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Dialectics of nature in materials science: binary cooperative complementary materials

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ABSTRACT Binary cooperative complementary materials, consisting of two components with entirely opposite physiochemical properties at the nanoscale, are presented as a novel principle for the design and construct of functional materials. By summarizing recent achievement in materials science, it can be found that the cooperative interaction distance between the pair of complementary property must be comparable with the scale of related physical or chemical parameter. When the binary components are in the cooperative distance, the cooperation between these building blocks becomes dominant and endows the macroscopic materials with unique properties and advanced functionalities that cannot be achieved by either of building blocks.

Keywords: dialectics of nature, binary cooperative complementary materials, cooperative interaction distance, material design

The binary cooperative complementary phenomenon, which refers to two entirely opposite but cooperative, complementary states, may be observed at many different levels of nature [1]. For example, we have a positive nucleus and negative electrons at the atomic level, hydrophilicity and oleophilicity in molecules, hard inorganic and soft organic components in biological tissues, male and female in biology, odd and even in mathematics, north and south in magnetism, and matter and anti-matter in the universe [2] (Fig. 1). The law of unity and interpenetration of opposites was proposed in "Dialectics of Nature," an unfinished 1883 work by Friedrich Engels. He stated "Everywhere we look in nature, we see the dynamic co-existence of opposing tendencies. This creative tension is what gives life and motion." [3] Dialectics was derived from the works of philosophers G. W. F. Hegel (1831) and Heraclitus (500 BC), who thought that everything was constantly changing and that all things consisted of two opposite elements that could change into each other. Ancient Chinese philosophers also utilized "Yin" and "Yang" as two basic polarities of the universe to interpret the binary cooperative complementary phenomenon in nature and the universe. However, Engels simply thought the idea of "Yin" and "Yang" was just an embryo of dialectics in ancient China. However, Chinese philosophers had already studied the evolution process and unity of two opposite elements quantitatively. For example, "I Ching" (1000–750 BC), an ancient Chinese book of changes, stated that 64 Yin-Yang combinations known as "64-gua" are possible with hexagrams (patterns of 6 broken and unbroken lines) [4]. This "64-gua" is remarkably consistent with 64 combined results of DNA genetic codons, which consist of 3 nucleobases from adenine (A), thymine (T), cytosine (C), and guanine (G) in a group and up to 4^3 or 64 amino acids can be encoded. Based on the complementary base pairing principle, genetic codons can be translated into proteins (amino acid sequences) by living cells. An extraordinary Chinese philosopher Laozi (600–400 BC) also pointed out, "Everything is formed by two opposing elements" (Fig. 2). In his *Daodejing*, he fully illustrated the interplay of "Yin" and "Yang" and proposed the principle of learning from nature, which now plays an important and profound role in modern science. As the essence of Chinese philosophy, "Yin" and "Yang" has already been proved in a wide variety of fields ranging from science and technology to politics and economy in the history of nature and human society.

In the development of materials science, the binary cooperative complementary principle has already been widely

