Low Temperature Physics/Fizika Nizkikh Temperatur, 2018, v. 44, No. 8, pp. 970–976

Dielectric properties of ferromagnetic Ni nanoparticles added $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_3O_{10-\delta}$ superconducting phase

M. Mumtaz and Mian A. Asghar

Materials Research Laboratory, Department of Physics, Faculty of Basic and Applied Sciences (FBAS)
International Islamic University (IIU), Islamabad 44000, Pakistan
E-mail: mmumtaz75@yahoo.com

Received November 30, 2017, revised February 15, 2018, published online June 27, 2018

Ferromagnetic nickel (Ni) nanoparticles were added in $(Cu_{0.5}Tl_{0.5})Ba_2Ca_2Cu_3O_{10-\delta}$ (CuTl-1223) superconducting matrix to get Ni_x/CuTl-1223; x = 0–1.00 wt% nanoparticles-superconductor composites. Temperature- and frequency-dependent dielectric properties of CuTl-1223 superconducting phase with different contents of Ni nanoparticles were studied. Different dielectric parameters such as dielectric constant (ε'_r, ε''_r), dielectric tangent loss (tan δ) and ac conductivity (σ_{ac}) were determined from experimentally measured capacitance and conductance at different frequencies from 10 kHz to 10 MHz and at different operating temperatures from 78 to 290 K. The values of ε'_r and ε''_r were found maximal at smaller frequencies and started to decrease at higher frequencies. The value of σ_{ac} is high at high frequency unlike to ε'_r and ε''_r, which is due to release of space charges at high frequencies. Peaks in tan δ graphs represent the resonance phenomenon at certain frequencies in these samples. Non-monotonic behavior in variation of dielectric parameters with temperature of Ni_x/CuTl-1223 samples was observed particularly at high temperatures which was due to thermal instability of the system at high temperatures.

PACS: 74.25.–q Properties of superconductors;
74.25. F– Transport properties;
74.72.–h Cuprate superconductors;
74.81.Bd Granular, melt-textured, amorphous, and composite superconductors.

Keywords: Ni_x/(CuTl-1223) composites, Ni nanoparticles, CuTl-1223 superconductor, dielectric properties.

1. Introduction

The dielectric properties of superconducting materials can play a vital role in growth of microelectronics to produce various robust devices such as memory devices and capacitor [1]. High values of dielectric constant of various high-temperature superconductors (HTSCs) made them highly suitable for practical applications in the field of electronics [2–5]. Different phases of $Cu_{0.5}Tl_{0.5}Ba_2Ca_{n-1}Cu_nO_{2n+4-\delta}$ HTSCs family can be synthesized at high pressure (~5 GPa) as well as at ambient pressure. The values of superconducting parameters of the materials synthesized at high-pressure are normally higher than those synthesized at ambient pressure [6]. The high pressure synthesis can play an important role for carriers' optimization and to enhance superconductivity [7,8]. The easy ambient pressure synthesis was the main motivation to select CuTl-1223 phase of CuTl-12(n–1)n HTSCs family for the study of dielectric properties of this phase [9,10]. In various electronic devices the conduction mechanisms strongly depends upon the frequency, temperature, surface charges, fabrication conditions, impurities and doping concentration. Therefore, it has become mandatory to study the dielectric properties of HTSCs over an extensive frequency and temperature ranges, which can yield useful information about their polarization and conduction mechanism. There are four primary mechanisms of polarizations: (i) electronic polarization (α_e), (ii) atomic and ionic polarization (α_a), (iii) dipolar and oriental polarization (α_o) and (iv) interfacial polarization (α_i) [11,12]. A short-range motion of charges in each mechanism of polarization leads to the total polarization. Interfacial polarization and oriental or dipolar polarization is generally active in HTSCs because at high frequency the dielectric constant becomes saturated for a temperature range from superconducting state to room temperature.

There are various techniques reported in literature to tune the dielectric properties of HTSCs but one of the easiest and very effective methods is the insertion of different nanostructures in appropriate amount at the grain boundaries of HTSCs matrices. Carrier density can be controlled in the material by varying the content of nanostructures at inter-granular sites. The nature of grain boundaries, oxygen contents, grains size and inter-grains connectivity of bulk HTSCs can also be influenced by the inclusion of nanostructures [13,14].

