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Resonant mode of Fabry-Perot microcavity regulated by metal surface plasmons

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Abstract: At present, the use of zinc oxide(ZnO) micro-nanowire structures in ultraviolet laser devices with natural resonant cavities has attracted wide attention at home and abroad. Aiming at the problems of the luminescence and stability caused by ZnO intrinsic defects, research on the local field luminescence enhancement of plasmons is very important for the application of ZnO-based UV laser devices. In this paper, ZnO microwire structure model is constructed by theoretical simulation and the micro-cavity optical loss and Fabry-Perot (F-P) resonant cavity mode evolution are theoretically analyzed. The relationship between the diameter change of ZnO microcavity and the evolution of the F-P resonance mode, optical loss and light intensity distribution is obtained. On this basis, the six surfaces of ZnO microwires are modified by metal Ag nanoparticles. It is found that the resonance coupling effect of metal local surface plasmons significantly inhibited the loss of light around the microcavity and the local field enhancement is realized by the resonance coupling effect at the intersection of the metal and the microcavity. The simulation results show that after the surface of the microcavity is modified with Ag nanoparticles, the confinement ability of the optical field increased by 6.72%, while the secondary coupling occurs along the X axis between the metal particles, and the electric field intensity is enhanced by 2 times.

Key words: F-P resonator; metal local surface plasmons; optical loss; local field enhancement

金属等离子激元调控 Fabry-Perot 微腔谐振模式研究

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摘要:目前,利用氧化锌(ZnO)微纳米线结构形成具有自然谐振腔的紫外激光器件引起国内外广泛关注。针对 ZnO 本征缺陷导致器件发光及稳定性不足等问题,开展金属局域等离子激元局域场发光增强方面的研究,对 ZnO 基紫外激光器

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件的应用具有十分重要的意义。本文通过理论仿真构建氧化锌微米线结构模型,对微腔光学损耗及 Fabry-Perot(F-P)谐振腔模式演化进行了理论分析。得到 ZnO 微腔直径变化与 F-P 谐振模式演化、光学损耗和光强分布的关系。在此基础上通过金属 Ag 纳米颗粒对 ZnO 微米线 6 个表面进行修饰,发现金属局域表面等离子激元共振耦合效应对微腔周围的损耗光有明显的抑制作用,并且在金属与微腔的交叉区通过共振耦合效应实现局域场增强。模拟结果表明,在损耗较大的微腔表面修饰 Ag 纳米颗粒以后,光场限域能力提高 6.72%,而在金属颗粒之间沿 X 轴方向产生二次耦合现象,其电场强度更有 2 倍的增强效果。

关 键 词:F-P 谐振腔;金属局域表面等离子激元;光学损耗;局域场增强

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1 Introduction

As a representative third-generation wide bandgap semiconductor, ZnO has a band gap of 3.37 eV and an exciton binding energy of 60 meV at room temperature. Therefore, ZnO materials have attracted extensive attention in the development of high-quality optical microcavities and high-efficiency ultraviolet optoelectronic devices[1-2]. Due to the different growth rates of the hexagonal wurtzite structure ZnO along the polar and non-polar planes, the ZnO material is easy to obtain abundant micro-nano structures such as tetragonal needle-like structures, micro-nano wires, rods, tubes, combs, *etc*. It is own end face, side face and optical feedback formed by random scattering make ZnO micro/nano-structures[3] have a natural resonant cavity that can attain various oscillation modes of laser emission. At present, the laser generated by micro/nano-structures can be classified into three categories: The first is a random laser obtained by randomly forming positive feedback based on particle interface scattering[4]. The second is based on micro-rods or other regular structures to form an Fabry-Perot(F-P) laser. The third is an whispering gallery mode(WGM) laser based on the total reflection of the inner wall of the microcavity[5-6]. However, due to the large optical loss of ZnO microcavities, high-efficiency laser output using micro-nano structures has been difficult to achieve. Improving the luminous efficiency of ZnO UV-emitting devices is currently the main problem[7].

In recent years, many research groups have found that metal surface plasmons(SPPs) can be used to enhance the luminous efficiency of semiconductor materials. SPPs refer to a hybrid electromagnetic wave mode in which free vibrational electrons on a metal surface are coupled with incident photons. It has spatial localization and local field enhancement properties which will have a huge impact on the recombination process of luminescent materials in surface plasmon electromagnetic fields[8-9]. At present, metal local surface plasmons resonance(LSPR) has made great progress in enhancing the photoluminescence(PL) of ZnO micro/nano-materials[10]. Many studies have shown that the combination of ZnO thin films and micro/nano-structures with metal nanoparticles or appropriate metal thin films can greatly enhance the ultraviolet light-emitting efficiency of ZnO through the LSPR coupling effect[11]. The use of metals to modify the optical properties of ZnO has attracted widespread attention. Chen et al. found that the UV and visible light emission intensity can be adjusted by gold nanoparticles[12]. Cheng *et al*. found that sputtering island-shaped Ag nanoparticles effectively enhance the light emission of ZnO thin films[13] and so on. In this paper, the natural resonant cavity of ZnO micron-line structure is used to achieve the F-P mode oscillation formed by the optical feedback of the upper and lower surfaces and the relationship between mode oscillation and optical loss in microcavity is theoretically analyzed. The electric field distribution characteristics of different diameter micro-wires were simulated by the FDTD solution and the optical loss and mode

evolution of different cavity diameters of ZnO microwire were studied. Finally, the LSPR effect of Ag metal nanoparticles were used to enhance the stimulated radiation of ZnO and improve the optical field confinement ability of ZnO micro-wire structures. It lays a foundation for the further application of local surface plasmons in the field of ultraviolet micro/nano-lasers.

