Critical Sheet Resistance for the Suppression of Superconductivity in Thin Mo-C Films

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(Received 30 January 1990)

We report resistivity and superconducting transition temperature measurements on ultrathin MoC films (down to 4.0 Å). We find that the critical sheet resistance separating the superconducting and insulating states at $T=0$ lies in the range 2.8–3.5 kΩ, which is about one-half of that reported for Bi films.

PACS numbers: 74.40.+k, 74.65.+n, 74.75.+t

The asymptotic behavior of a superconductor as one of its dimensions is reduced has attracted considerable interest. This interest increased following the development of various theories of electron localization and enhanced electron-electron interaction effects in disordered two-dimensional systems,[1,2] according to which there is no metallic state in two-dimensional systems at $T=0$ and, for a superconducting film, the disorder-enhanced effective Coulomb repulsion (μ^*) and the depleted density of states near the Fermi energy combine to reduce the superconducting transition temperature T_c.

Most of the data on superconducting films can, as suggested by Dynes *et al.*,[3] be viewed as involving two limiting cases: granular films[4,5] and homogeneous, microscopically disordered films.[6-8] In the limit of a granular film, the onset temperature of the transition remains essentially the same as the bulk value; however, the transition width broadens. The superconductivity is destroyed by a loss of long-range phase coherence. Support for these ideas came from Orr, Jaeger, and Goldman's work on *a*-Sn and *a*-Ga,[5] where the transition from global superconductivity (zero resistance) to local superconductivity (onset of a superconducting transition which is interrupted by an insulating behavior, resulting in a resistance minimum) was observed as the sheet resistance of the films increased. Critical sheet resistances for global superconductivity for both materials were found to be close to $h/4e^2$, which was later confirmed for Sn (Ref. 9) and for Pb films[10] and is also supported by the theories[11] involving Josephson arrays.

For homogeneous films in the weakly localized regime ($k_F l > 1$), many authors[6,12] have reported qualitative agreement of their data with various electron localization theories. However, Haviland, Liu, and Goldman[8] have recently reported some interesting results on the evolution of the temperature dependence of the sheet resistance with thickness of *a*-Bi films, in which the film is either superconducting or insulating (without any reentrance phenomena) in the $T \rightarrow 0$ limit, depending on whether the sheet resistance lies below or above a critical threshold value, which is close to $h/4e^2$. It is interesting that this value is very close to that quoted for the onset of global phase coherence for a granular film, despite the quite different behavior of the temperature dependence of the sheet resistance. Although the threshold value of $R_\square$ for *a*-Pb films was 9.5 $k\Omega$, a universal value of the threshold resistance of $R_\square = h/4e^2$, regardless of the length scale of the disorder, was suggested by Pang.[13] Recently, Fisher, Grinstein, and Girvin[14] also proposed a universal sheet resistance at the superconductor-insulator transition but they were not able to calculate the precise value.

One possible problem area is the role of the *a*-Ge (or other amorphous) underlayer generally required in preparing ultrathin, homogeneous films at cryogenic temperatures, as suggested in Ref. 8. Here we report on the thickness dependence of the superconducting and transport behavior of a series of thin Mo-C films and four selected films (6, 12, 20, and 40 Å thick) with a 10-Å-thick Si underlayer. Mo-C films turn out to be good candidates for such studies because they are electrically continuous down to 4.0 Å on either sapphire or MgO substrates (without any underlayer), when deposited at 200 °C; furthermore, thick (bulk) films deposited under the same conditions have a conveniently high T_c (8.3 K) and a very short mean free path with $k_F l \sim 1$.

The data reported here involve films prepared on polished single-crystal sapphire substrates by the reactive sputtering of Mo in an atmosphere of 5.0×10^{-5}-Torr acetylene and 3.0×10^{-3}-Torr of argon. The details of the deposition system have been reported elsewhere.[15] The thickness of a deposited film was inferred from the deposition time; the sputtering rate, calibrated with a film profilometer using thicker films, was in good agreement with that deduced from low-angle diffraction from multilayer films of MoC/TiC. T_c was taken as the midpoint in the resistive transition and a current density of 10–50 A/cm^2 was used.

