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Insight into Mechanisms of Electronic Sputtering in Gold-Silica Nanocomposite: Effect of Size of Au Nanoparticle

Vinay Kumar Srivastava
Department of Physics,
Deen Dayal Upadhyaya Gorakhpur University Gorakhpur, Gorakhpur, 273009, Uttar Pradesh, India.
E-mail: vinaypramod26@gmail.com

Shivani Chaudhary
Department of Physics,
Deen Dayal Upadhyaya Gorakhpur University Gorakhpur, Gorakhpur, 273009, Uttar Pradesh, India.
E-mail: chaudharyshivi16jan@gmail.com

Udai Bhan Singh
Department of Physics,
Deen Dayal Upadhyaya Gorakhpur University Gorakhpur, Gorakhpur, 273009, Uttar Pradesh, India.
Corresponding author: udaibhansingh123@gmail.com

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Abstract
This research examines the electronic sputtering characteristics of $Au:SiO_2$ nanocomposite thin films, focusing on size dependence, when subjected to 100 MeV Ag ion irradiation. $Au:SiO_2$ films were deposited on quartz substrates through atom beam sputtering, subsequently undergoing controlled annealing at 850°C in an Ar atmosphere to optimize the nanoparticle size. The progression of nanoparticle dimensions and their optical characteristics was observed via UV–Visible spectroscopy, validating an increase in size following the annealing process. Post-irradiation analysis employing Rutherford Backscattering Spectrometry (RBS) demonstrated a distinct inverse correlation between the size of nanoparticles and the sputtering yield. The results show that as nanoparticle size increases, the electronic sputtering yield decreases, highlighting the crucial role that nanostructure dimensions play in swift heavy ion–matter interactions.

Keywords- Sputtering, Nanocomposite, Surface plasmon resonance, RBS, Thermal spike.

1. Introduction

Ion beam irradiation allows the transfer of kinetic energy from ions to the atomic lattice and electronic systems of solids via mechanisms such as elastic collisions and electronic excitation or ionization (Chaudhary et al., 2024a). At elevated energies of 100 keV/amu, the primary mechanisms for energy deposition in ions are as follows (Imanishi and Ninomiya, 2004; Johannes et al., 2015; Möller, 2017). High-energy ions interact with a material's surface, dissipating their energy through collisions with electrons and atomic structures, leading to the formation of defects and an implantation cascade occurring on the order of femtoseconds (Johannes et al., 2015; Pandey et al., 2015). The extent of the collision cascade is influenced by the energy of the ions (Pandey et al., 2021), their mass, and the density of the target material (Shams-Latifi et al., 2024). The energy introduced by ions is partially transferred to phonons, which rapidly dissipate into the surrounding material (Kumar et al., 2017). The process of energetic ion irradiation in solid materials encompasses intricate interactions within the target, characterized by both nuclear and electronic energy loss mechanisms (Deoliet al., 2014; Shams-Latifi et al., 2024). Energy dissipation transpires via two mechanisms: nuclear energy loss (Sn) resulting from elastic collisions between the ion and target nuclei (Deoli et al., 2014), and electronic energy loss (Se) arising from inelastic interactions with the electronic subsystem of the target. leading to the generation of excitations and ionizations (Mammeri et al., 2018). The processes frequently result in the phenomenon known as sputtering, wherein atoms displacement, molecules, or clusters are expelled from the surface of the target material (Möller, 2017). The nuclear sputtering process, which results from nuclear energy loss, is well explained by Sigmund's linear cascade theory (Sigmund, 1969, 2005). This model does not explain the very high sputtering yields seen in regimes where electronic sputtering is dominated by electronic energy loss. Throughout the years, numerous theoretical frameworks have been developed to elucidate the phenomenon of sputtering resulting from electronic excitation mechanisms, especially in the context of metals and insulators.

