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Linear in magnetic field increase of the magneto-optical Kerr effect in multilayered Co/Cu films

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The Kerr rotation, steadily and linearly increasing in magnetic field was observed in the Co/Cu multilayered films at the Cu layer thickness providing the extreme exchange couplings between the Co layers. The enhancement of the Kerr effect in magnetic field is supposed to be caused by increase of the electron density at the Fermi level at the Co/Cu interfaces because of hybridization of the *d*-(Co) and *sp*-(Cu) electronic states subjected to the quantum size effect in the Cu layers.

В многослойных плёнках Co/Cu при толщинах Cu, обеспечивающих экстремальную обменную связь между слоями Co, наблюдалось монотонное линейное с полем увеличение угла вращения Керра. Предполагается, что увеличение эффекта Керра в магнитном поле вызвано возрастанием плотности электронов на уровне Ферми в интерфейсах благодаря гибридизации *d*-(Co) и *sp*-(Cu) электронных состояний, подверженных влиянию квантово-размерного эффекта в слоях Cu.

For recent years, attention of numerous research groups has been focused on studies of ultra thin films which exhibit properties essentially different from those of bulk materials [1]. The interest in the comprehensive studying thereof is caused, first of all, by the giant magnetoresistance (GMR) effect discovered in multilayered films of ferromagnetic/normal (FM/NM) metal type in the late 1980s [2]. The GMR properties of multilayered films make it possible to use them in magnetic field sensitive sensors [3], which are used as read heads of hard discs [4]. These materials also are promise for spintronics devices, for instance, in a new type of computer memory with magnetic tunnel junctions [5], spin-polarized current driven magnetization switching devices, nanoscale sources of radio-frequency radiation [6, 7]. In many cases, the difference in the properties of the ultra thin films with respect to those of the bulk materials is due to modified electronic structure of the thin film [8]. One of the reasons for these changes is the quantum size effect (QSE) arising when even one of the object geometrical dimensions is comparable with the de Broglie wavelength of conductivity electrons. Substantial changes of electronic spectrum also occur at interfaces of multilayered films due to hybridization of electronic states in adjacent metal layers.

Among physical properties sensitive to variations of the electronic spectra in multilayered systems, there are magneto-optical ones. For instance, the observed spontaneous polar Kerr effect oscillations as a function of the NM metal layer thickness in Au-wedge/Co, Fe/Au-wedge/Fe and Fe/Ag-wedge/Fe films were ascribed to QSE [9, 10]. Amplitudes of those Kerr effect changes amounted up to 25 %. It was shown that the influence of the electronic states hybridization on the electron band structure of interfaces in the multilayers is reflected in some cases also as an additional magneto-optical (MO) response [11, 12]. In [13], the field-induced Kerr effect in the Au/Ru-wedge/Co system has been reported. Oscillations of the "paramagnetic magneto-optical Kerr effect" as a function of the Ru layer were thickness explained by a considerably increasing paramagnetic susceptibility of the Ru layer conduction electrons at the certain Ru thickness caused by increasing of the electronic density-of-states at the Fermi level of Ru when quantum well resonance states approach the Ru Fermi level.

In this work, the longitudinal Kerr effect has been investigated in a series of Co(8 Å)/Cu(d_{Cu}) multilayers with different thickness of the Cu layers. The linear in magnetic field enhancement of Kerr rotation at increasing copper layer thickness has been first revealed in the Co/Cu multilayered system. The spontaneous Kerr rotation varies also as the copper layer thickness increases. The enhancement mechanisms of the field-induced and the spontaneous Kerr effects are supposed to be of a similar nature. Those are connected most likely with hybridization of the Co and Cu electronic states at the Co/Cu interfaces subjected to the quantum size effect in the Cu layers.

