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PRIMARY RESEARCH

A review on the formation of colors on SS304 stainless steel induced by laser color marking technique

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Abstract

This review comprehensively covers the research accomplished in the field of laser color marking process for various industrial applications. Laser color marking technology in recent years has been envisaged for numerous applications of all types of solid materials in a single step process. Likewise, the traditional laser marking process generally produces contrast marks which are often monochromatic. Therefore, there is more interest in color marking by laser processing for decoration and visual attraction in recent years. It is currently the most studied research to observe the effect of laser irradiation process parameters and external environmental factors on the formation of different colors on metals and non-metallic surfaces. This paper presents a detailed review of the recent advancement in laser coloring technologies. Furthermore, the process of laser color marking has been discussed based on the compatible parameters of the color mark and laser types. Moreover, the effect of laser processing parameters on microstructural features and the corresponding metallurgical defects encountered such as cracks and chemical composition are recapped. Finally, this article summarizes the present-day applications of the laser color marking technique used in many industrial sectors.

I. INTRODUCTION

Laser-based manufacturing processes such as laser surface modification [1], laser surface texturing, laser hardening [2], laser welding [3], laser cladding etc., are becoming popular due to their robust and rapid production time [4, 5, 6, 7]. Amongst these manufacturing processes, laser-induced color marking of metals is gaining recognition and is increasingly applied in various industries in the new era of technological development. This process is rapidly replacing traditional marking methods such as painting, etching, and printing [8, 9]. However, the process adds color to an object that is being marked merely "stains" on the surface. It has become a foremost industrial process today in many applications, such as labeling a serial number, date, barcode, Quick Response (QR) code, company logo, trademarking, and other product information. There are several types of color marking process including etching, marking, and lithography. However, Laser color marking boasts high quality, difficulty to be forged, wear-resistant and is a non-contact marking process that does not cause any contact pressure nor deformation whilst being sustainable [10].

The process of laser color marking ensues when the oxide layer is created on the ferrous metal such as steel and high-grade steel through localized heating. In general, the oxide layer that is created on the surface is black, but it can also be tailored to obtain a variety of colors such as red, blue,

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and green, etc. [11]. Hence, the color produced on the surface of a metal primarily depends on the temperature of the heated layers and the penetration of heat to the depth of the metal surface. The phenomena of color production are also dependent on chromium due to changes in molecular structure and electrons on the metal surface. The laser acts as a heat source allowing the formation of a thin transparent or semi-transparent oxide film. Suppose a beam of parallel monochromatic light falls, at an angle, onto a transparent or partly transparent layer. In that case, this beam is repeatedly reflected from the surfaces limited by the oxide layer (from the surface of both oxide and metal) [10]. By increasing the thickness of oxide on the surface and by changing the interference effect (each wavelength has a different degree of attenuation or amplification), a color effect can be observed and controlled.

The most common laser employed in the laser color marking is a pulsed wave mode nanosecond, pico-second, or femtosecond laser, due to the difference in the behavior of their pulse width and pulse frequency. Therefore, both femtosecond and pico-second lasers induced color on the metal surface using the Laser-Induced Periodic Surface Structure (LIPSS) technique to form color on the metal surface [12, 13]. Previous research has demonstrated the influence of the pulse frequency, mark depth width, and mark contrast on the color marking process which depend on the interaction process of the laser beam and the material which is influenced dramatically by the pulse frequency on the quality of the mark [8]. Most of the researchers have analyzed the parameters of the laser such as laser power, frequency, hatching distance, defocusing, and scanning speed on this color mark process, where they summarized the effect of these factors on altering the coloring effect on the metal surface [14, 15]. Hence, others have analyzed the mechanism of metal surface coloring effect on the thermal absorption, along with the thermodynamics of the laser process. However, the color marking implementation and development of the metal surface is still ongoing, since the formation of color on the surface is not uniform throughout the laser-scanned area, where the laser process color mark stability depends on many factors such as temperature, laser parameters and the quality of the metal surface [15]. Therefore, color marking is a versatile and aesthetics related technology that has been replacing traditional methods such as painting, etching, printing, etc. However, the limitations of using the conventional techniques to produce the color include time consumption and mass production of colors. The color can be produced by a relatively newer process known as laser marking in the modern era. By using this technique, the colors are produced depending on the oxide layer and the penetration of the heat through the depth of the surface of a metal to produce the color. Additionally, users can overcome the difficulties of producing color on the metal surface for numerous applications in industries. Although authors have correlated laser parameters with the colors produced, they have not investigated the effect of surface roughness on the formation of colors.

II. INTRODUCTION TO LASER COLOR MARKING PROCESS

In the current modern technological world, there is a continuous increase in demand for metal marking in various products in many industries. This includes the major manufacturing industry, security improvisation, and effectiveness, primarily when this marking is used for serial number printing or batch production numbers, in terms of QR or 2D barcodes [16]. This is mainly to overcome industrial infringement and IP protection, which is brought down to proprietary concerns. Moreover, the color reproduction on metal surfaces requires a specific technique that only can be employed for particular applications since color represents a unique marking that counters the differentiation [16].

In the exiasting color marking techniques a conventionally method has been employed for color marking where the metal surface are chemically treated to form the color on the surface of the metal [17]. In this technique, the formed layer is characterized by structural weaknesses and porosities during the process. It is essential to eliminate these defects using an electrochemical treatment to refurbish for the sustainability of color layer. Alternatively, the thermal oxidation treatment turns up as an option used for stainless steel coloring at high temperatures, approximately 500°C or higher [11, 18, 6, 7]. All these prevalence techniques are often based on the thermochemical treatment of the steel which has a high damage potential through penetrating at the micro and macro structure levels of the material. In a thermochemical process, a close loop of heat source would be applied to eliminate any damage in the material structure layers to obtain high quality and reliable products. In this perspective, laser beams with features such as monochromaticity, directivity, and Spatio-temporal coherence can achieve a controlled heat concentration onto surfaces of selective materials [8].

This objective can mainly be achieved by having the process of laser marking with two methods. Primarily marking the surface by using a specific pattern or shape of the mask and laser scanning marking onto the metal surfaces [19]. In the marking method, the focus of laser beam pen-

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etrating through a localized marking area before the light exposing the void area of marking onto the surface of the workpiece. Hence by using this method any complex feature of the product with the mark image can be imprinted over the selected surface by adapting the optical parameters set up. Technically, this marking method employs a high energy pulsed laser intensity. Moreover, the second method involves the displacement of the focused laser beam onto the material surface from the selected workpiece by using two galvanometric mirrors; each selected mirror guides the laser beam in the concentrated direction. By having the proper maneuver to the mirrors, it is feasible to obtain the desired trajectory for the laser beam. The earlier technique of metal color marking has shown the significance of using a laser source that tends to nail down to a very localized modification of the physical properties of the workpiece surface, without causing any changes to the bulk material properties of the structure. Ultimately, this lasering technique had a high success rate of producing various colors on small surfaces. So, based on the advancement of the rapid processing of laser coloring in the modern world, this method has become a leading industrial process.