2 Analysis

2.1 F-P Oscillation Mode Optical Loss

For laser devices, laser oscillation is generated by the interaction of optical feedback in the cavity with the laser gain medium[14]. Only a laser mode with lower loss can form a stable laser field distribution in the laser gain medium. There are two kinds of loss in the resonant cavity. One is the loss independent of the laser transverse mode, such as the internal loss of the laser working substance, the transmission loss of the cavity mirror, the absorption of the intracavity component, the scattering loss, *etc*. The other is the diffraction loss, which is closely related to the laser transverse mode. The diffraction loss has an important influence on the oscillation of the laser transverse modes with different orders. This characteristic is the physical basis for achieving laser transverse mode selection and obtaining high beam quality laser output. The ZnO microwire F-P resonant mode uses two mutually parallel reflecting surfaces as the cavity structure, as shown in Fig. 1(a). The F-P excitation mode in the ZnO microcavity can be observed in Fig. 1(b) and exhibits optical waveguide characteristics. This structure effectively limits the optical path between two mutually parallel end faces (M1 and M2) to realize gain amplification. The steady-state field of the optical wave of M1 on one of the side of the optical cavity is $E_1(x, y)$ and the steady-state field of the optical wave on the M2 plane is $E_2(x, y)$. The relationship between the two is as follows[15]:

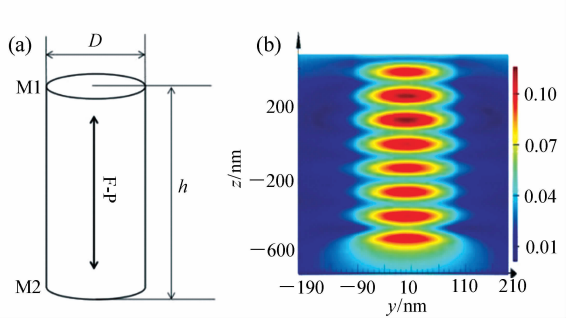

Fig. 1　(a) Structure diagram of an F-P laser cavity; (b) schematic diagram of laser propagation in ZnO microcavity

图 1　(a) 类 F-P 激光腔结构示意图;(b) 激光在 ZnO 微腔中传播的示意图

$$E_2(x,y) = \partial E_1(x,y) , \tag{1}$$

Where ∂ is a complex constant factor defined as the change in the amplitude and the phase of the light field after a single pass propagation. The total energy loss Ψ of light during propagation can be expressed as[16-17]:

$$\Psi = \frac{|E_1|^2 - |E_2|^2}{|E_1|^2} = 1 - |\alpha|^2. \tag{2}$$

If the loss of transmission, absorption, and output is not being considered, α is defined as the diffraction loss of the laser cavity.

2.2 Metal Surface Plasmon

The dielectric constant of the noble metal material has a negative real part and a small positive imaginary part. And, the outer free electrons are easy to oscillate under the excitation of the incident light field. When the free electron oscillation frequency matches the frequency of the incident light field, the metal surface plasmon resonance is generated[18]. According to the structural form of metal materials, this form of resonance is generally divided into two categories. The first is LSPR. The incident light field interacts with the metal nanoparticles. The free electrons on the conduction band collectively oscillate on the metal surface and the generated surface plasmon waves are localized near the nanostructure (Fig. 2(a)). In the resonant state, the energy of

the electromagnetic field is effectively converted into the collective vibrational energy of the free electrons on the metal surface, which in turn produces LSPR[19-20]. The second is surface plasmon resonance (SPR) on the propagation surface, in which light field acts on the continuous metal surface to produce a surface plasmon resonance wave that propagates on the metal surface (Fig. 2(b)). In this paper, the LSPR resonance coupling effect is used to generate near-field local enhancement, which effectively improves the optical field confinement of ZnO micro-wire structure and then enhances F-P stimulated radiation.

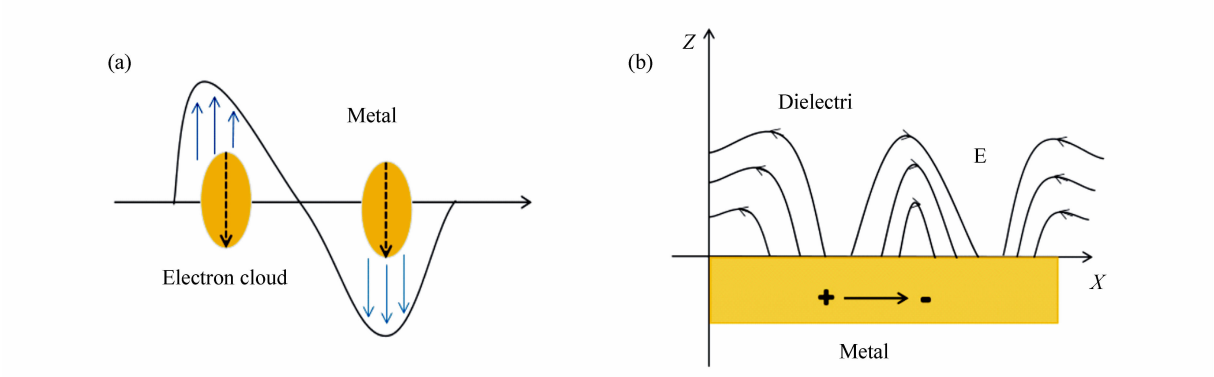

Fig. 2 (a) Localized surface plasmon oscillations; (b) spread surface plasmon

图 2 (a)局域表面等离子激元振荡图;(b)传播表面等离子激元图

3 Results and Discussion

3.1 Optical Loss Characteristics of ZnO Microwires with Different Cavity Diameters

In order to study the relationship between the ZnO micron-diameter and optical loss, the calculation and simulation are carried out by using three-dimensional space time domain finite-difference method for the hexahedral ZnO micro-wires. The effects of the electric field distribution of different cross-sections and the electric field intensity ratio inside and outside the cavity are used to analyze the effect of ZnO micron-diameter on optical loss. In the simulation, the ZnO microwires were placed vertically on the substrate. The height h of the microwires is set to 2.1 μm. Using a single-frequency light source and using a (n, k) material model for simulation, the refractive index of the ZnO material is set to n = 2.06, k = 1 by reference [21]. The corresponding model is used to simulate the change of the mode number in the resonant cavity formed by the ZnO nanowires. The Perfectly Matched Layer (PML) is selected as the simulation boundary condition by setting a special dielectric layer at the truncation boundary in the FDTD region so that the wave impedance of the layer medium and the wave impedance of the adjacent medium are completely matched. Therefore, the incident wave does not reflect at the interface and passes through before it finally enters the PML layer. Although PML has only a finite thickness, it still has a good absorption for incident waves. Therefore, PML is also an absorbing boundary that is often used in actual calculations.

