The effect of CoFe$_2$O$_4$, CuFe$_2$O$_4$ and Cu/CoFe$_2$O$_4$ nanoparticles on the optical properties and piezoelectric response of the PVDF polymer

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Abstract
Cobalt ferrite, Copper ferrite and cobalt doped copper ferrite nanoparticles have been synthesized and characterized using different characterization methods (XRD, FTIR and FESEM). The prepared nanoparticles have been used as promising fillers of the polyvinylidene fluoride (PVDF) polymer. The PVDF/(Cu–CoFe$_2$O$_4$, CoFe$_2$O$_4$, and CuFe$_2$O$_4$) nanocomposites films have been prepared via a simple solution casting technique. The optical properties and the piezoelectric response of the prepared nanocomposite films have been studied. This study showed that Cu–CoFe$_2$O$_4$ and CoFe$_2$O$_4$, have enhanced the interfacial polarization density and dielectric constant. The prepared nanofillers reduced the PVDF band gap energy value. The optical conductivity value of PVDF/(Cu–CoFe$_2$O$_4$ and CoFe$_2$O$_4$) increased five times compared with the pure PVDF. Also, an increase in the piezoelectric response has been recorded by adding the nano-fillers to the pure PVDF.

Keywords Pizoelectric · PVDF · PFM · Cobalt ferrite · Copper ferrite · Cobalt doped copper ferrite · Nanoparticles

1 Introduction
Energy harvesting or renewable energies, such as solar energy, wind energy, vibrational energy, and ocean energy, have been used recently as alternative energy to the usual forms of energy that pollute the environment [1]. One of the most important energy harvesting energies is piezoelectric technology. Piezoelectric materials could transfer energy between the electric field and force field. The piezoelectric polymer has been used as an excellent piezoelectric material for its excellent piezoelectricity and flexibility [2]. Compared with other polymer materials such as nylon and polyvinyl chloride, the polyvinylidene fluoride (PVDF) has better piezoelectric properties. (PVDF) is a low-density, non-reactive, easier to process, lightweight, chemical stable, flexibility to accept changing weather conditions, and has a much lower price [3–5]. The PVDF films are very flexible, it can be compatible with every kind of matrix material. PVDF filled with conducting materials such as metal nanoparticles, carbon nanotubes, and carbon fibers have recently been used to get higher composite materials based on the percolation phenomenon [6, 7].

PVDF is not only piezoelectric, but it also has excellent pyroelectric, flexoelectric, and dielectric properties. Ferroelectric materials could be considered a subclass of piezoelectric, a class of dielectric materials, and pyroelectric materials. Ferro electricity is the property in which certain materials can possess a spontaneous electric polarization by the application of an external electric field, this polarization can be reversed yielding a hysteresis loop [8]. A lot of various works have been carried out in exploring ferroelectric materials and their applications, such as the ferroelectric field-effect transistor, piezoelectric sensors, ferroelectric random-access memory. [8].

The incredible properties of CoFe$_2$O$_4$, CuFe$_2$O$_4$, and Cu–CoFe$_2$O$_4$ as high magnetic properties, environment-friendly, high conductivity, phase stability and outstanding photochemical, etc., make these nanoferrites one of the most researched among the numerous spinel systems [8].
The present work aims to obtain a new nanocomposite film, which is quite flexible, freestanding, and combines the excellent properties exhibited by both PVDF and the prepared nanoparticles. Consequently, PVDF films were prepared by a simple solution casting technique [9] selecting dimethyl formamide (NMP) as the solvent while, CoFe$_2$O$_4$, CuFe$_2$O$_4$, and Cu–CoFe$_2$O$_4$ nanoparticles have been used as the fillers. The effect of the nanofillers on the optical properties, dielectric constant, and piezoelectric response of the PVDF polymer is investigated.

2 Experimental techniques

2.1 Materials

Poly (vinylidene fluoride) (PVDF) powder, N-Methyl-2-Pyrrolidone (NMP, 99.5% of purity) (Merk Chemical, India), iron nitrate Fe(NO$_3$)$_3$·9H$_2$O (99%), cobalt nitrate Co(NO$_3$)$_2$·6H$_2$O, copper nitrate Cu(NO$_3$)$_2$·6H$_2$O (99%) and citric acid (C$_6$H$_8$O$_7$), were used as metal precursors. All materials were purchased from (Sigma Aldrich, USA).

