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Manifestation of quantum confinement in transport properties of ultrathin metallic films

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Abstract

The influence of quantum size effect on the transport properties of ultrathin Cu and Fe films in the thickness range between 1 and 32 monolayers is studied by solving the linearized Boltzmann equation in the relaxation-time approximation using *ab initio* calculations within the framework of spin density functional theory.

A strong manifestation of quantum confinement is found in the density of states. However, the plasma frequency shows a smooth or oscillating behavior for different film orientations. The results are in qualitative agreement to infrared-absorption experiments at Cu(111), Fe(001), and Fe (111) films.

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1. Introduction

Quantum confinement of electrons in low-dimensional systems causes oscillations in typical features of the electronic structure. In this respect the Ruderman–Kittel–Kasuya–Yosida-like interlayer exchange coupling between two ferromagnetic layers separated by a non-ferromagnetic spacer is well known. Oscillations of the interlayer exchange coupling for many spacer materials were observed [1–7] and were explained theoretically considering Fermi surface nesting vectors along the corresponding crystal growth direction [8,9].

Oscillations owing to the same nature are known also for transport properties of ultrathin metallic films. In this respect, the observation of quantum size effect (QSE) in the dependence of the specific conductivity on the thickness of ultrathin Pb(111) films [10,11] is well documented. A saw-tooth like oscillation of the in-plane conductivity with thickness was predicted by Trivedi and Ashcroft applying a free-electron model [12]. More sophisticated theoretical results obtained recently for this system are in good agreement with the experiment [11,13] and show the connection between quantum size oscillations of the conductivity and the density of states (DOS) at the Fermi level (E_F). As was shown in Ref. [13] this relation can be understood assuming the relaxation time to be inverse proportional to the DOS(E_F) [13,14]. The second contribution to the conductivity in the relaxation-time approximation, the plasma frequency, shows a smooth behavior for Cu films [14]. However, we have found a pronounced oscillation for Fe(111) film orientation. The main objective of this paper is to elucidate the different behavior of Fe and Cu films with surface orientations of (001) and (111).

2. The model

The electronic structure of the considered systems was calculated self-consistently using a screened Korringa–Kohn–Rostoker (SKKR) multiple scattering Green's function method [15–19]. A screening potential with a barrier height of 4 Ry is used and the screened structure constants include coupling to four shells of nearest neighbours. It ensures the same numerical accuracy as the traditional KKR scheme [18]. Spherical

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potentials in the atomic sphere approximation were used. The calculations employ the exchange-correlation potential in local density approximation of Vosko, Wilk, and Nusair [20]. An angular momentum cut-off of $l_{\max}=3$ is used for the Green's function, thus implying a cut-off for the charge-density components at $2l_{\max}=6$. The film unit cell contains N atoms of the considered material (each representing one atomic layer) and sufficient empty sites on both sides of film to ensure a proper treatment of the charge relaxation at the surfaces. Due to the slab geometry the states at the Fermi level are described by lines in the two-dimensional (2D) Brillouin zone (BZ). The Fermi surface integrals have to be replaced by line integrals [21]. In the considered geometry the irreducible part equals one eighth for (001) and one twelfth of the whole BZ for (111) surface orientation and contains about 100000 $\mathbf{k}_{\|}$-points to obtain convergence. We neglect the lattice relaxations at the surfaces. The lattice constants were taken equal to $A=0.3576$ nm for fcc Cu and $A=0.2865$ nm for bcc Fe [22]. The considered thicknesses of 1–32 monolayers (ML) correspond to 0.18–5.72 nm for Cu(001), 0.21–6.61 nm for Cu(111), 0.14–4.58 nm for Fe(001), and 0.08–2.65 nm for Fe(111) films.

The transport is treated quasiclassically by solving the linearized Boltzmann equation which in the *anisotropic* relaxation-time approximation gives the following expression for the components of the conductivity tensor $\hat{\sigma}$ of a crystal (per spin direction σ) [23,24]

$$\sigma_{ij}^{\sigma}=\frac{e^2}{V}\sum_k \delta(E_k^{\sigma}-E_F)v_{k,i}^{\sigma}v_{k,j}^{\sigma}\tau_k^{\sigma}, \tag{1}$$

where e is the electron charge and V the system volume. Here $v_{k,i}^{\sigma}$ denotes the Cartesian component of the group velocity $\mathbf{v}_k^{\sigma}$ of an electron with relaxation time τ_k^{σ} at energy E_k^{σ} (where k is a shorthand notation for crystal momentum vector $\mathbf{k}$ and band index n). For a cubic crystal and a 2D lattice with C_{4v} or C_{6v} symmetry the transport properties are isotropic $\sigma_{ij}^{\sigma}=\sigma^{\sigma}\delta_{ij}$.