The electric field distributions of the ZnO micro-wires with diameters of 1.6, 2, 4, 6, and 8 μm in the $E(x,y)$ and $E(x,z)$ sections were calculated using the established model, as shown in Fig. 3. The figure clearly shows that the ZnO microwire has optical waveguide characteristics and intracavity F-P mode oscillation. It is found by the electric field distribution that light is confined in the ZnO micron line and forms a standing wave propagating along the cavity. When the cavity diameter is 1.6 and 2 μm, a strong electric field distribution is observed outside the cavity, which indicates that the light will have a

severe energy loss during the propagation in the cavity. As the cavity diameter increases, more energy is added to the cavity to achieve laser resonance. When the cavity diameter is increased to 6 and 8 μm, most of the light can be locked in the cavity and a perfect optical resonance mode can be observed. Through the electric field distribution of the cross-section of the ZnO micron line, it can be found that as the diameter of the ZnO micron line increases, its ability to confine the optical field is enhanced, and the optical loss is reduced[22]. This phenomenon can be analyzed according to the theoretical formula for the laser cavity:

$$N = \frac{D^2}{\lambda h}, \tag{3}$$

where D is the lateral dimension of the microcavity (*ie*. the diameter of the microwire), h is the cavity length and λ is the wavelength. N is the Fresnel number, which is a characteristic parameter in the diffraction phenomenon. The larger the value, the smaller the diffraction loss. It can be seen from the formula (3) that, under conditions where the cavity length h is constant, the Fresnel number N gradually increases as the diameter D of the microwire increases at the same wavelength λ, thereby reducing the diffraction loss in the cavity.

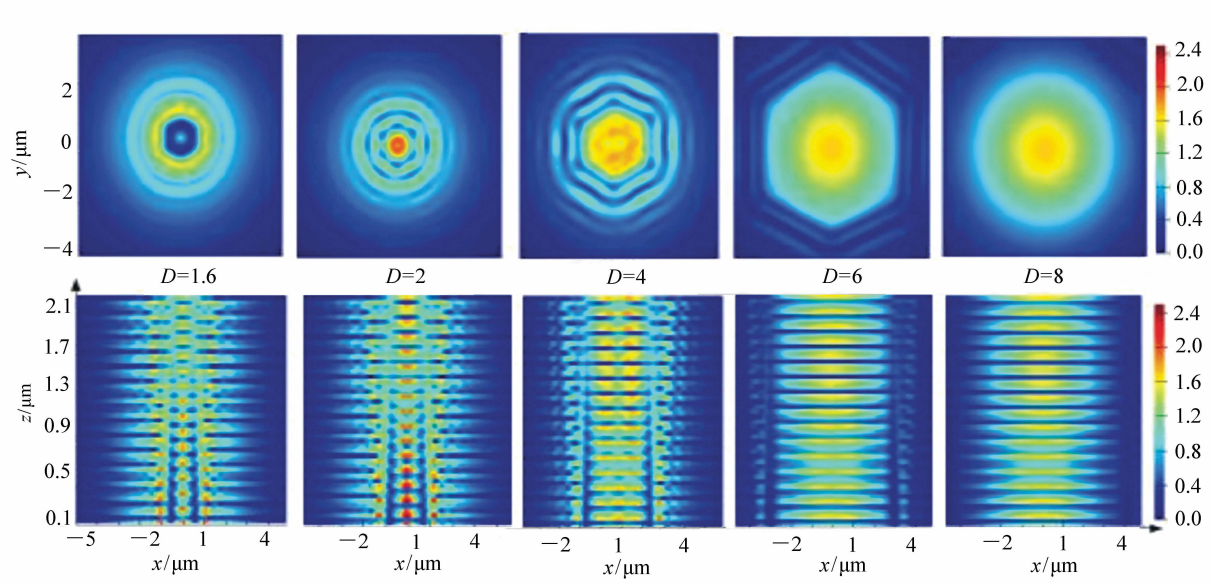

Fig. 3　The electric field distributions of (x,y), (x,z) planes of the ZnO micro-wires with different diameters

图 3　不同直径时的(x,y),(x,z)面电场分布图

Fig. 4 shows the distribution of light intensity and the efficiency of light confinement at different cavity diameters. The trend of the light field distribution in the cavity with different cavity diameters can be observed by Fig. 4(a). It can be seen as the cavity diameter increases, the intensity of the cavity increases gradually, while the intensity of the light field outside the cavity gradually decreases. Fig. 4 (b) shows the light confinement efficiency curve for different cavity diameters. The light confinement efficiency here is defined as the percentage of the light field intensity area in the ZnO micron-line cavity to the total light field intensity area. It can be found that when the diameter is 1.6 μm, the optical field energy loss is at its highest and almost no light exists in the cavity. On the contrary, as the diameter of the microcavity increases, the light field confinement efficiency reaches about 99%, indicating that the light field energy is almost entirely limited to the intracavity propagation when the diameter reaches a certain level. Therefore, the F-P microcavity mode oscillation can be constructed by using the ZnO micro-wire structure light field confinement ability, and the optical loss characteristic can be effectively regu-

lated by changing the micro-wire diameter.

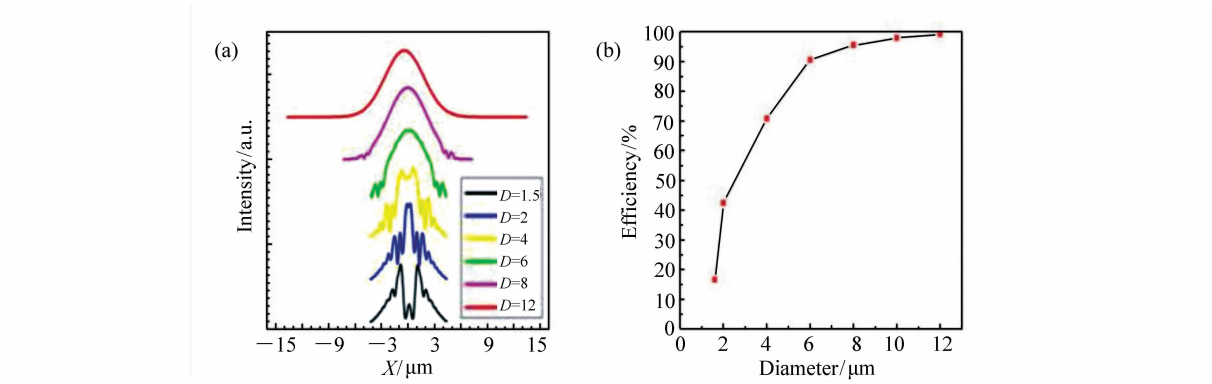

Fig. 4 (a) Light intensity curves corresponding to different cavity diameters; (b) light-confinement efficiency varies with cavity diameter

