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Non-Linear Behaviour of Thermally Deposited Silver Oxide Thin Films

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Silver oxide thin films were synthesized on a glass substrate using a thermal evaporation technique at room temperature, followed by post-annealing in a vacuum furnace at 150 °C for 30 minutes. The synthesized thin films were characterized for structural, morphological, and nonlinear optical properties. X-ray diffraction patterns revealed that the deposited films were cubic with a cuprite crystal structure. Morphology of the deposited thin films was studied using field emission scanning electron microscopy (FE-SEM). The optical limiting behaviour of the thin films was investigated using a resonant laser wavelength. These films were also experimentally investigated using a Fabry-Perot interferometer for their optical bistable nature. The prepared thin films showed optical limiting behavior at laser wavelengths of 632 nm.

Keywords: Silver oxide, XRD, Non-linear optics, Saturation, Optical bistability

1 Introduction

In recent years, there has been a lot of interest in the study of novel metal oxide nanomaterial thin films deposited on glass substrates, considering their uses in optical engineering[1], energy storage, biomedicine and sensors[2]. Thin silver films have drawn much attention because of their unique optical properties[3]. Many optical technologies, such as light-emitting diodes and solar cells, as well as methods to improve the properties of organic semiconductor materials and new metamaterials, are being investigated. It is well known that the deposition conditions, such as the substrate type and temperature, vacuum pressure, and deposition rate, regulate the aggregation of grains during the growth of thin films. To grow Ag_2O thin films, a variety of techniques, including PLD[4]. Vapor-liquid-solid process[5], chemical synthesis[6], RF, DC sputtering[7], the methods of chemical techniques8, such as anodic growth[9], electrodeposition[10], spray pyrolysis[11], and thermal evaporation are employed[12,13]. Research labs and industrial plants continue to use thermal evaporation, one of the earliest techniques for depositing thin films, to deposit metals and metal alloys[14].

Silver exists in various oxidation states and forms several oxides due to its d-shell electrons: Ag_2O, AgO, Ag_3O, and Ag_2O_3, creating different types of inorganic compounds. Among these oxides, the Ag_2O phase possesses the highest thermodynamic stability[15]. Different optical and electrical properties are made possible by the different shapes that constitute the Ag_2O crystal structures. Applications involving ultra-high-density optical data storage benefit from this characteristic in fluorescence imaging and the property of surface-enhanced Raman scattering (SERS) in plasmonic devices. Analysis of the surface morphology revealed that the films are continuous, fine-grained, and free of defects like pores and fissures. Moreover, silver oxide thin films have been used to create incredibly high-density optical storage devices. Additionally, they have been employing plasmon photonic devices and solar energy devices[16]. The growth technique and the experimental setup have a significant impact on the characteristics of thin films. On the other hand, there is a great deal of variation in silver oxide thin films, particularly in their electrical and optical characteristics. In addition to experimental effects, the interpretation of the experimental results and the identification of a specific film belonging to a particular phase of silver oxide are important contributing reasons for this large variation. Using pure oxide as the starting material is the most effective way to deposit metal oxide thin films. This partially bypasses the need for a reactive oxygen environment and the effects it has on the porosity, crystallinity, and rate of evaporation of the films.

The goal of this paper was to examine the characteristics of the films made from pure Ag_2O powder by thermal evaporation, as well as how these

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characteristics were impacted by the deposition conditions. Properties including surface topography, elemental composition analysis, and optical studies of the prepared material, Ag_2O thin films have been studied previously[17]. In general, silver is regarded as a non-reactive substance. According to Arenas *et al*[18] silver oxide (Ag_2O) is a p-type semiconductor with a band gap of 1.2 eV. While other studies show that silver oxide has a wide energy band gap from 1.2 eV to 3.4 eV [19,20]. Because of its wide optical band gap, it is transparent in the visible and infrared spectra[21]. These characteristic silver oxide thin films are useful for creating antireflective coatings for opto-electrical applications[22].

