

Physical Property Variation of Ferrite Nanoparticles under Heavy Ion Irradiation

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Abstract

The size-dependent behavior of nanostructured ferrite is well established. Various physical properties such as magnetic, optical and electrical exhibit strong size dependence. Thus, any treatment which causes a change in size is able to modify the characteristics of ferrites. The result of this effect can be seen when these ferrites are subjected to an intense heavy ion beam which modifies the physical properties of ferrites. This modification is related to cation redistribution owing to size change in most of the cases under heavy ion irradiation. However, few recent studies show that cation redistribution may occur under ion irradiation even though no size change is observed. The objective of this review is to highlight this effect in ferrite systems which ultimately may provide ample opportunity for its potential applications.

Keywords- Ion irradiation, Ferrites, Cation occupancy, Size independence.

1. Introduction

Ferrites have been a well-known system to mankind for a long time (Lord & Parker, 1960; Grimes, & Collett, 1971; Sato et al., 1987). These ferrites have the formula MFe_2O_4 , where, M is a transition metal (TM) ion and exhibits ferrimagnetic character in the bulk form (Brockman, 1951). One can alter the magnetic behavior of these ferrites by replacing M with appropriate transition metals (Smit & Wijn, 1954). Thus, the selection of TM ions is required while designing these ferrites for specific applications when they are used in bulk form. This leads to the growth of zinc ferrite, manganese ferrite, cobalt ferrite, nickel ferrite, nickel-zinc ferrite etc. (Table 1). Changes in microstructure or external treatment are also responsible for modification in the physical behavior of ferrites by altering cation inversion (Zhuravlev et al., 1990; Singh et al., 2013; Peng et al., 2017). Cation inversion is a frequently used term in ferrite and details can be found in many reports (Torruella et al., 2018; Nandy et al., 2022).

Table 1. Design of various ferrites while replacing M^{2+} in MFe_2O_4 with suitable TM ions.

M^{2+}	Magnetic Nature in Bulk Form at room temperature	Application
Zn^{2+}	Paramagnetic	Gas desulphurisation (Jha et al., 1988; Cilleruelo et al., 1995)
Ni^{2+}	Ferrimagnetic	Magnetic shielding (Narang et al., 2021)
Co^{2+}	Ferrimagnetic, Highly Coercive	Permanent Magnets (Lamouri et al., 2020a)
Mn^{2+}	Ferrimagnetic	Soft Magnetic applications (Aslibeiki et al., 2016)

These materials are still being explored because of the induction of numerous phenomena and applications that arise because of nano dimensions (Mohapatra et al., 2019; Singh et al., 2023a; Hublikar et al., 2023). The main characteristic of ferrite nanostructures is the size dependence of their physical properties when synthesized in nanoregime. Especially, these ferrite nanostructures are known for their altered magnetic behavior compared to their bulk counterpart (Liu and Zhang, 2001; Fantauzzi et al., 2019). Paramagnetic zinc ferrite (Singh et al., 2008) may transform to superparamagnetic (Singh et al., 2008; Hasirci et al., 2019), ferromagnetic (Li et al., 2011) and ferromagnetic (Raghavan et al., 2015) at room temperature depending upon size. Reports are available showing soft magnetic properties of $CoFe_2O_4$ when size is reduced to nano dimension (Zorai et al., 2023). Figure 1 depicts the magnetic behavior of ferrite nanoparticles and thin films which shows strong size dependence. Kamble et al. (2015) observed that the room temperature magnetic nature of nickel ferrite nanoparticles of size 14 nm (NF03), 15 nm (NF04), 18 nm (NF05) and 22 nm (NF06) remain the same. All nanoparticles exhibit a ferromagnetic kind of magnetic ordering but their saturation magnetization increases from 25 to 45 emu/g as the particle size increases from 14 to 22 nm (Figure 1(a)). Copper ferrite (Kumar et al., 2019), magnesium ferrite (Šepelák et al., 2007) and cobalt ferrite (Lamouri et al., 2020) also exhibit the same behavior. Change in corecivity (H_c) with crystallite size is also reported for ferrite nanoparticles (Lu et al., 2014). Parameters such as retentivity (M_r), and anisotropy (K) are also influenced by the crystallite size (Peddis et al., 2012; Das et al., 2020). In the case of thin films too, size-dependent behavior of saturation magnetization (M_s) is reported as shown in Figure 1(b) (Dixit et al., 2012a).

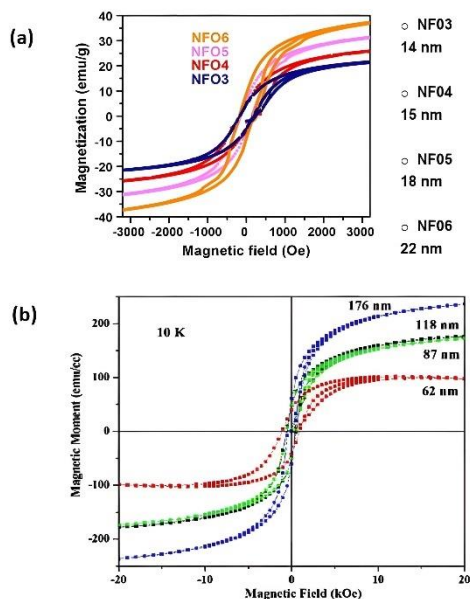


Figure 1. Magnetic Hysteresis curves of (a) nickel ferrite nanoparticles. Permission was obtained from Kamble et al. (2015) (Open access) and (b) nickel ferrite thin films. Reprinted with permission from Dixit et al. (2012a) (Open access).

Other characteristics of ferrite nanoparticles/thin films such as blocking temperature, T_B (Singh et al., 2012a; Singh et al., 2017) and Curie temperature, T_N (Sadeh et al., 2000; Fesharaki et al., 2016) also exhibit strong size dependence. Figure 2 summarizes the size dependence of various magnetic parameters for MnFe_2O_4 nanoparticles (Islam et al., 2020). All magnetic parameters exhibit a linear relation with the size of these nanoparticles. This figure also depicts that Yafet-Kittel (Y-K) canting angle and squareness (M_r/M_{sat}) is influenced by the size of nanoparticles.

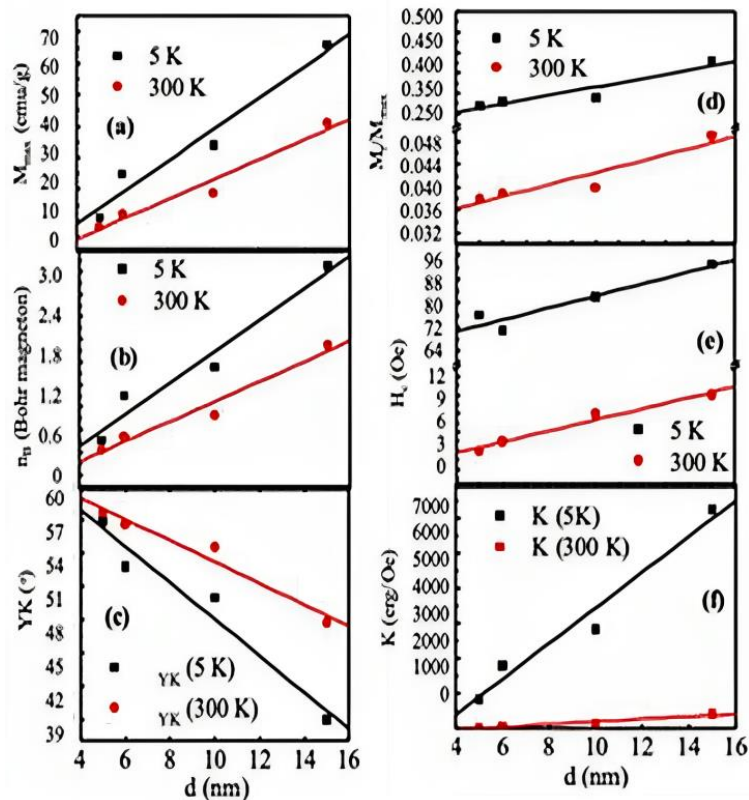


Figure 2. (a) Maximum magnetization (b) Bohr magneton, (c) Canting angles, (d) Squareness ratio, (e) Coercivity, and (f) Anisotropy with size variations of MnFe_2O_4 nanoparticles. Reprinted with permission from Islam et al. (2020).

