Superconducting and normal properties of the set of Mo/Si superlattices with variable Si layer thickness


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We report the results of the superconducting and kinetic parameter measurements (transition temperature $T_c$, parallel and perpendicular critical fields $H_{c1}$, resistivity in the normal state) on a set of Mo/Si superconducting superlattices with a constant metal layer thickness $d_{Mo} = 22$ Å and variable semiconducting one $d_{Si} (14-44)$ Å. Our data show a monotonic dependence of all measured parameters on $d_{Si}$. It is found that the Josephson interlayer coupling energy depends exponentially on the spacer thickness. The data obtained allowed us to determine the characteristic electron tunneling length for amorphous silicon with high precision. It is equal to 3.9 Å. Enhancement of interlayer coupling leads to the Mo/Si multilayer transition temperature increasing, in agreement with Horovitz theory and with the experimental data on high-$T_c$ materials.

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Introduction

Artificial superconducting superlattices have been of enduring interest for a long time as a perfect model system for the study of layered superconductor physics. Their tunability, i.e., the possibility of changing independently and in an arbitrary way the thicknesses of the superconducting layers and non-superconducting interlayers (spacers) and of using a wide spectrum of constituting materials, makes artificial multilayers very attractive objects for the investigation of many fundamental properties. The dimensionality effects, the role of the intrinsic anisotropy and its influence on $T_c$, the thermal and quantum fluctuation effects, etc., belong to such properties. It is especially important that for these systems the fine control over the interlayer Josephson coupling strength, which has a most profound influence on the behavior of layered superconductors, may be achieved [1,2]. In spite of an obvious importance of direct investigations of Josephson coupling, such studies are very scarce because of the necessity of preparing a large set of the variable layer thickness superlattices with a big number of bilayers $N$, very high regularity of the layering and with extremely small «steps» in the spacer thickness between neighboring samples in the set. The latter circumstance is associated with the expected exponential dependence of the interlayer coupling parameter on the insulating or semiconducting spacer thickness [1]. Such a dependence, consistent with the picture of quantum-mechanical tunneling of the charge carriers through a barrier, follows from obvious physical considerations. In Ref. 1 the
experimental data on the coupling parameter obtained on Nb/Ge superlattices were interpreted in such a way, although, unfortunately, the spread of the experimental data points was very large.

However, it has been found out recently that such a simple description of the interlayer coupling based on an usual tunneling mechanism is not applicable for all representatives of the $S/I$ multilayer class ($S$ is a superconductor, $I$ is a semiconductor). The anomalous oscillatory behavior of the superconducting and kinetic characteristics has been discovered on Mo/Si multilayers with a constant Si layer thickness ($d_{\text{Si}} = 25 \ \text{Å}$) and variable Mo layer thickness ($d_{\text{Mo}} = 8$–$200 \ \text{Å}$) [3,4]. The periodicity of all the oscillations was $\Delta d_{\text{Mo}} = 35 \ \text{Å}$. The most nontrivial effect among all observed phenomena is the oscillating behavior of the interlayer coupling strength at the constant thickness of the Si layers [5]. The background normal conductivity and superconducting transition temperature $T_c$ dependencies on $d_{\text{Mo}}$ for these samples are reasonably explained in terms of quantum interference effects in quasi-independent disordered films [6].

In spite of obviously similar features in the oscillatory behavior on Mo/Si superlattices and on single films of Sn and semimetals [7–9], the explanation of the oscillations found on superlattices in terms of the usual quantum size effect [10,11] appeared to be rather doubtful because of the quite small longitudinal transport mean free path of the electrons (about several interatomic distances) in the system under study [3]. It means that in the case of the usual quantum size effect the smearing of the quantum levels connected with the charge carrier scattering on the crystal lattice imperfections may exceed the distance between the energy levels, and the oscillations have to become practically indiscernible. The results of the investigations of the $T_c$ dependence on $d_{\text{Mo}}$ for single molybdenum film series with silicon underlayers and overlayers did confirm these expectations: in contrast to multilayered samples the molybdenum films have revealed a monotonous $T_c$ increase with $d_{\text{Mo}}$ [12]. Thus, the oscillatory behavior found is the property inherent only in the layered system.

