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Theoretical model investigating the magnetic properties of cobalt-doped ZnO

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Abstract

We propose a theoretical model to investigate the magnetic properties of cobalt-doped ZnO (ZnO:Co) thin films qualitatively. The model was built on the dilute Co dopants in the host of ZnO forming the magnetic Co^{+2} ions and the energy level of the magnetic ions crossing the band edge of ZnO resulting in a magnetic interaction between the Co^{+2} spins and the spins of the electrons from the conduction band of ZnO. The mechanism of the ferromagnetism revealed in the studied system is proposed here to be induced not only by the mediated conducting electrons via spin interactions but also by the Coulomb excitations, arising from the electrons localized by the oxygen vacancies. This approach of including Coulomb excitation in the modified carrier-mediated model could explain well the magnetic properties of ZnO:Co and solves the drawback of the carrier-mediated model in interpreting the appearance of ferromagnetism in the insulating ZnO:Co. We propose that the Coulomb excitations induced by the electrons captured by the oxygen vacancies are an essential element in the magnetic ZnO, which reveals the fact that the bound magnetic polaron model without considering the Coulomb excitation is insufficient to describe the magnetic properties of ZnO.

Keywords: Coulomb excitation, spin wave excitation, cobalt-doped ZnO, ferromagnetic ZnO, carrier-mediated model

(Some figures may appear in colour only in the online journal)

1. Introduction

Wide bandgap magnetic semiconductors are continuously studied for their important applications in spintronics [1–5], in which charge and spin properties are manipulated simultaneously resulting in novel emerging functionalities of the devices [6, 7]. In particular, owing to the wide bandgap, these semiconductors are transparent. In this paper, we are considering ZnO as an example of the wide bandgap transition metal oxide semiconductor, which has attracted lots of attention due to its easily controlled electric and magnetic properties [8–10]. Regarding electrical properties, many experiments evidenced that adding a small amount of Al or Ga to ZnO thin films results in effective n-type doping. Moreover, it was shown that ZnO becomes highly conductive by introducing more heavy dopants [11–13]. Besides, it was explicitly evidenced that annealing of ZnO thin films in low oxygen (O_2) pressure increases the conductivity [14, 15]. In fact, the conductivity

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of metal-doped ZnO has reached a competitive level of commercial indium tin oxides (ITO) for satisfying the application of transparent conductors [16, 17].

In contrast to the electric properties, the magnetic properties of magnetic ion-doped ZnO are relatively less clear [18–23]. Consequently, different magnetic properties of doped ZnO have emerged from doping with various magnetic ions, which arise probably from different dopant energy levels in ZnO [24]. In this paper, we focus on cobalt-doped ZnO (ZnO:Co), in which weak ferromagnetism appears at room temperature. In fact, there are lots of controversies concerning the origin of ZnO ferromagnetism including the exclusion of Co dopant origin. Moreover, it is well established that oxygen (O) vacancies also play an important role in the emergence of ferromagnetism, since ZnO:Co samples annealing in high O_2 pressure suppresses the ferromagnetism [25–27]. It can be expected that the correlation between oxygen vacancies and magnetic dopants should be very complex and its complexity arises from different charge states of Co [28, 29]. Nevertheless, the magnetic coupling between the Co dopants electrons and those from the ZnO conduction band is the key interaction determining the magnetic properties of Co-doped ZnO, which is also the main interaction considered in our proposed theoretical model [30–32].

Magnetic anisotropy is another interesting characteristic in ZnO:Co [24, 33–36]. In particular, it has been demonstrated that UV irradiation increases the conductivity of band electrons [37, 38] as well as enhances the magnetic anisotropy [39]. The increase of the electron conductivity enhances the ferromagnetism, which seems to be consistent with the increase of ferromagnetism with an increasing number of O vacancies. It has been demonstrated that the appearance of O vacancies is equivalent to electron doping [40, 41]. The magnetic anisotropy of ZnO:Co thin film arises most probably from the intrinsic single-ion anisotropy of Co [33, 42], which results in the anisotropic magnetic coupling between Co and ZnO conduction electrons. The valence of Co in ZnO is closely related to the presence of O vacancies which governs, in fact, the magnetic properties of ZnO:Co [43, 44]. We can make a conclusion that if the valence of Co is +2, i.e. completely ionized, the Co ions will have strong magnetic coupling with the conduction band electrons of ZnO; but if Co is only partially ionized the magnetic coupling should be reduced. Besides, if the clustering of Co dopants occurs the emergence of ferromagnetism arises mostly from the Co clusters.