In the same way, the optical behavior of ferrites is also affected by the crystallite size (Singh et al., 2010c; Jogi et al., 2022; Refat et al., 2022). Various ferrite nanoparticle exhibits absorbance which depends on the size of the ferrite nanoparticle. This causes a change in optical band-gap of ferrite material. An enhancement in the optical band-gap is reported for $\text{Ni}_2\text{Fe}_2\text{O}_4$ nanoparticles (Karmakar & Behera, 2020). CoFe_2O_4 nanoparticles also exhibit variations of optical band gap with crystallite size (Singh et al., 2020). Figure 3(a) shows the reduction in optical band-gap value with an increase of crystallite size. Similar behavior of optical band-gap is also reported by Massoudi et al. (2020) for $\text{Ni}_{0.6}\text{Zn}_{0.4}\text{Al}_{0.5}\text{Fe}_{1.5}\text{O}_4$ nanoparticles (Figure 3(b)). In the case of ferrite thin film, this parameter exhibits a strong dependence on film thickness. Figure 3(c) shows an inverse correlation between the optical band-gap and film thickness for $\text{Ti}_{0.5}\text{Li}_{0.5}\text{La}_{0.1}\text{Fe}_{1.9}\text{O}_4$ thin films (Abdelmoneim, 2010).

Figure 4(a) shows the ac conductivity behavior for cobalt ferrite nanoparticles of different sizes. This clearly shows the effect of particle size on the electrical behavior of ferrites (Yadav et al., 2017). DC conductivity of ferrite nanoparticles is influenced by the size of ferrite particles (Lahouli et al., 2019) as depicted in Figure 4(b) by the variation of conductivity with temperature.

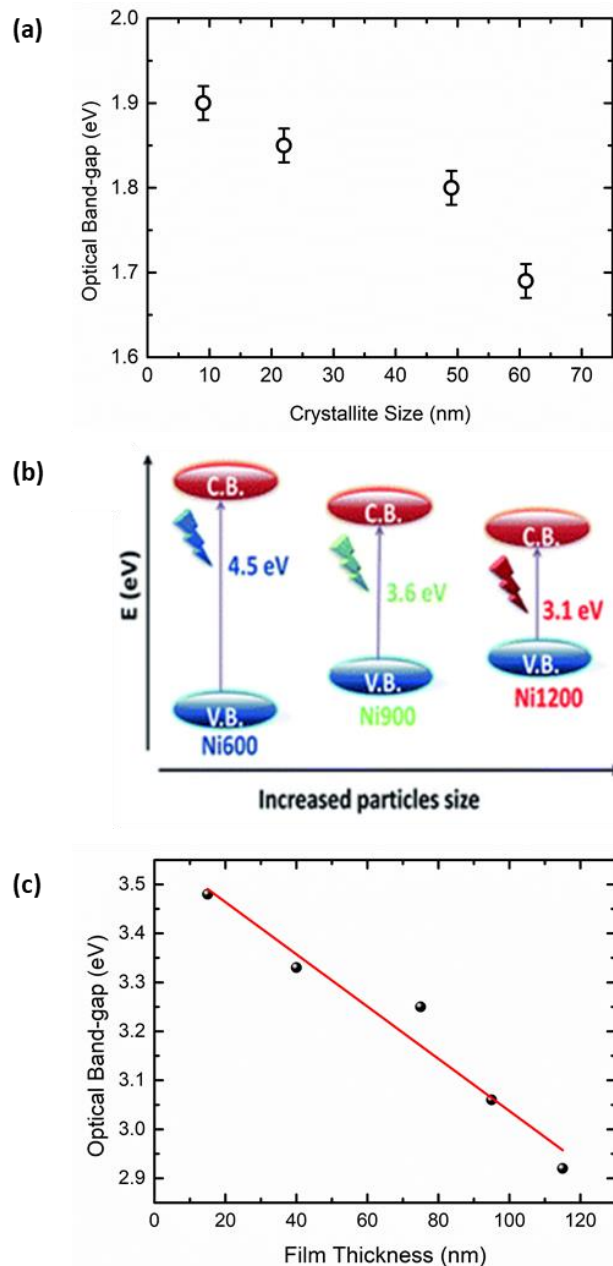


Figure 3. (a) Optical Band-gap for CoFe₂O₄ nanoparticles as a function of crystallite size. Permission obtained from RSC Advances (Singh et al., 2020) (b) Schematic representation of band structure at various particle sizes for Ni_{0.6}Zn_{0.4}Al_{0.5}Fe_{1.5}O₄ material. Preprinted with permission from Massoudi et al. (2020), Variation of optical band-gap with thickness for Ti_{0.5}Li_{0.5}La_{0.1}Fe_{1.9}O₄ thin films. Values of thickness and optical band gap are taken from Abdelmoneim (2010).

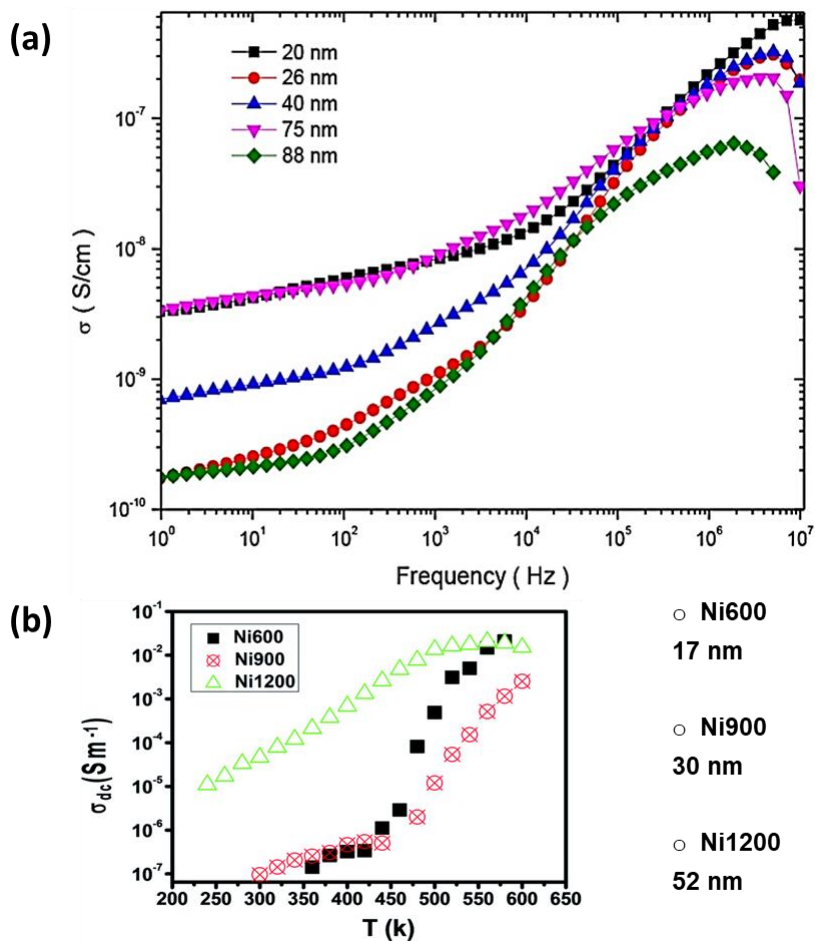


Figure 4. (a) Frequency dependent ac conductivity of cobalt ferrite nanoparticles of different sizes. Reprinted from Yadav et al. (2017) *Adv. Nat. Sci: Nanosci. Nanotechnol.* 8 045002 (Open access). The plot of DC conductivity versus temperature for $Ni_{0.6}Zn_{0.4}Fe_{1.5}Al_{0.5}O_4$ annealed at different temperatures. Reprinted with permission from Lahouli et al. (2019).

Not only the electrical behavior but dielectric behavior of ferrites is also affected by the size of ferrite nanoparticles. The dielectric behavior of $CoFe_2O_4$ nanoparticles of size 6 and 50 nm is depicted in Figure 5. This shows the increasing trend of dielectric permittivity with the decrease in crystallite size for $CoFe_2O_4$ nanoparticles (Vasundhara et al., 2013).

Thus, these examples show that size-dependent changes in physical properties are characteristics of ferrite nanoparticles. Depending upon the size, the physical behavior of ferrite having the same composition may differ (Mozaffari and Masoudi, 2014; Parmar et al., 2022a). The origin of size-dependent physical properties of ferrites is the result of a change in cation inversion in nanoregime (Siddique and Butt, 2010; Pham et al., 2023). A simple representation of various physical properties, their applications, size and cation distribution is shown in Figure 6 by taking the example of $CoFe_2O_4$ (Jauhar et al., 2016; Varma et al., 2016). Not only their physical behavior but their utility for particular applications is influenced by the size of ferrite nanoparticles (Noreen et al., 2017; Li et al., 2019).