Recently a new type of quantum size effect in metal-semiconductor superlattices has been predicted [13]. Giant oscillations of the tunnel current and the superlattice transverse conductivity should appear if size quantization of the electron spectrum in the metal layers occurs. This effect results from a sharp dependence of the probability of electron tunneling through a semiconducting interlayer on the incidence angle of the electrons moving to the interface. Due to this fact, it is expected that the effect of the size-quantization-controlled oscillations of the transverse conductivity may result. The spikes of transverse conductivity are bound to appear when the quantized electron levels pass through the Fermi level at the thickness of metal layer $d_m$ variation. One of the most interesting features of the theory considered is the prediction about the possibility of observing quantum oscillations of the transverse conductivity even in the case of rather disordered systems, provided that the electrons in the metal layers undergo soft (low-angle) scattering on the imperfections. It was shown [13] that a lifetime $\tau_c$ for the size-quantized states giving the main contribution to the tunnel current is $d_m^2/a$ times greater than a typical $\tau$ value determining the in-plane transport ($a$ is the interatomic distance). This fact is due to the structure of the size-quantized spectrum in an individual metal film. While the typical distance between quantized electron terms at a given energy is of the order of $\hbar/d_m$, the distance between the neighboring terms determining the tunnel probability is of the order of $p_F(a/d_m)^{1/2}$, i.e., noticeably larger. Under low-angle scattering conditions, when the changes in the electron longitudinal momentum are rather small, the electron transitions between these latter states are hampered, and, therefore, the lifetime is significantly enhanced.

It is clear that the predicted oscillations of the tunnel probability may at least lead to interlayer Josephson coupling strength oscillations. The conspicuous correlations between the oscillations of different physical characteristics are found on Mo/Si superlattices [3,5], and it allows one to believe that experimentally observed oscillation effects are close in its origin to the phenomenon considered theoretically.

It follows from the theory [13] that the oscillations arise at $d_m$ variation, while the thickness of the semiconductor layer $d_{\text{Si}}$ influences only the oscillation amplitude. Thus, one can expect that all multilayer parameters should depend monotonically

\* Only in the dependence of $dH_{2c}/dT$ on $d_{\text{Mo}}$, the slight hints on the oscillations with very small amplitude, may, appear, but a large spread of the experimental data does not allow us to state this positively.
on $d_s$ in a set of superlattices with a variable value of $d_s$ and a fixed value of $d_m$.

Here we report the results of the measurements of kinetic and superconducting parameters on such a set of Mo/Si superlattices.

Sample preparation and experimental methods

The superlattices were prepared by dc-magnetron sputtering onto optical quality glass substrates kept at 100 °C. The number of bilayers in all the set of superlattices was 50. The initial pressure in the deposition chamber was no worse than $10^{-6}$ Torr. Argon with the pressure 3-10^{-3} Torr was used as a sputtering gas. A time of exposition of the sample holder in the zone of every source and its displacement in another zone was regulated with a precise time controller. The thicknesses of the two layers in the bilayer were determined correspondingly by the deposition rates of both Mo and Si and by the time of exposition. In order to ensure the constant deposition rates, the stabilized power supplies for molybdenum and silicon sources and a precise gas admission system were used (with an accuracy to within 0.1% for both the gas pressure and the applied power). The deposition rates were 2-3 Å/s.

X-ray diffractometry was used for structural characterization of the superlattices. The low-angle diffractometry allowed us to determine the multilayer periods with the accuracy of 0.1 Å. For Mo/Si superlattices with the wavelengths exceeding 100 Å, which have been prepared under the same conditions, the number of satellites on the diffractograms was about 10. For short-period superlattices investigated here, this number was 4-5. These data as well as the small width of satellite lines testify to the high regularity of the layering. The separation between satellites and their position confirm the layer spacing expected from fabrication process. The relative difference of the Mo layer thickness for all the samples did not exceed 0.3 Å.

The recorded 6–20 diffraction patterns for superlattices revealed that molybdenum layers consist of small crystallites (with the size of 25–30 Å), while silicon ones are amorphous.

