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Study on Ion Bombardment Semiconductor Shaped Self-Assembled Nanostructures

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Abstract: *Through Surface nanopatterning induced by ion beam bombardment has become an effective nanostructure technology, in a short period, large-area patterning can be induced on the surface of various materials through ion bombardment technology, and island-like, porous, and corrugated morphology can be formed on the surface of the material, effectively improving its surface morphology. In this review, we provide an updated description of the progress made when ions bombard semiconductor surfaces, focusing on the influence of different parameters on the evolution of surface morphology. Secondly, the physical model of the evolution of surface nanostructures and the application of nanostructures generated by low-energy ion bombardment are summarized.*

Keywords: *Self-assembling nanostructures, Ion bombardment, semiconductor*

1. Introduction

In 1962, Navez et al[1]. reported that nano-ripple patterns were spontaneously formed on a glass substrate under the irradiation of an ionized air beam. After decades of development, different research directions have been derived, including the study of the formation mechanism of this self-organized nanostructure, the study of the nanostructure characteristics of the surface of different materials, and its application in optics, biology, and other fields.

Self-organized nanostructures induced by ion bombardment are not limited by the type of materials and can be formed on a variety of materials such as metals[2, 3], oxides[4, 5], semiconductors [6-13], polymers[14-16], etc. Various self-organized nanostructures are induced on the surface, such as nanodots [17-19], nanopores[20], and nanoripples[21, 22]. Moreover, low-energy ion bombardment can prepare large-area (cm^2 level) nanostructures without a mask, which has the advantages of low cost, high efficiency, and wide application range, and is a very potential preparation of surface.

To simplify the problem, the early studies usually used inorganic simple materials for bombardment experiments of pure ion beams, such as Si, Ge, Au, Fe, etc., and obtained quasi-periodic nano-ripple structures at oblique incidence, and analyzed the physical mechanism and evolution law [23-26]. With the deepening of research, the ion bombardment characteristics of SiO_2, GaSb, InP, and other binary inorganic compound materials have also been fully studied. The InP surface obtained the nanodot structure, in addition to forming nano-ripple structures under oblique incidence, normal-incidence ion bombardment also obtained nano-dot structures on GaSb and InP surfaces.

This paper first briefly introduces the continuum model of the evolution of surface nanostructures induced by ion bombardment and then introduces the experimental studies of surface nanostructures with different parameters, including pure ion beam bombardment of inorganic elemental materials, ion bombardment of impurity co-deposited inorganic elemental materials, and Pure ion beam bombardment of binary inorganic compounds causes the evolution of surface nanostructures. On the other hand, some applications of self-assembled nanostructures in magnetism, plasmonics, sensors, and other fields are summarized.

2. Continuum model

The generation of surface nanopatterns by ion bombardment dates back to the 1980s, and two pioneering contributions made this field popular. First in 1988, Bradley and Harper[27] proposed a continuous model Bradley and Harper (B-H) model for explaining ion bombardment-induced self-

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organized nanostructures, which established a theoretical framework for the formation of surface nanopatterns by ion bombardment. The model is based on the Sigmund projection theory, pointing out that the generation of surface self-organized nanostructures is the result of the joint action of surface growth caused by curvature-dependent sputtering and surface smoothness caused by surface thermal diffusion.

According to the Sigmund sputtering theory, the sputtering rate at a certain point on the material surface is proportional to the total energy deposited at that position. In the case of elastic collisions, the deposition energy distribution of incident ions on the solid surface is similar to a Gaussian distribution centered on its resting position. Taking into account the influence of the local surface topography on the deposition energy distribution, the sputtering rate at any point on the material surface can be determined more accurately. The study found that the total energy deposited at the concave position of the material surface is greater than the total energy deposited at the convex position of the surface, that is, the rate of incident ions eroding valleys is greater than that of eroding ridges.

Through continuous development and improvement, the B-H model can well explain the evolution of the surface morphology of inorganic elemental materials under oblique incidence conditions. For binary inorganic compound materials, due to the different masses and binding forces of different atoms, their sputtering yields are also different. Usually, atoms with lighter weight and less binding force are more likely to be sputtered out, that is, preferential sputtering radiation effect. Under the action of continuous ion bombardment, the surface composition of the compound material will change, and this composition change also has an important impact on the evolution of the surface nano-topography.