Although electronic sputtering yields in metallic systems are usually low, they may be higher than those predicted by Sigmund's theory (Sigmund, 1969),especially in nanostructured geometries or ultra-thin films. Insulating materials frequently demonstrate elevated sputtering yields when subjected to SHI irradiation, attributed to their limited abilities for electronic energy loss (Sigmund, 2005; Toulemonde et al., 2002). Although extensive studies have been conducted on electronic sputtering in bulk metals, insulators, and thin films (Gupta and Avasthi, 2001; Singh et al., 2011), there remains a notable lack of focus on metal-dielectric nanocomposite systems (Singh et al., 2011). Because of their nanoscale size and interfacial effects, these composites, especially those that include noble metal nanoparticles embedded inside dielectric matrices like SiO_2, display special physical features (Chaudhary et al., 2024b; Joseph et al., 2007). Electronic sputtering represents a non-thermal mechanism for material removal that occurs during swift heavy ion irradiation, driven by substantial electronic energy dissipation. The process can be elucidated through either the inelastic thermal spike model or the Coulomb explosion model (Khatter et al., 2022), conditioned upon the target material and the excitation density associated (Khan et al., 2011). Nanoscale metallic systems, ultra-thin films, and nanostructures can enhance electronic sputtering yields due to confinement effects, reduced electron mean free paths, and altered electron-phonon interactions (Dash et al., 2015; Singh et al., 2014). Metals exhibit a significant capacity for electron thermal conductivity. Insulators and semiconductors exhibit higher electronic sputtering yields as a result of defect-mediated bond breakage and the slower transfer of electronic heat (Gupta et al., 2025). Both insulating and metallic phases have an impact on electronic sputtering in nanocomposites; excited electrons are scattered and heat transport pathways are altered at the interface between phases (Arnoldbik, 2005; Gupta et al., 2025).

Gold (Au) nanoparticles exhibit remarkable characteristics, including exceptional chemical stability, pronounced surface plasmon resonance, and adjustable properties (Chaudhary et al., 2024b; Joseph et al., 2007). When incorporated within a SiO_2 matrix, Au nanoparticles establish a stable Au:SiO_2 nanocomposite (Jung et al., 2008; Terauchi et al., 1997), functioning as an effective model system for investigations involving ion irradiation (Chaudhary et al., 2024a; Kuiri et al., 2007). Nonetheless, the sputtering characteristics of these nanocomposites in high Se environments are not thoroughly investigated, especially regarding the impact of nanoparticle size (Kumar et al., 2007) on electronic sputtering yields (Breuer et al., 2018; Zhu et al., 2023). The applications of nanocomposites, such as in plasmonic sensing, photothermal therapy, nonlinear optics, and radiation-resistant coatings.

This research methodically examines the influence of size on electronic sputtering in Au:SiO_2 nanocomposite thin films when subjected to irradiation by 100 MeV Ag ions. The results indicate a distinct relationship between sputtering yield and nanoparticle size, where larger nanoparticles demonstrate reduced sputtering efficiency. This behavior highlights the essential influence of size effects in ion-matter interactions at the nanoscale and enhances the foundational comprehension of energy dissipation mechanisms in nanostructured systems subjected to electronic excitation conditions.

2. Experimental Details

Thin films of Au:SiO_2 were deposited on fused quartz substrates by atom beam sputtering , the details of the set up and synthesis are reported (Khan et al., 2011). Some samples were annealed at temperature of 850^{o}C in an inert atmosphere for an hour to increase the size of nanoparticles. Samples were irradiated with 100 MeV Ag ions with fluence 1×10^{13} ions/cm^2. Rutherford backscattering spectroscopy (RBS) is used for the determination of areal density of metal before and after irradiation of NCs using 2 MeV He+ ions. Optical properties of Au:SiO_2 nanocomposite is carried out using a dual beam Hitachi U3300 spectrophotometer in the spectral range of 400–800 nm.