As aforementioned, the O vacancies intrinsically exist in ZnO:Co thin films. It can be expected that the vacancies may accommodate or capture itinerant electrons to diminish the bonding instability resulting in a Coulomb repulsion between these captured electrons. It can be expected that the Coulomb repulsion will induce the on-site ferromagnetism by Coulomb excitation, called Stoner excitation. Evidently, the important aspect of the ferromagnetism in ZnO:Co arises from the spin interactions accompanied by on-site Coulomb excitations.

In this paper, we employed a Hamiltonian taking into account the magnetic interactions between the spins of Co electrons and ZnO conduction electrons and also the effective Coulomb interaction in the conduction band to simulate the magnetic properties of ZnO:Co. Consequently such model can simulate the magnetic properties very effectively at least in a qualitative manner. This investigation attempts to provide a guide for researchers to manipulate and design materials in the desired direction.

2. Theoretical description

Some theories were proposed to interpret the mechanism of ferromagnetism revealed in ZnO. One of the well-known models is the bound magnetic polaron model (BMP) [45, 46], which assumes that the ferromagnetism of ZnO is induced by a magnetic coupling between magnetic defects and bound carriers. In reality, no solid evidence has been demonstrated that the model is corresponding to the real mechanism of ferromagnetism in magnetic ion-doped ZnO. If the model would be right the magnetic properties should be not sensitive to the choice of magnetic ions in ZnO. Actually, not all magnetic ions can induce ferromagnetism in ZnO. Our theory is based on the fact that the ionized Co in ZnO couples strongly to the conduction band of ZnO, which results in the strong spin interaction between the Co spins and the spins of ZnO conduction electrons [47]. Obviously, the total Hamiltonian of ZnO:Co includes, at least, three main parts, $H = H_J + H_k + H_U$, i.e. the magnetic, kinetic and Coulomb interaction components, respectively.

The magnetic Hamiltonian can be written as

$$H_J = -\int \mathrm{d}x c_{\mathrm{co}} J(x)\ \vec{S}(x) \cdot \vec{\sigma}(x) \tag{1}$$

where $J(x)$ is the spin coupling between Co spin S and ZnO conduction electron spin σ which can be approximated to be a constant; x is the Co position index and c_{co} is the Co dopant concentration ratio. It should be reminded that the spin coupling can be anisotopic and in that case, the J can be decomposed to $J_{||}$ and $J_{\perp}$ for parallel and perpendicular directions relative to the applied magnetic field direction. The magnetic Hamiltonian of equation (1) can be written as

$$H_J = -\int \mathrm{d}x c_{\mathrm{co}} \{\frac{1}{2} J_{||}(x)\left(S^{+}(x)\sigma^{-}(x) + h.c\right) + J_{\perp}(x) S^{z}(x)\sigma^{z}\}. \tag{2}$$

The S^{+} and σ^{-} are spin raising and lowering operators corresponding to Co and ZnO conduction electrons, respectively. As ZnO is an intrinsic n-type semiconductor, the conduction band is only filled with some electrons and the valence band is completely filled. Moreover, there is no experimental evidence that Co dopant could introduce extra carriers to the conduction band of ZnO. For this reason, the kinetic Hamiltonian only needs to consider the ZnO conduction band:

$$H_k = \sum_s \int \mathrm{d}r(-\frac{\hbar^2}{2m}\nabla_{\mathrm{r}}^2)\psi^{+}(r)_{\mathrm{s}}\psi(r)_{\mathrm{s}}, \tag{3}$$

where $\psi(r)^{+}$ and $\psi(r)$ are field operators responsible for electrons creation and annihilation respectively; m and s are the electron mass and spin index.

