Voltage-Controlled Surface Magnetization of Itinerant Ferromagnet $\text{Ni}_{1-x}\text{Cu}_x$

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We argue that surface magnetization of a metallic ferromagnet can be turned on and off isothermally by an applied voltage. For this, the material’s electron subsystem must be close enough to the boundary between para- and ferromagnetic regions on the electron density scale. For the 3d series, the boundary is between Ni and Cu, which makes their alloy a primary candidate. Using Ginzburg-Landau functional, which we build from Ni$_{1-x}$Cu$_x$ empirical properties, ab-initio parameters of Ni and Cu, and orbital-free LSDA, we show that the proposed effect is experimentally observable.

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Further evolution of magneto-electronics [1] depends highly on the availability of materials, in which local magnetization can be turned on and off isothermally by an electric voltage. The hopes to achieve this are mainly laid on the dilute magnetic semiconductors (DMS) [2], in which the effect was recently demonstrated experimentally [3]. The voltage-controlled ferromagnetic ordering in DMS relies on the virtue of doped semiconductors to allow external variation of the free-carrier spatial density within the semiconductor depletion layer, typically measured in dozens of nanometers. The voltage variation of the high electron density in metals is possible only within the atomic size Thomas-Fermi (TF) surface layer. As a result, the voltage controlled ferromagnetism in a metal has not been considered a possibility lately.

We argue that by capacitively charging a metallic ferromagnet one can drive the surface electron subsystem in and out of its ferromagnetic state. At this, the electron system of the metal has to be paramagnetic at the device operation temperature (room temperature, $T_0$), but close enough to the ferromagnetic state on the temperature and/or electron density scales. The proximity of the ferromagnetic transition will play a twofold role: (i) the capacitive change in the electron density is relatively small, so that the transition has to be sufficiently close in order to reach the ferromagnetic region with reasonable voltages, and (ii) due to the critical collective spin correlations, the spin correlation length grows infinitely as one approaches the transition point. Consequently, even though the injected carriers are spatially limited to the TF-layer, the system must develop a much wider surface magnetization profile. In this paper we investigate the proposed possibility in Ni$_{1-x}$Cu$_x$.

The nature of ferromagnetism lies in the competition between the kinetic energy and the exchange interaction. The kinetic energy of the spatial quantization tends to equalize the numbers of spin up and down electrons by shifting the fermionic antisymmetry into the spin sector of the many-body wavefunction. In turn, the exchange interaction does the opposite, struggling to unbalance the up and down spins. For itinerant ferromagnets, - the subclass of materials elemental ferromagnets belong to, - the outcome of this competition can be predicted from the Stoner criterion [4]. According to the Stoner criterion, ferromagnets possess a high density of states (DOS) at the (paramagnetic) chemical potential. In the 3d series, the high DOS is provided by the 3d band on the back-}

![FIG. 1: (a) Schematics showing the paramagnetic chemical potentials of the elemental ferromagnets on the kinetic energy, $\epsilon$, and the spatial electron density, $\rho_0$ (outer shell electrons per atom), scales. The inset is the magnified dashed area, which shows the graphic solution for Eqs. (3) determining the spin density, $s_0$, the paramagnetic and ferromagnetic chemical potentials, $\mu_P$ and $\mu$, and the spin up and down kinetic Fermi energies, $\varepsilon_{F+}^0$, as functions of $\rho_0$ for pure Ni $\rho_0 = \rho_{0Ni}$. (b)-(d) The experimentally observed properties of Ni$_{1-x}$Cu$_x$: (b) zero-temperature magnetization vs. $\rho_0$; (c) Curie temperature vs. $\rho_0$; (d) magnetization vs. temperature, $T$.](cond-mat.mrl-sci)
stitional solid solutions at all compositions. Consequently, Ni$_3$xCu$_x$ can be thought of as an all-the-same structure solid, with the equilibrium electron density varying linearly with $x$, $\rho_0 = \rho_{Ni} + x/\alpha^3$. Within this picture, $x$ and $\rho_0$ are interchangeable.