We investigated the non-linear optical characteristics of Ag_2O thin films that are made by the direct thermal deposition technique at room temperature on pre-cleaned glass substrates and have a thickness of about 170 nm. Many studies have been conducted on the linear and nonlinear interactions of materials with light in the form of thin films or nanoparticles[23]. The Ag_2O thin films exhibit large third-order nonlinear susceptibility and nonlinear, refractive index, and response time making them attractive for use in optical data, systems of storage, and all-optical photonics systems[24]. The development of materials with high nonlinear optical response, low optical absorption for the spectral range of interest, small moisture sensitivity, high mechanical resistance, and thermal stability, large linear refractive index, and easy fabrication processing is being spurred by developments in photonic devices fabrication[25]. The spectral, structural, linear, and nonlinear studies of deposited thin films are conducted and examined in the corresponding sections. Among these ideas that were able to accomplish optical bistability are self-electro-optic devices (SEED) as well as laser-induced optical devices (LIOD). More recently, research has taken a different turn, and there is now a great deal of interest in using microelectromechanical systems (MEMS) for optical switching. Nevertheless, the transparent Ag_2O thin films show potential for use in optical limiting applications[26]. To obtain a solid understanding of the light-matter nonlinear interaction, experimental processes such as saturation and optical bistability are tried.

2 Experimental Details

2.1 Material Synthesis

The thermal evaporation system (model-TAS/2018/01, Technology Applications Services, New Delhi) was used to make thin films from a tungsten basket shown in Fig. 1. The thermal evaporation technique is one of the most basic vacuum deposition systems, which is the most straightforward and has the least controlling aspect, is a classifier for physical vacuum deposition processes (PVD). Vacuum evaporation involves heating the source material and providing precisely the right amount of pressure and temperature to the substrate. The films were deposited on unheated, well-cleaned glass substrates. The first component was a 10 mm diameter by 3 mm thick pellet of 99.9% pure black Ag_2O powder (Loba Chemie), which had a melting point of 280° C. After cleaning the glass slides with acetone, the substrates were placed in an ultrasonic bath with distilled water and a chemical agent for approximately 35 minutes. Lastly, fresh water was used to clean the substrates, and they were properly dried to prevent fingerprints and dust. First of all, the

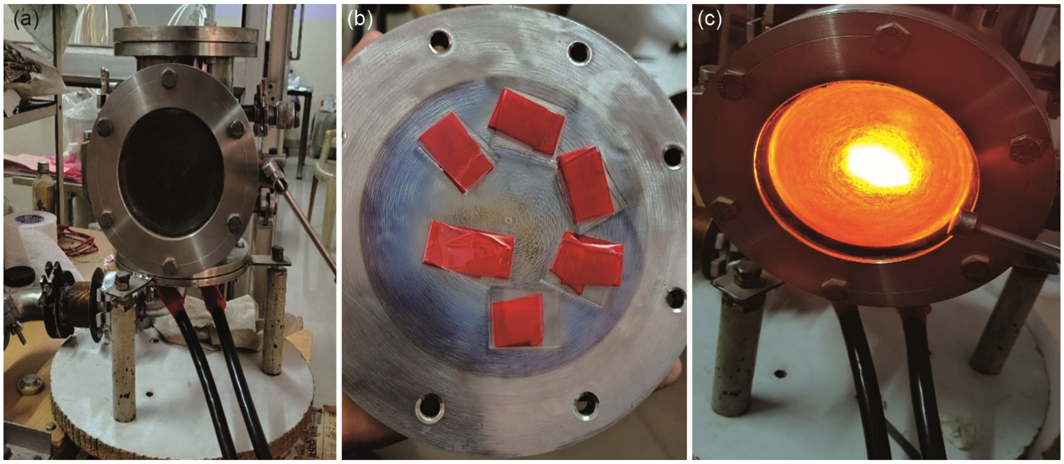

Fig. 1 — Deposition setup for the vacuum evaporator (a) deposition chamber, (b) substrate holder with glass slides assisted with the tape, and (c) hot boat glowing during the deposition process

