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Scattering mechanism of nonmagnetic phase on nano diluted magnetic semiconductors (DMS)

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Abstract: This paper shows the scattering mechanism at diluted magnetic semiconductors. The doped magnetic atom produces a scattering potential due to be coupled of itinerant carrier spin of host material with magnetic momentum of the doped magnetic atom. Formulas of scattering event were rewritten by the plane wave expansion and then the electron mobility of DMS was calculated. Calculations show Kondo effect on diluted magnetic semiconductors at nonmagnetic phase. Here has been supposed that the doping concentration is low and so the coupling coefficient between magnetic atoms is weak enough that DMS does not change its magnetic phase. In other words, material is on paramagnetic phase. For proofing our model, we have grown $Zn_{0.99}Mn_{0.01}O$ with Sol-Gel route. Pure ZnO has also grown with this method for a comparison. Experimental results proved our theoretical model. Therefore as a result, at diluted magnetic semiconductors similar to diluted magnetic metals in nonmagnetic phase can observe kondod's effect .

Key words: diluted magnetic semiconductor, Kondo effect, scattering event, relation time, electrical resistivity

1. INTRODUCTION

In physics, the Kondo effect describes the scattering of conduction electrons in a material due to magnetic impurities. The electrical resistivity (electron/hole) of a material is increased by decreasing temperature. Each physical quantity has a macroscopic description and a microscopic description. The macroscopic physical quantities are measureable at laboratory but microscopic physical quantities are described by mathematical relation. It has been known since 1930 that the resistivity has a shallow minimum at low temperature but there was not a theory for description it [1]. At 1963, the microscopic description of this

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minimum was shown by Jun Kondo [2,3]. The resistivity has a direct relation with scattering. The higher the resistivity is, the more the scattering will be. The factors which scatter all carriers (electron/hole) are extrinsic such as ionized impurities [4], neutral impurities [5], point defect [6], missing ions [7] or intrinsic such as lattice vibrations (acoustic and optical phonon) [8] and piezoelectric [9]. Nowadays, scattering mechanisms are quite well understood and measured in materials.

The mobility $\left(e\langle\tau\rangle/m^*\right)$ involves two physical parameters, the relaxation time and the effective mass which depend on the type of the material. The effective mass is essentially a property of the pure material, but the average relaxation time is strongly affected by scattering potential [10]. Semiconductors are intrinsically nonmagnetic materials but diluted magnetic semiconductors (DMSs) may be magnetic materials. DMSs have both magnetic property and semiconductor property. When a magnetic force is produced by alloyed magnetic atom at semiconductor, it interacts on the conduction carrier and changes the distribution function and consequently a scattering event is created. This scattering changes the resistance of host material. At this paper, we are investigating this effect and the form of these changes [11].

2. SCATTERING THEORY

At first, it is considered that a carrier interacts with an atom. The initial (ground) state of carrier (atom) is $|n\mathbf{k}\rangle$ ($|N\mathbf{K}\rangle$). The final (excited) state of carrier (atom) is $|n'\mathbf{k}'\rangle$ ($|N'\mathbf{K}'\rangle$). In this case, the interaction is an inelastic scattering because the kinetic energy of the final outgoing carrier is now less than that of the initial incoming carrier. The difference is used to excite the target atom. The interaction will be the elastic scattering if the target atom is not excited. Here, we consider the elastic scattering because the atom is much weightier than carrier. In a scattering event a current carrier makes a transition from the Bloch state $|n\mathbf{k}\rangle$ to the other Bloch state $|n'\mathbf{k}'\rangle$ and for intra-band scattering, $n=n'$. If we neglect multiple scattering, the Born approximation is used and also the scattering rate $W(\mathbf{k},\mathbf{k}')$ is quantum mechanically determined from Fermi's golden rule.

$$W(\mathbf{k},\mathbf{k}')=\frac{2\pi}{\hbar}\left|\langle\mathbf{k}'|H_{\text{int}}(\mathbf{r})|\mathbf{k}\rangle\right|^2\delta(E_{k'}-E_k) \tag{1}$$

