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Low-Temperature Dependence of the EPR Spectra of $\text{Gd}_{0.5}\text{RE}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ Compounds in the Tetragonal Phase

By

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The EPR spectra of perovskite ceramics of $\text{Gd}_{0.5}\text{RE}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ type (RE = La, Pr, Nd, Sm, Eu, Dy, Ho, Y, Er, Tm, Yb, and Lu) in tetragonal (non-HTC) phase are investigated as a function of temperature in the range 40 to 3 K. A signal arising from the isotropic exchange interaction of coupled pairs of Cu^{2+} ions is detected. The intensity of this signal is strongly dependent on the RE ions and its g -factor varies with temperature. Magnetic measurements show the existence of ferromagnetic anomalies at these low temperature. In the orthorhombic phase, these materials exhibit a superconducting phase transition in the range of 85 to 95 K, apart from the samples with La ions where a $T_C \approx 69$ K is found and the samples with Pr ions where a superconducting state can not be observed.

Die EPR-Spektren von $\text{Gd}_{0.5}\text{RE}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ -Perovskit-Keramiken (RE = La, Pr, Nd, Sm, Eu, Dy, Ho, Y, Er, Tm, Yb, and Lu) in der tetragonalen (nicht-HTC)-Phase werden in Abhängigkeit von der Temperatur im Bereich von 40 bis 3 K untersucht. Ein Signal von der isotropen Austauschwechselwirkung von gekoppelten Paaren von Cu^{2+} -Ionen wird nachgewiesen. Die Intensität dieses Signals ist stark abhängig von den RE-Ionen und sein g -Faktor ändert sich mit der Temperatur. Magnetische Messungen zeigen die Existenz von ferromagnetischen Anomalien bei diesen niedrigen Temperaturen. In der rhombischen Phase zeigen diese Materialien einen Supraleitungsphasenübergang im Bereich von 85 bis 95 K, bis auf die Proben mit La-Ionen, wo ein $T_C \approx 69$ K gefunden wird und Proben mit Pr-Ionen, wo ein supraleitender Zustand nicht beobachtet werden kann.

1. Introduction

In a previous work [1] we have reported the effect on the EPR spectrum of the partial substitution of gadolinium ions by other rare-earth ions in the $\text{Gd}_{0.5}\text{RE}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ compounds (RE = Y, Sm, Eu, and La) in both the non-HTC (tetragonal) and the HTC (orthorhombic) phases. The investigation [1] was carried out in the temperature range from 55 K up to room temperature. For all samples, in both phases, a broad EPR line from Gd^{3+} ions, centered at $g = 2.098(5)$ and with linewidth in the range $\Delta H = (0.14 \text{ to } 0.23)$ T has been observed. The lineshape and linewidth of this signal were almost temperature independent except for the HTC samples where a rapid narrowing below T_C appeared. However, the spectrometer seemed to work under impressively spoiled conditions [2]. For

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the non-HTC samples the EPR signal intensity increased as temperature decreased, a behavior which was attributed to the skin effect [1, 3]. According to this effect the EPR signal intensity decreases when the conductivity of the sample increases. This mechanism explains adequately the observed intensity variation if we take into account the semiconducting behavior of our non-HTC samples [4]. Furthermore, it has been suggested that the conductivity for these materials in the non-HTC phase is dominated by thermally activated hopping of small polarons [1, 3].

The work reported here extends the EPR study on the whole series of the polycrystalline oxides $Gd_{0.5}RE_{0.5}Ba_2Cu_3O_{7-\delta}$ ($RE = La, Pr, Nd, Sm, Eu, Dy, Ho, Y, Er, Tm, Yb,$ and Lu) to the low-temperature region below 40 K. In addition, detailed XRD and magnetic measurements were carried out in order to study the structural and magnetic properties of the materials.

2. Experimental Results

The samples were prepared by the standard solid state reaction technique, as described elsewhere [1].

