Anderson localization due to spin disorder: a driving force of temperature-dependent metal-semiconductor transition in colossal-magnetoresistance materials

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We study temperature induced metal-insulator transition in doped ferromagnetic semiconductors, described by s-d exchange model. The transition is a result of the mobility edge movement, the disorder being due to magnetic ions spin density fluctuations. The electrons are described in the tight binding approximation. Using ideas and methods of Anderson localization theory we obtain simple formulas, which connect the mobility edge with short-range order characteristics of the magnetic subsystem – static spin correlators. We discuss the application of the theory to several groups of novel colossal-magnetoresistance materials and include the reproduction of the paper [1] published by us 10 years ago.

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I. PRE-INTRODUCTION

Conducting ferromagnets exhibiting resistivity peak and colossal magnetoresistance near the Curie point such as europium oxide- and sulphide-based compounds Eu$_{\text{1-x}}$R$_x$X where X=O, S, R=La, Gd and chromium-chalcogenide spinels ACr$_2$X$_4$ where A=Cd, Hg, Cu, Fe, X=S, Se were studied from 60’s to late 70’s extensively. Because of difficulties in sample preparation and other reasons the interest of RD community in these compounds declined in early 80’s. The same, even earlier, happened to manganite perovskites La$_{1-x}$A$_x$MnO$_3$ where A=Ca, Ba, Sr, Pb. Nevertheless, some studies in this field continued through the 80’s. In 1993, in connection with new thin-film technology for manganite perovskites, the interest in these, called today colossal magnetoresistance (CMR), compounds has been revived. As a result, approaches and concepts developed and applied during 70’s and 80’s has again been brought into the focus of attention. Partial list includes: magnetic polaron versus FM-AFM phase separation, ferromagnetism in narrow conduction band, non-pole structure of Green function in s-d and Hubbard models. This concerns also the concept of localization driven by magnetic fluctuations. We published 10 years ago a series of papers on the resistivity peak and magnetoresistive effect in magnetic semiconductors. To the best of our knowledge, the present authors were the first to apply the concept of mobility edge for disorder determined by spin fluctuations. The concept of localization driven by magnetic fluctuations has become actual again, when the problem of resistivity peak and colossal magnetoresistance has again been brought into a focus of attention.

Below we reproduce our paper ”Anderson localization in ferromagnetic semiconductors due to spin disorder. I. Narrow conduction band” [1], the first one from this series.

II. INTRODUCTION

Magnetic semiconductors form a wide class of materials with unique physical properties, strong temperature dependence of the electroresistivity being one of them [1]. This phenomenon was most thoroughly studied experimentally in cubic ferromagnetics, such as CdCr$_2$Se$_4$, EuO and EuS. All these materials, when being deficient of chalcogen (oxygen) have n-type conductivity. The deficit being strong enough, at low temperatures these materials have temperature-independent metallic conductivity or conductivity with small activation energy (a few meV); when the temperature $T$ approaches the Curie temperature $T_c$ electroresistivity rises sharply. The rise can amount to four orders of magnitude in CdCr$_2$Se$_4$, six orders in EuS$_{1-x}$, and even twelve orders in EuO$_{1-x}$ [1]. The same behavior of the resistivity can be seen in Eu$_{1-x}$La$_x$S [2], Eu$_{1-x}$Gd$_x$O [3] and Eu$_{1-x}$Gd$_x$O$_{1-y}$ [4]. In the paramagnetic region there are two types of behavior: in CdCr$_2$Se$_4$ and EuO$_{1-x}$ ($x_i<4\%$) the resistivity has large activation energy ($\sim .2$ eV and $\sim .3$ eV...
respectively) in the whole temperature range. At the same time the resistivity of EuS_{1−x} and of EuO and EuS being doped by a three-valent rare earth metal (and also of Eu_{1−x}Gd_{x}Se, which at x > 0.01 is ferromagnetic) returns at high temperatures to the initial quasi-metallic behavior.

The common feature of all these materials is a strong interaction between electrons and spins of the magnetic d- and f-ions. It is commonly accepted now that the electrical of these materials are determined by the interaction of electrons with the magnetic subsystem. Pre-existing explanations of the above-mentioned resistivity behavior used the following two approaches:

1. magnetopolaronic approach (in various modifications), which was used for the description of metal-insulator transition in EuO_{1−x};
2. concentrational approach, when it was supposed that the resistivity rise is due to the movement of the conductivity band edge with the temperature relative to the fixed impurity levels.