3. Results and Discussion

The optical absorption spectra of as-deposited, annealed and irradiated NCs are shown in **Figure 1(a)** and **1(b)**. The absorbance at ~ 500 nm, is characteristic surface plasmonic resonance of Au nanoparticles, which confirmed the existence of Au NPs in these films. The surface plasmonic resonance frequency depends on the (i) size, (ii) shape and (iii) the embedding environment of the metal nanoparticles. It is observed that the size of the Au nanoparticles increases with annealing as revealed by the decrease in full width at half maximum (FWHM) of the peak. The average size of nanoparticles is estimated using the Fermi velocity formula (Mishra et al., 2007) as:

$$Particle\ size = {}^{2\hbar\vartheta_F}/_{\Delta E} \tag{1}$$

where, $\vartheta_F = 1.39 \times 10^8$ cm/s is the fermi velocity of electrons in case of bulk Au and ΔE is the FWHM (in eV). The as-deposited Au-silica films have Au NPs with average size of 4 nm whereas the annealed film with an average size of 7.8 nm.

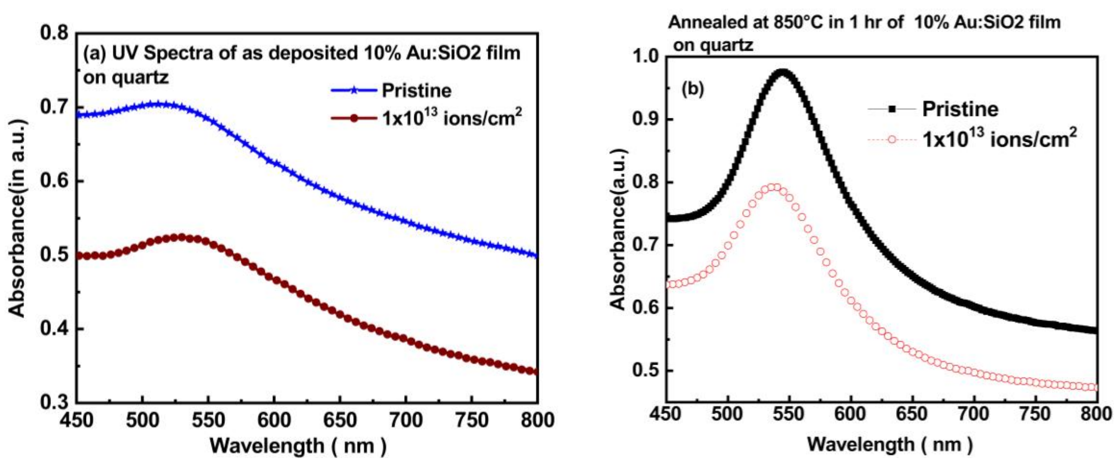

Figure 1. Optical absorption spectra of (a) as deposited and irradiated (b) annealed at 850°C and 100 MeV Ag ion beam irradiated annealed of Au:SiO_2 Nanocomposite (at fluence 1× 10 ^{13} ions/cm^2).

3.1 Quantification of Sputtering: Areal Concentration and Yield

The reduction in size of nanostructures significantly influences phonon dynamics. Smaller grains exhibit a decreased Debye temperature, which results in modified phonon spectra and enhanced electron-phonon interactions. This interplay leads to greater energy coupling and increased sputtering effects. In minuscule grains, the restricted phonon mean free path and heightened boundary scattering hinders heat dissipation, permitting the localized lattice temperature to elevate prior to energy diffusion, which escalates bond breaking and atom ejection, thereby augmenting sputtering efficiency in finer nanostructures.

Sputtering is generally assessed by the decrease in areal concentration (N_c) of the sputtered species, which is measured using Rutherford Backscattering Spectrometry (RBS). This measurement is derived from recoil counts (Y) through the following relation.

$$N_c = \frac{Y \sin\alpha}{N_p \frac{\partial\sigma}{\partial\Omega}\Omega} \tag{2}$$

Here, N_p is the number of incident ions, $\frac{\partial\sigma}{\partial\Omega}$ is the recoil cross-section, α is the incidence angle between beam and sample surface, and Ω is the solid angle subtended by the detector.

