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Anomalous temperature behavior of resistance in $C_{1-x}Co_x$ thin films grown by pulsed laser deposition technique

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ABSTRACT

We study the transport properties of $C_{1-x}Co_x$ thin films (with $x = 0.1$, 0.15 and 0.2) grown on *Si* substrate by pulsed laser deposition technique. The results demonstrate some anomalous effects in the behavior of the measured resistance $R(T,x)$. More specifically, for $0 < T < T^*$ range (with $T^* \simeq 220$ *K*), the resistance is shown to be well fitted by a small polaron hopping scenario with $R_h(T,x) \propto \exp\left\{\left[\frac{T_0(x)}{T}\right]^{0.5}\right\}$ and a characteristic temperature $T_0(x) \simeq T_0(0)(1-x)$ (with $T_0(0) = 120$ *K*). While for higher temperatures $T^* < T < T_C(x)$, the resistance is found to be *linearly* dependent on spontaneous magnetization $M(T,x)$, viz. $R_M(T,x) \propto M(T,x)$, following the pattern dictated by electron scattering on cobalt atoms formed robust ferromagnetic structure with the Curie temperature $T_C(x)$ obeying a percolation like law $T_C(x) \simeq T_C(x_m)(x/x_m)^{0.15}$ with $T_C(x_m) = 295$ *K* and the maximum zero-temperature magnetization reaching $M(0,x_m) \simeq 0.5\mu_B$ per *Co* atom for $x_m = 0.2$.

1. Introduction

Presently, the interest in nanoscale magnetic materials has increased dramatically due to the enormous potential for their applications in nanostructured devices [1,2], spintronics [3], recording magnetic media [4], etc. Therefore, it is important to better understand the basic principles governing the properties of these materials. In this respect, carbon-based thin films have become one of the most promising material for numerous applications [5–7]. In particular, by doping carbon films with different metallic atoms, it was possible to optimize their electrical and magnetoelectronic properties [8–11]. An important issue to be discussed is the method used for the production of thin films with desirable properties. Many several techniques have been used, including Cathodic Vacuum Arc [12], sputtering [13], Chemical Vapor Deposition [14], and Pulsed Laser Deposition [15–17] among others. In an attempt to achieve the optimal performance of nanodevices, the research has been focusing on finding interrelations between structure (morphology) and physical properties (both magnetic and transport) of high quality deposited nanofilms. Of particular interest are the properties of magnetically ordered metallic atoms implanted into non-magnetic carbon matrix [18–27].

This paper presents our latest results regarding structural and transport properties of $C_{1-x}Co_x$ thin films (with $0.1 < x < 0.2$) grown by using a pulsed laser deposition (PLD) technique. The obtained results demonstrate manifestation of some very unusual phenomena in the temperature and doping dependence of the measured resistance $R(T,x)$.

2. Experimental methods

High quality $C_{1-x}Co_x$ films (with thickness of about 85.3 *nm*) with $x = 0.10$, 0.15 and 0.20 have been grown by PLD technique. Graphite and cobalt superfine powders of high purity (99.98%) were used. Microstructure and crystallographic orientation of the

