Giant oscillations of coupling strength in Mo/Si multilayers with constant semiconductor thickness

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We report the observation of anisotropy ratio γ and interlayer-coupling-strength oscillations with variation of metal-layer thickness in Mo/Si multilayer series with constant Si-layer thickness. These oscillations correlate with previously found oscillations of T_c, R_{300}/R_n, and $dH_{c\perp}/dT$. The giant amplitude of γ oscillations makes one believe that all oscillation effects are due to the variation of the Josephson coupling. The possible origin of these unusual effects is discussed. [S0163-1829(97)05126-6]

Lately, the anomalous oscillation behavior of superconducting and kinetic parameters has been discovered on artificial Mo/Si superlattices.[1,2] For SL series with a constant Si-layer thickness ($s=25$ Å) and variable Mo-layer thickness ($d=8-200$ Å), the resistivity ρ_n, resistivity ratio ρ_{300}/ρ_n, superconducting transition temperature T_c, and the derivative of the upper critical magnetic field, $dH_{c\perp}/dT|_{T_c}$, were found to oscillate as a function of the Mo-layer thickness with a periodicity of $d=35$ Å.[1,2] The extremum positions for all dependences mentioned coincided. The T_c and the ratio ρ_{300}/ρ_n oscillated in phase (the minimum of T_c corresponded to the minimum of ρ_{300}/ρ_n and vice versa), and the oscillations of the $dH_{c\perp}/dT$ were out of phase with the both mentioned parameters. The oscillations of T_c and ρ_{300}/ρ_n on Mo/Si SL's resemble closely the thickness dependences of T_c and resistivity ratio inherent to the semimetal and metal films in the thickness range where the quantum size effect takes place.[3]

In spite of obvious similarity in the oscillatory behavior on Mo/Si SL's and on single films of Sn and Bi,[3] the explanation of the oscillations found on SL's in terms of usual quantum size effect appeared to be rather doubtful because of extremely small mean free path of electrons l in the system considered. For all samples the electron diffusion coefficient D_{el}, determined from $dH_{c\perp}/dT$ by the formula $D_{\text{el}}=4kc/(\pi e d H_{c\perp}/dT)$, appeared to be <1 cm^2/s, and thus l is only about interatomic distance. It means that the smearing of the quantum levels connected with the carrier scattering exceeds the distance between the levels, and the oscillations have to become indiscernible.[4]

The results of the investigations of the T_c vs d dependence on the single Mo films with silicon underlayers and overlayers in distinction to multilayered samples have revealed monotonous T_c increase with d.[2,5] Thus the oscillatory behavior found is the property inherent only to the layered system. For understanding of the origin of the oscillation effects, the additional experiments are necessary.

Here we report the results of parallel critical magnetic fields measurements on the same series of Mo/Si SL's. On all samples (with the single exception) near T_c, linear three-dimensional $H_{c\parallel}(T)$ dependences are observed. From the parallel critical field slope near T_c, $dH_{c\parallel}/dT|_{T_c}$, and from the slope $dH_{c\perp}/dT|_{T_c}$ the values of the anisotropy parameter γ are obtained for the samples with different wavelengths. The giant oscillations of γ are found which are in phase with the $dH_{c\perp}/dT$ oscillations. The amplitude of γ oscillations exceeds its average value. The parameter γ is usually considered as a measure of interlayer Josephson coupling strength. Thus one has to conclude that in the case of Mo/Si-layered system coupling strength can change essentially at the constant Si-layer thickness and this change is provoked only by the change in the metal layer thickness. The data on Mo/Si SL's are in obvious contradiction with those obtained on Nb/Ge SL's (Ref. 6) for which the exponential dependence of the interlayer coupling strength on Ge-layer thickness was obtained in accordance with the suggestion about quantum-mechanical tunneling through Ge layers, and no dependence on Nb-layer thickness was found. While the results on Nb/Ge SL's testify about the electrical passivity of Ge layers,[6] the oscillation behavior observed on a Mo/Si system may be considered as an evidence that some redistribution of charged carriers between metal and semiconductor layers may occur. Other possibilities to explain the unusual behavior observed are discussed.