The films were deposited by alternate condensation of Co and Cu metals onto a mica (fluoroflogopite) substrate. The magnetron sputtering in a vacuum setup at the residual pressure of ~10^{-6} Torr was used. During sputtering, argon pressure was kept fixed at $1.3\cdot10^{-3}$ Torr. Condensation rates of Co and Cu layers were 0.045 and 0.058 nm/s, respectively. At first, the Cu buffer layer (5 nm) was deposited onto the mica substrate. Then the Co layer and the 19 identical Cu/Co bilayers were deposited. The upper Cu layer thickness in all the films was 1.25 nm. Thicknesses of all magnetic Co layers, d_{Co}, were invariable in all the films and equal to 0.8 nm. Thicknesses of the Cu layers, d_{Cu}, except for the upper Cu layer, were identical in each film but different in various films. The d_{Cu} value was varied from 0.6 to 2 nm at a step of 0.1–0.2 nm in the film series. The layer thickness was determined basing on the sputtering duration, calibrated by multiple light beam interferometry. In accordance with the electron microscope measurements, each film has polycrystalline structure with the columnar grains extended along the normal to the film surface. The Co and Cu layers of the multilayered grains had fcc structures with the crystallographic fcc (111) planes of Co and Cu parallel to the films substrate. Lateral dimensions of the columnar grains in the films were from 8 to 10 nm. In spite of the fact that the Co layers have crystallographic anisotropy with "easy axis" [111], magnetic moments of the Co layers are oriented parallel to the layers providing the energy minimum in that way.

The magnetic field dependence of the Kerr rotation was examined in the longitudinal configuration. The magnetic field was parallel to the film plane and parallel to the light incidence plane. The electric field of the plane-polarized incident light was oriented transversely to the light incidence plane ($\mathbf{E}_s$ polarization). The light angle was about 55 deg. The measurements were carried out in the magneto-optical setup at room temperature of the samples using the modulation method. The working material of the Faraday modulator was the bismuth-iron-yttrium garnet. The light source was a He–Ne laser (λ = 632.8 nm). The maximum available field was 16.8 kOe. The measurement error was about 10^{-3} deg. The deviation of the film plane from the magnetic field vector direction did not exceed 1 deg. The light beam average diameter on the sample did not exceed 0.5 mm.

The magnetic field dependences of the Kerr rotation had hysteretic character at the field $H<100$ Oe for all the films, except for the film with the copper layer thickness d_{Cu} = 1.8 nm. The hysteresis loop shapes were dependent on the field direction orientation in the film plane. The hysteresis loop width was varied with the period of 180 deg during the film rotation about the normal thereto. This testifies to the "easy axis" anisotropy in the film plane. The coercive fields do not exceed 50 Oe. The curves $\theta(H)$ were independent of the magnetic field orientation in the film plane at $H>100$ Oe. Here, we focus our attention on the Kerr rotation θ behavior in the films with different thickness of the Cu layers in the fields $H>100$ Oe.

Approaching of the curves $\theta(H)$ to magnetic saturation are different for films having different Cu layer thickness. Almost all the films reach the magnetic saturation at the field H = 3 to 6 kOe, except for the films with the Cu layer thickness d_{Cu} = 0.9, 1.35, and 1.8 nm (Figs. 1 and 2). The curves $\theta(H)$ for these films are far from

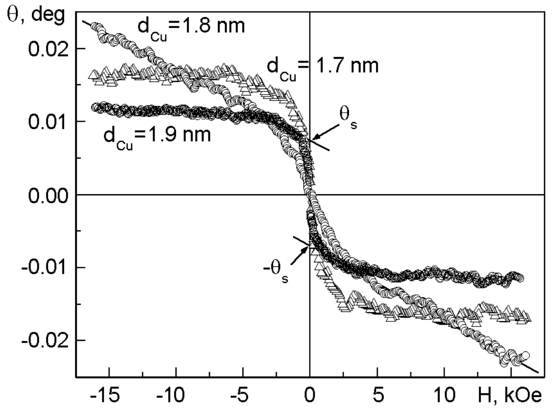

Fig. 1. The magnetic field dependences of Kerr rotation for the multilayers Co(0.8 nm)/Cu(1.7 nm), Co(0.8 nm)/Cu(1.8 nm) and Co(0.8 nm)/Cu(1.9 nm).

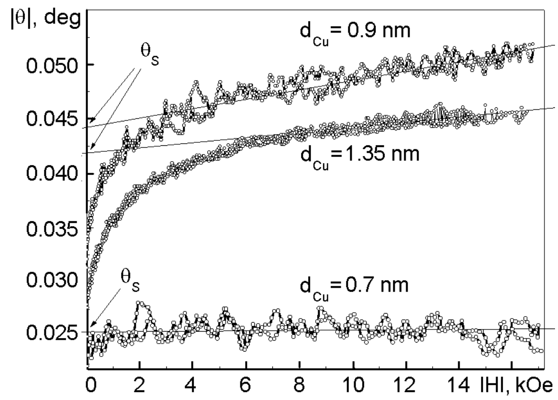

Fig. 2. Absolute value of Kerr rotation for Co/Cu films with d_{Cu} 0.7 nm, 0.9 nm, and 1.35 nm as a function of the magnetic field strength.