2.2 Samples preparation methods

2.2.1 Nano powder preparation

CoFe$_2$O$_4$, CuFe$_2$O$_4$ and Cu–CoFe$_2$O$_4$ nanoparticles were synthesized via the citrate auto-combustion method. The stoichiometric ratios of cobalt nitrate, copper nitrate, iron nitrate as well as citric acid had been dissolved with a small amount of distilled water under vigorous stirring. The pH value of the solution had been adjusted at 7. The temperature was raised up to 250 °C until all fumes ended. The resultant powder was calcined for 4 h at 800 °C with a rate of 4 °C/min.

2.2.2 (PVDF/CoFe$_2$O$_4$, CuFe$_2$O$_4$ and Cu–CoFe$_2$O$_4$) film preparation

Firstly, four samples of PVDF solution were prepared separately by dissolving 3 gm of PVDF powder in 10 mL of (NMP) at room temperature under continuous stirring for 12 h. until the resulting solution (PVDF/NMP) became transparent. Afterwards, 3 mg (0.1 wt%) of the CoFe$_2$O$_4$, CuFe$_2$O$_4$ and Cu–CoFe$_2$O$_4$ nanoparticles were added to three samples of the (PVDF/NMP) solutions which were prepared above and kept under magnetic stirring for 2 h. The fourth sample was kept as prepared. Finally, each sample was poured on a clean glass surface on a hot-plate kept at 60 °C until the solidification of the solution film. The obtained films were washed using distilled water to remove any contaminating particles and for full solidification of the films. The film thickness was 0.19, 0.12 and 0.21 mm for PVDF/(CoFe$_2$O$_4$, CuFe$_2$O$_4$ and Cu–CoFe$_2$O$_4$), respectively.

2.3 Nanoparticle's characterization

The crystalline phases of CoFe$_2$O$_4$, CuFe$_2$O$_4$ and Cu–CoFe$_2$O$_4$ nanoparticles were identified by the Fourier transform infrared spectroscopy (FT-IR) instrument (Perkin Elmer) working in the range of 4000–400 cm$^{-1}$. The degree of nanoparticles crystallinity has been investigated, using X-ray diffraction (XRD) analysis (Bruker, D8 Advance, X-ray diffractometer), the instrument operating at 40 kV and 40 mA of current with Cu–Ka radiation ($\lambda = 1.541$ Å). Field emission scanning electron microscopy (FESEM) JEM-ARM300F operated at 200 kV has been used for the topographical investigation of the prepared nanoparticles.

2.4 Optical investigations

UV–vis transmission and reflection spectra have been estimated to investigate the optical properties of the prepared nanocomposite films using (JASCO Corp., V-570, Rev. 1.00) spectrometer.

2.5 Piezoelectric response

2.5.1 Digital storage oscilloscope

The generated voltage by repeating human finger press and release on the surface of the investigated nanocomposite films have been recorded using a digital storage oscilloscope [GW Instek Gos-806 s]. Open circuit output voltage has been used to record the responses of the investigated samples, at room temperature.

2.5.2 Piezoresponse force microscopy (PFM)

PFM measurements were performed using a commercial Flex-Axiom AFM, head type FlexAFM with 75–18–163 μm.To measure the piezoresponse of the samples, an A.C. voltage was applied to the tip, at a frequency of 165.07 kHz. Scan rate was 0.5 Hz, and the scan area was (250×250) nm$^2$. All measurements were performed in an insulating chamber at room temperature.
3 Results and discussion

3.1 Nanoparticles characterization

3.1.1 XRD

Figure 1 illustrates the XRD patterns of CoFe$_2$O$_4$, CuFe$_2$O$_4$ and Cu–CoFe$_2$O$_4$ powder. The XRD results of all the prepared samples match with the reported standard phase in the XRD reference ICDD card. Cobalt ferrite and copper ferrite have crystallized as single-phase with rhombohedral and cubic structures, respectively, according to ICDD card numbers [04-005-7078] and [01-082-8784] for CoFe$_2$O$_4$, and CuFe$_2$O$_4$, respectively, while [00-065-0376] card for Cu–CoFe$_2$O$_4$. Some peaks were indexed of Hematite according to ICDD card [01–080-5405] and copper oxide reference card [04-014-5856]. The average crystallite size of prepared samples was calculated using the X-ray line broadening method via the Scherrer equation [10]

$$D = \frac{k \lambda}{\beta \cos \theta}$$  \hspace{1cm} (1)

where $D$ is the crystallite size (nm), $K = 0.9$ is the particle shape factor, $\lambda$ (nm) is the wavelength, $\beta$ is the full width at half maximum, and $\theta$ is the position (angle) of the peak at the maximum. The estimated crystallite sizes are 19.1, 24.1 and 22.2 nm. for the CoFe$_2$O$_4$, CuFe$_2$O$_4$ and Cu–CoFe$_2$O$_4$ nanoparticles, respectively.