In present article, the effects of ferromagnetic Ni nanoparticles on the dielectric properties of CuTl-1223 superconducting phase have been explored at different temperatures from 78 to 290 K and different frequencies from 10 kHz to 10 MHz. These nanoparticles can modify the nature of grain boundaries, grain size, inter-grain weak-links and oxygen contents, which can affect the superconducting and dielectric parameters of CuTl-1223 phase.We tried to tune the dielectric properties of CuTl-1223 phase by varying the content of ferromagnetic Ni nanoparticles, temperature and frequency.

2. Samples synthesis and experimental details

$Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_3O_{10-\delta}$ (CuTl-1223) superconducting phase was synthesized by solid-state reaction and nickel nanoparticles were prepared by sol-gel methods. These ferromagnetic Ni nanoparticles were added in CuTl-1223 matrix to get Ni_x/CuTl-1223; x = 0, 0.25, 0.5, 0.75 and 1.00 wt% superconductor-nanoparticles composites. These samples were characterized by using different experimental techniques such as x-rays diffraction (XRD), resistivity vs temperature measurements, scanning electron microscopy (SEM) and energy dispersive x-ray spectroscopy (EDX) [15].

The instrument used to measure the inductance (L), capacitance (C) and resistance (R) is commonly written as LCR meter. The frequency dependent dielectric measurements were carried out by LCR meter (Hewlett-Packard 4294A) in frequency range from 10 kHz to 10 MHz at different operating temperatures from 78 to 290 K. A conventional two-probe technique was used for these dielectric measurements. Silver paint was applied on both surfaces of rectangular bar-shaped sample and copper leads contacts were made by silver paint on two silver electrode surfaces of the sample.

3. Results and discussions

Structural, compositional, morphological and superconducting transport properties of Ni_x/CuTl-1223 composites samples were carried out by XRD, EDX, SEM and four-point probe resistivity vs temperature measurements and reported in our previously published manuscript [15].

In this articles, we are reporting only the frequency- and temperature-dependent dielectric properties of Ni_x/CuTl-1223; x = 0, 0.25, 0.5, 0.75 and 1.00 wt% superconductor-nanoparticles composites in frequency range from 10 kHz to 10 MHz at different operating temperatures from 78 to 290 K. The change in the values of ε_r' and ε_r'', tan δ and σ_{ac} by varying the contents of Ni nanoparticles from x = 0 to 1.00 wt%, were determined, compared and explained. The energy stored in the sample when exposed to external applied ac field is represented by real part of the dielectric constant (i.e., $\varepsilon_r' = Cd/A\varepsilon_0$). Where C is capacitance (F), d is the thickness of the dielectric/separation between the plates of the capacitor (m), ε_0 is the permittivity of free space ($\varepsilon_0 = 8.85\cdot10^{-12}$ F/m) and A is the area of the electrode (m^2). Variation in ε_r' of Ni_x/CuTl-1223 composites with different test frequencies and operating temperatures is shown in Figs. 1(a)–(e). The value of ε_r' is higher at lower frequencies demonstrating the gradual decrease with the increase in frequency and becomes almost zero at higher frequency indicating that the time period of applied ac signal becomes smaller than the time required for polarization. Active grain boundaries are responsible for high values of real part of dielectric constant at lower frequencies [16–18]. As space charge carriers need finite time for alignment in the direction of applied ac electric field, so ε_r' showed decreasing trend with increasing frequency of external applied ac field. Maximum values of ε_r' at lower frequency of 10 kHz varied from $3.4\cdot10^5$ to $3.6\cdot10^3$, $1.07\cdot10^4$ to $2.28\cdot10^3$, $2.5\cdot10^5$ to $4.25\cdot10^4$, $7.76\cdot10^4$ to $1.3\cdot10^4$ and $6.3\cdot10^5$ to $1.8\cdot10^4$ at various operating temperature from 78 to 290 K for Ni_x/CuTl-1223 composites with x = 0, 0.25, 0.50, 0.75 and 1.00 wt%, respectively. Overall decreasing trend in ε_r' with addition of Ni nanoparticles was observed with some non-monotonic behavior at different operating temperatures. Particularly, at higher temperatures the system was become thermally unstable due to which a slight anomalous behavior in variation of ε_r' with T was observed. Comparison of variation in maximum values of ε_r' with different contents of Ni nanoparticles at different operating temperature is shown in Fig. 1(f). The suppression in the values of ε_r' was observed up to x = 0.75 wt% and an increase was observed in the sample with maximum Ni nanoparticles, i.e., x = 1.00 wt%. The high values of ε_r' for the sample with x = 1.0 wt% of Ni nanoparticles can be caused by maximum trapping of mobile charge carriers at grain boundaries due to magnetic interaction with these magnetic Ni nanoparticles present there.