saturation state at the maximum available field 16.8 kOe. These curves are shown in Figs. 1 and 2 in comparison with those for the films with d_{Cu} = 1.7 nm, 1.9 nm, and 0.7 nm, as an example. It is noticeable that the curve $\theta(H)$ for the film with d_{Cu} = 1.8 nm has prominent linear rise of θ in the field range from 5 kOe to 16.8 kOe. Slopes of the linear parts ($d\theta/dH$) of the curves $\theta(H)$ for all films are shown in Fig. 3. The highest peak of $d\theta/dH$ is observed in the anhysteretic film Co/Cu (1.8 nm) and the smallest one, in the film Co/Cu(1.35 nm). The noticeable linear rise $\theta(H)$ occurred also in the film Co/Cu(0.6 nm).

Let us consider the possible reasons for the observed linear rise of θ in high fields. The linear in field rise of θ might have been connected with the magnetization process of the AFM-coupled Co layers. The fact that the maxima of $d\theta/dH(d_{Cu})$ are observed at the Cu layer thickness d_{Cu} = 0.9 and 1.8 nm, corresponding to the maxima of the AFM exchange coupling between the FM Co layers in the Co/Cu system [14, 15], requires to check this assumption. In connection with the fact that the anisotropy field in the film plane does not exceed 50 Oe, the spin-flip field for the layer magnetic moments (H_{flip}) will be defined mainly by the exchange constant and the FM layer thickness d_{FM} [16]:

$$H_{flip} = \frac{4(n-1)J}{nId_{FM}}. \quad (1)$$

Here I is the Co layer magnetization; J, the exchange coupling energy per unit area. The saturation magnetization of the Co-layers at the thickness d_{Co} = 0.8 nm is about 900 G [17], the exchange constant J in the second maximum of the AFM exchange coupling (d_{Cu} = 1.8 nm) between Co-layers in Co/Cu/Co films is within the range of 0.08 to 0.06 erg/cm^2 [16, 18], and the flipping field does not exceed 4 kOe. But in the Co/Cu(1.8 nm) film, the Kerr rotation θ and, consequently, the magnetization do not reach the saturation state even at the maximum available field 16.8 kOe (Fig. 1, 2). Moreover, $d\theta/dH \neq 0$ at $H>8$ kOe is observed also in the film with the Cu layer thickness d_{Cu} = 1.35 nm, corresponding to the maximum of the FM exchange coupling between Co-layers.

The linear in field rise of θ can be connected also with the contribution of an additional magneto-optical Kerr effect from the conduction electrons of the Cu layers. This contribution is supposed to be caused by the enhanced paramagnetic susceptibility of the conduction electrons. In accordance with the Stoner criterion, the Pauli paramagnetic susceptibility of the conduction electrons (χ_P) can be increased due to enhanced product of the atomic exchange integral (J_a) and the density-of-states at the Fermi level (N_F) [19]:

$$\chi_{St} = \frac{\chi_P}{1 - J_a N_F}. \quad (2).$$

χ_{St} is the enhanced paramagnetic susceptibility of the conduction electrons. The atomic exchange integral for copper in the Cu layers is unlikely to be changeable considerably, because J_a(Cu) is close to J_a for all FM metals [19], while the N_F(Cu) value could be changed in the thin Cu-layers of the Co/Cu multilayer films, because of electronic density redistribution in the energy spectrum [1]. The obtained value $d\theta/dH \approx 1\cdot 10^{-3}$ deg/kOe for the film Co/Cu(1.8 nm) exceeds the expected one (~$3.5\cdot 10^{-5}$ deg/kOe, taking into account [20]) for the longitudinal effect in the bulk copper by more than one order. Thus, in accordance with the expression (2), to explain the observed linear-in-field enhancement of the Kerr effect in the Co/Cu(0.9 nm), Co/Cu(1.35 nm) and Co/Cu(1.8 nm) films by the paramagnetic susceptibility enhancement of the conduction electrons in the Cu-layers, the density-of-states at the Fermi level in the Cu-layers should be increased more than by one order.