Since the B-H model is a linear continuous model, it cannot explain some other nonlinear characteristics of self-organized nanostructures. For example, it cannot predict the saturation of the ripple amplitude, surface dynamics coarsening, etc[28]. The anisotropic Kuramoto-Sivashinsky (K-S) model improves the B-H model, which has characteristics such as instability and nonlinearity[29]. The phenomenon of amplitude saturation and corrugation coarsening in the late stage of morphology evolution is explained.

The contribution of surface local curvature and slope-related sputtering effects, surface material etching, surface thermal diffusion and ion bombardment-induced surface diffusion, and quasilinear and nonlinear contributions to the formation and evolution of surface nanostructures is reflected in the K-S model, Saturation of ripple amplitude and formation of facet structure under prolonged ion bombardment[30]. And the nanodot and nanopore structures induced by ion bombardment under normal incidence are well explained.

In 2011, Norris et al. [31]proposed a Crater Function model which is based on the mass redistribution effect and sputtering effect during ion bombardment, predicting the formation of large-area graphics. And it is in good agreement with the related experimental results. The Crater Function model uses a multi-scale framework to expand the crater function representing the average change of local surface height into a continuous partial differential equation of surface evolution[32]. In this model, linear stability is strictly determined by the sign of the calculated coefficients, and only when these coefficients are negative, there is a linear instability mode, capable of producing a graph; while when these coefficients are positive, then a stable smooth surface is formed, and their signs are determined by the incident angle of ions, so the generation of surface self-organized nanostructures is related to the incident angle of ions.

The Crater Function model successfully predicts the variation of self-organized nanostructures with ion incident angle and theoretically proves the contribution of mass redistribution effect during ion bombardment in the formation of self-organized nanostructures. The physical mechanism of ion bombardment to produce self-assembled nanostructures mainly includes the sputtering effect, surface diffusion effect, mass redistribution effect, and stress-relaxation mechanism. The propagation of ripples on the surface of self-assembled nanostructures is mainly caused by the stress-relaxation mechanism.

Although ion bombardment produces the self-assembled nanostructure mathematical model and the research on the physical mechanism has made the above progress, a comprehensive theoretical understanding of IBI nanopatterning has not yet been obtained, which undoubtedly indicates that patterning is not a universal process because it depends on the combination of specific target material and bombarding ions, and the surface morphology depends on the setting of experimental parameters. Therefore, surface morphology evolution caused by ion bombardment is a problem worthy of continuous exploration.

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3. Parameter influence of ion bombardment to produce nanostructures

There are many factors affecting the self-assembled nanostructures produced by ion bombardment. The selection of incident particle source and target material is very important. According to whether impurity atoms are introduced during ion bombardment, it can be divided into pure ion beam bombardment and impurity co-deposition ion bombardment. According to the composition of elements in semiconductor materials, it can be divided into ion bombardment of single substance materials and ion bombardment of binary compound materials. This section focuses on exploring the effects of different parameters on the evolution of nanostructures on the surface of pure ion beam bombardment of simple material.

3.1. Elementary material is bombarded by pure ions beam

Most early theoretical modeling and experimental work involved single-element targets surface bombarded by noble gases ion beams. In this case, inert ions are implanted and eventually evaporate as the bombardment proceeds. Such targets are easier to study because they become amorphous under low-energy ion bombardment, thus avoiding the so-called Ehrlich– Schwoebel (ES) barriers cause surface diffusion instability. The typical research object on the surface of a single semiconductor is Si, and similar results have been obtained in many other types of research on single semiconductor materials. This subsection will mainly discuss experiments using Ar^+ ions as ion beams, and similar results can be obtained for heavier noble gases.