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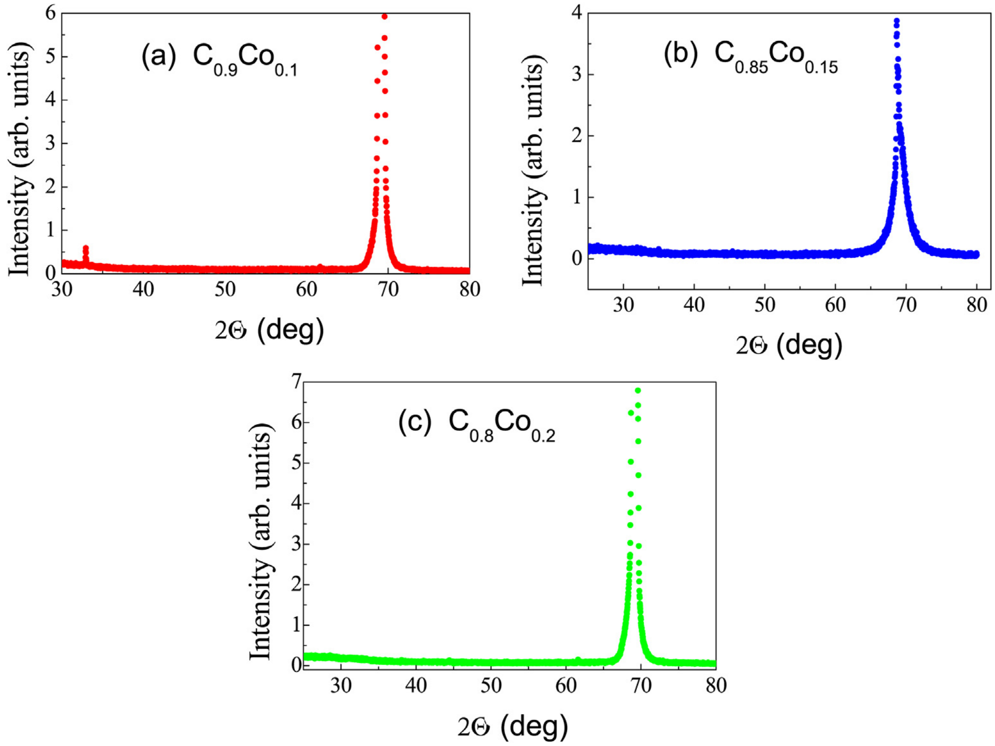

Fig. 1. XRD spectra of $C_{1-x}Co_x$ films with different content of *Co* atoms: (a) $x = 0.1$, (b) $x = 0.15$, and (c) $x = 0.2$.

films were characterized by X-ray diffraction (XRD) scans. The films thickness was confirmed by using field-emission scanning electron microscopy (FEG SEM). Typical XRD spectra and FEG SEM images for our $C_{1-x}Co_x$ films are shown in Fig. 1 and Fig. 2, respectively.

Thin films have been deposited on *Si*(100) (5×5 mm^2) substrates, using a *KrF* excimer laser (with wavelength $\lambda = 248$ *nm* and 25 *ns* pulse width). The laser is operated at a repetition rate of 8 *Hz* and energy density of 3.2J/cm^2 and was focused onto a 2.5 *mm* diameter $C_{1-x}Co_x$ pellet. Rotation speed of the target was 15 *rpm*. The substrate was first mounted, using silver paste, onto a non-rotating heater, which was positioned parallel to the target surface. The distance between the target and the substrate was

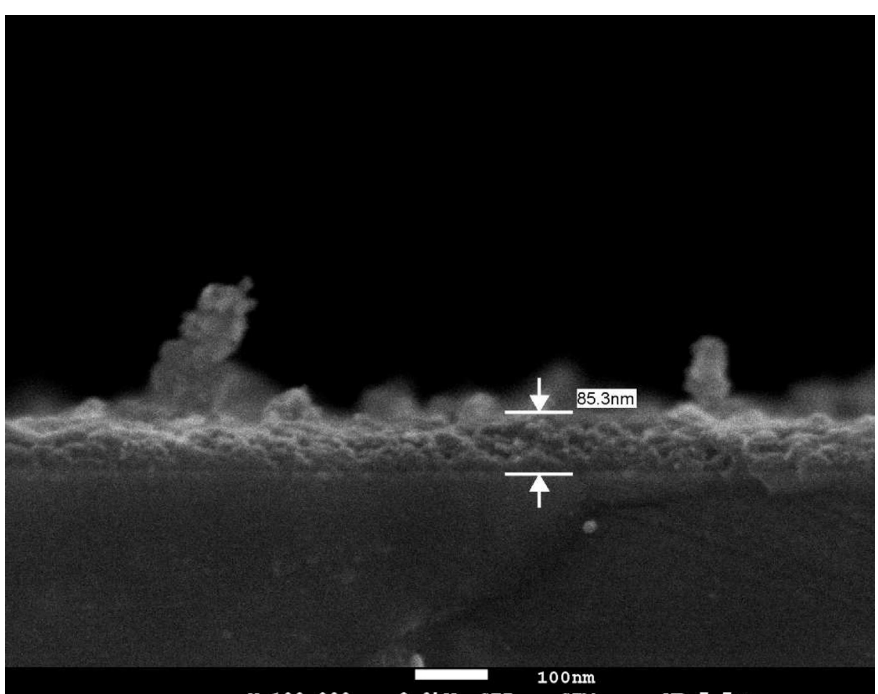

Fig. 2. Typical FEG SEM image of deposited $C_{1-x}Co_x$ films.