图 4 (a)不同腔径对应的光强曲线;(b)随腔径变化的光限制效率图

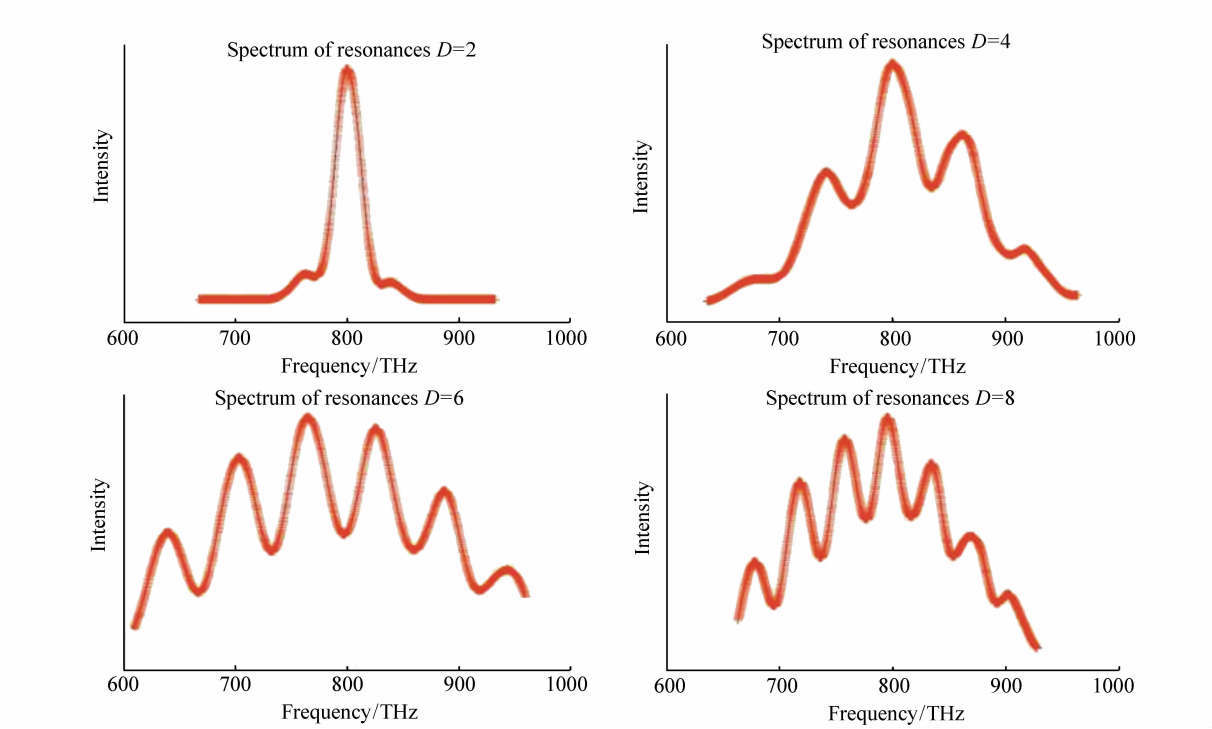

Fig. 5 ZnO F-P laser resonant spectra with different diameters

图 5 不同直径时的 ZnO F-P 激光腔共振谱图

3.2 Pattern Evolution of ZnO Microwires with Different Cavity Diameters

In order to study the influence of ZnO micron-cavity diameter on the F-P oscillation mode, the resonance mode analysis group was used to simulate the resonance spectrum evolution process in the micro-wire, as shown in Fig. 5. It can be seen from the above figure that as the diameter of ZnO increases, multiple resonant modes appear gradually in the resonant cavity. When $D = 2$ μm, there are three resonance peaks in the resonance spectrum, where the strongest peak appears at $f = 800$ THz. When D is increased to 4 μm, three strong resonance peaks appear and the strongest peak position is substantially

unchanged. As the diameter of the micron wire continues to increase to 6 and 8 μm, the multi-stage resonance peak increases significantly, and the resonance intensity increases accordingly. Studies have shown that as the diameter of ZnO increases, the number of intracavity resonance modes can also increase. Therefore, small-cavity ZnO micron-wire has the possibility of realizing a single-mode laser[23]. The reason for the formation of the single-mode laser is mainly due to the side mode suppression caused by optical loss. It is thus crucial to study the small cavity optical loss. In order to improve the performance of the traditional optical microcavity system, an optical cavity mode is proposed using the microcavity structure modified by metal local surface plasmons to achieve the coupling of the evanescent field loss light and the metal local plasmon wave. It is expected that metal local surface plasmons can be used to enhance light interaction with matter and to provide a limited waveguide mode in the sub-wavelength range.

3.3 Ag Nanoparticles Regulate the Light Field Characteristics of ZnO Microcavity

By using Ag nanoparticles to modify the ZnO micro-wire excitation to form the LSPR electric field, the local field enhancement and optical field confinement effect can be achieved on the surface of the micro-wire[24-25]. The problem of large optical loss in small-cavity ZnO micro-wires can be effectively suppressed by metal LSPR. To this end, Ag nanoparticles were used to modify the surface of ZnO microwires with a diameter of 2 μm and the electric field was recorded by an *x*-*y* section electromagnetic field monitor.