The Mo/Si SL's consisting of 30 bilayers have been prepared by magnetron sputtering on a glass substrate at a substrate temperature $T_s=100$ °C. In addition to former work on sample characterization,[7] we recorded standard θ-2θ diffraction patterns for SL's with $d_{\text{Mo}}=49$, 70, 90, 122, and 152 Å. We observe a peak located at $2\theta\approx39.6°$, which we attribute to the Mo (110) Bragg peak. From the peak position and the half-width, we obtain $a=(3.22\pm0.03)$ Å for the Mo lattice constant and $\Delta L\approx(25\pm3)$ Å for the grain size (with relatively large errors due to the poorer signal-to-noise ratio

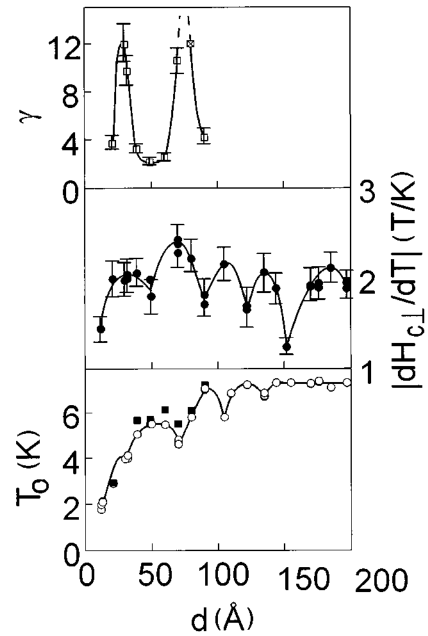

FIG. 1. T_c, $dH_{c\perp}/dT$, and γ as a function of metal-layer thickness. Error bars for perpendicular critical field derivative correspond to the transition width in a range $(0.1-0.9)R_n$. Relative error in γ values only slightly exceeds one for $dH_{c\perp}/dT$ due to essentially narrower transitions in parallel field. The width of transitions in zero field does not exceed 0.2 K, and error bars for the T_c are less than the point sizes.

for the thin Mo films). Comparing these values to $a=3.24$ Å and $\Delta L=(30\pm3)$ Å, obtained in Ref. 7 for Mo/Si SL's with $d_{\mathrm{Mo}}=176$ and 195 Å, it seems reasonable to exclude significant changes in the structural properties of the Mo layers throughout the whole sample series. For all samples Si layers are amorphous.

The resistivity measurements in magnetic fields up to 5 T were performed with an ac bridge using the four-probe method. Temperature was measured with an accuracy ±1 mK. T_c and H_{c2} were defined by the midpoints of the resistive transitions $R(T)$.

The comparison of the data for T_c and sheet resistance per layer, $R_\square$, with the results obtained on the same samples about 2 years ago revealed some aging effect. The comparison of T_c's obtained in two different runs is shown in Fig. 1. The aging effect is not very significant, and all the features of the oscillating $T_c(d)$ dependence were reproduced. The measurements of $H_{c\perp}(T)$ dependences were also repeated.

The upper critical magnetic fields $H_{c\parallel}$ and $H_{c\perp}$ as a function of temperature are shown in Fig. 2 for several Mo/Si SL's. The dependences $H_{c\perp}(T)$ on temperature are linear for all samples. The values of the slope $dH_{c\perp}/dT|_{T_c}$ obtained in recent measurements are presented in Table I. They also differ from obtained before (Fig. 1) only slightly and reveal oscillating behavior.

As for in-plane critical field $H_{c\parallel}(T)$, the different types of temperature dependence are found (Figs. 2 and 3). For some samples this dependence is linear in the entire accessible range of H. For other samples the crossover from linear three-dimensional (3D) dependence near T_c to the 2D square-root-like dependence at low temperatures is observed.