2.1 XRD measurements

Identifications were made through a Phillips X-ray powder diffractometer using CoK_{α} radiation. The major phase for oxygenated samples could be indexed on the basis of an orthorhombic unit cell and of a tetragonal unit cell for the samples with large oxygen deficiency. Table 1 presents the crystallographic constants of the unit cell of the $Gd_{0.5}RE_{0.5} \cdot Ba_2Cu_3O_{7-\delta}$ compounds for both the orthorhombic and tetragonal phases. The values of the various ion radii have been taken from [5]. In this series, the ionic radius of the ion in the rare-earth site is taken as the averaged $Gd^{3+}-RE^{3+}$ ion radius. The XRD patterns

Table 1

Crystallographic constants of the unit cells of members of the series $Gd_{0.5}RE_{0.5}Ba_2Cu_3O_{7-\delta}$ for both orthorhombic and tetragonal phases (IR and IV represent the averaged ionic radius and the averaged ionic volume, respectively)

pair	IR	IV	orthorhombic					tetragonal		
			T_C	a	b	c	V_{cell}	$a = b$	c	V_{cell}
	10^{-1} nm	10^{-3} nm^3	K	10^{-1} nm	10^{-1} nm	10^{-1} nm	10^{-3} nm^3	10^{-1} nm	10^{-1} nm	10^{-3} nm^3
GdLa	0.977	3.925	69	3.8526	3.9014	11.6580	175.2260	3.8934	11.7382	177.9342
GdPr	0.976	3.906	—	3.8359	3.9018	11.7122	175.2955	3.8863	11.8302	178.6754
GdNd	0.967	3.792	92	3.8480	3.9036	11.7087	175.8770	3.8877	11.8140	178.5600
GdSm	0.951	3.605	94	3.8391	3.9012	11.6940	175.1822	3.8801	11.8156	177.8859
GdEu	0.944	3.524	95	3.8400	3.8953	11.6990	174.9931	3.8757	11.8186	177.2779
GdDy	0.923	3.296	86	3.8271	3.8915	11.6847	174.0221	3.8673	11.7984	176.4570
GdHo	0.916	3.225	89	3.8336	3.8903	11.6757	174.1297	3.8703	11.7964	176.7009
GdY	0.916	3.220	89	3.8330	3.8878	11.6744	173.9712	3.8699	11.8056	176.8021
GdEr	0.910	3.161	85	3.8253	3.8878	11.6858	173.7912	3.8662	11.8236	176.7333
GdTm	0.904	3.108	91	3.8229	3.8833	11.6856	173.4782	3.8619	11.8213	176.3061
GdYb	0.898	3.051	92	3.8176	3.8828	11.6704	172.9901	3.8549	11.7954	175.2826
GdLu	0.894	3.015	91	3.8173	3.8814	11.6816	173.0801	3.8613	11.8184	176.2081

were typical for the RE123 perovskites and usually no impurities were detected. The substitution of gadolinium atoms for half the atoms of the RE in the Gd123 perovskite does not affect the general trends of the crystallographic constants, a fact also observed for the series with samarium atoms [6]. Thus the Gd-RE system could be considered as a single continuous lattice. The values of the crystallographic constants of the unit cell are systematically lower than those of the members of the $\text{Sm}_{0.5}\text{RE}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ compounds for both orthorhombic and tetragonal phases, because the averaged ionic radius of the Gd-RE system is smaller than that of the Sm-RE system (Table 1) [6].

2.2 Magnetic measurements

A PAR 155 vibrating sample magnetometer was used to study the magnetic properties of the samples in the range 4.2 to 100 K. The applied field was 0.02 T. The magnetic data for the HTC samples revealed a T_C onset between 85 to 95 K (Table 1) for all members of the series except for the $\text{Gd}_{0.5}\text{La}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ sample, which has a significantly smaller T_C value (Table 1) and $\text{Gd}_{0.5}\text{Pr}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ which is not superconducting for temperatures above 4.2 K, a behaviour also exhibited by the $\text{PrBa}_2\text{Cu}_3\text{O}_{7-\delta}$ compound [7]. The obtained T_C values usually are smaller than the corresponding ones for the samarium series, except