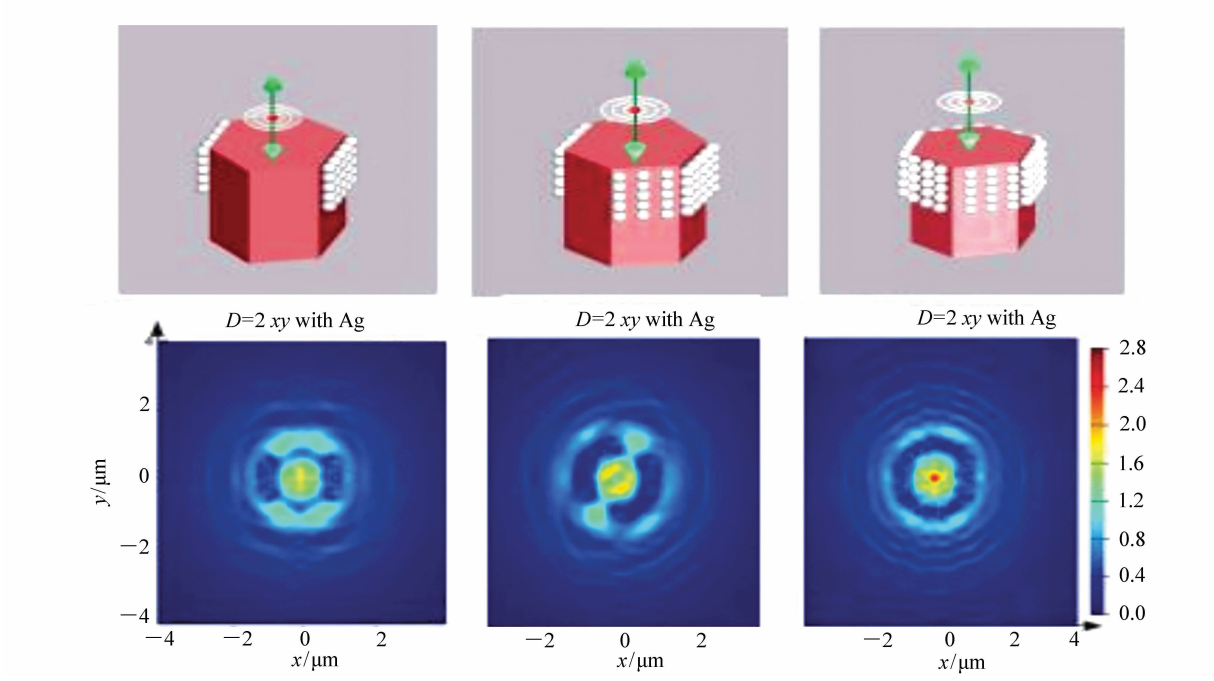

Fig. 6　Simulation diagram of LSPR microcavity modified by Ag nanoparticles and corresponding electric field distributions

图 6　Ag 纳米颗粒修饰的 LSPR 微腔的模拟示意图和对应电场分布

Fig. 6 is a structural diagram and a light field distribution diagram of 2, 4, and 6 surfaces of Ag-modified ZnO microwires. It can be easily seen from the electric field distribution diagram of the *x*-*y* cross-section that the optical field in the cavity is enhanced by the modification of the Ag nanoparticle, which proves the ability of the LSPR to limit the optical field. Since the work function of Ag equates to 4.35 eV and the electron affinity of ZnO is 4.26 eV, the electrons are easily transferred to each other

at the interface of Ag/ZnO. In addition, the ZnO emission energy is close to the energy of the plasmon resonance of the Ag surface, so the enhancement in light field intensity is the result of the coupling between the local surface resonance plasmon of the Ag nanoparticle and the photons from the stimulated emission of the ZnO microrod.

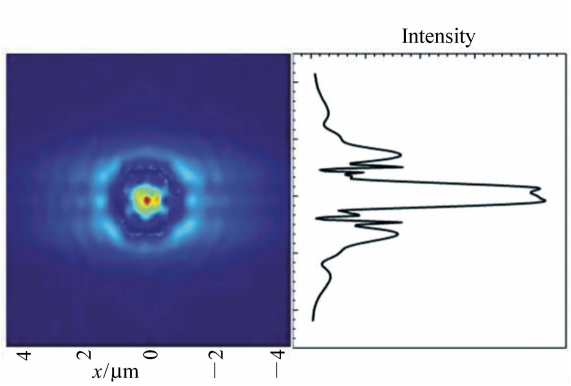

Fig. 7 Distribution of the electric field intensity of the Ag nanoparticle modified microcavity

图 7 Ag 纳米颗粒修饰微腔的电场强度分布图

Fig. 7 is an *x*-*y* plane electric field distribution diagram of the Ag particle modified ZnO microwire and its electric field intensity curve. It can be seen that there is a clear light field enhancement effect in the area where the Ag particles are in contact with the micron lines, corresponding to the two sharp enhancement peaks of the intensity curve of Fig. 7. The formation of these two enhancement peaks is the result of the resonant coupling of the local surface plasmon with the loss light in the microwire.

Since the micron-scale ZnO micro-wire structure is much larger than that of the nano-scale Ag particles, the contact surface of ZnO and Ag is equivalent to a plane. A local simulation of the Ag/ZnO microwire interface was performed to monitor electric field in the *x*-*y* cross-section of the local field and the *x*-*z* end surface, respectively, as shown in Fig. 8. 5 × 5 equal-sized Ag nanoparticles were placed on the ZnO plane. Through the electric field distribution diagrams of the cross-section and the end surface, it can be found that there is coupling light enhancement at the edge of the Ag particle and that between the two metal particles, the electric field strength along the *X*-axis direction has a two-fold enhancement effect. This phenomenon is mainly due to the secondary coupling phenomenon between metal particles. First, the excitation of the plasmon and the micron-line photons create resonant coupling and amplification. Second, the oscillating coupling between the *X*-direction metal particles is generated by the resonance-enhanced energy.

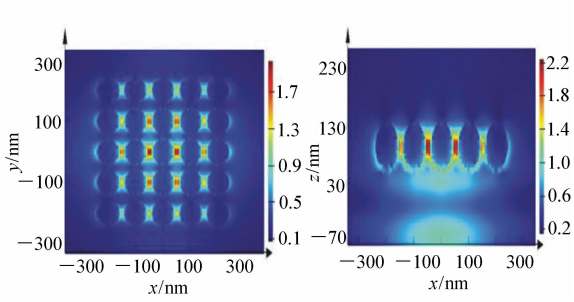

Fig. 8 Local electric field diagrams of Ag nanoparticle modified ZnO microwire

图 8 Ag 纳米颗粒装饰 ZnO 微米线的局部电场图

In order to explain and quantify the effect of metal LSPR on the optical loss of the ZnO micron wire structure, Fig. 9 simulates the distribution curve and light confinement efficiency of ZnO micron lines with 2, 4 and 6 sides modified by Ag nanoparticles respectively. Fig. 9(a) is a graph of the light intensity change of different amount of Ag nanoparticle modification, and Fig. 9(b) is a graph of the corresponding light-confinment efficiency of ZnO micron line. It can be seen from the figure that as the decorative surface of Ag nanoparticles increases, the light field confinement efficiency improves significantly. Through calculation for the six-face-modified ZnO micro-wires with Ag nanoparticles, the light field-confinement efficiency of the ZnO micro-wires increased by 6.72% and the light field in the cavity is enhanced significantly.