3.1.2 FTIR

Figure 2 shows the FTIR spectra of CoFe$_2$O$_4$, CuFe$_2$O$_4$ and Cu–CoFe$_2$O$_4$ nanoparticles which were recorded in the range 4000–400 cm$^{-1}$. From Fig., it can be noted that the band at 1108 cm$^{-1}$ is related to the residual Fe-OOH. At 537.9 cm$^{-1}$ and 471 cm$^{-1}$ bands appeared which corresponded to the stretching vibrations of metal oxide in the octahedral group and tetrahedral group [11]. 534.97 cm$^{-1}$ and 430.2 cm$^{-1}$ could be related to vibrations of M–O (M denoted to copper or iron) [12]. Generally, the transmittance bands in the region 400–600 cm$^{-1}$ indicate the existence of copper ferrites. Moreover, the band $v_1$ that is appeared around 526 cm$^{-1}$ is corresponding to the metal cations intrinsic stretching vibration in tetrahedral sites while the band $v_2$ which is observed at 466 cm$^{-1}$ is attributed to the oxygen stretching vibration bands in the B sites [13, 14].

3.1.3 FESEM

The FESEM micrographs shown in Fig. 3a–c of CoFe$_2$O$_4$, CuFe$_2$O$_4$ and Cu–CoFe$_2$O$_4$, respectively showed a homogenous distribution of polycrystalline nanometer particles. The present particles showed agglomeration where some particles form large clusters. However, as shown in Figs. 13, 14, the obtained PFM images reveal homogeneous distribution of nanofillers.

3.2 Nanocomposite characterization

Figure 4 illustrates the (XRD) patterns of neat PVDF polymer and the nanocomposite films under investigation compared to the reference card of PVDF. Figure 4a shows peaks corresponding to (100), (020), (110), and (021) these peaks

Fig. 1 XRD pattern of CoFe$_2$O$_4$, CuFe$_2$O$_4$ and Cu–CoFe$_2$O$_4$ nanoparticles

Fig. 2 FTIR spectrum of CoFe$_2$O$_4$, CuFe$_2$O$_4$ and Cu–CoFe$_2$O$_4$ nanoparticles

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characterize the $\alpha$-phase of PVDF polymer. However, in Fig. 4b corresponding to the (PVDF/CoFe$_2$O$_4$) nanocomposite sample the XRD peaks (110) and (021) are broader, indicating the presence of a semi-crystalline mixture of $\alpha$, $\beta$- and $\gamma$-phases. These results confirm that the (PVDF/CoFe$_2$O$_4$) nanocomposite sample has a higher percentage of the polar $\beta$-phase, $\beta$ phase possesses the highest dipole moments improving the PVDF piezoelectricity [15].

3.3 Optical properties

Figure 5 shows the variation of the reflectance of UV–Vis. Spectrum of CoFe$_2$O$_4$, CuFe$_2$O$_4$, and Cu–CoFe$_2$O$_4$ powder with the wavelength. From Fig., it can be observed that the reflectance values increase with increasing wavelength values. Also, the prepared PVDF/(CoFe$_2$O$_4$ and Cu–CoFe$_2$O$_4$) nanocomposites films have the highest reflectance values with an intensive peak at 854 nm. were observed in Fig. 6a.
The effect of CoFe$_2$O$_4$, CuFe$_2$O$_4$ and Cu/CoFe$_2$O$_4$ nanoparticles on the optical properties of PVDF nanocomposites was investigated. Figure 6b shows the change in transmittance of pure PVDF and PVDF/(CoFe$_2$O$_4$, CuFe$_2$O$_4$ and Cu–CoFe$_2$O$_4$) nanocomposites with the wavelength ($\lambda$). From Fig., it can be noted that the transmittance of PVDF decreased with the addition of nanoparticles, especially with the CoFe$_2$O$_4$ and Co-CuFe$_2$O$_4$ samples. The decrease in transmittance observed for nanocomposite samples is attributed to light scattering caused by the CoFe$_2$O$_4$, CuFe$_2$O$_4$ and Cu–CoFe$_2$O$_4$ nanoparticles.