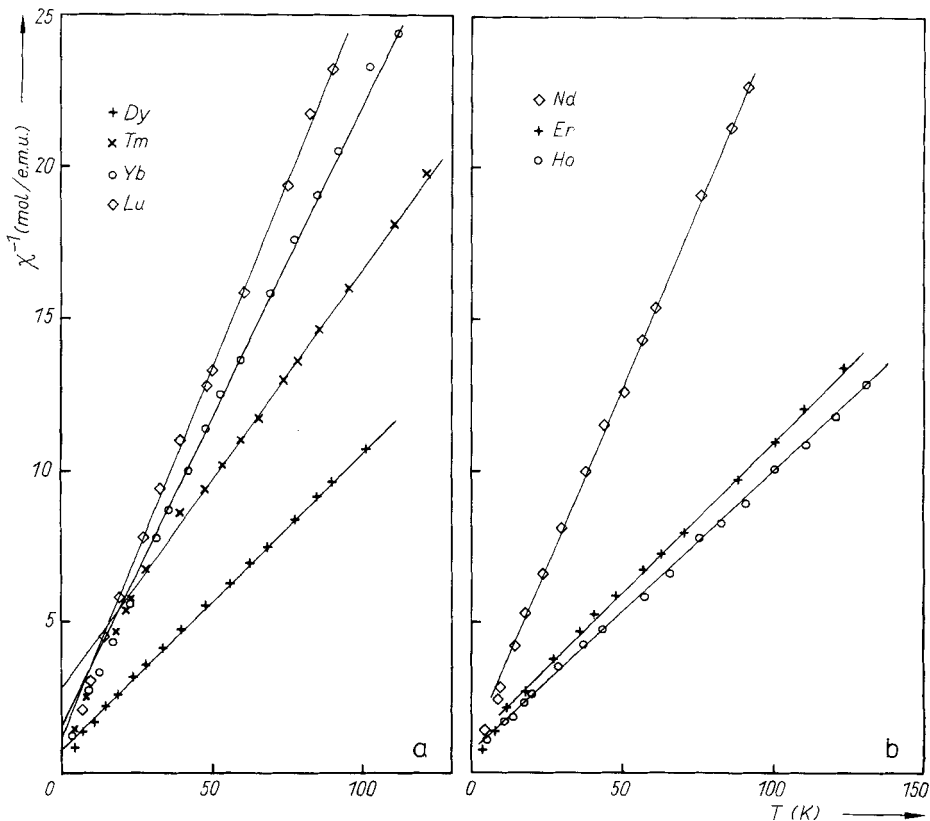


Fig. 1. Temperature dependence of the reciprocal susceptibility of the non-HTC samples of $\text{Gd}_{0.5}\text{RE}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$

Table 2

The effective magnetic moments (μ_{eff}) per formula unit (in units of Bohr magnetons, μ_{B}) for some $\text{Gd}_{0.5}\text{RE}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ compounds in tetragonal phase, theoretical and experimental (the values of μ_{eff} for the trivalent ions were taken from [8]). T_{p} is the temperature at which $\chi^{-1} = 0$

ions	$\mu_{\text{eff}}(\mu_{\text{B}})$			T_{p} (K)
	RE	(Gd-RE) _{calc}	(Gd-RE) _{exp}	
Gd ³⁺	7.94	7.94	7.76	- 5.0
Nd ³⁺	3.62	5.78	5.68	- 3.0
Dy ³⁺	10.63	9.29	8.95	- 7.5
Ho ³⁺	10.60	9.27	9.20	- 6.0
Er ³⁺	9.59	8.77	8.79	- 8.0
Tm ³⁺	7.57	7.76	7.58	-15.0
Yb ³⁺	4.54	6.24	6.15	- 5.0
Lu ³⁺	-	3.97	5.83	- 5.0

for the samples with Eu, Tm, Yb, and Lu ions for which comparable values were found [6]. For the HTC sample with europium atoms the phase transition temperature T_{C} was found to have the largest value, i.e. ≈ 95 K, the same as that of the Sm-Eu system [6].

Fig. 1 presents the plot of the reciprocal static susceptibility (χ^{-1}) as a function of temperature (applied magnetic field 0.1 T) for the non-HTC samples $\text{Gd}_{0.5}\text{RE}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ (RE = Nd, Dy, Ho, Er, Tm, Yb, and Lu). All the compounds exhibited Curie-Weiss behavior above 5 to 10 K with the exception of Gd-Lu. The lines extrapolated from the data in this region cross the abscissa at a negative temperature (Table 2) indicating an antiferromagnetic ordering in the systems. The experimental effective moments μ_{eff} , derived from the linear part of the plot χ^{-1} versus temperature, are close to the calculated values, assuming trivalent rare-earth ions (Table 2). These are in a good agreement with the theoretical values of the average magnetic moment of the trivalent gadolinium and the respective rare-earth ion. Assuming that lutetium ions are trivalent the difference between the experimental and the calculated effective moments for Gd-Lu cannot be understood. Below 5 to 10 K the extrapolation of the χ^{-1} lines shows a positive divergency of the magnetic susceptibility (Fig. 1) similar to systems possessing ferromagnetic ordering [9, 10]. In a previous work [11], in order to identify the origin of this anomaly, we introduced BaCuO_2 into the $\text{TmBa}_2\text{Cu}_3\text{O}_{7-\delta}$ compound and searched for possible additional phases. An addition of up to 5% BaCuO_2 in this compound showed a negligible effect on the magnetic susceptibility.

