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Progress and Prospects of Chromium-Based *p*-Type TCOs: Next-Generation Transparent Electronics

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Abstract

The field of active devices has recently shown a great interest in p-type transparent conducting oxides (TCOs). In the realm of optoelectronics, a new generation known as "Invisible Electronics" began with the introduction of transparent junctional devices. In recent years, many different p-type transparent conducting oxide materials with enhanced optical and electrical properties have been produced. These p-TCOs are not stoichiometric and are doped with suitable counter-cations/anions. For p-TCOs, chromium (Cr)-based materials stand out as the best option among all previously employed p-type materials. Despite their electrical properties lie on the insulator side, appropriate doping has been shown to greatly increase their electrical conductivity. In this review, an up-to-date and detailed explanation of TCO materials based on chromium (Cr) is provided. The development of highly efficient p-TCO films could lead in an innovative era in the form of "Transparent Electronics", as it could pave the way for the fabrication of transparent active devices.

Keywords: Optoelectronics, TCO, chromium, *p*-type, invisible electronics

INTRODUCTION

It was in the form of CdO that Badekar created the first TCO in 1907, thereby marking the beginning of TCO [1]. TCOs find widespread use in many different kinds of optoelectronic devices. But due to a scarcity of higher performing *p*-type TCOs, active devices (e.g., bipolar diodes and transistors) are in short supply. In order to create functional electronic devices, *p-n* junctions are required. However, *n*-type TCOs predominate in the commercial supply. Therefore, only unipolar devices can be realized in optoelectronics applications [2, 3]. The band structure of *p*-type TCOs is a problem because of oxygen's strong electronegativity. This causes the battle to generate holes with low effective masses and a shallow acceptor level. There are also ongoing efforts to create transparent *p-n* homojunction by introducing acceptor level in existing *n*-type TCOs [4]. There is currently no known TCO material capable of demonstrating ambipolarity. Many attempts have been made, but all have failed, to change the *n*-type conductivity of materials like ZnO and SnO_2 to *p*-type conduction [5]. According to the results of a DFT study, oxygen vacancies and cation interstitials are two examples of intrinsic defects that can remove *p*-type dopants by acting as donor levels. Considering the band structure, theoretical studies show that the requirements for *p*-type and *n*-type dopability are in direct opposition to one another.

Transparent *p-n* junctions with more intricate circuits will be feasible once similar *p*-type TCOs are commercially available for production. For instance, silicon technology allows for the creation of optical complementary metal-oxide semiconductor (CMOS) devices with comparably complex circuitry. Studies of highly efficient *p*-type TCOs consequently formed an important topic of

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"invisible electronics" research. The unique optical and electrical properties of transition metal oxides (TMOs) have garnered a lot of interest in comparison to *p*-type TCO materials. These characteristics can be modified for use in optoelectronics by adding the appropriate dopants [6]. Extensive research has been conducted to improve the properties of valuable Cr-based materials and add new useful capabilities to them. The optical and electrical properties of Cr-based nanomaterials are amenable to tuning via the substitution of a suitable dopant. Consequently, a wide range of dopants has been employed to tailor and enhance the electrical properties of chromium-based materials [1].

The present study offers a comprehensive review of the current understanding of the optoelectronic performance of chromium-based oxide materials, highlighting the key challenges encountered by researchers in the development of high-performance *p*-type transparent conducting oxides (TCOs).

STRUGGLE FOR DEVELOPING *P*-TYPE TCO

P-type TCOs, which have a different valence band structure, cannot compete with *n*-type TCOs in terms of performance because the conduction band minimum (CBM) in *n*-type oxides is dominated by metal ns-orbitals. In addition, the s-orbital is well-known to be highly delocalized and hybridize adequately even in amorphous crystal formations. Electrons benefit from a low effective mass and high mobility because to the CBM's highly delocalized or dispersed nature. On the other hand, *p*-type TCOs do not conduct electricity for the reasons given below:

a. Holes are limited in their production due to the high formation energy of intrinsic acceptors and cation vacancies (V_c).
b. Donors that are already present and have low formation energies, such as oxygen vacancies (V_o).
c. Holes' effective mass is increased, and their mobility is decreased in the VBM due to oxygen 2p orbitals.
d. The low VBM energy level and restricted dispersion of the valence band make hole doping undesirable [7].