Moreover, Figs. 6a and 7a have shown a significant increase in the reflectance and absorbance coefficient percentages of the prepared nanocomposite films compared with the pure PVDF. The absorption coefficient ($\alpha$) of the investigated samples was calculated using the following equation [15].

$$\alpha = \frac{2.303 \cdot A}{l} \quad (2)$$

where A: absorbance, l: thickness of the specimen.

The very high absorbance in the UV region for the investigated films makes them of interest in UV protection applications [16]. The curves in Fig. 7a demonstrate a clear peak in the UV region at $\lambda \approx$ 230 nm for both pure and doped PVDF which is corresponding to $\pi$–$\pi^*$ transitions.
transitions of aromatic C–C bonds [16]. At the same time, the PVDF/CoFe$_2$O$_4$ and PVDF/Co–CuFe$_2$O$_4$ samples showed a high reflectance peak at the near-infrared region of $\lambda \approx 854$ nm.

Figure 6b indicates a remarkable increase in the extinction coefficient (K) of the PVDF/(Cu–CoFe$_2$O$_4$) nanocomposite. The extinction coefficient is the measure of the portion of the light that is lost, absorbed, or scattered per unit length of penetration medium. The extinction coefficient (K) is given by the following equation [15]:

$$K = \alpha/\lambda$$

$$\alpha$$ is absorption coefficient percentage, and $\lambda$ is the wavelength.

The refractive index is another mainly important elemental property of material because of its direct relationship with the electronic polarizability of ions and the local field in the optical material. The refractive index (n) of composite materials was calculated using equation [17, 18],

$$n = \frac{(1 + R)}{(1 - R)} + \sqrt{\frac{4R}{(R - 1)^2} - K^2}$$

Figure 8 shows the variation of the refractive index n versus $\lambda$ for all investigated samples. A significant increase in the refractive index n was observed in the samples by the addition of the nanoparticles to the PVDF polymer. This increase can be attributed to the intermolecular interactions that occurred between the filler and the adjacent PVDF chain segments which lead to enhance the density of the films resulting in higher refractive indices.

The optical conductivity $\sigma_{opt}$ of the prepared nanocomposite films was estimated from the following equation, [15]:

$$\sigma_{opt} = \frac{\alpha n c}{4\pi}$$

$\alpha$ is the absorption coefficient, n is the refractive index of the samples, c is the speed of light.

Figure 9 shows the variation of optical conductivity $\sigma_{opt}$ of the investigated samples with the incident photon energy $h\nu$. It is observed that the PVDF optical conductivity increased by more than five times with the addition of nanoparticles. This increase is attributed to the increase of
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concentration of the excited electrons as the nanoparticles introduced into the PVDF polymer [19]. The largest optical conductivity value $\sigma_{\text{opt.}}$ was observed for the samples with CoFe$_2$O$_4$ content.

The complex dielectric constant reveals the basic intrinsic property of materials. The real part of the dielectric constant indicates the amount of light that slows down inside the material, although the imaginary part of the dielectric constant represents the energy absorbed in dielectric material from electric field due to dipole motion where dipoles rearranged, and a transformation takes place. The real and imaginary parts of dielectric constant have been calculated using the following equations [18, 20, 21].

$$\varepsilon_r = n^2 - k^2$$

$$$\varepsilon_i = 2nk$$

where, $\varepsilon_r$ is real part of dielectric constant, $\varepsilon_i$ is imaginary part of dielectric constant.

Figure 10a, b shows the real ($\varepsilon_r$) and imaginary ($\varepsilon_i$) parts of the dielectric constant as a function of $\lambda$, of the investigated samples. Fig. shows that both ($\varepsilon_r$) and ($\varepsilon_i$) of the PVDF/(Cu–CoFe$_2$O$_4$, CoFe$_2$O$_4$, and CuFe$_2$O$_4$) have higher values compared with pure PVDF. This increase is caused by the interfacial polarizations at the conductor–insulator interface [22]. The filler nanoparticles significantly increased the value of the PVDF relative permittivity. In the case of CoFe$_2$O$_4$, an increase in the relative permittivity of up to 10 times the original value was observed. The higher polarization process was observed in the PVDF/(CoFe$_2$O$_4$) sample, which can be due to the existence of a huge number of trap states at the grain boundaries of the synthesized sample. These trap states are produced due to the presence of vacancies or defects [23].