III. TYPES OF LASERS USED IN COLOR MARKING

A. Nd: YAG LASER

A laser is an optoelectronic instrument that generates irradiation rays. In this context, the basic type of laser device running under solid-state laser as an active medium is known as Nd: YAG (Neodymium-doped Yttrium Aluminium Garnet), which is also called crystal laser. The basic construction of this laser consists of 4 level subsystems or 4 stages of energy bands to multiply and magnify the reproduction of a laser beam. Therefore, the overall system is operated by either pulsed or continuous mode, and the laser light spectrum falls closer to the infrared regions of 1064 nanometers (nm). Moreover, the laser wavelength range covers 1440 nm, 1320 nm, 1120 nm, and 940 nm [20].

The system connectivity consists of three elements, including an energy source, active medium, and optical resonator. Lasering this process, creation to achieve population inversion the ignitor serves as a pump source to achieve population inversion which is responsible for supplying a fragment of photons toward the active medium for the oscillation. This process is accomplished using a flash tube or laser diodes as an energy source. In the early era of laser inventions, flash tubes become a primary medium to serve as a source of pumps due to cost-effectiveness and technological limitation. However, over the years of laser improvement with nanotechnology advancement, MEMS has been leading Tec in the semiconductor industry. Thus, laser diodes are now favored over flash tubes due to their high performance [21]. In the second stage, the Nd: YAG laser consists of a synthetic crystalline material (Yttrium Aluminium Garnet (YAG)) doped with a chemical element (neodymium (Nd)) which serves as an active medium. Principally, the active medium provides lasing action whereby lower energy state electrons of the neodymium ions will be excited to a higher state of energy. A simple illustration of the working process is shown in Fig. 1, the construction of a laser beaming subsection placed in-between two mirrors that have been optically coated with silver, one mirror fully coated while the opposite mirror is partially coated. The fully coated mirror has a high tendency to bounce every particle without losing, but the partially coated mirror which is known as a partially reflecting mirror, is designed to allow a small portion of the light to penetrate and then accumulate to become a laser beam.

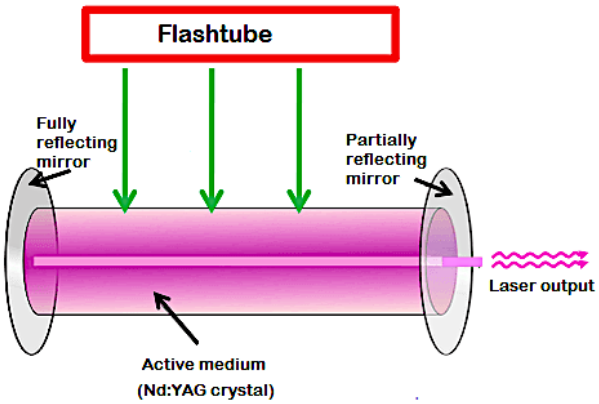

Fig. 1. Schematic diagram of Nd: YAG laser [22]

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To understand the working concept of Nd: YAG laser, the crystalline atom that bonds could be subdivided into a four-level quantum energy layer with N number of electrons, E1, E2, E3, and E4.Suppose the energy levels are assumed to be E1<E2<E3<E4, then the initial population should be N1>N2>N3>N4. The level of energy for E1 is called the ground state, and it elevates to E2, which is the next higher energy state or excited state and moves on to E3, which is the excited state or metastable; finally, E4 is the state of a pump or excited state [22]. This chain of reaction firstly starts when the light energy is supplied to the active medium from the source of either flash tube or laser diode. So the electron from the stable valence receives a sufficient amount of energy to agitate and crossover till reaching the maximum boundary layer of the valence to reach a steady-state condition which is E4. However, the pump-state lifetime at E4 which has a higher energy-state, only sustains a very short fragment of time (230 microseconds (μs)). When the electron decay retunes to its original state, each layer of electrons collapses into a lower energy state until reaching the electron equilibrium. For instance, from E4 to E3, the rapid reactive action releases non-radiation energy (releasing energy without producing photons). At E3, it is called a metastable state lifetime of the metastable state is small. The electrons thus entry to E3 is much quicker than leaving E3. This leads to an increase in the number of electrons in the metastable E3, thereby achieving population inversion. After a certain duration, by releasing photons of light, the electrons on metastable state E3 will step down into the next lower energy state E2, as shown in Fig. 2, which describes every step of energy emission. The emission of photons is called spontaneous emission. Finally, the electrons in the energy state E2 will return to the ground state E1 after a short period by releasing radiation-free energy. The entire process is associated with harvesting photon that is released as a result of spontaneous emission. Subsequently, two photons are released, and the continuity of chain reactions emits millions of photons. In this scenario, the emittance of photons is considered as stimulated radiation emission because the productivity of the irradiation wave is controlled using a certain mechanism compared to spontaneous emission which a natural process. Furthermore, the photons produced in the active medium between the two mirrors will bounce back and forth, reaching certain strength of spatial coherence before passing through the partially reflective mirror; the light produced within the active medium is mirrored several times between both mirrors.

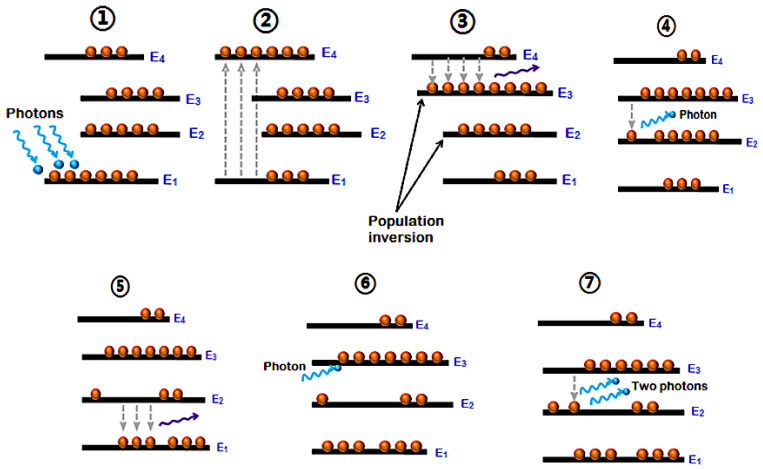

Fig. 2. Working Process of Nd: YAG laser (1) ground state receiving energy from source, (2) electrons at ground state moves up to pump state, (3), (4) electrons in the metastable state E3 fall into the lower energy state E2, (5) electrons in state E2 return to the ground state E1, (6) and (7) emission of two photons due to stimulated emission [23]

B. Fiber Laser

In a modern generation, fiber laser has become the best choice for most industrial applications as well for instrumentations. However, based on the operating principle, it has quite a several similarities except for the active medium made of an optical fiber doped with rare elements. For example, fiber is usually combined with Erbium, ytterbium, neodymium, thulium, praseodymium, holmium, or dyspro-

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sium. Moreover, the most important physical part of this system that is able to distinguish different types of laser which are mainly used of the active medium [20]. As described earlier that the Rare Earth Elements (REE) are used to dope the fiber before it is used as the central core; commonly the chosen REE would be Erbium because of the stability of its chemical compounding. Additionally, this rare element contains extremely useful energy valence layers, in which the high output of energy can be stimulated from a cheaper diode laser pump source. An example of doping fiber in Erbium is the energy level that can oscillate the photons with a 980 nm wavelength and decayed to a metastable equivalent of 1550 nm. In other words, with a laser pump source of 980 nm, high-quality laser irradiation plus high energy and high power of laser beam at 1550 nm can be regenerated. When the REE atoms serve as the laser core medium in the doped fiber, the emitted photons remain within the center of the fiber. Fibber Bragg Grating (FBG) is applied to create the cavity where the photons remain trapped, as shown in (Fig. 3).