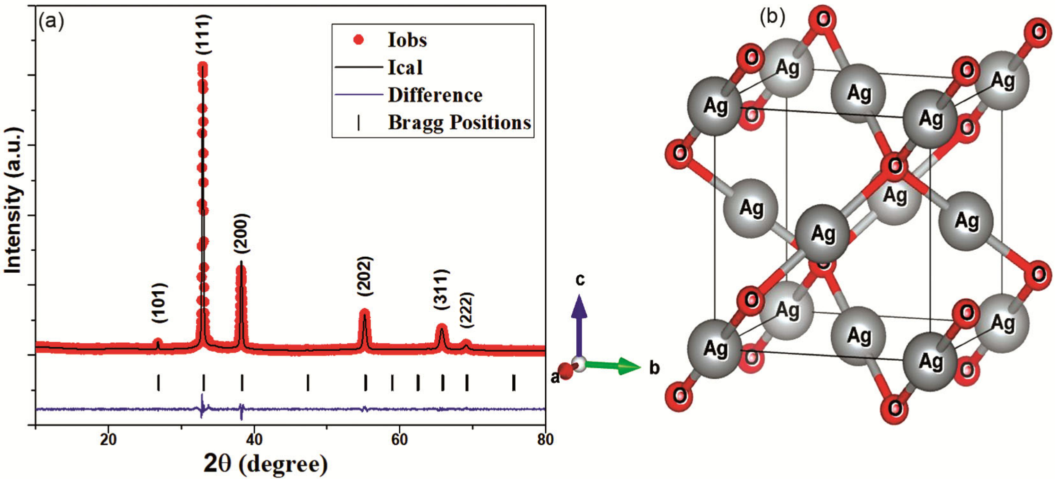

Fig. 2 — Structural analysis of Ag2O thin films (a) X-ray diffraction pattern of Ag_2O thin film, and (b) Cuprite Structure of deposited thin films

glass substrates were fixed with the help of tape in the substrate holder and sealed in the vacuum chamber as shown in Fig. 1 (b). The target and substrate were 10 cm apart inside the evaporation chamber. The whole process was done at room temperature under the base pressure of 10^{-5} torr, at which the system was pumped by using a rotary pump and maintained during the coating. We slowly heated the source to degas the material. Using a tungsten basket filament and resistive heating, the Ag_2O target was evaporated and condensed onto a pre-cleaned glass substrate. To maintain the temperature difference, the sample under investigation was placed on a tungsten basket, and the module's wires were attached to a continuous power source to supply the power voltage to the thermoelectric module. After the synthesis, thin films were annealed at 150 °C for 30 minutes.

2.2 Characterisation Techniques of Ag_2O Thin Films

Many methods have been used to examine the various characterizations of thin films, including their structural, and non-linear optical characteristics. The synthesized Ag_2O thin layers were examined using the XRD technique using an X-ray diffractometer (Cu-Kα radiation λ=0.154056 nm) to ascertain the crystal structure of the prepared samples. The resulting transmittance, Saturation behavior, and non-linear optical characteristics of the thin films were measured using a non-linear optical bistability setup.

3 Results and Discussion

3.1 XRD Analysis

The prepared samples have been characterized using the XRD technique for the confirmation of the formation of the crystalline phase and their crystallite size (Fig. 2). The powder XRD pattern of Ag_2O is recorded on a Rigaku miniflex II X-ray diffractometer. The scan rate is set at 2°/min with the scan range selected from 2θ = 10° to 80° [27]. To get structural information, Rietveld refinement was performed using the full prof program, space group Pn-3m (224), a= 4.7139 Å, b= 4.7139Å, c= 4.7139Å, RB= 2.182, RF= 2.431 and visual representation of the unit cell obtained using VESTA software (Table 1).

Table 1 — Structure parameters of prepared samples

Atoms	Wyckoff position	x	Y	z	Occupancy
Ag	4b	0.00000	0.00000	0.00000	1
O	2a	0.25000	0.25000	0.25000	1

The phase identification has been confirmed using the Crystallography open database (COD) ID 4318188 [28]. XRD results revealed that the as-deposited silver oxide thin film is cubic with a cuprite crystal structure[29] (Fig. 2 (b)).

3.2. FE-SEM Analysis

The deposited Ag_2O thin film's surface morphology was investigated using FE-SEM. The nanoparticles in the films have an approximately spherical shape (similar to nanoglobular agglomeration) and are homogeneous but not highly uniform. The FE-SEM image displays smaller, randomly distributed grains (Fig. 3).