The thick Mo-C film (0.4 μm) shows the Mooij correlation[16] as found for the transition-metal alloys; $\rho = 169$ $\mu\Omega$ cm and $(1/\rho)d\rho/dT = 40\times10^{-6}$ K^{-1}. The transition temperature was $T_c = 8.32$ K and $dH_{c2}(T)/dT|_{T=T_c} = 3.09$ T/K. The temperature dependence of the sheet resistance $R(T)$ as a function of the Mo-C film thickness is shown in Fig. 1(a). The overall features are in agreement with those of *a*-Bi films,[8] except that the critical sheet resistance separating the insulating and superconducting states at $T=0$ lies between 2.8 and 3.5 kΩ,

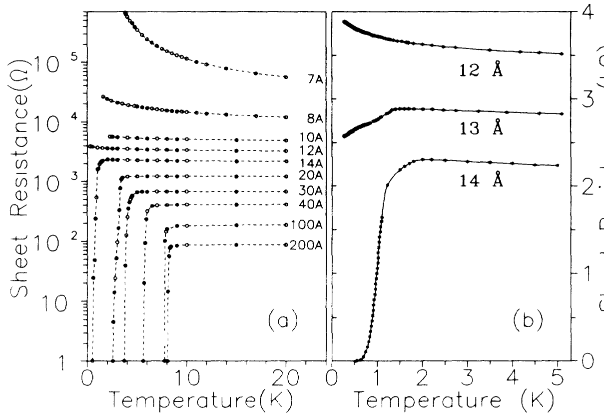

FIG. 1. (a) Temperature dependence of the sheet resistance of Mo-C films with a variety of thicknesses (7.0–200 Å). (b) Same as (a) down to 0.25 K for films 12, 13, and 14 Å thick.

which is considerably below $h/4e^2$ (6.5 kΩ). Clearly, quasireentrant phenomena (a resistance minimum) are not seen and the onset of the superconducting transition decreases with the thickness. Near the superconducting-insulating transition, three samples (with thicknesses of 12, 13, and 14 Å) were measured down to 250 mK, using an Oxford top-loading ^{3}He-^{4}He dilution-refrigerator system; these measurements are shown in Fig. 1(b). The superconductor-insulator boundary appears to lie between 12 and 13 Å, although the transition is broad and not completed by 250 mK. Ohmic response was examined and all samples displayed a linear I-V behavior for current densities in the range 10–30000 A/cm^2.

The effect of a 10-Å-thick Si underlayer on T_c turns out to be minor as seen in Fig. 2(a); it causes only a slight reduction of T_c. This is in contrast to its effect on the transport properties especially for ultrathin films. The Si underlayer acts as a 2-Å-thick effective layer in both the magnitude and the temperature dependence of the sheet resistance; a 12-Å-thick Mo-C film on a Si underlayer has a similar sheet resistance and temperature dependence to that of a 14-Å-thick Mo-C film without the underlayer but is not superconducting. Therefore, the threshold value for the superconducting-insulating transition in Mo-C films is lowered below 2.3 kΩ due to the underlayer.

