Disorder and superconductivity in Mo/Si multilayers

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T_c vs $R_\square$ dependence in thin Mo films and Mo/Si multilayers is simultaneously of three-dimensional (3D) and 2D nature: the main effect of disorder consists in essential increase of "bulk" T_{c0} which appears to exceed many times T_{c0} for pure Mo. The x-ray data enable one to explain the origin of T_{c0} enhancement in relatively thick layers. The depression of T_c with increasing sheet resistance was observed on Mo/Si multilayers in agreement with the theory of superconductivity in disordered 2D metals. It is found that $T_c(R_\square)$ dependence for multilayers is quite close to one for single Mo films.

Recently, the interplay between disorder and superconductivity has attracted considerable interest. It is known that in disordered metals quantum interference effects are essential giving rise to unusual temperature dependence of resistivity and magnetoresistivity. According to the recent theories[1,2] these effects in two-dimensional (2D) systems have to influence also the superconducting transition temperature T_c. The superconductivity is suppressed with the increasing sheet resistance $R_\square$. $R_\square=\rho/L$ is considered as a quantitative characteristic of disorder in the 2D case (here ρ is the resistivity, L is the thickness of the metal film). These expectations were confirmed in the experiments on thin Mo-Ge films.[3] For the sheet resistance $R_\square<500\ \Omega$ rather good agreement with the perturbation theory[1] was obtained with the use of the single fitting parameter $g^*N(0)$. However in the wider $R_\square$ range up to complete suppression of superconductivity the experimental data agree better with the results of the more sophisticated theory of Finkel'stein[2] obtained by renormalization-group analysis of the electron gas in the presence of disorder. The similar T_c depression with increasing $R_\square$ per layer was also observed in the multilayered system NbTi/Ge.[4] Because data on $H_{c2||}(T)$ dependences for these superlattices show decoupled 2D behavior, it is quite natural that $T_c(R_\square)$ dependence also has a two-dimensional character. However, 3D behavior was also observed for many multilayers, and this property cannot be considered as common for all multilayers of the S/I type (S is superconductor, I is a semiconductor). Coupling strength and the dimensionality in the superconducting multilayers depend on the material and the thickness of the I layers. Therefore there is no universal rule determining T_c behavior for multilayered systems.

Here we report on the change of T_c with the sheet resistance for the multilayered system which is very unusual from many points of view. The measurements have been carried out on Mo/Si multilayers with constant Si interlayer thickness 25 Å and variable Mo layer thickness 12–200 Å. In these multilayers T_c at $L_{\mathrm{Mo}}>120$ Å exceeds 7 K,[5] though for bulk molybdenum T_c is only 0.92 K. Other interesting anomalies are found for this artificially layered system: in the above-mentioned series of samples resistivity ratio R_{300}/R_n, transition temperature T_c, and the derivative of the upper critical field $dH_{c2\perp}/dT|_{T=T_c}$ were found to oscillate as a function of Mo layer thickness with a periodicity of $L\simeq 35$ Å.[5] The T_c oscillations are seen against the background of a rapid change of transition temperature with L_{Mo} at small Mo layer thicknesses, as well as against the background of constant T_c at larger L_{Mo}.[5] Recently the temperature dependence of resistivity and magnetoresistivity has been investigated on many samples of the same series of Mo/Si multilayers,[6] and quantum corrections to conductivity and magnetoconductivity have been determined. The analysis of these corrections on samples with $L_{\mathrm{Mo}}=32$–90 Å showed that at $T<25$ K all these multilayers display 2D behavior characteristic for single-metal layers.