According to Tauc’s relation [24], $a\nu = B(\hbar\nu - E_g)^{1/2}$, the optical band gap ($E_g$) of PVDF and PVDF/(Cu–CoFe$_2$O$_4$, CoFe$_2$O$_4$, and CuFe$_2$O$_4$) nanocomposite have been estimated. The direct bandgap of the prepared sample was obtained by plotting ($a\nu^2$) versus the incident photon energy and extrapolating the linear parts of obtained curves as shown in Fig. 11a–d. The optical band gap ($E_g$) values were 3, 1.85, 2.75 and 2.2 eV for the pure PVDF and PVDF/(CoFe$_2$O$_4$, CuFe$_2$O$_4$, and Cu–CoFe$_2$O$_4$) nanocomposite respectively. It can be noted that CoFe$_2$O$_4$, CuFe$_2$O$_4$, and Cu–CoFe$_2$O$_4$ nanofiller decreases the PVDF bandgap this decrease in optical band gap could be due to the creation of new levels in the bandgap which facilitates the flow of electrons from the valance band to the conduction band. The prepared nanocomposite narrow bandgap
structure causes the electrons to release after absorbing photons “photoelectric effect property” [25].

### 3.4 PVDF/(Cu–CoFe$_2$O$_4$, CoFe$_2$O$_4$, and CuFe$_2$O$_4$) energy harvest performance

The piezoelectric response of the prepared samples has been investigated using two different methods. The first one uses a digital storage oscilloscope (DOS) and the other uses piezoresponse force microscopy (PFM).

#### 3.4.1 Piezoelectric response using a digital storage oscilloscope (DOS)

The working principle of the piezoelectric energy harvester of PVDF depends on it being an insulating material that generates an internal piezoelectric field as it is subjected the material to mechanically applied force [26].

The prepared nanocomposite films have been tested using a digital storage oscilloscope (DOS), where each film was placed between two copper layers. With respect to the open circuit (with forward and reverse connection), the generated piezo potential was recorded because of a repetitive finger stress that has been applied on the upper surface of nanocomposite films. A compressive stress on the surface of films produced by the finger tapping, causes the displacement of positive and negative charges in the nanocomposite films.

It is well known that PVDF mainly has four-phase structures, α phase, β phase, and δ phase, depending on the chain conformations [27]. The alpha phase is the most energy-stable, but it has very weak piezoelectric characteristics compared with the high polarity beta phase. The β phase possesses the highest dipole moments, where PVDF has

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**Fig. 11** (a–d): $(\alpha h \nu)^2$ verse incident photon energy of (a): PVDF (b): PVDF/(CoFe$_2$O$_4$), (c): PVDF/(CuFe$_2$O$_4$) and (d): PVDF/(Co-CuFe$_2$O$_4$)
polymeric chain of repeated units of \([\text{CF}_2-\text{CF}_2\]_n\) monomers which consist of CH₂-positively charged dipoles and CF₂-negatively charged dipoles and as a result, best piezoelectric performance [28].

By mechanical deformation and polarization, the structure of alpha phase can be transformed into polar beta phase to achieve piezoelectric characteristics [29].

The nanoparticles improved the piezo-potential as shown in Fig. 12a–d where an interaction between the CoFe₂O₄, CuFe₂O₄ and Cu–CoFe₂O₄ nanoparticles and the dipoles of PVDF. The largest recorded output voltage has been observed in the PVDF/CoFe₂O₄ film sample, as shown in Fig. 12b.

This enhancement of piezo-potential in PVDF composite films is due to the role of CoFe₂O₄, CuFe₂O₄ and Cu–CoFe₂O₄ nanofillers in PVDF matrix, which provides a conducting particle that could help charges induced inside the film to move to the film surface. Also, the interaction between Co²⁺, Cu²⁺ and Fe³⁺ nanoparticles with CF₂-dipoles and O²⁻ nanoparticles of CoFe₂O₄, CuFe₂O₄ and Cu–CoFe₂O₄ with CH₂-dipoles could enhance the piezoresponse of PVDF polymer.

It can be noted that the PVDF composite films with higher values of complex dielectric constant (PVDF/CoFe₂O₄ and PVDF/Cu–CoFe₂O₄), have higher values of piezo potential Figs. 10, 12. As the polarization causes a slight increase in the dielectric constant, where the beta phase dipoles rearranged and a transformation of alpha to beta takes place when energy is applied [30].