The low-temperature measurements were carried out in a $^4$He cryostat equipped with a 5 T superconducting coil. The temperature was measured with a carbon-glass thermometer, calibrated against a Ge thermometer in a zero field. Temperature was controlled within 1.4–10 K with the accuracy of 1 mK. Due to a small magnetoresistance a maximum deviation of temperature was about 10 mK in a magnetic field of 5 T as compared to the temperature at zero magnetic field. The resistivity measurements were performed with an ac bridge using the conventional four-probe method. Both the $T_c$ and the $H_{c2}$ were defined as the midpoints of the $R(T)$ and $R(H)$ transitions.

Experimental results and discussion

The typical dependencies of the resistance $R$ on the temperature in a range 4–100 K for Mo/Si multilayers are presented in Fig. 1. A negative temperature coefficient of resistivity was observed for all samples investigated between 300 K and a temperature where the resistance starts to decrease due to the approach to the superconducting transition. The similar $R(T)$ behavior was previously observed on Mo/Si multilayer series with $d_s$ = const = 25 Å and a variable $d_m$ in the $d_m$ range 13–90 Å [14]. Analysis of these dependencies as well as of the data on the magnetoresistance in magnetic fields perpendicular to the layers has revealed that in the Mo/Si system with disordered metal layers the quantum corrections to the conductivity are essential [14]. As the data obtained show, the same is true for the set of superlattices investigated here.

The dependence of the resistance ratio $R_{300}/R_m$ on the thickness of silicon layer $d_s$ is shown in Fig. 2. $R_m$ is the maximum resistance before superconducting transition. There is an obvious tendency towards diminishing of $R_{300}/R_m$ with $d_s$ increase which suggests that the magnitude of the quantum interference corrections to the resistivity for the multilayers with the better separated metal layers are enhanced.

Figures 2,b and 2,c demonstrate the transition temperature $T_c$ and the derivative of the perpen-

![Fig. 1](image)

**Fig. 1.** The resistance ratio as a function of temperature for some of Mo/Si samples ($1 - d_s = 30$ Å; $2 - d_s = 43$ Å; $3 - d_s = 34$ Å).
Fig. 2. The dependencies of multilayer parameters on silicon spacer thickness \(d_{\text{Si}}\): resistance ratio \(R_{300}/R_m\) (see text) (a); transition temperature \(T_c\) (b); derivative of the perpendicular critical magnetic field \(dH_{c \perp}/dT\) (c).

distinct critical magnetic field \(dH_{c \perp}/dT\) as a function of \(d_{\text{Si}}\). While \(dH_{c \perp}/dT\) does not in practice depend on \(d_{\text{Si}}\), \(T_c\) noticeably diminishes with \(d_{\text{Si}}\) increase. The \(T_c\) vs \(d_{\text{Si}}\) dependence shows a tendency to saturation at large \(d_{\text{Si}}\) values. Only the derivative of the parallel critical magnetic field and, correspondingly, the anisotropy parameter \(\gamma = (M/m)^{1/2} = (dH_{c \parallel}/dT)/(dH_{c \perp}/dT)\) exhibits a very pronounced dependence on the spacer thickness \((M/m)\) is an effective mass ratio).

In Fig. 3 the magnetic critical field dependencies on temperature are shown for the samples with different \(d_{\text{Si}}\). For the case of a field orthogonal to the layer planes the dependencies \(H_{c \perp}(T)\) are linear. Parallel critical fields \(H_{c \parallel}\) for the multilayers with the smallest values of \(d_{\text{Si}}\) are also linear with a temperature in all accessible range of magnetic fields. For the samples with \(d_{\text{Si}} \geq 34\) Å the change in the temperature dependence of \(H_{c \parallel}\) is observed at low temperatures which points out to the dimensional 3D-2D crossover. The anisotropy becomes larger with increase in the spacer thickness, and \(\gamma\) amounts to the value of 23 at \(d_{\text{Si}} = 43\) Å. The anisotropy parameter \(\gamma\) as a function of \(d_{\text{Si}}\) is presented in Fig. 4.