H_{int} is scattering potential, δ-function expresses that the scattering is elastic. The matrix element of H_{int} with using Bloch's theorem that state $|\mathbf{k}\rangle$ is as the product of a plane wave function and the periodic function $u_{\mathbf{k}}(\mathbf{r})$ becomes

$$\langle \mathbf{k}'|H_{\text{int}}(\mathbf{r})|\mathbf{k}\rangle = \int e^{(\mathbf{k}-\mathbf{k}')\mathbf{r}} u^*_{k'}(\mathbf{r}) u_k(\mathbf{r}) H_{\text{int}}(\mathbf{r}) d^3r \tag{2}$$

$u_k(\mathbf{r})$ and $u_{k'}(\mathbf{r})$ are the periodic function then $u^*_{k'}(\mathbf{r})u_k(\mathbf{r})$ has the periodicity of the crystal and can be expanded in the Fourier series. Here, $\mathbf{G}$ is a reciprocal lattice vector in crystal. For $\mathbf{q} = \mathbf{k} + \mathbf{G} - \mathbf{k}'$, $e^{\mathbf{q}\cdot\mathbf{r}}$ can also be written as

$$e^{i\mathbf{q}\cdot\mathbf{r}} = \sum_{i=0}^{\infty} i^l (2l+1) j_l(qr) P_l(\hat{\mathbf{q}}\cdot\hat{\mathbf{r}}) \tag{3}$$

If scattering potential is a weak interaction potential with varying slowly, we may neglect all small argument except $G = 0$. The matrix element of $H_{\text{int}}(\mathbf{r})$ simplifies to

$$\langle \mathbf{k}'|H_{\text{int}}(\mathbf{r})|\mathbf{k}\rangle = \frac{1}{V}\sum_{i=0}^{\infty} i^l (2l+1) \int j_l(qr) P_l(\hat{\mathbf{q}}\cdot\hat{\mathbf{r}}) H_{\text{int}}(\mathbf{r}) d^3r \tag{4}$$

3. EXCHANGE SCATTERING

At each temperature, carriers in semiconductors are scattered by different potentials but at diluted magnetic semiconductors are added a new scattering event. one of these is named exchange interaction between the localized moments and itinerant carriers both paramagnetic phase and ferromagnetic phase. Many features of DMS, such as the special electronic properties, unique phase diagrams, and important magnetic and magneto-optical characteristics, are induced by the exchange interaction between the localized *d* shell electrons of the magnetic ions and the delocalized band states (of *s* or *p* origin). The s-d, p-d exchange, and its consequences and origin have been pointed out from the very beginning of the history of DMS and the Heisenberg form of the exchange interaction Hamiltonian was successfully used for this interaction.

$$H_{\text{int}} = \sum_i J(\mathbf{r} - \mathbf{R}_i)\, \mathbf{s}\cdot\mathbf{I}_{\text{i}} \tag{5}$$

Where, r and R are the position vectors of the band electron and the ith magnetic ion, respectively. $J(\mathbf{r}\text{-}\mathbf{R})$ is the exchange coupling coefficient of the band electron with the 3*d* electrons in the i$^{\text{th}}$ magnetic ion. $\mathbf{I}_{\text{i}}$ is the total angular momentum of the 3*d* electrons in the i$^{\text{th}}$ magnetic ion. If doping concentration of

magnetic atom in DMSs is low that their wave function cannot overlap one another, we will neglect the interaction of other magnetic atoms. With neglecting the interaction of the far magnetic ions can write:

$$H_{\text{int}} = J(\mathbf{r}-\mathbf{R})\,\mathbf{s}\cdot\mathbf{I} \tag{6}$$

The stable configuration of the magnetic atom at DMS is considerate to the form ${}^{2I+1}L_J$. Suppose the state of Mn ions in DMS material is Mn^{2+}. The electronic structure of Mn^{2+} is $1s^2\ 2s^2\ 2p^6\ 3s^2\ 3p^6\ 3d^5$, in which $3d^5$ is a half-filled shell. According to Hund's rule, the spin of these five $3d^5$ electrons will be parallel to each other, so the total spin is I = 5/2. These five electrons are in states in which the orbital angular momentum quantum number l = 0; ± 1; ± 2. Thus the total orbital angular momentum L = 0. The total angular momentum for a Mn^{2+} ion then is J = S = 5/2. The stable configuration is ${}^6S_{5/2}$. The Lande g-factor is