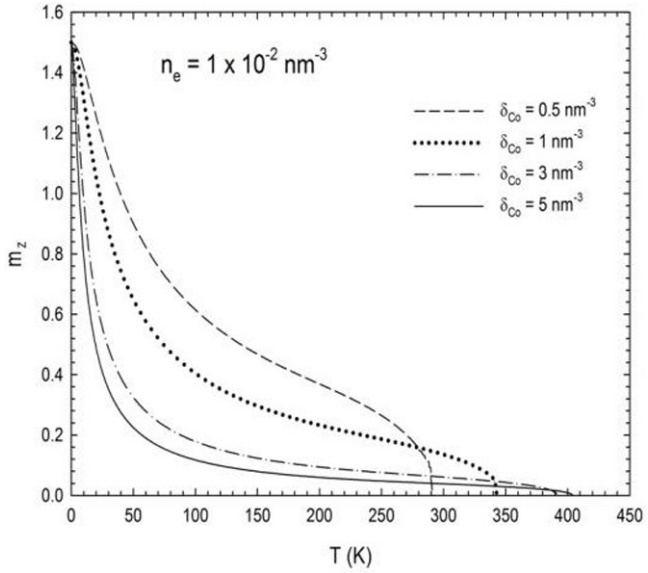

Figure 1. Temperature dependence of Co spin moment m_z calculated for various Co dopant densities δ_{Co} (called also atomic molar fraction). The electron density in the conduction band is $n_{\mathrm{e}} = 1 \times 10^{-2}\,\mathrm{nm}^{-3}$. The magnetic coupling is isotropic and both coupling constants are $J_{||} = J_{\perp} = 0.85\,\mathrm{eV/nm}^{-3}$.

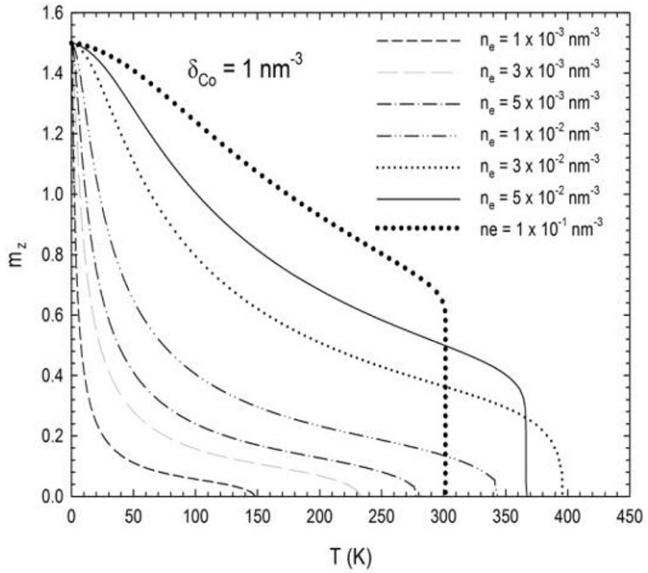

Figure 2. The temperature dependence of Co spin moment m_z calculated for various electron densities n_{e}. The Co dopant density $\delta_{\mathrm{Co}} = 1\,\mathrm{nm}^{-3}$. The magnetic coupling is isotropic and both couplings $J_{||} = J_{\perp} = 0.85\,\mathrm{eV/nm}^{-3}$.

As already discussed, the oxygen vacancies will effectively increase the Coulomb interactions among electrons in ZnO. The Coulomb interaction Hamiltonian can be represented as,

$$H_U = U \int \mathrm{d}x n(x)_{\uparrow} n(x)_{\downarrow} \tag{4}$$

where U is the effective Coulomb constant arising from electrons captured by the O vacancies; $n_{\uparrow}$ and $n_{\downarrow}$ are electron carrier densities in the conduction band with different spin indexes, $\uparrow$ and $\downarrow$.