Here we present the experimental evidence that the background dependence T_c vs L_{Mo} for Mo/Si superlattices may be also explained in terms of localization theory[1,2] for disordered 2D metals despite nonstandard behavior of many properties of this system. However, the most interesting issue consists of the essentially more complicated influence of disorder on superconductivity than in the usual 2D systems studied in Refs. 3, 4. According to Ref. 1 the interplay between the interaction and disorder leads to the enhancement of the Coulomb repulsive interaction and to the depressing of the electronic density of states $N(0)$. Both effects contribute to the depression of T_c, however, the influence of the $N(0)$ change for dirty 2D superconducting films is negligibly

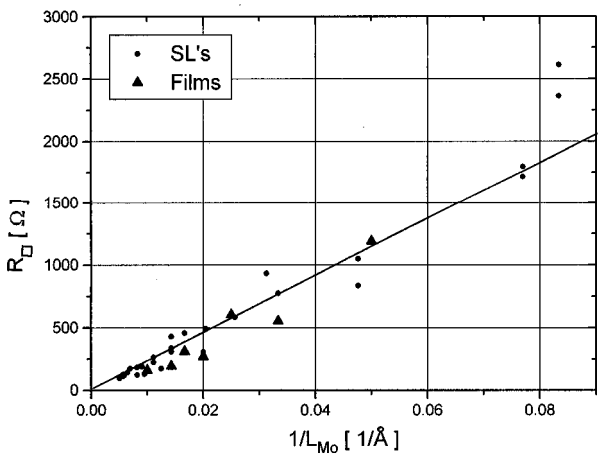

FIG. 1. $R_\square$ vs L_{Mo}^{-1} for Mo/Si multilayers (●) and for single Mo films (▲). The solid line is the constant resistivity 230 $\mu\Omega$ cm.

small as compared with that of the increased Coulomb interaction.[3] In Mo/Si multilayers the main effect of disorder consists in an essential increase of "bulk" T_{c0} for Mo by virtue of considerable $N(0)$ change which is intrinsically a 3D effect and is due to the changed crystal structure in thin layers. Due to the two-dimensionality of the samples under study, this effect is accompanied by T_c depression with increasing sheet resistance connected with enhanced electron-electron repulsion characteristic for weakly localized systems. Occurring simultaneously, both the 2D and 3D effects give rise to the observed $T_c(R_\square)$ dependence which extrapolates at large thickness to the high T_c value 7.3 K. Transition temperatures for single Mo films are also high and they reveal the same dependence on the sheet resistance as the background T_c for multilayers. Two-dimensional T_c behavior is observed along with 3D properties displayed by some of the Mo/Si multilayers in the small wavelength range.

Mo/Si multilayers were prepared by magnetron sputtering on the glass substrates kept at $T=100\,^\circ$C. The number of bilayers in all samples is 30. Other details of Mo/Si multilayer preparation and characterization are described in Ref. 5. Alongside with multilayered samples the thin films of Mo have been also studied. The film thickness has varied in the range 20–100 Å. Each film had silicon under- and overlayers of thickness 100 Å. The T_c values were determined resistively, using the four-point contact geometry. The transition width did not exceed 0.3 K (0.1–$0.9R_n$). T_c's were determined at the middle point of resistive transitions $R(T)$, i.e., at $R=0.5R_n$.

When studying localization and Coulomb interaction effects on T_c it is important to deal with the samples having the same bulk resistivity. It allows one to avoid the influence of an additional mechanism leading to T_c variation through resistivity change.[7] Namely, such a situation takes place for samples investigated. This fact is illustrated by Fig. 1 where $R_\square$ dependence on L_{Mo}^{-1} is shown. The sheet resistance was calculated from the measured resistance per single layer. If the resistivity $\rho(L_{\mathrm{Mo}})=$ const for all the samples, the experimental points have to lie on a straight line with the slope equal to ρ. As Fig. 1 shows, there is a scattering of data with respect to such a straight line which is connected with the oscillation effect mentioned above and with the error in the