Other experimentally observed properties of the alloy \cite{9} can be well approximated as: (see Fig[1]-d) the linear dependence of the zero temperature spin density, $s_0(\rho_0, 0)$, and the Curie temperature, $T_C(\rho_0)$, on $\rho_0$, and the Landau dependence of the spin density, $s_0(\rho_0, T)$, on the temperature, $T$:

$$2s_0(\rho_0, 0)/\gamma = (\rho_{sd} - \rho_0)\theta(\rho_{sd} - \rho_0), \quad (1a)$$

$$T_C(\rho_0)/\kappa = (\rho_{sd} - \rho_0)\theta(\rho_{sd} - \rho_0), \quad (1b)$$

$$s_0(\rho_0, T)/s_0(\rho_0, 0) = (1 - T)^{1/2}\theta(\rho_{sd} - \rho_0), \quad (1c)$$

where $T = T/T_C(\rho_0)$, $\theta$ is the Heaviside step-function, $\rho_{sd} = \rho_{Ni} + 0.53/\alpha^3$ is the position of the 3d band edge on the density scale, and $\gamma$ and $\kappa$ are the slopes of the $2s_0(\rho_0, 0)$ and $T_C(\rho_0)$ lines. For nickel $2s_0(\rho_{Ni}, 0) = 0.06/\alpha^3$ and $T_C(\rho_{Ni}) = 627K$, so that $\gamma \approx 1.25$ and $\kappa \approx 1.2 \times 10^3\text{K}^2$.

A rather accurate quantitative look at the itinerant ferromagnetism can be taken via the Stoner approximation \cite{1}. In a homogeneous system case, the zero-temperature energy per volume is the sum of the kinetic and exchange parts:

$$E_{K\times}(\rho_0, s_0) = \sum_{\nu} K(\rho_0^{1\nu}) - J s_0^2, \quad (2)$$

where $\rho_0^{1\nu} = \rho_0/2 \pm s_0$ are the up and down spin densities, $J$ is the Heisenberg exchange interaction constant \cite{[1]}, and the kinetic energy is defined as $K(\rho_0^{1\nu}) = \int_{-\infty}^{\infty} \nu(\varepsilon)\varepsilon d\varepsilon$, with $\varepsilon^{1\nu} \equiv \varepsilon F(\rho_0^{1\nu})$ being the kinetic spin up and down Fermi energies, and $\nu$ being the DOS (per spin). \cite{[1]} The function $\varepsilon F$ defines the one-to-one correspondence between the (spin) density and the position of the Fermi energy on the kinetic energy scale. It can be defined via its inverse function as $\rho = \int_{-\infty}^{\varepsilon F} \nu(\varepsilon) d\varepsilon$, so that $\partial \varepsilon F/\partial \rho = \nu(\varepsilon F)^{-1}$ and $\partial \varepsilon F/\partial \rho = \varepsilon F$, i.e., $\varepsilon F$ and $\rho$ are the Legendre conjugates with respect to $K$.

For the ground state, vanishing variations of $E_{K\times}$ in $\rho_0$ and $s_0$ yield the equations determining the ground state’s spin density and the chemical potential as functions of $\rho_0$:

$$2s_0(\rho_0, 0)f = \varepsilon F - \varepsilon F, \quad \mu(\rho_0) = (\varepsilon F + \varepsilon F)/2. \quad (3)$$

To get \cite{13} from Eqs.\cite{13} (the graphic solution for pure Ni is given in the inset of Fig[1]) the DOS has to be of the following simple form ($\varepsilon_{3d}$ is the 3d band edge):

$$\nu(\varepsilon) = \nu_{Ni}(\varepsilon_{3d} - \varepsilon) + \nu_{Cu}(\varepsilon - \varepsilon_{3d}).$$

Then, $\gamma = (\nu_{Cu} - \nu_{Ni}^{-1})/[(\nu_{Ni}^{-1} + \nu_{Cu}^{-1} - 2J)$, and

$$\mu(\rho_0) = \mu_F(\rho_0) - \Delta \mu(\rho_0), \quad (4)$$

where

$$\mu_F(\rho_0) = \varepsilon_{3d} + 1/2(\rho_0 - \rho_{3d}) \times \left\{ \begin{array}{ll} \varepsilon_{Ni}^{-1}, & \rho_0 < \rho_{3d}, \\ \varepsilon_{Cu}^{-1}, & \rho_0 > \rho_{3d}, \end{array} \right. \quad (5)$$

is the chemical potential, corresponding to the paramagnetic solution ($s_0 = 0$, $\mu_F = \mu = \varepsilon F = \varepsilon_F(\rho_0/2)$), and

$$\Delta \mu(\rho_0) = (2\Delta \nu)^{-1}(\rho_0 - \rho_{3d})\theta(\rho_{3d} - \rho_0), \quad (6)$$

is the chemical potential shift due to the switching from the paramagnetic to the ferromagnetic state, with $\Delta \nu^{-1} = \gamma(J - \nu_{Ni}^{-1})$. \cite{14} Numerical estimates for the parameters of the model can be obtained by fitting $\mu_F(\rho_0, \nu_{Ni}) - \varepsilon_{3d}$ from Eq.\cite{5} and the exchange splitting in Ni, $2s_0(\rho_{Ni})f$, with their ab-initio values \cite{2}: $\nu_{Cu,Ni} = 6.7, 0.79eV\text{a}^3$, $J = 1.3eV\text{a}^3$, and $\Delta \nu^{-1} = 0.61eV\text{a}^3$.

