

Electric field for tuning quantum entanglement in supported clusters

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Abstract

We show that quantum entanglement, nowadays so widely observed and used in a multitude of systems, can be traced in the atomic spins of metal clusters supported on metal surfaces. Most importantly, we show that it can be voluntarily altered with external electric fields. We use a combination of *ab initio* and model Heisenberg–Dirac–Van Vleck quantum spin Hamiltonian calculations to show, with the example of a prototype system (Mn dimers on Ag(001) surface), that, in an inherently unentangled system an electric field can ‘switch on’ the entanglement and significantly change its critical temperature parameter. The physical mechanism allowing such rigorous control of entanglement by an electric field is the field-induced change in the internal magnetic coupling of the supported nanostructure.

Keywords: quantum entanglement, electric field, tuning

(Some figures may appear in colour only in the online journal)

1. Introduction

In recent years, electric fields have become increasingly popular in the surface science community as the tool of choice, when it comes to: tuning the morphology and energetics of thin films and nano-islands [1, 2]; tailoring magnetic spins and their anisotropy/coupling in a reversible, controlled and, most importantly, local way [3–9]; or tuning spin dynamics [10] and exchange biases [11–13]. Recently, it has been suggested [14] that this might also affect such fascinating phenomena as quantum entanglement (QE) [15–17], nowadays so generally accepted and utilized in quantum optics for cryptography and dense coding.

While one is used to speaking of entanglement in the context of optical systems and experiments, QE does not only exist and play an important role in optical systems, such as bi-photons [18]. It can also be found on the nanoscale in solid state [19], [19–24] and molecular [25, 26] systems. For a system of spins, we can speak of entanglement, when the many-spin state cannot be represented as a product of single-spin states. In other words, entanglement is non-local correlation present in the system.

Condensed matter systems are especially interesting subjects for studying entanglement for several reasons. First and foremost, condensed matter is the basis of a sizeable portion

of today's research and development in electronics and information industries. Understanding QE in solids and nanostructures could open a completely new perspective on those research directions. Moreover, the source of QE of spins is the exchange interaction, which is known to be strong in condensed matter systems. While molecular magnets were shown to be in an entangled state at low temperatures, exchange interaction of their constituent parts does rarely exceed tens of meV [25, 26]. At the same time, exchange interaction in solids can reach hundreds and thousands of meV, which may help QE to survive at much harsher conditions [27–29].

In the case of solids and molecules, however, detecting QE can be tricky. Measuring microscopic quantum properties is not always easy or even possible. It was shown, however, that entanglement can also affect the macroscopic properties of solids [19, 30], making itself traceable by thermodynamic quantities such as magnetic susceptibility [31]. In the present paper we shall use this macroscopic ‘entanglement witness’ property to study entanglement in supported metallic clusters, which continuously attract the attention of the solid state community [32, 33], and the influence of external electric fields thereon. It shall be shown that quantum entanglement can be ‘switched on’ in an initially unentangled Mn dimer on a Ag(001) surface by applying an external field (EEF).

2. Methodology used

As has already been mentioned, in condensed matter systems the entanglement of atomic spins can be traced by the effect it has on the thermodynamic properties of the system, most notably so in the magnetic susceptibility [31]:

$$\chi = \left(\frac{\partial M}{\partial B} \right)_{H \rightarrow 0}. \quad (1)$$

While the magnetization of the system is an inherently local property, its magnetic susceptibility depends on both the local magnetism and non-local correlations. It has been shown [31] that for a system of N spins of length s in a separable state, the following inequality is true:

$$\chi \geq \frac{Ns}{kT}. \quad (2)$$

Thus any susceptibility lower than Ns/kT necessarily implies the presence of non-local correlations of spins of the system, or quantum entanglement thereof. One can define a quantity

$$EW = \chi \cdot \frac{kT}{Ns} - 1, \quad (3)$$

which, according to (2), is larger than, or equal to, zero for a system in a separable state. $EW < 0$, on the contrary, is a signature of entanglement in the system. EW is one of the class of macroscopic quantities called entanglement witnesses (EW) [34] which act as indicators and a sufficient condition of entanglement presence.