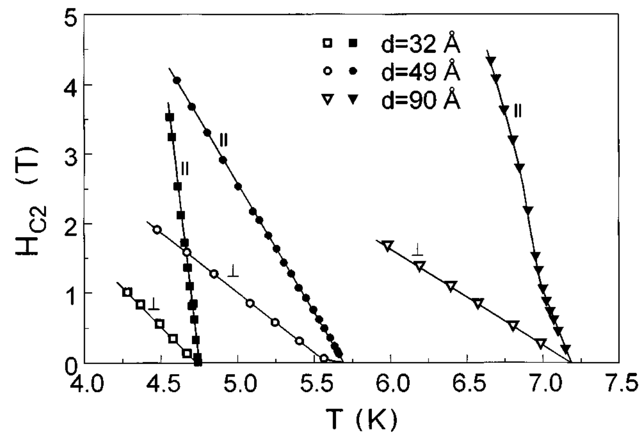

FIG. 2. Critical magnetic fields as a function of temperature for three Mo/Si SL's.

For SL's with $d=80$ Å, the 2D square-root-like dependence starts directly from T_c, testifying at the first sight about full decoupling of superconducting layers (Fig. 3). This dependence will be commented in more detail below.

The behavior of $H_{c\parallel}(T)$ described above is characteristic for many superconducting SL's of S/I type (S is a superconductor, I is a semiconductor) with varying semiconducting spacer thickness. The data demonstrating all three types of $H_{c\parallel}(T)$ dependence were obtained on V/Si (Ref. 8) and Nb/Ge (Ref. 6) SL's. The coupled 3D behavior was observed at small spacer thicknesses $s<24$ Å, decoupled 2D behavior at $s>45$ Å (Ge), or $s>70$ Å (Si). The crossover was observed at intermediate values of s. In distinction to the cited above results, on Mo/Si SL's three different types of $H_{c\parallel}(T)$ behavior are found at the same constant s value when only the metal-layer thickness changes. It means that for multilayers investigated the usual simple description of $H_{c\parallel}(T)$ behavior with the assumption about the Josephson coupling depending only on s fails. This statement is confirmed in the most dramatic way by the data of Fig. 1 where the variation of anisotropy parameter or effective mass ratio $\gamma=(M/m)^{1/2}=(dH_{c\parallel}/dT)/(dH_{c\perp}/dT)$ with d is shown. The effective mass ratio oscillates with practically the same period as other parameters of Mo/Si SL's and with a giant amplitude exceeding the average value of γ. It appears that

TABLE I. Values of the SL parameters obtained in recent measurements.

d (Å)	$\lvert dH_{c\perp}/dT\rvert$ (T/K)	$\lvert dH_{c\parallel}/dT\rvert$ (T/K)	H_{cr} (T)	$\tilde{T}_c$ (K)	γ (1)	γ (2)	γ (3)
21	2.0	7.4			3.7		
30	1.98	23.7	0.9	3.95	11.9	24.2	17.2
32	1.92	18.7			9.7		
39	1.87	6.0	1.4	5.64	3.2	11.1	18.1
49	1.7	3.7			2.2		
60	1.77	4.5			2.6		
70	1.84	19.5	0.7	5.46	10.6	9.7	10.2
80	2.13					12.5[a]	
90	1.41	5.9	1.3	6.98	4.2	3.8	4.0

[a]The way of obtaining $\gamma=12.5$ is described in the text.

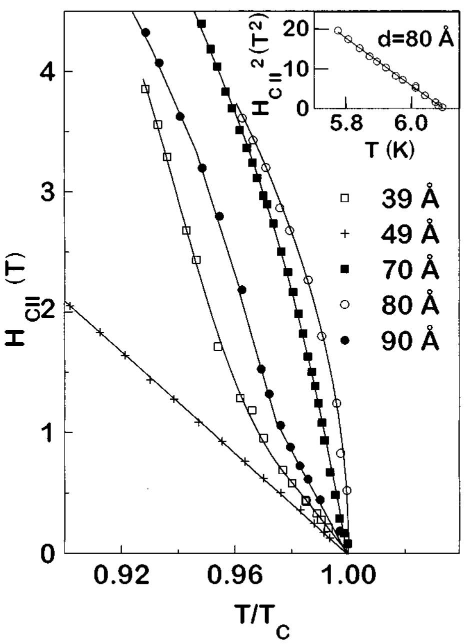

FIG. 3. Parallel critical magnetic fields as a function of the reduced temperature T/T_c for several samples. Inset: $H_{c\parallel}^2$ vs T for SL with $d=80$ Å.

the parameters $dH_{c\parallel}/dT$ and γ (their values are listed in Table I) oscillate in phase with $dH_{c\perp}/dT$ and in opposite phase with T_c. For SL's with $d=80$ Å, the 2D behavior visually starts directly from T_c and the value of γ cannot be determined straightforwardly. This fact as was mentioned before may be considered as the evidence of full decoupling of the superconducting layers ($\gamma\to\infty$).