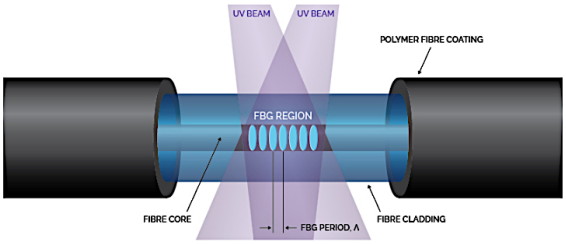

Fig. 3. Illustration of a Fiber Bragg Grating region in fiber laser [20]

Fig. 3 shows an illustration of a FBG region in a fiber laser. This FBG is a specific constructive section of glass that contains stripes, whereby the refractive index potentially changes. Therefore, whenever a light crosses the intended boundary between one refractive index and its neighbor, the energized ray refracts back a little amount of light energy. The Bragg grating was essentially making the laser fiber behave like a mirror. It becomes a significant feature in the fiber core because the core itself is too small and when a low-quality diode laser focuses on it, the pump laser is based on cladding all over the fiber core. The laser is bounced inside by injecting the laser into the cladding around the core, and each count it passes through the core, the core absorbs more and more of the pump light. The remaining sector of this laser is unchanged compared to Nd: YAG laser.

C. *Advantages of Fiber Laser*

At a glance, these two-laser types, which were discussed above, were widely used in the Laser color marking process to achieve superior laser properties, which are Nd: YAG, versus fiber laser. Previous work [24] has made a comprehensive study to show a comparison between these two types of laser with high power in terms of energy, power, beam efficiency, wavelength, maintenance, footprint, and proprietorship cost to study the performance of each type of laser. All the comparison factors are illustrated in Fig. 4. This Pugh matrix (decision-matrix method) (Fig. 4) shows the best characteristic sets for each type of laser which can be selected for the best Return of investment [25]. Compared to other types of laser, fiber laser has several advantages such as higher energy efficiency than a diode or lamp-pumped laser rod, simple installation due to a compact design, good quality of the beam, possibility to produce several kilowatts of power, and longer service life. All these advantages have led to low operating and ownership costs. Moreover, the cost of using fiber laser is typically half of what CO_2 can offer, this is due to lesser electrical consumption and higher electrical efficiency of the fiber laser. This type of laser is also equipped with a much more modern cooling system than the laser, which came from its invention such as CO_2 and Nd: YAG. This system helps to ensure the heat is kept low and uniformly distributed throughout the surface engineering process. Heat is also minimized from the fiber laser system so that the risk of machine damage is also at the minimum.

Fig. 4. Characteristic comparison between most commonly used high power lasers [25]

IV. LASER COLORING PROCESS

A. *Parameters Involved in Laser Color Marking*

The formation of the multicolor on the substrate surface of the selected material can be customized according to the design needs or graphical, pictorial base on the adjustment of the laser process parameter, which constitutes primarily the laser beam exposed magnitude and intensity. To research this behavior, various authors have been study-

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ing the effect of the obtained color by adjusting the process parameters made a suitable method to be used for this laser coloring processing. Therefore, most experimental researches agree that laser coloring is influenced by the lasering processing parameters. Besides the laser color processing parameters, the studies have proven that the method used to obtained color plays a significant role in obtaining a vivid color on the metal surfaces. The influence of the lasering method has been explained in detail in the following subsection. The details of the experimental work, including the use of process parameters, have been summarized in Table 1. It can be generally observed that fiber laser has been the most common choice adopted, with a power range of 5 to 20W. However, the pulse width used is commonly in the range of nanoseconds and frequency has not been greater than 500 HZ. The most critical parameters reported are laser power, scanning speed, frequency, pulse width, scanning direction, hatching distance, etc.

TABLE 1
LASER COLOR MARKING PROCESS AND A SUMMARY OF THE PARAMETERS INVOLVED, AND COLORS PRODUCED

Steel Series (REF)	Laser Type	Applications	Laser Power (W)	Laser Frequency (KHz)	Scanning Speed (mm/s)	Pulse Width (ns)	Beam Diameter	Significant Parameter	Color Produced
SS304L & SS316L [26]	Nanosecond-pulsed laser	Color marking and bar code	10	35	1-1000	10	-	Scanning speed	Grey, Yellow. Green and Brown
SS304L [27]	Nanosecond-pulsed laser	Automotive and biomedical industries	5W-8	225	100-600	119	-	Power and scanning speed	-
SS304L [8]	Pulsed fiber laser	Manufacturing industries for security improvement	20	20-100	10-500	100	-	Making velocity(mm/s), Frequency and laser beam power	Yellow, Blue, Brown and green
SS304L [10]	Q-switched third-harmonic Nd: YVO4	Product identification, enhancing styles and aesthetics	7W-10	40	400-500	25	30 micro meters	Laser power, focal plane offset, scanning direction and scanning speed	Green, Blue, Grey and Purple
SS304L [28]	MOPA Yb:glass fibre laser	Biomedical and Automotive industries	1W-10	20-100	100-300	100	-	Temperature, laser power, scan speed and marked area	Yellow, Red, Green, Blue, Purple, Silver and Black
SS304L [16]	Nanosecond-pulsed laser	Beauty product and jewellery	20	45-500	1000-3000	4-260	-	Scanning speed, Repetition rate, and Pulse width	Red, Green and Blue

B. The Effect of Laser Parameters on the Formation of Colors

A previous report [16] has studied the effect of the pulse frequency of the laser grating on the mark depth & width that contribute to the marked contrast. The result has shown that the marking depth and mark contrast depend on the interaction process of the laser beam (energy stimulation) and the material which was influenced by the pulse frequency. Fundamentally, the Laser Color marking process is one of the versatile techniques that can be adopted in the modern industry due to its ability to permanently stains the color onto the surface, and it can be one of the important processes used in industrial applications. The laser coloring process is more advantageous for the industry's usability to avoid the marking defect with the existing conventional method and it can be marked faster than the traditional method due to process finishing. However, to use the laser coloring method, the process parameters have to be precisely calibrated in the laser to avoid the mismatch in producing the color by the laser energy. Whereby this becomes one of the key factors leading to many studies on the process parameters which prove the consistency of regeneration of the color (basically it defines as R&R – Repeatability and Reproducibility). Another study [19] has covered the major influence of parameters on laser color marking where the authors concluded that the sum of the parameters such as laser power, pulse repetition rate, scanning speed, the spacing between successive lines, the temperature of the material, and location of sample relative focal plane affects the repeatability of the color obtained. The authors recommended the use of the optical spectrometer parameter to analyze the color changes based on their experimental work showing the effects of parameters on the colors as presented in Table 2 and Fig. 5.