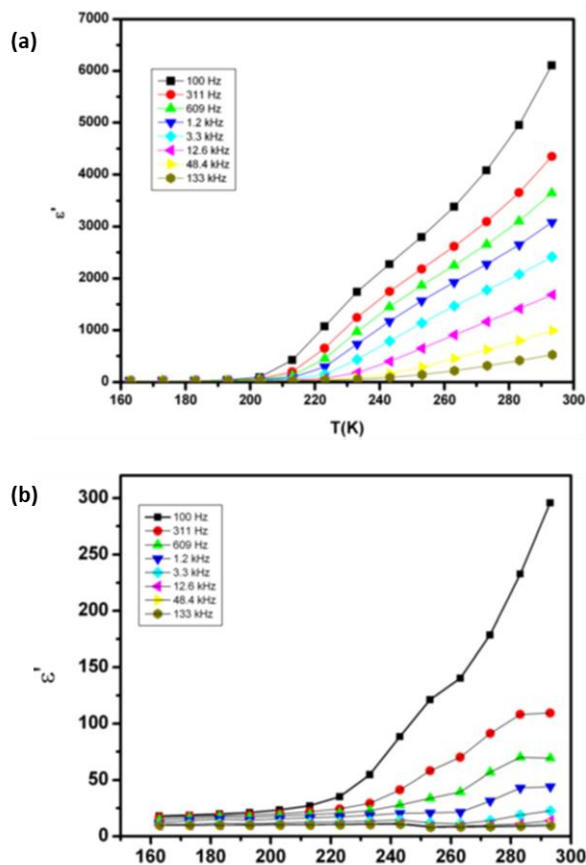


Figure 5. Variation of real part of relative permittivity with temperature in CoFe_2O_4 samples ((a) 6 nm sample and (b) 50 nm sample). Reprinted with permission from Vasundhara et al. (2013).

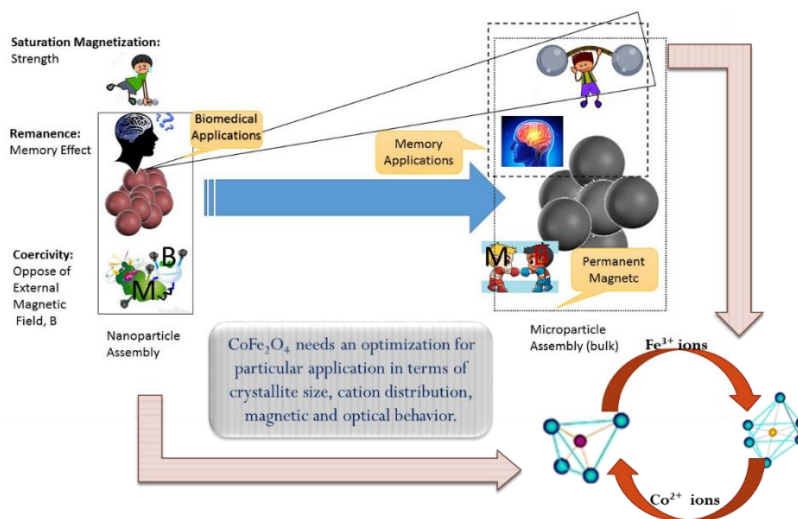


Figure 6. Schematic depicting the size dependent variation in magnetic parameters along with their applications and its correlation with cation distribution (Drawn based on the properties of CoFe_2O_4).

Apart from size dependence, doping of various elements is also able to change in physical properties of ferrites (Dixit et al., 2012b). Doping of particular ions too influences the crystallite and cation inversion. Thus, cation inversion is responsible for the changes in the properties of ferrites. There are certain methods such as laser irradiation (Tawfik et al., 2002; Ateia et al., 2019), ion implantation (Sharma et al., 2023a), gamma-irradiation (Raut et al., 2018) and heavy ion irradiation (Nongjai et al., 2023) which are well known to change the physical behavior of ferrites in a controlled way. This review focuses on the cation inversion manipulation and associated changes with swift heavy ion irradiation.

2. Swift Heavy Ions

Swift heavy ions (SHI) are well known to modify characteristics of the materials (Kuzmann et al., 2023; Wu et al., 2023) and devices such as spintronics (Singh et al., 2016; Zhao et al., 2019), electronic (Bharathi et al., 2016; Kumar et al., 2023) and sensing (Ramola et al., 2022). These ions have the potential to design ferrites for spintronic (Garg et al., 2023) and gas sensing applications (Bagwan et al., 2022). The basic characteristics of these ions and their production procedure can be found elsewhere (Kumar et al., 2022). These ions when penetrates through the materials lose energy into the medium. The energy loss per unit length in the medium is known as stopping power (S) which is estimated from the stopping and range of ions in matter (SRIM) code (Ziegler et al., 1985). This code also gives information of the projected range in the medium. This calculation shows that energy loss in the medium is determined by the composition of the material. Energy loss or stopping power in the medium is further divided into two categories- (1) electronic stopping, S_e and (2) nuclear stopping, S_n .

2.1 Choice of Ions and their Energy

Figure 7 shows the electronic stopping power and projected range of 100 MeV O^{7+} , 200 MeV Ag^{15+} and 150 MeV Si^{7+} for various ferrite materials (Ziegler et al., 2010). The projected range for O^{7+} is almost 4 times that of Ag^{15+} , however, it is almost twice compared to that Si^{7+} ions. This means a larger thickness material may be used to observe irradiation effects in the case of O^{7+} ion beams. On other hand, the electronic stopping power value is less compared to that of other ions. This indicates that it will cause less damage compared to other ions. To get an understanding of the damage process in these systems, the ratio of electronic stopping to nuclear stopping was determined (Ziegler et al., 2010). These values for 100 MeV O^{7+} , 200 MeV Ag^{15+} and 150 MeV Si^{8+} are collated in Table 2.

At first glance, these calculations show that the ion beam induced effects depend on the composition of ferrites and are supported by most of the studies carried on ferrites from 70's to early 90's (Pascard and Studer, 1997). In all cases, it is clear that S_e/S_n ratio is almost 10^3 (Table 1), hence, the inelastic collision of ions with target nuclei is the dominant mechanism (Péter, 1977; Chandramohan et al., 2007), if the irradiation is carried out by these ions of energy mention in the table caption. This shows that the order of S_e is greater than S_n which is the reason for the dominance of electronic energy loss over nuclear ones for all swift heavy ions in different ferrite samples (Weber et al., 2015).