$$g = 1+\frac{J(J+1)+S(S+1)+L(L+1)}{2J(J+1)} = 2 \tag{7}$$

$$\begin{aligned}\mathbf{s}\cdot\mathbf{I} &= s_z I_z + \frac{1}{2}\left(s^+ I^- + s^- I^+\right) = \\ &= \frac{\hbar^2}{2}\left(F(F+1)-s(s+1)-I(I+1)\right)\end{aligned} \tag{8}$$

$\mathbf{F}=\mathbf{s}+\mathbf{I}$ is the total spin. For example, $s=\frac{1}{2}$ and $I=\frac{5}{2}$, the total spin are 3 (seven degenerated states) and 2 (five degenerated states). For F=3, this is $\frac{5}{4}\hbar^2$ and for F=2, it equals $-\frac{7}{4}\hbar^2$. Matrix element for intra band scattering is

$$\begin{aligned}&\langle \mathbf{k}'\mathbf{s}',\mathbf{I}'|H_{\text{int}}|\mathbf{k}\mathbf{s},\mathbf{I}\rangle = \\ &\frac{1}{V}\sum_{l=0}^{\infty} i^l\sqrt{4\pi(2l+1)}\,j_l(qr)Y_{lo}^*(\theta,\varphi)\langle s'|H_{\text{int}}(\mathbf{r})|s\rangle d\mathbf{r} \\ &\frac{\langle s',I'|\mathbf{s}\cdot\mathbf{I}|s,I\rangle}{V}\sum_{l=0}^{\infty} i^l\sqrt{4\pi(2l+1)}\,j_l(qr)Y_{lo}^*(\theta,\varphi)J(\mathbf{r}-\mathbf{R})d\mathbf{r}\end{aligned} \tag{9}$$

$A=\langle \mathbf{s}',\mathbf{I}'|\mathbf{s}\cdot\mathbf{I}|\mathbf{s},\mathbf{I}\rangle$ based on transition rule $(\nabla S=0)$ is $\frac{5}{4}\hbar^2$ or $-\frac{7}{4}\hbar^2$. Consider origin on the magnetic atom (**R**=0) and $J(\mathbf{r})=f(r)Y_{l'm}$:

$$\langle \mathbf{k}'\mathbf{s}', \mathbf{I}' | H_{\text{int}}(\mathbf{r}) | \mathbf{k}\mathbf{s}, \mathbf{I} \rangle =$$
$$\frac{A}{V}\sum_{l=0}^{\infty} i^l \sqrt{4\pi(2l+1)} \int_0^{\infty} j_l(qr) f(r) r^2 dr \int_{-1}^{1}\int_0^{2\pi} Y_{lo}^*(\theta,\varphi) Y_{l'm'}(\theta,\varphi) d\Omega \tag{10}$$
$$= \frac{A}{V} i^l \sqrt{4\pi(2l+1)} \int_0^{\infty} j_l(qr) f(r) r^2 dr$$

This relation say that the coupled constant of exchange interaction cannot be a function of φ . This is a physical inherent phenomenon so

$$J(\mathbf{r}) = f(r) Y_{l0}(\theta,\varphi) = \sqrt{\frac{2l+1}{4\pi}} f(r) P_l(\cos(\theta)) \tag{11}$$

In the following, matrix element was calculated to the different forms for evaluating resistance.

For $f(r) = j_{l'}(kr)/(kr)^2$

$$\langle \mathbf{k}'\mathbf{s}', \mathbf{I}' | H_{\text{int}}(\mathbf{r}) | \mathbf{k}\mathbf{s}, \mathbf{I} \rangle =$$
$$= \frac{A}{V} i^l \sqrt{4\pi(2l+1)} \int_0^{\infty} j_l(qr) \frac{j_{l'}(kr)}{(kr)^2} r^2 dr$$
$$= \frac{A}{Vk^2} i^l \sqrt{4\pi(2l+1)} \int_0^{\infty} j_l(qr) j_{l'}(kr) dr \tag{12}$$
$$= \frac{A}{Vk^2} i^l \left(\frac{\pi^3}{2l+1}\right)^{1/2} \frac{(k_<)^l}{(k_>)^{l+1}}$$

