Magnetic interactions of substitutional Mn pairs in GaAs

T. O. Strandberg and C. M. Canali
School of Pure and Applied Natural Sciences, Kalmar University, 391 82 Kalmar, Sweden

A. H. MacDonald
Department of Physics, University of Texas at Austin, Austin, Texas 78712, USA

We employ a kinetic-exchange tight-binding model to calculate the magnetic interaction and anisotropy energies of a pair of substitutional Mn atoms in GaAs as a function of their separation distance and direction. We find that the most energetically stable configuration is usually one in which the spins are ferromagnetically aligned along the vector connecting the Mn atoms. The ferromagnetic configuration is characterized by a splitting of the topmost unoccupied acceptor levels, which is visible in scanning tunneling microscope studies when the pair is close to the surface and is strongly dependent on pair orientation. The largest acceptor splittings occur when the Mn pair is oriented along the $\langle 110 \rangle$ symmetry direction, and the smallest when they are oriented along $\langle 100 \rangle$. We show explicitly that the acceptor splitting is not simply related to the effective exchange interaction between the Mn local moments. The exchange interaction constant is instead more directly related to the width of the distribution of all impurity levels – occupied and unoccupied. When the Mn pair is at the (110) GaAs surface, both acceptor splitting and effective exchange interaction are very small except for the smallest possible Mn separation.

I. INTRODUCTION

Experimental progress in the past 5 years has led to a large number of experimental studies of magnetic and nonmagnetic transition-metal impurities in semiconductors using advanced scanning tunneling microscope (STM) techniques.[1–7] This effort was motivated in part by the hope that the high-resolution imaging and spatially resolved spectroscopic power of the STM could help in developing an accurate microscopic picture of dilute magnetic semiconductor (DMS) magnetism. In DMSs magnetic impurities provide local moments, which can couple to yield a collective ferromagnetic state. In the prototypical DMS, (Ga,Mn)As, the Mn impurities act as acceptors providing itinerant holes that can mediate long-range interactions between local moments. Among the open issues in DMS physics[8] are the precise character of the hole states, the nature of the coupling of the holes with the local magnetic moments, and the properties of the ensuing magnetic interaction between local moments. STM experiments performed recently[4–7] are playing a decisive role in clarifying some of these issues.

In Ref. [4] STM substitution techniques were used to incorporate individual Mn atoms into Ga sites in a GaAs (110) surface. Real-space spectroscopic measurements in the vicinity of an isolated Mn impurity revealed the presence of a mid-gap resonance arising from a Mn induced acceptor state. High-resolution imaging showed that the acceptor wave function is strongly anisotropic with respect to the crystal axes of the host. When two Mn atoms were incorporated close to each other, two resonances appeared in the gap, split by approximately 0.5 eV. The splitting was found to be strongly dependent on the Mn pair orientation with respect to the GaAs crystal structure and on Mn separation. A simple toy model, describing acceptor states coupled to the Mn ion local moments of the two impurities,[9] suggested that a measurable splitting of the acceptor levels could only occur if the two Mn local moments were ferromagnetically aligned.[4] Since acceptor-level splitting is an observable indicator of ferromagnetic coupling, it seemed plausible that the dependence of this splitting on separation should be related at least qualitatively to the Mn-Mn exchange interaction. If so, the STM experiment could be used to measure exchange interactions between the Mn moments and test theories of this interaction. One of the purposes of the present study is to examine this relationship quantitatively.

Several of the experimental features uncovered in Ref. [4] could be qualitatively accounted for theoretically by a tight-binding model calculation for Mn in bulk GaAs presented in the same paper. A more thorough comparison between experiments and theoretical modeling, both for Mn atoms in the bulk *and Mn near the surface* is nevertheless necessary to interpret the experiments, motivating the present theoretical work. Here we consider a kinetic p-d exchange tight-binding model[10] in which the effective exchange interaction between the hole states and the local Mn moments arises from hybridization of the impurity d levels with p levels of the host. The model is solved numerically for large super-clusters containing up to 3200 atoms. This approach allows us to place the Mn pair either in bulk GaAs or on the (110) surface.

Within this model, we study the electronic and magnetic properties of Mn pairs in GaAs, assuming that the two local magnetic moments are collinear, having either parallel [ferromagnetic (FM)] or antiparallel [antiferromagnetic (AFM)] relative orientation. One of our goals is to study the spin-orbit induced magnetic anisotropy energy of the system and see how this quantity is related to the properties of the mid-gap acceptor states. In the FM configuration the magnetic moment tends to point along the direction of the pair, while in the AFM configuration there is typically a quasi-easy plane per-