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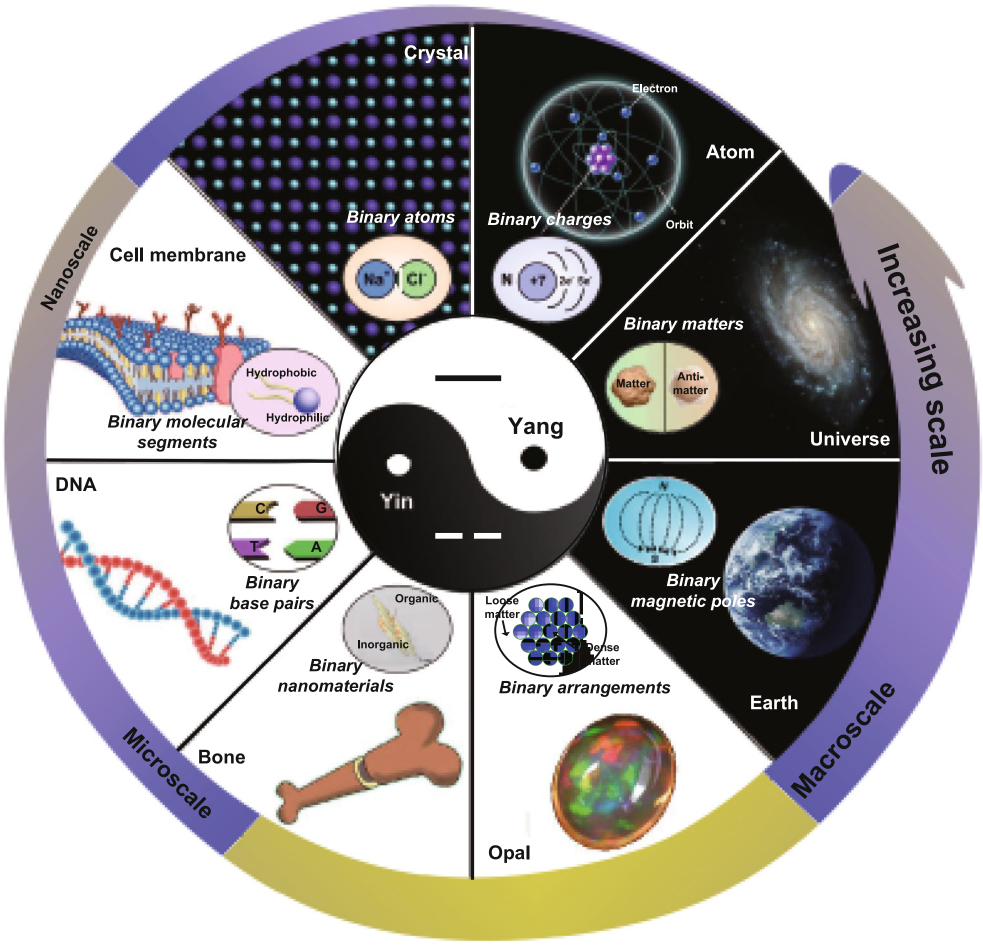


Figure 1 Binary cooperative complementary phenomena may be observed at many different levels of nature. Reprinted with permission from Ref. [2], Copyright 2014, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

Figure 2 Images of ancient Chinese philosopher Laozi, Ban Gu, and German philosopher Frederick Engels.

utilized to design and create novel functional materials. To date, various types of binary atomic materials and binary molecular materials have been successfully developed and have shown tremendous potential in both basic research and practical applications, as shown in Fig. 3. For example, in DNA, two long polymer chains bearing four kinds of