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TABLE 2
THE PARAMETERS USED AND THE OBTAINED COLORS [?]

No	Color	Power (W)	Speed (mm/s)	Frequency (KHz)	Hatching (mm)
1	Yellow	3	100	80	0.02
2	Red	3.1	95	80	0.01
3	Green	3.2	75	80	0.01
4	Blue	3.1	125	80	0.01
5	Purple	5.0	150	80	0.01
6	Silver	7.0	75	100	0.05
7	Brown	8.0	50	80	0.01
8	Black	9.0	25	80	0.01

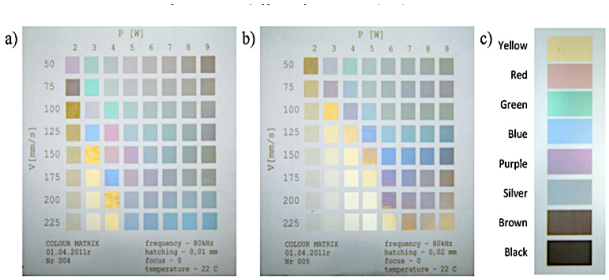

Fig. 5. The matric form analysis of produced colors [28]

Table 3 describes the experimental parameters used by the study to produce the colors [8]. The readjustment of the process parameters is the most challenging part of the experiment to repeat the required color due to laser process sensitivity in the laser color processing. However, the researchers [8] have managed to stabilize the reproduction of few colors to narrow down their analysis in the obtained color. From their finding and research, the authors have signified that the less impact on color variation is due to the temperature and oxide thickness. At the same time, the process parameters such as scanning speed, power, frequency, and focal plane are the major influencers on the reproducibility of the intended color. Knowing the degree of the repeatability of the laser coloring has been achieved successfully. A subsequent reliability test is necessary to prove the laser marking capabilities in a harsh environment. Thus, the material aging test towards the surface coloring, which is oxidized layer, has been conducted [28] to assess the environmental resistivity by limiting the chemical penetration over the thin film of oxide. Then the international std of Mil-Std-810G (810G covers accelerated corrosion using salt fog in Method 509.5), in a similar fashion the salt test was conducted to provide more evidence on the intrinsic characteristic of laser coloring. This test was conducted to observe the laser color marking sustainability on the metal surface after the process.

TABLE 3
THE PROCESS PARAMETERS AND THE COLORS OBTAINED [?]

Case	Laser Beam Power (W)	Marking Velocity (mm/s)	Frequency (KHz)	Obtained Color
1	2	10	40	Yellow
2	6	100	20	Blue
3	9	100	20	Brown
4	10	500	40	Green
5	10	80	80	Dark Red

C. *Physio Chemical Mechanism for Color Formation*

The subsequent study [8] has investigated the physical mechanism that occurs in the color formation of an oxide layer on the surface of the metal. The process parameters used to obtain the color has been shown in Table 3. Therefore, in this paper, the research has highlighted the effect of oxide growth with the number of lines passes through a surface of the process on the growth of the oxide layer. Fig. 6 shows the line passes for one and two passes in a single color where the oxide growth is more significant on the two-line passes.

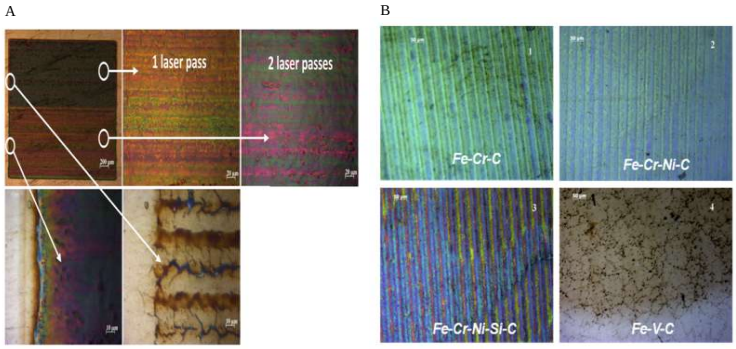

Fig. 6. a) The effect of laser number of passes on the metal surface and b) Chemical composition influence on the obtained color [8]

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To analyses the exact influential factor, researchers' have performed analysis using Energy Dispersion Spectroscopy (EDS) to retrieve the sample chemical composition on a non-treated and treated sample, Fig. 7. When comparing the results of non-treated and treated samples, the oxygen (KeV) has peaked in the treated sample, which confirms that an oxidation phenomenon has occurred in the result of the color obtained. EDS has shown that the treated sample at higher power has a higher oxygen peak [27]. This can be explained by the increase of laser power, which increases the temperature leading to a higher rate of oxidation, indicating the acceleration of the oxidation process. To nail-down the detail, a comparison was made for the oxygen intensity peaks for two samples treated with different laser powers. Indeed, it has been observed that the increase of temperature can facilitate the cation/anion diffusion through the metal-oxide interface [27]. Moreover, the study has shown that the obtained color on the metal surface by the laser process also depends on the treated material composition, as shown in Fig. 6. In conclusion, the authors have clearly defined different colors that can be produced by varying the process parameters.

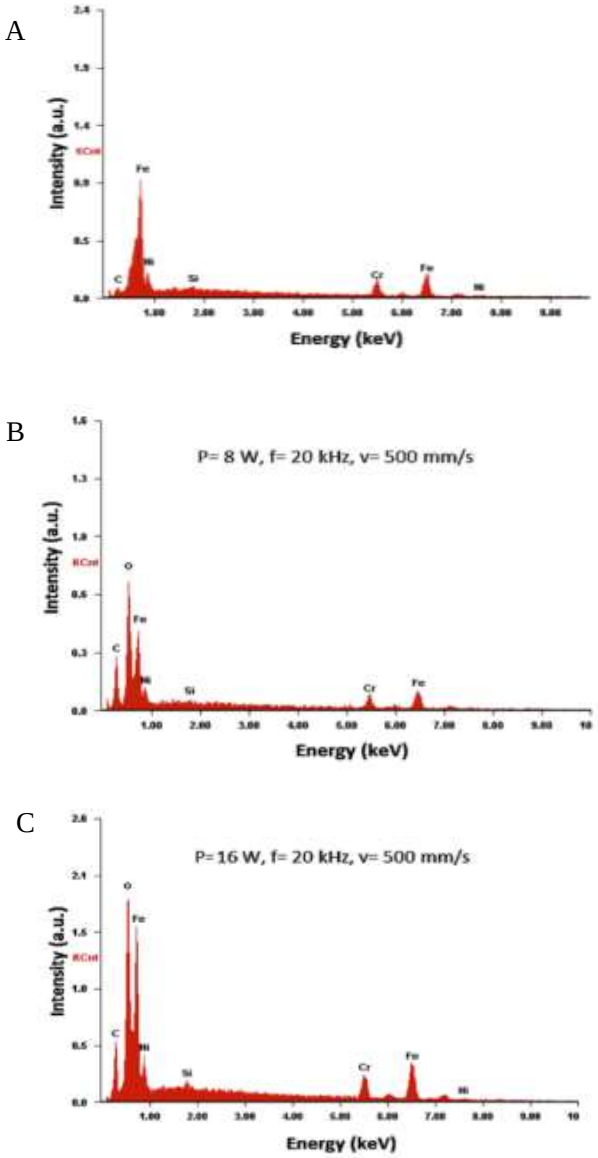

Fig. 7. The oxygen peak shown by EDS to a) non-treated sample, b and c) for treated sample with different parameters [27]