It was shown[8] that if the dependence $H_{c\parallel}(T)$ reveals a 3D-2D crossover, the value of γ may be determined from the experimental data using the theory for S/I SL's in three independent ways. Beside the usual way of defining γ from the ratio of the critical field slopes

$$\gamma=(dH_{c\parallel}/dT)/(dH_{c\perp}/dT), \tag{1}$$

this value can be obtained from the crossover field $H_{\rm cr}$ and from crossover temperature $\tilde{T}_c$ using expressions[8]

$$\gamma=\varphi_0/\pi H_{\rm cr}D^2, \tag{2}$$

$$\gamma=[\xi(0)/D]\sqrt{2T_c/(T_c-\tilde{T}_c)}. \tag{3}$$

Here $D=d+s$ is the SL wavelength, $\xi(0)$ is the in-plane coherence length at $T=0$, and $\tilde{T}_c$ is the crossover temperature which is defined by the extrapolation of low-temperature $H_{c\parallel}(T)$ dependence to $H=0$.

The experimental values of $H_{\rm cr}$ and $\tilde{T}_c$ along with the values of γ obtained by all three methods are presented in Table I. For samples with $d=70$ and 90 Å, the agreement between the γ's obtained by different methods is rather good. But for the two samples with the smaller d ($d=30$ and 39 Å), there is pronounced discrepancy in three values, the γ's obtained from $H_{\rm cr}$ and $\tilde{T}_c$ being larger than the one obtained from the ratio of critical field slopes. It is known that in strong perpendicular magnetic fields the anisotropy parameter becomes field dependent, and it grows with H due to thermal fluctuations of vortices, weakening the Josephson coupling between layers.[9] It is not obvious whether this result might be used for explaining relatively large γ values obtained from $H_{\rm cr}$ and $\tilde{T}_c$ for SL's with the small D.

At last we would like to comment the $H_{c\parallel}(T)$ dependence on the sample with $d=80$ Å. Due to a very large slope of this curve, the first value of $H_{c\parallel}$ which could be registered is about 0.5 T. If we try to consider this field as an upper limit for crossover field, then, according to formula (2), the lower limit for γ may be defined: $\gamma_{\rm min}=12.5$. Thus, for the sample considered, the situation with the large γ value is possible as well as the situation of fully decoupled layers.

It is also worth mentioning that the absolute values of both $dH_{c\perp}/dT$ and $dH_{c\parallel}/dT$ are extremely high, and they exceed ones even for high-T_c oxides. For superconductors of the YBCO group, the $|dH_{c\perp}/dT|$ values in the range 0.4–1.9 T/K (Refs. 10,11) are reported, for Bi-based oxides in the range 0.3–1.2 T/K,[12,13] while for Mo/Si multilayers they attain values above 2 T/K. In the case of an in-plane field for the first group of compounds, the values of $|dH_{c\parallel}/dT|$ are in the range 1.4–10.5 T/K,[10,11] and for Bi-based compounds $|dH_{c\parallel}/dT|_{\rm max}$ is 14 T/K.[10] The maximum value of $|dH_{c\parallel}/dT|=23.7$ T/K is obtained for Mo/Si SL's. Thus, not only oscillations found, but also extremely high values of the critical magnetic fields in Mo/Si multilayered system may be regarded as anomalous.

Considering the results obtained, one must bear in mind that T_c oscillations are not found on single Mo films, and this fact may be treated as important evidence against the explanation based on conventional quantum size effect. In the mean-field approximation the T_c of the infinite S/I-type SL's should coincide with T_c of the constituting superconducting layers (the slight difference between T_c's of the SL's and film may be observed only at small number of bilayers $N<10$).[14] It is obvious that the mean-field GL approximation fails in the case of Mo/Si SL's as well as in some other cases. In terms of this theory the depression of T_c in Nb/Ge SL's with the growth of s (Ref. 6) and anomalously large enhancement of T_c with N on W/Si SL's (Ref. 15) cannot be explained either. It is noteworthy that on Mo/Si SL's the same correlation between coupling strength and T_c is found like in Ref. 6: the more coupling strength, the higher T_c.