These problems highlight how difficult it is to develop very efficient *p*-type TCOs. Chemical manipulation of the valence band (CMVB) was first proposed in 1997 by Kawazoe *et al*. [8], as illustrated in Figure 1. This theory proposes that the conductivity of *p*-type TCOs can be enhanced by incorporating suitable countercations whose energy levels are comparable to or higher than those of the oxygen 2p orbitals. By forming covalent bonds between the oxygen 2p orbitals and the cation's s, p, or d orbitals, greater delocalization of charge carriers can be achieved, thereby improving the material's overall conductivity. As the VBM broadens, the charge carriers' effective mass decreases, and holes' mobility rises.

The effective *p*-type TCO in the structure of $CuAlO_2$ was developed by Kawazoe *et al.*, marking a major step forward in our understanding of *p*-type TCOs. The optical band gap of this material increases to 3.1 eV [8], and its conductivity increases to 1 S/cm. While this material has improved in terms of electrical conductivity, it still falls short of the efficiency of *n*-type TCOs. Improving *p*-type TCOs may seem like an insurmountable issue, but this work shows that it is not. A number of *p*-type TCOs followed, allowing for the creation of cutting-edge "invisible electronics" devices that are difficult to produce with traditional Si technology.

$SrCu_2O_2$ and $CuAO_2$ (where A=Mg, Al, Cr, Ga, In, Sc, and Y) are among several oxide materials recognized as *p*-type transparent conducting oxides (TCOs), attributed to their degenerately doped atomic structures. The successful development of p-type TCOs has confirmed the feasibility of fabricating transparent p-n junctions. Following extensive investigations into Cu-based p-type TCOs, the concept of a chemically modified valence band (CMVB) was extended to chalcogen-based materials. In this approach, chalcogen elements such as sulfur (S), selenium (Se), and tellurium (Te) were proposed as substitutes for oxygen to further enhance *p*-type conductivity. As a result, hybridization between chalcogen *p*-orbitals and metal *d*-orbitals will occur. Candidates for p-type TCOs include layered LaCuOS, LaCuOSe, and LaCuOTe [5].

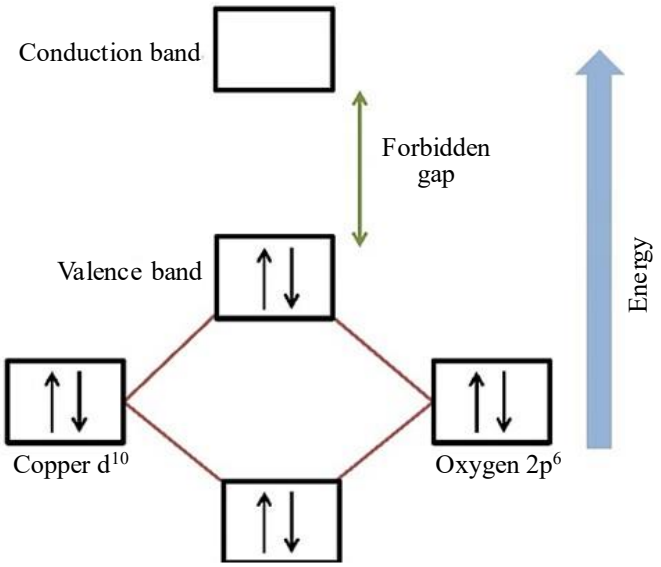

Figure 1. Illustration of CMVB technique [8].