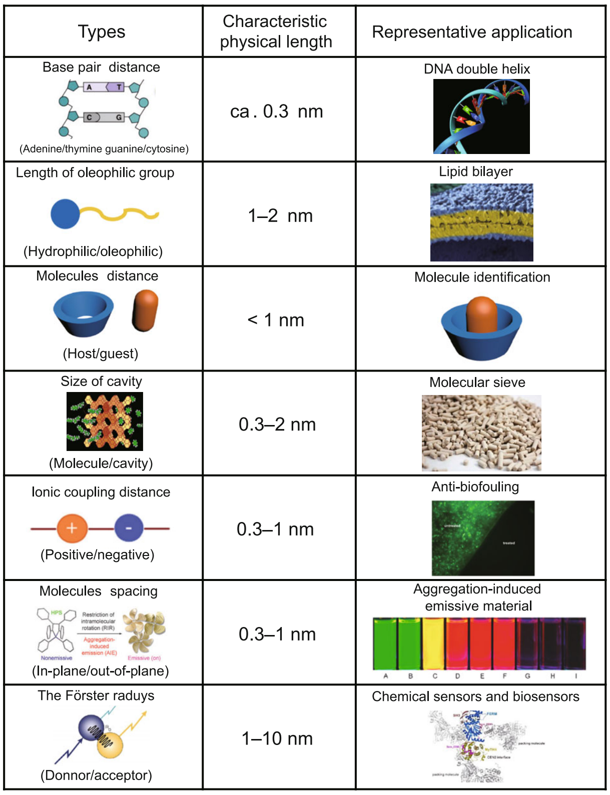
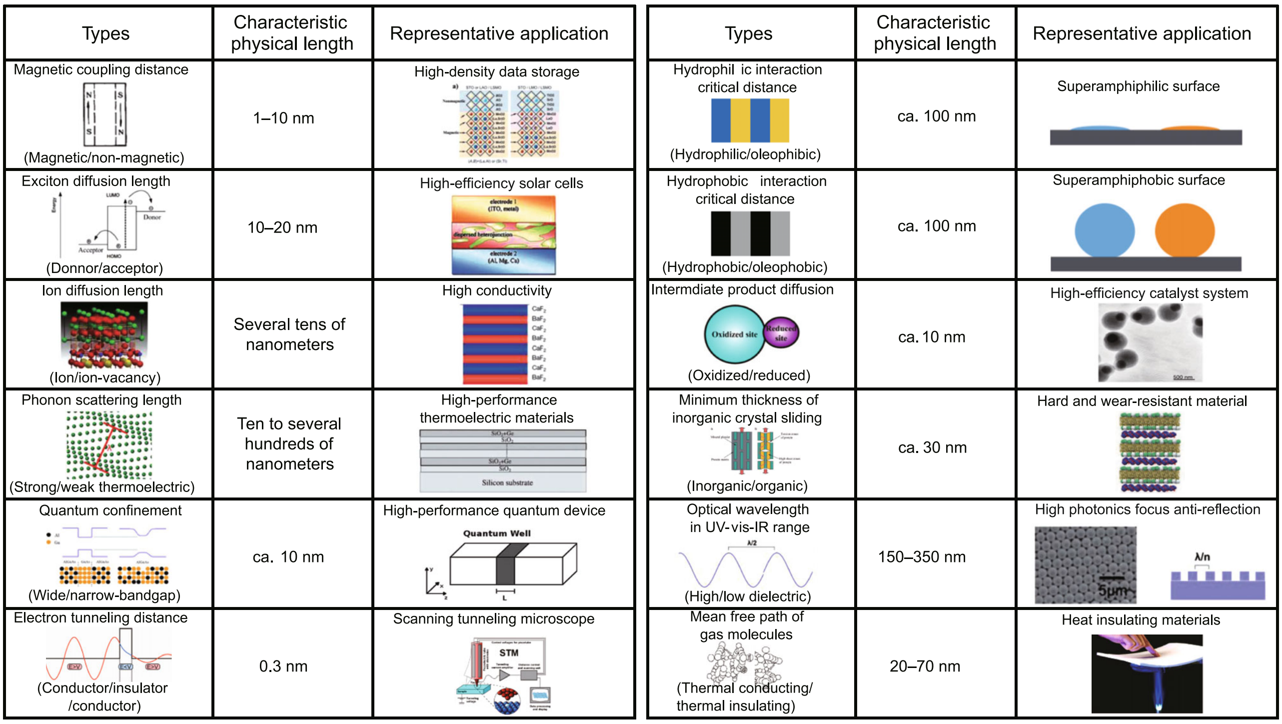


Figure 3 Examples of the binary cooperative complementary phenomenon in molecular design.

nucleobases form double-helix structures through hydrogen-bonding interactions between a pair of nucleobases (A to T and C to G) at a distance of approximately 0.3 nm [5]. Amphiphilic molecules, also known as surfactants, have both hydrophilic and oleophilic groups in the same molecular structure [6]. They can assemble into a stable lipid bilayer when the length of the oleophilic groups is approximately 1–2 nm. Host molecules combining with guest molecules with an appropriate spacing (< 1 nm) by the intermolecular force have been widely used in molecule identification [7]. A molecular sieve with a cavity size of approximately 0.3–2 nm can be used to separate different molecules with different sizes [8]. Zwitterionic molecules with both positive and negative groups in the same molecular structure at a distance approximately 0.3–1 nm (ionic coupling distance) are strongly hydrated through ionic solvation and thus create a strong repulsive force on protein. Such molecules can be used to prepare non-fouling materials [9,10]. When the conjugate molecules aggregate at a certain distance (< 1 nm) that restricts intramolecular rotation, the aggregation-induced emission (AIE) phenomenon can be observed [11,12]. When a donor molecule in an excited state approaches an acceptor molecule at a distance equal to the Förster radius (1–10 nm), energy transfer may occur through non-radiative dipole–dipole coupling [13]. This energy transfer has been widely used in the design of chemical sensors and biosensors.