The ferromagnetism of ZnO:Co arises mainly from the correlation between Co ions mediated by the ZnO itinerant electrons. The first step of the calculation is to utilize Green's function method to calculate the spin moment of Co ions. The Co spin time-dependent Green's function can be written as,

$$G_{xx'}(t) = -\mathrm{i}\theta(t) \left\langle \left[S_x^t(t), S_{x'}^-(0) \right] \right\rangle, \tag{5}$$

where t is the time, $\theta(t)$ is the step function and S_x^+ and $S_{x'}^-$ are spin raising and lowing operators on positions x and x' at time t and 0, respectively. By means of the equation of motion method and in mean-field approximation scenarios for evaluating of the magnetic moments and Coulomb interactions, after tedious derivations, the spin Green's function, $G(q,\omega)$, can be represented in the momentum q and frequency ω domains as:

$$\left(\omega - \frac{J_{||}^2}{2} c_{\mathrm{co}} \langle m_z \rangle \sum_p \frac{\left\langle c_{p\downarrow}^+ c_{p\downarrow} \right\rangle - \left\langle c_{p+q\uparrow}^+ c_{p+q\uparrow} \right\rangle}{\omega + \epsilon_{p+q} + U n_{\downarrow} - \epsilon_p - U n_{\uparrow} - J_{\perp} c_{\mathrm{ox}} \langle m_z \rangle} - J_{\perp} \langle \sigma_z \rangle \right) G(q,\omega) = \varphi, \tag{6}$$

where $\langle m_z \rangle$ and $\langle \sigma_z \rangle$ are the mean values of spin moments from Co ions and itinerant electrons, respectively. It should be noted that the fully polarized spin moment of a Co ion is $+3/2$. $\left\langle c_{p+q\uparrow}^+ c_{p+q\uparrow} \right\rangle$ and $\left\langle c_{p\downarrow}^+ c_{p\downarrow} \right\rangle$ are electron densities with momentum $\vec{p} + \vec{q}$ and $\vec{p}$ with spins $\uparrow$ and $\downarrow$, and their corresponding kinetic energies in the conduction band are ϵ_{p+q} and ϵ_p, respectively; $n_{\uparrow}$ and $n_{\downarrow}$ are the mean value of up and down electron densities and φ is a constant. It should be noted that in the calculations of both electron densities, the chemical potential obtained from the particle conservation and the Coulomb interaction is not considered here. The singularity poles of ω in the prefactor of $G(q,\omega)$ are the magnetic excitation energies called also a magnon dispersion, $\omega(q)$. From Callen's formula the spin magnetic moment $\langle m_z \rangle$ is a function of excitation number $\Phi(\omega(q))$.

$$\langle m_z \rangle = \frac{[S - \Phi][1 + \Phi]^{2S+1} + [S + 1 + \Phi][\Phi]^{2S+1}}{[1 + \Phi]^{2S+1} - \Phi^{2S+1}} \tag{7}$$

where

$$\Phi(\omega(q)) = \left(1 + e^{\beta\omega(q)} \right)^{-1} \tag{8}$$

In order to investigate the correlation between the Coulomb and spin interactions, we have considered the magnon