In addition, the highest p-type conductivity (910 S/cm) has been reported in Mg-doped LaCuOS [5]. A 2.8 eV optical band gap limits the material's transparency to light. Materials having quasi-closed shell topologies, such as d^6 and d^3, have recently seen use of the CMVB method. As a result, new TCO materials can be created, such as zinc oxide (ZnO) and chromium oxide (Cr_2O_3) [1].

CONDITIONS TO SELECT *P*-TYPE TCOS

Most of the CBM in *n*-type TCOs is made up of s-orbitals of metal atoms, which are spread out in different locations. Very high conduction is feasible due to the low effective mass of electrons and the high electron doping density. An *n*-type TCO often has high electrical conductivity like this. However, in *p*-type oxides, the valence band maximum (VBM) is predominantly composed of localized oxygen 2p orbitals, which significantly restrict hole mobility and result in a larger effective mass of holes. *P*-type TCOs have a serious problem with positive hole localization at the oxide's VBM. This localization is caused by the highly electronegative oxygen and an energy level well below the VBM of metallic atoms [8]. The following considerations should be made during the design of p-type TCOs if high visible-spectrum transparency is a design goal:

1. It needs to have an optical band gap energy greater than 3 eV.
2. Select a host material where the orbital energy of the host cation is larger than or equal to that of the oxygen 2p energy level.
3. Introduction of an appropriate countercation so that VBM is dispersed, and the holes' mobility is increased.
4. It was previously believed that host-material cations or countercations required closed-shell electronic structures to prevent coloring. However, more recent studies have shown that the optical transparency is not significantly affected by the inherent electronic transition generated by partially filled d-orbitals. As a result, oxides that are either partially or completely filled electronic structure, can be employed as a host material or dopant. Materials based on chromium are a good illustration of this.
5. Improving a material's electrical properties also rely on the coordination number of its crystal structure. Materials with high hole conductivity benefit from tetrahedral coordination because it can disrupt localization around oxygen anions. The oxidizing anions in this mixture are sp^3 hybridized. Oxygen anions have less localization at the VBM. For example, Cu_2O has high *p*-type conductivity because of its tetrahedral coordination and the fact that the orbital energy of the copper atom is greater than that of the oxygen 2p level [9].

ROLE OF BURSTEIN-MOSS (BM) SHIFT

When a material is degenerately doped with a suitable countercation, the excess charge carriers fill the lower energy states in the conduction band, thereby shifting the Fermi level to a higher energy position. In such cases, electrons can only be excited to higher energy levels if they possess enough energy to overcome the filled states at the bottom of the conduction band and occupy the higher unfilled states. This results in an apparent widening of the optical band gap, which is beneficial for the material's optical properties. This phenomenon is referred to as the Burstein-Moss (BM) shift, and its magnitude (ΔE_{BM}) depends on the carrier concentration (n) and the effective mass of the charge carriers (m^*).

In heavily doped transparent materials, a significant increase in carrier concentration can cause the plasma edge to shift toward the red region of the visible spectrum, thereby impacting optical transparency. Moreover, once the lowest conduction band states are fully occupied, electrons at the elevated Fermi level (E_{FL}) may possess sufficient energy to transition into the next available higher conduction band. The energy separation between these two conduction bands is referred to as the "secondary band gap" (E_{sg}), as depicted in Figure 2. A large secondary band gap (>3.1 eV) is essential for TCO materials to suppress intra-band optical transitions and maintain high levels of transparency.

A comparable mechanism governs *p*-type TCOs, wherein holes serve as the dominant charge carriers instead of electrons. This interdependence of structural, electronic, and optical properties indicates that achieving all desirable features in a single TCO material is extremely challenging. Among all these factors, the nature of the charge carrier (*p*-type or *n*-type) is the most fundamental and difficult to alter. Although devices utilizing a single carrier type can exhibit fundamental functionalities, the integration of both *p*-type and *n*-type materials is essential for achieving the optimal performance demanded by most modern electronic and optoelectronic applications.