3.3 Non-Linear Optical Properties

3.3.1 Saturation Behaviour

A basic experimental setup (Fig. 4) is employed to study the saturation absorption behavior of Ag_2O thin

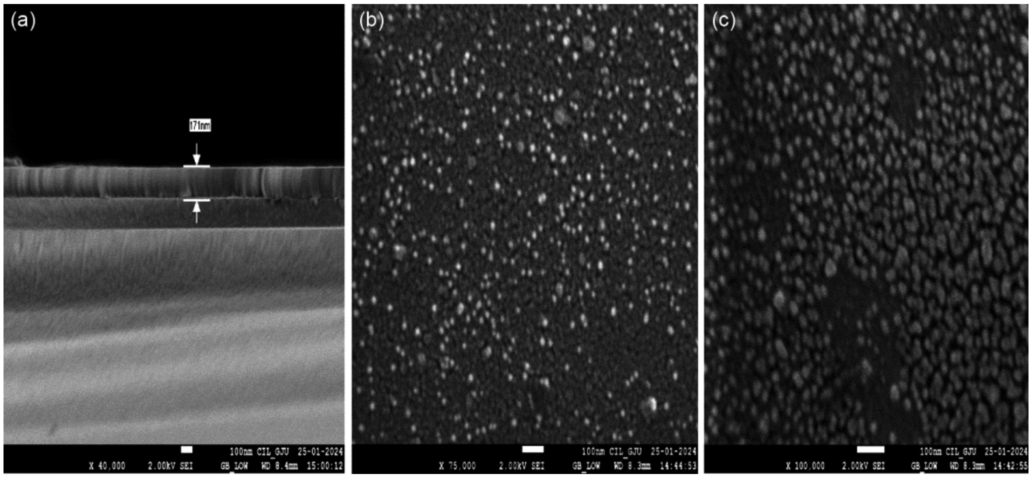

Fig. 3 — FE-SEM micrographs of Ag_2O thin film (a) cross-section area for deposited thin films with rough thickness around 171 nm, (b) magnification at 75000x, and (c) magnification at 100000x

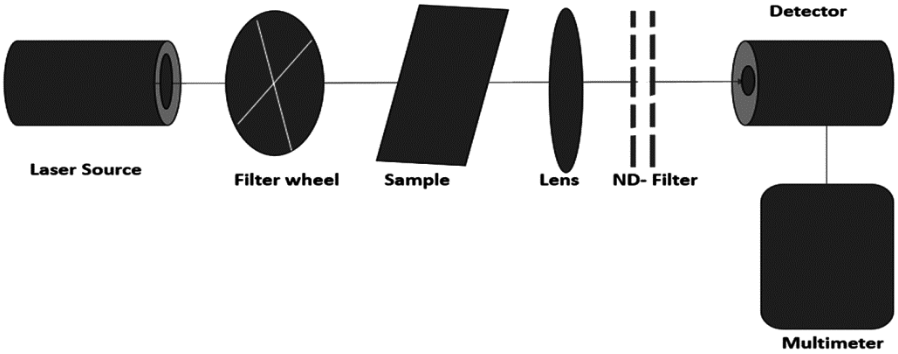

Fig. 4 — Schematic diagram for measuring saturation absorption

film samples by using a He-Ne laser with a wavelength of 632.8 nm and a maximum output power of 23 mW. A filter wheel is designed with 12 variable natural density filters (Edmund Optics) as shown in Figure 4. The transmittance energy density of the filters starts from 0.1 % to 79 % transmittance. The sample is mounted between the variable ND filter wheel and a focusing lens. The laser beam passed through the variable ND filters having different transmittance, then traversed the samples after crossing the focusing lens. The transmitted intensity is passed through a separate ND filter, which is placed behind the focusing lens to reduce the intensity falling on the detector. The single is connected to the multimeter to get the output intensity. This setup enables the measurement of laser light absorption as a function of varying incident light intensity. The resulting data plots the saturation absorption curve, illustrating the nonlinear relationship between absorption and transmittance intensity as shown in Fig. 5.

The saturation absorption curve of Ag_2O thin films demonstrates how light absorption varies with increasing incident intensity flux. At low intensities, absorption increases proportionally, and later it becomes saturated. This characteristic is crucial for optical limiting applications, where the material effectively attenuates high-intensity light, protecting sensitive components. Nevertheless, we do not see a marked saturation behavior, which is expected for an inorganic crystal of silver oxide. Since the material is silver ions in the form of thin films. Saturation is generally not expected. Ions should be highly resonant with the laser wavelength to achieve better saturation. Molecules generally show pronounced saturation since they possess rotational and vibrational degrees of freedom. Many molecules show confirmation changes when an intense laser beam interacts with them.