The loss or attenuation of energy across the interfaces of the samples in an external applied electric ac field can be estimated by the imaginary part of the dielectric constant ($\varepsilon_r'' = Gd/(\omega A\varepsilon_0)$). Where G is conductance and ω (= $2\pi f$) is the angular frequency. Conductance is an expression of the ease with which electric current flows through any material and is the reciprocal of resistance R. The standard unit of conductance is Siemens (S). The variation of ε_r'' with test frequency from 10 kHz to 10 MHz at different operating temperatures from 78 to 290 K of Ni_x/CuTl-1223 composites samples are shown in Fig. 2(a)–(e). A decreasing trend in ε_r'' has been ob-

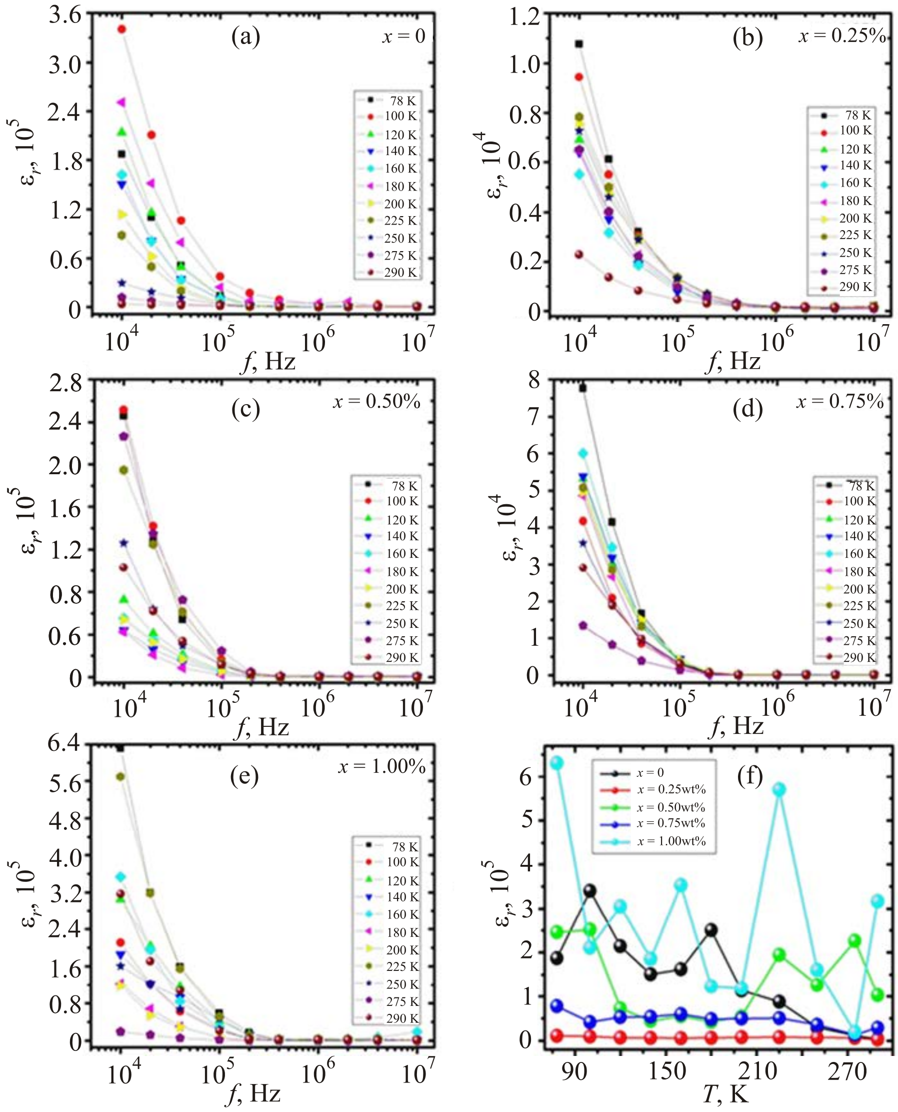

Fig. 1. Variation in ε_r' as a function of frequency at different operating temperatures of Ni_x/CuTl-1223 composites with (a) $x = 0$; (b) $x = 0.25$ wt%; (c) $x = 0.50$ wt%; (d) $x = 0.75$ wt%; (e) $x = 1.00$ wt% and (f) variation of maximum ε_r' at 10 kHz with operating temperature.

