Voltage sensitivity of Curie temperature in ultrathin metallic films

Igor V. Ovchinnikov* and Kang L. Wang†
Department of Electrical Engineering, University of California at Los Angeles, Los Angeles, California 90095-1594, USA
(Received 9 May 2009; revised manuscript received 23 June 2009; published 20 July 2009)

In ferromagnetic metals, the surface enhancement of magnetization and the surface magnetic anisotropy are known to be coupled to the surface charge, σ, controllable by bias voltage. In the critical region, these surface parameters are nonlocal relevant fields, from which the Curie temperature of an ultrathin film, T_C, should borrow the dependence on σ. Studying this phenomenon in the $3d$ transition metals, we find a significant sensitivity of T_C to the charging. Our results suggest the possibility of the near-critical voltage control of the very existence of magnetization in ultrathin metallic films, which can be useful for the future development of spintronics devices.

DOI: 10.1103/PhysRevB.80.012405 PACS number(s): 75.70.Rf, 85.75.−d, 85.70.Ay

Magnetoelectronics is rightfully considered one of the most likely future alternatives to the semiconductor technology. Metallic ferromagnets as a material basis are especially promising due to the robustness of the metallic ferromagnetic order, which persists even well-above room temperature, and the high conductance and/or carrier density, which allows for going further in the miniaturization.[1] The energy-dissipation issues make the manipulation of the magnetization by voltage[2,3] more advantageous over that by electric currents.[4] These are the reasons why among modern directions of frontier research is the study of the possibility to control by voltage the magnetization of tiny metallic parts,[5–13] which are typically of a film geometry.

The voltage control of the ferromagnetic transition in nanometer scale metallic films may seem impossible due to the screening of the electric field within a surface atomic layer. This first-sight conclusion, however, is not correct. The point is that the spatial width of the influence of the surface charge, σ, on the magnitude of the magnetization (soft sector) is determined by the soft-sector correlation length, ξ_S,[13] and *not* by the electric field penetration depth. In turn, ξ_S grows infinitely in the critical region, and sufficiently close to the transition, when ξ_S becomes on the order of the film width, σ couples to the soft sector across the entire film. This mechanism of the critical nonlocality is universal and, in particular, is responsible for the recently observed bulklike behavior of surface quasiparticles in near-critical Mott insulators.[14] In our case, it should make the Curie temperature, T_C, dependent on σ. The $T_C(\sigma)$ dependence assumes the possibility to isothermally manipulate by voltage the very existence of the metallic magnetization, which is quite remarkable from the fundamental point of view and may be of use in the future development of the low-power magnetoelectronics devices. Studying this phenomenon, we find that the T_C of $3d$ transition metals ultrathin films is considerably sensitive to the charging.

Our strategy to determine the $T_C(\sigma)$ dependence is rather straightforward. The coupling of σ to the soft sector at low temperatures is known as the surface magnetoelectric effect (SMEE),[6,7] represented graphically in Fig. 1(a). At $T \ll T_C$, the mean-field theory is applicable and the SMEE coefficients directly provide the information on the σ dependence of surface parameters of the Ginzburg-Landau (GL) functional. To the best of our knowledge, the only SMEE coefficients for the $3d$ transition metals available in the literature are those of Ref. 7 for Ni[001], Co[0001], and Fe[001] free surfaces, to which we limit our consideration.

In the critical region, in which the SMEE develops into the $T_C(\sigma)$ dependence [see Fig. 1(b)], the fluctuations are important and it is necessary to go beyond the mean-field theory. Accordingly, using previous experimental and *ab initio* data for $T=0$ we construct the GL functional, perform the mean-field analysis, and then "dress" the mean-field results by the fluctuational corrections.

Only relevant operators should be kept in the "minimal" GL functional to be used in the critical region. They are known to be $\sim\partial^2\boldsymbol{m}^2$, $\boldsymbol{m}^2$, and $\boldsymbol{m}^4$, where $\boldsymbol{m}\equiv\boldsymbol{M}/M_0$ is the dimensionless vector magnetization and M_0 is the magnetization at $T=0$. The explicit T dependence can be dropped for all the operators but one—the "mass" term of the spherically symmetric Landau potential $U_L=(\boldsymbol{m}^2+\tau)^2/4$, where $\tau=(T-T_C^b)/T_C^b$ with T_C^b being the bulk transition temperature. The "kinetic" nonlocal term can always be brought to the spherically symmetric form, $(\partial\boldsymbol{m})^2\equiv\Sigma_{ij}(\partial\boldsymbol{m}_j/\partial\boldsymbol{x}_i)^2$, by the appropriate transformation of the spatial coordinates. The cubic anisotropy, being of the fourth order, $\propto\Sigma_i\boldsymbol{m}_i^4$, may also seem