The binary cooperative complementary principle has also seen significant success in the field of superwettability. By learning from the natural surfaces with special wettability, we recognized that the cooperative effect of surface micro/nano structures and surface energy is key in superwettability [14–19]. A surface roughness that matches the characteristic length of hydrophobic interaction of ~100 nm [20] can enhance the hydrophilicity of a substrate with water contact angles (CAs) less than 65° and enhance the hydrophobicity of a substrate with water CAs greater than 65° [21]. Following this principle, interfacial materials with extreme wetting states, such as superhydrophobic, superhydrophilic, superoleophobic, and superoleophilic states, can be fabricated from metals, polymers, ceramics, semiconductors, insulator, and so on. By combining micro/nanostructured substrates and responsive molecules, we can also fabricate smart interfacial materials possessing two complementary properties of superwettability that can be switched using light [22–25], pH [26–31], electric, chemical, or multi-stimuli [32–34]. If the design principle is transferred from an air environment to water or other liquid media, underwater superoleophobic and superareophobic surfaces can be developed [35–42]. Furthermore, novel functional solid/liquid interfacial systems, including three-dimensional integrated materials, two-dimensional membranes, one-dimensional fibers/channels, and zero-dimensional nanoparticles, can be generated and integrated into devices by combining different super-wettability properties [43,44]. These systems could be used for oil/water separation, electrochemical catalysis and energy conversion, water collection, control of cell adhesion, and robust surfaces in cosmetics as well as industrial applications [45–52].

In order to gain a deeper understanding of the binary cooperative complementary principle, we present more examples from recent achievements in materials science in Fig. 4. Magnetic/nonmagnetic alternative multilayered nanostructures will lead to remanence enhancement or magnetism-responsive properties. For example, in a magnetic tunnel junction consisting of two ferromagnetic layers isolated by a thin insulator layer (typically a few nanometers in thickness), electrons can be allowed to or prevented from tunneling through the insulating layer by tailoring the orientation of the magnetization, yielding two



Types	Characteristic physical length	Representative application	Types	Characteristic physical length	Representative application
Magnetic coupling distance (Magnetic/non-magnetic)	1–10 nm	High-density data storage	Hydrophil ic interaction critical distance (Hydrophilic/oleophibic)	ca. 100 nm	Superamphiphilic surface
Exciton diffusion length (Donnor/acceptor)	10–20 nm	High-efficiency solar cells	Hydrophobic interaction critical distance (Hydrophobic/oleophobic)	ca. 100 nm	Superamphiphobic surface
Ion diffusion length (Ion/ion-vacancy)	Several tens of nanometers	High conductivity	Intermdiate product diffusion (Oxidized/reduced)	ca. 10 nm	High-efficiency catalyst system
Phonon scattering length (Strong/weak thermoelectric)	Ten to several hundreds of nanometers	High-performance thermoelectric materials	Minimum thickness of inorganic crystal sliding (Inorganic/organic)	ca. 30 nm	Hard and wear-resistant material
Quantum confinement (Wide/narrow-bandgap)	ca. 10 nm	High-performance quantum device	Optical wavelength in UV-vis-IR range (High/low dielectric)	150–350 nm	High photonics focus anti-reflection
Electron tunneling distance (Conductor/insulator /conductor)	0.3 nm	Scanning tunneling microscope	Mean free path of gas molecules (Thermal conducting/ thermal insulating)	20–70 nm	Heat insulating materials

Figure 4 Examples of the binary cooperative complementary phenomenon in materials science.

states of electrical resistance. This mechanism has already been used in the read-heads of modern hard-disk drives [53]. When the electron donors and acceptors are combined at the nanoscale and the distance between the two components is 10–20 nm (the exciton diffusion length), high-performance photovoltaic devices can be achieved [54]. To facilitate the ion diffusion, nanoscale ion/ion-vacancy binary building blocks need to be assembled with a distance of tens of nanometers, which should match the ion diffusion length in the solid [55,56]. Examples of this type of materials are alternative CaF_2/BaF_2 layers [57], Y_2O_3-ZrO_2/$SrTiO_3$ hybrids, and Ag_2S nanoparticle-GeS_2 mixture [58–60]. Thermoelectric efficiency can be greatly enhanced if strong and weak thermo-electric building blocks hybrid in nanoscale adopt alternative layered structures, in which the thickness of each layer should match the phonon scattering length. Quantum wells (2D) can be fabricated by integrating wide/narrow-bandgap semiconductors, in which the cooperative distance is the average electron free path [61]. A superconducting tunnel junction (Josephson junction) consists of a superconductor and insulator at a distance of 2–5 nm, which is the coupling length of Cooper pairs [62]. Scanning tunneling microscopy and surface-enhanced Raman spectroscopy were designed based on the electron tunneling distance (0.3 nm) between conductor/insulator/conductor [63,64]. A high-efficiency catalyst system can be developed if the distance between the oxidized site and reduced site is comparable with the diffusion radius of the intermediate product. Biological tissues, such as teeth and bone, utilize organic/inorganic hybrid nanostructures to enhance their mechanical strength. The minimum thickness of inorganic crystal sliding domains is approximately 30 nm [65]. Periodically arranged nanoarchitectures of high/low dielectric constant, such as photonic crystals or plasmonic metamaterials, can guide photon directional transport and manipulate light at the nanoscale. In this case, the distance between high/low dielectric building blocks should match the wavelength of light [66]. The aerogel shows high thermal insulation performance if the average pore size is comparable with the mean free path of gas molecules (20–70 nm) [67]. By introducing thermoresponsive segments into the hydrophilic network, nonswellable hydrogel can be prepared [68].