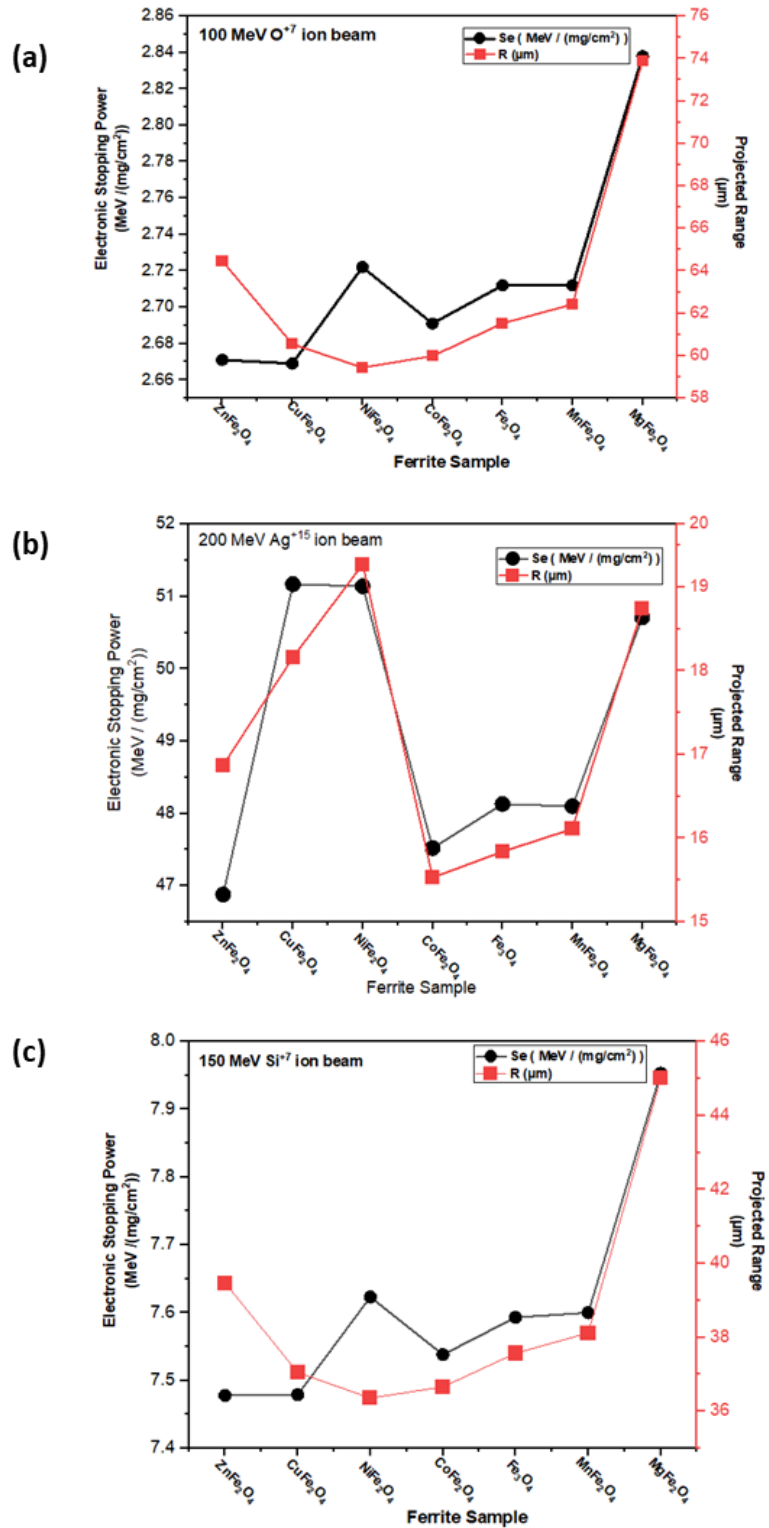


Figure 7. Electronic stopping power and projected range of various ferrites for (a) 100 MeV O^{7+} and (b) 200 MeV Ag^{15+} and (c) 150 MeV Si^{7+} ions. Values are estimated from SRIM code (Ziegler et al., 1985).

Table 2. Stopping power (S) and projected range (R) for various ions in ferrites. (Values are obtained from SRIM calculations).

Ferrite	Se (MeV / (mg/cm ²))	Sn (MeV / (mg/cm ²))	S _e /S _n Ratio (10 ³)	Nature of defects
100 MeV O ⁷⁺				
ZnFe ₂ O ₄	2.67	1.55 × 10 ⁻⁰³	1.76	Point defects
CuFe ₂ O ₄	2.67	1.51 × 10 ⁻⁰³	1.76	
NiFe ₂ O ₄	2.72	1.55 × 10 ⁻⁰³	1.76	
CoFe ₂ O ₄	2.69	1.52 × 10 ⁻⁰³	1.77	
Fe ₃ O ₄	2.71	1.54 × 10 ⁻⁰³	1.77	
MnFe ₂ O ₄	2.71	1.52 × 10 ⁻⁰³	1.78	
MgFe ₂ O ₄	2.84	1.59 × 10 ⁻⁰³	1.78	
200 MeV Ag ¹⁵⁺				
ZnFe ₂ O ₄	46.90	1.28 × 10 ⁻⁰¹	0.37	Columnar Defects
CuFe ₂ O ₄	51.17	1.33 × 10 ⁻⁰¹	0.39	
NiFe ₂ O ₄	51.14	1.33 × 10 ⁻⁰¹	0.38	
CoFe ₂ O ₄	47.52	1.28 × 10 ⁻⁰¹	0.37	
Fe ₃ O ₄	48.13	1.29 × 10 ⁻⁰¹	0.37	
MnFe ₂ O ₄	48.10	1.28 × 10 ⁻⁰¹	0.37	
MgFe ₂ O ₄	50.71	1.33 × 10 ⁻⁰¹	0.38	
150 MeV Si ⁷⁺				
ZnFe ₂ O ₄	7.48	5.14 × 10 ⁻⁰³	1.45	Point/cluster defects
CuFe ₂ O ₄	7.48	5.13 × 10 ⁻⁰³	1.46	
NiFe ₂ O ₄	7.62	5.26 × 10 ⁻⁰³	1.45	
CoFe ₂ O ₄	7.54	5.16 × 10 ⁻⁰³	1.46	
Fe ₃ O ₄	7.59	5.21 × 10 ⁻⁰³	1.46	
MnFe ₂ O ₄	7.60	5.17 × 10 ⁻⁰³	1.47	
MgFe ₂ O ₄	7.95	5.38 × 10 ⁻⁰³	1.48	

Additionally, the S_e value gives information on the nature of defects formed in the system during irradiation. If S_e is less than the threshold value, then point defects are created in the system, however, if it is larger than this value, then columnar defects are produced in the system. In the case of $S_e \sim S_{eth}$, both point/columnar defects are observed in the system. In the case of ferrite, the threshold value is almost 13 keV/nm as mentioned in previous work (Kumar et al., 2005). Hence, oxygen ions are known to produce point defects in ferrite materials (Singh et al., 2013). However, modifications are governed by the production of latent tracks/columnar defects in the case of Ag ions (Singh et al., 2014). In the case of Si ions, point/cluster defects are expected to induce effects in the system.

2.2 Sample Choice

In case of heavy ion irradiation, another criteria is related with the thickness of the target materials, which should always be less than the projected range. Generally, films are ideal choices for heavy ion irradiation studies as they have thickness ranging from few nm to several hundred nm, which is always less than the projected range (Panda et al., 2021; Vershney et al., 2023). Though ions penetrate through the samples, however, they reside in the substrate attached to the film (Figure 8(a)). Hence, proper care should be taken while studying the magnetic behavior of these samples.

Sometimes pellets of thickness 2-3 nm / single crystals are used for irradiation experiments (Sharma et al., 2023b; Aithal, 1997), however, post-irradiated pellets are a mixture of irradiated and unirradiated counterparts. Thus, information specific to the irradiation phenomena on the bulk physical properties is hard to obtain. In this case, only surface-related phenomena can be investigated (Figure 8(b)). To investigate, bulk behavior, very high energy of ion beam will be required in this case (Studer et al., 1993).

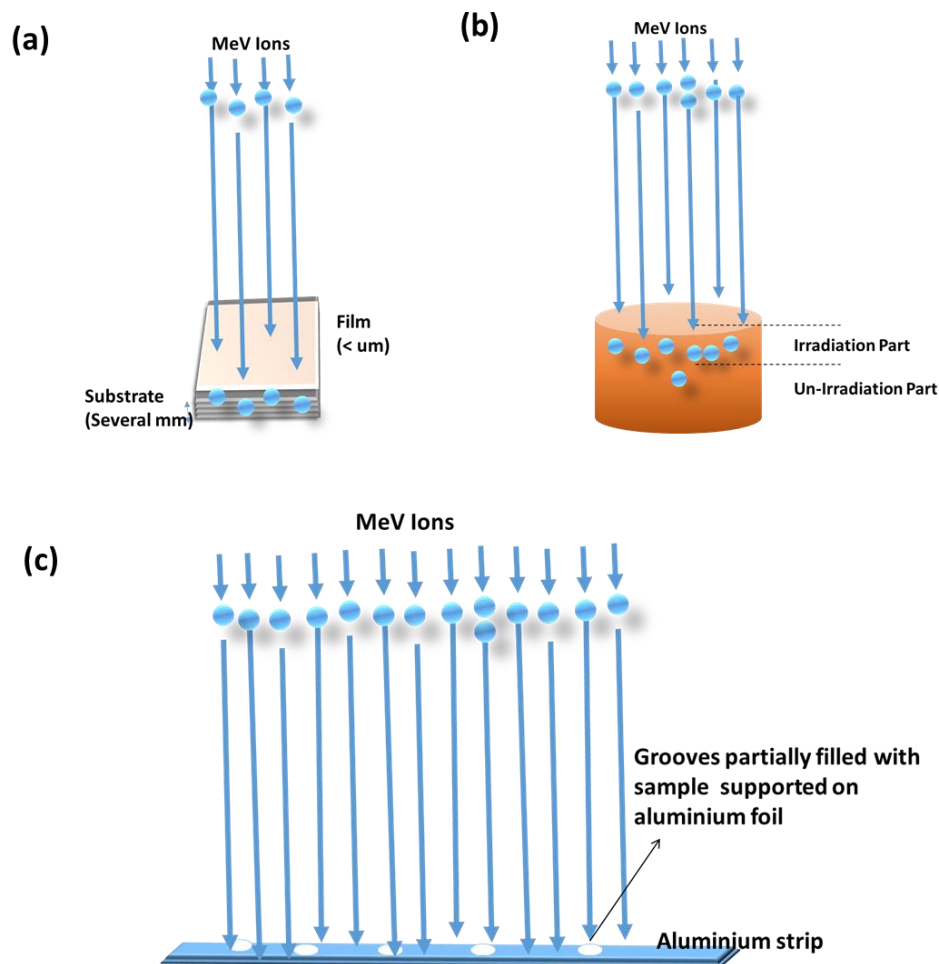


Figure 8. Irradiation procedure for (a) thin films, (b) pellets and (c) nanoparticles/powder samples.