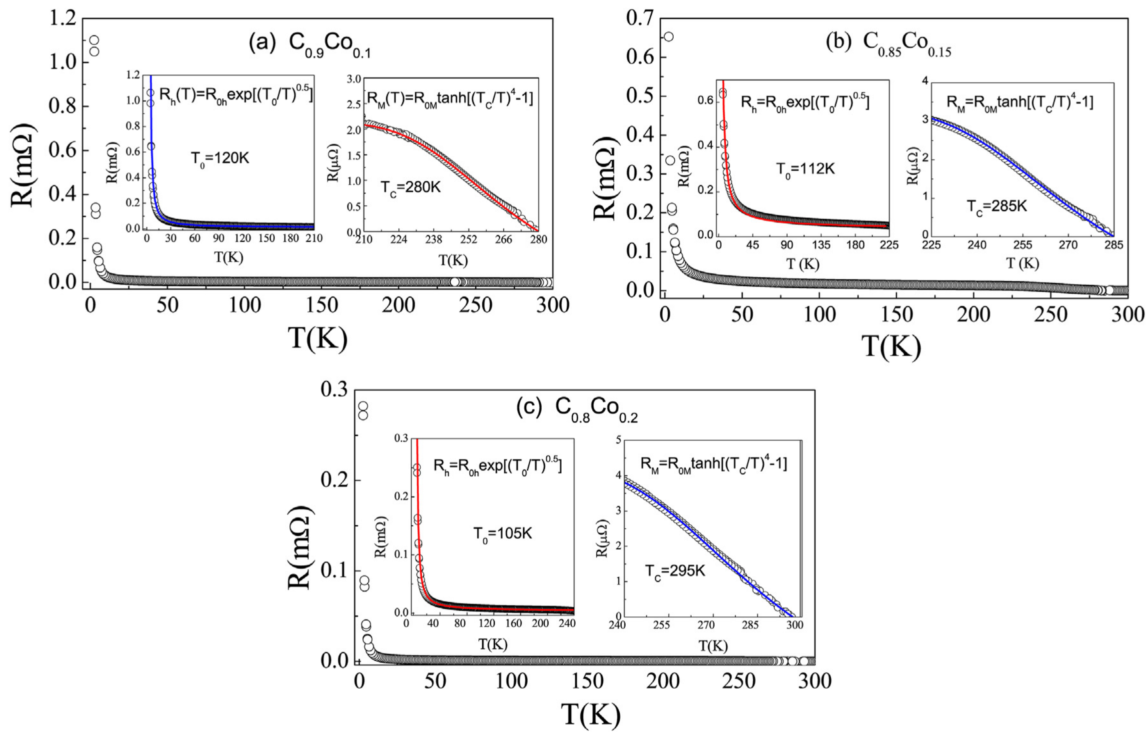

Fig. 3. Temperature dependence of the resistance $R(T)$ measured for $C_{1-x}Co_x$ thin films with different content of *Co* atoms: (a) $x = 0.1$, (b) $x = 0.15$, and (c) $x = 0.2$. The solid lines are the best fits for hopping $R_h(T,x)$ and magnetic scattering $R_M(T,x)$ contributions.

maintained at 3.8 *cm*. This distance was defined as the best fit according to the shape of the plume. In this way, the tip of the plume could just cover the substrate during deposition. Before starting the deposition, the chamber was evacuated with a turbo pump to a background pressure of 2×10^{-7} *mbar* and the substrate was maintained at a temperature of 800 °*C* for 30 min. During the deposition process, the substrate was maintained at 500 °*C* in argon pressure of 0.5 *mbar*. The temperature of the heater block was measured with *K*-type thermocouples placed in contact with the heater block directly behind where the samples were mounted. When the deposition was completed the argon pressure was hold constant and the temperature was decreased to 400 °*C* where it remained for 30 min for in-situ annealing of the films and thereafter the substrate temperature was cooled naturally. Before deposition, the *Si* substrates were ultrasonically cleaned. For the purpose of removing formation of SiO_2 from the surface of the substrate, a final cleaning was performed through using the chemical reaction in Ref. 10% dilute of HF.