2.3 EPR measurements

EPR measurements were carried out using a standard X-band ($\nu = 9.3$ to 9.4 GHz) spectrometer of Varian E-112 and a Bruker ER-200 type spectrometer, both working with a 100 kHz magnetic modulation. An Oxford flow cryostat system was used operating between 4.2 and 100 K. The Varian E-112 spectrometer was working with a reflection double cavity and a static magnetic field varying between 0.025 to 1.0 T. The powdered samples weighed about 30 mg each and were placed into quartz tubes with diameter of 2 mm. Fig. 2 shows the EPR spectra at RT of the ceramics $\text{Gd}_{0.5}\text{Tm}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ in the tetragonal

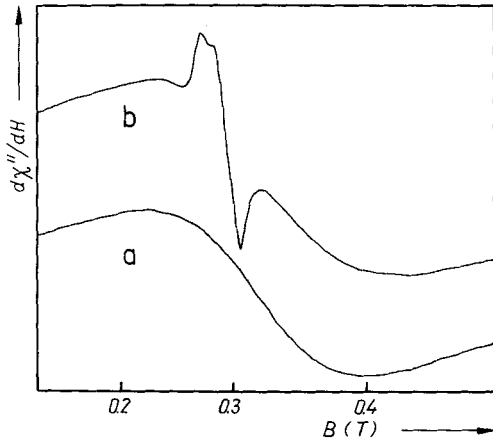


Fig. 2. The EPR spectra of $Gd_{0.5}Tm_{0.5}Ba_2Cu_3O_{7-\delta}$ compound at RT in (a) tetragonal and (b) orthorhombic phases ($\nu = 9.3$ GHz)

and orthorhombic phases. For all members of the series $Gd_{0.5}RE_{0.5}Ba_2Cu_3O_{7-\delta}$ a similar broad EPR line was observed. The EPR spectrum of all the HTC samples consists of the superposition of two signals, arising from the trivalent gadolinium ions and the divalent copper ions in orthorhombic local symmetry, the latter being similar to the EPR spectrum of the RE123 compounds [7]. The simulated powder spectrum of the $Gd_{0.5}Eu_{0.5}Ba_2Cu_3O_{7-\delta}$ compound in the orthorhombic phase shown in Fig. 3a is in good agreement with the experimental data, Fig. 3b. Fig. 4 shows the Cu^{2+} signal in orthorhombic local symmetry obtained after decomposition of the initial spectrum. This spectrum is similar for all RE123 compounds in the orthorhombic phase [7]. In Table 3, values of the linewidth ΔH for HTC and non-HTC samples are shown for all the members of the series, except for La, Sm, Eu, and Y compounds which have been reported previously [1]. Usually the linewidths for the non-HTC samples are broader than those for the HTC samples, with the exception of the samples with La, Ho, and Er. For the oxygenated and non-oxygenated $Gd_{0.5}Pr_{0.5}Ba_2Cu_3O_{7-\delta}$ compounds the Gd line has not been observed. The lineshape and linewidth of the Gd signal were independent of temperature [1, 3]. We should notice again that below

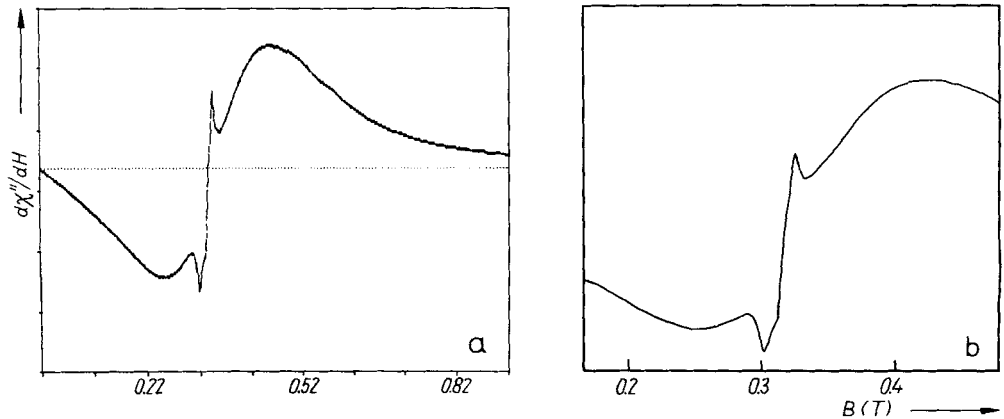


Fig. 3. The EPR spectra of $Gd_{0.5}Eu_{0.5}Ba_2Cu_3O_{7-\delta}$ compound in the orthorhombic phase a) simulated and b) experimental