In the case of powder materials, irradiation experiments are tricky and samples are prepared by mixing with polyvinyl alcohol (PVA) in grooves of a specific size (Srivastava et al., 2010). In these grooves, materials have volume less than the area of the groove multiplied by the projected area. Samples after irradiation are carefully taken out from the grooves and heated at 100°C for removal of PVA (Figure 8(c)).

4. Experimental Results for Swift Heavy Ion Irradiation Induced Effects

4.1 Structural Properties

Heavy ions are known to induce crystallization in few systems such as ZrC films (Jiang et al., 2021), MAX nanostructures (Cannavò et al., 2021), however, most of the materials exhibit crystalline disorder after irradiation (Sathyavathi et al., 1999; Ai et al., 2019; Pang et al., 2021). Ferrites are known to be stable materials, hence, their crystalline phase remains unchanged under heavy ion irradiation as reported in most of the work. However, they too exhibit a reduced degree of crystallisation under the effect of the intense ion beam. This is evident from the X-ray diffraction studies of ferrites under heavy ion irradiation (Dixit et al., 2012c; Dolia et al., 2012; Satalkar et al., 2016; Raghuvanshi et al., 2019). This motivated a number of researchers to work in this direction and the effect of swift heavy ion irradiation on the structural properties of spinel ferrite nanoparticles was investigated by various researchers. Kumar et al. observed an increase in

crystalline disorder on irradiating $\text{CoFe}_{1.90}\text{Dy}_{0.10}\text{O}_4$ nanoparticles with 100 MeV O^{7+} ions at varying fluences. It was inferred that with higher fluence more energy was deposited inside the material which resulted in deterioration of crystalline structure. Figure 9(a) shows XRD patterns of Dy doped CoFe_2O_4 nanoparticles. These patterns exhibit that the crystalline phase remains unchanged under irradiation of 100 MeV O^{7+} ions (Kumar et al., 2015a). However, onsets of decreased crystalline order are observed from the decreased intensity of XRD peaks. Similar behavior of XRD patterns is observed for zinc ferrite nanoparticles under the same ion beam (Figure 9(b)).

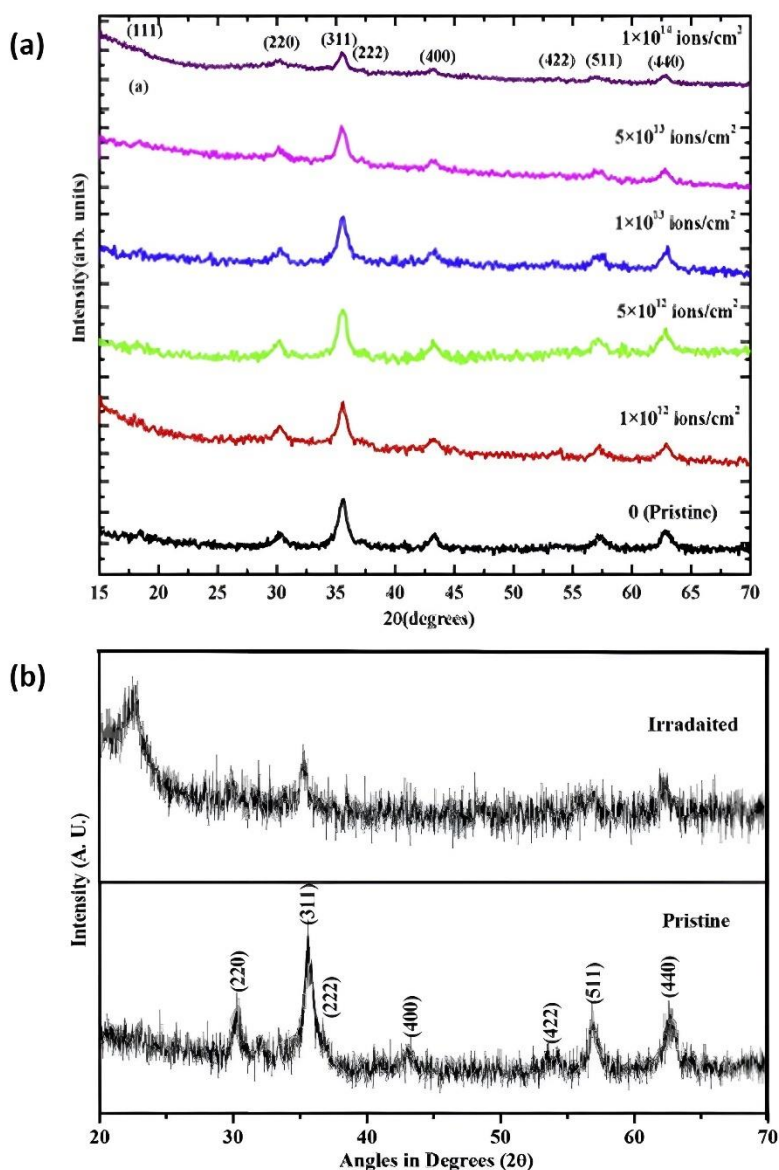


Figure 9. (a) XRD patterns of Dy doped CoFe_2O_4 nanoparticles under irradiation of 100 MeV O^{7+} Reprinted with permission from Kumar et al. (2015a), (b) XRD patterns of pristine and irradiated ZnFe_2O_4 (5×10^{13} ions/cm²) reprinted with permission from Singh et al. (2010b).

Satalkar et al. (2016) in their study of effect of 80 MeV O^{6+} ions on manganese-zinc ferrite nanoparticles also observed the rearrangement of cations at tetrahedral and octahedral sites (Satalkar et al., 2018). Similar results were reported by Raghuvanshi et al. (2019) and Dhyani et al. (2020). Thus, from several studies, it can be inferred that cation redistribution can take place on irradiation. Zinc ferrite nanoparticles annealed at 300, 500, 800, and 1000°C were subjected to irradiation with 200 MeV Ag^{15+} ions and studied by Singh et al. (2012b) XRD analysis showed that the presence of impurity peaks after irradiation for samples annealed at 300 and 500°C. However, nanoparticles annealed 800 and 1000°C were in pure cubic spinel phase even after irradiation. Increased crystallite size was observed for all samples upto a certain fluence due to increase in local temperature as suggested by Kaoumi et al. (2008). At higher fluence crystallite sizes decreased due to the splitting of crystallites. The properties of zinc ferrite thin films formed via RF sputtering technique and irradiated with 100 MeV Ag ions were studied by Raghavan et al. (2017). Decrease in crystallite size was observed on irradiating the pristine samples. It was attributed to the amorphisation of the irradiated material which was explained on the basis of the thermal spike model. When the pristine thin films were irradiated, high-temperature localized zones can melt the material which on rapid cooling led to partial amorphisation of the material. On further irradiating the material with higher fluence, an increase in crystallite size was observed which was attributed to the breaking of crystallites and joining again to form larger crystallites. Nongjai et al. (2015). observed decrease in the intensity of peaks corresponding to planes in the XRD pattern with increase in irradiation fluence on cobalt ferrite thin films. It was also attributed to the partial amorphisation of the material. Progressive amorphisation with increasing fluence in the material was also observed by Dixit et al. (2011) in their study of nickel ferrite thin films irradiated with 200 MeV Ag^{15+} ions. Figure 10 shows that crystalline Nickel ferrite thin films become amorphous after irradiation (Dixit et al., 2011). Balaji et al. (2011) in their study of 150 MeV Ni^{11+} ions irradiated tetragonal phase $CuFe_2O_4$ thin films observed complete amorphisation at higher fluences.