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Later the studies have extended to the next level; previous work [26] has investigated the thickness of the thin-film and the composition of the different oxide layers produced on the metal surface to determine the consequence of laser irradiation that restructures the substrate micro & macro lattice. Hence, the analysis has included TEM and EDS characterization, which showed the oxide layer thickness that emerged in their experiment was 400nm thick, which is the dark volume on the underlying substrate, Fig. 8(a). The individual grains have been noticed within the oxide layer, where the width of the grain is on the order of the oxide layer thickness. Meanwhile, Fig. 8(b) shows the three EDS representative spectra obtained from dissimilar regions of the specimen marked with different colors which have been shown in Fig. 7(a). The spectrum obtained in the bottom half of the thick oxide coating reveals a Cr-rich oxide solution containing Mn, where the top half of this coating is a Fe-rich oxide. Fig. 8(c) and (d) show the compositional gradients through the thickness of the regions. In summary, this study has performed TEM, EDS, and XDR analysis and concluded that laser irradiation and oxide growth had been significantly changed by composition and structure underlying the substrate.

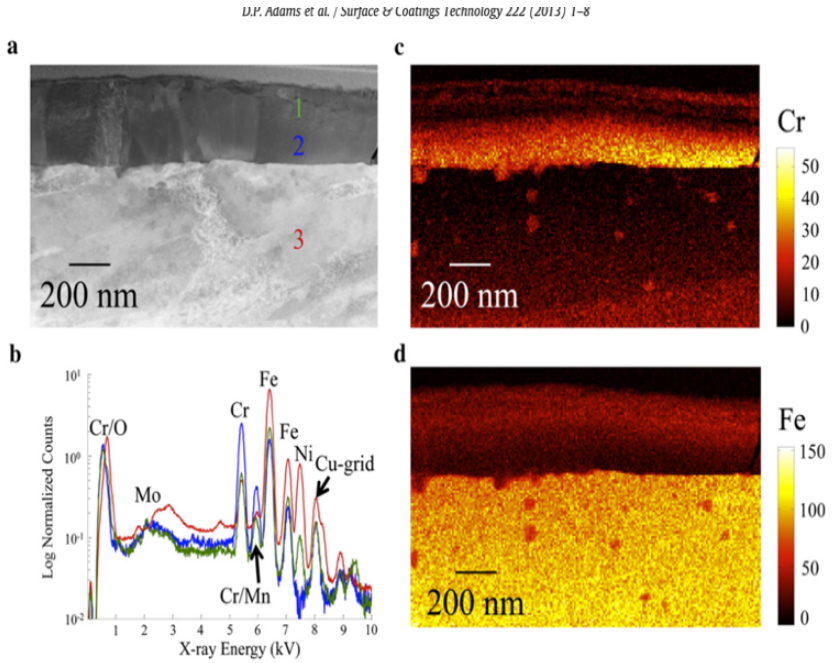

Fig. 8. Cross-section transmission electron micrographs showing (a) a portion of an oxide layer grown on SS 304L EDS spectra in (b) were obtained from three different points identified in the micrograph using colored numbers. Specifically, the green, blue and red spectra are taken from the top half of the oxide layer, the bottom half of the oxide layer and the underlying substrate, respectively. Images (c) and (d) map the Cr and Fe composition [27]

Liu et al. [10] have investigated the laser coloring process by varying the parameters with a different experimental method. The authors have proven that laser coloring processing is a unique technique for surface coloration. It offers clear benefits of high resolution with a smaller pitch and is capable of the single-step process. According to the laser coloring mechanism, three methods could be employed for the surface coloring, which are (i) laser-induced oxidation and thin-film effect, (ii) LIPSSs and their diffraction leading to the rise of iridescent colors and (iii) laser-induced nanostructure and nanoparticles to the high absorption at some wavelength due to the SPR effect [13].

Table 4 shows a comparison of the employed methods using different process parameters to control the color on the substrate. The surface oxidation method is based on creating an oxide layer on the surface to produce the color. This process occurs when the fast gaseous corrodes the surface with a high temperature. The oxide growth layer method involves five stages which are (1) heating of the metal surface by laser radiation, (2) reaction between the oxygen of air and metal, (3) the dissociation of the oxygen-oxygen liaisons and formation of ionic liaisons between oxygen-free

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electrons and the metallic ions (4) the oxide germs grow laterally which induces the metal oxide film on the lasering area and (5) the cation/anion diffusion at the interface of metal. Hence, the LIPSSs method is the composition approach for the color formation, which is described as colorations. This technique employs optical interaction such as light ray interference, light diffraction, and the scattering caused by micro/nanostructure. Fig. 9 shows the visible color by the angle of degree after employing the LIPSSs method processing to produce the color [10]. However, the nanoparticle and nanostructure method an exact realm of photonic crystallization structure and the multi-layer dielectric structure is obliged in femtoseconds constructiveness.

TABLE 4
COMPARISON BETWEEN THE EMPLOYED METHODS USING DIFFERENT PROCESS PARAMETERS [?]

Method	Substrates	Laser	Parameter
Surface oxidation	SS304L	Yb: glass fiber laser	1062 nm; 100 ns; 20–100 KHz; 50–225 mm/s; 10–50 μm
LIPSSs	SS304L	Yb:glass fiber laser	800 nm; 90 fs; 1 kHz; 1–4 mm/s; 50 μm
Nanoparticle and nanostructure	Aluminum	Ti:sapphire fs laser	800 nm; 65 fs; 100 KHz; 1 mm/s

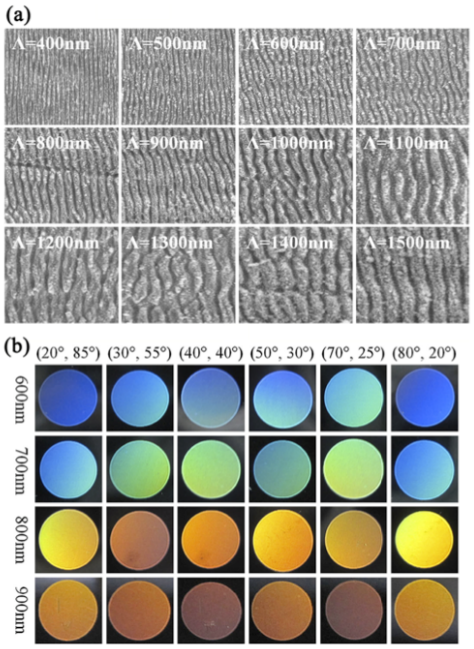

Fig. 9. (a) LIPSSs method with different spatial periods with dissimilar laser wavelengths and (b) Various iridescent colors on stainless steel sample with different periods 5,9].