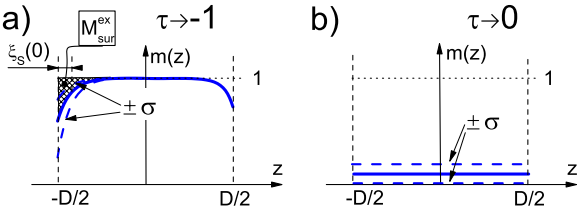

FIG. 1. (Color online) (a) The dimensionless magnetization, $m(z)$, as a function of the spatial coordinate, z, across the film at low temperature, $\tau=(T-T_C^b)/T_C^b\to-1$, with T_C^b being the bulk transition temperature. D is the film width. The difference in the magnetic properties of the surface and the bulk atoms, represented by Eq. (3), results in the excess surface magnetization, $M_{\mathrm{sur}}^{\mathrm{ex}}$, which is shown as the shaded area and the spatial width of which is determined by the $T=0$ soft-sector correlation length, $\xi_S(0)$. The charge, σ, injected into the (left) surface approximately linearly varies $M_{\mathrm{sur}}^{\mathrm{ex}}$, which constitutes the low-temperature SMEE. (b) In the critical region, $\tau\to0$ and $\xi_S>D$, the coupling of σ to the soft sector enables the ultimate manipulation of the very existence of the magnetization across the film.

 ©2009 The American Physical Society

relevant. However, for the number of components of the order parameter, $N<4$, the system should flow to the O(N) Heisenberg stable point.[15] Thus the cubic anisotropy can be most likely dropped. The above reasonings limit the volume part of the "minimal" GL functional to the following

$$\mathcal{F}_V = T_C^b \Lambda^{-3} \int_V [\Lambda^2 (\partial \boldsymbol{m})^2/2 + U_L + U_A]. \tag{1}$$

Here the *dimensional* T_C^b and the real-space cutoff, Λ, are the primary energy and length scales, over which all the energies and lengths in the problem lose their dimensions. Note, that all the variables and coefficients have to be brought to the dimensionless form in order to be able to appropriately shift the power laws later, while accounting for fluctuations.

Adopting the convention of Ref. 16 that positive anisotropy prefers the out-of-plane order, the local potential of the uniaxial bulk anisotropy in Eq. (1) can be given as

$$U_A = -K \boldsymbol{m} \boldsymbol{A} \boldsymbol{m}/2, \tag{2}$$

where the traceless $\boldsymbol{A}_{ij} = \delta_{iz}\delta_{jz} - \delta_{ij}/3$ (z is the coordinate perpendicular to the film) and the bulk anisotropy $K = K_u - q^2$ consists of the magnetocrystalline contribution, K_u, and the magnetic dipole-dipole "shape" contribution represented by the parameter called quality, q.[16]

The "minimal" GL functional is the sum of the volume part Eq. (1) and the relevant surface-integral terms:[17]

$$\mathcal{F}_S = T_C^b \Lambda^{-2} \sum_\alpha \int_{z=z_\alpha} (c_\alpha \boldsymbol{m}^2 - k_\alpha \boldsymbol{m} \boldsymbol{A} \boldsymbol{m})/2, \tag{3}$$

where $z_\alpha = (-1)^\alpha D/2$ are the z coordinates of the left ($\alpha=1$) and right ($\alpha=2$) surfaces, and c_α and k_α are the surface enhancement and surface anisotropy coefficients.

The bulk GL functional parameters relate to the experimentally obtained "exchange" constant, A, the bulk uniaxial anisotropy constant, K_1, and the $T=0$ magnetization, all reviewed in Ref. 18, as $\Lambda = T_C^b/(2A) \approx (3.71, 3.04, 4.03)$ Å, $K_u = 2\Lambda^3 K_1 / T_C^b \approx (0, 1.2, 0) \times 10^{-3}$, and $q^2 = \mu_0 M_0^2 \Lambda^3 / T_C^b \approx (1.8, 3.7, 16) \times 10^{-3}$. Here and in the following the triples of the numerical values correspond, respectively, to Ni[001], Cu[0001], and Fe[001]. The surface anisotropy can be expressed via K_s of Ref. 16 as $k = 2\Lambda^2 K_s / T_C^b \approx (-1.4, -0.26, 2.1) \times 10^{-2}$.