3.3.2 Non-Linear Optical Bistability

Passive non-linear optical bistability is being carried out in prepared materials. An optical system is said to exhibit optical bistability if, under a given incident light intensity (I_I), it can have two distinct transmitted light intensities (I_T). The system can switch between these two states rapidly and reversibly. Figure 5 shows that output power can take two different values for the same incident power within a certain range of I_I. This behavior is nonlinear, meaning the transmitted intensity I_T is not simply proportional to the incident intensity I_I (i.e., I_T is not just a constant multiple of I_I). Instead, the system shows two possible output states (I_T) for a particular input intensity (I_I), making it bistable. Such a system is capable of maintaining one of the two states until a perturbation or change in input intensity forces it to switch to the other state. Input intensity I_I is systematically varied, first by increasing and then decreasing, and noting down the transmittance at each step, which exhibits a hysteresis type loop. The schematic diagram for field propagation in the Fabry-Perot (F-P) cavity is shown in Fig. 6[30,31]. In an F-P cavity, detuning of the wave vector from the resonant state takes place. The intensity of light causes a refractive index (n) of the medium, which is as follows:

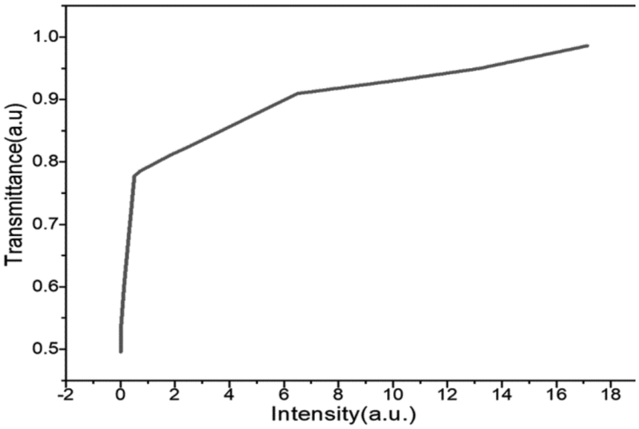

Fig. 5 — Saturation absorption curve of deposited Ag_2O thin film sample

$$n = n_0 + n_2 I \qquad \dots (1)$$

Here, n_0 is the linear refractive index of the medium, n_2 is the optical Kerr coefficient, and I is the light intensity inside the medium.

The transmission of the Fabry-Perot cavity is defined as

$$I = \frac{I_T}{I_I} = \frac{1}{[1+F\sin^2(\frac{\Phi}{2})]} \qquad \dots (2)$$

Here, I_T stands for transmitted intensity, I_I for incident intensity, and F for cavity finesse. Which is expressed as $F = 4R/T^2$ with $R(T = 1-R)$ for mirror reflectivity and $1/[1+ F \sin^2(\Phi/2)]$ is referred to as Airy function, $\Phi = 2\pi n_0 L(\lambda/2)$ is the round-trip phase shift where λ is the wavelength of the radiation and L is the length of the cavity.

The transmittance plot shows a saturation at input intensity. The bistable nature exhibited as shown in Fig. 7, is absorptive bistability, therefore Ag_2O films can be promising materials for optical switching devices, and further doping with ions (rare earth, transition metals) may enhance the optical limiting behavior[32-33]. The hysteresis type loop of the optical bistability based on Fabry–Perot interferometry for Ag_2O thin films (Fig. 8).

