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NEGATIVE MAGNETORESISTANCE IN MANGANESE-DOPED ZINC OXIDE

Thin films of zinc oxide have a unique combination of piezoelectric, conductivity, and optical properties. Zinc oxide (ZnO) is a direct, wide bandgap semiconductor material with many promising properties for blue/UV optoelectronics, transparent electronics, sensor applications and especially for spintronic devices.

Thesp-d(f) exchange interaction between conduction carriers and localized spins in diluted magneticsemiconductors (DMS) gives rise to anomalous optical, magnetic and transport properties. This interactionis at the origin of the giant Faraday and Zeeman effects, were observed in wide band-gap magnetic semiconductors likediluted ZnO [1-3].

Several attempts have been unsuccessfully made to model the effects by using standard theory of spin-disorder scattering, possibly fromboundmagnetic polarons. But the negativemagnetoresistance is always observed for appliedmagnetic fields wellabove the saturation magnetization. Moreover, the authors assume that all the donors are activated in theconduction band no matter the temperature, therefore excluding additional formation or annihilation of boundmagnetic polarons. Surprisingly the possibility of the existence of an hopping conduction channel was not considered yet.

We here report our study on the negative magnetoresistance in Mn:ZnO thin films. As the temperature isreduced, the resistivity of the films increases with distinct signatures of a transition from band- to hopping-conduction. A sharp decrease of resistance was measured when an external magnetic field was applied. Thechange of resistivity was found to increase with the concentration of Mn. By using magneto-photoluminescence measurements, we demonstrate that the external magnetic field

reactivates the carriers in theconduction band, with a consequent sharp reduction of the filmresistivity. In analogy with the case of 4f semiconductors, we callthe effectgiantnegative magnetoresistance.

The study was carried out on 300 nm thick films of $Zn_{1-x}Mn_xO$ withx=50 (pure ZnO), 0.02, 0.04 and 0.08 grown by pulsed laserdeposition (PLD) on sapphire substrates. The films shown-typeconductivity, regardless temperature. The concentration of oxygenvacancies ($V_{O's}$) in the films was tuned by changing the temperatureof the substrate and the oxygen partial pressure during growth. It isrelevant here to note that in films with x=0.08 we have carefullyexcluded the presence of Mn in chemical valence different from by resorting to low temperature x-ray absorption spectroscopy.Therefore we can reliably exclude that magnetism is due to double-exchange interaction in our films. In addition, photoluminescence (PL) measurements have excluded the presence of Znvacancies, which are sources of magnetic moment. The films showed a coercivity of 20 mT and a saturation field of 200 mTat low temperatures.

A large decrease of resistivity is observed when $Eu_{1-x}Gd_xSe$ is cooled through its Curie temperature [4]. This anomalous phenomenon has beenwell studied and explained[5]. Gd substitutes $Eu^{2+}$with valence 3+ and the excess electrons form an impurity band thatoverlaps with the conduction band of EuSe. At room temperaturethe impurity electrons are thermally excited in the conduction band.As the temperature is reduced, the electrons localize on the Gd ionsand a hopping conduction channel appears. In an occupied site, theimpurityselectron will mediate exchange interaction between the 4felectrons of the Gd^{3+}, as well as the 4f electrons of the surroundingEu^{2+}. As the impurity electron gains the energy of the s-fexchangeinteraction, its activation energy increases. Hopping requires largeactivation energy because the 4f spins in the unoccupied sites arerandomly oriented. Yet, if magnetic order appears, or an externalmagnetic field is applied that aligns the 4fspins, the activation energysharply decreases and so does the resistivity.

Mn is isovalent to Zn in thewurtziteZnOcrystal and, unlikeGd in EuSe, is not a dopant. The carrier spins that mediate exchange interaction are provided by the $V_{O's}$.

Exchange interaction is establishedwhen apelectron from a $V_O$delocalizes on a Mn^{2+}energy level andmediates exchange interactionbetweendelectrons of theMn^{2+}sites.More properly one should think of this process as a delocalization ona$Mn\text{-}V_O$complex, rather than on aMn ion. The chemical valence of the Mn will remain 2+. The magnetic alignment is not due to double-exchange interaction between Mn in mixed valence but formation of $Mn\text{-}V_O\text{-}Mn$polarons. As the temperature is reduced,impurity electrons from $V_{O's}$ start freezing out and a hopping channel is established. Hopping occurs between $Mn\text{-}V_O$complexes thatare randomly oriented, therefore belonging to different polarons. Amagnetic field that tends to align$Mn\text{-}V_O$complexes results in a sharpreduction of the resistivity. Unlike the case of $Eu_{1-x}Gd_xSe$, the effectis not limited to the temperature range near the Curie temperature because of the fundamentally differentmechanismof magnetic interaction in the twomaterials. Mn:ZnOdoes not have a sharpmagnetictransition, if amagnetic transition can be defined at all. The molecular field plays a marginal role as compared to the externally appliedfield in the case ofMn:ZnOand the giant negativemagnetoresistanceexists as long as an hopping channel can be created. The peculiarform of magnetism in Mn:ZnO that has confined this material to more academic curiosity might represent its uniqueness and fortunewhen it comes tomagnetotransport properties, with potential applications in magnetic sensing.Due to the strongsp-d exchange interaction, and thereforegiant Zeeman splitting, delocalization occurs even for field of the order of hundreds of mT. Theactivation of these electrons, whether due to the temperature or themagnetic field results in agiantreduction of resistivity.

We report a large negative magnetoresistance in Manganese-substituted Zinc Oxide thin films. This anomalous effect was found to appear in oxygen-deficient films and to increase with the concentration of Manganese. By combining magnetoresistive measurements with magneto-photoluminescence, we demonstrate that the effect can be explained as the result of amagnetically induced transition from hopping to band conduction where the activation energy is caused by the sp-d exchange interaction.

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