The theory of the critical magnetic fields for layered systems with the Josephson coupling between the superconducting layers is highly developed [2,15–18]. Using the effective-mass model of

Fig. 3. Critical magnetic fields \(H_{c \perp}(T)\) as a function of the reduced temperature \(t = T/T_c\) for two Mo/Si samples \((d_{\text{Si}} = 34\) Å: ▪️ \(- \ H_{c \parallel}(T)\); ▼ \(- \ H_{c \perp}(T)\), \(d_{\text{Si}} = 18\) Å: ■ \(- \ H_{c \parallel}(T)\); ▲ \(- \ H_{c \perp}(T)\). Inset: \(d_{\text{Si}} = 18\) Å, \(H_{c \parallel}(T)\).

Fig. 4. The anisotropy parameter \(\gamma = (M/m)^{1/2}\) as a function of \(d_{\text{Si}}\) for multilayers investigated.
Lawrence and Doniach [19], which is a good approximation for the weak field range, one can estimate the Josephson coupling energy by the formula [15]

\[ \eta_j = \frac{\hbar^2}{2m^* \gamma} . \]

(1)

Here, \( s = d_m + d_s \) is a superlattice period.

Using the experimental \( \gamma \) values (Fig. 4) one can determine the Josephson coupling parameter \( \eta_j \). The coupling parameter as a function of \( d_{Si} \) is shown in Fig. 5. As follows from the experimental data, the \( \eta_j \) decreases exponentially with an increase in the silicon layer thickness according to the relation:

\[ \eta_j = \eta_{j0} \exp \left(-\frac{d_{Si}}{d_0} \right) . \]

(2)

As it was mentioned in the Introduction, Ruggero, Barbée and Beasley [1] have found the same type of relationship between the interlayer coupling energy and the spacer thickness on Nb/Ge superlattices. From the observed exponential relationship between \( \eta_j \) and \( d_{Si} \) (Fig. 5) we obtain the \( d_0 \) value of 3.9 Å which may be considered as a characteristic tunnel length for amorphous silicon. For amorphous Ge the value \( d_0 = 8 \) Å was derived [1]. In the latter case, the accuracy of \( d_0 \) evaluation was not very high because of large spread of the data points. This spread in the experiments mentioned was, probably, inevitable because as \( d_{Ge} \) varied, the thickness of Nb layers did not remain constant like in our experiments (\( d_{Nb} \) was varied in a range 30–65 Å). It is possible that the variation of \( \gamma \) with \( d_m \) at a constant \( d_s \) value, observed on Mo/Si superlattices, cannot be excluded for \( S/I \) multilayers having different constituent materials. It may be the reason for large spread of the points.

All the above results provide a clear answer to the important issue arising in the context of the observation of the oscillation effects found on a series of Mo/Si samples with variable \( d_m \). The issue is whether the semiconductor interlayer thickness variation leads to the appearance of the oscillation effects, just as it occurs when \( d_m \) varies at constant \( d_s \). All dependencies of kinetic and superconducting parameters on \( d_s \), if any, appeared to be monotonic. Simultaneously fair evidence for the exponential dependence of coupling energy on the spacer thickness is obtained for the first time.

It should be mentioned that there are experiments on V/C layered system [20,21] where the oscillations of \( T_{c} \), \( dH_{c2}/dT \) and normal resistivity were observed with the variation of the carbon spacer thickness. These oscillation effects were explained by Kagan and Dubostvii [22], who assumed that the oscillations in the metal-semiconductor system are mainly associated not with the quantization of transverse motion of electrons in metal films, but with a change in the boundary conditions at the interface between a metal and semiconductor layers. These conditions depend on a spacer thickness. Such an effect should be expected in the case of the crystalline semiconductor interlayer. Our experimental results indicate that such effects are absent in a case of Mo/Si layered system with amorphous silicon spacers.