served with increasing frequency, which is in accordance with Wagner and Koop's theory [19,20]. According to these models, dielectric medium is a combination of well conducting grains and poorly conducting grain boundaries. The grain boundaries are responsible for high value of ε_r'' at lower frequencies. The maximum value of ε_r'' at lower frequency of 10 kHz varied from $4.79\cdot10^6$ to $1.1\cdot10^6$, $9.9\cdot10^5$ to $6.3\cdot10^5$, $5.6\cdot10^6$ to $4.99\cdot10^5$, $5.78\cdot10^6$ to $2.9\cdot10^6$ and $1.05\cdot10^7$ to $5.5\cdot10^6$ at different operating temperatures from 78 to 290 K for Ni_x/CuTl-1223 composites with $x = 0$, 0.25, 0.50, 0.75 and 1.00 wt%, respectively. At lower frequencies, the value of ε_r'' was found to be highest, which was gradually decreased with the increase of frequency and became linear with very lower values at higher frequencies. This was because at lower frequency the carriers can follow the frequency of external applied ac field and at high frequency the time period become very short and most of the carriers can't follow the external electric frequency and the response of the material becomes very small. Variation in maximum value of ε_r'' at lower frequency of 10 kHz versus operating temperatures is shown in Fig. 2(f). Maximum values of ε_r'' initially decreases with the addition of Ni nanoparticles at all operating temperatures and then starts to increase with higher content of these nanoparticles contents. Increase in maximum value of ε_r'' at $x = 0.75$ and 1.00 wt% showed enhanced energy loss due to high potential barrier offered by ferromagnetic Ni nanoparticles present at grain boundaries. The mobile charge carriers can be trapped across grain boundaries due magnetic interaction of Ni nanoparticles and large energy loss can take place at higher contents of Ni nanoparticles in CuTl-1223 matrix.