The difference in the magnetization of the surface and the bulk atoms at $T=0$, δm_{sur} (in Bohr magnetons, μ_B) is known from *ab initio* studies and reviewed in Ref. 18.[19] The excess surface magnetization [see Fig. 1(a)] is given as $M_{\mathrm{sur}}^{\mathrm{ex}} = \mu_B \delta m_{\mathrm{sur}} s$, where s is the area density of surface atoms.[20] From the GL functional formalism at $T=0$ and for $|c| \lesssim 1$ the mean-field $M_{\mathrm{sur}}^{\mathrm{ex,MF}} \approx (-c) M_0 \xi_S(0)$, where the "width" of the excess magnetization is given by the $T=0$ correlation length, $\xi_S(0) = \Lambda/\sqrt{2}$. For all the three materials $\xi_S(0)$ is smaller than the lattice constant and $M_{\mathrm{sur}}^{\mathrm{ex,MF}}$ can be attributed to the surface atoms only so that $M_{\mathrm{sur}}^{\mathrm{ex,MF}} \approx M_{\mathrm{sur}}^{\mathrm{ex}}$ or $c \approx -\mu_B \delta m_{\mathrm{sur}} s / [M_0 \xi_S(0)] \approx (-0.11, -0.04, -0.16)$.

In Ref. 13 it was suggested that σ couples to the soft sector via the surface enhancement: $c \to c + \alpha_c \sigma$, where α_c is the surface enhancement sensitivity. This assumption is in accord with the SMEE at $T=0$,[6,7] in which $M_{\mathrm{sur}}^{\mathrm{ex}}$ varies linearly with σ. The results of Ref. 7 also suggest the linear coupling of σ to the surface anisotropy: $k \to k + \alpha_k \sigma$. The sensitivities $\alpha_c = (2\alpha_S^{\parallel} + \alpha_S^{\perp})/[3\varepsilon_0 \mu_0 M_0 \xi(0)] \approx (1.88, 0.47, 0.43)/\sigma_0$ and $\alpha_k = (\alpha_S^{\parallel} - \alpha_S^{\perp})/[\varepsilon_0 \mu_0 M_0 \xi(0)] \approx (0.37, -0.03, -0.09)/\sigma_0$, where $\sigma_0 = 10^{-3}$ C/cm^2 and α_S's are the $T=0$ SMEE coefficients of Ref. 7 with superscripts specifying direction of the magnetization.

Without a contact, free surfaces of the metals are "extraordinary" ($c<0$) due to the reduced coordination number for the surface atoms resulting in the increased surface density of states and consequently positive $M_{\mathrm{sur}}^{\mathrm{ex}}$. In situations important for applications, however, the metallic surface is in contact with an insulating material withstanding the voltage. In order to maximize the controllable surface charge it is meaningful to have a ferroelectric material as a contact.[5] Even if no considerable reconstruction of the lattice structure of the metal has happened at the surface, the effect of the contact material is in the charge redistribution preserving the electrochemical equilibrium and putting a constant surface charge $\Delta\sigma_{\mathrm{contact}}$ on the metallic surface. Thus, c and k should acquire constant contact-specific shifts ($\sim \alpha_{c,k} \Delta\sigma_{\mathrm{contact}}$). This, together with the other interface effects, e.g., materials' interdiffusion, will often render the surface ordinary ($c>0$)—the situation shown in Fig. 1(a). We believe, however, that the above values of c and k for free surfaces properly reflect the order of the magnitude of the constants and, in particular, $\xi_c \lesssim \xi_{k,K}$ (see below) is a reasonable assumption. Note that within the above oversimplified picture, $\alpha_{c,k}$, along with the upcoming estimations for the T_C controllability by σ in the ultrathin film limit, are not contact specific.