The absence of T_c oscillations on single Mo films and the giant amplitude of the coupling strength oscillations on SL's enable one to suppose that oscillation effects appear only as a result of multilayering. The large amplitude of the γ oscillations and the correlation between all oscillating parameters makes one believe that other oscillation effects may be a consequence of γ oscillations.

Most probably, the oscillations of the transverse resistivity ρ_t on SL's which obviously occur when the barrier transparency oscillates should give rise to the oscillations of longitudinal resistivity ρ_l. These latter oscillations were directly observed on Mo/Si SL's. In the presence of this effect, the oscillations of T_c and $dH_{c\perp}/dT$ can be explained synonymously. According to the theory of type-II superconductors, $dH_{c2}/dT|_{T_c}=-2.55kceN(0)\rho_n$, and the fields $H_{c\perp}$ of SL's of S/I type do not differ from H_{c2} for bulk superconductors if the value ρ_l instead of isotropic resistivity is used. From the formula for $dH_{c\perp}/dT$, it follows that the derivative of the critical field should oscillate in phase with ρ_l if the density of electron states $N(0)=\text{const}$ for all samples. Most probably, this is the case, because the amplitudes of ρ_l and $dH_{c\perp}/dT$ oscillations are practically the same, and the both parameters vary with d in phase.

The T_c oscillations are also directly connected with those of ρ_l or, more precisely, with the oscillations of the sheet resistance $R_\square=\rho_l/d$. In disordered 2D superconductors, T_c should decrease with enhancement of $R_\square$ due to localization and Coulomb electron-electron interaction effects.[16] The behavior of the background T_c vs $R_\square$ dependence on the Mo/Si SL's investigated is in agreement with the results of this theory.[7] It means that the ''local'' increase of $R_\square$ at definite values of d in the respect to its background value should give rise to the diminishing of T_c. As may be expected from these considerations, the experimental T_c and ρ_l values oscillate in opposite phase.

At this stage of the investigations, it is difficult to interpret the oscillations of γ and of the Si barrier effective transparency in an inequivocal way. One possible assumption may be connected with some redistribution of the charged carriers between metal and semiconducting layers. Such effect was considered theoretically for the metal/semiconductor interface in Ref. 17. However, this theory cannot be applied to our case directly, not only because of a constant s, but also because it is valid only for clean semiconductors with clearly defined band structure. In our case Si layers are amorphous, and in amorphous semiconductors only a dense system of random local states exists.[18] The essential change of the semiconducting barrier transparency arises when resonant tunneling through the local states in the barrier occurs, and it is an attractive idea for explaining the results obtained. However, the experiments on Mo/α-Si/Mo tunnel junctions[18] have shown that at silicon thickness less than 70 Å there is direct tunneling and the contribution to conductivity from resonant processes appears only at larger Si thickness.

At last, it is worth mentioning that for Mo/Si SL's, according to the results of the investigations of the quantum corrections to the conductivity, the characteristic phase-breaking length for electron L_ϕ at low temperatures is about 200 Å;[19] i.e., L_ϕ is more than or comparable with d. Possibly, the conservation of the electron phase during its motion across the metal layer may be essential from the point of view of its tunneling through barrier.

In summary, oscillations of the anisotropy parameter with extremely high amplitude are found on superconducting Mo/Si SL's with $s=\text{const}$, while the thickness of metal layers changes. These oscillations correlate with the oscillations of the other superconducting and kinetic parameters of Mo/Si SL's previously observed. The oscillations of γ may be considered as an evidence of the oscillating behavior of Si barrier effective transparency, of the SL transverse resistivity, and of the Josephson coupling strength. Being absent on Mo single films, the oscillation effects should be considered as the exceptional property of a mutlilayered system. Now it is difficult to explain the oscillation phenomena in terms of the known solid-state physics concepts. The explanation of the quantum oscillations found seems to be a serious challenge to physicists, especially if we take into account the strong disorder in the system considered.

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