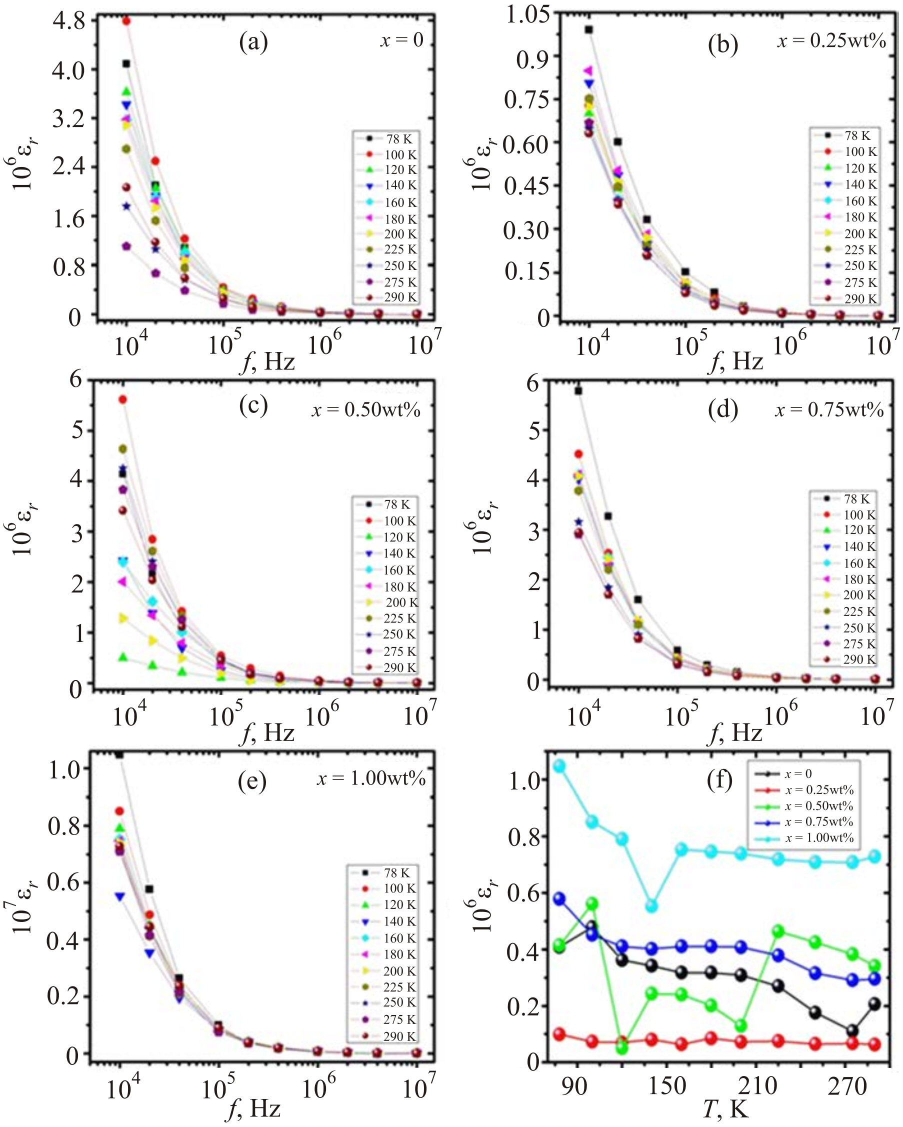

Fig. 2. Variation in ε_r'' as a function of frequency at different operating temperatures of Ni_x/CuTl-1223 composites with (a) $x = 0$; (b) $x = 0.25$ wt%; (c) $x = 0.50$ wt%; (d) $x = 0.75$ wt%; (e) $x = 1.00$ wt% and (f) variation of maximum ε_r'' 10 kHz with operating temperature.

Dielectric tangent loss $(\tan\delta = \varepsilon_r''/\varepsilon_r')$ is the ratio of energy dissipated to energy stored in material when exposed to ac field. Variation of $\tan\delta$ with frequency from 10 kHz to 10 MHz at different operating temperatures from 78 to 290 K of Ni_x/CuTl-1223 composites is shown in Figs. 3(a)–(e). Value of $\tan\delta$ is very small and almost constant at low frequency and a peak is observed for all these samples in frequency range from 10^5 to 10^7 Hz, which was due to resonance phenomenon resulting from the superposition of hooping frequency of charge carriers with frequency of external applied electric ac field. Variation in $\tan\delta$ at 10 kHz with operating temperature of these samples is shown in Fig. 3(f). Maximum values of $\tan\delta$ were observed at higher operating temperatures for all these samples. Overall increase in $\tan\delta$ showed the enhanced potential barriers at grain boundaries with the addition of ferromagnetic Ni nanoparticles in CuTl-1223 superconducting host matrix. At high temperature the system may become unstable due to which a slight anomalous behavior has been witnessed.

Variation in ac conductivity (σ_{ac}) with frequency from 10 kHz to 10 MHz at different operating temperatures from 78 to 290 K of Ni_x/CuTl-1223 composites is shown in Figs. 4(a)–(e). Prominent increase in σ_{ac} was observed at higher frequencies, which was attributed to strong conduction mechanism by hooping of charge carriers. Maximum value of σ_{ac} (s/m) varied from $4.75\cdot10^{-3}$ to $6.17\cdot10^{-7}$, $1.7\cdot10^{-4}$ to $1.27\cdot10^{-5}$, $2.08\cdot10^{-3}$ to $4.2\cdot10^{-6}$, $5.8\cdot10^{-4}$ to $3.7\cdot10^{-6}$ and $1.3\cdot10^{-2}$ to $9.3\cdot10^{-6}$ at maximum frequency of 10 MHz with different operating temperatures from 78 to 290 K for Ni_x/CuTl-1223 composites with $x = 0$, 0.25, 0.50, 0.75 and 1.00 wt%, respectively. Variation of maxi-