The electron density-of-states at the Fermi level in Cu layers of the multilayered Co/Cu system can be increased due to the quantum size effect, when the quantum well resonance states reach the Fermi level of Cu. In [13], the oscillations of the "paramagnetic Kerr susceptibility" (slope of linear in field parts of $\theta(H)$) in the three-layered Au/Ru-wedge/Co system as a function of the Ru layer thickness is explained by just that mechanism. The maxima of the "paramagnetic Kerr susceptibility" were observed at the Ru layer thicknesses the AFM exchange interaction between the Co layers in the Co/Ru/Co sandwiched system. As mentioned above, the maxima of the AFM exchange coupling between the FM Co layers in the Co/Cu system are observed around $d_{\mathrm{Cu}} = 0.9$ and 1.8 nm [15]. It is known that exchange coupling between the FM layers arises with formation of the spin-polarized quantum-well states and the maxima of the indirect exchange coupling are observed when the quantum-well states cross the Fermi level [21]. So, the fact that the observed linear-in-field rise of θ in the multilayered Co(0.8 nm)/Cu system at the Cu layer thickness $d_{\mathrm{Cu}} = 0.9$ nm, 1.35 nm and 1.8 nm, corresponding to the exchange coupling maxima, can testify the QSE as a cause of the peaks $d\theta/dH$ in $d\theta/dH(d_{\mathrm{Cu}})$ dependence. However, N_F is unlikely to be increased by one order due to the QSE only.

Increase in the electronic density-of-states at the Fermi level at the Co/Cu interfaces may be caused also by hybridization of the Cu *sp*-electron states with the Co *d*-electron states. Besides, it is naturally to

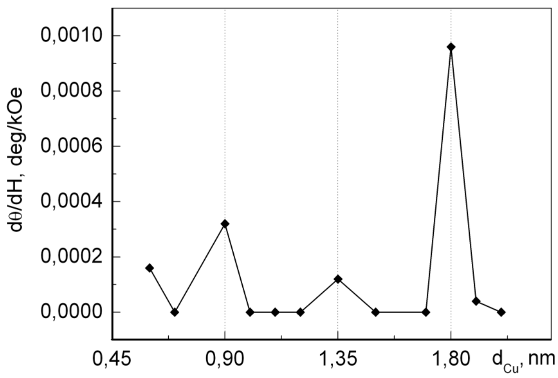

Fig. 3. Cu-layer thickness dependence of the slope of the $\theta(H)$ curves in high fields (H>10 kOe).

expect changed optical and magneto-optical constants at the interfaces due to changing oscillator strengths of the optical transitions. Calculations of magneto-optical properties of multilayered FM/NM films taking into account the hybridization of the electronic states at the interfaces have shown that the spontaneous magneto-optical properties are heavily dependent on the quality of the interfaces [11, 12, 22, 23]. It is known too, that the space distribution of the conduction electron spin density in the presence of QSE can influence the structure and properties of the MLF interfaces [24–28]. In several works, influence of the QSE on the atomic interlayer spacing near interfaces during sputtering [24], on the film surface energy [25] and the island height [26] during annealing, on twin formation [27] and on the magnetic anisotropy of films [28] has been revealed. Consequently, the QSE is supposed to influence the interface structure of the Co(0.8 nm)/Cu multilayers with $d_{\mathrm{Cu}} = 0.9$ nm, 1.35 nm and 1.8 nm during sputtering and it results in increase of the electronic density-of-state the Fermi level at the interfaces. This circumstance may cause an enhanced paramagnetic susceptibility of the interfacial Co/Cu conduction electrons and, at last, the enhancement of the paramagnetic magneto-optical effects in the films. If this mechanism is considered as the most probable, linear extrapolation of the $\theta(H)$ from high field to $H = 0$ can be accepted as a correct determination of the spontaneous Kerr rotation (θ_s) (Fig. 1, 2).

The Cu layer thickness dependence of the spontaneous Kerr rotation $|\theta_s|(d_{\mathrm{Cu}})$ is shown in Fig. 4. It consists of a monotonously decreasing background and the prominent peaks (100 % changes of $|\theta_s|$) at d_{Cu} = 0.9 and 1.35 nm. The small minimum at d_{Cu} = 1.8 nm is also noticeable. The monotonic decrease of $|\theta_s|$ can be explained by the light attenuation in the Cu layers with increasing thickness of the latter. The expression (3) for $\theta_s(d_{Cu})$, obtained in the ultrathin FM layers approximation using the work [29],