3.1.1. Influence of incident angle on pure ion bombardment of simple material

Different incident angles have different effects on the evolution of nanostructures on the surface of the Si substrate. As the incident angle increases, the patterns on the substrate surface continue to evolve. In 2011, Madi et al. [33] changed the incident angle of the Ar^+ ion beam and the ion bombardment energy, and found that when the ion incident angle is less than the critical angle of 48°, the Si surface is smooth, and when the ion incident angle is in the range of 48° to 80°, will produce parallel mode ripples (the wave vector of the ripples is parallel to the projection direction of the ion beam); and when the ion incident angle is 85°, it will produce vertical mode ripples (the wave vector of the ripples is perpendicular to the projection direction of the ion beam), The findings are in good agreement with those predicted by the C-F model.

Not only does the surface nano topography change with the incident angle but also the wavelength and amplitude of the ripples change with the incident angle. Just as Sandeep Kumar Garg et al. [34] bombarded the Si(100) surface with a 60keV Ar ion beam, the study found that the corrugated morphology began to appear at an incident angle of 45° and became more prominent at higher incident angles. Atomic Force Microscope (AFM) studies have shown that although the ripple wavelength decreases with the increase of the incident angle, the amplitude increases, it is further found that the evolution of the surface topography is the result of the joint action of sputter erosion and ion-induced atomic redistribution.

3.1.2. Influence of ion beam current on pure ion bombardment of simple material

It is not only the incident angle of ions that affects the nanostructure of the substrate surface, but the size of the ion current beam also has a crucial influence on the evolution of the nanostructure of the surface. The research group of Professor Lu Ming of Fudan University [35] used a combination of simulation and experiment methods to bombard the Si surface with an Ar^+ ion beam at a normal incident and studied the effect of the ion flux (Ion flux) on the formation of nanostructures on the Si surface. The results show that the surface roughness, the diameter of the surface nanodots, and the sample temperature are all affected by the ion beam parameters. It is also pointed out that self-assembled nanostructures appear on the surface only when the ion beam current is greater than a certain threshold (the threshold in this experiment is 220nA/cm^2), and the nanodots diameter and surface roughness of Si surface decay exponentially with the increase of ion flux.

Professor Liu Weiguo and Dr. Chen Zhizhi from Xi'an Technological University studied the influence and regulation of different parameters on the self-organized nanostructure of single-crystal silicon. The research results show that the characteristic wavelength of the corrugation is not affected by the ion beam current; the characteristic wavelength of the corrugation increases with the increase of ion energy. Through the influence of different parameters such as ion beam current and ion incident angle on the evolution of surface morphology in this section, the controllability of self-assembled nanostructures is verified, and it is further possible to manufacture defect-free large-area regular nanostructures. Its application in various fields has laid the foundation.

3.2. Impurity co-deposited ion bombardment of a pure element

It is difficult to obtain large-scale structures and faceted nanostructures (blazed grating-like structures with asymmetric triangular cross-sections) simply by relying on pure ion beam bombardment-induced nanostructures. This defect greatly limits the application of ion beam technology in the field of optics. Therefore, this subsection explores the evolution of nanostructures on the surface of single-element targets (silicon) co-deposited with anisotropic or isotropic metals under inert gas irradiation depending on the experimental setup.

3.2.1. Co-deposition of anisotropic impurity ion

As in the experimental setup designed by Höfsassetal et al., when the impurity flux is directional, under the modulation of anisotropic metal impurities, a corrugated structure appears on the substrate surface. In this type of setup, both the silicon target and the inclined steel plate are simultaneously irradiated by a vertical ion beam, the sputtered atoms from the inclined steel plate fall on the bombarded silicon surface, and the impurity ion flux follows along the silicon target decreases as the distance increases to the plates. The research team used this device to explore the evolution of self-assembled nanostructures bombarded by anisotropic impurities co-deposited ions on the surface of the substrate. By changing the incident angle of the ion beam and the inclination angle of the steel plate, the influence of impurity ions on the self-assembled nanostructures was observed.