To access the susceptibility of the system theoretically, we first calculate, within the framework of density functional theory (DFT), the electronic and magnetic properties of the studied systems. Using those properties as an input, we use the irreducible tensor operator technique, as implemented in the MAGPACK code [35, 36], to evaluate the Heisenberg–Dirac–Van Vleck quantum spin Hamiltonian of the system limited to bilinear and Zeeman terms:

$$\hat{H}_{\text{spin}} = - \sum_{i,j} J_{ij} \hat{S}_i \hat{S}_j - g_s \mu_B \sum_i \hat{S}_i \hat{B}, \quad (4)$$

where J_{ij} is the Heisenberg exchange parameter between sites i and j ³, \hat{S}_i is the spin operator on the i -th site and \hat{B} is the magnetic field operator. The magnetic susceptibility χ can be derived from magnetization

$$M = NkT \frac{\partial \ln Z}{\partial B}, \quad (5)$$

where Z is the partition function of the system with Hamiltonian \hat{H}_{spin} . The latter is given by:

$$Z(H) = \text{Tr}(e^{-\hat{H}/kT}). \quad (6)$$

The DFT part of the procedure is carried out with the Vienna *Ab initio* Simulation Package (VASP) [37, 38]. In the calculations of Mn dimers on a Ag(001) surface we follow the established procedure described—see our previous paper by Negulyaev

³ Shall be extracted from first-principles calculations of exchange interaction energy of two adatoms E_{exch} as $J_{ij} = E_{\text{exch}}^{ij} / 2S^2$.

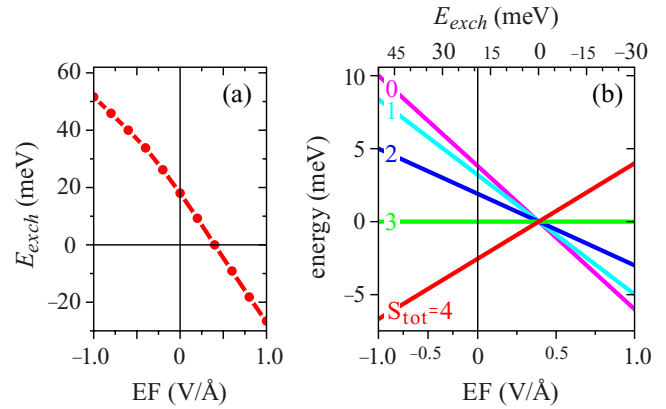


Figure 1. Exchange interaction (a) and spin spectrum (b) of a Mn dimer on a Ag(001) surface as a function of the external electric field (a). For details see the discussion in the text.

et al [39]. In short, the results are obtained using the projector augmented-wave technique [40] as implemented in VASP code [37, 38] within the local Ceperly–Alder exchange and correlation functional (LDA) [41], as parametrized in Perdew and Zunger [42]. Negulyaev *et al* have shown that gradient on-site Coulomb interaction corrected functionals yield qualitatively identical (marginally quantitatively deviating) results for the our prototype system. The geometry of the system was always relaxed self-consistently to correctly capture all aspects of surface and EEF-related physics.

3. Tuning quantum entanglement with electric field

As a model system we shall take Mn dimers on a Ag(001) surface. The reason for that choice is the exchange interaction strength between Mn atoms in that system, which, being ferromagnetic with a strength of ~ 18 meV, is large enough to allow for robust magnetism in a wide temperature range yet still small enough to be easily changeable by external influences. This requirement, however, does not strongly limit the generality of obtained conclusions, save through the necessity to scale the amplitude of external fields with the internal exchange strength in the system.

The principal parameters governing the entanglement in the system are the interaction strengths between the individual parts. For the case of atomic spin entanglement this shall be the exchange interaction, which is reflected in the Heisenberg–Dirac–Van Vleck quantum spin Hamiltonian discussed above. In our case we shall also be interested in the change of those parameters under the external electric field influence. To obtain those parameters we turn to the DFT calculation, which has been shown to nicely capture the physics of our model system [39].