### 3.4.2 PFM studies

Another tool to investigate the piezoelectric response of the particles, piezoresponse force microscopy (PFM) was
Fig. 13 (a–l): Z-axis direction, phase and amplitude PFM micrographs of PVDF/(Cu–CoFe$_2$O$_4$, CoFe$_2$O$_4$, and CuFe$_2$O$_4$) nanocomposite films
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used. An alternating voltage $V_{ac}$ is applied to the tip causing the production of an alternating electric field inside the investigated sample. The piezoelectric materials undergo a periodic deformation, along the tip. Such deformation leads to the deflection of the cantilever in any direction. [31]. Out of plane deformation (in the z-axis), in-plane deformation (a diameter change in the y-axis), and a second in-plane deformation (a length change in the x-axis). PFM generates a highly resolved lateral mapping of the piezoelectric properties of the investigated sample. The domain structures can be obtained using piezoresponse force microscopy (PFM) by combining the in-plane and out-of-plane PFM images [32].

The pure PVDF and PVDF/(Cu–CoFe$_2$O$_4$, CoFe$_2$O$_4$, and CuFe$_2$O$_4$) nanocomposites films with the area of 2.5 $\mu$m × 2.5 $\mu$m was scanned with alternating Vac of 510 mV applied to the cantilever tip. Figure 13a–l showed the PFM responding in the z-axis direction, phase, and amplitude micrographs of the investigated samples, respectively. The out-of-plane PFM phase images Fig. 13a, d, g and j for all samples, the observed negative (white) and positive (black) areas corresponding to the piezoelectric polarization indicating antiparallel ferroelectric nanodomains with 180° domain walls. The negative domains (white areas) related to polarization oriented downward perpendicular to the surface of the PVDF film, while the positive domains (black areas) related to polarization oriented upward [33, 34]. A direct correlation between the ferroelectric domains and the photoelectric signal where ferroelectric domains are related to the photocurrent [35, 36]. Electrons and holes created by optical absorption will be separated by the application of the electric fields, where electrons will move to the positive domains and holes to the negative domains [37]. These carriers are then localized at the surface. Well-defined piezoelectric domains shown in Fig. 13a indicating elongated crystallites which correspond to the homogeneous $\beta$-phase. Consequently, the domains in the PVDF nanocomposite films align perpendicular to the sample and along the z-axis, Fig. 14). The PFM amplitude (Fig. 13b and c) shows a piezoelectric contrast as a result of the defects caused by the application of the alternating field.

The images of in-plane PFM (Fig. 13d–j) were analyzed both for the phase and amplitude dependence of the PFM signal of PVDF/(Cu–CoFe$_2$O$_4$, CoFe$_2$O$_4$, and CuFe$_2$O$_4$) nanocomposites films. The images Figs. 13, 14) reveal the formation of periodic stripe domains acquired at $\theta\approx 180^\circ$. All the images of PVDF/(Cu–CoFe$_2$O$_4$, CoFe$_2$O$_4$, and CuFe$_2$O$_4$) nanocomposites films show stripe domains aligned along the last saturating field direction.

In Fig. 13d–f, the PVDF/(CoFe$_2$O$_4$) sample indicating that the nanodomains merged into micro-sized domains, the micro-sized domain is produced because of the merged nanodomains, supported by the cross-section phase variation.

### 4 Conclusion

Cobalt ferrite, Copper ferrite and cobalt doped copper ferrite nanoparticles have been synthesized and characterized. The prepared Cu–CoFe$_2$O$_4$, CoFe$_2$O$_4$, and CuFe$_2$O$_4$ nanoparticles have been introduced to the PVDF polymer.

PVDF optical and piezoelectric properties have been studied using three nanofillers, (Co, Cu and Co/Cu) ferrite. This study showed that the PVDF polymer optical properties have improved by introducing the prepared nanoparticles. The dielectric constant of the PVDF polymer multiplied 10 times in the case of PVDF/(Cu–CoFe$_2$O$_4$ and CoFe$_2$O$_4$) nanocomposite samples. However, the optical conductivity of pure PVDF increased five times by adding (Cu–CoFe$_2$O$_4$ and CoFe$_2$O$_4$) nanofillers. The PVDF band gap energy decreased by the addition of the investigated nanofillers.

As well as, the prepared nanoparticles enhanced the PVDF piezoelectricity, the PVDF/(Cu–CoFe$_2$O$_4$, CoFe$_2$O$_4$, and CuFe$_2$O$_4$) nanocomposites films show stripe domains aligned along the last saturating field direction when examined using PFM., and the domains in the PVDF film align along the z-axis direction perpendicular to the sample and the bottom electrode.

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