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# Interdot exchange coupling in superferromagnetism

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**Abstract.** The electronic structure of the arrays of transition-metal quantum dots separated by nonmagnetic tunnel barriers is considered on the basis of the Anderson localization theory. From the analysis of the grand canonical potentials, we identify the temperature-dependent interdot superexchange coupling constant.

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Quantum dot (QD) arrays allow one to study the interactions, transport processes, and phase features of a system with widely varying sizes, energy scales, and electronic densities (cf. [1]). Particular effort is currently devoted to the investigation of the magnetic properties of such structures [2–6]. Recent experimental work on one- and two-dimensional (1D and 2D, respectively) self-organized (i.e., quasi-periodic) arrays of nanosized transition-metal dots show magnetic ordering [3–6] of mostly dipolar type. However, in the case of a 2D system of Fe dots on an insulator substrate, a long-range order has been found [2] that has been attributed to a contribution of superexchange coupling between the dot supermoments.

In the work to be reported here, such a superexchange mechanism in the magnetic ordering of QD arrays is analyzed theoretically by the employment of the band-structure-based shell model [7, 8]. Within this model, the band structure of $s$ and $d$ (or $f$) electrons is modulated, because of their confinement in a finite volume. This gives rise to the well-known gross-shell behavior of the level density (cf. [9]), which results in the oscillatory size dependence of the supermoments and minority spin holes. Such a picture reproduces rather well [7] the experimental observations [10, 11] of oscillating magnetic supermoments of free individual transition-metal dots as a function of their size.

If such dots are arranged in an array of sufficiently dense packing, their coupling modifies the level density; this can result in various types of magnetic coupling between the dots [7], similar to the ones for atoms in a lattice. Given that the single-particle level density difference $\delta\varrho = \varrho_{\rm f}(\varepsilon) - \varrho(\varepsilon)$ between a ferromagnetically arranged and an uncoupled dot system is known, the temperature- ($T$-) dependent superexchange coupling constant $J$ is calculated as the difference of the respective grand canonical potentials

$$J = \Omega_{\rm f} - \Omega_{\rm d} \approx -(k_{\rm B}T)\int_{-\infty}^{\infty} {\rm d}\varepsilon\ \delta\varrho(\varepsilon)\cdot\ln[1+\exp\{(\mu-\varepsilon)/k_{\rm B}T\}]\,, \tag{1}$$

where $\mu$ is the electronic chemical potential.

One of the questions that arises with respect to the coupling-induced change in the level density is related to the formation of a coherent Bloch state from the dot supermoment wave functions in an array. In the Anderson localization theory (see, for example, [12] and references therein), such conditions are given by $\Gamma/B < 2$ , where $B$ is the width of the miniband splitting caused by the coupling effects (see (3)), and $\Gamma$ represents the level broadening caused, e.g., by the uncertainties in the dot linear sizes $\delta R$. Within the jellium model, applied to a single QD, we estimate $\Gamma \approx 2\varepsilon_{\rm F}\cdot\delta R/R$, where $\varepsilon_{\rm F}$ denotes the Fermi energy and $R$ is the average radius of the dots. Since the variation of the linear dot sizes is related to the variation of atom number $N$ in the $d$-dimensional dot, $\delta R/R \approx \delta N/N$, the condition referring to the coherent states in a QD array becomes $\delta N/N < 2B/\varepsilon_{\rm F}$. Having defined this condition, we will further model the electronic properties.

We assume a simple periodic structure of the QD array with a period $a_i$ in the $i$th direction ($i = 1, ..., D$). The valence electron states in this periodic field can be described by Bloch functions associated with the quasi-energy, $\varepsilon_\alpha = \varepsilon_{\bar\alpha} + \Delta\varepsilon(\mathbf{k})$, and quantum numbers $\alpha = \{\bar\alpha, \mathbf{k}\}$; the electron quasi-momentum in $D$ dimensions is $\mathbf{k}$, and the miniband quantum number $\bar\alpha$ characterize the energy levels $\varepsilon_{\bar\alpha}$ in a single dot. Then the level density change is expressed as

$$\delta\varrho \approx \int\prod_{i=1}^{D}{\rm d}\Big(\frac{k_i a_i}{2\pi}\Big)\,[\varrho(\varepsilon-\Delta\varepsilon(\mathbf{k}))-\varrho(\varepsilon)]. \tag{2}$$