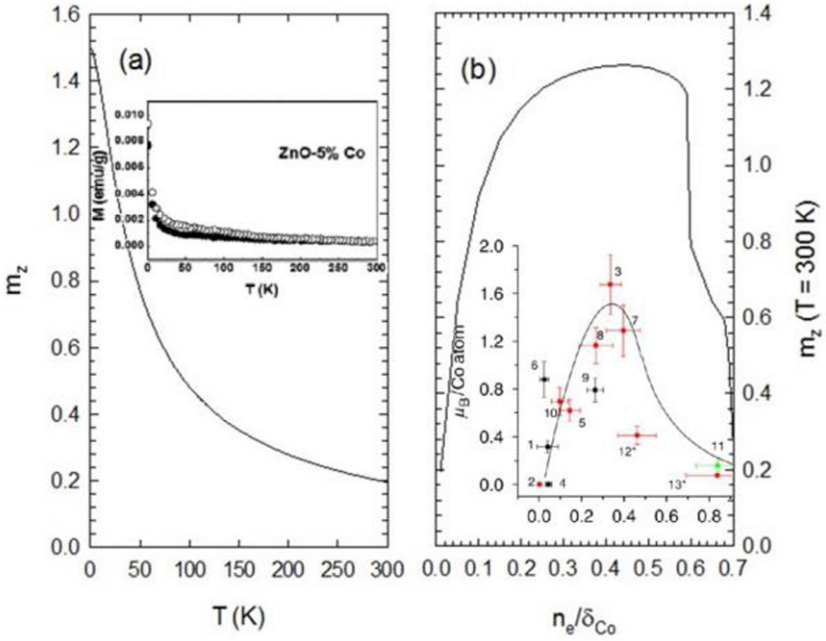

Figure 3. (a) The concave profile in the temperature-dependent magnetization for 5% Co ($\delta_{Co} = 3.6\,nm^{-3}$) with $n_e/\delta_{Co} = 1 \times 10^{-2}$. The inset figure displays the field and zero-field cool experimental results from the [52]. (b). The calculated result of the room temperature magnetization as a function of n_e/δ_{Co} for 5% Co. The inset figure is the experimental results from [53]. The magnetic coupling on both calculations are $J_{||} = J_{\perp} = 1\,eV/nm^{-3}$.

excitations (magnetic excitations of cobalt spins) under different magnitudes of Coulomb interactions.

3. Results and discussions

ZnO doped with a certain amount of Co exhibits evident ferromagnetism at room temperature [48, 49]. Moreover, it is expected that Curie temperature T_C is Co dopant density δ_{Co} dependent [50–52]. Evidently, our model can confirm this experimental fact, as shown in figure 1, whereas the increase of T_C is shown to be not linear with δ_{Co}. This is due to the fact that our ferromagnetic model is based on the carrier-mediated mechanism resulting in different profiles of the magnetization with different δ_{Co}. The increase in δ_{Co} increases initially T_C, but in the case of too high δ_{Co} densities the ferromagnetism is weakening, i.e. giving a more concave profile, which is due to an insufficient number of mediating carriers to correlate the Co spins. Besides, it can be predicted for a given ratio of n_e/δ_{Co} whether the ferromagnetic states would lower to a finite value or would completely break down.

However, too large electron density n_e will produce suppression of ferromagnetism (negative ferromagnetism), called also a magnetic screening effect, where too many electrons surround the Co ions leading to the suppression of magnetic interactions between Co ions. In particular, the negative ferromagnetism effect has been reported recently [30], where the magnetization as a function of temperature in field cool conditions decreases with increasing carrier concentration, even though it was interpreted by the BMP model. In order to investigate the electron concentration effect on the ferromagnetism we change n_e keeping a constant δ_{Co}.The results are shown in figure 2. T_C increases with increasing n_e for relatively small n_e values, i.e. in the large electron dilution range. Oppositely, for the large n_e range T_C decreases with n_e. This decreasing trend is due to the magnetic screening effect. Apparently, for a constant δ_{Co} there exists a maximum T_C at a specific n_e, which is consistent with the experimental results [53, 54].

At present, we realize that the magnitude of ferromagnetism of ZnO:Co is dependent on many parameters, which implicitly explains why there are so many controversies in different published papers. Based on our theoretical investigations, we can separate the influence of different magnetic contributions on particular parameters corresponding to experimental results, showing the ZnO carrier-concentration dependent ferromagnetism in Co-doped ZnO [55, 56], which implies that the ferromagnetism is carrier-mediated.

In order to confirm the possibility of this mode further, we compare our calculation results with some experimental results qualitatively in a plot. Figure 3 display the comparison from the results of figures 1 and 2. Evidently, the profile of the temperature-dependent magnetization from the experimental results shows significantly concave in figure 3(a). Obviously, the result is qualitatively consistent with our theoretical results as the density of the mediated carriers is relatively low. As the aforementioned results of figure 2, the magnetization in room temperature with a constant δ_{Co} increases with n_e and then decreases after reaching an optimal n_e. Apparently, figure 3(b) shows a consistent trend in comparison with the experimental results. It should be noted that 5% of Co dopants in the unit cell of ZnO are corresponding roughly to $\delta_{Co} = 3.6\,nm^{-3}$. We have to express that both comparisons can only be qualitative instead of quantitative because of the lack of information about the real interaction parameters.