$$\theta_s = \mathrm{Re}\left[V(\varphi)d_{Co}\frac{\varepsilon_1^{Co}}{\varepsilon_0^{Co}}N^{Cu}\sum_{n=1}^{20}\eta(d_{Cu})\right] \quad (3)$$

can be applied to fit the observed background satisfactory. The function

$$V(\varphi) = \frac{2\pi N_{Cu}\sin 2\varphi}{\lambda(N_{Cu}^2 - 1)\cos\varphi\sqrt{\varepsilon_0^{Cu} - \sin^2\varphi} + \sin^2\varphi}$$

describes mainly the incidence angle dependence of the effect,

$$\eta(d_{Cu}) = \exp\left(\frac{4\pi i\sqrt{\varepsilon_0^{Cu} - \sin^2\varphi}\,(d_{Cu}^{top} + nd_{Cu})}{\lambda}\right),$$

taking into account the magneto-optical response attenuation because of increasing light path in the absorbing Cu-medium between the entrance surface and the n-th Co layer. The approximations of the experimental $|\theta_s|(d_{Cu})$ dependence by the expression (3) are shown in Fig. 4 as the smooth decaying lines *1* and *2*. The approximation using the optical constants of bulk Co and Cu (ε_0^{Co} = $-12.50-i\cdot18.46$ [30] and $\varepsilon_1^{Co} = -0.47-i\cdot0.65$ [31] — diagonal and off-diagonal components of the Co permittivity tensor, respectively; $N_{Cu} = 0.24+i\cdot3.47$ [32] — the complex reflectivity index of Cu) is labelled with 1. It is seen from Fig. 4 that this simulation explains the monotonous decay of the $|\theta_s|(d_{Cu})$ dependence qualitatively. But a more satisfactory approximation can be obtained by changing optical parameters of Cu and Co. For instance, the line *2* is obtained if we take N_{Cu} as $0.21+i\cdot4$.

Comparison of Fig. 3 and 4 shows that the positions of the first two maxima of $d\theta/dH(d_{Cu})$ dependence coincide with those in the $|\theta_s|(d_{Cu})$ one. The most intense maximum of the $d\theta/dH(d_{Cu})$ coincides with the weak minimum of $|\theta_s|(d_{Cu})$ one. The correlation between the $d\theta/dH(d_{Cu})$ and $|\theta_s|(d_{Cu})$ dependences may testify the coupling between the origins of the $d\theta/dH$ peaks and

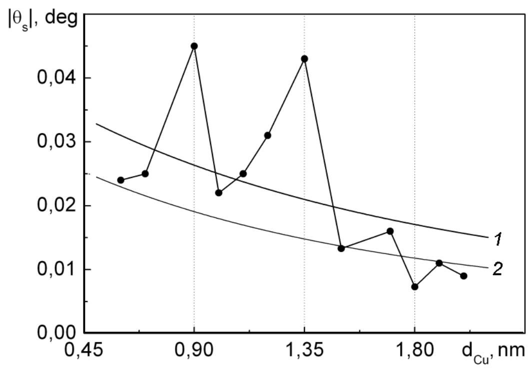

Fig. 4. Experimental (dark circles) and calculated (smoothly decaying lines) Cu layer thickness dependence of the spontaneous Kerr rotation for the Co(0.8 nm)/Cu(d_{Cu}) multilayers. Line *1*, approximation by Eq.(3) with the bulk Co and Cu optical constants ($\varepsilon_0^{Co} = -12.50-i\cdot18.46$ [30] and $\varepsilon_1^{Co} = -0.47-i\cdot0.65$) — diagonal and off-diagonal components of the Co permittivity tensor, respectively; $N_{Cu} = 0.24+i\cdot3.47$ [32] — the complex reflectivity index of Cu). Line *2*, approximation by Eq.(3) with the changed complex reflectivity index of Cu $N_{Cu} = 0.21+i\cdot4$.

the θ_s increase at least in two films Co/Cu(0.9 nm) and Co/Cu(1.35 nm).