Assuming in addition an *isotropic* relaxation-time we can write the above expression for σ^{σ} as a product of the relaxation time τ^{σ} and a squared Drude-type plasma frequency ω_P^{σ} [25]

$$\sigma^{\sigma}=\tau^{\sigma}\frac{e^2}{V}\sum_k \delta(E_k^{\sigma}-E_F)v_{k,i}^{\sigma}v_{k,i}^{\sigma}=\frac{\tau^{\sigma}(\omega_P^{\sigma})^2}{4\pi}. \tag{2}$$

In the case of a film (with C_{4v} or C_{6v} symmetry) of thickness d the expression for the spin-dependent plasma frequency is evaluated in the following form

$$(\omega_{\mathrm{P,film}}^{\sigma})^2=\frac{e^2}{2\pi\hbar d}\oint_{E_k^{\sigma}=E_F} dl_k^{\sigma}|v_k^{\sigma}(E_F)|, \tag{3}$$

where dl_k^{σ} is a differential Fermi line element arising from the n-th band.

According to Eq. (2), conductivity is an interplay of host band structure effects and scattering processes. Whereas, $\omega_{\mathrm{P}}^{\sigma}$ is determined by the one-particle energy spectrum E_k^{σ} of the ideal system only and has a clear physical meaning. It measures the mobility of the conduction electrons as an integrated value. The relaxation time accounts for the scattering mechanisms acting in the considered system. At $T=0$ K the inelastic scattering by phonons vanishes. In this case τ^{σ} is determined by the scattering at impurities inside the slab as well as imperfections at the surfaces and interfaces. So, the residual resistivity of the system has to be considered only. It is well known that the scattering properties depend drastically on the type of defects [24]. Nevertheless, some common features can be determined without a detailed consideration of the type of impurities. As it was shown in an earlier publication [14], the band structure of the ideal film strongly influences the relaxation time by the spin-dependent total film density of states at the Fermi level (normalized to a film unit cell)

$$n_{\mathrm{film}}^{\sigma}(E_F)=\frac{1}{\hbar S_{\mathrm{BZ}}}\oint_{E_k^{\sigma}=E_F}\frac{dl_k^{\sigma}}{|v_k^{\sigma}(E_F)|}. \tag{4}$$

Here S_{BZ} is the area of the 2D surface Brillouin zone. The DOS defines on average the number of states available for the scattering at defects equally distributed inside a film (called bulk defects). Thus, some features of the transport properties of ultrathin films can be derived from the one-particle energy spectrum E_k^{σ} of the ideal system.

3. Results

3.1. Cu films

The total film density of states at the Fermi level n_{film} (E_{F}) determined by Eq. (4) is presented in Fig. 1 for the cases of Cu (001) and Cu(111) films. The theory of the oscillatory interlayer exchange coupling predicts two periods of 5.88 ML and 2.56 ML for Cu in (001) orientation and one of 4.5 ML for Cu in (111) orientation [8]. In the investigated thickness range for Cu (111) films a pronounced oscillation with a period of 4.3 ML could be identified by Fourier analysis. This value is in good agreement to the calculation in Ref. [8]. The small difference is as a result of different lattice constants (0.3576 nm and 0.3602 nm, respectively). Experiments at the systems fcc(111)

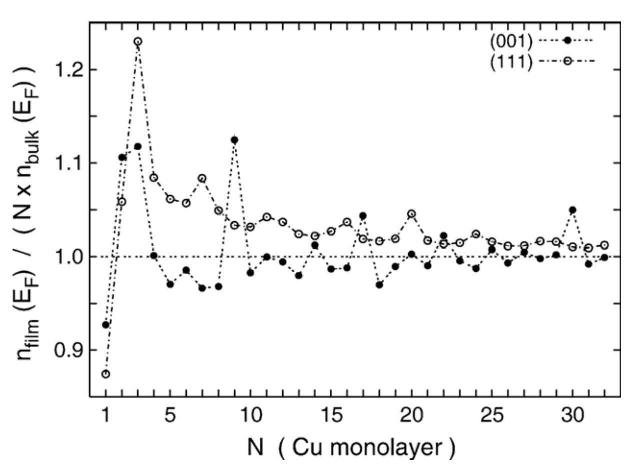

Fig. 1. Total film density of states at the Fermi level (normalized to the bulk value) as a function of Cu layer thickness for (001) and (111) orientation.