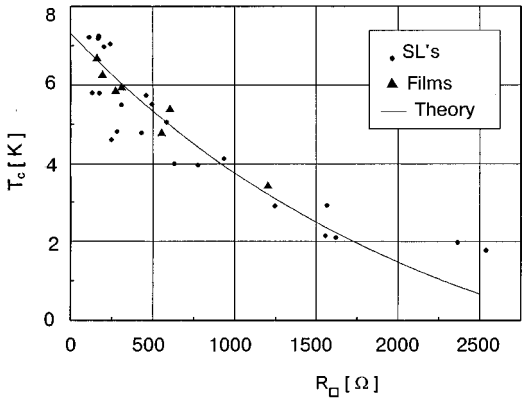

FIG. 2. T_c vs $R_\square$ for Mo/Si multilayers (●) and for Mo films (▲). The solid line is a fit to Eq. (1). Fitting parameter $\gamma^{-1}=-7$, $T_{c0}=7.3$ K.

determination of the sample cross section. Nevertheless all data points concentrate around the same line corresponding to the resistivity $\rho=230$ $\mu\Omega$ cm. From previous measurements of $dH_{c\perp}/dT$,[5] it follows that the electronic diffusion coefficient

$$D=-4kc/(\pi^* dH_{c2\perp}/dT|_{T=T_c})$$

for all multilayers does not exceed 1 cm^2/s. It means that the mean free path of electrons is ~ 5 Å, i.e., essentially less than layer thickness for the samples with $L_{\mathrm{Mo}}>20$ Å, and one need not take into account finite-size corrections to the resistivity. As Fig. 1 shows, the films and multilayers have the same bulk resistivity.

The transition temperature dependence on the sheet resistance $R_\square$ for multilayered samples and thin Mo films is presented in Fig. 2. In this figure the theoretical dependence $T_c(R_\square)$ obtained with the Finkel'stein formula[2]

$$\frac{T_c}{T_{c0}}=\exp(-1/\gamma)\left[\left(1+\frac{(t/2)^{1/2}}{\gamma-t/4}\right)*\left(1-\frac{(t/2)^{1/2}}{\gamma-t/4}\right)^{-1}\right]^{1/\sqrt{2t}} \tag{1}$$

is shown as a solid line. In this formula $\gamma=1/\ln(kT_{c0}\tau/\hbar)$, $\gamma<0$, $t=e^2R_\square/2\pi^2\hbar$, T_c is the transition temperature of the film, T_{c0} is the bulk value of transition temperature for the material considered, and τ is transport relaxation time. In calculations by formula (1) we used γ as a fitting parameter. The best fit is achieved with $\gamma^{-1}=-7$. As the bulk T_{c0} value we used the magnitude of T_c for the superlattices with the wavelengths exceeding 170 Å (i.e., 7.3 K). The absence of a ρ_n systematic dependence on L_{Mo} allows us to suppose that T_{c0} values for all samples investigated are practically the same, and the universal T_c vs ρ_n dependence obtained for disordered 3D systems[7] gives no contribution to experimentally observed T_c's. Due to this fact, the agreement with the theory[2] is rather good, and the value of $\tau=0.95\times10^{-15}$ sec obtained with $\gamma^{-1}=-7$ and $T_{c0}=7.3$ K is quite reasonable. The high value of the bulk transition temperature will be commented on below.

It should be stressed that the T_c vs L_{Mo} dependence for single Mo films and the background T_c vs L_{Mo} dependence for Mo/Si multilayers almost coincide. Such a coincidence

should be expected for S/I superlattices in the mean-field theory approximation.[8] Although it is evident that in this approximation some subtle features of the multilayer T_c behavior cannot be explained (in particular, the T_c oscillation effect on the Mo/Si multilayers), nevertheless most of experimental data confirm this expectation.

The main distinction of data represented here from other experimental results concerning the depression of T_c in disordered films, consists of the requirement of using 7.3 K as T_{c0} for the bulk material, exceeding by almost an order of magnitude the bulk T_{c0} for Mo. Thus the behavior observed corresponds to the necessary assumption that electronic properties of single and multilayered Mo films differ essentially from the properties of pure bulk molybdenum.