This is because the evanescent wave field intersects with the plasma exciton localized on the surface of the ZnO, providing an environment in which the

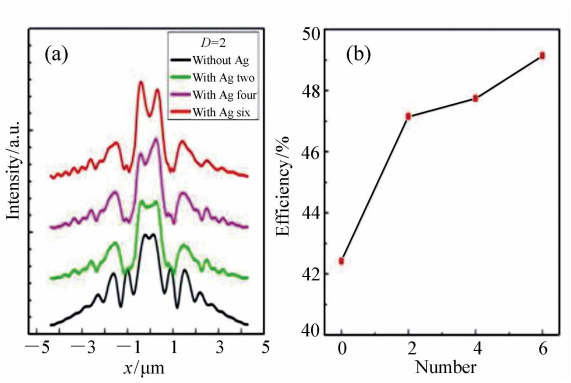

Fig. 9　(a) Light intensity curve as a function of the number of decorative surface; (b) light confinement efficiency map as a function of the number of decorative surfaces

图 9　(a)随装饰面数目变化的光强曲线;(b)随装饰面数目变化的光限制效率图

surface plasmon resonance mode is coupled with the traditional F-P resonant cavity mode. Thus, the energy distribution and oscillation process in the microcavity are regulated.

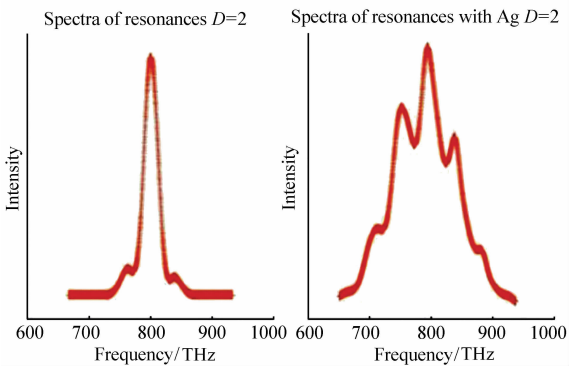

Fig. 10　Resonance spectra of ZnO microcavity without and with Ag particle decoration when $D = 2$

图 10　$D = 2$ 时无 Ag 颗粒装饰和有 Ag 颗粒装饰时 ZnO 微腔的谐振谱图

The resonance pattern of the decorative Ag particles modified micro-wires was also simulated by the resonance mode analysis group, as shown in Fig. 10. It can be seen that the multi-stage resonance peak in the ZnO micron-wire resonator modified by Ag nanoparticles becomes strong and increases from the original three resonance peaks to five, and that the stable resonance mode in the micro-wire increases significantly. This is mainly due to the surface plasmon resonance effect produced by Ag nanoparticles and the optical field confinement effect of the intersection region, allowing the ZnO cavity with same diameter to have a stronger light field. This also reduces the band edge loss, helps improve the performance of the laser, and achieves more resonant modes.

4　Conclusion

In this paper, the optical loss characteristics and F-P mode evolution characteristics of ZnO micro-wire structures with different diameters were analyzed by simulation. It could be seen by the electric field distribution that as the diameter of the ZnO micro-wire increased, their ability to confine the optical field increased and the optical loss decreased. Through the resonance mode analysis group, it could be seen that more resonant modes were generated in the cavity as the diameter of the ZnO microwire increased. In addition, a ZnO microcavity structure with an Ag nanoparticle decoration was established. An LSPR electric field was formed by exciting Ag nanoparticles and the local field enhancement and optical field confinement effects were achieved on the surface of the micrometer line. Moreover, the metal LSPR effectively limited the problem of optical loss in the small cavity diameter ZnO micro-wire. It is found that the limiting ability of the optical field increases by 6.72% after the Ag nanoparticles were modified on the surface of the microcavity with high loss. A secondary coupling phenomenon occurs between the metal particles along X-axis direction, the strength of the electric field is twice as strong as that of the original electric field. Finally, LSPR was used to effectively control the evolution of the ZnO microwire resonance mode. The research in this paper lays a theoretical foundation for the design and implementation of high-performance UV laser devices.

——中文对照版——

1 引 言

氧化锌(ZnO)材料作为具有代表性的第三代宽禁带半导体,其禁带宽度达到3.37 eV,且室温下激子束缚能高达60 meV,因此ZnO材料用于制作高品质光学微腔和高效率紫外光电子器件方面引起广泛的关注[1-2]。由于六角纤锌矿结构ZnO沿极性面与非极性面的生长速率不同,导致ZnO材料容易获得丰富的微纳米结构,如四角针状结构、微纳米线[3]、棒、管、梳等。其自身端面、侧面以及随机散射形成的光学反馈,使得ZnO微纳米结构具有自然谐振腔并可以获得多种振荡模式激光发射。目前利用微纳米结构可产生3种类型的激光:一是基于颗粒界面散射,随机形成正反馈而得到的随机激光[4];二是基于微米棒等规则结构的端面反射形成F-P激光;三是基于微腔内壁全反射形成的回音壁模(WGM)激光[5-6]。然而,由于ZnO微腔的光学损耗较大,基于微纳结构的高效率激光输出一直难以实现,因此有效提高ZnO紫外发光器件发光效率是目前面临的主要问题[7]。

近年来,许多研究组都发现金属表面等离子激元(SPPs)可以用来增强半导体材料发光效率。SPPs是指金属表面的自由振动电子与入射光子相互耦合而形成的一种杂化电磁波模式,它具有空间局域性和局域场增强特性,这一性质将对处在表面等离子激元电磁场中的发光材料的复合过程有着巨大的影响[8-9]。目前,金属局域表面等离子激元共振(LSPR)在增强ZnO微纳米材料的光致发光(PL)方面取得了巨大的进展[10]。已有不少研究表明,将ZnO薄膜、微纳米结构等与金属纳米粒子或适当的金属薄膜相结合,通过LSPR耦合效应,可极大增强ZnO的紫外发光效率[11]。利用金属对ZnO光学性能进行改性已引起人们的广泛关注。Chen等人发现,紫外和可见光发射强度可以由金纳米粒子来调整[12]。Cheng等人研究发现溅射岛形Ag纳米颗粒可有效增强ZnO薄膜的光发射[13]等。本文利用ZnO微米线结构具有的自然谐振腔,实现上下表面光学反馈形成的F-P模式振荡,并对微腔中的模式振荡与光学损耗之间的关系进行理论分析。通过FDTD Solution模拟计算不同直径微米线中的电场分布特性,研究分析不同腔径ZnO微米线的光损耗和模式演化规律。最后,利用Ag金属纳米粒子的LSPR效应,实现ZnO受激辐射的增强,并提高ZnO微米线结构的光场限域能力。为局域表面等离子激元在紫外微纳米激光器领域的进一步应用奠定研究基础。