The θ_s increase in MLFs can be due to several reasons. The spontaneous magneto-optical properties of the multilayered films can be noticeably enhanced due to the changing optical parameters of the interfaces between NM and FM layers, caused by electronic states hybridization of the adjacent metal layers [11, 22]. The changes of the spontaneous magneto-optical Kerr effect in the Co/Cu multilayered films due to the modified optical parameters of the interfaces at λ = 632.8 nm attain 100 % [33]. The hybridization of atomic electronic states of the adjacent NM and FM layers in MLFs can influence the spin-orbit splitting of electronic bands in the FM layer [34]. But, this is unable to cause any marked changes in the MO response because of small difference between the spin-orbit coupling of Cu and Co [12]. The hybridization of Cu-p band with Co-d band can cause Cu polarization at the Cu/Co interfaces. But the Cu magnetic moment amounts only $0.02\mu_B$ [35] (for the Co(1.2 nm)/Cu(0.8 nm) multilayer); in contrast, the Co magnetic moment in the bulk fcc material is about $1.67\mu_B$ (1400 G [35])). The NM layer magnetization in MLFs also may be due to the interference of the spin-polarized conductivity electrons in the NM layer. However, its possible values (about $0.05\mu_B$, calculated for $[Co/Cu(0.4\ nm)]_{20}$ [36], and about $0.09\mu_B$, experimentally obtained for $[Co/Cu(1\ nm)]_{50}$ [37]) are much less than ~ $1.67\mu_B$ (bulk Co), too. Thus, Cu polarization in the Co/Cu multilayers due to both the hybridization of the Cu and Co states at the interfaces and the interference of the spin-polarized conductivity electrons in the Cu layer may enhance the Kerr effect only by no more than 10 %.

In several works, the enhancement of the spontaneous Kerr effect in the Au-wedge/Co [9], Fe/Au-wedged/Fe, and Fe/Ag-wedged/Fe [10] films was observed. The authors proved that such phenomenon was caused by the QSE influence on the energy spectrum of the NM layer and, consequently, the appearance of new magneto-optical transitions to the spin-polarized quantum well states of NM metal. As the NM layer thickness increases, the intensities of these new transitions oscillated periodically as the quantum well state crosses the Fermi level, giving rise to enhancement of the Kerr effect by up to 25 %.

Among the considered reasons for the spontaneous enhancement of MO properties, the hybridization of atomic electronic states in adjacent Cu and Co layers is seemed to be the most probable one. It has the most influence on the MO response.

It should be noted the absence of the Kerr effect spike at $d_{Cu} = 1.8$ nm. Moreover, at this thickness, the weak minimum in the $|\theta_s|(d_{Cu})$ dependence is noticeable. Such behavior is supposed to be connected with the peculiarities of the interface structure between the Co and Cu layers. As shown in our previous work [38], the film Co/Cu(1.8 nm) has pronounced superparamagnetic properties, testifying the weak FM coupling between its grains and the small FM pinhole amount between the Co layers. The particular magnetic and magneto-optical properties of the Co/Cu(1.8 nm) are likely to be connected with the peculiarities of the interface structure between the Co and Cu layers in this film, caused by the known influence of the QSE on the structure and properties of the MLFs interfaces [24–28]. As mentioned above, the spontaneous magneto-optical properties of the multilayered FM/NM films depend heavily on the quality of the interfaces [11, 12, 22, 23].

Thus, the magnetic field induced Kerr rotation has been observed in the Co/Cu multilayers having the Cu layer thicknesses equal to 0.9, 1.35, and 1.8 nm, providing the extreme AFM or FM exchange couplings between the Co layers. The Kerr rotation angle increases in magnetic field steadily and linearly with the magnetic field strength. In the films with $d_{Cu} = 0.9$ and 1.35 nm, the enhanced spontaneous Kerr rotation is also observed. The enhancement mechanisms of the field-induced and spontaneous Kerr effects may have a common reason, i.e., hybridization of the Co and Cu electronic states at interfaces. The Kerr effect enhancement in magnetic field is supposed to be caused by rise of the electron density at the Fermi level at the Co/Cu interfaces because of the d-(Co) and ps-(Cu) electron hybridization, subjected to the quantum size effect in the Cu layers.

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Лінійне збільшення магнітооптичного ефекту Керра у магнітному полі у багатошарових плівках Co/Cu

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У багатошарових плівках Co/Cu при товщині шарів Cu, які забезпечують екстремальний обмінний зв'язок між шарами Co, спостерігалося монотонне лінійне з полем збільшення кута обертання Керра. Припускається, що збільшення ефекту Керра у магнітному полі викликано збільшенням густини електронів на рівні Фермі в інтерфейсах завдяки гібридизації *d*-(Co) і *sp*-(Cu) електронних станів, що зазнають впливу квантового розмірного ефекту.