The point of view exists[9] that strong disorder influences superconducting T_c in the transition metals mainly through the change of the electronic density of states because of the specific structure of $N(\varepsilon)$ dependences in these materials [sharp peaks in the $N(\varepsilon)$]. Lattice disorder leads to a smearing of $N(\varepsilon)$ and to a displacement of the Fermi level. Hence for Nb, V, and Ta in which the Fermi level coincides with the $N(\varepsilon)$ peak, T_c has to decrease with disordering, while in Mo and W, in which the Fermi level lies in the vicinity of $N(\varepsilon)$ minimum, the values of T_c must increase. This idea finds confirmation in many experiments on amorphous and ion-implanted films of transition metals (see Ref. 10 and references therein).

It is worthwhile noting here that a high transition temperature is observed for Mo films even at quite large thickness. $T_c=8$ K is reported for a film with $L_{\mathrm{Mo}}=300$ Å.[11] According to the data of Ref. 12, $T_c=4.8$–7.2 K for Mo films in the thickness range 1–5 μm. For thin Mo film ($L=100$ Å) $T_c=3.1$ K is obtained in Ref. 13. For Mo/Si multilayers investigated in Ref. 14, the maximum T_c value is 6.1 K at $L_{\mathrm{Mo}}=L_{\mathrm{Si}}=12$ Å. Thus the large enhancement of T_c is often observed in evaporated Mo films which are obtained without any contact with Si and in Mo/Si superlattices. The same is true for W films and multilayers where T_c enhancement is even more dramatic. Transition temperatures in the range 2.7–4.2 K are observed[15] for W/Si multilayers with different wavelengths 20–110 Å, while bulk T_{c0} for W is 0.01 K.

On the other hand, low T_c values for Mo films are reported in Refs. 16 and 17: 0.62 K for a 3000 Å thick film and $T_c\sim 1$ K for 1000–1500 Å thick films (for the latter films $\rho(300\ \mathrm{K})=15\ \mu\Omega$ cm). Thus the literature data concerning the T_c of evaporated Mo films are rather controversial, and the reason for so large a discrepancy is not clear especially because in many cases the detail structure characteristic is absent.

Some explanation for these data may be obtained from our x-ray-diffraction examination of multilayered samples prepared at different substrate temperatures. These data for two samples with equal wavelengths ($L_{\mathrm{Mo}}=195$ Å, $L_{\mathrm{Si}}=25$ Å) are presented in Fig. 3. Superconducting transitions for the same samples are shown in the inset to Fig. 3.

For sample A from the series of multilayers investigated ($T_{\mathrm{subst}}=100$ °C) on an x-ray diffractogram, only one wide peak corresponding to the (110) line of Mo is present which is shifted to a smaller angle as compared with the bulk Mo. From analysis of this diffractogram it follows that lattice

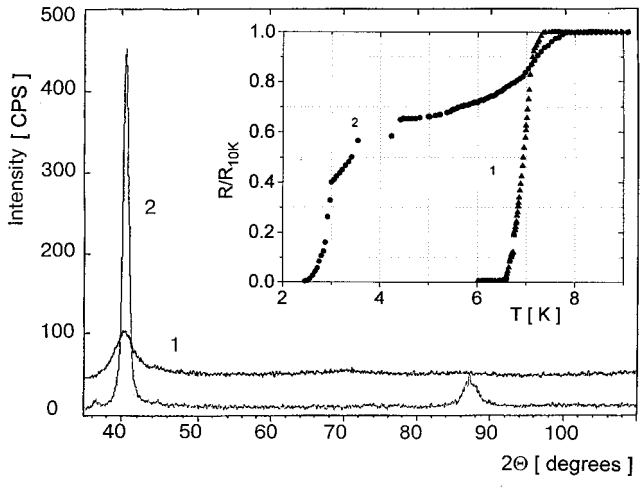

FIG. 3. X-ray diffractograms for two Mo/Si samples: (1) sample A, (2) B. Small peak at $2\theta\simeq 36°$ is connected with $\lambda_{\mathrm{Cu}\text{-}\beta}$ radiation. Inset shows the resistive transitions for the same samples, $A(1)$ and $B(2)$.

parameter is equal to $a_0=(3.246\pm 0.005)$ Å ($a_0=3.147$ Å for bulk Mo). Quite probably, this difference in a_0 is connected with the stretching macrodeformation of the crystal lattice in the multilayer. The strain is about 3.1%, i.e., rather large. The grain size, as it follows from the peak half-width, is about 30 Å. The same only peak is seen on the diffractogram for the multilayer from series A with $L_{\mathrm{Mo}}=175$ Å.

