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Effect of increasing disorder on superconductivity of Mo/Nb superlattices

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Abstract
We investigated the superconducting properties of Nb/Mo superlattices (SLs). The structural changes as a function of Nb and Mo layer thickness allow us to investigate the effect of disorder on the superconducting properties in a controlled fashion. Systematic structural studies provide quantitative measures of disorder parameters, such as roughness, interdiffusion, and strain, which allow separating their effect on the individual superconducting layers. The Mo critical temperature does not change as the layer thickness decreases below its coherence length. Thus, the SL critical temperatures in the presence of disorder and proximity effects can be modeled by considering only the effects of the Nb mean free path and coherence length. With increasing layer thickness, the SL critical temperatures approach Nb bulk values. Contrary to expectations the $T_c$ of Mo remains below the Nb $T_c$. We discuss the results using existing theories based on Coulomb repulsion or changes in the density of states at the Fermi surface as a function of disorder. Questions about current understanding of the effect of disorder on superconductivity arise from the results.

Keywords: superconductivity, superlattice, disorder, sputtering

(Some figures may appear in colour only in the online journal)

1. Introduction

An avenue for the engineering of novel physical properties and new physical mechanisms was originally proposed in semiconductor-semiconductor [1], semiconductor/metal [2, 3] and metal/metal [4] superlattices (SLs). As with all modern materials produced by artificial means, disorder plays an important role. In general, disorder of a thin layer in the presence of roughness, interdiffusion, and lattice strain increases with decreasing layer thickness. As a consequence, in many cases disorder can also strongly modify and determine the materials properties.

A reduction of layer thickness contributes to increasing disorder in the layer in several ways. Thinner layers show larger crystalline disorder due to strain and interface defects. Secondly, roughness and interdiffusion have a larger impact in the properties of the material when the layers are thin. Lastly, a reduced layer thickness limits characteristic length scales of the materials (mean free path, penetration depth or coherence length) and effectively induces disorder in the electronic properties of the layers.

Theoretical studies [5] claim that the superconducting critical temperature in dirty superconductors (SCs), i.e. systems with a mean free path substantially smaller than the coherence length, should not be affected by 'potential scattering' e.g. non-magnetic impurities. However, strong dependencies of the critical temperature as a function of disorder are observed in hundreds of SC. To overcome this contradiction, it is argued that the critical temperature is not intrinsically dependent on the disorder, but is caused by different phenomena in each case.

For Nb, the superconducting critical temperature ($T_c$) decreases from 9.2 K to below 2 K with increasing structural and substitutional disorder, for example metallic impurities or absorbed residual gas [6]. A similar behavior is observed in other SC, for example bismuth [7–9]. In contrast, the behavior...
of Mo as a function of disorder is opposite and the $T_c$ increases from 0.9 to 8 K with increasing disorder [10, 11]. These deviations from Anderson’s theorem [5] are explained by a disorder induced smearing of the density of states (DOS). Nb DOS has a maximum at the Fermi energy, while Mo has a minimum. The smearing of the DOS reduces the DOSs in the case of Nb, and increases it in the case of Mo. Consequently, $T_c$ depends on the DOS and not directly on disorder [12]. However, Finkel’stein argued that the effect of disorder on the Coulomb repulsion is not properly addressed in Anderson’s calculations [13].

In order to investigate the mechanisms of superconductivity in disordered systems, we use the expected opposite behaviors of Nb and Mo when incorporated into a single SL. Within a SL, the layer thickness and all other disorder parameters can be controlled and measured precisely and have the same magnitude in both materials. Our results indicate that Finkel’stein’s prediction as a function of disorder explains quantitatively the experimental results. However, the mechanisms behind the changes of the superconducting properties of Nb and Mo with layer thickness are not straightforward, and the results suggest that the understanding of the effects of disorder is incomplete.

