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MICROSCOPIC EXPLANATION OF THE MULTIFERROIC PROPERTIES IN M-DOPED (M = Co, Cr, Mg) ZnO THIN FIMS

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***Keywords*:** *ZnO thin film, Co-, Cr- and Mg-ion doping, multiferroic properties*

***Research area*:** *physical science; solid state physics*

ABSTRACT

The origin of multiferroism is still an open problem in ZnO. We propose a microscopic model to clarify the occurrence of multiferroism in this material. Using a Green's function technique we study the influence of ion doping and size effects on the magnetization and polarization of ZnO thin films. The calculations for magnetic Co- and Cr-ions are based on the s-d model, the transverse Ising model in terms of pseudo-spins and a biquadratic magneto-electric coupling, whereas in case of nonmagnetic Mg-ions the model takes into account the Coulomb interaction and an indirect coupling between the pseudo-spins via the conduction electrons. We show that the magnetization M exhibits a maximum for a fixed concentration of the doping ions. Furthermore M increases with decreasing film thickness N. The polarization increases with increasing concentration of the dopant and decreasing N. The results are in good agreement with the experimental data.

1. Introduction

Zinc oxide (ZnO) has been a subject of technological interest because of its potential applications in ultraviolet light emission devices, chemical or biological sensors, and piezoelectric devices. Since the mechanical, optical and vibrational properties can be modified by size reduction, shape modification, and surface properties ZnO thin films are

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drawing attention as well. Recently, oxide-based materials are found to exhibit multiferroic properties where both ferroelectric and ferromagnetic orders coexist.

The origin of the room temperature ferromagnetism (RTFM) observed in ion doped ZnO is still controversial. Although RTFM for Cr-doped ZnO has been predicted in some theoretical studies using first principle calculations [1-4] there are still some conflicts within experimental results. Whereas Roberts et al. [5] and Zhuge et al. [6] have observed a decrease of the saturation magnetization M_S with increasing of Cr-ions, Fu et al. [7] and Duan et al. [8] have offered an increasing of M_S. Several studies reveal that the magnetic behavior of Cr-doped ZnO is very sensitive to the preparation methods. Meanwhile, several authors have reported that the Cr dopant is incorporated into the ZnO lattice in form of Cr^{3+} ions [7,8], while other authors have observed that Cr ions in the Cr-doped ZnO film exist mainly in the form of Cr^{2+} [6].

A similar situation is observed in ZnO doped with Co. This material is the first ZnO-based RTFM semiconductor that was theoretically predicted and experimentally examined [9,11]. Despite being one of the most representative and extensively investigated wide-gap magnetic materials [11], Co-doped ZnO lacks a conclusive explanation of its magnetic properties. There are several speculations concerning the origin of the observed ferromagnetism ranging from electron-induced (or enhanced) ferromagnetism [10-12], defect-induced ferromagnetism by the formation of bound magnetic polarons [13,14] up to extrinsic process due to the precipitation of a second phase [15,16].

Ferromagnetism in ZnO nanoparticles doped with a nonmagnetic element like Al is reported by Gao et al. [17], which is related to the doping-induced oxygen vacancies. This situation is related to the problem of the so-called d_0 ferromagnetism which could explain also the origin of RTFM in bulk ZnO and undoped ZnO thin films [17,18].

Additionally, ferroelectricity has been observed in doped ZnO bulk crystals and thin films where Zn sites are substituted by Li, Be, Mg, V, Cr, Cu. ZnO has a hexagonal wurtzite structure which lacks the center of symmetry. As a result it is expected that the material exhibits ferroelectric behavior. Due to the difference in ionic radii of Zn^{2+} and M^{2+} ions, the Zn-O bond length will be reduced and a lattice distortion takes place which contributes to the ferroelectric behavior of this oxide too.