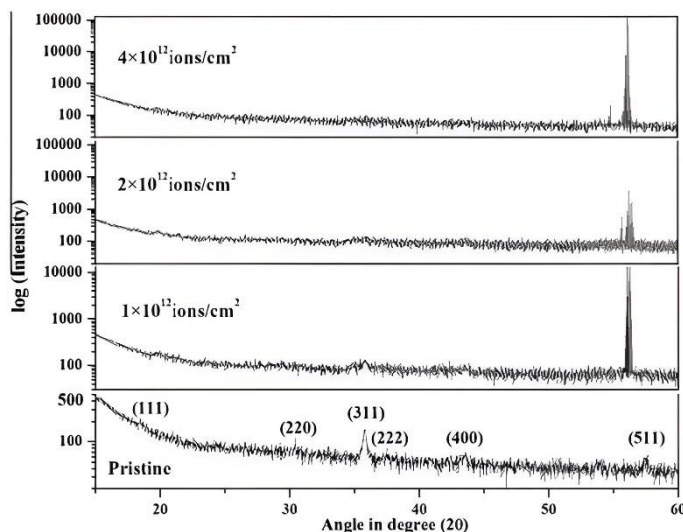


Figure 10. XRD patterns of the pristine and irradiated films of thickness 150 nm at fluences of 1×10^{12} , 2×10^{12} and 4×10^{12} ions/cm² of 200 MeV Ag^{15+} ions. Reprinted with permission from Dixit et al. (2011).

Thus, it can be inferred on irradiating the spinel ferrite nanoparticles and thin films with swift heavy ions, deterioration of the crystalline structure, cation redistribution among lattice sites and amorphisation of the material takes place. This leads to a change in crystallite size after irradiation depending on the crystallite size of pristine ferrite as can be seen from Figure 11 (Singh et al., 2011; Singh et al., 2012b).

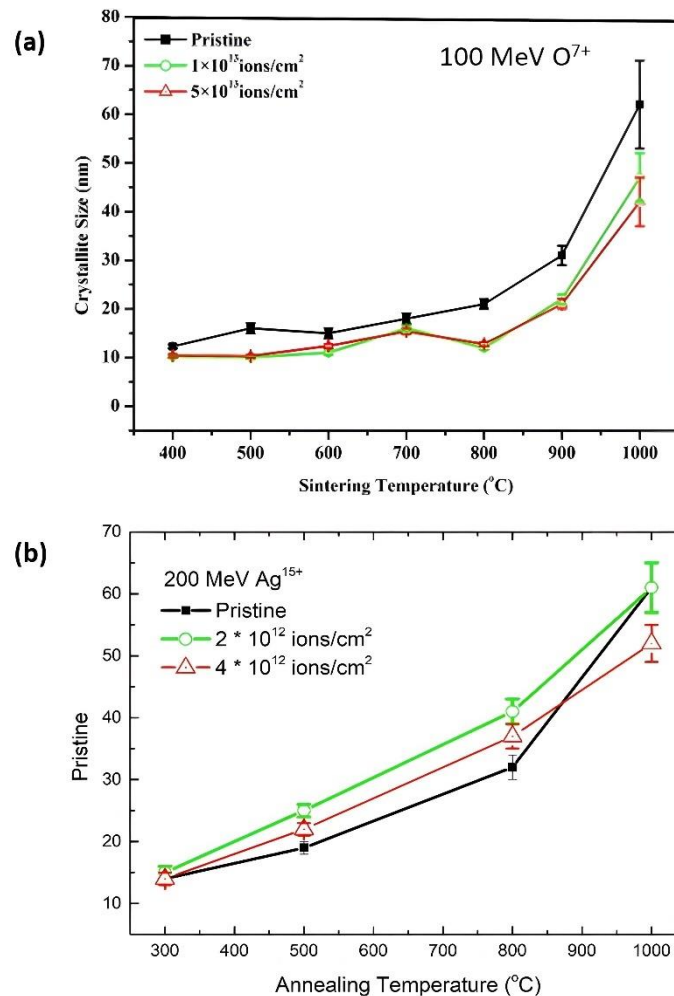


Figure 11. Crystallite size of zinc ferrite nanoparticles under different fluences of (a) 100 MeV O^{7+} reprinted with permission from Singh et al. (2011), and (b) 200 MeV Ag^{15+} . reprinted with permission from Singh et al. (2012b).

4.2 Magnetic Properties

Heavy ion irradiation is known to modify the magnetic behavior of various systems (Kato et al., 2005). Hence, this tool is widely applied to ferrite nanoparticles and modification is associated with either point or columnar defects. The dissipation of energy in the materials during ion beam irradiation is responsible for the formation of latent tracks. It induces the rearrangement in the distribution of cations, the oxygen as well as inversion parameters and produces significant changes in magnetic properties of a typical ferrite (Nongjai et al., 2015). The rearrangement of cations in ferrites depends on the type of selected ions as well as on the energy and fluence range of the ions. The variation in the magnetic parameters (M_s , H_c) of irradiated ferrites in comparison to pristine samples are shown in Table 2.

Ion beam irradiation leads the formation of a non-magnetic layer (also known as dead layer) depending on the ion beams. The non-magnetic layer plays a significant role in the variation of magnetic parameters of ferrites upon SHI irradiation (Raghuvanshi et al., 2019). The canting of the surface spin, high surface anisotropy and heavy ion irradiation are the general causes which leads the formation non-magnetic layers on the ferrites surface (Maehara et al., 2005; Li et al., 2010). Irradiation leads to the formation of a dead

layer on the surface, thus affecting magnetic properties, via spin canting at the surface. The increased thickness of dead layer is ascribed to the effect of spin disorder at the surface of the particle due to irradiation. The thickness (t) of dead layer can be evaluated using the following expression (El-Sayed et al., 2017):

$$t = \frac{D}{6} \left(1 - \frac{M_s}{M_B} \right) \quad (1)$$

where, D , M_s and M_B is grain size, saturation magnetization, bulk magnetization of the ferrite sample, respectively.

The mathematical expression (1) suggests the inverse relationship between M_s and t , which is due to the effect of surface spin disorder upon SHI irradiation. The decrease in the value of M_s can be attributed to the low concentration of Fe^{3+} ions on the B site or the reduction in the inversion parameter upon SHI irradiation (Parmar et al., 2022b). The reduction in oxygen position parameter (u), which is a measure of disorder leads to diminishing strain, hence reducing the coercivity of the sample due to irradiation. Further, It can be verified with the corresponding changes in the cation distribution.

The SHI-irradiated ferrite sample is composed of two regions: one with spins that are aligned with the latent track and another with spins that are randomly distributed. In addition, irradiation may result in a cation inversion, which raises the temperature of the materials exposed to radiation. As a result, the production of directed spin zones in the direction of latent tracks and induced cation inversion is observed.

Also enhanced magnetization and the development of hysteresis were observed after irradiation (Balaji et al., 2011, Raghavan et al., 2017). The physical and chemical characteristics of the irradiated ferrites also vary as a result of extended defective patches and changed cation inversion.

At low fluence, as the accumulation of defects in the ferrite sample induced by SHI irradiation, results in an increase in H_c while the reduction in M_s . It is mainly because of the significant increase in the number of grain boundaries which act as pinning sites of the magnetic domain. Si^{9+} ion-irradiation results in a decrease in oxygen positional parameter, strain, and saturation magnetization as well as an increase in inversion degree, and a reduction in dead-layer thickness. The modification in the A-O-B, A-O-A, and B-O-B super-exchange interaction is also observed upon Si irradiation. The analyzed samples fall in an overlap region between many domains or inside a single domain, according to the linear dependency of coercivity on grain diameter (Parmar et al., 2022b). At high fluence, the electron-phonon coupling is responsible for the change in crystal orientation and the dissociation. After irradiation, the temperature of the sample's surface is enhanced due to the very high energy of incident ion beams (Singh et al., 2022). The thermal spike model suggests that the time required to transfer the energy from an incident ion to an excited electron is far less than the normal time scales of lattice vibrations. The electrons instantly receive the energy from the incident ion and transfer to the excited states (Ghosh et al., 2006) which leads to a momentary increase in temperature of the confined zone along the path of the ion and heat is transmitted to the lattice in the form of energy via electron-phonon coupling. The enhanced local temperature results in the variation in the crystallinity of the sample and alterations in various structural properties.

The nature of strain-induced defects, production mechanisms, atomic motion, relaxation, thermal spike, coulomb explosion, surface morphology, the electronic-excitation effects, etc. is used to interpret changes in magnetic properties after irradiation (Kumar et al., 2009; Singh et al., 2010a; Sun et al., 2012). The structural analysis, surface morphology, and magnetic characteristics of ferrite nanoparticles showed a significant variation upon irradiation, which was supported by a number of ion irradiation experiments. A few of them are summarized in Table 3.

Table 3. Magnetic characteristic of SHI irradiated ferrite materials.