### 2. Experiment

The superconductivity of Nb/Mo SLs as a function of layer thickness is investigated using transport, magnetization and magnetic field modulated microwave spectroscopy (MFMMSS). Each sample is designed with different amount of disorder. The interface roughness is intentionally minimized to provide a well-defined layer thickness which allows to separate the properties of Nb from Mo. The reduction of mean free path, coherence length and the increase of crystalline disorder due to strain are expected to be the main contribution to disorder as the layer thickness is reduced. We find a systematic dependence of the SL critical temperatures, with layer thicknesses, and interface roughness extracted from x-ray reflectivity (XRR). These results are compared to a simple model which has one essential input; the electronic mean free path obtained from independent transport measurements. Our results show that Nb dominates the superconducting behavior and Mo only reduces the SL $T_c$ through the proximity effect. Even in the presence of large amount of disorder (SLs grown at 100 K), the Mo $T_c$ remains below that of Nb contrary to earlier expectations. Therefore, earlier findings explaining Mo $T_c$ enhancements as due to disorder may have a different origin.

Nb/Mo SLs were grown by RF sputtering from two targets arranged in a confocal configuration, i.e. the sample was kept stationary while depositions of materials were alternated using mechanical shutters. The growth process was optimized so as to minimize interface roughness and maximize $T_c$ for each individual layer thickness. Over 40 samples were grown, to optimize the growth parameters. The final set of samples was grown at room temperature (RT) at an RF power of 200 and 300 W for Nb and Mo, respectively. The chamber base pressure was $1 \times 10^{-7}$ Torr, while the pressure during growth was kept at 2 mTorr of Ar. The $T_c$ of 100 nm thick Nb layers grown under these conditions was 8 K, while the critical temperature of 100 nm Mo layers was below 2 K. In the SLs, ten bilayers with equal Nb and Mo thickness (1–23 nm) in each stack were grown. In order to assess the effect of crystalline disorder induced by low temperature growth a 100 nm Mo film and a SL sample with layer thickness of 2 nm were grown at a substrate temperature of 100 K.

The samples were structurally characterized by XRR and x-ray diffraction (XRD) using a Rigaku Smartlab operating with CuKα radiation. The fitting and simulation of the reflectivity and diffraction data was performed using Motofit and Suprex software [4, 14–16]. The SC transition was investigated using a SQUID magnetometer (Quantum Design MPMS) and MFMMSS [17]. In addition, we obtained the critical currents from transport measurements.

### 3. Results

The XRR measurements are summarized in figure 1(a). The data was fitted with a slab model using the nominal Nb and Mo material densities. Interfaces between the two materials were modeled using an error function profile of the materials scattering length densities (SLDs) [18]. The fitted interface roughness, which is defined as the standard deviation of the error function [18], varies between 0.5 and 1 nm towards larger layer thickness. The fitted layer structures (figure 1(a)) agree with the nominal values within the roughness values.

The $\theta$–$2\theta$ XRD measurements ($k$ vector perpendicular to the sample surface) and the simulations are shown in figure 1(b). The sample with the largest period could not be simulated using Suprex due to software restrictions for the maximum thickness. The diffraction data of the 1 nm sample shows a single peak located between the Mo and Nb diffraction angles. The diffraction remains centered between Nb and Mo for intermediate layer thicknesses, with additional satellite peaks related to the SL structure. Only in samples with layer thicknesses above 5 nm, diffraction maxima corresponding to the Mo and Nb BCC (110) peaks are observed. The results show that the layers have a BCC structure textured along the (110) direction even for the lowest thickness, but strain values vary as a function of thickness. The out-of-plane lattice distance of Nb is initially under tensile strain and Mo under compressive strain (figure 1(c)). The lattice parameters approach bulk values as the layer thickness increases. The relaxation of the layers is complete for layer thicknesses above 5–6 nm.