2 理论分析

2.1 F-P振荡模式光学损耗

对于激光器件来说,激光振荡是通过谐振腔[14]内的光学反馈与激光增益介质的共同作用产生的,只有损耗较低的激光模式才能在激光增益介质中形成稳定的激光场分布。在谐振腔内存在两种不同性质的损耗,一种是与激光横模阶数无关的损耗,如激光工作物质的内部损耗、腔镜的透射损耗,腔内元件的吸收、散射损耗等;另一种则是与激光横模阶数密切相关的衍射损耗。衍射损耗对激光的各阶横模的振荡都有重要影响,这一特性是实现激光横模选择并得到高光束质量激光输出的物理基础。

ZnO微米线F-P谐振模式采用两个相互平行的反射面作为腔体结构,如图1(a)所示。图1(b)中可以观察到ZnO微腔中的F-P激发模式,并表现出光波导特性。

此结构有效将光路限制在两个相互平行的端面M1、M2之间进行振荡,从而实现增益放大。光腔M1两侧面的光波稳态场为$E_1(x,y)$,M2面的光波稳态场为$E_2(x,y)$,两者之间的关系如下[15]:

$$E_2(x,y) = \partial E_1(x,y), \tag{1}$$

式中,∂为一复常数因子,定义为一次单程传播后,光场振幅和位相的变化。光在传播过程中的总能

量损耗 Ψ 可以表示为[16-17]:

$$\Psi = \frac{|E_1|^2 - |E_2|^2}{|E_1|^2} = 1 - |\alpha|^2, \quad (2)$$

如果不考虑传输、吸收和输出等损耗,α 定义为激光谐振腔的衍射损耗。

2.2　金属表面等离子激元

贵金属材料介电常数具有负实部和小的正虚部,且外层自由电子在入射光场的激发下易发生振荡,当自由电子振荡频率与入射光场的频率相匹配时产生金属表面等离激元共振[18]。根据金属材料结构形式的不同,这种共振形式一般分为两类:第一种为 LSPR,入射光场与金属纳米颗粒相互作用,导带上的自由电子在金属表面发生集体振荡,产生的表面等离子激元波被局域在纳米结构附近(如图 2(a))。共振状态下,电磁场的能量有效地转化为金属表面自由电子振动能,进而产生 LSPR 效应[19-20]。第二种是传播表面等离子共振(SPR),光场作用在连续金属表面,从而在金属表面产生传输型的表面等离子激元共振波(图 2(b))。本文利用 LSPR 共振耦合效应产生近场局域增强,有效提高 ZnO 微米线结构光场限域能力,进而增强 F-P 受激辐射。

3　结果与讨论

3.1　不同腔径 ZnO 微米线的光学损耗特性

为了研究 ZnO 微米线腔径变化对其光学损耗造成的影响,本文对正六面体 ZnO 微米线进行三维空间时域有限差分法的计算模拟,利用不同横截面的电场分布及腔内腔外电场强度的比值,分析 ZnO 微米线腔径变化与光学损耗之间的联系。在仿真模型中,将 ZnO 微米线垂直放置在衬底上,微米线的高度 h 设置为 2.1 μm,使用单频光源,并在仿真模拟时利用(n,k)材料模型创建 ZnO 材料,通过文献[21]估算设置其折射率 n = 2.06,k = 1 的对应模型来模拟 ZnO 微米线形成的谐振腔里模式数量的变化。选择完美匹配层(Perfectly Matched Layer,PML)作为其模拟边界条件。方法是在 FDTD 区域内的截断边界处设置一种特殊介质层,使得该层介质的波阻抗与相邻介质波阻抗达到完全匹配,从而确保入射波在分界面上不发生反射,穿过分界面最后进入 PML 层,而且 PML 层存在有耗介质,可以使进入 PML 层的透射波迅速衰减,尽管 PML 的厚度有限,但它对入射波却有很好的吸收效果。因此,PML 在实际计算中也是一种常被使用的吸收边界。

利用建立的模型分别计算直径为 1.6、2、4、6 和 8 μm 的 ZnO 微米线在 $E(x,y)$ 截面及 $E(x,z)$ 截面的电场分布,如图 3 所示。图中可以清晰反映出 ZnO 微米线具有光波导特性,以及腔内 F-P 模式振荡。通过电场分布可以发现光被限制在 ZnO 微米线中并形成驻波沿腔体传播。当腔径为 1.6 和 2 μm 时,观察到腔外具有很强的电场分布,这说明光在腔内传播的过程中会产生严重的能量损失。随着腔径的逐步增加,腔内会加入更多的能量来实现激光的共振。当腔径增加到 6 和 8 μm 时,绝大部分光可以被锁定在腔中并可观察到完美的光学共振模式。通过 ZnO 微米线横截面上的电场分布可以发现,随着 ZnO 微米线腔径的增加,其对光场的限域能力增强,光学损耗降低[22]。这一现象可以根据激光谐振腔理论公式来分析:

$$N = \frac{D^2}{\lambda h}, \quad (3)$$

其中,D 为微腔横向尺寸(即微米线的直径),h 为腔长,λ 为波长。N 为菲涅耳数,它是衍射现象中的一个特征参数,其数值越大表征衍射损耗越小。通过公式(3)可知,在腔长 h 不变的情况下,在同一波长 λ 下,随着微米线直径 D 的增加,菲涅耳数 N 逐步增大,进而导致谐振腔内的衍射损耗降低。

图 4 为不同腔径下光强的分布曲线及光限制效率。通过图 4(a)可以观察到不同腔径下腔内光场分布的变化趋势。随着腔径的增加,腔内光强逐渐增强,而腔外的光场强度逐渐减弱。图 4(b)给出不同腔径对应的光限制效率变化曲线。这里的光限制效率定义为:ZnO 微米线腔内光场强度面积占总的光场强度面积的百分比。可以发现当直径为 1.6 μm 时,光场能量损耗最大,腔内几乎没有光场存在。相反,随着微腔直径的增加,光场限制效率达到了 99% 左右,说明直径达到一定程度时,光场能量几乎全部限制在腔内传播。因此利用 ZnO 微米线结构的光场限域能力可以