The binary cooperative complementary principle has also been applied to design "multiferroic" materials (Fig. 5). For example, in order to enhance magnetoelectric effects, a promising strategy is to introduce strain coupling between two materials such as a ferromagent and a ferroelectric [69]. When a piezomagnetic (or magnetostrictive)

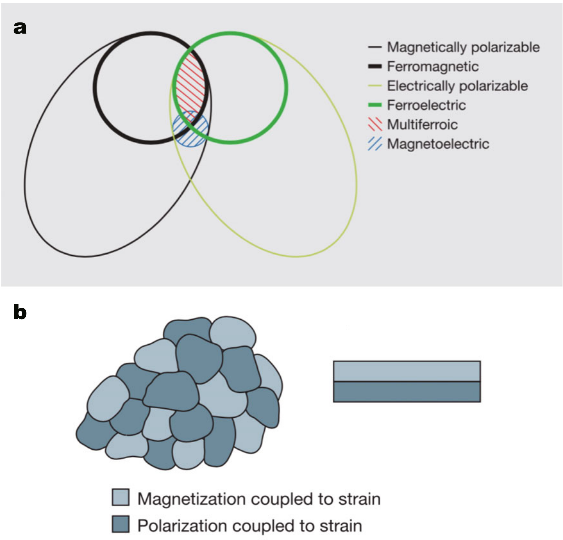


Figure 5 Example of the multiple cooperative complementary effect. (a) The relationship between multiferroic and magnetoelectric materials. (b) Strain-mediated magnetoelectric coupling in two-phase systems. Suitable structures include mixtures of grains and thin-film heterostructures. Reproduced with permission from Ref. [69], Copyright 2006, Nature Publishing Group.

material and piezoelectric (or electrostrictive) material come into intimate contact with each other to form composites, laminates, or epitaxial multilayers, strain coupling can be realized. The coupling constant depends on the frequency of the a.c. applied magnetic field. Such multiferroicstructures could be applied in microwave-frequency transducers.

As the most general laws in the historical development of nature and human society, "*Dialectics in nature*" can be reduced at a fundamental level to three laws: 1) the law of transformation of quantity into quality and vice versa; 2) the law of interpenetration of opposites; and 3) the law of negation of the negation. Furthermore, by summarizing the recent achievements in materials science, the law of cooperative interaction distance might be considered the fourth law of "*Dialectics in nature*". The cooperative interaction distance between a pair of complementary properties must be comparable with the scale of related physical or chemical parameters.

In 2000, we extended this principle from the molecular level to the nanostructural level and developed a new material system, i.e., binary cooperative complementary interfacial materials (BCCIMs) [1]. We expected that unique macroscopic functional properties could be generated when two components with entirely opposite properties were introduced at the nanoscale. The design principle and assembly principle of BCCIMs are shown in Fig. 6. These opposite properties could be hydrophilic and oleophilic, oxidizing and reducing, thermally conducting and thermally insulating, ferromagnetic and antiferromagnetic, p-type and n-type, positive swelling and negative swelling, high dielectric constant and low dielectric constant, strong thermoelectric and weak thermoelectric, convex and concave, ion and ion vacancy, electron donor and electron acceptor, and so on. The key parameter for designing BCCIMs is the cooperative distance Δ between the pair of complementary components. Numerous achievements indicate that the cooperative distance Δ must be comparable with the scale of the related physical or chemical parameter. When the binary components are separated by the cooperative distance, the cooperation between these building blocks becomes dominant, endowing the macroscopic materials with unique properties and