The aim of the present paper is to propose a microscopic model for doped ZnO based on the s-d model. In particular, we have shown that the experimentally observed RTFM is an intrinsic property which can be originated due to doping effects in ZnO nanoparticles. As demonstrated in this paper the situation is more complex in ZnO thin films. So, we analyze the origin of the multiferroic behavior of ZnO films which are ion-doped by magnetic and nonmagnetic substances.

2. The Model and the Green's Functions.

The Hamiltonian describing the magnetic properties of transition metal or rare earth ion doped ZnO thin films is the s-d(f) Hamiltonian which is proposed for ferromagnetic semiconductors

$$H_{s-d} = H_{sp} + H_{el} + H_{sp-el} \tag{1}$$

H_{sp} is the Heisenberg model of the d or f-electrons in the transition metal ions defined by

$$H_{sp} = -\sum_{i,j} J_{ij}(S_i^+ S_j^- + S_i^z S_j^z) - \sum_i D_i (S_i^z)^2 - g\mu_B H \sum_i S_i^z \ . \tag{2}$$

Here S_i^+, S_i^- and S_i^z are the spin-operators for the localized spins at lattice site *i*, J_{ij} is the magnetic exchange interaction between neighboring sites i and j, D_i is the single-site anisotropy parameter, and H is an external magnetic field. Surface effects are included by assuming different coupling parameters between the localized spins within the surface layer J_s and within the bulk J_b.

H_{el} represents the Hamiltonian of the conduction band electrons

$$H_{el} = \sum_{i,j,\sigma} t_{ij} c_{i\sigma}^+ c_{j\sigma} , \tag{3}$$

where t_{ij} is the hopping integral, $c_{i\sigma}^+$ and $c_{i\sigma}^-$ are Fermi-creation and annihilation operators.

The operator H_{sp-el} couples the two subsystems, given by Eqs. (2) and (3), respectively. With the intra-atomic exchange interaction Ii the s-d interaction is defined by

$$H_{sp-el} = \sum_i I_i S_i s_i . \tag{4}$$

The spin operators si of the conduction electrons at site *i* can be expressed by Fermi-operators $s_i^z = c_{i+}^+ c_{i-}$ and $s_i^z = (c_{i+}^+ c_{i+} - c_{i-}^- c_{i-}) / 2$, where ± characterizes the spin orientation.

There is a great evidence that ion doped ZnO bulk crystals and thin films exhibit a ferroelectric behavior which could be due to the different ionic radii of Zn^{2+} and the dopant ions. The bond length (Zn-O) is found to be reduced when the Mg content is enhanced from zero to 0.05 [19]. The small structural distortion caused by the bond-length variation is likely responsible to drive the ferroelectric behavior in $Zn_{1-x}Mg_xO$ systems. Wang et al. [1] have suggested that the ionic displacements of Cr dopants with respect to the Zn positions are able to occur in the ZnO-Cr system. Apparently, this kind of distortions could locally induce electric dipoles in the CrO_4 tetrahedral and thereby contribute to polarizations. In case of the Mg-doped ZnO, there is a large difference in size between Zn and Mg ions. It is expected that Mg ions may occupy off-centered positions, replacing the host Zn ions and forming electric dipoles. Therefore, we assume that the origin of the ferroelectricity can be described by an order-disorder model expressed by the Ising model in a transverse field (TIM):

$$H_{TIM} = -\Omega \sum_i B_i^x - \frac{1}{2} \sum_{i,j} J'_{ij} B_i^z B_j^z . \tag{5}$$

Here the pseudo-spin operator B_i^z characterizes the two position of the ferroelectric unit at the lattice point *i*. The interaction between adjacent lattice sites is denoted by J'_{ij}. The flip-dynamics of the model is determined by the operator B_i^x, the strength of it is denoted by Ω. The ordered phase is characterized by non-zero averages $< B^x > \neq 0$ and $< B^z > \neq 0$. The situation can be simplified by introducing a new coordinate system for Eq. (5). Rotating the frame in the *xz* plane the rotation angle θ is determined by the requirement $< B^x > = 0$ in the new coordinate system.

