

On the occasion of the 80th birthday of Professor Yitai Qian: Celebrating 60 years of innovation in solid-state chemistry and nanoscience

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Professor Yitai Qian, a pioneer in nanomaterials, an academician of the Chinese Academy of Sciences (CAS) and a professor at the University of Science and Technology of China (USTC), has committed his entire career to the development of solid-state chemistry and nanoscience in China. Prof. Qian is recognized for his pioneering contributions to the development of emerging high- T_c superconducting materials, and the development of solvothermal methodology for the synthesis of a wide range of low-dimensional nanomaterials. Beyond the landmarking scientific advancements he has made in his own trailblazing research, Prof. Qian has directly mentored more than 100 excellent PhD students, in addition to many more master and undergraduate students, many of them have gone on to become the leading scientists in China and worldwide in the field of nanoscience and beyond. Without exaggeration, the academic offspring and extended family of Prof. Qian have contributed a substantial portion of nanoscience and nanotechnology development over the past several decades.

In celebrating Prof. Qian's 80th birthday and his 60 years of continued innovation in solid-state chemistry and nanoscience, *Nano Research* publishes this special issue with a collection of 39 papers from Prof. Qian's former mentees and collaborators. To reflect Prof. Yitai Qian's contributions to the advancement of solid-state chemistry and nanoscience development in China over the past 60 years, here we provide a brief account of his career path and major academic achievements over the past six decades.

1. Establishing the research program in solid-state chemistry

Solid-state chemistry mainly involves the preparation, structure and the properties of solid-phase materials, particularly crystalline materials. Prof. Qian began his research on the growth of microcrystals under the guidance of Prof. Chenghuang Xu in Shandong University sixty years ago and discovered that Al^{3+} could induce the ammonium dihydrogen phosphate (ADP) to form crystalline microwires, and completed his undergraduate thesis of "The effects of impurities on the crystal growth of ADP".

Prof. Qian joined the division of inorganic chemistry of the newly founded USTC in Beijing in 1962, where he first assisted

Prof. Heng Fu of the CAS in teaching "Crystal Chemistry", and then served as the leading instructor of the course since 1966.

In 1975, the CAS charged USTC, which had been moved to Hefei, to establish the strategic development plan for each division. Prof. Qian, then as the director of the division of inorganic chemistry, advocated to place high priority on solid-state chemistry and strengthen collaborations with the solid-state physics division of the USTC and other research institutes within the CAS.

The solid-state chemistry represented a major research focus within the CAS around this period, with the Fujian Institute of Research on the Structure of Matter focusing on the growth of new crystals, Changchun Institute of Applied Chemistry on developing luminescent materials, and Shanghai Institute of Ceramics on polycrystalline silicates. Thus, by exchanging students with these institutes through the institute-division joint program within the CAS, the solid-state chemistry program at USTC ramped up quickly.

Within this period, Prof. Qian began to recruit graduate students within the discipline of Irradiation Chemistry in 1978. In the master thesis of Yicheng Wu, they conducted the first electron microscopy studies to discover that rare earth element doping could induce the growth of one-dimensional γ - Fe_2O_3 nanowires with enhanced magnetic properties [1]. After cultivating a batch of master students in inorganic chemistry, Prof. Qian proposed to apply for the establishment of the Master Program in 1982, which was approved by the Ministry of Education in 1984.

2. Solid-state chemistry of heterogenous catalysts

In 1982, Prof. Qian conducted research in the Solid-State Laboratory of Prof. A. Wold at Brown University as a visiting scholar, where he conducted a fundamental investigation of the phase transition behavior of iron oxide catalysts for Fischer-Tropsch process under the reducing atmosphere of H_2 and CO. Through this three-year visiting program, Prof. Qian developed modern experimental skills in solid-state chemistry, and published five research papers. At the same time, he recognized the critical importance of the laboratory platform for scientific research and the thesis proposal for the PhD training program, which has been immensely helpful for him in establishing a

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leading research laboratory and a PhD program in solid-state chemistry at USTC. Later, Prof. Qian recommended four colleagues at USTC to visit Brown University and conduct research in solid-state chemistry.

3. Exploration of new oxide superconductors

After returning to USTC in 1985, Prof. Qian supported Prof. Qirui Zhang's funding application of "*The exploration of new oxide superconductor materials*" from the National Science Foundation of China (NSFC), which predates the discovery of high- T_c superconductor in 1986 and the international rush on the yttrium lanthanide copper oxides. For its extraordinary innovative nature, the proposal was granted with special award funding by NSFC. Later, the preparation and property exploration of superconductor crystalline materials was awarded the third prize of CAS Natural Science and further received funding support from the "863 program".