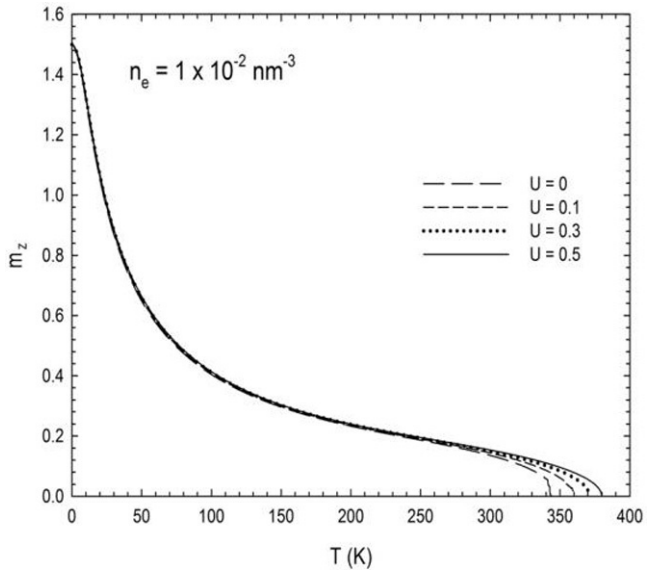

Figure 4. The temperature-dependence of Co spin moment m_z calculated for different Coulomb potential U and low electron density $n_e = 1 \times 10^{-2}\,\mathrm{nm}^{-3}$.

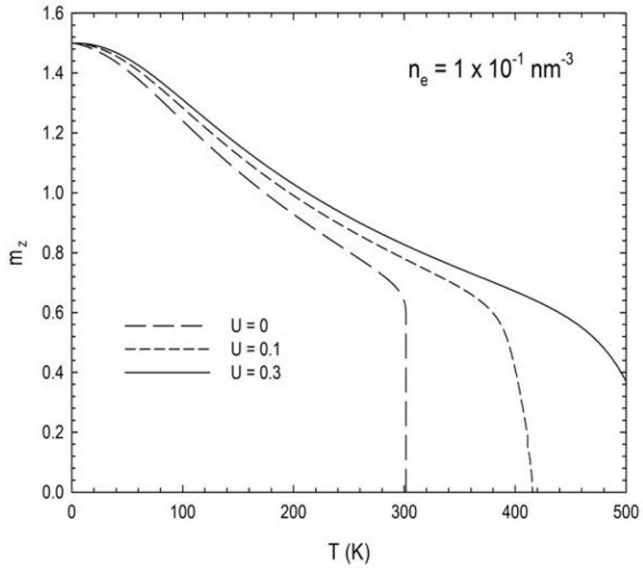

Figure 5. The temperature-dependence of Co spin moment m_z calculated for different Coulomb potential U and high electron density $n_e = 1 \times 10^{-1}\,\mathrm{nm}^{-3}$.

As we have proposed, the existence of the oxygen vacancies in ZnO:Co is an important parameter for the induction of ferromagnetism. Many experiments have revealed the fact that ZnO:Co films with appearing presence of ferromagnetism have to be annealed in vacuum. Briefly, there are two competing contributions from the O vacancies: one is the electron donation and the other is the electron capture. Apparently, both contributions fall into a balance. The former one is due to the fact that O is a high oxidation element which tends to absorb electrons to form atomic bonds. The latter one arises from the fact that O vacancies tend to capture electrons to stabilize the structure, which in turn results in the Coulomb excitation of the captured electrons. In the mean-field approximation, this