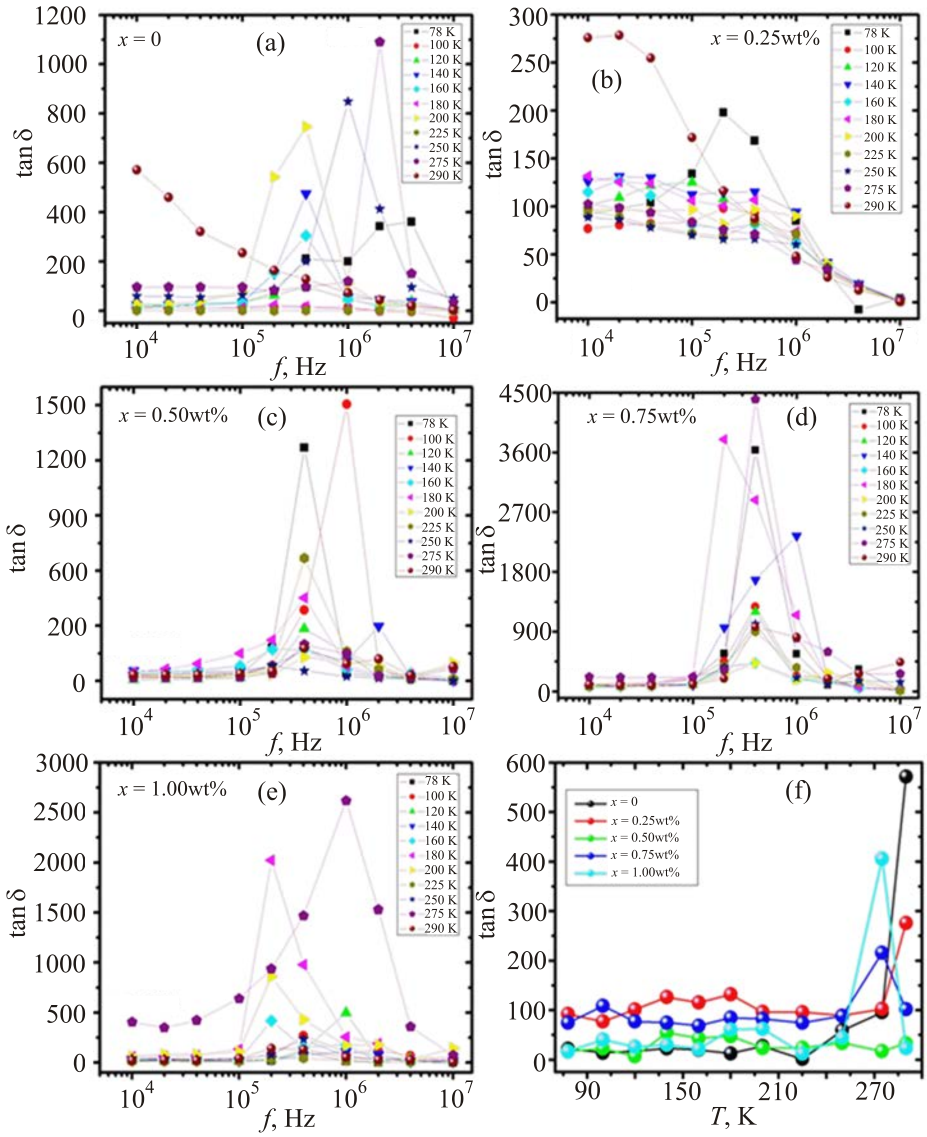

Fig. 3. Variation in $\tan\delta$ as a function of frequency at different operating temperatures of Ni_x/CuTl-1223 composites with (a) $x = 0$; (b) $x = 0.25$ wt%; (c) $x = 0.50$ wt%; (d) $x = 0.75$ wt%; (e) $x = 1.00$ wt% and (f) variation of $\tan\delta$ at 10 kHz with operating temperature.

mum σ_{ac} as a function of operating temperatures is shown in Fig. 4(f). Increase in maximum value of σ_{ac} was observed at higher operating temperatures, which shows enhanced conduction mechanism due to thermal excitation. Overall suppression in σ_{ac} was observed at higher contents of ferromagnetic Ni nanoparticles in CuTl-1223 superconducting matrix for all operating temperatures, which showed the enhanced resistance in the transportation of carriers due to increased barriers across the grain boundaries with these ferromagnetic Ni nanoparticles. Maximum value σ_{ac} has initially been increased for $x = 0.25$ and 0.75 wt% and then started to decrease at higher contents due to enhanced trapping mechanism promoted by the strong interaction of magnetic Ni nanoparticles settled at grain boundaries of the host CuTl-1223 matrix. The slight anomalous trend in variation of σ_{ac} at higher temperature can be attributed to thermal instability of the system.

4. Conclusion

Ni_x/CuTl-1223 nanoparticles-superconductor composites were synthesized and their dielectric properties were explored at different operating temperatures and frequencies. The values of ε_r' and ε_r'' were decreased with addition of Ni nanoparticles at low concentration and enhancement was observed at high concentrations. The values of $\tan\delta$ were increased with inclusion of Ni nanoparticles in CuTl-1223 superconductor. Occurring of resonance phenomenon was evidenced from the peaks in $\tan\delta$ graphs at certain frequencies in all Ni_x/CuTl-1223 samples resulting from the superposition of hooping frequency of charge carriers with frequency of external applied electric ac field. Higher content of Ni nanoparticles at grain boundaries created higher barriers to the mobility of charge carriers, which reduced the ac conduction and enhanced