The critical temperatures were measured by magnetometry, and confirmed by MFMMSS (data not shown) and transport measurements in selected samples. The MFMMSS technique, capable of detecting minute amounts of SC embedded in a non-superconducting matrix [17], proves the absence of minority phases in the Nb/Mo SLs. A single transition is observed at a temperature similar to those measured by magnetometry. Therefore, the averaged $T_c$ measured
by magnetization and transport is a representative measure of the full SL and not of localized regions within the structure. The MFMMS further confirms that Mo does not become superconducting below the $T_c$ of Nb. The critical currents were measured for the samples with 2, 19 and 21 nm layer thicknesses using 500 $\mu$m wide and 3 mm long bridges fabricated by photolithography and reactive-ion-etching. The results as a function of temperature are shown in figure 2. Theoretically, the temperature dependence of the critical current close to the critical temperature is expected to be given by $[19, 20]$: 

$$J_c \approx 4J_c(0)\left[1 - \frac{T}{T_c}\right]^{3/2}. \quad (1)$$

Fitting the resistivity data to equation (1) gives an estimate of the critical current at 0 K and the $T_c$, which are consistent with independent magnetometry results within the accuracy of the measurement ($\pm 0.5$ K). The critical currents at 0 K are $3 \times 10^5$ A cm$^{-2}$ for the two samples with larger layer thickness and $1 \times 10^6$ A cm$^{-2}$ for the sample with 2 nm layer thickness. These values are consistent with the ones previously reported for pure Nb, $\sim 5 \times 10^6$ A cm$^{-2}$ [21].

$T_c$ as a function of layer thickness obtained from magnetometry is shown in figure 3. The $T_c$ increases with the layer thickness until it saturates above 8 nm layer thickness. This shows that the strain of the SL determined in figure 1 does not affect the $T_c$. No increase of the critical temperature is observed in the low temperature growth, even though larger crystalline disorder is expected. Instead, the samples grown at 100 K show similar critical temperatures and roughness values to the ones grown at RT. This is in contrast to previous
therefore suggest that the $T_c$ of Mo never exceeds the $T_c$ of Nb despite the increase of disorder due to the reduction of layer thickness. [22] shows a decrease of the critical temperature of Mo layers as the thickness of the layers is reduced, however in that particular case the superconductivity of Mo is most probably related to the fact that the Mo layers are amorphized by the Si used as buffer and capping layers.

4. Discussion

The observed critical temperature in the SLs can be compared to values expected from model calculations relating $T_c$ and disorder. Here we calculate the critical temperature for a Nb/Mo SL, assuming that Nb is superconducting and that Mo with a critical temperature much lower than Nb acts as a normal metal. The calculations take into account disorder and the proximity effect between superconducting Nb and metallic Mo.

The theoretical models used here are only based on the changes in the mean free path ($l$) and coherence length ($\xi$) due to disorder and proximity between Nb and Mo. The critical temperature of Nb as a function of disorder can be estimated from our resistivity measurements, and is related to both $l$ and $\xi$. The product of the RT resistivity ($\rho$) and mean free path ($l$) is a constant, as it is predicted by transport theory [27] and estimated for pure Nb from the data from Asada and Nose [28], $\rho \times l = 1.5 \times 10^{-11} \Omega \text{ cm}^2$. From this we calculate a mean free path of our samples between 4 and 10 nm. The coherence length of Nb is estimated using $\xi = 0.852(\xi_0 \times l)^{1/2}$, where $\xi_0 = 38$ nm is the coherence length in the clean limit [29]. The estimated coherence lengths for the superconducting SL are between 10 and 15 nm.

In order to take into account the effect of disorder we use a model which includes renormalization group corrections in the analysis of the electron gas [13]. The interplay between Coulomb interaction and homogeneous disorder increases the Coulomb repulsion, decreasing $T_c$. The model proposed by Finkel’stein predicts the dependence of $T_c$ to the mean free path taking into account the Coulomb repulsion:

$$T_c^d = \exp \left( -\frac{1}{\gamma} \left[ \left( 1 + \left( \frac{\gamma}{\gamma - 1/4} \right)^{1/2} \right) \times \frac{1}{T_C^0} \right] \right)$$

with $t = \left( e^2 / 2 \pi \hbar \right) R$ and $\gamma = 1 / \ln \left( T_C^0 / \tau \right)$, $e$ the electron charge, $\hbar$ Planck’s constant, $R$ the sheet resistance and $T_C^0$ the critical temperature in the absence of disorder. The mean-free-path time ($\tau$) was estimated using the calculated mean free path and assuming a Fermi velocity $1.37 \times 10^5 \text{ m s}^{-1}$ [27]. It is important to note that this model considers the enhancement of Coulomb repulsion due to disorder.