For non-zero temperatures, the free energy of a homogeneous system can be assumed a function of $\rho_0$, $s_0$, and $T$. Its spatial density can be given as the sum of the paramagnetic and ferromagnetic (Landau) parts:

$$F_{K\times}(\rho_0, s_0, T) = F_P(\rho_0, T) + \Delta F(\rho_0, s_0, T). \quad (7a)$$

$F_P$ is determined mostly by the crystal structure. For not very high temperatures, such that all the structural phase transitions are far on the $(T, \rho)$-plane, $F_P$ can be assumed temperature independent and consequently equal to the paramagnetic part of the zero-temperature energy, so that $\partial \rho_0 F_P = \mu_P(\rho_0)$.

The ferromagnetic properties of the alloy (Eqs.\cite{1}) determine the Landau part of the free energy up to an unknown factor $f$:

$$\Delta F = f \left(2(\rho_0 - \rho_{3d})^2(\tilde{T} - 1)(2s_0/\gamma)^2 + (2s_0/\gamma)^4\right). \quad (7b)$$

The para-to-ferro shift of the chemical potential provided by $\Delta F$ is:

$$\Delta \mu(\rho_0, T) = -\partial \rho_0 \left(f(\rho_0 - \rho_{3d})^4(\tilde{T} - 1)^2 \theta(1 - \tilde{T})\right). \quad (7c)$$

The comparison of the previous equation at $T = 0$ with Eq.\cite{1}, together with the assumption that $f$ is also temperature independent, uniquely defines the function $f$:

$$f(\rho_0) = (\rho_0 - \rho_{3d})^{-2}/(4\Delta \nu). \quad (7d)$$

Accordingly, $\Delta \mu(\rho_0, T) = \Delta \mu(\rho_0)(1 - \tilde{T})\theta(1 - \tilde{T})$, with $\Delta \mu(\rho_0)$ from Eq.\cite{6} (see Fig[2]).

Now, in the spirit of the orbital-free local spin density approximation (LSDA) we turn to the inhomogeneous case by letting the electron and the spin densities spatially vary: $\rho_0 \to \rho(x), s_0 \to s(x)$. Capacitively charging the conductor we make it an open system, which is governed by the $\Omega$-functional:

$$\Omega = \int d^3x (W + \mathcal{F}_{K\times} - \mu_0 \rho + e\varphi(\rho - \rho_0)/2). \quad (8a)$$
FIG. 2: (a) The chemical potential provided by the proposed model vs. temperature (b) The area on the \((T, \rho)\)-plane under consideration (dashed rectangle). The parameters \(\Delta \rho, \rho_C, \Delta \rho, \rho, \) and \(\delta T\) are introduced in the text. The arrow symbolizes the effect of the (positive) bias on the surface electron subsystem.

(c) The widths of the spatial profiles of the magnetization and the density are determined by the spin and density correlation lengths (Eqs.(9)), and \(\lambda_s \gg \lambda_p\). (d) The surface magnetization, \(m_s\), vs. surface charge, \(\sigma\). (insets) The threshold charging, \(\sigma_{th}\), and the characteristic magnetization, \(m^*\), vs. the alloy’s detuning from the transition, \(\delta T\).

Here \(F_{KX}\) is defined by Eqs.(7), \(\mu_0 \equiv \mu(\rho_0, T_0)\), \(\varphi = e \int d^3x' (\rho(x') - \rho_0)/|x - x'|\) is the direct interaction potential with \(e\) being the electron charge, and the nonlocality of the kinetic energy functional is accounted for by the \(\alpha\)-von Weiszacker term \(W = \alpha h^2/(8m^*) \sum_{i}|\nabla \rho|_{1i}^{2}/\rho_h^{2i}\), with \(m^*\) being the effective mass of the 3d holes and with \(\rho_h^{2i} \equiv \rho_{sd}/2 - \rho_{s1}^{1i}\) being the spin up and down hole densities. We adopt \(\alpha = 1/9\) - the case when the \(\alpha\)-von Weiszacker approximation is asymptotically correct for long wavelengths \(\delta\), i.e., the domain of the applicability of the Ginzburg-Landau (GL) theory below.