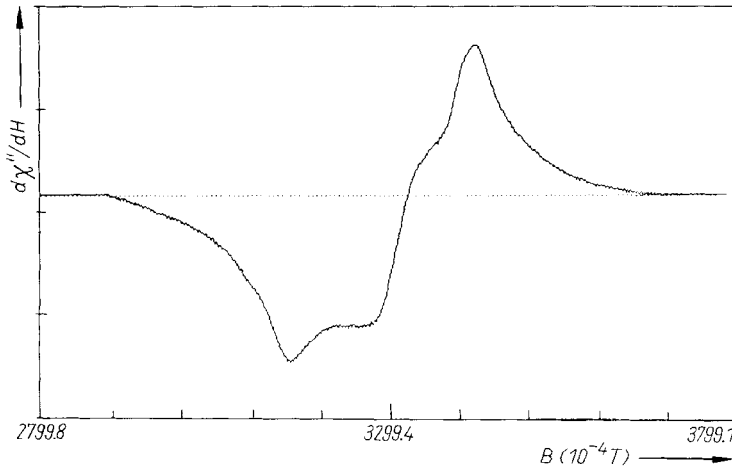


Fig. 4. The simulated powder EPR spectrum of Cu ions in orthorhombic crystal field symmetry

T_C , the spectrometer worked under spoiled conditions. The intensity of the EPR spectrum of the non-HTC samples increases as the temperature decreases. This phenomenon could be explained by invoking the “skin effect” [1].

Fig. 5 presents the temperature dependence of the EPR spectrum of DPPH in a double cavity with and without a non-HTC sample ($Gd_{0.5}Y_{0.5}Ba_2Cu_3O_{7-\delta}$ and $Gd_{0.5}La_{0.5}Ba_2 \cdot Cu_3O_{7-\delta}$). The more intense signal was obtained without a sample. For all investigated non-HTC samples, the intensity seems to decrease as the temperature decreases (Fig. 5a). However, the sample with La atoms shows the opposite behavior (Fig. 5b). The temperature dependence of the intensity may be due to thermal losses of the EPR cavity quality factor Q_0 as the dielectric properties of the sample change. The EPR spectrum of Gd^{3+} ions is not affected by this phenomenon [1].

In the case of the La atom, a strong zero-field non-resonant behavior was recorded at liquid nitrogen temperature (LN). Similar observations have been made on the $Sm_{0.5}La_{0.5} \cdot Ba_2Cu_3O_{7-\delta}$ compound in the tetragonal phase [6]. In the $LaBa_2Cu_3O_{7-\delta}$ compound in the tetragonal phase some periodic pulses on the recorder were observed [12]. We think that these phenomena and the observed increase of the intensity of the DPPH signal with temperature may have the same origin.

Fig. 6 shows temperature dependence of the EPR spectra of non-HTC samples in the low-temperature region. The EPR spectrum coming from the exchange coupled Cu^{2+} pairs, [3, 13] has been detected for all the samples. This spectrum is superimposed on another very broad line coming from Gd^{3+} ions. The signal from Cu^{2+} pairs is very weak in the

Table 3

The values of the EPR linewidths ΔH of $Gd_{0.5}RE_{0.5}Ba_2Cu_3O_{7-\delta}$ compounds in orthorhombic and tetragonal phases at RT

RE	Pr	Nd	Dy	Ho	Er	Tm	Yb	Lu
$\Delta H(T)$ (HTC)	–	0.17	0.17	0.23	0.23	0.17	0.17	0.17
$\Delta H(T)$ (non-HTC)	–	0.21	0.20	0.23	0.23	0.20	0.20	0.20