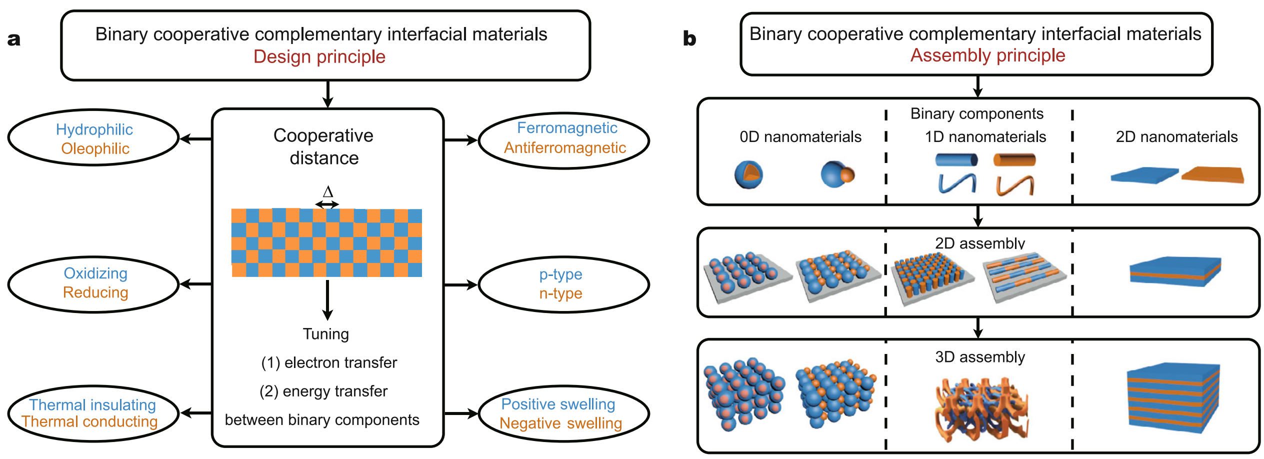


Figure 6 Design principle and assembly principle of binary cooperative complementary interfacial materials.

advanced functionalities that cannot be achieved individually by either of building blocks. Reversible switching between two entirely opposite macroscopic properties can also be realized on the same interfacial materials. By following the design principle and assembly principle in Fig. 6, we expect that the combination of hydrophilic/oleophilic polymers in 3D cross-linked networks can yield high-performance organohydrogels. Alternative layered structures of thermally conducting/thermally insulating materials can lead to anisotropic thermally insulating materials. A combination of positive-swelling and negative-swelling materials can result in wide-temperature-range non-swelling materials. By sandwiching a photo-conductive organic layer between two inorganic magnetic layers, photo-controlled giant magnetoresistance devices can be fabricated and developed. Considering that so many opposing physical properties exist in the universe, the binary cooperative complementary principle will no doubt lead to the generation of numerous possibilities to construct new functional materials.

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Author contributions Jiang L proposed the concept of Binary cooperative complementary materials. Liu M and Jiang L wrote the paper and contributed to the general discussion.

Conflict of interest The authors declare that they have no conflict of interest.

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Lei Jiang received his BSc degree in solid state physics (1987), and MSc degree in physical chemistry (1990) from Jilin University in China. From 1992 to 1994, he studied at the University of Tokyo in Japan as a China-Japan joint course PhD student and received his PhD degree from Jilin University of China with Prof. Tiejin Li. Then, he worked as a postdoctoral fellow in Prof. Akira Fujishima's group at the University of Tokyo. In 1996, he worked as researcher in Kanagawa Academy of Sciences and Technology, Prof. Hashimoto's project. In 1999, he joined the Institute of Chemistry, CAS. In 2015, he moved to the Technoligical Institute of Physics and Chemistry, CAS. Since 2008, he also served as the dean of School of Chemistry and Environment in Beihang University. He was elected as members of the Chinese Academy of Sciences and The World Academy of Sciences in 2009 and 2012. In 2016, he was also elected as a foreign member of the US National Academy of Engineering. His research is focused on bioinspired, smart, multi-scale surfaces with super-wettability and his publications have been cited more than 36000 times with an H-index of 89.

材料科学中的自然辩证法: 二元协同材料

刘明杰[1,2], 江雷[1,3*]

摘要 “二元协同材料”这一新概念,不同于传统的单一体相材料,是在材料的宏观表面或体相内建造二元协同纳米界面结构. 该材料设计原理是,在介观尺度引入不同甚至完全相反理化性质的纳米微区,在某种条件下具有协同的相互作用,以致在宏观上呈现出超常规物性的材料. 这一新原理的关键是找出这两种组分间的协同距离,该协同距离应该与物理或化学中的某一特征常数相关. 这一设计原理可以拓展到材料科学的多个领域,用于指导制备各种新型功能材料.