Hofsäss et al.[36] used Xe^+ with an ion energy of 5 keV as the incident ions to bombard the Si target, at an incident angle of 70°, relative to the surface of the silicon substrate. The impurity co-deposition ions were selected from Si and Au. The use of Si as co-deposited atoms was found to have no effect on the evolution of the surface morphology. However, Au atoms can significantly improve the wavelength and amplitude of the ripples, and the surface roughness decreases and gradually flattens out as it gets farther and farther away from the Au source location. No nanopattern is formed on the surface at the location closest to the Au source (no impurity atoms are sputtered onto the substrate at the location closest to the Au source), thus showing that Au acts as a surfactant during the ion bombardment.

Using the same experimental setup, Macko et al [37] further investigated the impurity-induced morphological evolution of the substrate concerning the distance from the steel plate. It was found that as the distance increases, the surface changes from a plane with nanopores to a ripple with a wave vector parallel to the direction of ion beam projection. As the distance increases, combined ripples and dot patterns appear, and for greater distances, the substrate surface pattern becomes dotted. Finally, for large enough distances, the surface remains flat. When the flux of impurity ions is fixed, the effect of the incident ion flux on the evolution of the surface morphology is a question worth investigating. Zhang et al [38] bombarded Si with a constant ion flux of Fe impurities and found that the pattern varied with the incident ion flux for sufficiently high Fe content while evolving from a weakly mixed dotted ripple pattern to a well-defined ripple pattern, where the ripple increases its length and amplitude, and the pattern becomes more regular. The location of the impurity source also affects the surface nanostructure evolution when the impurity flux is certain. k. Zhang et al. obtained surface nanostructures with different morphologies and symmetries on the Si surface by placing Fe impurity targets at different locations around the Si substrate during ion bombardment. The results show that the deposited Fe has a significant modulating effect on the morphological characteristics of the Si surface nanostructures.

Macko et al [37]found that patterns can be formed on the target surface only when the angle between the incident ions and the impurity ions is large enough, and the ripple vector is not along the direction of the projection of the incident ion beam, but along the direction of the projection of the incident impurity flux. Therefore, the introduction of impurity co-deposition during ion bombardment enriches the types of self-organized nanostructures and also enhances the ability to modulate the nanostructure morphology and feature size.

3.2.2. Co-deposition of isotropic impurity ions

Stable nanodot patterns can be obtained when the metal flux is isotropic and the ion beam is incident normal to the target surface. The most systematic and controllable method to induce nanodot patterns is to use a mask with a circular hole in the center, which is placed on top of the silicon target for irradiation normalization. In this way, iron atoms are ejected from the mask and land isotropically in the center of the target surface, with the metal content increasing first from the periphery and then decreasing slightly near the center. As Ozaydin et al [39] used positive incidence Ar^+ bombardment of the Si substrate,

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providing a small amount of each homogeneous Mo atom, it was found that a nanopore-like pattern is produced very close to the edge of the mask, where the metal content is small, and when moving towards the center, a ripple pattern is found, and to the right of the positive center position, a clear dot pattern is observed. Stress measurements were also performed on bombarded samples that initially showed compressive stresses that eventually transformed into greater tensile stresses upon further irradiation, and it is believed that stress plays an important role in nanodot patterning.

This mask device allows further investigation of the effect of different parameters on the patterning properties. J A Sánchez-García et al [19] used a positive incidence of 1 keV Ar ions on the Si (001) surface to selectively generate self-organized nanopore and nanodot patterns. For a fixed ion flux, relatively low ion current densities induce nanopore patterns that evolve into nanodot patterns at higher current densities. Muñoz-García J et al [23] found that the ripple wavelength increased with the bombardment time and gradually reached saturation. M. Cornejo et al [40] studied the effect of simultaneous doping of Fe on Si(001) by low-energy ion beam sputtering in self-organized pattern formation, where the doping of Fe affects the evolution of the topography and is necessary for the formation of ripples close to normal incidence, where Fe is not uniformly distributed on the surface, but the concentration at the crest is higher than at the trough.

As explored by Redondo-Cubero A et al[41], these length scales also vary with the energy of the incident ion, with much larger characteristic wavelengths observed for Xe ions at 20 keV than when using Ar ion beams at 1.2 keV. In addition, the fact that nanostructures with higher aspect ratios can be produced using medium energy ion bombardment in co-deposition devices can be used for different applications[42].

