Ph.D Thesis Booklet

Theoretical study of magnetic impurities in low-dimensional systems

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Background

Thin films and nanoparticles are new devices of high-density magnetic recording and spintronics. Because of challenging quantum phenomena these systems are also preferred subjects of modern solid state research. Important features of the electronic and magnetic structure of nanosystems are often determined/modulated by relativistic spin-orbit (SO) interaction that gives rise, e.g., to the magnetic anisotropy, the anisotropic magnetoresistance, the Kerr and Faraday effects, the Rashba effect, and the anomalous and spin Hall effects.

The suppression of the Kondo effect in thin films and wires of dilute magnetic alloys has been observed by G. Chen and N. Giordano [1] in the early 90's. A few years later, Újsághy et al. [2] proposed an explanation of the experimental observations based on a spin-orbit induced magnetic anisotropy that is larger than the Kondo temperature, $k_B T_K$, up to a distance of 150-200 Å from the surface of the film. In this 'dead layer' a level-splitting of the impurity spin can then block the spin-flip processes responsible for the Kondo effect. To explain the unexpectedly large width of the dead layer, Újsághy et al. proposed a model of an impurity with a half-filled *d* shell (S=5/2) immersed in a host metal, where conduction electrons experience spin-orbit scattering through hybridizing with low-lying valence *d* orbitals of the host (HSO model).

Later on a rather different mechanism has been proposed by G. Zaránd and L. Szunyogh [3] that assumes strong local SOC on the impurity's *d* level (LSO model). The basic observation leading to this mechanism is that, for partially (but not half) filled *d* shells, the spin states have also a large orbital content, being strongly coupled to charge Friedel oscillations in the vicinity of a surface. Despite of some numerical estimations the question remained open which mechanism was responsible for the suppression of the Kondo-resistance in a reduced dimensional system.

Theoretical [4,5] and experimental [6] works examined the fluctuating spin g-tensor in small disordered mesoscopic grains using the methods of random matrix theory (RMT). These works motivated us to investigate the level splitting (magnetic anisotropy) distribution caused by the LSO mechanism in ordered and disordered Au particles.

The Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between magnetic impurities in bulk materials [7] has been known for a long time and has become a textbook knowledge. Theory predicts an oscillatory behavior with a magnitude decaying as $1/R^3$, where R is the distance between the impurities, and a frequency related to the extremal vectors of the Fermi surface. The effect of the SO coupling to the exchange interaction, however, has not yet been investigated. Furthermore, a thorough investigation of the RKKY interaction for different metals and surfaces was missing in the literature.

Metallic surfaces, such as Au(111), often exhibit Shockley-type surface states located in a relative band gap of the bulk band structure, and forming a two-dimensional electron gas [8]. One of the most intriguing manifestation of SOC at surfaces is the splitting of the corresponding dispersion relation, known as Bychkov-Rashba (BR) splitting. Describing and controlling the BR splitting of surface states is crucial for spintronics applications. In case of Au(011) surface and some surface alloys, like Bi/Ag(111) or Bi/BaTiO_3(001), numerical calculations predicted a BR splitting that is anisotropic in reciprocal space, however, no careful theoretical exploration of this effect occurred so far.

Objectives

The main objective of my Ph. D work was to study spin-orbit induced effects on magnetic impurities placed, in particular, in low-dimensional systems. In order to access the problems mentioned above it was necessary to work out a link between the quantum-impurity models and realistic bandstructure calculations of the valence and conduction electrons, for both surfaces and nanoparticles. For this purpose I employed a tight-binding Green's function (TB-GF) technique, which allowed for a perfect treatment of the semi-infinite geometry of a surface and made also possible a nonperturbative treatment of the SO interaction.

By using the TB-GF method, a quantitative comparison of the HSO and LSO models became possible for the realistic cases of Cu and Au host surfaces. In this way we hoped to make reliable predictions in favor of one of these mechanisms for the surface-induced anisotropy proposed originally by Ujsaghy et al. to explain the Kondo effect in reduced dimension.

Motivated by previous theoretical studies on the spin g-factor distribution, I planned to investigate the magnetic anisotropy of randomly distributed impurities in geometrically ordered and disordered mesoscopic metallic (Au) nanoparticles in terms of the LSO model and real-space TB calculations. The ultimate goal of this study was to establish universality of the magnetic anisotropy in mesoscopic nanograins.

The TB-GF method provided also possibility to calculate the exchange interaction between two impurities in bulk and at surfaces. Here I planned to study how the SO coupling in the host influences the RKKY interaction. In relation to this phenomenon, I also aimed at investigating the Bychkov-Rashba effect in two-dimensional electron states at Au(111) and Au(110) surfaces, with emphasis to the anisotropic BR splitting.

Thesis statements

1. I derived analytical expression of the HSO and LSO magnetic anisotropy constants, *K(d)*, and performed an asymptotic analysis for large distances d from the surfaces. As a result of this analysis the period of the oscillations could be identified as the length of an extremal vector of the Fermi surface of the bulk host and the amplitude decayed as $1/d^2$ in both models [Pubs. 1,2].

2. In terms of the TB-GF method, I performed calculations of the anisotropy constant of impurities near the (001) surface of copper and gold. The numerical results confirmed the analytical expressions for *K(d)*. In case of the HSO model the magnitude of *K(d)* remained well below the Kondo temperature, while within the LSO model it turned to be in the desired energy range up to $d \sim 100$ Å. I, therefore, concluded that the LSO mechanism can explain the suppression of the Kondo resistance observed in thin films [Pubs. 1,2].

3. Using Random Matrix Theory, I performed an analysis of the spectral statistics of the Hamiltonian of disordered gold nanoparticles. I showed that, within satisfactory numerical accuracy, the distribution of eigenenergy spacings follows a universal behavior: without SOC the distribution refers to a Gaussian Orthogonal Ensemble (GOE), while SO coupling on the gold atoms implies a Gaussian Symplectic Ensemble (GSE) for the level-spacing distribution. [to be published]

4. I showed that, within the LSO mechanism, the magnetic anisotropy can, in general, be described by a 4x4 self-adjoint matrix containing six real parameters. I explored the structure of this matrix and defined magnetic anisotropy terms related to a quadrupole operator basis. In the case of ordered nanograins, I numerically found symmetry constraints in the six-dimensional parameter space. Using representation theory of double groups I fully explained the observed symmetry relations of the anisotropy parameters. [to be published]

5. By using a T-matrix technique combined with the TB method, I examined the RKKY interaction between impurities embedded in Cu and Au bulk host. My numerical calculations confirmed within a relative accuracy of 1 % that magnitude of the oscillations of the exchange interaction decays as $1/R^3$, R being the distance of the impurities, and that the period is related to the length of an appropriate extremal vector of the Fermi surface. I pointed out that strong SO coupling in Au drastically reduced the magnitude of the exchange energy. The underlying physics is attributed to the spin-mixing induced by SO coupling. [Pubs. 4,5]

6. I performed detailed calculations for the dispersion relations of the Shockley-type surface states of Au(110) and on Au(111) surfaces. In agreement with analytical $k \cdot p$ perturbation theory and first principles calculations, I confirmed that the Bychkov-Rashba splitting is anisotropic for Au(110) that can be described with a Hamiltonian in first order of k. Furthermore, my calculations evidenced that for Au(111) surface (case of C_{3v} symmetry) an anisotropy of the BR splitting arises according to an effective Hamiltonian in third order of k. [Pub. 3 & to be published]

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List of Publications

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