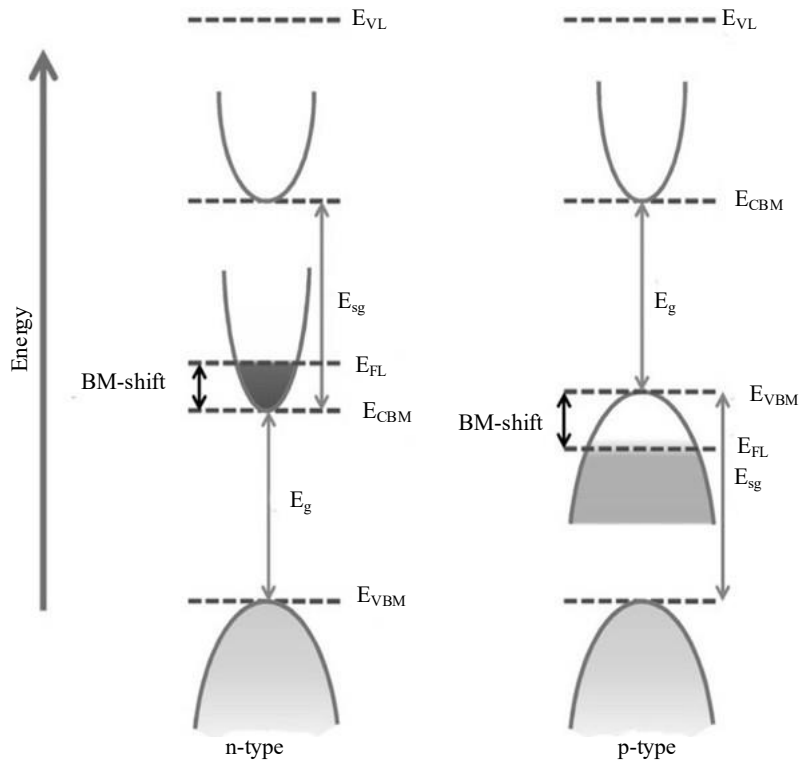

Figure 2. Schematic illustration of the band structures for degenerately doped n-type and p-type materials, emphasizing critical features such as the Burstein–Moss (BM) shift, Fermi level (E_{FL}), vacuum level (E_{VL}), fundamental band gap (E_g), and secondary band gap (E_{sg}). The dark-shaded regions denote occupied electronic states, whereas the unshaded regions represent unoccupied energy levels, as adapted from [5].

$CuCrO_2$ MATERIAL

Nagarajan *et al*. investigated $CuCr_{1-x}Mg_xO_2$ material with a high hole conductivity value [10]. Both bulk and thin films of $CuCr_{1-x}Mg_xO_2$ materials have been manufactured. Bulk undoped $CuCrO_2$ was discovered to be a black solid with decent conductivity. In addition, the conductivity of an undoped $CuCrO_2$ thin film was ~0.1 S/cm. The addition of 5% Mg to $CuCrO_2$ boosted its conductivity to a factor of 10^3, as depicted in Table 1. The conductivity of this was unparalleled by any other *p*-type TCO material available at the time. Although its thickness was only 250 nm, the optical transmittance was only 30% in the region of visible spectrum. Additionally, post-annealing took place at a variety of temperatures for the thin film. Crystallinity, conductivity, and transparency all remain unchanged up to 600ºC. However, at 900ºC, both transmittance and resistance increased. Adding oxygen through intercalation to pre-existing thin films also failed to produce the desired results. Thermogravimetric analysis (TGA) showed that up to 1200ºC, the $CuCrO_2$ combination did not absorb any oxygen due to its small lattice volume. At this temperature, $CuCr_2O_4$ is formed when oxygen is consumed. Spray pyrolysis was used by Farrell *et al*. to create Cu-deficient $CuCrO_2$ thin film in 2015 [11]. They achieved maximal conductivity and a decent transparency, enlisted in Table 1. In an experiment conducted by Ahmadi *et al*. [12], Mg and N were co-doped into $CuCrO_2$ thin films. In order to create thin films of $CuCrO_2$ doped with Mg and N on a quartz substrate, radio frequency (RF) sputtering was used. Electrical conductivity increases dramatically at 2.5% of Mg, N doping into $CuCrO_2$, along with transparency of 69.1% in the visible range. Thin films of co-doped $CuCrO_2$ also have a significant carrier concentration (1.18×10^{21} cm^{-3}) and the lowest hall mobility (0.006 cm^2/V-s) of any known material.