Having obtained the necessary GL functional parameters, we turn to the mean-field picture, in which the ground-state magnetization profile across the film can be found by the solution of the Euler equation with the appropriate boundary conditions

$$\Lambda^2 \partial_z^2 \boldsymbol{m} = \partial_{\boldsymbol{m}} (U_L + U_A), \tag{4a}$$

$$(-1)^\alpha \Lambda \partial_z \boldsymbol{m} = (c_\alpha - k_\alpha \boldsymbol{A}) \boldsymbol{m}|_{z=z_\alpha}. \tag{4b}$$

At low temperatures, the soft sector is frozen ($\boldsymbol{m}^2 \approx 1$) and $\boldsymbol{m}$ is allowed to vary only in its orientation (Goldstone sector). The orientation is governed by the nonlocal and the anisotropy terms of the GL functional, which define the (mean-field) bulk Goldstone-sector correlation length, $\xi_K^{\mathrm{MF}} = \Lambda |K|^{-1/2}$ (>5 nm for the $3d$ transition metals). In case when $D \lesssim \xi_K^{\mathrm{MF}}$, the orientation of the magnetization across the film acts as one and the effective anisotropy can be introduced, $K_{\mathrm{eff}}^{\mathrm{MF}} = K + \Sigma_\alpha k_\alpha \Lambda / D$. Provided $K_{\mathrm{eff}}^{\mathrm{MF}} \to 0$, disregards of the fact that σ resides only in, e.g., the left surface atomic layer, it is possible to externally control the sign of $K_{\mathrm{eff}}^{\mathrm{MF}}$ along with the orientation of the magnetization across the entire film. A considerable experimental progress has been recently achieved on the way to the realization of the possibility to control the orientation of the low-T metallic magnetization by sheer voltage.[8–10] This possibility stands conceptually aside from cases, in which the control is mediated by, e.g., piezoelectric strain,[11] or the effective surface field (exchange bias) in the antiferromagnet-ferromagnet structures.[12]

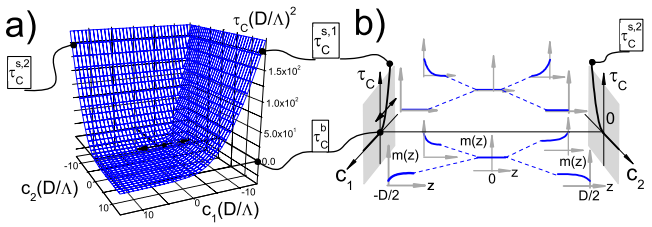

FIG. 2. (Color online) (a) The mean-field scaled Curie temperature, $\tau_C(D/\Lambda)^2$ (where Λ is the real-space cutoff) as a function of the scaled left and right surface enhancements $(c_1,c_2)\times(D/\Lambda)$. The thin-film limit corresponds to the neighborhood of the origin [cf. Eq. (9)]. The possibility to control the transition by the injected surface charge on the (left) surface $(c_1\to c_1+\alpha_c\sigma)$ is indicated by the double-headed arrow. The points on the phase-plane away from the origin correspond to (b) the wide-film limit, in which the two surfaces separate from the bulk and develop independent phase diagrams on the (τ_C,c) plane: when $c<0$, surface orders before the bulk at $\tau_C^s\approx(-c)^{1/\Phi}>\tau_C^b$ (for $\xi_c\lesssim\xi_{K,k}$ see text). The insets show the magnetization profile in the corresponding point of the phase diagram and space. The possibility to control the (left) surface transition (Ref. 13) is indicated by the double-headed arrow.

Turning back to finding the $T_C(\sigma)$ dependence, we notice that right below the transition the magnetization is vanishingly small and the nonlinear term in U_L of Eq. (4a) can be dropped, so that

$$\boldsymbol{m}(z)=\sum_{i=\pm}\boldsymbol{m}_i\exp\pm(\tau_C^{\perp,\parallel})^{1/2}(z/\Lambda), \tag{5}$$

where $\tau_C^{\parallel}=\tau_C-K/3$ and $\tau_C^{\perp}=\tau_C+2K/3$ for the in- and out-of-plane components of the magnetization. The boundary conditions (4b) provide linear homogeneous equations on $\boldsymbol{m}_{\pm}$, solvable only if

$$\Theta^{\mathrm{MF}}=\theta(\tau_C^{\perp},c_\alpha^{\perp})\,\theta(\tau_C^{\parallel},c_\alpha^{\parallel})^2=0, \tag{6}$$

where $c_\alpha^{\parallel}=c_\alpha-k_\alpha/3$, $c_\alpha^{\perp}=c_\alpha+2k_\alpha/3$ and

$$\theta=(c_1c_2+\tau_C)\tanh(\tau_C^{1/2}D/\Lambda)+(c_1+c_2)\tau_C^{1/2}. \tag{7}$$

Equations (5) and (7) are given for the $\tau_C>0$ case, which is limited on the (c_1,c_2) plane by $\Sigma_\alpha c_\alpha^{-1}>-1$ and $c_\alpha<0$. In case of negative τ_C, the corresponding equations are obtained by the analytical continuation $\tau_C^{1/2}\to i(-\tau_C)^{1/2}$.