As we already mentioned, the TF layer’s spatial electron density variation, due to the injection of the carriers, is relatively small. Therefore, to be able to reach the ferromagnetic region from an initially paramagnetic state, we need an alloy which is close to its transition at \(T_0\):

\(T_C \equiv T_C(\rho_0) < T_0, \delta T = T_0 - T_C \ll T_0\) (see Fig.2), so that \(\delta \rho = \rho_0 - \rho_C = \delta T/\kappa \ll \Delta \rho, \) where \(\rho_C = \rho_{sd} - \Delta \rho\) and \(\Delta \rho = T_0/\kappa \approx 0.25/a^3\). Having chosen the composition, we can focus on a small area on \((\rho, T)\) plane, such that \(|\rho - \rho_C| \ll \Delta \rho\). Within the area, \(\rho_h^{2i} \approx \Delta \rho/2\), and the von Weiszacker term simplifies as

\[ W \approx \alpha h^2/(8m^*) \int d^3x \left( \nabla \rho \right)^2/\Delta \rho. \]  

In the Thomas-Fermi picture of a Fermi liquid, which the von Weiszacker correction relies on, the DOS at the chemical potential is given as \(\nu_{TF} = k_F m^*/(2\pi^2 h^2)\), where \(k_F = (3\pi^2 \Delta \rho)^{1/3}\) is the Fermi wave-vector. To make the von Weiszacker term energy-wise consistent with the rest of the \(\Omega\)-potential, obtained by the physical arguments different from the orbital-free LSDA, we can require that \(\nu_{TF} = \nu_{Ni}, \) or \(m^* \approx 8.2 m_0\). This value of the effective mass lies well within the wide range of the 3d sub-bands’ effective masses, which vary from several \(m_0\) to almost \(30 m_0\) (see, e.g., Ref.[14]).

Even though the orbital-free LSDA has proven successful in some atomic-scale non-homogeneous problems, in our case any atomic-scale results obtained from Eqs.8 would have a rather qualitative character. Indeed, the very notion of the composition of a solid solution is well defined only on scales larger than the lattice constant. Furthermore, the microscopic properties of the electron subsystem of the TF-layer differ from those of the bulk and are interface-material dependent. The \(\Omega\)-potential \(\delta\), however, can be used to find the characteristic distances of the spatial variations of \(\rho\) and \(s\), which are given by the the density and the spin correlation lengths, \(\lambda_p\) and \(\lambda_s\). Omitting intermediate derivations, the correlation lengths obtained from (8) in a linear response manner are:

\[ \lambda_p = 2^{-1/2} \lambda_{TF} \sqrt{1 + \left(1 - k_F/(\pi^2 \Delta \rho \lambda_{TF}^2) \right)} \]  

\[ \lambda_s = \lambda_p (\Lambda/\delta T)^{1/2}. \]  

Here the Thomas-Fermi radius \(\lambda_{TF} = (4\pi e^2/2\gamma_{Ni})^{-1/2}\) and \(\lambda = b k_F^2 \Delta \nu/(4m^* \lambda_{TF}^2)\).

The numerical estimations lead to \(\Lambda \approx 7 \times 10^2\) K. In the vicinity of the transition, the inequality \(\delta T \ll \Lambda\) is well satisfied, so that the spin and the density scales separate, \(\lambda_p \gg \lambda_s\) (see Eq.(9b)). Thus, we arrived at a typical picture of the critical phenomena theory. The spin density is a "ready-to-condense" soft variable, behavior of which is governed by the large-scale low-energy GL functional (the \(s\)-dependent part of \(\delta\)), which in the \((T, \rho)\)-area under consideration has the following form:

\[ F_{GL} = \int d^3x \left(A(\nabla s)^2 + b(\rho - \rho_C)s^2 + C s^4/2\right), \]  

with \(b = 2/(\gamma^2 \Delta \rho \Delta \nu), \) \(A = k_b, C = 4b/(\gamma^2 \Delta \rho)\). The high-energy stiff variable, \(\rho,\) plays a guiding role via the \(\rho\)-dependence of the effective chemical potential for the magnetization (the overall coefficient in \(s^2\)-term of Eq.(10), \(B = b(\rho - \rho_C)\)). The feedback action of the soft variable, \(s\), on the stiff variable, \(\rho,\) is weak and/or unimportant.