The diameter of the mask plate also affects the substrate surface morphology, and the different co-deposited metals affect the pattern characteristics. As Gago R et al [43] found that by varying the surface diameter, both the wavelength and surface roughness of the pattern changed, and varying the co-deposited metal type found that the Mo mask template modulated a neater and more ordered pattern than the Fe mask template.

3.3. Binary inorganic compounds are bombarded by ions beam

When ions bombard a compound, chemical heterogeneity in the target atomic species needs to be taken into account, which can lead to different erosion rates as well as different surface diffusivity. (a) Due to the different sputtering rates and the different ion-induced transport processes, kinetic phase separation occurs at the most surface of the target, causing elements with higher sputtering yields to be preferentially located at the pattern peaks; For the evolution of nanostructures, this subsection focuses on III-V semiconductors (typical GaSb) as well as IV-IV semiconductors (typical SiC) to investigate the effect of different parameters on ion bombardment of binary inorganic compounds. Allmers T et al [44] investigated the transition from point to ripple on the GaSb surface during the bombardment in relation to the ion incidence angle. Point structures were formed at incidence angles less than 10°, where small amounts of impurities were observed to act as nucleation centers for forming nanostructures of different shapes. Corrugated structures started to form at incidence angles greater than 20°.

3.3.1. Ion bombardment of GaSb

For GaSb has been explored by many groups, Teichert C et al [45]studied the size distribution and dot shape of ion bombardment-induced GaSb quantum dots (QD) with sputtering time by high-resolution atomic force microscopy (AFM). The results show that the particle size distribution range widens when the sputtering time is less than 150 s; it starts to decrease when it exceeds 150 s and then remains stable for a long period. At the same time, GaSb quantum dots transform from partial domes to cones in shape and eventually develop into complete domed structures.

Oluwole E Oyewande et al [46] investigated the effects of different ion energies, different incidence angles, and different ions on the sputtering rate of GaSb surfaces based on Monte Carlo simulations. It was found that the maximum sputtering yield does not occur at a specific angle, but in the range of 65° and 85° (for 1 keV ion energy) and between 75° and 85° (for 10 keV ion energy). Moreover, the sputtering yield increases with increasing energy, which indicates that the ion energy is directly related to sputtering, and Kr+ leads to the lowest sputtering yield at 10 keV. Therefore, when one is interested in high sputtering parameters for the binary compounds AlSb, GaSb, and InSb, one should use high-energy heavy rare gas ions. Thomas Bobek et al [47] have discussed methods for transferring dot patterns created on semiconductor-forming layers to buried metal films. Self-organized nanodot patterns formed on the overlying GaSb film can be successfully transferred to the embedded Co layer during vertically incident ion erosion, resulting in short-range hexagonally ordered arrays of nanoparticles, the size of which can be controlled by adjusting the ion energy. M A. Lively et al [48] used 500 eV Kr^+ ions incident on GaSb semiconductor surfaces to elucidate the nature of the coupled compositional and morphological pattern formation mechanisms. It was found that the altered composition depth distribution is critical to induce surface morphology changes and that the morphological evolution of the surface follows the nucleation and kinetic growth equations.

3.3.2. Ion bombardment of SiC

The degradation of self-assembled nanostructures formed by ion bombardment of surfaces with time is a problem that limits the application of ion bombardment technology. With the rapid development of nanotechnology, some nanomaterials have been found to have unique properties such as excellent creep resistance, fatigue resistance, irradiation resistance, and corrosion resistance. SiC is rapidly becoming a hot spot for research due to its superior mechanical properties, chemical stability, high-temperature resistance, electrical properties, and other unique properties.

Y.S. Katharria et al [49] studied the bombardment of 6H-SiC surfaces with 100 keV Ar+ oblique incidences (angle of incidence $\theta = 60°$). The topographic changes induced by ion beam bombardment were observed using scanning electron microscopy, and the results showed that the surface morphology consists mainly of nano-islands with an average diameter of 30 nm for a short period, followed by the appearance of highly modulated nano-ripples with a period. Corrugated structures developed on physically stable materials (e.g., SiC) will exhibit very little time degradation compared to corrugated structures grown on conventional semiconductors (e.g., Si, GaAs, InP, etc.), and corrugated structures are more favorable for various technical applications.