The coupling between the magnetic and the electric subsystems in the ferroic compound is given by the Hamiltonian H_{mf}

$$H_{mf} = -g\sum_{ijkl} B_i^z B_j^z (S_i^+ S_j^- + S_i^z S_j^z)\,. \tag{6}$$

The coupling parameter g describes the strength between the magnetic and the electric order parameters. Such a biquadratic coupling between the pseudospins and magnetic moments as in Eq. (6) implies that the magnetic and the ferroelectric orderings underlie independent mechanisms. In particular, such a coupling gives rise for different well separated transition temperatures T_C^{FE} for the ferroelectric subsystem and T_C^{FM} for the magnetic one.

The magnetization M of the local spins system of the transition metal ions; the polarization P of the pseudo-spin system and the magnetization of the n-th shell of the conduction electron $< s_n^z >$ are calculated usihf the following Green's functions:

$$G_{ij}(M) = << S_i^-; S_j^+ >>;\; G_{ij}(P) = << B_i^-; B_j^+ >>;\;\; g_{ij\sigma} = << c_{i\sigma}; c_{j\sigma}^+ >>\,.$$

In order to investigate the doping effects and their influence on the magnetic and electric properties of ZnO thin films we have to complete the Hamiltonian by corresponding terms. In case of **nonmagnetic dopant ions** such as Mg we propose

$$H = H_{TIM} + H_{el} + H_{TIM-el}\,. \tag{7}$$

The first term is the TIM already defined in Eq. (5). It describes now the interaction between the local dipole associated with the ionic mismatch between Mg and Zn ions which causes Mg to occupy an off-centered position, giving rise to an electric dipole moment. We suppose that all off-center ion dipole moments have two possible directions along the z-axis. The second term in Eq. (7) characterizes the conduction electrons which are subjected to the Coulomb interaction. The general form is

$$H_{el} = \sum_{i,j,\sigma} t_{ij} c_{i\sigma}^+ c_{j\sigma} + \frac{1}{2}\sum_{ijkl,\sigma\sigma'} v_{ijkl} c_{i\sigma}^+ c_{j\sigma'}^+ c_{k\sigma} c_{l\sigma}\,. \tag{8}$$

The last term in Eq. (7) describes the indirect coupling between the pseudospins via the conduction band electrons

$$H_{TIM-el} = \sum_{i\sigma} I'\sigma (c_{i\sigma}^+ c_{i\sigma})(B_i^x + B_i^z)\,. \tag{9}$$

It seems possible that carrier-electric dipole interaction may lead to a new kind of dipole-dipole interaction. This indirect interaction of electric dipoles through carriers could be considered to be a Rudermann-Kittel-like interaction. It should be mentioned that the Hamiltonian in Eq. (7) is likewise relevant for the description of the multiferroic properties of undoped ZnO materials.

Following the approach proposed in the first part of this section we obtain the contributions to the magnetization M in the form:

$$M = \frac{1}{2}\sum_i (n_{i\sigma} - n_{i-\sigma}) \tag{10}$$

with

$$n_{i\sigma} = \frac{1}{\exp(E_{i\sigma} / k_B T) + 1} \text{ and}$$

$$E_{i\sigma} = \sum_j [t_{ij} - \sigma I' < B_i^z > \cos\theta_i \delta_{ij} + \sum_{l\sigma'} (v_{illj} - v_{iljl}\delta_{\sigma\sigma'}) < n_{lj\sigma'} >] . \quad (11)$$

The contribution to the polazization P_n is:

$$P_n = < B^z >_n = \frac{1}{2} \sum_i \tanh(\varepsilon_i / k_B T) \cos\theta_i \quad (12)$$

with

$$\varepsilon_i = 2\Omega_i \cos\theta_i + 2\cos\theta_i I' \sum_\sigma \sigma n_{i\sigma} + \sum_j J'_{ij} \cos\theta_i \sin\theta_j < B_j^z > + \\ + \frac{1}{2} \sum_j J'_{ij} \cos\theta_i \sin\theta_j < B_j^+ B_j^- > . \quad (13)$$