Ferrite	Magnetic Parameters			SHI		Ref.
	D (nm)	M _s (emu/g)	H _c (Oe)	Ion	Fluence (ions/cm ²)	
ZnFe ₂ O ₄	16	7.8	~	100 MeV O	0	Singh et al. (2010a)
	10.3 ± 0.3	6.6	~		1 × 10 ¹³	
	12 ± 0.2	~	18	90 KeV, Ne	0	Gafton et al. (2016)
	13 ± 0.8	~	74		3 × 10 ¹⁴	
	17.4	0 μB	~	80 MeV, O	0	Satalkar et al. (2018)
15.7	8.5 μB	~	1 × 10 ¹¹			
NiFe ₂ O ₄	42.82	33.02	157	120 MeV, Si	0	Sharma et al. (2018)
	36.33	20.32	163		1 × 10 ¹²	
Ni _{0.5} Zn _{0.5} Fe ₂ O ₄	33.10	49.68	85.45	120 MeV, Si	1 × 10 ¹²	Sharma et al. (2018)
	32.51	18.60	89.0			
Ni _{0.65} Zn _{0.375} In _{0.25} Ti _{0.025} Fe _{1.7} O ₄	6.8	~	~	190 MeV	0	Rao et al. (2006)
	5.2	~	~	Ag	1 × 10 ¹³	
MnFe ₂ O ₄	10.04	5	~	80 MeV, O	0	Satalkar et al. (2016)
	9.98	~	~		1 × 10 ¹³	
Mn _{0.75} Zn _{0.18} Fe _{2.07} O ₄	3.9	~	~	190 MeV,	0	Rao et al. (2006)
	2.4	~	~	Ag	1 × 10 ¹³	
MgFe ₂ O ₄	34.7	23.8	106	120 MeV, Si	0	Raghuvanshi et al. (2019)
	31.0	15.6	103		1 × 10 ¹¹	
CuFe ₂ O ₄	10	5.50	22.698	150 MeV, Ni	0	Balaji et al. (2011)
	~	4.195	54.875		5 × 10 ¹⁴	

4.3 Optical Behavior

Though the optical behaviour of ferrite is an important characteristic but it is rarely reported for irradiated nanoparticle systems. To investigate the effect on the optical properties of irradiated zinc ferrite system, optical measurements were performed on the zinc ferrite nanoparticles having crystallite sizes of 10 (ZF300), 16 (ZF500), 21 (ZF800) and 62 nm (ZF1000). UV-VIS absorption spectra of the pristine (P) and irradiated (I) samples recorded at room temperature are shown in Figure 12. All the spectra exhibit strong absorption in the UV-region which is characteristic of ferrites (Muret, 1974). This effect is observed for both pristine and irradiated counterpart

The absorption coefficient, α , for both the pristine and irradiated samples was determined by using the relation (Fox, 2001; Joshi et al., 2003).

$$\alpha = 2.303 \frac{\text{Absorbance}}{t}$$

t is the thickness of the cuvette ($t=1\text{cm}$).

Figure 13 shows α versus $h\nu$ plot for the pristine and irradiated samples. We have estimated the optical band gap of samples by using the Tauc's relation (Tauc, 2012). The optical gap from this relation was estimated by plotting $(\alpha h\nu)^{1/m}$ versus $h\nu$ and by extrapolating a straight line for $m=1/2$ (as elaborated by Singh et al., 2010b). The value of the optical band-gap varies from 3.8 eV to 3.65 eV with respect to the sintering temperature for the pristine samples. The detailed investigation related to this effect has been attributed to the quantum confinement and discussed elsewhere (Singh et al., 2010b). The values of optical band-gap were found to increase to a value of ~4.37 eV in the case of irradiated samples as compared to the pristine sample. The values of optical band-gap remain constant for irradiated materials at all sintering temperatures. The values of the band-gap for irradiated ferrites are of the order of 4.4 eV. This suggests a dominant contribution from intra-d and d-s transition to the UV-VIS spectra (Schmitt et al., 1987).

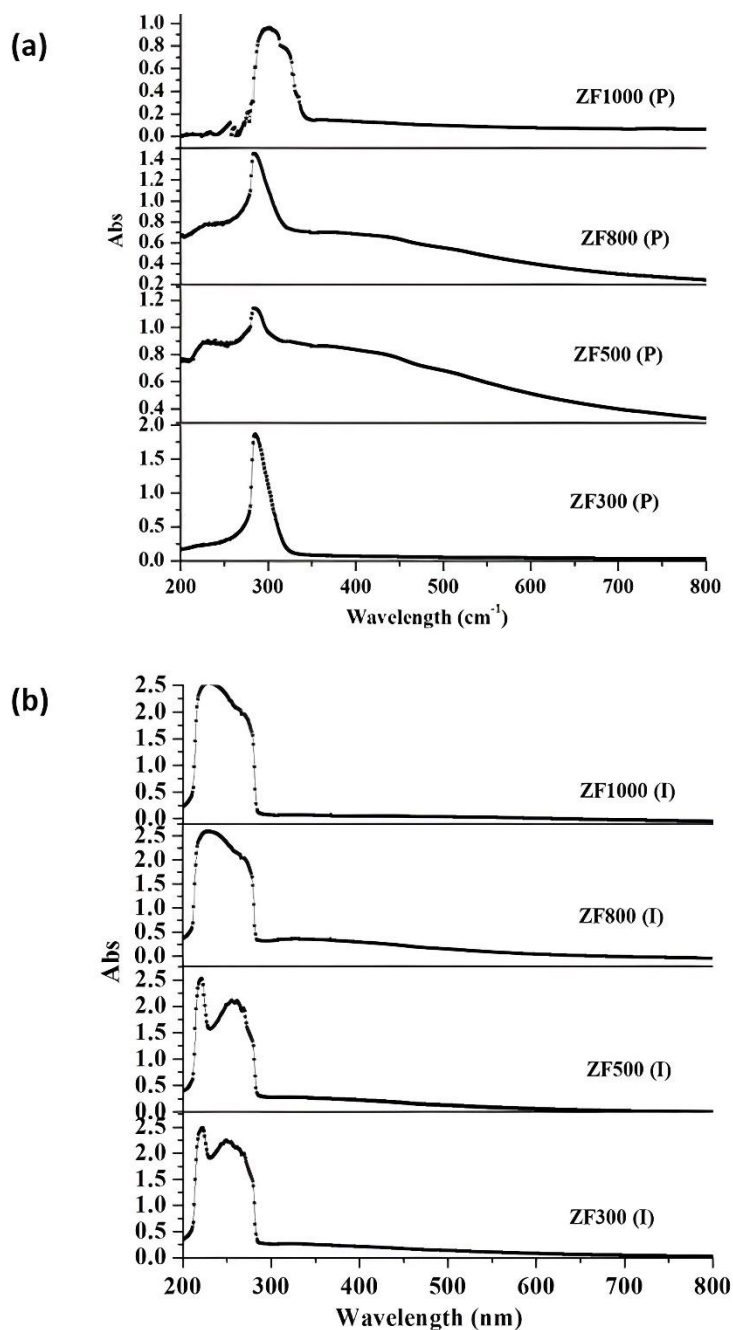


Figure 12. UV-Vis absorption spectra of the (a) pristine (P) Reprinted with permission from Singh et al. (2010b), and (b) irradiated (I) counterpart at room temperature for ZnFe₂O₄ nanoparticles under 100 MeV O⁷⁺ ions.