The sputtering yield (Y) is determined by taking the difference in areal concentration between two fluences and normalizing it by the ion fluence.

$$\text{Sputtering Yield (Y)} = \frac{N_{c\phi_1} - N_{c\phi_2}}{\phi_1 - \phi_2} \text{ (atoms/ion)} \tag{3}$$

Here, ϕ_1 and ϕ_2 represent the ion fluences prior to and subsequent to the sputtering measurement, $N_{c\phi_1}$ and $N_{c\phi_2}$ are areal concentration for pristine and at any fluence. For smaller particles, a sharper decrease in N_c verifies greater sputtering rates, which are in line with the confinement-based and thermal spike theories. Martinez et al. (2015) studied that how electronic sputtering change when the different parameters changes. One very intriguing finding is that the yields of sputtered particles vary a lot on the thickness of the coating. As the film becomes thicker, the number of positive sputtered ions likewise increases. For films that are quite thick ($z > 12$ nm), the sputtered ion yields stop increasing because the track core becomes fully ionized (Martinez et al., 2015).

The metal concentration in pristine, annealed and irradiated samples is determined by Rutherford Backscattering Spectroscopy and spectra are shown in **Figure 2**. The areal concentration of as-deposited film is decreased with irradiation from 4.87×10^{16} atoms/cm^2 to 3.80×10^{16} atoms/cm^2 at fluence of 1×10^{13} ions/cm^2. The RBS spectra were analyzed and fitted using the SIMNRA 7.03 simulation software.

The sputtering yield is estimated to be 1070 atoms/ion in as deposited case having particles with average sizes 4 nm. In contrast to as deposited case, the annealed samples having average size 7.8 nm shows very less sputtering yield 66 atoms/ion as estimated from RBS. The metal concentration decreases from 3.61×10^{16} and 3.55×10^{16} atoms/cm^2. The size dependent sputtering rate of NCs can be understood in terms of the thermal spike model, in which a reduced mobility of the electrons and a more energy confinement due to scattering of the excited electrons from the surface boundaries of particle results in an increased sputtering in compare to large particle.

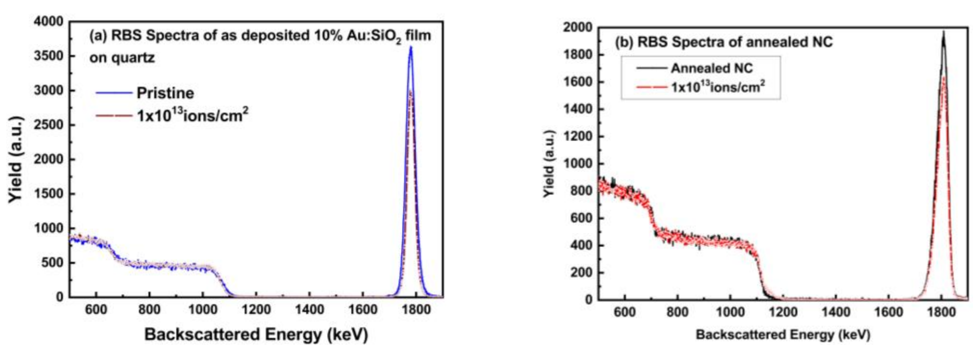

Figure 2. Rutherford backscattering spectra of Ag ion irradiated a) as deposited and irradiated (b) annealed at 850°C and irradiated annealed of Au:SiO_2 Nanocomposite. (100 MeV Ag ion beam irradiation at fluence 1×10^{13} ions/cm^2).