Cr_2O_3 MATERIAL

Recently, there has been a lot of focus on chromium oxide (Cr_2O_3) *p*-type TCO. Cr_2O_3 is *p*-type because of the low formation energy of Cr-vacancies (V_{Cr}). Cr_2O_3 has an energy of 3.1 eV for its optical band gap. Two more kinks occurred at 2 and 2.6 eV [6] with the dipole-prohibited d-d transition. Transparency in the visible spectrum is not significantly affected by these transitions in thin films. Even though stoichiometric Cr_2O_3 is an insulator, it was demonstrated that with the right dopants, the material's conductivity may be significantly improved. A study investigated the possibility that dopants like Li, Mg, and Ni could improve conductivity [13]. Spin coating technique was employed to grow doped Cr_2O_3 thin films. They have not, however, provided accurate conductivity or resistivity values. They simply showed the temperature-resistance relationship and discovered that Ni, Mg, and Li-doping reduces resistance. In Li-doped materials, the reduction in resistance was negligible. However, it was found that Mg-doping increased the absorption because the addition of Mg-dopant leads to the formation of ions with mixed valences (Cr^{3+} and Cr^{6+}). Arca *et al*. conducted the studies on the (Mg, N)-doped Cr_2O_3 thin films [14]. To stop the creation of the Cr mixed-valence state, researchers have also tried co-doping with magnesium and nitrogen. Using a spray pyrolysis method, they created thin films of N-doped Mg and Cr_2O_3. The optical transparency of undoped and Mg-doped Cr_2O_3 thin films was somewhat typical for a 150 nm thin film due to the poor film quality. The greatest transmittance was reported to be 65% in the NIR region, however transparency improved with Mg and N co-doping. But the electrical resistance went down from 400 to just 15 Ω-cm. The best outcomes were achieved at Cr:Mg ratio of 9:1 and Cr:N ratio of 1:4. In this research, 3 Ω-cm was the lowest resistivity measured. All thin films were *p*-type, with Hall mobilities of 0.1 cm^2/V-s and carrier concentrations of 10^{19} cm^{-3}.

Table 1. $CuCrO_2$ material as p-type TCO with various dopants, deposition techniques and optoelectronic parameters.

Host material	Dopant used	Growth technique	Optical transmission (%)	Band gap (eV)	Electrical conductivity, σ (S/cm)	Ref.
$CuCrO_2$	Mg	RF sputtering	30–40	3.1	220	[10]
$CuCrO_2$	Cu-deficient	Spray Pyrolysis	55	-	12	[11]
$CuCrO_2$	Mg and N	RF sputtering	69.1	3.25	277.7	[12]