A compact Mn dimer on a Ag(001) surface without any external influences is coupled ferromagnetically with an exchange energy of $E_{\text{exch}} \approx 18$ meV figure 1(a). Magnetic moments of Mn are $\mu_{\text{Mn}} = 3.9 \mu_B$ (which gives us a spin of $S = 4/2 = 2$ for the quantum Hamiltonian in equation (4)). As was shown by Negulyaev *et al*, application of an external electric field, via the spin dependent screening of the field by

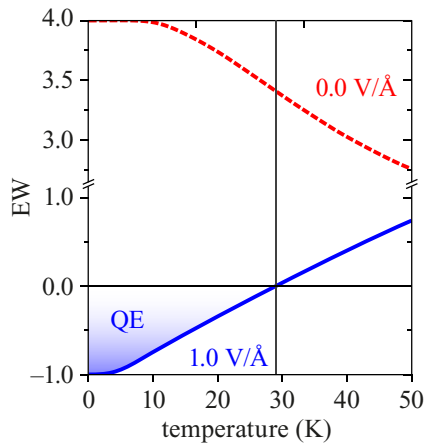


Figure 2. Entanglement strength of a Mn dimer without an external electric field applied (red dashed) and in an external field of 1.0 V/\AA (blue solid).

surface and dimer electrons, can change the exchange coupling of the dimer. In figure 1 we plot the dependence of the exchange interaction between Mn atoms on the strength of the applied EEF strength. If an electric field of 1 V/\AA is applied in the direction away from the surface the exchange coupling between Mn atoms is increased from 9 to over 50 meV ($EF < 0$ in figure 1(a)). If an opposite field of 1 V/\AA ⁴ is applied the dimer changes its coupling to an antiferromagnetic one, with $E_{\text{exch}} \approx -24 \text{ meV}$. Since E_{exch} is a bilinear coefficient in our spin Hamiltonian, the change in the coupling will have a linear effect on the spin structure of the dimer (figure 1(b)). As a system with $2 \times 4 = 8$ spins with non-zero exchange coupling our dimer shall have five distinct spin states. Here we neglect the effect of anisotropy, which would lift the internal degeneracy of each of those states, since this effect is of the second order and would not affect the conclusions of the present study. The spin spectrum of our dimer with no EEF applied is shown in figure 1(b) (intersections of the lines in the graph with $EF = 0$). The spin ground state is the ferromagnetic one with $S_{\text{tot}} = 4$ and lower-spin states lying 3–8 meV higher. In negative electric fields the high-spin ground state is retained and the spectrum stretches, with lower-spin states now lying 7–17 meV higher in energy. In positive electric fields the situation changes dramatically. At about 0.4 V/\AA , the exchange interaction between the two Mn atoms is fully quenched and the spin spectrum becomes degenerate. In higher positive fields the interaction becomes antiferromagnetic, which is reflected in the $S_{\text{tot}} = 0$ spin state now being the lowest energy one. At $EF = 1 \text{ V/\AA}$ the zero-total-spin state is 6–10 meV lower than the higher-spin states.

We are, however, not interested here in the fine details of the spin-spectra—which are the subject of trivial quantum mechanics—but rather in the thermodynamic properties of the system which can be deduced from those spectra, as described above. The red dashed curve in figure 2 shows the temperature dependent entanglement strengths for the Mn dimer in

zero EEF (calculated via the magnetic susceptibility). In the absence of EEF the entanglement witness (EW) exhibits values in the range 2.75–4 and a temperature range up to 50 K, giving, according to equation (3), no hint of quantum entanglement presence. At $EF = +1.0 \text{ V/\AA}$ (blue solid curve in figure 2), however, the EW is negative, up to temperatures of about 30 K. According to equation (3) this is a conclusive sign of the presence of entanglement in the system. At intermediate EF strengths (down to 0.4 V/\AA) the entanglement is always present in the system. Its critical temperature, however, decreases, until at $EF = 0.4 \text{ V/\AA}$ it disappears altogether. This is quite consistent with the notion that entanglement can only survive in an antiferromagnetic system [43–47]. This makes the physics of entanglement control by EEF a two-step process. The field changes the exchange interaction between constituent parts of the system, which in turn is directly reflected in the change of the entanglement. For the sake of completeness it has to be noted that, in general, the entanglement is affected by the coupling of the magnetic system on the surface to the bulk ‘bath’ of electrons and nuclei. This coupling in itself deserves further study. For our prototype system, however, it is known that this coupling does not destroy the entanglement, since such systems have been shown to exist in an entangled ground state experimentally [47].

4. Conclusion

Thus, our calculations suggest that, by just exposing the surface-supported nanostructure to EEF it seems to be possible to switch the quantum entanglement on an off, which should be easily realized in a normal scanning probe (STM, FIM) experiment. This opens up new ways of looking at the effect of QE on the nanoscale physics of surfaces.

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⁴ In the following, compliant with the notation used in [39], we shall speak of positive electric fields if the field vector is directed toward the surface and of negative ones in the opposite case.

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