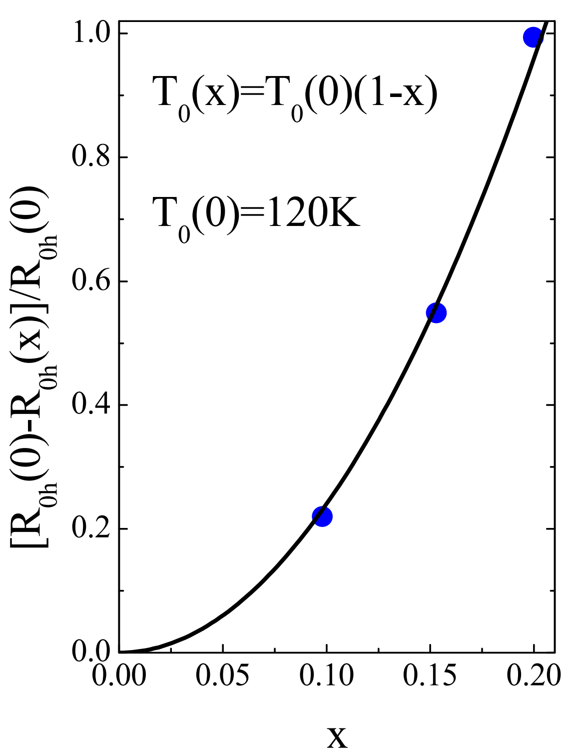

Fig. 4. The experimentally determined variation of the normalized hopping driven resistivity $[R_{oh}(0) - R_{oh}(x)]/R_{oh}(0) \simeq x^2$ and hopping temperature $T_0(x) \simeq T_0(x)(1-x)$ with the concentration of *Co* atoms x.

The electrical resistance $R(T)$ was measured using the conventional four-probe method. To avoid Joule and Peltier effects, a dc current $I = 100\mu A$ was injected (as a 1 s pulse) successively on both sides of the sample. The voltage drop V across the sample was measured with high accuracy by a *KT*256 nanovoltmeter.

3. Results and discussion

Fig. 3 shows the typical results for the temperature dependence of the resistance $R(T)$ in our $C_{1-x}Co_x$ thin films with different content of *Co* atoms. The left and right insets describe a low-temperature and high-temperature regions. The solid lines are the best fits of the experimental data according to the suggested scattering scenarios (see below).

Since no tangible structural changes have been observed upon cobalt doping, the Jahn-Teller mechanism can be safely ruled out and the most reasonable cause for the resistance drop in the doped material is the reduction of the spin-polaron tunneling energy $E_\sigma(x)$ which within the localization scenario [28–30] is tantamount to an

