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Unraveling the nature of carrier-mediated ferromagnetism in diluted magnetic semiconductors

Elucider la nature du ferromagnétisme induit par des porteurs dans les semiconducteurs magnétiques dilués

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ABSTRACT

After more than a decade of intensive research in the field of diluted magnetic semiconductors (DMS), the nature and origin of ferromagnetism, especially in III–V compounds, is still controversial. Many questions and open issues are under intensive debates. Why after so many years of investigations, Mn-doped GaAs remains the candidate with the highest Curie temperature among the broad family of III–V materials doped with transition metal (TM) impurities? How can one understand that these temperatures are almost two orders of magnitude larger than that of hole-doped (Zn,Mn)Te or (Cd,Mn)Se? Is there any intrinsic limitation or is there any hope to reach room-temperature ferromagnetism in the dilute regime? How can one explain the proximity of (Ga,Mn)As to the metal–insulator transition and the change from Ruderman–Kittel–Kasuya–Yosida (RKKY) couplings in II–VI compounds to double-exchange type in (Ga,Mn)N? In spite of the great success of density functional theory-based studies to provide accurately the critical temperatures in various compounds, till very lately a theory that provides a coherent picture and understanding of the underlying physics was still missing. Recently, within a minimal model, it has been possible to show that among the physical parameters, the key one is the position of the TM acceptor level. By tuning the value of that parameter, one is able to explain quantitatively both magnetic and transport properties in a broad family of DMS. We will see that this minimal model explains in particular the RKKY nature of the exchange in (Zn,Mn)Te/(Cd,Mn)Te and the double exchange type in (Ga,Mn)N and simultaneously the reason why (Ga,Mn)As exhibits the highest critical temperature among both II–VI and III–V DMS's.

RÉSUMÉ

Après plus d'une décennie de recherches intensives dans le domaine des semiconducteurs magnétiques dilués (DMS), la nature et l'origine du ferromagnétisme, en particulier dans les composés III–V, restent controversées. De nombreuses questions et problèmes ouverts sont toujours sujets à d'intenses débats. Pourquoi, parmi la grande famille des matériaux III–V, et pour une concentration donnée en métal de transition, le composé (Ga,Mn)As

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reste-t-il le candidat présentant encore la température critique la plus élevée ? Comment peut-on comprendre que ces températures soient presque de deux ordres de grandeur supérieures à celles observées dans (Zn,Mn)Te dopé en trous ou (Cd,Mn)Se ? Subsiste-t-il pour ces matériaux dilués un espoir d'observer un ordre ferromagnétique au-delà de la température ambiante, ou est-il fatalement anéanti par des limitations physiques intrinsèques ? Comment expliquer que (Ga,Mn)As soit si proche de la transition métal–isolant ? Comment comprendre la nature des couplages magnétiques passant typiquement de RKKY dans les composés II–VI à double échange dans (Ga,Mn)N ? Des études, basées sur la théorie de la fonctionnelle de la densité, ont pu fournir avec précision les températures critiques dans divers composés. Cependant, un modèle théorique en mesure de fournir une vision unifiée et une compréhension de la physique sous-jacente manquait toujours. Très récemment, dans le cadre d'un modèle minimal, il a été possible de montrer que, parmi les paramètres physiques, la clé réside dans la position du niveau accepteur de l'impureté magnétique. En adaptant ce dernier, il devient en effet possible d'appréhender la diversité des propriétés magnétiques et aussi de transport dans une large famille de DMS. Nous verrons alors que le modèle minimal explique non seulement la nature RKKY des couplages magnétiques dans (Zn,Mn)Te/(Cd,Mn)Te ou leur caractère de double échange dans (Ga,Mn)N, mais aussi la raison pour laquelle (Ga,Mn)As présente les températures de Curie les plus élevées parmi les DMS II–VI et III–V.

1. Introduction

In recent years, the rapidly growing field of diluted magnetic semiconductors (DMS) [1–3] has attracted a considerable interest owing to their potential for spintronic devices. One of the main goals is to combine the traditional electronic functionality (charge) and the spin degree of freedom of the electrons/holes. This requires optimal candidates that exhibit room-temperature ferromagnetism. Recent progress in growth processes of TM-doped III–V semiconductors has boosted the interest for such novel materials. Among III–V DMS, Mn-doped GaAs that could be considered as the prototype is certainly the most widely studied (both transport and magnetic properties). However, the understanding of the fundamental physical properties in these doped compounds involves theoretical speculations that are subject to controversy. The quest for a model able to capture quantitatively the physics and identify the key physical parameters that control both magnetic and transport properties was a clear open issue over the last decade. Till recently, DMS-based theoretical studies could be split into two main distinct types: (i) first-principle based approaches [2] and (ii) Zener Mean Field type theories [1,4]. The first kind is based on density functional theory (DFT) such as local spin density approximation (LSDA) or generalized gradient approximation (GGA), for instance. They require no adjustable parameters and are essentially material specific. The second type is a model approach that includes a realistic description of the host band structure within a 6-band or 8-band Kohn–Luttinger Hamiltonian [5,6] and a local p–d exchange between itinerant holes and localized impurity spins. In Zener Mean Field theory the p–d coupling is treated perturbatively and the dilution effects at the lowest order, also known as Virtual Crystal Approximation (VCA). As a consequence, the Fermi level lies inside the unperturbed valence band (VB) leading to the so-called valence band scenario (see Fig. 1a). Regarding the specific case of (Ga,Mn)As, the perturbative VB picture is inconsistent with first-principle-based studies. Indeed, density functional calculations clearly predict the existence of a well-defined preformed impurity band (see Fig. 1b), which for a sufficiently large concentration of Mn (beyond 1%) overlaps with the valence band [2,7]. It was found that the Fermi level lies in the resonant impurity band. Thus, ab initio studies clearly support the so-called "impurity band picture" (IB). It is worth noticing that both optical conductivity measurements [8–10] and proximity of Mn-doped GaAs to the metal–insulator transition [11–13] fully support the IB picture. In spite of all that, the issue of VB versus IB scenario is still controversial. On the other hand, the VB scenario remains suitable to describe the physics in II–VI materials such as Mn-doped ZnTe, CdTe, ZnSe, for instance. The reason for this is the absence of hybridized p–d states in the vicinity of the top of the valence band in these alloys. In other words, treating the substitution by Mn as a perturbation remains a good approximation in II–VI materials. In the following, we present a two step approach that allows to describe both magnetic and/or transport properties of a wide range of diluted magnetic semiconductors. Concerning the magnetic properties that are our main concern in this paper, the two steps are described as follows. The first one consists in calculating the magnetic couplings between localized spins randomly distributed in the semiconductor host. To this end, one can use first-principle calculations or suitable model approaches. The purpose is to build the effective Heisenberg Hamiltonian of the problem. This spin Hamiltonian is diagonalized during the second step within the self-consistent local random-phase approximation [15,16] (SC-LRPA) procedure, which is described in the next section.

2. Self-consistent local RPA diagonalization of the dilute Heiseinberg Hamiltonian

The Hamiltonian that describes N_{imp} interacting spins $\mathbf{S}_i$ (classical or quantum) randomly distributed in the host lattice is the dilute/disordered Heisenberg model,