For sample B prepared at the elevated substrate temperature two peaks are seen corresponding to the (110) and (220) lines of Mo, lattice parameter $a_0=(3.163\pm 0.001)$ Å differs essentially less from one for the bulk Mo, the mean grain size is (170 ± 20) Å, i.e., about Mo film thickness. These data testify about better crystallinity of sample B and about texture growth of Mo layers [(110) planes are oriented orthogonal to the layers]. For sample B, the transition temperature defined by criterion $R=0.5R_n$ is only 3.5 K.

Thus, taking into account these data and $T_c=8$ K for the amorphous Mo film,[11] one can detect a definite correlation between transition temperature and disordering in relatively thick films where two-dimensional effects do not yet operate: the more disorder, the more T_c. This correlation enables one to suppose that disorder leads to the change of electronic structure in accordance with the considerations of the one-electron approximation[9] and to the enhancement of "bulk" T_{c0}. This assumption is confirmed by the data on Mo films implanted with heavy ions which show T_c increasing with the increasing of the irradiation dose.[18] The conclusion that disordering entails T_c enhancement in Mo films is also supported by the fact that in Mo films low T_c goes together with small bulk resistivity[17] and vice versa.

Thus "bulk" properties of Mo films as well as of Mo layers in Mo/Si multilayers are essentially modified by disorder. The bulk resistivity for all samples investigated is practically the same, and T_{c0} values also coincide. It allows us to study the influence of disorder on the superconductivity in the 2D case without interfering bulk effects. Two-dimensional behavior of T_c on $R_\square$ dependence is in accordance with low-temperature two-dimensional behavior of resistivity and magnetoresistivity on Mo/Si multilayers in the

normal state.[6] With respect to all quantum interference effects, the metal layers in Mo/Si superlattices with $L_{Si}=25$ Å behave like decoupled films.

It is noteworthy, however, that some of the Mo/Si multilayers investigated reveal also 3D behavior. This concerns, in particular, the behavior of the ''in-plane'' critical magnetic fields $H_{c2\perp}(T)$.[5] From the observations mentioned it follows that coupling strength in the multilayered system cannot be considered a universal characteristic with respect to different physical properties.

In conclusion, we have shown that T_c vs $R_\square$ dependence in thin Mo films and Mo/Si multilayers is simultaneously of 3D and 2D nature connected with disorder and the influence of the Coulomb interaction effect in a strongly disordered medium. In distinction from more usual cases studied in Refs. 3 and 4, the main effect of disorder is the enhancement of bulk T_{c0} stipulated by the change of the electronic structure. This 3D effect is accompanied by a 2D effect, i.e., by T_c depression with the rise of sheet resistance as L_{Mo} diminishes. This latter effect is in a good agreement with the localization and electron-electron interaction theory for two-dimensional superconductors[2] if one uses as the ''bulk'' T_{c0} value 7.3 K, taking into account the modified electronic structure of ''bulk'' sputtered molybdenum. Background T_c dependence on sheet resistance per layer for Mo/S superlattices practically coincides with T_c vs $R_\square$ dependence for single Mo films. It means that in the first approximation the conclusion of the mean-field theory[8] about the coincidence of S/I superlattice T_c with the transition temperature of constituting superconducting films, is confirmed. Despite the 2D behavior of T_c in superlattices with small wavelength, some of them reveal 3D coupled behavior below T_c in a parallel magnetic field. It is evidence that the coupling strength in the multilayered systems cannot be considered as a universal parameter for describing different physical phenomena.

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