Equation (6) has an infinite number of solutions. Only the highest solution has the meaning of the mean-field τ_C, as below it the magnetization is already condensed and, in particular, approximation Eq. (5) is not valid. Thus τ_C corresponds to either the out-of-plane $\theta(\tau_C^{\perp},c_\alpha^{\perp})=0$ or the in-plane $\theta(\tau_C^{\parallel},c_\alpha^{\parallel})=0$, whichever provides the highest τ_C depending on the surface parameters. The appropriate solution of $\theta(\tau_C,c_\alpha)=0$ is given in Fig. 2(a).

In systems of low dimensionality and/or large number of components of the order parameter, the fluctuations weaken the tendency of the magnetization to form a long-range order. For any finite $D<\infty$, the system is formally two dimensional (2D) and is known to support only the out-of-plane Ising-type long-range order[21] and the in-plane Kosterlitz-Thouless-Berezinskii (KTB) quasi-long-range order[22] with no global spontaneous magnetization (the global spontaneous magnetization exists, however, in finite 2D systems[23]). Before the two transitions may happen, the system has first to pass a rounded transition (or crossover)[24] into the state with the local (in the 2D sense) magnetization across the film, experimentally observable by, e.g., the spatially resolved magnetic measurements. It suffices to study only the σ dependence of the critical temperature of the rounded transition, τ_C, as the Ising-type and the KTB transitions temperatures are related to (and lower than) τ_C.

The rounded transition can be located as a peak in the magnetic susceptibly. This can be expressed mathematically as an implicit equation on τ_C

$$\Theta(\Lambda/D,\tau_C,K,c_\alpha,k_\alpha)=0, \tag{8}$$

the mean-field version of which is Eq. (6). In the critical region, the onset of the renormalization group flow is in the vicinity of the $N\&d=3$ stable point $(\Lambda/D,\tau_C,K,c_\alpha,k_\alpha\to 0)$, in which the fields scale as $D^{-1}\sim\mu^{-1},\tau_C\sim\mu^{-1/\nu}$, and $x_i\sim\mu^{-\phi_i/\nu}$, where the notation x_i stands for (K,c_α,k_α) and $\nu\approx 0.68$, $\phi_K\equiv\varphi\approx 1.22$, $\phi_c\equiv\Phi\approx 0.67$, and $\phi_k\equiv\Psi\approx 0.89$ up to the second order in $\epsilon=4-d$.[17,25] Besides D, the reciprocal of which is considered to be an additional relevant field in the spirit of the finite-size scaling,[25] the system has three characteristic lengths: the extrapolation length, $\xi_c=\Lambda|c|^{-\nu/\Phi}\approx(3.4,7.3,2.7)$ nm, and the lengths associated with the anisotropies $\xi_K=\Lambda|K|^{-\nu/\varphi}\approx(12.3,8.4,3.9)$ nm and $\xi_k=\Lambda|k|^{-\nu/\Psi}\approx(9.2,27,7.6)$ nm.

The shifts in the critical exponents is the most pronounced effect of fluctuations, which we are going to take into account by scaling arguments. Compared with the mean-field picture, the fluctuations have two other effects: the crossover functions acquire factors of order unity, e.g., $\sin\pi\nu\sim 1$, which, however, can be dropped to a good approximation; the absolute values of T_C^b and c_α acquire fluctuational shifts,[17] which can be absorbed into the parameters by their redefinition.