We describe the conductivity behavior in all above-mentioned materials in a novel approach, which is based upon the ideas and methods of localization theory. The differences in the resistivity behavior between materials, as it will be shown later, are due to the different relations of the band width to the spin exchange splitting and also to the different electron-impurity interactions.

### III. HAMILTONIAN AND THEORETICAL FORMULATION

Hamiltonian of the electron is

\[ H_e = \sum_{l,l',\sigma} V(l-l')c_{l\sigma}^\dagger c_{l'\sigma} - 2I \sum_{l,\sigma,\sigma'} S_{l\sigma} s_{l\sigma'} c_{l\sigma'}^\dagger c_{l\sigma} + V_{e-1}, \]

where \( V(l-l') \) is the hopping integral, \( I \) is the s-d(f) exchange integral, \( S_{l\sigma} \) is spin localized on \( l \) site, \( c_{l\sigma}^\dagger \), \( c_{l\sigma} \) are electron creation and annihilation operators, and \( V_{e-1} \) is the potential of electron-impurity interaction. (We suppose that magnetic and crystallographic lattices coincide and that there is one electron orbit per site).

It is convenient to separate out two limiting cases \[13\]

1. narrow conduction band \( W \ll |I|/S \), where \( W \) is the band width;
2. wide conduction band \( W \gg |I|/S \).

As it was shown in band calculations, the conduction band in CdCr_{2}Se_{4}, formed by p-d\(\gamma\) orbitals (3d\(\gamma\) Cr + 4p Se) is narrow \[14\], and p-d\(\gamma\) conduction bands in EuX are wide \[15\]. It is worth noting however, that strong inequalities for these materials do not hold.

In Part I of our paper we shall be interested in the narrow band case. In this case it is convenient to consider Hamiltonian \[1\] in the representation which diagonalizes the main term – s-d exchange. We restrict ourselves to the quasiclassical limit \( 2S \gg 1 \), when we can consider spins as vectors with fixed length,

\[ S_l = S n_l; \]

The transformation of the initial creation and annihilation operators \( c_{l\sigma}^\dagger \), \( c_{l\sigma} \) into new ones \( C_{l\alpha}^\dagger \), \( C_{l\alpha} \) is equivalent to the quantization of the electron spin in the local reference frame, the OZ axis being parallel to \( n \). We have

\[ \left( \begin{array}{c} C_{l+} \\ C_{l-} \end{array} \right) = \left( \begin{array}{cc} u_l & v_l \\ -v_l \exp(i\phi_l) & u_l \exp(-i\phi_l) \end{array} \right) \left( \begin{array}{c} c_{l+} \\ c_{l-} \end{array} \right), \]

where \( u_l = \cos(\theta_l/2), v_l = \sin(\theta_l/2) \exp(-i\phi_l) \), Hamiltonian \((1)\) after the transformation \((2)\) takes the form

\[ H_e = \sum_{l< l'} V^{\alpha\alpha'} (l, l') C_{l\alpha}^\dagger C_{l'\alpha'} + \sum_{l, \alpha, \alpha'} S I^{\alpha\alpha'} C_{l\alpha}^\dagger C_{l\alpha'} + V_{e-1}, \]

where matrices (in spin space) \( \hat{V}(l, l') \) and \( \hat{I} \) are

\[ \hat{V}(l, l') = V \left( \begin{array}{ccc} u_l u_{l'} + v_l v_{l'} & -u_l v_{l'} + v_l u_{l'} \\ u_l v_{l'} - v_l u_{l'} & u_l u_{l'} + v_l v_{l'} \end{array} \right), \]

\[ \hat{I} = \left( \begin{array}{cc} I & 0 \\ 0 & -I \end{array} \right), \]

and nearest neighbors approximation is used.