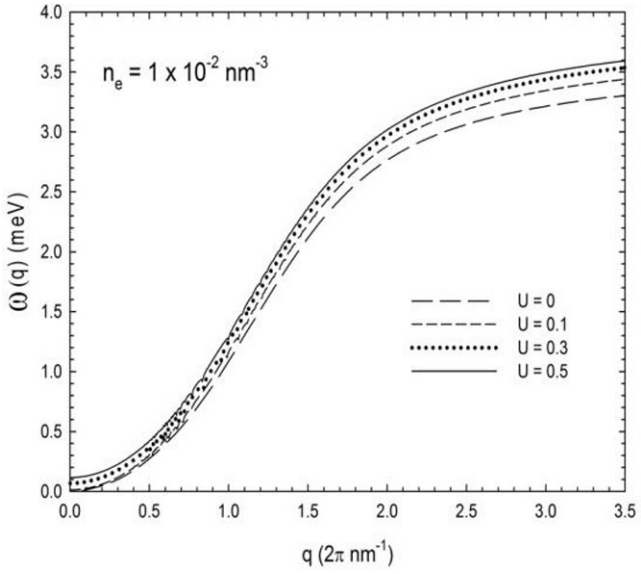

Figure 6. The magnon dispersion calculated for different Coulomb potential U for low electron density $n_e = 1 \times 10^{-2}\,\mathrm{nm}^{-3}$.

Coulomb excitations occurring on the O vacancies can be seen as an effective Coulomb interaction applying to the conduction electrons described in the equation (4). Here, we have simulated the Coulomb potential U affecting ferromagnetism for both, low and high electron densities, i.e. $n_e = 1 \times 10^{-2}\,\mathrm{nm}^{-3}$ and $n_e = 1 \times 10^{-1}\,\mathrm{nm}^{-3}$, corresponding results are shown in figures 4 and 5, respectively. Obviously, in both cases T_C increases with U, whereas the impact of U is more effective in the case of high electron densities. Therefore, we propose that O vacancies enhance the ferromagnetism due to simultaneous contribution of n_e and U.

In order to take into account the coupling between the Coulomb and spin excitations, we have calculated the magnon excitation for different values of U. Similarly, we have investigated it for low and high n_e cases. Both results are demonstrated in figures 6 and 7, respectively. It should be noted when the area of m_z versus q dispersion diagram is larger, ferromagnetism is stronger. From the magnon dispersion calculations, we find that Coulomb potential U can improve the ferromagnetism. In particular, for a certain value of U the bandgap of magnon dispersion appears, namely, $\omega(q)$ is finite at $q = 0$. Clearly, the bandgap increases with U and this arises from the Coulomb excitations.

In fact, the O vacancies could be seen as defects in ZnO and these defects as it was aforementioned could capture electrons, and they correspond to the defect centers in the BMP model [57] inducing the ferromagnetism. In fact, our proposed carrier-mediated model is also a magnetic polaron model but the polarons are not bound and the proposed model includes the Coulomb excitations between captured electrons, which explains well the magnetic properties of Co-doped ZnO. It should be noticed that in our model the Coulomb excitation is in a collective mode and is a key factor for the appearance of ferromagnetism because it promotes the carrier-mediated interactions. On the other hand, the Coulomb interaction is not included in the BMP model, which seems insufficient to

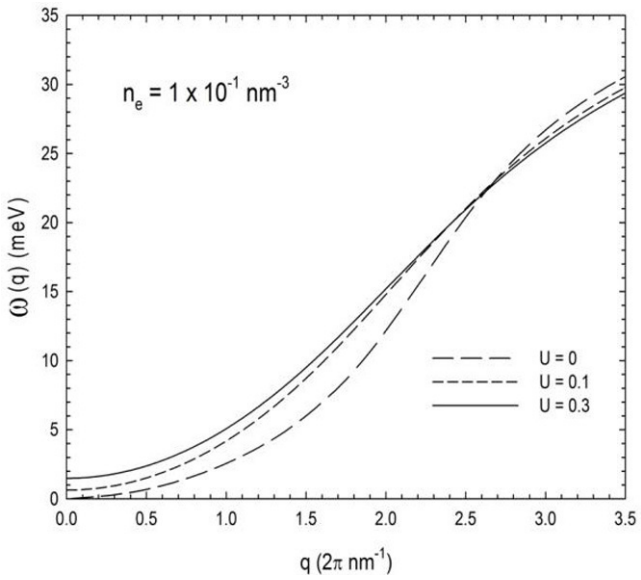

Figure 7. The magnon spectrum calculated for different Coulomb potential U and for high electron density $n_e = 1 \times 10^{-1}\,\mathrm{nm}^{-3}$.