构建 F-P 微腔模式振荡,并且通过改变微米线直径可以有效调控光学损耗特性。

3.2 不同腔径 ZnO 微米线的模式演化

为了研究 ZnO 微米线腔径对 F-P 振荡模式的影响,利用谐振模式分析组对微米线中的谐振谱演化过程进行了模拟仿真,如图 5 所示。

通过图 5 可以看出,随着 ZnO 直径的增加,谐振腔中逐渐出现多个谐振模式。当 $D = 2$ μm 时,谐振谱中共有 3 个谐振峰,其中 $f = 800$ THz 处表现出一个最强峰。当 D 增加到 4 μm 时,出现 3 个较强的谐振峰,且最强峰位置基本不变。随着微米线直径继续增加到 6 和 8 μm 时,多级谐振峰明显增多,谐振强度也相应增加。有研究表明随着 ZnO 直径的增加可以使腔内谐振模式的数量增加,而小腔径 ZnO 微米线具备实现单模激光的可能性[23]。形成单模激光的机理主要是光学损耗引起的边模抑制,因此对小腔光学损耗的研究至关重要。为了改善传统光学微腔系统的性能,提出利用金属局域表面等离子激元修饰微腔结构,来实现微腔侧面倏逝场损耗光与金属局域等离子激元波耦合的一种光学腔模式。这将有望利用金属局域表面等离子激元加强光与物质相互作用并提供一个亚波长范围的限制波导模式。

3.3 Ag 纳米颗粒调控 ZnO 微腔光场特性

利用 Ag 纳米颗粒修饰 ZnO 微米线激发形成 LSPR 电场,可以在微米线表面实现局域场增强及光场限域效应[24-25]。对于小腔径 ZnO 微米线光学损耗较大的问题,可以通过金属 LSPR 进行有效抑制。为此,应用 Ag 纳米颗粒修饰直径为 2 μm 的 ZnO 微米线表面,并通过 x-y 截面电磁场监视器记录电场的变化。图 6 为 Ag 修饰 ZnO 微米线 2、4、6 个面的结构示意图及光场分布图。通过 x-y 截面的电场分布图可以看出,经 Ag 纳米颗粒修饰后腔内光场明显增强,这证明 LSPR 对光场具有限域能力。由于 Ag 的功函数为 4.35 eV,ZnO 的电子亲和能为 4.26 eV,在 Ag/ZnO 交界面上电子容易相互转移。此外 ZnO 发射能量接近于 Ag 表面等离子激元共振的能量,因此光场强度的增强是 Ag 纳米颗粒的局域表面等离激元共振与 ZnO 微米棒受激发射的光子相耦合的结果。

图 7 为具有 Ag 颗粒修饰 ZnO 微米线的 x-y 面电场分布图以及电场强度曲线图。可以发现,在 Ag 颗粒与微米线相接触的区域有明显的光场增强效果。对应于图 7 的强度曲线可以观察到两个尖锐的增强峰,这两个增强峰的形成正是局域表面等离子激元与微米线中损耗光共振耦合的结果。

由于微米尺度的 ZnO 微米线结构远大于纳米尺度的 Ag 颗粒,所以将 ZnO 与 Ag 的接触面等效成一个平面,对 Ag/ZnO 微米线界面进行局部模拟,分别监视局部场的 x-y 截面以及 x-z 端面电场,如图 8 所示。在 ZnO 等效平面上放置 5 × 5 个等直径的 Ag 纳米颗粒。通过截面和端面的电场分布图可以发现在 Ag 颗粒边缘有耦合光增强,而且在两个金属颗粒之间,沿 X 轴方向的电场强度更有 2 倍的增强效果。这种现象主要是因为金属颗粒之间可以产生二次耦合,即首先激发的等离子激元与微米线光子之间产生共振耦合及放大;其次是经过共振耦合增强的能量在 X 方向金属颗粒之间产生的振荡耦合。

为了解释及量化金属 LSPR 对 ZnO 微米线结构光学损耗的影响,图 9 仿真计算了 Ag 纳米颗粒分别修饰 ZnO 微米线 2、4、6 个侧面下光强的分布曲线及光限制效率,如图 9 所示。图 9(a)为不同数量 Ag 纳米颗粒修饰面对应的光场强度变化曲线图;图 9(b)为对应的 ZnO 微米线光限制效率的曲线图。通过图 9 可以看出,随着 Ag 纳米颗粒修饰面的增加,光场限制效率有了明显的提高。经过 6 面修饰 Ag 纳米颗粒后,ZnO 微米线的光场限制效率提高了 6.72%,并且腔内的光场有了明显的增强。这是因为倏逝波光场与局域在 ZnO 表面的等离子激元形成交叉区,提供了一个实现表面等离子激元共振模式与传统的 F-P 谐振腔模式耦合的环境,从而调节微腔中的能量分布和振荡过程。

同样利用谐振模式分析组模拟得到了微米线修饰 Ag 颗粒前后的谐振谱图,如图 10 所示。可以看出,通过 Ag 纳米颗粒修饰后,ZnO 微腔内多级共振峰变强且由原来的 3 个谐振峰增加到 5 个,微米线内稳定的谐振模式明显增多。这主要因为 Ag 纳米颗粒产生的表面等离子共振效应和交叉区的光场限域效应,使得相同直径的 ZnO 腔

内具有更强的光场,且降低了带边损耗,有助于提高激光的性能,并获得更多的谐振模式。

4　结　论

本文通过仿真计算分析不同直径 ZnO 微米线结构的光学损耗特性及 F-P 模式演化特性。通过电场分布发现随着 ZnO 微米线腔径的增加,其对光场的限域能力增强且光学损耗降低。通过研究谐振模式分析组发现,随着 ZnO 微米线直径的增加腔内产生了更多的谐振模式。此外,建立具有 Ag 纳米颗粒修饰的 ZnO 微腔结构。利用 Ag 纳米颗粒激发形成 LSPR 电场,在微米线表面实现局域场增强及光场限域效应。此外,通过金属 LSPR 有效改善小腔径 ZnO 微米线光学损耗较大的问题。分析发现,在损耗较大的微腔表面修饰 Ag 纳米颗粒以后,光场限域能力提高了 6.72%。而且在金属颗粒之间沿 X 轴方向产生二次耦合现象,其电场强度更有 2 倍的增强效果。最后利用 LSPR 有效调控 ZnO 微米线谐振模式的演化。本文的研究为设计和实现高性能紫外激光器件奠定理论基础。

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