The conduction band being narrow, we must retain in Eq. \((1)\) the creation and annihilation operators with \( \alpha = sgn I = +1 \) only. The Hamiltonian then takes the form

\[ H_e = \sum_{l< l'} V_{l\ell} C_{l+}^\dagger C_{l+} + V_{e-1}, \]

\[ V_{l\ell} = V(n_l, n_{l'}) = V \left[ \cos \left( \frac{\theta_l}{2} \right) \cos \left( \frac{\theta_{l'}}{2} \right) + \sin \left( \frac{\theta_l}{2} \right) \sin \left( \frac{\theta_{l'}}{2} \right) \exp i(\phi_l - \phi_{l'}) \right]. \]

When there is a complete order \((T = 0)\) \( \theta_l = 0 \). When \( T > 0 \) the angles \( \theta \) and \( \phi \) and hence the Hamiltonian \( H_e \) are the functions of time. Luckily for us there is a small parameter ratio of typical fluctuations frequency to electron energy \( \sim kTc/I \). Then the adiabatic approximation is applicable, and what we need to solve is static problem with the random Hamiltonian \( H_e(t) \). Hence we obtain the disordered system with an off-diagonal disorder \[10\]. We suppose also that the influence of the electron-impurity interaction on the conduction electrons is much less than the influence of magnetic disorder, and completely neglect the former.

The analysis of Hamiltonian \((1)\) is conveniently started with the density of states.
IV. THE MOBILITY EDGE CALCULATION

Let us turn to Hamiltonian (1). As is well known, the problem of the mobility edge calculation is equivalent to the calculation of the convergence radius for a non-averaged Green function self-energy perturbation series expansion. The site diagonal representation of matrix elements of the Green function being written in the form

$$G_{ii}(E) = [E - M_i(E)]^{-1},$$

we obtain for the self-energy $M_i(E)$ the series

$$M_i(E) = \frac{1}{E} \sum_{l_2} V_{ll_2} V_{l_2l_i} + \frac{1}{E^2} \sum_{l_2 l_3} V_{ll_2} V_{l_2l_3} V_{l_3l_i} + \ldots$$

$$= \sum_{L} X_L. \quad (11)$$

Every term in $X_L$ corresponds to a definite path with $L$ steps starting from the site $l_1$ and ending at it. To each step from a site $l$ to one of its nearest neighbors $l'$ there corresponds the factor $V_{ll'}$ and the passage of every intermediate site produces multiplication by the factor $1/E$.

It is worth noting that the analysis of the series (11) is much easier than that of the series involved in the case of the traditional Andersen model with diagonal disorder. It is due to the fact that the expectation value of every term in Eq. (11) is finite and, hence, the convergence of this series with probability one is equivalent to its convergence in some norm. Indeed we can use the theorem according to which the series of independent random variables, say $\sum_L X_L$, converges with probability one if and only if the series $\sum_L <X_L>$ and $\sum_L <X_L^2>$ converge, where the brackets mean the average $\langle \rangle$ in our case over the distribution $H_L(t)$, that is due to the ergodic hypothesis the average over all spin configurations. Let us analyze now the averaged series. We introduce formally a small parameter $1/z$, where $z$ is the number of nearest neighbors, and then calculate the averages in the leading approximation with respect $1/z$. Since the ratio of the number of intersections to the total number of sites for a typical path is of the order of $1/z^p \quad (p > 0)$, we shall on averaging neglect the intersections at all, i.e., when calculating $<V_{l_1 l_2} \ldots V_{l_k l_s}>$ we consider all the sites involved as different ones, though the number of paths of of the given Length $L$ being approximated by $z^L$. Note that our analysis deviates from the traditional one for the case of the off-diagonal disorder (see e.g. Ref. [21]). Neglecting the correlations due to self-intersections of the paths we, however, take into account the correlations created by magnetic short-range order, which is more important in the problem.