Let us point out that there appears a term of the form $I' < B^z >$ in the energy of the conduction band electrons $E_{i\sigma}$, see Eq. 11. This term occurs due to the indirect coupling of the electrons and the dipoles in Eq. (9) and contributes accordingly to the magnetization. Additionally it leads to an additive shift of the two bands. Moreover, because of $P =< B^z >$ the electron energy and the magnetization *M*, compare Eq. (10), could be also depend on an external electric field E. In a similar manner the pseudo-spin-wave energy, see Eq. (13), includes a term $2\cos\theta_i I' \sum_\sigma \sigma n_{i\sigma} = 2\cos\theta_i I' M$. Since the magnetization *M* can be altered by an external magnetic field *H*, the polarization *P* could be controlled by the magnetic field *H*. This is an evidence of the multiferroic properties of non-magnetic ion doped ZnO thin films.

3. Results and discussion

In this section we present the numerical results based on our theoretical calculations for magnetic and nonmagnetic ion-doped ZnO thin films. Bulk ZnO crystallizes in a wurtzite structure and reveals a diamagnetic behavior. The doping of different ions into the ZnO lattice induces different strains and leads to a variation in the lattice parameters depending on the dopant concentration. Such changes are able to modify the electronic and magnetic properties of the material.

From the numerical calculation, using model parameter extracted from the experimental observation for ZnO:(Co, Cr and Mg); we can conclude: 1/ For all doped ZnO thin film we get RTFM i.e the observed ferromagnetism of the ion-doped ZnO thin films should be an intrinsic property of the material.; 2/ The spontaneous magnetization M_S increases firstly and achieves a maximum value after that it rapidly decreases (the magnetism of the ion-doped thin ZnO films depends strongly on the ion-doping content). 3/ Furthermore, the exchange interaction between the magnetic-ions (Co, Cr) can be mainly ferromagnetic due to the s-d interaction or antiferromagnetic due to the super-exchange Me-Me interaction where Me=Co,Cr for low and high Co, Cr-concentrations, respectively. 4/ For

a high Co-, Cr- concentration where antiferromagnetic super-exchange Me-Me interactions predominate the magnetization decreases and the changes in M_S are not so distinctive large, Ms is nearly x independent, in agreement with the experimental data of Pan et al. [11]. In case all Zn ions are substituted by Co ions, $x = 1$, the magnetization will not be zero because there is a contribution to M due to surface effects and due to uncompensated surface spins. 6/ The spontaneous magnetization Ms increases with the single-ion anisotropy D and with the s-d interaction constant I . With increasing I the peak in Ms shifts to higher dopant concentration values. 7/ We obtain that Ms decreases with increasing of J and the peak shifts to smaller x-values. 8/ Ms increases with increasing of the magnetoelectric coupling constant g. The increase of g leads to a shift of Ms to higher Me-concentrations. 9/Because Mg is a nonmagnetic ion the conduction electron magnetization is obtained from the modiffed model Eq. (7) including the Coulomb interaction between the conduction electrons and an indirect coupling between the pseudo-spins and the carriers. The results of Ms(x) for different numbers of layers, N = 5, 7, 11, are shown in Fig. 2. One observes the maximal value of Ms at x = 25%, which is in agreement with the experimental data. The value is smaller compared to the maximum value of Co- and Cr-doped ZnO thin films and its position is at a higher value compared to Cr and Co.