$T_C^0$ is chosen to be 7.5 K, close to the maximum experimental $T_c$ of 100 nm Nb layers grown under similar conditions to the SLs. The $T_c$ obtained from equation (2) alone disagrees with our data (figure 3). This implies that disorder is not the only mechanism acting in SL samples and different influences on $T_c$ must be taken into account. Therefore in addition to the disorder in the Nb layers, we include the proximity effect between Nb and the metallic Mo.

The $T_c$ of a SC decreases when in contact with a normal metal. A general description for normal metal–SC systems was developed by DeGennes and Guyon, and subsequently modified by Werthamer (DGGW-model) [30, 31]. The DGGW-model is based on the Green’s functions study of the spatial dependence of the electron–electron interaction as a function of the superconducting gap. This model is dependent on the coherence lengths and the thickness of the two materials. In this model the critical temperature of a SC sandwiched between two thick normal metal layers with thickness $D_n \gg \xi$ is given by:

$$d_s = \frac{\pi/2}{1 - \theta_p^0 - \frac{1}{\sqrt{\theta_p^0}}} \cot^{-1} \left[ \left( \frac{\theta_p^0}{\theta_p^0} \right)^{1/2} \right]$$

with $\theta_p^0 = T_C^s / T_C^0$ and $d_s = D_s \left( \hbar v_F / \pi l \sigma_{\text{B}} T_C^0 \right)^{1/3}$, where $D_s$ represents the thickness of the superconducting layer, $v_F$ is the Fermi velocity, $l$ the mean free path of the superconducting layer, $\sigma_{\text{B}}$ the Boltzmann constant, and $T_C^0$ the original critical temperature of the SC when not in proximity with a normal metal. Equation (3) was solved using numerical methods.

If the thickness of the superconducting layers is comparable to its coherence length, the critical temperature can be calculated by applying a correction to the solution of equation (3). The corrected critical temperature ratio...
is given by:

\[ \theta_P \cong 1 - \left( \frac{t}{\theta_{0}^d} \right) \left( 1 - \theta_{0}^p \right) \tanh(D_n/\xi). \] \hspace{1cm} (4)

By solving equations (3) and (4), we can calculate the critical temperature after taking into account disorder and proximity using \( T_C^p = \theta_P \times T_C^d \). Figure 3 shows a good agreement between the model \( T_C^p \) and the experiment. A \( T_{CO} \) of 8 K, as observed in 100 nm Nb films, leads to a small shift along the \( y \)-direction but does not affect the shape of the curve. This shows that the \( T_C \) dependence of the SL can be modeled assuming that Mo is a normal metal in proximity with superconducting Nb. This implies that the superconducting behavior of the SLs is dominated by the Nb superconductivity. This comparison does not contain any contribution from superconducting Mo in spite of the short mean free path expected due to interfacial scattering. The very short Mo mean free path is not sufficient to increase its critical temperature above 2 K.