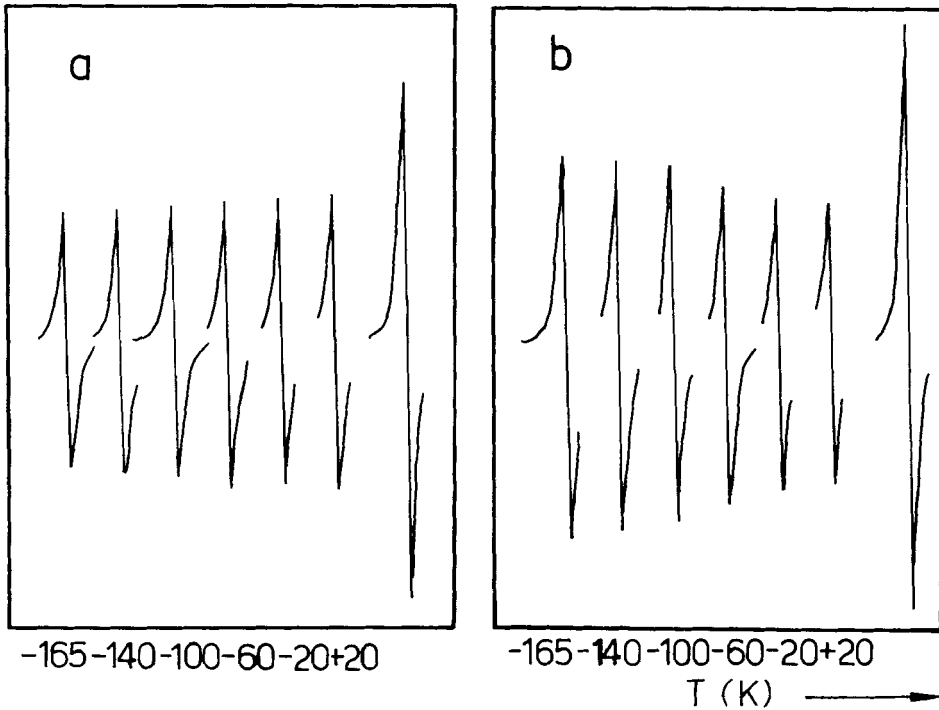


Fig. 5. Temperature dependence of the intensity of the EPR signal of DPPH a) with and b) without non-HTC sample for $Gd_{0.5}Y_{0.5}Ba_2Cu_3O_{7-\delta}$ (I) and $Gd_{0.5}La_{0.5}Ba_2Cu_3O_{7-\delta}$ (II) in the double-cavity system

case of the Ho and Lu samples (Fig. 6c). The values of the effective g -factor and the peak-to-peak linewidth ΔH as a function of temperature are shown in Table 4. A strong temperature dependence of the g -factor in the range 10 to 30 K is observed (Table 4). For some of the samples linewidth changes are also measured. An ordering of the compound at these low temperatures may exist [13]. Magnetic measurements systematically show anomalies in the magnetization data at low temperature (Fig. 1). For the $YbBa_2Cu_3O_{7-\delta}$ compound in the tetragonal phase this phenomenon was interpreted in terms of the fluctuation theory of magnetization [10, 14, 15] predicting a similar effect of ferromagnetic ordering. No anomalous behavior of the magnetic susceptibility was observed for the compounds with Ho and Lu. This is presumably the cause of the weak intensity of the exchange interaction line for these samples. The value of the g -factor is also affected by the rare-earth substitutional ions. The values of g -parameters decrease with decreasing temperature. This could be explained by considering the influence of an additional internal magnetic field. In this case the resonance condition may be described by a relation of the form: $H = (H_0^2 - aH_E)^{1/2}$ [16], where $H_0 = \hbar\omega/gH$ is the resonance field in the paramagnetic state at temperature $T > T_C$, H the resonance field in the ferromagnetic state, and H_E the internal magnetic field.

