrate on the synthesis of porous silicon from various starting materials, including an invited article by J.-N. Chazalviel and coworkers (Ecole Polytechnique) which describes porous silicon from hydrogenated amorphous silicon films (a-Si:H). Most of the remaining papers in Part VI concern the process/property relationships in porous silicon, its structure and surface chemistry, and the role of the surfaces in various aspects of the photoluminescence process. Also included in Part VI are several papers that describe the deposition of polymers or other semiconductors into porous silicon. An article describing studies of porous SiC concludes Part VI. The papers in Part VII describe studies of the basic photoluminescence properties and mechanisms of porous Si, particularly focusing on the emitting nanocrystals within porous Si. Other papers in this part describe the optical properties and Raman spectra of porous Si. The emphases of the papers in Parts VIII and IX are the applications of nanocrystal and porous semiconductors, as well as the specific properties that relate to these applications, such as electronic transport. The applications in Part VIII include the non-light-emitting applications, starting with applications in biotechnology, as described in the initial invited article by L.T. Canham and co-workers. The subsequent series of papers in Part VIII present photochemical, photoelectronic, and photovoltaic properties and applications. The final papers describe passive optical applications including sensors, gratings, and interference filters. The first set of papers in Part IX describe light-emitting properties and applications of porous Si, including the electroluminescence process and the performance of light-emitting diodes. In addition, the integration of Si optoelectronics is demonstrated, and the performance of optical microcavities formed from porous Si is described at the end of Part IX.

Papers in Parts X-XII present research results on the nano-, micro-, and polycrystalline thin films, including their preparation, properties and applications. The distinguishing character of these studies from many in the earlier parts is that the intended applications are more often electronic or photovoltaic than light-emitting. Many of the materials and films described in earlier parts are prepared with a wide-gap intergranular phase designed to promote quantum confinement and passivate the surfaces, whereas the intergranular phase of the films in Parts X-XII is most often incidental and detrimental. Part X begins with papers that concentrate on the preparation and properties of micro/nanocrystalline silicon, including an invited one by F. Finger and coworkers (Forschungszentrum Jülich), continues with papers on the properties of these films, and concludes with those describing materials other than Si, such as ITO and PZT. Papers in Part XI describe the applications of the nano/microcrystalline films in devices. These include, in order of presentation, photovoltaic devices, thin-film transistors, photodiodes, light-emitting diodes, and photoconductors. An invited article by H. Keppner and coworkers highlights Part XI. Many papers in Part XII deal with the preparation, characterization, and application of crystalline Si films having large grain sizes, using inexpensive substrates such as glass. The initial papers in this part describe laser and thermal annealing methods for the preparation of these materials, including invited articles by K.S. Choi and M. Matsumura (Tokyo Institute of Technology) focusing on thin-film transistor applications and M. Tanaka and co-workers (Sanyo Electric) focusing on photovoltaic applications. Succeeding papers describe materials preparation by enhanced chemical vapor deposition (CVD) and low-pressure CVD techniques, and materials characterization. The final papers of the volume describe the preparation and properties of crystalline materials other than Si that exhibit novel properties with potentially important applications.

In concluding, the organizers wish to thank the many referees who dedicated their free time at the symposium in Boston to review all the articles in this volume. The authors also deserve praise for revising their articles within a very narrow time frame to ensure rapid publication. All invited speakers and session chairs should be commended for their essential contributions in making the symposium in Boston a success. Special thanks go to K. Tanaka, L. Brus, and L.T. Canham for their extra efforts in providing comprehensive surveys of prior research and anticipated future directions in the three focus areas of the conference. Our thanks also go to Jennifer Barker for her help in preparing and indexing the volume. Finally, the organizers are indebted to the sponsors below for their generous contributions in making this symposium possible:

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