In the limit of wide films, $D\gg\xi_{x_i},\forall i$, the interior of the film is a bulk, what reflects itself in the factorization $\Theta(0,\ldots)\to\Theta^{\mathrm{bulk}}(\tau_C,K)\Pi_{\alpha=1,2}\Theta^{\mathrm{sur}}(\tau_C,K,c_\alpha,k_\alpha)$. The "bulk" part of Eq. (8) defines $\tau_C^{\mathrm{bulk}}=\Xi^{\parallel,\perp}|K|^{1/\varphi}$ of the in- and out-of-plane bulk transitions, respectively ($K\lesseqgtr 0$), where the values of the corresponding crossover functions $\Xi^{\parallel,\perp}$ are such that $\Theta^{\mathrm{bulk}}(\Xi^{\parallel,\perp},\mp 1)=0$. The two surfaces have separate phase diagrams [see Fig. 2(b)]. In case $c<0$, the surface should order before the bulk at $\tau_C^{\mathrm{sur}}\approx\Xi^{\mathrm{sur}}(-c)^{1/\Phi}$ (for $\xi_c\lesssim\xi_{k,K}$), where Ξ^{sur} is such that $\Theta^{\mathrm{sur}}(\Xi^{\mathrm{sur}},0,-1,0)=0$.[26] The possibility to control the surface transition by charge injection was discussed in Ref. 13. At this, the (2D local) magnetization controllably exists only in a relatively small surface layer of the film, making the effect of a limited interest for applications. Moreover, the voltage control is possible only if $c\to-0$, which is not satisfied in general.

Of the most importance for applications is the thin-film limit, in which the two above disadvantages with the controllable surface transition disappear. When

$$D\lesssim\xi_c, \tag{9}$$

the surface and the bulk transitions lock into one crossover. As long as Eq. (9) is satisfied, D becomes the shortest scale

in the problem, $\Lambda < D \lesssim \xi_c \lesssim \xi_{k,K}$, which allows to Taylor expand Eq. (8), after having performed the scaling transformation $\Lambda/D \to 1$, $\tau_C \to \tilde{\tau}_C \equiv \tau_C (D/\Lambda)^{1/\nu}$, $x_i \to \tilde{x}_i \equiv x_i (D/\Lambda)^{\phi_i/\nu}$ $[i=(K,c_\alpha,k_\alpha)]$, and arrive at

$$\tau_C \approx \Xi_0 (\Lambda/D)^{1/\nu} + \sum_{x_i} \Xi_i x_i (\Lambda/D)^{(1-\phi_i)/\nu}. \tag{10}$$

Here Ξ_0 is the solution of $\Theta(1,\Xi_0,0,\ldots)=0$ and

$$\Xi_i = \partial_{\tilde{x}_i}\Theta(1,\tilde{\tau}_C,\ldots)/\partial_{\tilde{\tau}_C}\Theta(1,\tilde{\tau}_C,\ldots)\big|_{\tilde{\tau}_C=\Xi_0,\tilde{x}_i=0}. \tag{11}$$

The first term in Eq. (10) is the well-known finite-size scaling correction with the "shift" exponent $1/\nu$.[24] The rest is the contribution from the other relevant fields.

Finally we are in the position to conclude with the estimations of the surface-charge-induced shifts in T_C. From Eq. (6), the necessary values of the mean-field crossover functions $\Xi_c=1$ and $\Xi_k=-1/3,2/3$ for the in- and out-of-plane orderings. Combining this result with Eq. (10) and the above values for the critical exponents, Λ and $\alpha_{k,c}$, we find that the T_C shift

$$\Delta T_C \approx \sigma T_C^b [\alpha_c \Xi_c (\Lambda/D)^{(1-\Phi)/\nu} + \alpha_k \Xi_k (\Lambda/D)^{(1-\Psi)/\nu}],$$

induced by typical saturation polarization of lead zirconate titanate (PZT) ($\sigma \approx 30\ \mu C/cm^2$) in a $D=2$-nm-wide film is (13.6,7.9,6.9) K and (19.1,7.0,4.7) K for the in- and out-of-plane orderings, respectively.

In summary, in this Brief Report we studied the possibility to control the ferromagnetic critical temperature of ultrathin metallic films by charging, which should enable the bias-voltage control of the existence of the metallic magnetization. The coupling of the surface charge to the critical temperature is due to the surface-charge dependence of the surface enhancement and the surface anisotropy, which are relevant fields in the critical region. Using scaling arguments and the GL functional derived from data previously obtained experimentally and by zero-temperature *ab initio* studies we found a considerable sensitivity of the critical temperature to the charging in the 3*d* transition metals ultrathin films. For the room-temperature applications, the critical temperature should be brought down to the desired operation temperature. This can be achieved by, e.g., the use of alloys.[13]

At the time of the submission we became aware of the very recent experimental demonstration[27] of the critical temperature of a 4-nm-wide layer of the complex ferromagnetic oxide (LaSrMnO) is subject to the 20 K variation by the surface charge of a PZT contact. At this, the density of free carriers is almost at the metallic level (10^{21} cm^{-3}). This result supports our prediction that disregard of the high density of free carriers, the critical temperature in the metallic ferromagnetic films is externally controllable.