This investigation was expanded in the same group. Spray pyrolysis has been used to generate thin films of Mg and N-doped Cr_2O_3 [14], and the effects of various chemical precursors on their optoelectronic properties have been investigated. Producing thin films with substantial resistance can be done with either chloride or acetate, however the resistivity values obtained with nitrate at pH 0 are the lowest (4 Ω-cm). It has been hypothesized that low pH values promote the development of highly reactive molecules like NO and NO_2. A film with minimal resistivity at neutral pH was the result. Nitrogen addition decreased the concentration of Cr^{6+} ions, which in turn expanded the optical band gap. Mg-doped Cr_2O_3 thin films were synthesized using molecular beam epitaxy (MBE), followed by oxygen post-annealing treatment. This combination of techniques was employed to achieve precise control over film composition and defect states, thereby optimizing the structural and electronic properties of the doped Cr_2O_3 system [15]. They demonstrated that thin film conductivity can be enhanced through post-annealing under oxygen environment. Moreover, it has been demonstrated that the electrical conductivity in Cr_2O_3 is predominantly governed by the small polaron hopping (SPH) mechanism, which also accounts for hole conduction in both undoped and Mg-doped Cr_2O_3 thin films. In a related study published in 2017, the optical and electrical characteristics of Ni-doped Cr_2O_3 thin films fabricated via pulsed laser deposition (PLD) on sapphire substrates were investigated, offering further insight into the impact of transition metal doping on the functional properties of Cr_2O_3-based systems [16]. *P*-type electrical conductivity be reliably achieved only by doping Cr_2O_3 thin films with Ni, summarized in Table 2. Experiments confirmed that Ni was more soluble than Mg, and DFT calculations demonstrated that it increased delocalization of Cr_2O_3 valence band. These two benefits make Ni a preferable dopant for use in Cr_2O_3 powder. Ni-doped Cr_2O_3 thin films have a visible-range transmittance of 35–55%, with average being 40% transparent. This is because light is being reflected. The optoelectronic properties of Cr_2O_3 thin films have been improved with the addition of Mg, Al, and Ni as substituents. All thin films were deposited using the pulsed laser deposition (PLD) technique under oxygen-rich conditions, with sapphire substrates serving as the growth platform. This controlled environment facilitated the formation of desired phase compositions and defect structures, which are critical for tuning the electronic and optical properties of the films. However, insulating behavior resulted in Al substitution. But adding Mg and Ni boosts electrical conductivity with only a slight reduction in optical transparency. In case of Ni-substituent, electrical conductivity was found to be improved to 1.17 S/cm, while increase in electrical conductivity in case of Mg-dopant was of the order of 0.2 S/cm [6, 13].

$LaCrO_3$ MATERIAL

Perovskite-structured $LaCrO_3$ has been studied for its potential as a *p*-type TCO. In addition to being an insulator, $LaCrO_3$ also has a band gap of 4.6 eV, which makes transparent materials possible. Using a thin layer of $LaCrO_3$ doped with Sr, Zhang *et al*. demonstrated the optoelectronic properties [17]. An insulating sample is produced when a thin layer is not doped. The maximum conductivity was achieved at 50% Sr-doping, and the hole conductivity improved with increasing Sr-doping, as reflected in Table 3. Experimental observations indicate that increasing the Sr content in $LaCrO_3$ thin films, i.e., from x=0.04 to x=0.5, leads to a notable rise in carrier concentration, accompanied by an enhancement in Hall mobility from 0.0095 to 0.04 cm^2/Vs. Despite these improvements in electrical performance, the incorporation of Sr adversely affects optical transparency, with the transmittance decreasing significantly from 69.1 to 42.3%.

Table 2. Thin films of Cr_2O_3 material with different dopants, deposition techniques and optoelectronic parameters.

Host material	Dopant used	Growth technique	Optical transmission (%)	Band gap (eV)	Electrical conductivity, σ (S/cm)	Ref.
Cr_2O_3	Mg and N	Spray Pyrolysis	65	-	0.33	[14]
Cr_2O_3	Mg	MBE	55	-	0.17	[15]
Cr_2O_3	Ni	PLD	40	-	28	[16]
Cr_2O_3	Mg	PLD	68	3.51	0.18	[13]
Cr_2O_3	Ni	PLD	78	3.49	1.17	[6]

Table 3. Different dopants, deposition techniques and optoelectronic parameters of $LaCrO_3$ material.