The scattering rate between state *k* an state *k*′ (Fermi's golden rule) can rewrite to form

$$W(k,k') = N\frac{2\pi}{\hbar} \left| \langle \mathbf{k}'\mathbf{s}', \mathbf{I}' | H_{\text{int}}(\mathbf{r}) | \mathbf{k}\mathbf{s}, \mathbf{I} \rangle \right|^2 \delta(E_{k'} - E_k)$$
$$= \frac{A^2 N}{V^2 k^4 \hbar} \left(\frac{2\pi^4}{2l+1}\right) \frac{(k_<)^{2l}}{(k_>)^{2(l+1)}} \delta(E_{k'} - E_k) \tag{13}$$

N is the number of magnetic impurities. The inverse relaxation time is

$$\frac{1}{\tau(E_k)} = \text{the sum over all stases at the phase space}$$
$$= \left[\int W(k,k')(1-\cos\theta_{kk'}) \frac{d\mathbf{r}d\mathbf{k}'}{(2\pi)^3} \right] \times (\text{spin states}) \tag{14}$$

Based on Pauli's exclusion principle, each state $|k\rangle$ has two degenerate states with up and down spin so spin states are 2. It means that the spin of carrier after a scattering event can change from up to up or up to down. If there is nondegenerate, the spin of carrier doesn't change and the number of states will be one. Equation (14) is multiplied with $z=\{1 \text{ or } 2\}$. So:

$$\begin{aligned}\frac{1}{\tau(E_k)} &= \frac{zV}{(2\pi)^3}\int W(k,k')(1-\cos\theta_{kk'})d\mathbf{k}' \\ &= \frac{zV}{(2\pi)^4}\frac{\pi^2\hbar}{(2m)^{1/2}}\int_0^{\infty}\int_0^{2k} W(k,k')\frac{q^3}{E'^{3/2}}dE'dq\end{aligned} \tag{15}$$

Here, $E'=\frac{\hbar^2}{2m}k'^2$ and $q=|\mathbf{k}-\mathbf{k}'|$ and $\frac{N_I}{V}=n_I$ as the concentration of the magnetic atom so:

$$\begin{aligned}\frac{1}{\tau(E_k)} &= A^2 n_I\left(\frac{\pi^5}{2l+1}\right)\left(\frac{\hbar^4}{(2m)^{5/2}}\right)\int_0^{\infty}\frac{\delta(E_{k'}-E_k)}{E'^{7/2}}dE'\int_0^{2k}\frac{(k_<)^{2l}}{(k_>)^{2(l+1)}}q^3dq \\ &= A^2 n_I\left(\frac{\pi^5}{2l+1}\right)\left(\frac{\hbar^4}{(2m)^{5/2}}\right)\int_0^{\infty}\frac{\delta(E_{k'}-E_k)}{E'^{7/2}}dE'\left[\int_0^{k}\frac{(q)^{2l}}{(k)^{2(l+1)}}q^3dq+\int_k^{2k}\frac{(k)^{2l}}{(q)^{2(l+1)}}q^3dq\right]\end{aligned} \tag{16}$$

Because of the fact that the relaxation time is positive $\left(\tau(E_k)\rangle 0\right)$, the angular momentum l, in equation (16), can only take the values zero or one. So $\tau(E_k)$ is obtained as

$$\tau(E)=\frac{1}{xA^2n_I}\left(\frac{2m^{3/2}}{\hbar^2\pi^5}\right)E^{5/2} \tag{17}$$

Here x is 7/4 ((1+6ln2)/6) for l=0 (1). Average value $\tau(E)$ is:

$$\langle\tau(E)\rangle=\frac{\int\tau(E)e^{-\beta E}E^{3/2}dE}{\int e^{-\beta E}E^{3/2}dE}=\frac{64}{xA^2n_I}\left(\frac{m^{3/2}}{\hbar^2\pi^{11/2}}\right)(k_BT)^{5/2} \tag{18}$$