Closely following the international research rush on the superconductor of YBaCuO, Dr. Rukang Li prepared a new layered cuprate of $\text{TaSr}_2(\text{NdCe})_2\text{Cu}_2\text{O}_y$ as a Ta analog of the Tl-1222 phase superconductor, and conducted the detailed structural and electrical characterization, which attracted considerable attention [2]. For this reason, Prof. Linzhao Qian, an academician of the CAS, wrote a letter to encourage Prof. Qian and pointed out that it was necessary to use high-resolution electron microscopy for further structural analysis. Soon, the USTC purchased and installed a high-resolution electron microscope, which provided substantial help to Prof. Qian's group on the studies of nanomaterials in the following years. Later, Dr. Xianhui Chen further studied the bismuth-based copper oxide superconductor materials [3]. In 1993, Prof. Qian was approved to be the Ph.D. advisor in Solid-State Physics.

The copper-based oxide superconductor materials exhibit high superconducting temperature, but are generally plagued by limited stability. For example, it was reported that the superconductivity of the yttrium-based copper oxides can be weakened under water exposure, and the structures of bismuth-based copper oxides become amorphous under grinding. Dr. Wenbing Wu found that bismuth-based single crystal superconductor materials could decompose at the temperature of 500 °C [4]. During this period, Prof. Qian's group recommended many excellent undergraduate students for graduate studies abroad, including Peidong Yang to Harvard University.

During this period, Prof. Qian also developed and wrote the textbook of "*Crystal Chemistry*", which was published in 1988 and became the primary textbook for the course of "*Crystal Chemistry*" at USTC and a key reference for studying superconductor materials.

4. Chemical synthesis of nanomaterials

At the same time, the division of inorganic chemistry at USTC began to explore new research directions. Under the suggestions of Prof. Tingsui Ge, an academician of CAS, Prof. Qian embarked his effort in the chemical synthesis of nanomaterials. For the development of the chemical synthesis of nanomaterials and the talent cultivation, Prof. Qian proposed to apply to establish the PhD program of Inorganic Chemistry, which was finally approved by the Ministry of Education in 1996.

To strengthen the relationship with the peers in inorganic chemistry, Prof. Qian also applied for funding support of inorganic chemistry with the proposal entitled "*Low-temperature and non-aqueous synthesis of III-V group nanomaterials and*

their optical properties". Due to the highly innovative nature at the time of the proposal, the application was not successful in regular fund track, but supported through a venture funding track aiming to fund high-risk, high-reward project.

With the support, Dr. Yi Xie et al. developed a benzene-thermal approach to synthesize nanocrystalline GaN at relatively low temperature, which was published in *Science* [5]. Later, Dr. Yadong Li et al. developed a solvothermal co-reduction route to synthesize nanocrystalline III-V semiconductor InAs [6], and further synthesized diamond powder in mild condition by reducing CCl_4 using sodium metal, which was also published in *Science* in 1998 [7]. These landmarking advancements inspired a rapid ramping up of the relevant research in Prof. Qian's group and worldwide. For instance, Dr. Yang Jiang et al. synthesized multi-wall carbon nanotubes [8]. Dr. Shuhong Yu et al. synthesized chalcogenide nanorods in monodentate ligand of butylamine [9]. During this period, many undergraduate students associated with Prof. Qian's lab was admitted top graduate programs worldwide, including Xiangfeng Duan, Yu Huang, Yue Wu and Guihua Yu who conducted their graduate study in Harvard University.

For his pioneering contribution to nanomaterials and superconductors, Prof. Qian was elected as an academician of the CAS in 1997. In 2001, Prof. Qian received the second prize of the National Natural Science Award for his pioneering contributions to the development of solvent-thermal synthesis method. At the same time, Prof. Qian and Prof. Xiaoguang Li in Solid-State Physics jointly led a team to apply for the PhD program in materials science, which was approved by the Ministry of Education in 2001.

5. Preparation and industrialization of rechargeable battery materials

With the rapid development of chemical synthesis of nanomaterials, Prof. Qian forged further ahead to explore the material functions and potential applications. In particular, he started to investigate secondary battery materials. The long-term research experience on superconductors and nanomaterials has established the solid foundation for the team to rapidly adjust the research directions. Secondary battery materials have basic requirements for ionic conductivity and electronic conductivity, and often share similar layered structures of superconducting materials. Prof. Qian worked as the principal investigator for "*Micro-nanocrystallization and the performance improvement rules of the key materials for secondary batteries*", a sub-project of the Ministry of Science and Technology 973 program led by Prof. Jun Chen.