Meanwhile, the author Ma et al. [17] have studied the effect of nanosecond pulsed laser parameters on the color marking of 304 stainless steel. In this research, the authors aimed to determine the surface morphology, the element of the composition, phase composition, and the film thickness of the marking color. The research has concluded that the most dominant parameters on the color marking process are the scanning speed and the hatching distance. However, by adjusting these two parameters, various colors have been marked during the experiment. Using optical microscope, they found six-mark color and found out that the surface of the color marking is cracked, the cracks are surrounded with red circles, as shown in Fig. 10. This is because the laser irradiation causes rapid heating with the environmental gas surrounding the laser beam cooling the material in a very short time.

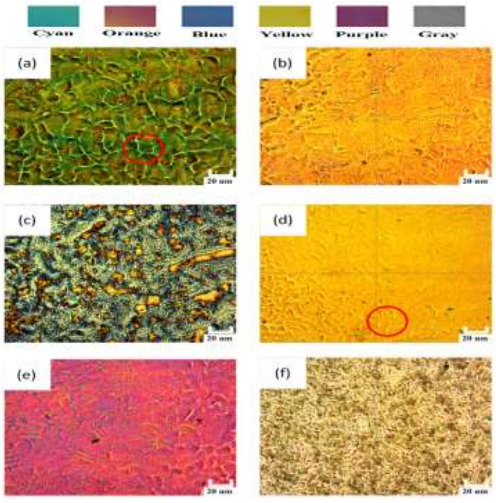

Fig. 10. Optical micrographs of six colors a) cyan, b) orange, c) blue, d) yellow, e) purple and d) gray [17]

Moreover, the research [17] analyzed the oxide surface morphology by using the FESEM for the same 6 six color mark surface, where the untreated sample hasshown no crack. However, for the surface microstructure after laser irradia-

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tion, it could be seen that the surfaces of the six colored samples are covered with ubiquitous, interconnected, through-thickness cracks, as shown in Fig. 11. The whole oxide layer is irregularly divided into many small pieces. Cracks appearing in thin films are thought to be a product of eliminating residual stress due to different thermal expansion coefficients between the substrate and the top thin film. All the six-sample surfaces have been cracking but the volume fraction of cracks on the surface varies from one sample to another. From the Fig. 11, it can be observed that the cyan and blue samples have fewer surface cracks and a larger size of clumps. Therefore, the crack in the yellow sample is more defined while the size of the fragments is very small. The authors have also stated that they figure out more cracking in the treated surface by ranging the hatching distance parameter. The microstructure of the orange surface is significantly cracking occur than the blue due to the significance difference in hatching distance of both which are 5 and 7 microns, respectively. As shown in Fig. 11 (d–f), the number of surface cracks increases with the escalation of the scanning speed (see Table 5). However, the study [17] has concluded that the continuous increase of scanning speed will lead to the decrease of pulse energy absorbed by the sample surface, resulting in very thin oxide film without color change on the sample surface.

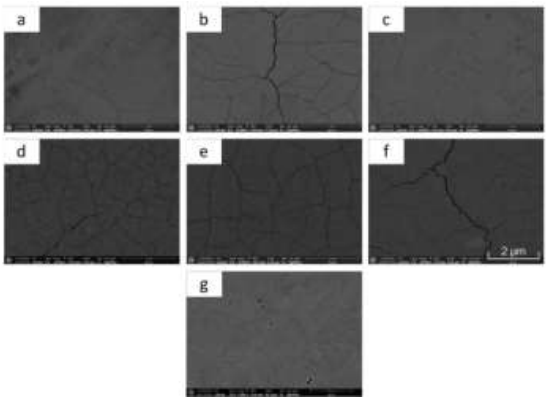

Fig. 11. FESEM images of different sample surfaces: (a) cyan (b) orange (c) blue (d) yellow (e) purple (f) gray and (g) an untreated sample [17]

TABLE 5
THE LASER COLOR MARKING PARAMETERS [?]

Color	Scanning Speed(mm/s)	Defocusing Distance(mm)	Hatch Distance(mm)	Repetition Frequency (kHz)	Pulse Width(ns)	Average Power(w)
Purple	350	4	10mm	225	15	15
Green	350	5	8	225	25	15
Gray	350	6	8	225	50	15
Blue	350	4	8	225	100	15

Finally, Garcell et al. [24] have studied the femtosecond laser-induced structural colorization in water and air. As most of the color marking research has been carried out on SS304l stainless steel in air, this study has conducted the color marking experiment in the water to see the colour's formation on the surface of copper. It has been concluded that colors created in liquid are formed by nanoparticle-induced plasmonic absorption and result in a range of colors transitioning from purple to orange, as shown in Fig. 12. Surface structures formed in liquid are less hierarchical and more uniform than those formed in air, producing a surface with a much higher reflectance due to reduced light trapping, resulting in more vibrant color. However, colorization formed in water suffers from less uniform colorization due to turbulence at the air-water and water-target interfaces, resulting in slight changes to the laser beams focus during processing. Moreover, varying the laser scan speed can successfully create four non-iridescent colors ranging from purple to orange using femtosecond laser ablation in water.

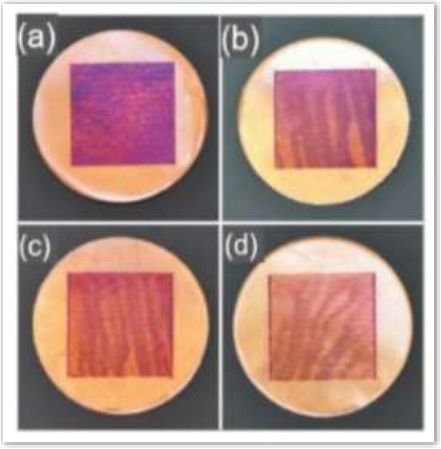

Fig. 12. Photographs of colored copper samples irradiated in double distilled water with laser scanning speeds (a) 0.5 (b) 1.0 (c) 1.5 and (d) 2.0 mm/s. [24]

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D. Three distinct physical mechanisms

1) Thin film: This cutting-edge technology of laser color marking has been started with the technique of surface oxide layer causing the i) thin film interference effect, ii) (LIPSSs) as an acronym standing for laser-induced periodic surface structures and iii) finally plasmonic structure form from nanoparticles and nanostructures enable to produce vibrant colors.