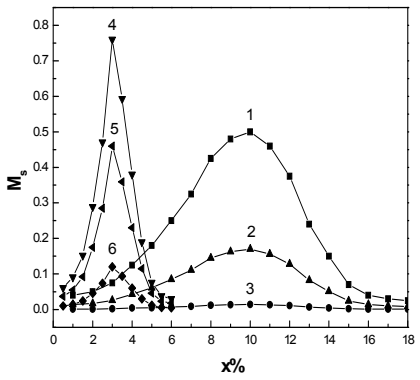

Fig. 1. Dependence of the spontaneous magnetization Ms on the doping concentration x for Co: (1) N = 5, (2) 7, (3) 11 layers; and Cr: (4) N = 5, (5) 7, (6) 11 layers for T =300 K

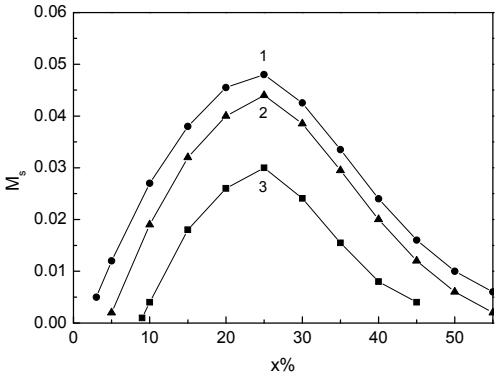

Fig. 2. Dependence of Ms on the Mg-doping concentration x for T = 300 K and different layer numbers N: (1) 5, (2) 7, (3) 11

The spontaneous polarization Ps is calculated for ZnO thin films doped with Cr- and Co-ions in Eq. (10) and with Mg-ions from Eq. (20). The results are represented in Fig. 3. For small dopant concentration x the polarization increases nearly linearly with increasing concentration x and remains constant for larger doping concentration. Such an increasing of Ps is observed experimentally for different ions, such as Cr, Mn, Fe [20], Mg [21], Cu [22], V [23], Li [24]. We find that the polarization of Cr-doped ZnO thin films is larger compared to that of Co- and Mg-doped ZnO thin films. Moreover, we have calculated the film thickness dependence of Ps in the case of Mg-doping. The polarization increases with decreasing film thickness as depicted in Fig. 3, curves 1 and 2. The qualitative explanation is: By doping with non-central ions the ZnO films display ferroelectric behavior. When host Zn ions are replaced by Me ions then a small structural distortion is induced along the polar c-axis. The large ionic size mismatch between Zn and Me ions, gives reason that Me occupies an off-center position thereby inducing permanent electric dipoles. Ferroelectricity

of a non-perovskite ferroelectric semiconductor of ZnO:Me is be due to an indirect interaction between Me impurities that become dipolar via the conduction electrons of the host ZnO crystal. We assume that the polarization in ion-doped ZnO thin films appears due to ionic displacements. The mechanism is similar to the displacive mechanism in perovskite ferroelectrics. From our numerical calculation is clear that: 1/ The polarization increases with decreasing film thickness; 2/ P_S increases strongly with an increasing of the interaction constants J' and I'. 3/ the ferroelectric phase transition temperature T^{FE}_C increases also with the Mg-concentration.

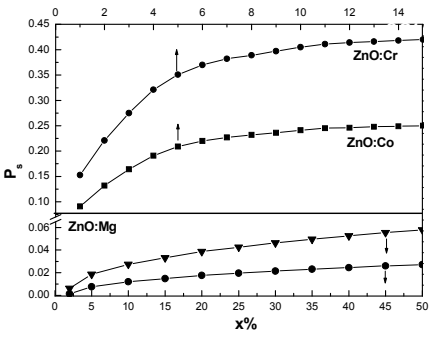

Fig. 3. Dependence of the spontaneous polarization P_S on the doping concentration x for Cr, Co and Mg: (1) N = 7, (2) 11 layers for T = 300 K