3.2 Discussion of the Thermal Spike Model, Electron-Phonon Coupling, and Size-Dependent Electronic Sputtering

Electronic sputtering is often modelled using the inelastic thermal spike model. According to this theory, energy is deposited in the electronic subsystem by a high-energy ion along a cylindrical track. This energy is then redistributed via electron-electron interactions and eventually transmitted to the lattice by electron-phonon coupling. This leads to a swift increase in lattice temperature adjacent to the ion track. The efficiency of this energy transfer is determined by the electron-phonon coupling constant (g)

$$g = \frac{D_e(T_e).C_e(T_e)}{\lambda^2} \tag{4}$$

Here, D_e is the electronic thermal diffusivity, C_e is the electronic specific heat, T_e is the temperature and λ is the mean diffusion length of excited electrons.

The phenomenon of electronic sputtering in nanostructured thin films and nanocomposites exhibits a significant size dependence, which arises from confinement effects, changes in electron transport properties, and alterations in electron-phonon interactions. The dimensions of nanoparticles are essential factors influencing sputtering behaviour due to increase in electron phonon coupling factor. The size of the nanoparticles emerges as the dominant variable influencing the sputtering behaviour. At the nanoscale, the phenomena of quantum confinement and the effects of surface-to-volume ratio play a crucial role in determining the behaviour of materials. The film surface in these materials serves as a confinement barrier for electron movement, thereby altering electron mobility and scattering dynamics. As the size of grains reduces, there is a corresponding increase in the density of grain boundaries, which act as efficient scattering centres for excited electrons. This minimizes the average diffusion length of electrons, thereby constraining their energy transport away from the ion track region. This leads to an increased concentration of deposited energy and heightened local heating, thereby amplifying sputtering in smaller grains. Reduction in thermal conductivity is also influencing confinement in energy in nano dimensional. It leads to increase in temperature of lattice. Increase in the lattice temperature due to increase in electron phonon coupling factor and reduction in thermal conductivity causes huge sputtering in comparison to bulk. The role of various parameters for thermal spike in various literatures has been showed. Ridgway et al. (2011) examined the morphological changes of elemental metal nanoparticles (NPs), with particular emphasis on shape alterations induced by Swift Heavy Ion Irradiation (SHII). Metal-specific variations were observed in the threshold diameter of spherical nanoparticles and the width of elongated nanoparticles, correlating with the dimensions of the molten ion track and the energy density per atom necessary for vaporization (Ridgway et al., 2011). Seznes and Solids (2020) showed about how the properties of materials affect track formation and shows that there is a clear link between melting temperatures T_m and ion-induced track radii R_e in various solids. 14 different insulators, such as $LiNbO_3$ and $BaFe_{12}O_{19}$, have been shown to be valid. The research also challenges earlier findings that came to opposite conclusions, showing that an arbitrary ion energy value changed the results a lot (Szenes and Solids, 2020).

4. Conclusions

Under the condition of swift heavy ion irradiation with 100 MeV Ag ions, this research examines the size-dependent electronic sputtering behaviour of Au:SiO_2 nanocomposite thin films. Controlled annealing was employed to increase the size of the embedded Au nanoparticles. Annealing caused the nanoparticles to enlarge, with diameters rising from around 4 nm to 7.8 nm (is increased to approximately double), as confirmed by UV-Visible spectroscopy. Rutherford Backscattering Spectrometry analysis showed a clear decrease in sputtering yield with increasing nanoparticle size, highlighting a strong inverse correlation between particle dimension and sputtering efficiency. Sputtering yield decreased considerably for bigger (7.8 nm) nanoparticles compared to smaller (4 nm) ones, reducing from 1070 atoms/ion to 66 atoms/ion.

This phenomenon is explained by size-dependent thermal spikes, which make sputtering more effective for smaller nanoparticles than bigger ones due to their enhanced energy confinement and lower electron mobility. This study indicates that the results may contribute to the development of plasmonic devices exhibiting improved stability when exposed to irradiation, and facilitate precise nanofabrication through the utilization of ion beams for optimizing surface morphology and functionality.

Conflict of Interest

The authors declare no conflict of interest.

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AI Disclosure

The author(s) declare that no assistance is taken from generative AI to write this article.

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