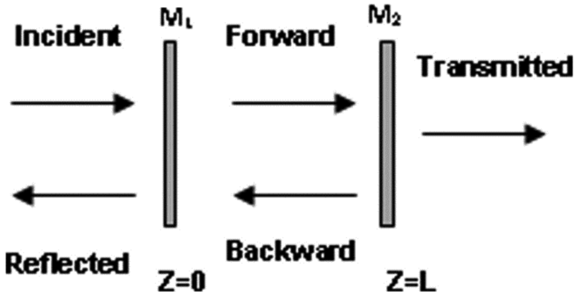

Fig. 6 — Schematic diagram of the Fabry-Perot cavity for field propagation

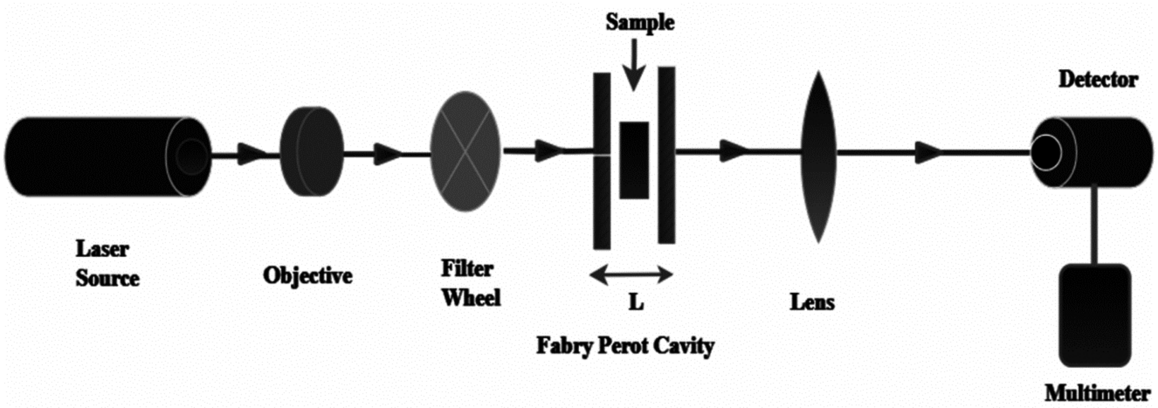

Fig. 7 — Schematic illustration of a Fabry-Perot interferometer for optical bistability

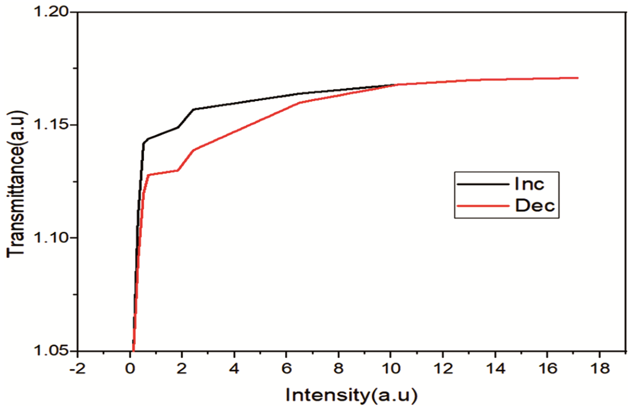

Fig. 8 — Hysteresis type loop of the optical bistability based on Fabry–Perot interferometry for Ag_2O thin films

4 Conclusion

Ag_2O thin films were synthesized at room temperature utilizing thermal evaporation treatment on glass substrates. However, the linear and non-linear optical properties of Ag_2O thin films can be changed by adjusting the deposition conditions. The current study is to investigate the structural, morphological and non-linear optical characteristics of Ag_2O thin films. XRD pattern revealed that the deposited Ag_2O thin film is cubic, which provides the [hkl] values and indices to the various planes. FE-SEM analysis revealed the formation of spherical nanoparticles (like a cluster of nanoglobules). The thin films exhibit partial saturation behaviour, indicating potential applications as optical limiting materials. The transmittance plot shows a saturation at increasing incident intensity. F-P cavity gives the non-linear optical feedback of silver oxide thin films. The two stable states at a single input state of the synthesized Ag_2O thin film are demonstrated. According to our investigation, Ag_2O thin films are promising materials for optical bistability applications and can be used in nonlinear optical switches for data processing systems and communication. Measurements of non-linear parameters of deposited thin films, which are reported here give a good response so that Ag_2O thin films can be utilized for photonic applications. However, investigating third-order susceptibility $\chi^{(3)}$ by using the nonlinear Z-scan technique is needed. Because the third-order susceptibility $\chi^{(3)}$ is expected to play a major role, as the thin film shows a cubic crystalline structure. Films of this feature can be used in applications that require two, three, and four-wave mixing, and device applications include optical limiting and phase conjugation mirrors.

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