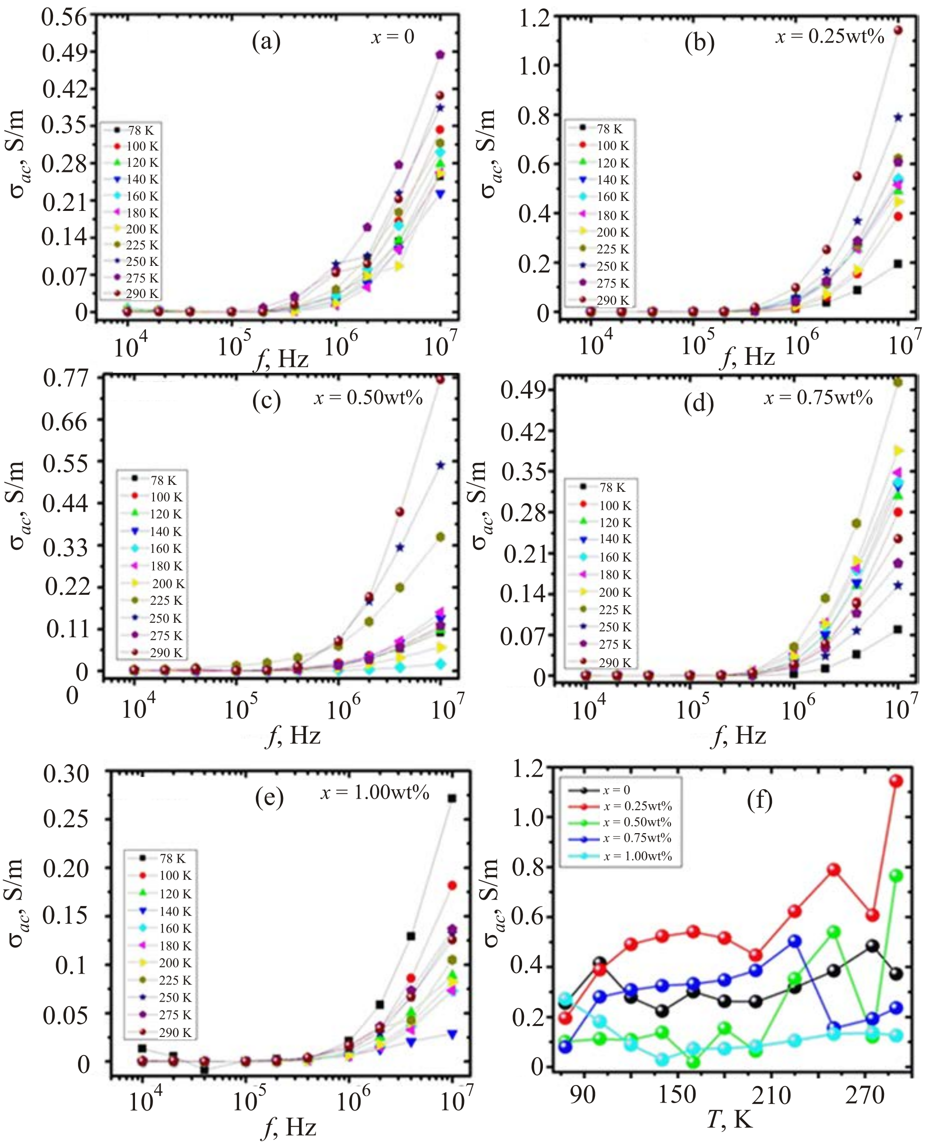

Fig. 4. Variation in σ_{ac} as a function of frequency at different operating temperatures of Ni_x/CuTl-1223 composites with (a) $x = 0$; (b) $x = 0.25$ wt%; (c) $x = 0.50$ wt%; (d) $x = 0.75$ wt%; (e) $x = 1.00$ wt% and (f) variation of maximum σ_{ac} at 10 MHz with operating temperature.

loss factors in the materials. The variation in dielectric properties can be attributed to the enhanced accumulation of charge carriers across the grain boundaries of CuTl-1223 superconducting phase due to magnetic interaction of ferromagnetic Ni nanoparticles present there. The most probable reason for suppression of σ_{ac} at high content of Ni nanoparticles is the reduction of optimum carriers' density desired for high conductivity after addition of these magnetic Ni nanoparticles settled at grain boundaries.

So, we can tune the dielectric properties of CuTl-1223 superconductor by homogeneous distribution of nanostructures at grain bombardiers, varying test frequency and operating temperature according to the requirement of applications.