In the field of lithium-ion batteries, Prof. Qian's group have made great progress in silicon, carbon and the Si-C composite anodes. In 2004, Dr. Guifu Zou et al started to use carbon materials as the anodes of lithium-ion batteries [10]. Dr. Xiaona Li et al. synthesized a selenium-sulfur solid solution with better electrochemical performance than elementary molecules [11]. Dr. Ning Lin et al. prepared silicon nanocrystalline by thermal reduction of SiCl_4 in molten salt condition, which was highlighted as "simply silicon" by *Nature Materials* [12].

Recently, Prof. Qian has focused his attention on the industrialization potential of aqueous secondary batteries for their superior safety. In 2017, Prof. Qian established a pilot scale laboratory in Jiangsu Institute of Technology, Changzhou. Dr. Zhiguo Hou scaled up the $\text{LiMn}_2\text{O}_4/\text{NaTi}_2(\text{PO}_4)_3$ system by 50,000 times to make a large battery of 12 Ah capacity [13]. After 4-year efforts, they have successfully achieved practical demonstration of the first water-based secondary battery with

energy density superior to that of the commercial lead-acid batteries.

Building upon the seminal contributions by Prof. Qian along with many other pioneers in the field, we have witnessed tremendous progress in nanoscience and nanotechnology, from synthetic control, to fundamental investigation of size-scaling relationship, emerging properties and novel functions, to broad technological opportunities in catalysis, energy, electronic and optoelectronics. This special issue includes 39 papers contributed by Prof Qian's former mentees and collaborators, aiming to highlight the rapidly expanding field of nanoscale materials that Prof. Qian has helped define.

The synthetic control has always been central for functional nanomaterials. Extraordinary efforts have been devoted to developing methodologies for rational synthesis of a wide range of nanomaterials with highly elaborated control of chemical compositions, sizes, shape, crystal structures, interfaces/surfaces, and heterostructured architectures, which in turn allows to precisely tailor their properties and functions. In the first example of this special issue, Di Sun from Shandong University has shown that, by tuning the surface ligands, two kinds of superatom gold selenido nanoclusters can be produced with distinct geometric and electronic structures [14]. Introducing heteroatoms into nanocrystals significantly affects their electronic and optical properties. In this way, Dabin Yu from the National University of Defense Technology demonstrates that introducing Ce^{3+} into $CsPbI_3$ quantum dots results in optimized photoluminescence properties [15]. Xuefeng Qian from Shanghai Jiao Tong University further shows that creating copper vacancies in Cu_3SnS_4 yields plasmonic nanocrystals with broad solar-spectrum absorption, efficient charge separation and enhanced reaction activity, leading to superior solar-to-hydrogen conversion efficiency [16]. Synergistically introducing cation dopants and anion vacancies, as shown by Chong Xiao and Yi Xie from University of Science and Technology of China, now offers a new route to regulate the localized electron distributions and achieve enhanced solar-driven water oxidation efficiency [14]. Analogously, Jun Lu from Beijing University of Chemical Technology reports a new class of ultrathin Ni/V-layered double hydroxide nanosheets, wherein the abundant oxygen vacancies enable efficient photocatalytic nitrogen reduction [18].

Guozhen Shen from Institute of Semiconductors, Chinese Academy of Sciences reports a surface sulfide treatment strategy to substantially reduce surface electron traps in Cd_3As_2 nanowires, thereby enhancing their NIR photoresponse [19]. By exploiting the epitaxy principle, Qing Yang from University of Science and Technology of China presents catalytically grown PbSe nanorods with unconventional growth direction and excellent broadband photodetection performance [20]. As for two-dimensional (2D) nanomaterials, Yu Huang and Xiangfeng Duan from University of California, Los Angeles now demonstrate that the solution-processed 2D van der Waals thin films can be integrated as active matrix for high-performance spatially resolved pressure sensing [21]. Guangcheng Xi from Chinese Academy of Inspection and Quarantine develops a new kind of MoO_2 hollow nanospheres, with plasmonic effect for surface enhanced Raman scattering [22]. Hongcai Zhou from Hunan University reports a stable porous hollow nanocage with great potentials for mechanobiology-related applications [23].