In an array of dots separated by nonmagnetic tunnel barriers with heights $U_i > \mu$, the supermoment wave functions are nearly undistorted by the small overlap in the coupling region. Then the miniband structure can be approximated by a cosine shape

$$\Delta\varepsilon(\mathbf{k}) = \sum_{i=1}^{D} B_i \, \sin^2(k_i a_i), \quad B_i = 2\omega_{\mathrm{e}} \, P_i \, , \tag{3}$$

where $\omega_{\mathrm{e}}$ is the frequency of electron oscillation within the dot (i.e., collision with the wall) which is different for *sp*, minority ($e = \downarrow$) and majority ($e = \uparrow$) spin bands. As a measure for the overlap integral of the supermoment wave functions of two neighboring dots, the tunneling probability $P_i$ in the $i$th direction can be estimated within the WKB approximation [13] to be

$$P_i \approx \exp\left\{-\sqrt{(U_i - \varepsilon)/\varepsilon_b^i}\right\} \, , \tag{4}$$

where $\varepsilon_b^i = \hbar^2/2m^*(\xi b_i)^2$, where $m^*$ is the effective mass of an electron in the substrate and $b_i$ is the tunneling length. The parameter $\xi$ depends on the shape of the barrier; for instance, $\xi = 2$ is for a square well potential, and $\xi = \pi/2$ is for the inverted harmonic oscillator. One sees that for the barriers of sufficiently large heights ($U_i = U$) and widths ($b_i = b$), only the levels in the vicinity of the Fermi energy will give a noticeable contribution to the integral in (1). Therefore, in the case of *strong* ferromagnets, like the iron series transition metals Co and Ni, we can neglect the influence of the filled majority spin band since it is located below the Fermi energy $\varepsilon_{\mathrm{F}}$.

Next, the coupling constant for a 2D array is calculated from (1)–(4),

$$J \approx J_{\mathrm{D}} \cdot J_{\mathrm{B}}, \tag{5}$$

with the component $J_{\mathrm{B}}$, related to the barrier properties and temperature

$$J_{\mathrm{B}} = \frac{4\varepsilon_b \alpha}{\sin(\alpha)} \exp\left\{-\sqrt{(U-\mu)/\varepsilon_b}\right\}, \tag{6}$$

where $\alpha = \pi T/T_\alpha$, $T_\alpha = 2\sqrt{(U-\mu)\varepsilon_b}/k_{\mathrm{B}}$, and the component

$$J_{\mathrm{D}} = (\mu - U)\left[\varrho_s' \omega_s + \varrho_\downarrow' \omega_\downarrow\right] \tag{7}$$

which is related to the dot electronic structure, with the level density $\varrho_s$ and $\varrho_\downarrow$ of *sp* and minority-spin bands, respectively. The *prime* denotes the energy derivative taken at the Fermi energy.

Equation (5) quantifies the direct superexchange interdot coupling originating from tunneling between the dots. It can be seen that the coupling constant shows nonoscillatory dependence on the interdot separation distance similar to the coupling properties of ferromagnets abutted by a nonmagnetic insulator [14]. The exponential decrease of the coupling strength (i.e., the factor $J_{\mathrm{B}}$ (6)) with increasing separation distance arises from the exponentially decaying overlap of supermoment wave functions extending their tails into the barrier. This restricts the interdot separation at which the exchange can contribute to the magnetic ordering. This is in agreement with recent experiments (cf. [2], for example); in these studies, no long-range magnetic ordering was found in a system of self-organized Fe islands with too low surface coverage. As is seen from (6), reducing the height of the barrier yields exponentially larger exchange fields at the same interdot separations. Therefore, using, e.g., a semiconductor substrate may allow one to observe the direct interdot exchange coupling in an array of less dense packing (due to lower tunnel barriers and a smaller $m^*$) than on an insulator substrate. For example, in the case of carbon, the barrier height could be reduced to about 1 eV (cf. [15]) and the magnetizing field would be enhanced by the factor $\sim \exp\{5.7\xi b/\mathrm{nm}\}$. Finally, the quantity $J_{\mathrm{B}}$ grows with increasing temperature. Such a behavior is caused by the exponentially increasing tunnel exchange current for higher energy levels. At finite temperature, the electrons within an energy range $k_{\mathrm{B}}T$ above the Fermi energy $\varepsilon_{\mathrm{F}}$ contribute to the interdot superexchange coupling at the expense of decreasing occupation of the energy levels below $\varepsilon_{\mathrm{F}}$. Since the relative contribution from the levels above $\varepsilon_{\mathrm{F}}$ is larger, the factor $J_{\mathrm{B}}$ increases as a function of temperature. We note, however, that the factor $J_{\mathrm{D}}$ is expected to decrease with increasing temperature because the shell structure of the dots is washed out. This may result in a nonmonotonic thermal behavior of the interdot superexchange coupling [7].

In summary, we have discussed the direct superexchange coupling in an array of nanosized QD of *strong* ferromagnets. Such a coupling can be associated with the array structures in which the dots are abutted by an insulator or semiconductor. We have seen that the respective coupling constant decreases exponentially with increasing interdot separation distance. The superexchange coupling is expected to react sensitively to the temperature, since it is determined by the relation between the contributions coming from tunnel spin current (increasing with temperature) and the dot structure. Evidently, a variety of possibilities exist for the manipulation of the magnetic properties of such arrays.

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