The mobility associated with magnetic potential is obtained by multiplying Eq. (17) by e/m*. The resistivity is proportional to $1/\langle\tau\rangle$. As seen, the temperature dependence of the resistivity is contained in the factor $(k_BT)^{-5/2}$. The increase in the resistivity with decreasing temperature is associated with the Konto effect.

$$\rho = 1/qn\mu_n = = \left[\frac{64ne^2}{xA^2n_I}\left(\frac{m^{1/2}}{\hbar^2\pi^{11/2}}\right)\right]^{-1}(k_BT)^{-5/2} \quad (19)$$

Being lessened the relation of the mobility with being diminished temperature depends on the coupling constant. The more the mobility is, the weaker the coupling constant will be. The coupling constant has a complex form and changes at different materials (metals or semiconductors). Therefore we cannot write temperature dependence for mobility due to magnetic scattering similar to phonon $(T^{-3/2})$ or impurity $(T^{3/2})$ scattering. In table 1, we have summarized a physically selected form of the scattering potentials and calculated their electron mobility.

TABLE 1

the exchange coupling coefficient, matrix element and electron mobility for various scattering

$J(r,\theta,\varphi)$	Matrix element	Electron mobility
$\frac{1}{r^2}$	$\frac{2\pi^2}{\Omega}\frac{A}{q}$	$\frac{8e\hbar^2}{3\pi^{7/2}(2m)^{3/2}n_IA}(k_BT)^{1/2}$
$e^{-\alpha r}$	$\frac{8\alpha}{\pi\Omega}\frac{A}{(\alpha^2+q^2)^2}$	$\left(\frac{2m}{\pi}\right)^{\frac{1}{2}}\frac{12a^2}{\pi An_I}\frac{1}{\left(1-\frac{1+3T/T_o}{1+T/T_o}\right)}(K_B)^{3/2}$, $T_o=\frac{\hbar^2\alpha^2}{24mK_B}$
$j_{l'}(kr)Y_{l'm}$	$\frac{\pi A}{2V}i^l\sqrt{4\pi(2l+1)}\frac{\delta(q-k)}{q^2}$	$\frac{140e}{A^2\pi^2\hbar n_I(2l+1)}(k_BT)^2$

3. EXPERIMENTAL REPRESENTATION

For experimental representation of our model, we have grown $Zn_{0.99}Mn_{0.01}O$ with the Sol-Gel Technique. Pure ZnO was also been grown for comparison by this method. Growth conditions and primary materials are the same with paper of reference [11]. Hysteresis loops and magnetic properties of samples were recorded by using superconducting quantum interference device (SQUID). Carrier concentration and mobility were measured by standard resistivity and Hall effect set-ups in the Van der Pauw configuration. Fig. 1 illustrates magnetic properties of samples at temperature 100 K. As seen, ZnO and

$Zn_{0.99}Mn_{0.01}O$ have paramagnetic property. Electronic stability configuration of isolated atom Mn in orbital shell d^{10} is $\left({}^{6}S_{5/2}\right)$ but for Zn (d^{10}) is $\left({}^{1}S_{0}\right)$. Ion Mn^{2+} has magnetic property. The half-filled 3d-shell of Mn^{2+} ions, which have the largest ionic moment ($5\mu_B$), plays a particular role in both theoretical and experimental studies. As seen, susceptibility of ZnO is smaller than that of $Zn_{0.99}Mn_{0.01}O$

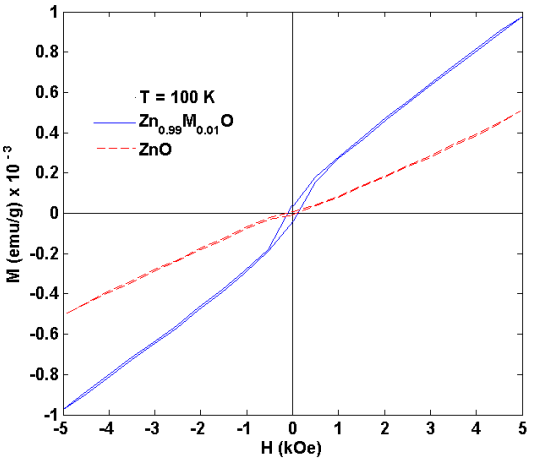

Fig. 1. Magnetization loops of ZnO and $Zn_{0.99}Mn_{0.01}O$ at temperature 100 K