We are interested in the convergence radius of the series

\[\text{FIG. 1. The temperature dependence of the density of states in doped ferromagnetic semiconductor (the lower local band).} \]
Then the convergence of the series (12) is equivalent to
\[ < M(E) >= E \sum_{L} \frac{z^{L}}{E^{L}} M_{L}, \] (12)
where the following notation is introduced:
\[ M_{L} = < < V_{11} V_{12} \ldots V_{1L} > > = \]
\[ < \int d\mathbf{n}_{1} \ldots d\mathbf{n}_{L} V(\mathbf{n}_{1}, \mathbf{n}_{2}) \ldots V(\mathbf{n}_{L}, \mathbf{n}_{1}) > > \]
with \( P(\mathbf{n}_{1}, \ldots, \mathbf{n}_{L}) \) being the probability density of a
given spin configuration and the brackets \(< \ldots >_{L} \) denote the average over all paths of a given length. It is
worth noting that in our approximation the convergence
of the series \( \sum_{L} < X_{L} > \) is equivalent to the convergence
of \( \sum_{L} X_{L}^{2} > > . \)

Let us consider two limiting cases:

1. \( T = 0 \)
   In this case the hopping integral does not fluctuate and the series take the form
   \[ < M(E) >= E \sum_{L} \frac{z^{L} V^{L}}{E^{L}}, \] (14)
   and its convergence radius \( |E_{c}| = zV \) naturally coincides with the exact band edge (the center of the band corresponds to \( E = 0 \)).

2. \( T = \infty \)
   In this case the only factorizable. The eigenfunction of \( \hat{V} \) corresponding to the larger of the two eigenvalues has the form
   \[ f(\mathbf{n}) = \left| P(\mathbf{n}) \right|^{1/2} \cos(\theta/2). \] (22)
Using this trial function in the variational principle we obtain
\[ |E_{c}| = \frac{zV}{2} \frac{1 + 2 < n^{z}> + < \mathbf{n}_{0} \cdot \mathbf{n}_{1} >}{1 + < n^{z}>} = \]
\[ \frac{zV}{2} \frac{1 + 2 < S^{z}> /S + < S_{0} \cdot S_{1} > /S^{2}}{1 + < S^{z}> /S}. \] (23)

Hence the result is the mobility edge expressed in terms of magnetization and pair correlation function of the nearest-neighbor spins. Our method of mobility edge
evaluation can be generalized also to the case of arbitrary relation between IS and \( W \) \( [23] \). For arbitrary temperature
the calculation of the mobility edge is equivalent to solving the cumbersome integral equation. For \( T \gg T_{c} \), however, this equation can be easily solved; the mobility
edge then is
\[ |E_{c}| = \frac{W}{4} + IS - \left[ (IS)^{2} + \frac{W^{2}}{16} \right]^{1/2}. \] (24)

V. COMPARISON WITH EXPERIMENT

Though Eq. \( [23] \) is obtained for classical spins we’ll use it to explain the temperature dependence of the resistivity of CdCr2Se4 (Fig. 2). The temperature dependence
of the resistivity at low temperatures \( (T < 50 \text{ K}) \) shows that the chemical potential lies in the close vicinity of the band edge. At those temperatures the resistivity is defined by the exact position of \( \xi \) relative to the band
edge; at higher temperatures when $E_c$ moves upward, the dependence of $\rho$ on $T$ is determined mainly by the shift of $E_c$ and for the chemical potential we may use the approximation $\xi = -zV$. Inserting into Eq. (6) the expression for the mobility edge we obtain

$$\rho = \rho_0 \exp \left[ \frac{1 - <S_0 \cdot S_1 > / S^2}{1 + <S^2> / S} \left( \frac{W}{4kT} \right) \right]$$

where $W = 2eV$ is the conductivity band width. Using for the correlator $<S_0 \cdot S_1>$ the results of Ref. 24, and for $<S^2>$ the molecular field approximation ($S = 3/2$), and choosing the appropriate values of $W$ and $\rho_0$, we obtain the plot presented on Fig.3.

![Fig.2](image.png) ![Fig.3](image.png)

**FIG. 2.** Electrical resistance of Cd$_{0.99}$In$_{0.01}$Cr$_2$Se$_4$ (reproduced from Ref. [1]).

**FIG. 3.** Electrical resistance calculated on the basis of Eq. (25).

The chosen value for the band width is $W = 0.8$ eV, which well agrees with other estimations [13,14]. We can deduce that Eq. (25) can claim for a quantitative description of the experimental dependence. This formula also explains the colossal negative magnetoresistance of the materials under consideration, as the magnetic field strongly influences the spin correlators in the exponent, especially near $T_c$.