Generally, when the laser strikes an area, it induces oxidation in which laser energy creates a transparent or semi-transparent thin film. This well-known phenomenon efficiently induces a reaction with the exposed substrate under the assist of laser heating. The thickness of the oxide layer has a unique characteristic to disintegrate the incoming white light spectral into multicolor illumination. This phenomenon happens when the light ray reflects a different layer of the oxide film at the top and bottom surfaces. The constructive interference is due to varying angles of refractive blending the light waves into the intended colors. This effect is mainly determined by the oxide layer thickness and its respective refractive index [14]. In certain studies, [14, 29], it has been found the metal oxides had an intrinsic appearance of colors giving an additional color rendering as well. Thus, the control of oxide film formation by laser processing technique on sample surfaces generate permanent colors. An extensive study [30] has been conducted on this technique to understand how the color is made by numerous experiments involving many types of metals and alloys subjected to a variety of laser sources. Stainless steel has become a novel alloy that researchers have widely studied. Therefore, in laser colorization, various types and grades of stainless steels have been used, for instance, 304, 304L, 316, 316L, and even home-made stainless steel, which specific compositions [30]. Each grade of these alloys had a different tendency for surface oxidation which is governed by a technique in lasering. Then the conservative of energy with the intended proportions are induced oxide films which are essential to maintain the exposed surface performance. Thus, a systematic study [29] has been conducted to quantify and analyze the formation of the oxide layer and the influence of processing parameters to generate multiple colors on the selected alloy. It was clear that laser process parameters can generate a set of color gradients that are displayed in a pallet form. Having controllable growth of the oxide layer could be achieved by beaming in raster scanning with nanosecond, using a variant of the line spacing and different scanning speeds to produce accruable intended colors [29]. So, the oxide film structure and its substrate composition reveal how the laser color marking works well in a certain physical mechanism. Multiple studies highlight a broader range of the color spectrum that has been achieved in alloys and metals. Varying speeds of scanning and laser power gives series of colors. However, a certain range of colors cannot be achieved in some selected metals and alloys but the intensity of the laser would be able to colorize oxide substrates whereby thermal energy bleaches the natural color of this amorphous film [10].

Fundamentally, the metal oxidation is induced by laser at high temperature and fast gaseous reactions. This process has been described in 5 stages [10], especially the oxide layer growth. The process begins with generating heat energy by laser irradiation on the metal surface. When the metal or alloy surface is exposed to oxygen within the ambient environment's laser irradiation given excessive energy to the electron, which it allows too excited from the lower to higher energy layer in valence [31]. Then, to stabilize the natural state, the energized electron will rapidly decay and the excess energy will be released to lattice whereby the irradiated area will be subjected to temperature incremental. When the metal and oxygen are in contact with the heat energy, the chemical reaction occurs. This state is called oxidation, whereby the process initiates the bonding between oxygen and substrate surfaces. During the chemical reaction, the stable oxygen atomic bond will be dissociated to form an ionic state which allows the free electrons of metal to bond together to form an oxide germ. Subsequently, the metal oxide layer grows laterally until the laser irradiation is exposed to a localized area till a certain fragment of time [32]. The continuous diffusion of cation/anion on top of oxide results in perpendicular growth of the thin film. From the thermodynamic principle on the kinetic coefficient, the thermal reaction results in laser oxidation, but in the real condition, the thermal energy causes many complex factors such as no isothermal oxidation. The nonlinear process leads to nonequilibrium circumstances. Quite complicated the overall process of forming the laser oxidation, the slow rate of chemical reactive compared to rapid incremental of temperature due to nano pulse. Therefore, it is hard to describe the formation of a thin-film as a universal chemical principle since the oxidation kinetic dominating the metal reaction in comparison to the isothermal process. However, there is a possibility of mapping the correlation between the selected metal and the oxide layer through experimentation [20, 21]. Therefore, the quantitative analysis has been conducted by tuning the laser parameter to characterize the oxidation layer characteristics such as thickness, surface crystallinity, and chemical bonding. This de-

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tailed characterization was used for different incremental of laser to understand the fluences on metal substrates. It has been proved that oxidation kinetic directly impacts laser parameters such as wavelength, pulse-width repetition- rate, scanning speed, and line spacing.

The optical illumination could be illustrated by two main working principles or mechanisms after forming the thin-film oxide layer. The first effect is based on optical interference due to the thin film [33]. As shown in Fig. 13, when the incoming white ray strikes the oxide thin film, the penetrable light source tends to reflect at two boundaries: i) air and oxide ii) oxide and metal. This incident causes the reflective beam to split into two different optical paths (δ) whereby the following formula expresses the reflective beam differences [10]:

$$\delta = 2\,\mathrm{h}\sqrt{\mathrm{n21} - \sin 2\theta\mathrm{i}} \quad (1)$$

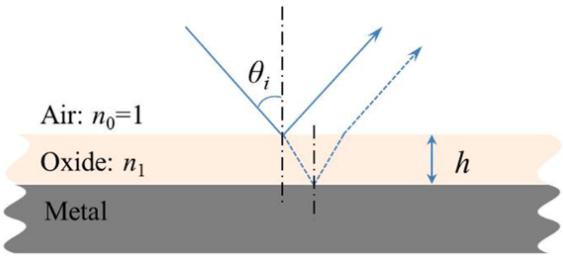

Fig. 13. Schematic diagram of the interference effect of the oxide film. n0 and n1 are the refraction indices of air and oxide, respectively [10]

Each of these factors belongs to the oxide thin film h, n1, and θi based on the thickness and refractive index of the oxide film and the incidence angle, respectively. Equation 1 indicates that the multitude of color reproduction is propositional to three factors, mainly the formation of thin-film quality and type of oxidation which would define the i) refractive index, ii)visible angle, and iii)degree of light interferences [34]. The formulation is further simplified in terms of wavelength δ = kλ (k is an integer); it was very straightforward to further understand the behavior of constructive interferences [12, 24]. It has been observed from the experiment exercise [10] when the viewing direction has been changed for the refractive light the laser induces marking will vary in color which changes according to the angle. Moreover, the change was not limited to the reflectance of light spectrum color brightness but also color hue which is the dominant wavelength and saturation or chroma which would have shifted as the increase of incident angle [35].

In the second laser coloring mechanism, the thin film metal oxidizes itself will be colorized by laser energy due to the formation of chemical compositions. Thus, the appearance of metal oxide color was blended based on final substrates surface discoloration and the opaqueness of oxidizing film, which means the level of transparencies of the thin layer [36]. It shows that there are strong correlations between a thin film of oxidized metal compositions and its surface colorfulness based on substrates due to spinel compounds which is presented as pigmentation [19, 27]. These interactions become natural behavior that helps to distinguish colors that mark through the laser process. Therefore, when these two laser coloring principles combine, it results in a formation of desirable colors, which could be predicted by tune important factors and processing parameters.

2) Laser Induced Periodic Surface Structure (LIPPSS): Several compositions could approach the color formation which is described as structural colorations. This technique is predominated by optical interaction such as light ray interferences, light diffraction, and the scattering caused by micro/nanostructure rather than chemically pre-process dyes or pigments. Thus, in laser processing, there is a unique approach called LIPSSs that could be performed to induce structural colors [10, 13]. The acronym of LIPSSs could be explained as laser-induced solid etching. Whereby the characterization focuses on how the periodic structure surface could be nanograting when the induced laser beam is exposed in a femtosecond. In the past decades, this intrinsic feature was found on semiconductor material which was discovered by Birnbaum. Then, the search was evolved to include various conductor materials, dielectrics as well on semiconductors [37]. It was fascinating among researchers that the one-dimensional grating or grooving structures produced by a laser beam or irradiation with the influence of frequency and wavelength at different scales could regenerate various ranges of colors on metal substrates [38]. Still, in the LIPSSs technique, the color generation was highly dependable on viewing angle which turns up as one of the primary factors for interferences of the light ray. Moreover, the LIPSSs process forms a surface ripple during nanograting which has a much lesser spatial-period than the actual laser wavelength. Furthermore, the process of engraving the parallel lines is highly dependent on laser polarization and the orientation of the periodic lattice structure to be perpendicular [39]. When this two-mechanism is re-oriented in such an optimistic face angle, it is possible to control laser marking with the proper selection of beam polarization. Based on the type of material selection, a unique grating or ripple of nanostructure will produce the array pattern which influences the multiple color creations as the structural laser coloring have a high tendency in predefining the

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metal colorization by tweaking the specific laser parameters and the laser processing parameters [40]. It has been expanded to observe the influence of the groove shape, including length and depth, and the narrow scribbling pattern created through the irradiated laser beam. In the normal condition, the pattern present in one to two-dimensional high-density arrays with nanoholes simulates the reproduction of a broader range of visible light spectrum with having vivid colors [10].