To illustrating effect of magnetic scattering potential on carriers ZnO, mobility of two samples is drawn at fig.2 has been drawn. This figure shows that firstly, mobility of pure ZnO is more than that of Zn0.99Mn0.01O because the isolated atom Mn is a magnetic atom and consequently scattering potential due to electron coupling of orbital s and p of host material ZnO by orbital d of alloyed impurity Mn will be stronger. Secondly, mobility is decreased by decreasing temperature because at low temperature, magnetic coupling potential is stronger than other factors such as phonon and ionized impurity. Then, each atom Mn or Zn produced two scattering potentials; one is ionized impurity potential Mn^{2+} or Zn^{2+} and another is sp–d exchange interaction potential between the band electrons of host material and the localized d electrons of the ion Mn^{2+} or Zn^{2+}.

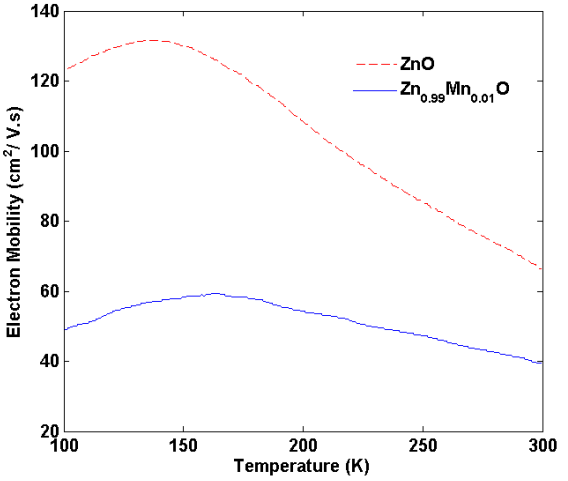

Fig.2. electron mobility for pure ZnO and $Zn_{0.99}Mn_{0.01}O$

The mobility of samples can divide to high temperature and low temperature. Phonon is main factor for decreasing the mobility at high temperature. There are two factors at low temperature that decrease the mobility. One is ionized impurity scattering and the other is exchange scattering. At ionized impurity scattering of atoms Mn^{2+} and Zn^{2+} that are isovalent, the low concentration of atom Mn must not change not only path of decreasing or maximum point but also amount of mobility very much. As a result, we can say that there is the other scattering factor at ZnO and $Zn_{0.99}Mn_{0.01}O$ in the name of exchange scattering. This test proved our theoretical model. Here, this is noticeable that samples have not magnetic property at temperature above 100 K. If samples have magnetic property, spin relaxation happens [12,13, 14,15].

4. CONCLUSION

Pressure, volume and temperature are thermodynamic parameters that change the stability of materials. It is expectable that with being decreased temperature, electrical resistance is decreased. Electrical resistance is the same the scattering of carriers. Parameters which decrease the scattering events decrease electrical resistance. Main factor at DMS's for low temperature is interaction between the spin of carrier with the spin of the doped magnetic atom. The more the exchange coupling coefficient is, the stronger the scattering will be. The exchange interaction happens between localized moments of the magnetic atom and itinerant carriers that can also cause to spin flip. Here, Kondo effect on diluted magnetic semiconductors has been investigated. It is seen that if the doping concentration of the magnetic is low so that the exchange coupling coefficient will not overlap and DMS is nonmagnetic phase. At this case, carriers were scatter and the electrical resistivity of material goes up the same as

what happens at metals. For proving our model, we have grown ZnO and $Zn_{0.99}Mn_{0.01}O$ by sol-gel method. At temperature above 100 K, there is not hysteresis loop and materials have nonmagnetic property. The mobility of samples shows that there is another scattering potential in the name of the magnetic potential due to itinerant carrier spin with the doped magnetic atom. Therefore, as a result, at diluted magnetic semiconductors similar to diluted magnetic metals in nonmagnetic phase can observe kondod's effect.

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