In Refs. [24] the data for high resistivity samples of CdCr$_2$Se$_4$ was presented. In this case the resistivity as the function of temperature has no maximum, but the dependence of $\log \rho$ on $1/T$ changes its slope after the transition to the paramagnetic region. Such a behavior is also described by Eqs. (6) and (25) with the chemical potential $\zeta < -zV$.

**VI. DISCUSSION (10 YEARS AFTER)**

It is expedient to discuss the applicability of the approach of this ancestor paper to new classes of materials and its place among more recent theoretical approaches. Our principal point was that strongly interacting system of electrons and magnetic ions (s-d exchange model) in classical approximation for the ions spins and narrow electron band, reduces to a random-bond model with off-diagonal disorder depending on an instant spin configuration. This double-exchange (DE) Hamiltonian (Eq. (3) in the present manuscript) has been reopened quite recently [28].

The model, we believe, can be applied with minimal changes to manganite pyrochlores discovered recently [27,28]. Its application to high- carrier concentration CMR compounds such as manganite perovskites demands discussions and probably additional ideas. Formally, in this approximation the difference between, say, LaSrMnO$_3$ and CdCr$_2$Se$_4$ (except for carriers concentration) is that in the latter the doping electrons appear (due to Se vacancies) in the empty d-band, whereas in the former the doping holes appear in the filled d-band (due to capture of d-electrons by Sr atoms). According to our concept, it is the movement of mobility edge, with temperature and magnetic field, relative to the Fermi level that explains the resistivity peak and CMR effect. It is concordant with the recent conclusion of Varma [29] who suggested the decisive role of the above spin-dependent disorder in metal-semiconductor transition and CMR effect in manganite perovskites.

The viewpoint that spin disorder alone can not localize carriers at temperatures close to $T_c$ and in paramagnetic state has been argued by Millis et al. [30] on the basis of a perturbational calculation. Later this group presented very subtle arguments concerning probability distribution of spin-dependent hopping integral [31] in favor of their conclusion. The matter is that, as with every random-bond model, DE Hamiltonian keeps a large portion of the states at the band center delocalized irrespective of how the bond fluctuations are strong. Therefore, it may turn out that the weight of the localized states be insufficient to embed large number of holes (0.2 to 0.3 per cell). After numerical simulation with the use of one-parameter scaling localization theory [32] it has been found that this is really the case for pure DE mechanism. Note that for SC lattice our result of 1988 $E_c = -3V$ (see manuscript above) is in a reasonable agreement with the new more exact $E_c = -3.55V$ [32].

Thus, an agreement with experiment for manganites may be achieved by introducing some additional mechanism. Millis et al. [31] developed and advocated Jahn-Teller polaron mechanism to explain the resistivity peak and CMR. On the other hand, in 1988 we proposed the mechanism based on the pinning of the Fermi energy by the donor levels, due to very weak overlap of the vacancy states with magnetic d-states in CdCr$_2$Se$_4$ (see present paper). Recently our mechanism has been recognized [27,28], and applied to LaBaMnO$_3$ by Bebenin et al. [33]. In the case of manganite perovskites one may speculate...
that the Fermi level is more or less fixed by impurity band formed by Sr, Ba, etc. acceptor states. The band emerges in Mn d-band at low temperatures and holes are mobile because the d-band is delocalized. With increasing the temperature the mobility edge shifts towards higher energies and holes remain in localized part of the band.

Recently an approximate pinning of the Fermi level has been found in a model where diagonal disorder is added to DE hopping Hamiltonian [34]. Numerical simulation using one-parameter scaling localization theory have shown that the states at the Fermi energy are localized in PM region if the intensity of diagonal disorder is as intense as about the bandwidth [33]. Though this model is very attractive, the physical origin of such strong fluctuation of Mn d-levels is not evident. Sheng et al. [34] attribute it to potential fluctuations due to random substitution of La by Sr, Ca etc. using experimental data on resistivity of polycrystalline samples. However, contrary to the assumption of [33] there exist single crystals with zero-temperature resistivity smaller than $10^{-4}$ $\Omega\cdot$cm which do exhibit large resistivity peak [33]. This is an evidence in favor of weak substitutional disorder, and impurity band formation.

The mechanism of conduction in PM region is not established until now but it lies out of scope of this discussion. We hope to return to discussion on this problem in a forthcoming paper.

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