The work was supported in part by the Western Institute of Nanoelectronics (WIN) funded by the NRI, Center on Functional Engineered NanoArchitectonics (FENA) funded by FCRP, and ARO MURI.

*Also affiliated with Laboratory for Nanophysics, Institute for Spectroscopy of Russian Academy of Sciences, Troitsk 142190, Russia. iovchinnikov@ucla.edu

†wang@ee.ucla.edu

[1] I. V. Ovchinnikov *et al.*, Appl. Phys. Lett. **92**, 093503 (2008).
[2] Ch. Binek *et al.*, J. Phys.: Condens. Matter **17**, L39 (2005); M. Bibes *et al.*, Nature Mater. **7**, 425 (2008).
[3] A. S. Logginov *et al.*, Appl. Phys. Lett. **93**, 182510 (2008).
[4] C. Chappert *et al.*, Nature Mater. **6**, 813 (2007).
[5] C.-G. Duan *et al.*, Phys. Rev. Lett. **97**, 047201 (2006).
[6] J. M. Rondinelli *et al.*, Nat. Nanotechnol. **3**, 46 (2008).
[7] C. S. Duan *et al.*, Phys. Rev. Lett. **101**, 137201 (2008).
[8] S. Sahoo *et al.*, Phys. Rev. B **76**, 092108 (2007).
[9] T. Maruyama *et al.*, Nat. Nanotechnol. **4**, 158 (2009).
[10] M. Weisheit *et al.*, Science **315**, 349 (2007).
[11] T. K. Chung *et al.*, Appl. Phys. Lett. **92**, 112509 (2008).
[12] P. Borisov *et al.*, Phys. Rev. Lett. **94**, 117203 (2005); V. Laukhin *et al.*, *ibid.* **97**, 227201 (2006); X. Chen *et al.*, Appl. Phys. Lett. **89**, 202508 (2006).
[13] I. V. Ovchinnikov *et al.*, Phys. Rev. B **79**, 020402(R) (2009); **78**, 012405 (2008).
[14] F. Rodolakis *et al.*, Phys. Rev. Lett. **102**, 066805 (2009); G. Borghi *et al.*, *ibid.* **102**, 066806 (2009).
[15] E. Brezin *et al.*, Phys. Rev. B **10**, 892 (1974).
[16] M. T. Johnson *et al.*, Rep. Prog. Phys. **59**, 1409 (1996).
[17] H. W. Diehl, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic, London, 1986), Vol. 10.
[18] C. A. F. Vaz *et al.*, Rep. Prog. Phys. **71**, 056501 (2008).
[19] In order to be consistent with the calculations of Ref. 7, among the values obtained by different computational methods we choose the ones by the FLAPW.
[20] $s=2a^{-2}, 2/\sqrt{3}a^{-2}$ and a^{-2} for the fcc[001], hcp[0001], and bcc[001], respectively (a is the lateral lattice constant).
[21] M. Bander *et al.*, Phys. Rev. B **38**, 12015(R) (1988).
[22] A. M. Polyakov, Phys. Lett. B **59**, 79 (1975).
[23] S. T. Bramwell *et al.*, J. Phys.: Condens. Matter **5**, L53 (1993).
[24] M. E. Fisher *et al.*, Phys. Rev. Lett. **19**, 169 (1967); M. N. Barber, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic, London, 1983), Vol. 8.
[25] D. J. Amit, *Field Theory, the Renormalization Group and Critical Phenomena* (McGraw-Hill, New York, 1978).
[26] In case when k and K are of opposite signs, Fe[001] only, and $\xi_k \lesssim \xi_K$, the $\pi/4$ domain wall can be formed at the surface. This situation, however, looks unlikely from the above estimations of the lengths, so that the surface and the bulk order most likely in one direction.
[27] H. J. A. Molegraaf *et al.*, Adv. Mater. **21**, 1 (2009).