Beyond single-component nano-objects, integrating them into multicomponent heterostructures can enable new functions

and improved performances. Hao Wang and Guifu Zou from Soochow University observe ambipolar photoresponse in the 2D van der Waals heterostructures of MoS_2 and $Ti_3C_2T_x$ MXene [24]. Thanks to the multi-junction synergy and surface adsorption of TiO_2 top layers, Tao Li from Shenzhen Technology University achieves the adjustable multi-color detector with controlled photoelectric response to ultraviolet (UV) and visible light [25]. Furthermore, when upgrading the conducting TiO_2 layers into well-arranged nanopillar arrays in solar fuels, Jie Zhao and Guifu Zou of Soochow University report improved light-harvesting and carrier-transport abilities in the resultant multi-layered heterostructure devices [26]. By engineering type-II heterojunction between $CuBi_2O_4$ and CuO , Dekun Ma from Shaoxing University widens spectral response range and promotes the charge separation efficiency, resulting in enhanced photoelectrochemical activity towards H_2O_2 production [27].

Electrochemical conversion technologies, such as water electrolysis, fuel cells, and metal-air batteries, are now of increasing importance for renewable energy generation, conversion and utilization. The core of these technologies relies on a number of electrocatalytic reactions, including oxygen evolution reaction (OER), oxygen reduction reaction (ORR), hydrogen evolution reaction (HER), and hydrogen oxidation reaction (HOR), that often require costly precious metal electrocatalysts. Understanding these catalytic processes and developing high-performance catalysts are thus central for practical implementation of these technologies. Xiaogang Yang from Suzhou University of Science and Technology delivers a comprehensive review on the OER process, including reaction intermediates, reaction mechanisms, and factors for OER reaction kinetics [28]. Gongming Wang from University of Science and Technology of China reports a highly efficient HER catalyst of Cr-doped Ni_3N/Ni with an impressive overpotential [29]. To reduce the voltage for water splitting, Shenglin Xiong from Shandong University develops a bifunctional electrocatalyst of Mo-doped $NiS/Ni(OH)_2$, which promotes both OER and HER [30]. Changhua An from Tianjin University of Technology also reports a bifunctional OER and HER catalyst based on $FeNi@MXene@Ni$ foam with remarkable water-splitting performance [31]. Meanwhile, Yadong Li from Tsinghua University develops an efficient OER/ORR electrocatalyst featuring Co and Ni-based dual atomic sites, which serves as cathode in the zinc-air battery and delivers a high maximum power density [32]. Cheng Wang from Tianjin University of Technology designs a V-doped Ni_3N/Ni heterostructure catalyst for both HER and HOR with superior activity for efficient hydrogen production and utilization [33].

Besides these oxygen- and hydrogen-involving energy conversion reactions, other electrocatalytic reactions, such as CO_2 reduction reaction (CO_2RR), formic acid oxidation reaction (FAOR), and nitrogen reduction reaction (NRR), have also attracted extensive attention. Yu Huang from University of California, Los Angeles reports a Cu-Ag interfaced catalyst by introducing Ag sites on Cu nanowires, leading to significantly improved CO_2RR selectivity toward methane [34]. Yi Ding from Tianjin University of Technology reports the core-shell structured Au-Pd-Au as an efficient FAOR electrocatalyst and realizes high intrinsic mass activities [35]. Peidong Yang from University of California, Berkeley develops a NRR catalyst with ultrafine Au_{25} nanoclusters loaded on sulfur-doped graphene, and obtains remarkable activity and stability for

ammonia production [36]. Additionally, Jinhua Zhan from Shandong University reports ultrathin porous Co_3O_4 nanosheets with synergistic Lewis acid-base sites, which can serve as high-performance nanozymes for peroxidase-like catalysis [37].