Jiaming Zhang et al [50] formed well-aligned corrugated structures on the surface by focused ion beam bombardment of 3C-SiC single crystal films and characterized the resulting morphology using scanning electron microscopy as well as atomic force microscopy. The characteristic wavelength of the corrugated structures formed after ion sputtering beyond a critical incidence angle of 50° varies from 158 to 296 nm with the incidence angle and ion beam flux. By varying the ion beam incidence angle and ion beam flux, the geometry, orderliness, and uniformity of the ripples can be well controlled, laying the foundation for the use of SiC nanostructures in optics and electricity.

4. Application of ion bombardment to produce nanostructures

Among the main features of IBI nanopatterning, the technology allows patterning larger areas, shorter processing times, and cost-effective production, and the technology has been applied in many fields such as magnetics, plasmonics, nanoelectronics, optoelectronics, biomaterials, and sensors in the last few years.

Teichert.C. et al [51] proposed the application of low-energy ion sputtering in magnetics. Patterns are formed spontaneously during the epitaxial growth or ion etching of semiconductor wafers. By tuning the film thickness and growing superlattices, various patterns with different symmetries can be obtained, and since these arrays of self-organized nanostructures cover the entire wafer on which they are grown, they can be used as large-area nanopatterned substrates for subsequent magnetic film deposition.

Thomas W. H. Oates et al [52] applied low-energy ion irradiation of nanostructures in plasma with low-energy ion bombardment in silicon corrugated templates, where physical vapor deposition of silver atoms preferentially forms nanoparticles in the troughs with adjustable periodicity, which is cost-effective for medium-scale nanostructures and suitable for large-scale production.

Nanostructures bombarded with low-energy ions have applications in nanoelectronics, optics, biomaterials, and sensing as follows. Xin Ou et al [53] fabricated horizontal single-crystal silicon nanowire arrays on insulating substrates by irradiating insulator silicon materials with Xe ion beams based on a pattern self-assembly formation mechanism. Daniele Chiappe et al [54] synthesized transparent metallic nanowire arrays using a method based on scattered focus ion beam sputtering. The nanostructured metal electrodes were used as an alternative to conventional transparent conducting oxides due to the reduced resistance of the thin layer compared to transparent conducting oxides and the support of local plasmon resonance. Matteo Barelli et al [55] demonstrated an innovative and fully self-organizing method based on wrinkle-assisted ion beam sputtering. It can be used to fabricate large-area (cm2 level) nanoribbon sodium-calcium templates supporting ultrathin Au films by physical deposition. The potential of such templates for a wide range of applications in optoelectronics was demonstrated, aiming at applications in plasma-enhanced photon harvesting for molecular or biological sensing.

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Self-organized nanostructures induced by low-energy ion bombardment can also be used as masks to transfer nanostructures to substrate surfaces by direct etching or other graphic transfer techniques. The Mongeot team in Italy has successfully transferred self-organized nanostructures from Au film surfaces to glass substrate surfaces using ion bombardment, and due to the different etching rates of glass and Au, the aspect ratio of nanostructures on glass surfaces is increased compared to nanostructures on Au surfaces Ion bombardment of surface nanopatterns have also been used to study radiation damage and repair of 2D materials, as well as by ion trapping to modify their properties.

5. Conclusion and Outlook

Ion bombardment is a versatile and effective technique that can change the surface properties of substrate materials in a controlled manner without changing the overall properties of the substrate material. Different parameters such as incident ion type, ion beam current, ion incidence angle, ion energy, bombardment time, impurity ion co-deposition, and substrate material properties have different effects on the surface morphology evolution. Based on the previous studies, it is possible to select the appropriate parameters to fabricate large-area regular nanostructures without defects. The application of ion bombardment to generate self-assembled nanostructures in various fields such as magnetism, plasma, nanoelectronics, optoelectronics, biomaterials, and sensors is further explored.

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