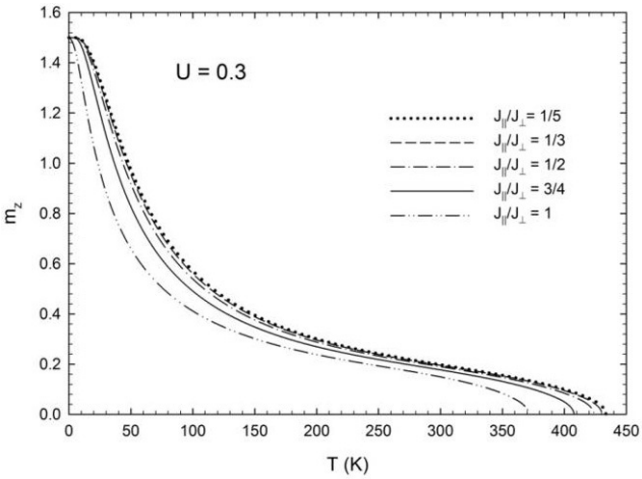

Figure 8. The temperature-dependence of Co spin moment m_z calculated for different anisotropic magnetic couplings. The electron density is $n_e = 1 \times 10^{-2}\,\mathrm{nm}^{-3}$ and Coulomb potential $U = 0.3$.

picture the real situation. For instance, in the BMP model, the ferromagnetism appearance is explained by the polaron orbits overlap. However, taking into account the Coulomb interaction in the BMP model will produce a counter effect—the Coulomb interaction will also increase with the increase of the orbitals overlap what should result in suppressing the orbitals overlap and as a consequence also the ferromagnetism. Although, the drawback of the carrier-mediated model is the ferromagnetism reported in the insulating Co-doped ZnO where there is not enough number of itinerant carriers to induce the ferromagnetism [46]. However, consideration of the Coulomb excitation is solving this problem as the captured electrons will compensate the carrier insufficiency. It gives a contribution equivalent to high energy virtual states in the conduction band enabling the electrons to occupy those states and leading to the carrier-mediated induced ferromagnetism reported experimentally.

In ZnO:Co thin films, the Co-ions couple to the conduction band electrons. Based on our discussion above, the Coulomb excitations should also affect spin interactions. Since the collective spin of Co is determined predominantly by electrons from d orbitals, we suppose that the Coulomb excitations will also excite those Co electrons. On the other hand, the Co orbitals are responsible for the spin interactions of the conduction electrons, which leads to the anisotropy of the magnetic coupling between Co electron spins and ZnO electron spins. Figure 8 shows the temperature-dependence of magnetization for different magnetic anisotropies. As the structure of ZnO:Co is hexagonal the preferred magnetic easy axis is along the c axis and it can be expected that $J_{||} < J_{\perp}$. Obviously, the maximum T_C occurs at $J_{||} = 0$ because $J_{||}$ produces magnetic fluctuations which will suppress the ferromagnetism.

4. Conclusions

We propose here the carrier-mediated model modification by taking into account the Coulomb excitations enabling to explain the magnetic properties of n-type ZnO:Co. This model interprets the role of cobalt ions and oxygen vacancies on the appearance of ferromagnetism consistently and it is in accordance with experimental results. Coulomb excitations coupling with the spin interactions are predicted to exist in oxygen-vacancies of ZnO:Co, which are enhancing the ferromagnetism as well as opening a bandgap of magnon spectrum. In addition, in the presented model the Coulomb excitations induce the magnetic anisotropy which results in ferromagnetic enhancement. We suppose that the BMP model without considering the Coulomb excitations is insufficient to describe correctly the magnetic properties of ZnO.

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