The proposed form of the near-critical \(s-\rho\) coupling can be obtained on a more general footing. Indeed, Taylor expanding \(B\) around the transition point, at which \(B(T_0, \rho_0) = 0\), and noticing that in itinerant ferromagnets the \(s-\rho\) coupling can only be local (especially on the \(\lambda_s\)-scale), we arrive at \(B \approx b(\rho - \rho_0) + b'(T_0 - T_C) \equiv

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...b(ρ - ρc), with ρc = ρ0 - δρ, δρ = (T0 - Tc)/κ, κ ≡ b/β'.
For quantitative studies, however, it is crucial to possess reliable values of the three material-specific parameters
for Eq. (10). The way the parameters for Ni1-xCux are derived in this Letter can now be summarized as follows:
A is obtained from the orbital-free LSDA considerations (Eq. (8)): the mutual relation between b and C - from the
empirical properties of the alloy and from the Landau theory for the II-order phase transitions (Eq. (75));
the overall energy factor for b and C - from previous ab-initio parameters of Ni and Cu and from the Stoner
type of itinerant ferromagnetism (Eqs. (1) and (7c)).

On the λ-scale, the microscopic effects are scaled out. The magnetization is ignorant to the microscopic details of the
electron density profile, e.g., the Friedel oscillations. The injected carriers' spatial density in the TF
layer, with a width of order of λ, can be assumed infinitely narrow. Therefore, a positively biased surface
can be represented as a 1D semi-infinite solution (z < 0, see Fig. (2)) of the GL Euler equation with ρ(x) → ρ0 and
with the following boundary condition imposed by the form of the s-ρ coupling: λsΩs ln s|x|=0 = σ/σtr, where
σs = f → ∞(ρ(z) - ρ0)dz is the surface density of the excess holes and σtr = λsδρ is the threshold charging. The solution is
σ = s*(sinh(coth−1(σ/σtr) - z/λs))−1θ(σ - σtr), where s* = (2bδρ/C)1/2. The magnetic response of the surface
can be characterized by the surface density of Borh magnetons, m = ρsΩs ln s|x|=0 2s(z)dz (see Fig. (2)): m = m* log(σ/σtr) + ((σ/σtr)2 - 1)1/2θ(σ - σtr), where m* = (2A/C)1/2. The characteristic magnetization, m*, is
independent of δT, so that the effective magnetic susceptibility to the charging, m*/σtr ∝ δT−1/2, grows infinitely as one approaches the transition point (see Fig. 2 inset).

In reality, the surface charge is limited by the breakdown of the insulating interface material. For SiO2, the breakdown electric field $E_{\text{SiO}_2} ≈ 10^7 \text{V/cm}$, which corresponds to the following charging $\sigma_{\text{SiO}_2} = eE_{\text{SiO}_2}/(4\pi\epsilon) \approx 2.5 \times 10^{13} \text{cm}^{-2}$, where $\epsilon = 4.5$ is the SiO2 dielectric constant. According to our calculations, for the Ni1-xCux/SiO2 interface, which is $\delta T = 5$K away from
the transition, the threshold charging $\sigma_{\text{tr}} ≈ 0.15 \times \sigma_{\text{SiO}_2}$, and on the edge of the semiconductor breakdown ($\sigma < \sigma_{\text{SiO}_2}$) the surface magnetization is $m \approx 1.3 \times 10^{14} \mu_\text{B/cm}^2$.

The magnetic properties of the surface are in fact different from the bulk on their own, without charging. This difference is modeled in the surface phase transition
theory as a delta-functional jump of the local Curie temperature at the surface of the semi-infinite system (see, e.g., Ref [11] and Refs. therein). Within the proposed approach the jump can be taken into account as an intrinsic shift of the charging density $\sigma \rightarrow \sigma_{\text{int}} + \sigma$ (the cases $\sigma_{\text{int}} \geq 0$ are known respectively as ordinary and extraordinary transitions). A reliable estimate for the material
and interface-dependent $\sigma_{\text{int}}$ can only be obtained from ab-initio studies or experimental data.

Another issue is the quantum fluctuations, due to which the critical exponent of the spin correlation length
must acquire the renormalization group correction: $\lambda_s \rightarrow \lambda_s^* \approx \rho_s(T_c/\delta T)^1/2 + \delta$ (δ ≈ 0.14 for, e.g., the d = 4 - ε approximation [12]). Accordingly, the exponential tail of the magnetization profile will be elongated and the magnetic response should acquire an enhancement factor $\propto (T_c/\delta T)^\delta$. On the other hand, the intrinsic substitutional irregularity in a solid solution must shorten $\lambda_s$ due to the Anderson localization mechanism. Near the transition, the quantum fluctuations' effect is dominant and in reality the magnetic response of the surface is stronger than the one obtained here on the mean-field level.

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ferromagnetic region, however, the paramagnetic solution is unstable with respect to fluctuations in $s_0$ (the Stoner criterion): $\frac{\partial^2 \mathcal{E}_{\mathcal{K},X}}{\partial s_0^2} \bigg|_{s_0=0} < 0$. 