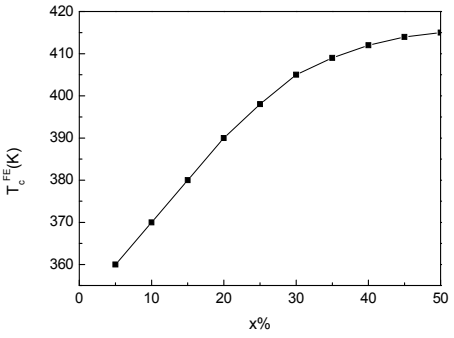

Fig. 4. Dependence of the ferroelectric critical temperature T^{FE}_C on the Mg-concentration x for N = 7 layers

4. Conclution

In the present paper we have investigated and discussed the multiferroic properties in magnetic- and nonmagnetic-ion doped ZnO thin films. To our understanding the origin of ferroelectricity is the small structural distortion caused by the different ionic radii of the doping and host ions. It is expected that the doping ions may occupy off-centered positions, replacing the host Zn ions and forming electric dipoles along the polar c-axis. Such an observation suggests to describe the ferroelectric properties by the transverse Ising model.

The spontaneous magnetization M_S in ZnO thin films doped with transition metal ions such as Co and Cr, is studied on the basis of the s-d model. Because experimental observations have suggested that the magnetic and ferroelectric orderings are subjected to independent mechanisms, we have taken into account a biquadratic coupling between the two order parameters magnetization and polarization and their counterparts.

In case of doping with nonmagnetic ions, such as Mg, the microscopic model has to be modiffed by including Coulomb interaction in the Hamiltonian of the conduction band electrons and an indirect coupling between the pseudo-spins. The last realization is similar to a Rudermann-Kittel interaction. From here we conclude that this indirect coupling offer the facility that an electric field can change the magnetization and vice versa a magnetic field can alter the polarization. Such a mechanism gives a clear evidence of multiferroism. Notice that our model could describe the multiferroic properties in undoped bulk and ZnO nanostructures, too.

For all doping ions we obtain a RTFM which should be the intrinsic property of the material. The polarization P_S and the phase transition temperature T^{FE}_C increase with an increase of the doping concentration. The spontaneous magnetization M_S and the spontaneous polarization P_S exhibit the largest values for Cr-doped and the smallest one for Mg-doped ZnO thin films. They increase strongly with decreasing film thickness. The ferroelectric critical temperature is significantly larger than the ferromagnetic one which leads to a biquadratic magnetoelectric coupling in the case of Co and Cr doping. The results are in qualitative agreement with the experimental data.

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МИКРОСКОПИЧЕН АНАЛИЗ НА МУЛТИФЕРОИЧНИТЕ СВОЙСТВА НА M-ДОТИРАНИ (M = Co, Cr, Mg) ZnO ТЪНКИ ФИЛМИ

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***Ключови думи*:** *ZnO тънки филми, Co-,Cr-,Mg-йонно дотиране, мултифероични характеристики*

***Научна област*:** *физически науки, физика на твърдото тяло*

РЕЗЮМЕ

Произходът на мултифероизма в ZnO е все още нерешен въпрос за физиката на твърдото тяло. Ние предлагаме микроскопичен модел с цел изясняване на наблюдавания мултифероизъм в тези вещества. Чрез метода на функциите на Грийн е изследвано влиянието на йонното дотиране и размерните ефекти върху намагнитеността и поляризацията в тънки филми на ZnO. Пресмятанията за магнитните Co- и Cr- йони се базира на s-d модела, напречния изинг модел в представяне на псевдоспиновите оператори и биквадратична магнетоелектрична връзка. В случая на немагнитни Mg-йони моделът отчита и Кулоновото взаимодействие и индиректното сдвояване между псевдоспиновете и проводящите електрони. Показано е че намагнитеността има максимум при определена концентрация на примесните йони и нараства при намаляване на дебелината на филма N. Поляризацията нараства с нарастване на концентрацията на примесите и намалява с N. Резултатите са в добро съвпадение с експерименталните резултати.

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