Another interesting question that is widely discussed [23–28] concerns the nature of the superconducting phase transition in the layered superconductors with the Josephson-type coupling and the influence of the interlayer coupling on the transition temperature \( T_c \). According to the mean field theory [29], the \( T_c \) of \( S/I \) multilayers should coincide with that of the separate superconducting layers. When the fluctuations are taken into account the situation changes essentially. The most extensive study of this issue was performed in the works of Horovitz [24–26]. It was shown that at a finite coupling the layered system should exhibit at the critical temperature the three-dimensional phase transition. The \( T_c \) value differs from that for non-coupled individual films. In the system of coupled layers the \( T_c \) is determined by the competing influence of two types of the topological excitations (the thermally excited vortices and antivortices piercing the superconducting layers which should appear in a two-dimensional superconductor in the vicinity of
the Berezinsky-Kosterlitz-Thouless transition [30–32] and the vortex loops proliferating between superconducting layers [23]). It is predicted that increased coupling should lead to the enhancement of \( T_c \). As the data of Figs. 2 and 5 show, namely, such a type of dependence of \( T_c \) on the interlayer coupling is observed in our experiments. The data on Mo/Si sample series with the variable \( d_m \) and \( d_{Si} = \text{const} \) [5] confirm such an observation as well. The minima on the oscillating \( T_c \) vs \( d_m \) dependence correspond to the \( d_m \) values where the anisotropy parameter is the largest (i.e., the coupling parameter has its minimum). As with the multilayer set investigated here, relatively small \( T_c \) variation is observed whereas the \( \eta_f \) changes almost by an order of magnitude.

The data, which are qualitatively similar to those reported in this work, were obtained on Nb/Ge multilayers [1]. There is the same trend to \( T_c \) enhancement when the Ge spacer thickness decreases. However, the latter data do not permit estimating the role of the interlayer coupling due to the interfering effect of inhomogeneities in the metal layers. The inner structure of Nb films consisting of layers with essentially different conductivity allows the authors to provide another explanation for the \( T_c \) vs \( d_{Ge} \) dependence in terms of the proximity effect. Nevertheless, according to the recent concepts of the layered superconductor physics the alternative interpretation of the experiments [1] cannot be excluded either. Considerably stronger effect of coupling on \( T_c \) is observed on the multilayers including high-\( T_c \) materials [33–35]. When superconducting layers of YBa\(_2\)Cu\(_3\)O\(_7\) are separated by the insulating layers of PrBa\(_2\)Cu\(_3\)O\(_7\), the \( T_c \) of the superlattices diminishes drastically with the increase in the insulating layer thickness, reaching the saturation at 10–20 K for the spacer thickness of 6–10 unit cells of PrBa\(_2\)Cu\(_3\)O\(_7\) [33–35]. There are other ways to control the anisotropy parameter \( \gamma \) in high temperature compounds: intercalation, variations of the oxygen concentration, doping with some specific impurities [35–37]. In all above mentioned cases the superconducting transition temperature decreases with the coupling weakening, but for YBa\(_2\)Cu\(_3\)O\(_7\) the dependence of \( T_c \) on \( \gamma \) is not universal; it is different for chain doping and for the plane doping [37]. While the relative \( T_c \) change in YBa\(_2\)Cu\(_3\)O\(_7\) is quite noticeable with the variation of the oxygen deficiency, its change at plane doping (the variation of Zn concentration) is far greater. All existing experimental data are in good qualitative agreement with the predictions of the theory [24–26], but a more detailed comparison with theory will be interesting. For the present set of Mo/Si superlattices it is difficult to make such a comparison because the necessary conditions of the constant normal resistivity through all the set of the samples were not fulfilled.

**Conclusions**

1. The exponential dependence of the Josephson coupling energy on the silicon spacer thickness is found on Mo/Si superlattices with the constant thickness of molybdenum layers and variable \( d_{Si} \). The characteristic electron tunneling length for amorphous silicon determined from the experimental data is equal to 3.9 Å.

2. All the dependencies of superconducting and kinetic parameters on the Si interlayer thickness (if any) are monotonic according to the expectations based on the theory [13].

3. The diminishing of the Josephson coupling parameter leads to a decrease in the superconducting transition temperature, in agreement with the theory of Horovitz [24–26] and with the experimental data on high temperature materials.

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