The outcome of LIPSSs had a systematic controllable self-organized laser marking. The formation of wavy ripples on the surface indicates a strong interaction mechanism between laser light parameters against surface lattice [32, 41]. It means that LIPSSs have time coherency close to irradiate wavelength and angle at 90° to the polarization. In the past, it has been theorized when the irradiate wave bounces back from the substrate, by following the pattern or the roughness of the surface, this wave becomes a singular source to provide the integrity to be coupled with incident laser light [29, 31]. In a flat surface with no-surface roughness, it is regarded as an identical reflectance angle opposite to incident light whereby no additional substance called fragmental of laser light is superimposed. However, when the surface has random roughness, a high potential scatters wave, which is the remnant of the earlier laser beam, will interfere in the direction of incident laser light. This incident is known as "coupling", and it was described as a wave vector in the Fourier series by having surface roughness "g" to represent harmonic components [12][26] . Equation 2 represents the total vector functions:

$$Kip \pm mg = Ksp. \quad (m = 1, 2, 3) \tag{2}$$

This equation satisfies energy conservation law, whereby Kip is the vector components to the incident light, and Ksp is the wave vector to the surface wave in which both vectors are parallel to the substrate surface. This wave-vector relation is shown in the geometrical form in the equation below: The given mathematical model further reduces when m = 0 the interference pattern Λ= 2π/|g|, i.e., the period of the LIPSS can be derived;

$$\Lambda = \frac{\lambda}{(n \pm \sin\theta i)} \tag{3}$$

whereby λ = laser wavelength,

θi = laser striking angle

n = refractive index of material

This correlation was examined using continuous wave (CW); the laser pulse has oscillated at a range of nano to pico-second. The derivative of the present mathematical model has a significant challenge in the advancement of laser technology because a more complex pattern has been produced using femtosecond when using different material compositions. Moreover, when the overall system operates with femtosecond to produce nanograting, its influence of laser pulse with a larger wavelength (nanosecond) could not explain well by referencing Equation 3 because at that point of time the irradiation energy had a dependency on the laser fluence. To further explore the behavior of quantum energy and its manipulation, the regime of surface plasmon, which is called plasmon polarization (SPPs) is introduced [12]. This will be explained in detail in the subsequent sections.

3) Plasmon color by nanoparticle and nanostructure: In the exact realm of the existence of photonic crystallization structure and the multi-layer dielectric structure only obliged in femtosecond constructiveness [12, 13]. Naturally, the plasmons polarization contributes to the color generation by the visible light interferences when the light source passes through the miniature structure. This phenomenon could be explained via two natural creatures, morpho butterfly and beetle [33, 34]. Their wings have a multi-layer of cuticle and air passes restructure for the iridescent of vivid colors. The dielectric nanostructure is made on a micro-scale which is much larger than the diffractive limit. However, the artificial dielectric metamaterial and met surfaces tend to mimic the similar natural appearances of structured colors when the microelements are smaller than the spatial wavelength of striking light [42]. This proves that the metallic nanostructure can absorb resonant energy and scatter the waves across the lattice. Therefore, the surface plasmons have many free electrons that oscillate and travel between the interface valence of metal and dielectric material. The polar energy on the surface of substrates could be manipulated when a light beam with the selective wavelength strikes the quantum layer. Hence the electrical field intensifies the free electron on metal which will resonate to reach the peak of oscillation, creating the valley of the optical absorption [35, 36]. At this point, the system resonate would be addressed as (SPR-surface plasmon resonance). Therefore, it is understandable that the incident light energy can be conjugated with surface atomic structure due to SPP-surface plasmon polarization via nano-roughness diffraction. The wave interferences modify the spatial modulation resulting in the new pattern of the periodic structure. Thus, the interference pattern incorporates dielectric function [10].

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which gives as below:

$$\Lambda = \frac{\lambda}{\mathrm{Re}[\eta] \pm \sin\theta i} \tag{4}$$

where the refractive index has been given as:

$$\eta : \left[\varepsilon_d \varepsilon_{\text{metal}} / \left(\varepsilon_d + \varepsilon_{\text{metal}}\right)\right]^{1/2}$$

ε_d: Dielectric constant of the ambient medium

$\varepsilon_{\text{metal}}$: Dielectric constant of metal

In the current modern advancement of instrumental efficiency, there is an enhancement version of LIPSS, which is often described as high spatial frequency because the period is much lesser than beam wavelength irradiation [43]. Therefore, the polarization model created by frequencies interference forms a nanostructure that has a period equal to or fractions of laser wavelength (λ/2m, m = 0, 1, 2, ...). Additionally, when high spatial frequency LIPSSs distorts the entire spinel compound microlayer, the behavior can be only explained using second-harmonic generation. This process could explain the manner of photon energy retention, likewise, when the initial laser pulse strikes at once the substrate surface nanostructure, it will be a little disordered. Then, the continuum of pulses focuses on the same locus agitate the molecular energy for the self-organized polarization [44, 45, 46]. At this point, the irradiated area bound to be coupled both energies formed the spatial modulation, which is called a nascent periodic structure. This relation is given in Equation 4, whereby influential of laser wavelength, the beam striking angle as well the effective optical refractive ratio equated in the formulation. It proves varying the effective refractive index, which intertwines with dielectric constant and laser fluence. Therefore the tunable wavelength at different periods produce a range of various iridescent controllable colors at a different angle of views [37, 47].

V. CONCLUSION

The current shift towards product miniaturization is expediting the intensification of manufacturing processes. It is highly anticipated that the technology is drifting towards photon driven manufacturing systems. Concerning light beam-manufacturing technologies, laser color marking on the steel material has been sporadically so far. Thus, the inherent sensitivity of laser processing demands precision in controlling the parameters of the laser in the formation of color on the metal surface. For instance, the material that is reactive and sensitive to oxidation or cracking requires proper selection of the parameters such as laser power, frequency, hatching distance, defocusing distance and pulse width for this process. Hence, the challenge lies in obtaining a precise parameter that is suitable and compatible to form a solid color on the metal surface. The following conclusion can be drawn for laser color marking of metal surface;

1. Laser color marking is a single step process compared to traditional marking. It is a safe and fast process in color marking.
2. The defects in the microstructure, including laser-induced porosity in the heated area of the metal surface, can contribute to the accumulation of stress raise that accelerates the initiation of cracks.
3. Laser color marking on the metal surface remains an engineering challenge, and it can have non-solid color due to the processing parameters, where the process parameters have to be precisely adjusted to achieve a solid color using this process.

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