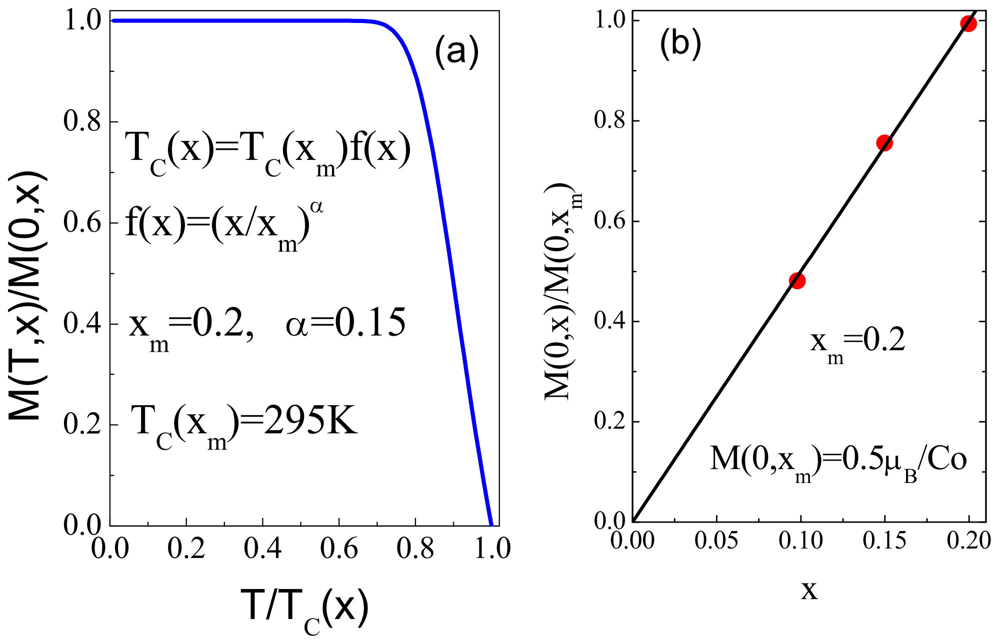

Fig. 5. (a) The temperature dependence of $M(T,x)$ predicted by Eq. (3) for all T and x. (b) The extracted x dependence of the normalized magnetization $M(0,x)/M(0,x_m)$ with $M(0,x_m) \simeq 0.5\ \mu_B$ (μ_B is the Bohr magneton).

increase of the charge carrier correlation length $L(x) \simeq \hbar/\sqrt{2mE_\sigma(x)}$ (here m is an effective carrier mass). In the low-temperature ferromagnetic (FM) region ($0 < T < T^*$), the resistance is assumed to be due to polaron hopping

$$R_h(T,x) = R_{oh}(x)\exp\left\{\left[\frac{T_0(x)}{T}\right]^{0.5}\right\} \tag{1}$$

with a characteristic temperature $T_0(x) \simeq E_\sigma(x)/k_B$.

The experimentally determined variation of the normalized hopping driven resistivity $[R_{oh}(0) - R_{oh}(x)]/R_{oh}(0) \simeq x^2$ and hopping temperature $T_0(x) \simeq T_0(x)(1-x)$ with the concentration of Co atoms x are depicted in Fig. 4. In turn, the above dependencies result in the following reasonable estimate of the charge carrier correlation length in our films $L(x) \simeq L(0)/\sqrt{1-x}$ with $L(0) \simeq 2\ nm$ (assuming a free electron mass value for m).

At the same time, for $T^* < T < T_C(x)$, the resistance seems to be dominated by electron scattering on cobalt created FM structure with spontaneous magnetization $M(T,x)$, namely

$$R_M(T,x) = \gamma M(T,x) \tag{2}$$

with

$$M(T,x) = M(0,x)\tanh\left\{\left[\frac{T_C(x)}{T}\right]^4 - 1\right\} \tag{3}$$

Here, $M(0,x)$ is the saturation magnetization.

In fact, Eq. (3) is an analytical (approximate) solution of the well-known Curie–Weiss mean-field equation on spontaneous magnetization (valid for all temperatures), viz [28–30].

$$\frac{M(T,x)}{M(0,x)} = \tanh\left\{\left[\frac{M(T,x)}{M(0,x)}\right]\left[\frac{T_C(x)}{T}\right]\right\} \tag{4}$$

Fig. 5(a) shows the temperature dependence of $M(T,x)$ predicted by Eq. (3) for all T and x. Notice a rather strong FM ordering till $T < 0.8T_C(x)$. Besides, the dependence of the Curie temperature $T_C(x)$ on x is found to follow a percolation (non-linear) law $T_C(x) \simeq T_C(x_m)(x/x_m)^\alpha$ with $T_C(x_m) = 295\ K$, $x_m = 0.2$ and $\alpha = 0.15$. Finally, Fig. 5(b) shows the extracted x dependence of the normalized zero-temperature magnetization $M(0,x)/M(0,x_m)$ with $M(0,x_m) \simeq 0.5\ \mu_B$ per Co atom (μ_B is the Bohr magneton).

4. Conclusion

In summary, some very unusual transport properties of $C_{1-x}Co_x$ thin films (with $x = 0.1$, 0.15 and 0.2) grown on Si substrate by pulsed laser deposition (PLD) technique have been reported and attributed to manifestation of small polaron hopping (at low temperatures) and a strong electron scattering on cobalt atoms created robust ferromagnetic structure (at high temperatures).

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