With the ever-increasing development of consumer electronics, electric vehicles, and grid-scale energy storage, there is a growing demand for high-energy and high-power rechargeable batteries. Regardless of engineering design at the battery level, the development of new active materials and the elaborated hierarchiral structural design are essential for boosting the fundamental capacity limit or rate capability. In terms of battery materials, Jian Yang from Shandong University presents N,P-codoped reduced graphene oxide supported few-layered MoS_2 nanosheets, which exhibit high specific capacity and ultra-long cycle life for potassium-ion storage [38]. Weixin Zhang from Hefei University of Technology reports Zn/Mg dual-doping in $\text{P2-Na}_{0.67}\text{MnO}_2$ to adjust the material composition and enlarge interplanar spacing, simultaneously improving the Na-ion diffusion coefficient and the structural stability of the layered material during cycling [39]. In terms of the effect of active sites on the behavior of vanadium redox reactions, Chuankun Jia from Changsha University of Science and Technology reports a large-scale graphene-modified carbon felt with a high voltage efficiency and long-cycle stability for vanadium redox flow battery [40]. Qianwang Chen from University of Science and Technology of China designs a core-shell structure consisting of an active bismuth sulfide core and a highly conductive sulfur-doped carbon shell ($\text{Bi}_2\text{S}_3@\text{SC}$) as a novel anode material for potassium-ion batteries [41]. Meanwhile, Liqiang Xu from Shandong University further reveals the electrochemical mechanism of hierarchical composite as high-performance anode for potassium ion batteries [42]. In terms of structural design, Guihua Yu from University of Texas at Austin provides a comprehensive overview on the fabrication, advances, and challenges of vertically aligned 2D material electrodes for scalable energy storage systems [43]. Jinkui Feng from Shandong University demonstrates a flexible and freestanding MXene/COF framework to stabilize the Li anode in Li-S battery [44]. Zhaoping Liu from Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences reports porous carbon nitride microspheres as uniform lithophilic coatings on Cu/Li foils to achieve dendrite-free lithium plating [45]. Junhao Zhang from Jiangsu University of Science and Technology develops a superior zephyranthes-like Co_2NiSe_4 arrays grown on butterfly wings derived carbon framework (Z- $\text{Co}_2\text{NiSe}_4/\text{BWC}$), which exhibit excellent Na^+ storage performances [46].

Beyond those technologies, the emerging synthetic materials have also fueled a broad range of energy- and environment-related applications, including thermoelectrics, spintronics, superconductors, flexible electronic devices, and water purification. Yue Wu from Iowa State University provides a focused review highlighting the significance of porosity in thermoelectric materials on enhancing their figure of merits [47]. By identifying the phonon dispersion mismatch at device interfaces, Xinqiang Wang from Peking University now offers the design principles for optimizing thermal conductance in electronic devices [48]. Lingfei Wang and Wenbin Wu from University of Science and Technology of China find that asymmetric interfaces and ferroelectric-like polar distortion in atomically thin superlattices cooperatively trigger the ferromagnetic phase with high Curie temperature, offering guidelines for designing novel oxide-based spintronic devices

[49]. Guanghan Cao from Zhejiang University develops a block-layer model to rationalize the essential roles of lattice match and charge transfer in the formation of intergrowth structures, which may be implementable for discovering new superconducting materials [50]. To fabricate flexible electronic devices, Shu-Hong Yu from University of Science and Technology of China develops a solvothermal polymerization process to change the cross-linking networks of polymer elastomer [51], producing highly stretchable, soft, and sticky elastomer that holds potentials for artificial electronic skins. Yingjie Zhu from Shanghai Institute of Ceramics, Chinese Academy of Sciences realizes a flexible, high-strength, and versatile hydrogel with fiberboard-and-mortar hierarchically ordered structures, offering a new solution to water purification and pollution treatment [52].

Overall, this special issue provides an excellent collection of reviews and research articles on the topic of solid-state chemistry and nanoscience. We are grateful to all authors for their contributions. The editorial board of *Nano Research* congratulates Prof. Yitai Qian for his long-term contributions to this booming field and wishes him a happy and healthy birthday. We look forward to seeing his continued innovation and relentless spirit in science in the years and decades to come.

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Biography of Professor Yitai Qian

Professor Yitai Qian was born in Wuxi, Jiangsu Province in 1941. In 1962, he received BS from the Department of Chemistry of Shandong University. Then, he joined the inorganic chemistry faculty at the University of Science and Technology of China (USTC). From 1982 to 1985, he worked as a visiting scholar in Brown University and was engaged in Fischer Tropsch process research and film preparation of iron catalysts. He visited Brown University again from 1989 to 1990. In 1986, he was appointed as an associate professor at USTC. In 1992, he studied the thermal analysis technology as a visiting scholar in Purdue University. In the same year, he was appointed as a full professor. He was then appointed as the doctoral supervisor of condensed matter physics (1994) and inorganic chemistry (1996). In October 1997, he was elected as an academician of the Chinese Academy of Sciences. From June 2000 to June 2006, he served as the vice president of the Council of Chinese Chemical Society. He served as the Dean of the School of Chemistry and Materials at USTC from 1999 to 2004. In 2005, he joined the School of Chemistry and Chemical Engineering of Shandong University, and was appointed as the Director of the academic committee of the Key Laboratory of Colloid and Interface Chemistry of the Ministry of Education, China. In 2008, he was appointed as the Dean of the School of Chemistry and Chemical Engineering of Shandong University. In the same year, he was elected as a Fellow of the Royal Society of Chemistry. He received the 2nd Prize of National Natural Science Award in 2001 and the Science and Technology Innovation Award of Ho Leung Ho Lee Foundation in 2015.