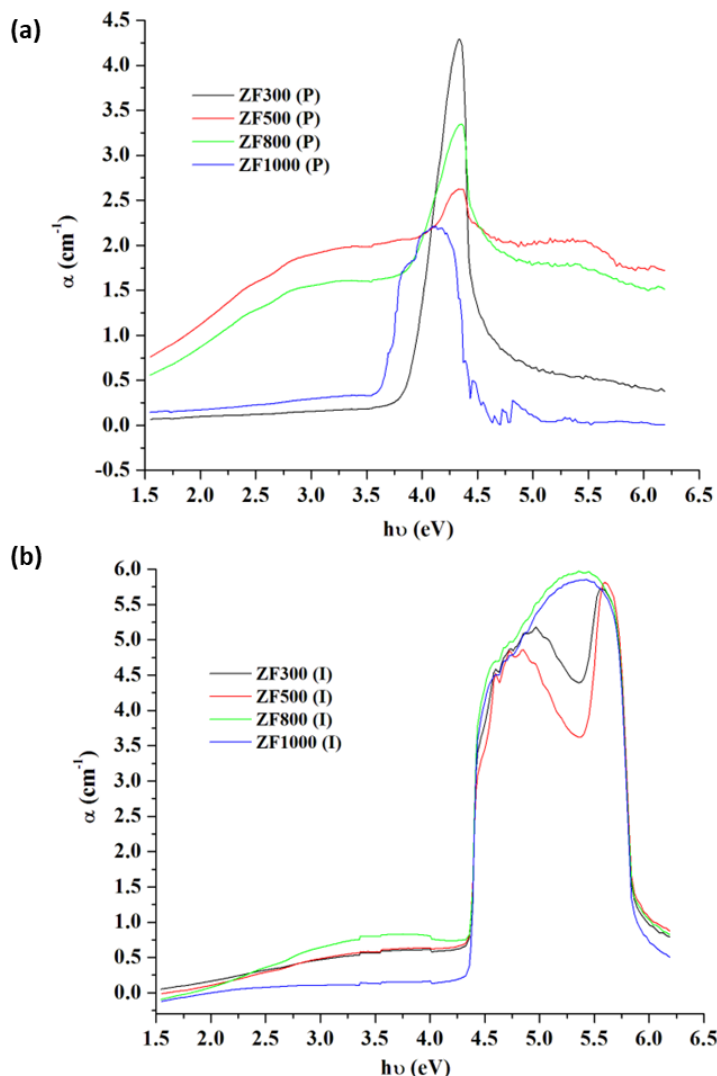


Figure 13. (a) α versus $h\nu$ for the (a) pristine (P), and (b) irradiated (I) samples.

The particle size of irradiated ferrites is small as compared to that of pristine as shown in Figure 11(a), therefore optical band-gap increases after irradiation. Besides this, the presence of point defects induced by irradiation in ferrites may cause an effective mass of carriers to decrease, resulting in enhancement in the band-gap of these ferrites (Singh, 2002). Hence, the increase of optical band-gap may be attributed to the cumulative effect of both effects. To elucidate the effect of defects on UV-VIS spectra, the position of maxima and full width at half maximum (FWHM) of the absorption peak were estimated and shown in Table 4. The FWHM of the irradiated samples is very much large (~ 1.38 eV) as compared to the pristine (~ 0.33 eV). This large value of FWHM is attributed to the increase in defect density in the case of irradiated samples. Similar behavior of irradiated material was also depicted for In_2O_3 on the basis of photoluminescence spectroscopy (Tripathi & Rath, 2014). The maxima of absorption peaks are of the order of 4.34 eV for the pristine samples and these shifted to a value of ~ 5.5 eV for the irradiated samples. In the case of irradiated samples, we also observe maxima at 4.95 eV for the sample ZF300 (I) and at 4.76 eV for

ZF500 (I). This shows the formation of sub-levels in the small-sized irradiated samples, thereby affecting the optical behaviour of irradiated nanoferrites.

Table 4. Maxima, FWHM of UV-VIS absorption peak for the pristine (P) and irradiated (I) zinc ferrite at different sintering temperatures.

Sintering Temperature (°C)	Crystallite size (nm)	Maxima (eV)		FWHM (eV)	
		P	I	P	I
300	10	4.34	4.95, 5.56	0.34	1.39
500	16	4.34	4.76, 5.60	0.25	1.40
800	21	4.34	5.36	0.33	1.39
1000	62	3.75	5.36	0.60	1.38

Apart from the changes in structural, magnetic and optical properties, few reports are available showing the modification in dielectric behavior of ferrites under heavy ion irradiation (Dolia et al., 2012, Dolia et al., 2013). Fe₃O₄ thin films are investigated to observe modification in electrical behavior under 190 MeV Ag¹⁵⁺ ions (Kumar et al., 2006). Both the electrical and dielectric behavior are subjected to changes in microstructure, cation redistribution. Thus, almost reports studying the irradiation induced effects strongly emphasize the role of cation redistribution as well as the change in the size of nanoparticles under heavy ion irradiation. This behavior of ferrites corroborates the behavior of native ferrite. However, research work from our group confirms the possibility of cation redistribution without changing crystallite size. These are discussed in next section.

5. Heavy Ion Irradiation Towards Control of Cation Inversion without Changing Size

Thus, these studies clearly depict variation of various properties with heavy ion irradiation and depict the usefulness of heavy ion irradiation. These studies show that size dependent behavior of ferrites is critical even though they are subjected to external effects. However, irradiation carried out on the zinc ferrite nanoparticles shows the reduction of cation inversion factor from 0.56 to 0.24 as estimated from in-field Mössbauer spectroscopy after irradiation from 100 MeV O⁷⁺ ions. In this case, crystallite size remains to be same (~10 nm) after irradiation (Singh et al., 2010b). Similar behavior is reported for zinc ferrite nanoparticles of almost 12 nm using X-ray absorption fine structure measurements (EXAFS) and X-ray diffraction measurements (Singh et al., 2018). Fe K-EXAFS measurements reveal decrease of cation inversion from 0.39 to 0.24 (Figure 14).

In another work from our group, we observed that the crystallite size of CoFe₂O₄ remains almost the same within experimental error under the different fluence of 100 MeV O⁷⁺ ions (Figure 15).

Mössbauer spectroscopy studies carried out on these sample shows changes in cation occupancy which results modification in cation inversion parameters (Kumar et al., 2015b). Thus, these studies may provide an opportunity to researchers to think of heavy ion irradiation with ferrite in different ways. It can provide a scope to manipulate ferrite characteristics without altering crystallite/particle size. Thus, heavy ions can persist a new way to design ferrite for different applications.

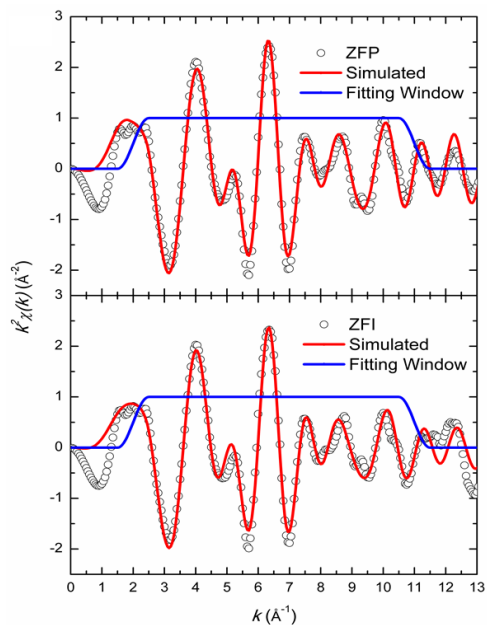


Figure 14. Simulated Fe K-edge EXAFS spectra of pristine (ZFP) and irradiated (ZFI) zinc ferrite. Reprinted with permission from Singh et al. (2018).

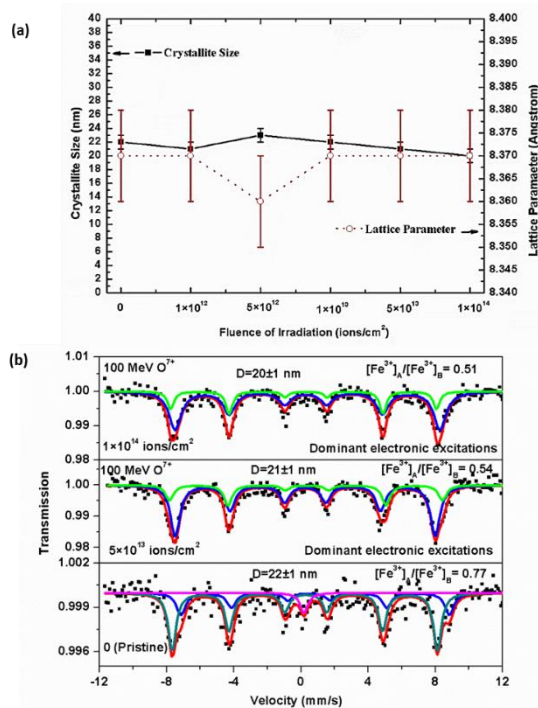


Figure 15. (a) Crystallite size and lattice parameter as a function of the fluence of irradiation (b) Mössbauer spectra of pristine and irradiated ferrite sample showing cation redistribution. Permission is required from Kumar et al. (2015b).

6. Conclusion

Thus, we have provided a critical and in-depth review of irradiation studies in ferrites. Ferrite which is well known to exhibit size-dependent behavior shows this behavior even when subjected to external tools such as swift heavy ion irradiation. Recent research also shows that it can be an effective tool to control cation occupancy in ferrites without changing the crystallite size. This can ultimately be used to tailor the physical properties of ferrites by keeping crystallite size as a constant parameter.

Conflict of Interest

The authors confirm that there is no conflict of interest to declare for this publication.

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