Since the absolute value of the interface roughness varies from sample to sample, a dependence of the \( T_C \) on roughness might be expected. The roughness value effectively defines a layer thickness that is highly affected by interdiffusion and breaking of the lateral homogeneity due to height variations. Structural refinement of the SLs provides a roughness parameter which is characterized as the width of the Gaussian profile between the SLDs at the interface between the two materials. Therefore, XRR does not distinguish between interdiffusion and height variations at the interface. The behavior of roughness as a function of layer thickness is similar to the prediction for Stranski–Krastanov growth [32] which is similar to the critical temperature as a function of layer thickness (inset of figure 4). This similarity is effectively taken into account in figure 4, which shows a linear behavior of \( T_C \) in the measured range as a function of rms roughness normalized by the layer thickness. This ratio quantifies the percentage of the layer that is affected by roughness. Therefore, figure 4 shows the behavior of \( T_C \) as a function of the portion of layer affected by interdiffusion or height variations. The critical temperature decreases linearly as the layer thickness decreases and the amount of disorder in the layer increases. In contrast, the roughness shows a strong increase of 40% only within the first 60 Å and appears to saturate at 240 Å layer thickness (figure 4, inset). Therefore, the absolute roughness does not play a determining role, but the \( T_C \) is inversely proportional to the absolute ‘clean’ layer thickness. Therefore, the disorder with decreasing layer thickness is fully quantified by changes in crystal structure of Mo and Nb due to strain, the limitation of the coherence length and mean free path.

While the disorder has a definite effect on the Nb \( T_C \), Mo remains normal and only influences the SL superconductivity through the proximity effect. This seems contrary to earlier claims that explain a \( T_C \) enhancement by disorder with a smeared DOS in the Mo band structure. McMillan argues that the energy scale in a metal–insulator transition due to disorder depends directly on single particle DOS at the Fermi level [33]. Finkel’slein, however, proposed that \( T_C \) changes due to changes in Coulomb repulsion, not due to changes in DOS. Finkel’slein’s model has been used before to describe a number of superconducting systems when the thickness of the layers is reduced [13]. Our results indicate that Nb critical temperature agrees with Finkel’slein model, which can be attributed to the Coulomb repulsion increase associated with the increased disorder. On the other hand, since no change in the Mo \( T_C \) with increasing disorder is found, this may be attributed to a different origin.

The low \( T_C \) of Mo may indicate that the disorder introduced by reducing the thickness of the layers is too low and does not affect the single particle DOS of Mo and Nb. Therefore we suggest that the reduction of \( T_C \) we observe in the SLs is due to changes in Coulomb repulsion by weak disorder. Finkelstein’s model fits well our observations on the behavior of Nb in the SLs if proximity effect with metallic Mo is considered. This interpretation disagrees with the initial Anderson theorem [5], but it reconciles our results with the results shown in [13] and previous understanding of \( T_C \) degradation in 2D systems [34–38]. Crow et al show for a 25 nm Nb film that a decrease of the mean free path to 1 nm causes a \( T_C/T_{CO} \) of \(~77\%\), agreeing with Finkel’slein’s calculations that show an expected \( T_C \) ratio of \(~80\%). This suggests that extremely large disorder is needed to affect DOS of Nb and Mo. For the case of Mo, this interpretation is supported by other works i.e. an increase of \( T_C \) is only obtained by amorphization with incorporation of impurity elements, or by growth at He temperatures [8, 12, 22]. Comparison with literature indicates this may not be a general conclusion, since other materials seem to be more sensitive to DOS changes [36].

5. Conclusions

We present a strong correlation of disorder and superconducting properties of Nb and Mo in Nb/Mo SLs. Changes
in the lattice parameter and roughness do not affect crucially the $T_c$ of the SL. Therefore, the disorder controlling the SC properties of Nb is the layer thickness only. Our results can be quantitatively explained with a simple model taking into account variations in mean free path and coherence length. No extra effects need to be invoked related to smearing of the DOS at the Fermi surface of Nb and Mo. The decrease of the critical temperature of Nb is associated with the increase of Coulomb repulsion with increased disorder. Our results also indicate no change in the $T_c$ of Mo with increased disorder, suggesting that an extremely large disorder is necessary to increase Mo $T_c$. Two possible questions arise from these results: first whether it is possible for a thin layer of Mo to exhibit an enhanced $T_c$ since Coulomb repulsion setting in at weak disorder may counteract the effect of a change in the DOS at larger disorder. Second, whether only the amorphous phase of Mo is superconducting and the observed increase in $T_c$ is related only to its appearance.

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