The intensity I of the EPR line may be described by the following relation [17]:

$$I = 1/T[3 + \exp(J/k_B T)], \tag{1}$$

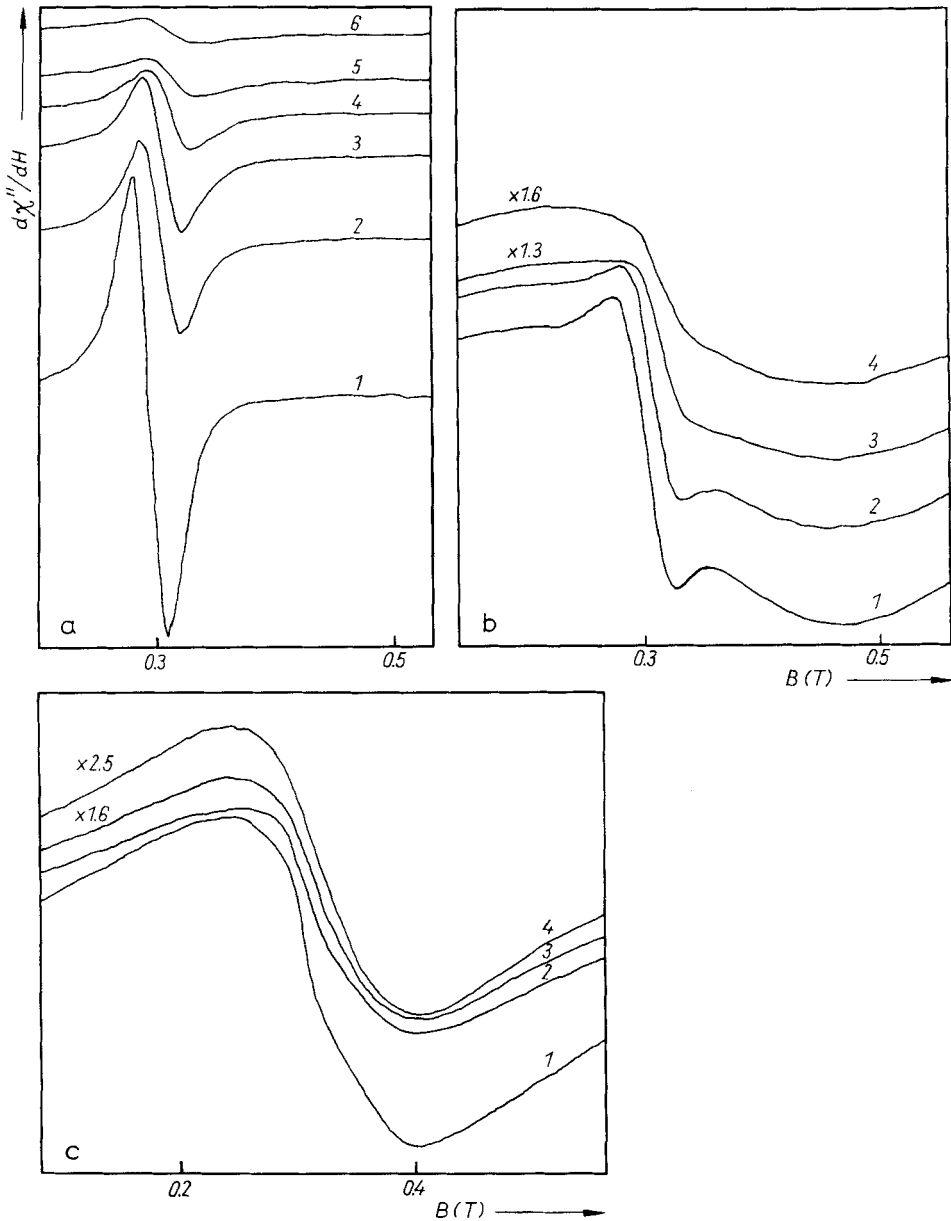


Fig. 6. Temperature dependence of the EPR spectra of $Gd_{0.5}RE_{0.5}Ba_2Cu_3O_{7-\delta}$ compounds in the tetragonal phase for a) Dy: (1) $T = 4.4$, (2) 8, (3) 10, (4) 15, (5) 23, (6) 25 K; b) Er: (1) $T = 4.6$, (2) 10, (3) 20.6, (4) 29.3 K; c) Lu: (1) $T = 4.5$, (2) 14, (3) 25, (4) 34 K.

where J is the exchange interaction constant and k_B the Boltzmann constant. Using (1), for our non-HTC samples (with Nd, Dy, Y, and Er) a value of $J \approx 20$ to 30 cm^{-1} was estimated. These values are in good agreement with those obtained for $Pr_{0.5}Dy_{0.5}Ba_2Cu_3O_{7-\delta}$ and $Gd_{0.5}Sm_{0.5}Ba_2Cu_3O_{7-\delta}$ compounds [13].