1. X. Xu, Z. Jiao, M. Fu, L. Feng, K. Xu, R. Zuo, and X. Chen, *Physica C* **417**, 166 (2005).
2. S. Cavdar, H. Koralay, N. Tugluoglu, and A. Gunen, *Supercond. Sci. Technol.* **18**, 1204 (2005).
3. R.K. Nkum, M.O. Gyekye, and F. Boakye, *Solid State Commun.* **122**, 569 (2002).
4. J. Konopka, R. Jose, and M. Wołcyrz, *Physica C* **435**, 53 (2006).
5. N. Mohammed, *J. Supercond. Novel Magn.* **25**, 45 (2012).
6. H. Ihara, K. Tanaka, Y. Tanaka, A. Iyo, N. Terada, M. Tokumoto, M. Ariyama, I. Hase, A. Sundaresan, and N. Hamada, *Physica C* **341**, 487 (2000).
7. C. Jin, *High Pressure Res.* **24**, 399 (2004).
8. G. Samara, *J. Appl. Phys.* **68**, 4214 (1990).

9. H. Yamamoto, K. Tanaka, K. Tokiwa, H. Hirabayashi, M. Tokumoto, N.A. Khan, and H. Ihara, *Physica C* **302**, 137 (1998).
10. H. Ihara, *Physica C* **364**, 289 (2001).
11. S. Çavdar, H. Koralay, and S. Altındal, *J. Low Temp. Phys.* **164**, 102 (2011).
12. X. Xu, Z. Jiao, M. Fu, L. Feng, K. Xu, R. Zuo, and X. Chen, *Physica C* **417**, 166 (2005).
13. Nawazish A. Khan, M. Mumtaz, and A.A. Khurram, *J. Appl. Phys.* **104**, 033916 (2008).
14. P.B. Ishai, E. Sader, Y. Feldman, I. Felner, and M. Weger, *J. Supercond.* **18**, 455 (2005).
15. Irfan Qasim, M. Waqee-ur-Rehman, M. Mumtaz, Ghulam Hussain, K. Nadeem, and Khurram Shehzad, *J. Magn. Magn. Mater.* **403**, 60 (2016).
16. P. Lunkenheimer, V. Bobnar, A.V. Pronin, A.I. Ritus, A.A. Volkov, and A. Loidl, *Phys. Rev. B* **66**, 052105 (2002).
17. P. Lunkenheimer, R. Fichtl, S.G. Ebbinghaus, and A. Loidl, *Phys. Rev. B* **70**, 172102 (2004).
18. S. Krohns, P. Lunkenheimer, S.G. Ebbinghaus, and A. Loidl, *Appl. Phys. Lett.* **91**, 022910 (2006).
19. K.W. Wagner, *Ann. Phys.* **345**, 817 (1913).
20. C.G. Koops, *Phys. Rev.* **83**, 121 (1951).

Вплив додання феромагнітних наночастинок Ni на діелектричні властивості надпровідної фази $(Cu_{0,5}Tl_{0,5})Ba_2Ca_2Cu_3O_{10-\delta}$

M. Mumtaz and Mian A. Asghar

Для отримання композитів наночастинка–надпровідник Ni_x/CuTl-1223 (x = 0–1,00 мас.%) в надпровідну матрицю $(Cu_{0,5}Tl_{0,5})Ba_2Ca_2Cu_3O_{10-\delta}$ (CuTl-1223) було додано феромагнітні наночастинки Ni. Вивчено температурно- та частотно-залежні діелектричні властивості надпровідної фази CuTl-1223 з різним вмістом наночастинок Ni. Різні діелектричні параметри, такі як діелектрична стала (ε'_r, ε''_r), діелектричні тангенціальні втрати (tg δ) та провідність змінного струму (σ_{ac}), визначалися по експериментально виміряних ємності та провідності на різних частотах від 10 кГц до 10 МГц при різних робочих температурах від 78 до 290 К. Значення ε'_r та ε''_r були максимальні при менших частотах та зменшувалися при більш високих частотах. Значення σ_{ac}, на відміну від ε'_r та ε''_r, було високе на високій частоті, що пов’язано з виходом об’ємних зарядів на високих частотах. Піки на tg δ графіках представляють собою резонанс на деяких частотах в цих зразках. Немонотонна поведінка температурних залежностей діелектричних параметрів зразків Ni_x/CuTl-1223 спостерігалася, зокрема, при високих температурах, що викликано тепловою нестійкістю системи.

Ключові слова: Ni_x/(CuTl-1223), наночастинки Ni, надпровідник CuTl-1223, діелектричні властивості.