Host material	Dopant used	Growth technique	Optical transmission (%)	Band gap (eV)	Electrical conductivity, σ (S/cm)	Ref.
$LaCrO_3$	Sr	PLD	42	-	54	[17]
$LaCrO_3$	Ba	-	-	-	2.27	[18]
$LaCrO_3$	Ca	-	-	-	35.1	[18]

It has also been doped with a thin layer of $LaCrO_3$ at a Sr concentration of x=1. In a remarkable turn of events, optical transmittance was reduced to 29% while hole conductivity was raised to 720 S/cm. Hole-induced phase shift from insulator to metal in epitaxial $LaCrO_3$ thin films was studied in the same year [17]. At x=0.65, it is shown that lattice mismatch causes thin layers to compress in-plane, leading to the insulator-to-metal transition. The resistance value entered the semiconductor regime below this point. Given the emergence of Sr-doped $LaCrO_3$ as a promising *p*-type transparent conducting oxide (TCO) material [18], first-principle's calculations have been employed to investigate the formation energies and electronic structures associated with various intrinsic defects. These theoretical studies suggest that under oxygen-rich conditions, the introduction of Sr leads to the formation of shallow acceptor states with minimal formation energy, thereby enhancing hole conductivity when a thin film of Sr-doped $LaCrO_3$ material is deposited. Many intrinsic defects, such as O-vacancies and Cr on the O-site in low oxygen environment, made up for the shallow acceptors. The electrical properties of $LaCrO_3$ can be improved by doping with divalent cations like Ca and Ba, just as they are with Sr. The use of Ca and Ba dopant in solid oxide fuel cell applications has already been explored [19]. Comparative studies have reported that Ca-doped $LaCrO_3$ exhibits superior electrical conductivity compared to its Ba-doped counterpart. It is noteworthy, however, that these findings are primarily based on measurements conducted on powdered forms of the doped $LaCrO_3$ samples, and comprehensive investigations on materials' thin films remain limited. Despite promising optoelectronic properties, no experimental results based on these characteristics have been published for Ca and Ba-doped $LaCrO_3$ thin films.

CONCLUSION

The exploration and development of *p*-type TCOs have made significant steps, with Cr-based materials emerging as one of the most promising candidates due to their tunable optoelectronic properties. Despite their inherent insulating nature, Cr-based oxides can exhibit appreciable electrical conductivity through suitable doping strategies, enabling their integration into transparent electronic devices. The advancement in this domain not only addresses the long-standing challenge of achieving efficient p-type conductivity in transparent materials but also paves the way for a new class of optoelectronic applications. One of the most effective ways to enhance the functionality of Cr-based *p*-TCOs is through the substitution of countercations such as magnesium (Mg), strontium (Sr), nickel (Ni), and nitrogen (N). These dopants play a pivotal role in modulating the electronic structure, reducing defect densities, and enhancing carrier concentration and mobility, contributing to improved electrical and optical performance. In addition to chemical doping, physical modifications such as the irradiation of thin films with swift heavy ions (SHI) offer another way for tailoring material's properties. SHI irradiation can induce beneficial structural reconfigurations, grain size refinement, and defect engineering, all of which contribute to enhanced optoelectronic behavior. Furthermore, the deposition conditions, particularly the ambient gas environment during synthesis, play a crucial role in dictating the final properties of Cr-based thin films. A highly enriched oxygen environment has been shown to promote superior optoelectrical characteristics by minimizing oxygen vacancies and stabilizing desirable oxidation states. Overall, combining doping strategies, ion irradiation, and optimized deposition conditions holds great promise for advancing Cr-based *p*-TCOs in transparent active devices, enabling innovations in displays, sensors, and energy applications.

Conflict of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this study.

Data Availability Statement

The data that support the findings of this study are available on request from the corresponding author.

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