Table 4
 The values of spin-Hamiltonian parameter, g , and linewidth, ΔH , for the EPR spectrum of paired copper ions for $Gd_{0.5}RE_{0.5}Ba_2Cu_3O_x$ compounds in the tetragonal phase

T (K)	Gd, La		Gd, Nd		Gd, Dy		Gd, Er		Gd, Y		Gd, Eu		Gd, Tm		Gd, Yb	
	g	ΔH (T)	g	ΔH (T)	g	ΔH (T)	g	ΔH (T)	g	ΔH (T)	g	ΔH (T)	g	ΔH (T)	g	ΔH (T)
3.5	2.187	0.0325	-	-	-	-	-	-	2.202	0.0325	2.368	0.0550	-	-	-	-
4.6	-	-	2.158	0.0500	2.288	0.0305	2.232	0.0500	-	-	-	-	≈ 2.23	≈ 0.05	2.228	0.0360
8.0	2.166	0.0325	-	-	2.215	0.0350	-	-	-	-	-	-	-	-	-	-
10.0	-	-	2.109	0.0460	2.198	0.0340	2.186	0.0500	2.184	0.0325	2.347	0.0550	-	-	2.203	0.0360
15.0	-	-	-	-	2.169	0.0370	-	-	-	-	-	-	-	-	-	-
20.0	-	-	2.109	0.0460	2.158	0.0430	2.135	-	2.123	0.0325	-	-	-	-	-	-
25.0	-	-	2.109	0.0460	2.158	0.0430	-	-	-	-	2.229	0.0425	-	-	≈ 2.16	-
30.0	-	-	-	-	-	-	2.135	-	2.108	0.0325	2.210	0.0375	-	-	-	-
42.0	-	-	-	-	-	-	-	-	2.108	0.0325	2.210	0.0300	-	-	-	-

Table 5
Relative intensities and conductivities for the samples $\text{Gd}_{0.5}\text{RE}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$

T (K)	I/I_0	w	$\ln \sigma$ ($\Omega^{-1} \text{cm}^{-1}$)
(Gd, Eu)–Ba–Cu–O			
3.3	1	—	—
9.4	0.922	0.72	−6.17
18.4	0.750	1.40	−4.83
30.5	0.583	2.03	−4.09
42.4	0.417	2.84	−3.42
(Gd, Ho)–Ba–Cu–O			
4.6	1	—	—
11.0	0.700	1.58	−4.59
22.0	0.472	2.54	−2.54
30.0	0.355	3.26	−3.14
(Gd, Er)–Ba–Cu–O			
4.6	1	—	—
10.0	0.847	1.03	−5.44
20.6	0.600	1.96	−4.16
29.3	0.467	2.56	−3.62
(Gd, Tm)–Ba–Cu–O			
4.8	1	—	—
13.5	0.462	2.59	−3.60
22.0	0.346	3.33	−3.10
(Gd, Yb)–Ba–Cu–O			
4.6	1	—	—
11.0	0.718	1.52	−4.67
22.0	0.468	2.56	−3.62
(Gd, Lu)–Ba–Cu–O			
4.5	1	—	—
14.0	0.694	1.60	−4.56
25.0	0.482	2.49	−3.68
34.0	0.370	3.15	−3.14

The intensity of the EPR line of Gd ions drastically increases as temperature decreases. The conductivity properties essentially influence the intensity of the EPR spectra through the so-called “skin effect”. The relative intensity of the EPR signal as a function of the electrical conductivity (σ) is given by the relation [18, 19]

$$I/I_0 = \{2 \exp(-w) + [1 + \exp(-2w)]/w\} / [1 + \exp(-w)]^2, \quad (2)$$

where $w = d/\delta$ ($d = 2 \text{ mm}$ is the thickness of the conducting sample), $\delta = (2/\mu_0\omega\sigma)^{1/2}$ is the skin depth, and ω a microwave frequency.

Table 5 presents the relative intensities and the conductivities ($\ln \sigma$) for the samples $\text{Gd}_{0.5}\text{Re}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ (Re = Eu, Ho, Er, Tm, Yb, and Lu) in the tetragonal phase at low temperatures.

In this paper we presented experimental results concerning the behavior of non-HTC materials which might be useful for the study of their HTC state. It is clear that other phases or oxygen deficiency essentially influence the EPR spectrum of Gd^{3+} ions and the spectrum due to exchange-coupled pairs of copper ions. We think that this kind of investigation on other series of 123 compounds at low temperatures could shed more light on the extraordinary physical properties of these materials.

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