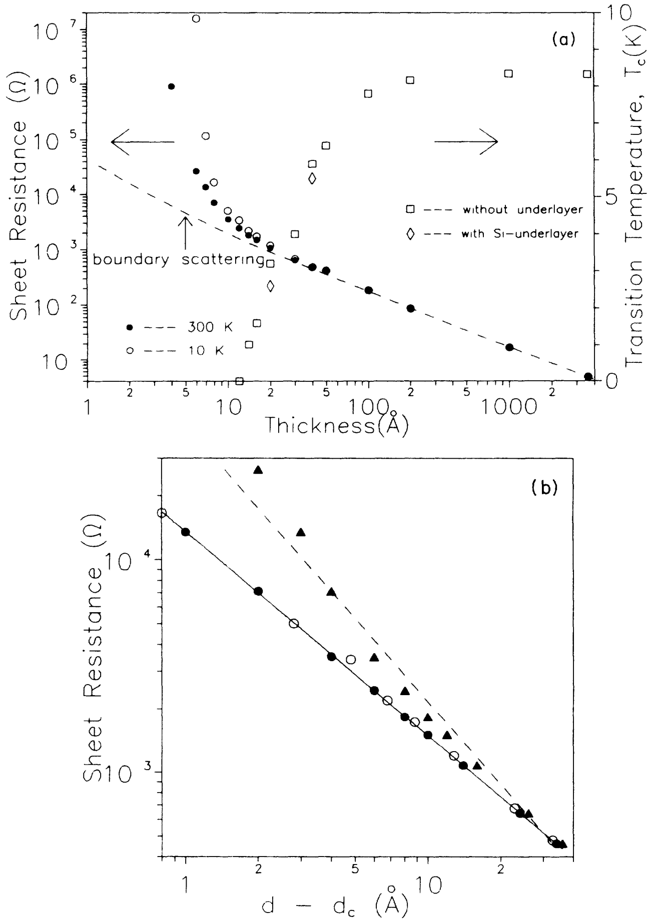

FIG. 2. (a) Thickness dependence of the sheet resistance and the superconducting transition temperature of Mo-C films: Solid (open) circles, the sheet resistance at 300 K (10 K); dashed line, the theoretical variation of $R_\square$ due to the classical-size effect assuming pure diffuse scattering at the boundary; squares (diamonds), T_c of the Mo-C films without (with) a 10-Å-thick Si underlayer. (b) $R_\square$ vs $d-d_c$: Triangles, sheet resistance at 300 K with $d_c=4.0$ Å; solid (open) circles, sheet resistance at 300 K (10 K) with $d_c=6.0$ Å (7.2 Å); dashed line, $R_\square(d)\propto(d-d_c)^{-1.3}$; solid line, $R_\square(d)\propto(d-d_c)^{-0.96}$.

Figure 2(a) shows that the resistivity of the film starts to deviate significantly from its bulk value at a thickness of about 20 Å. If we take the mean free path as 4 Å, the contribution from surface effects and/or 2D localization (and interaction) effects would appear to involve more than just the boundary scattering.[17] A fit of the data to the relation $R_\square(d)\propto(d-d_c)^{-t}$ is shown in Fig. 2(b). The sheet resistance at 300 K for one set (triangles) is plotted with $d_c=4.0$ Å, which is the minimum thickness for (room-temperature) electrical continuity. They are to be compared with $R_\square(d)\propto(d-4.0)^{-1.3}$, which is predicted by classical percolation theory if we assume that the thickness is proportional to the areal occupation probability in the percolation problem. On the other hand, two sets of sheet-resistance data at 300 and 10 K (solid and open circles) fall on the same line for the exponent $t=0.96$ with a d_c of 6.0 and 7.2 Å, respectively. (Note that, due to the cutoff, data for $d<d_c$ are not included in the fit.) The latter behavior could be explained by a percolation-localization crossover.[18,19] According to this idea, as the metallic fraction is reduced, Anderson localization sets in before the classical percolation threshold is reached (unless the microscopic resistivity is zero). Since the localization effects dominate the resistance before the transition, $R_\square(d)\propto(d-d_c)^{-\nu}$, with

$\nu \sim 1$.[20] Assuming that the phase diagram for the metal-insulator transition by Deutscher, Goldman, and Micklitz[18] can be extended to two dimensions at finite temperatures, the reduction of T_c is caused by the localization and the localization-percolation crossover occurs at a thickness of about ~ 100 Å, where T_c degradation starts. In any case, all three critical thicknesses are smaller than 13 Å, the minimum thickness required for the onset of superconductivity, which suggests that our superconducting films behave homogeneously.

With regard to electron localization or interaction theories, we could not find convincing evidence for the relation between the weak localization and the T_c degradation. Since the diffusion constant D is 3.6×10^{-5} m^2/sec [as determined from the slope of the $H_{c2}(T)$ curve at T_c], the Thouless length $[L_T=(4hD/kT)^{1/2}]$ is $330T^{-1/2}$Å. If we take $T=100$ K and $L_T=33$ Å, we should observe a logarithmic temperature dependence of the resistivity for films with thicknesses less than 33 Å when $T<100$ K. However, the temperature dependence of the sheet resistance is rather weak for the films thicker than 16 Å. Applying the localization theory, which predicts that $\Delta\sigma=(e^2/2\pi^2\hbar)\alpha p\ln(T/T_0)$, we obtain a value $\alpha p=0.9$–1.2 in the range 10–50 K for the thicknesses between 12 and 16 Å. In dirty metal films, it is found that $p=1$ due to a diffusive nature of the electron propagation.[21] But the T_c degradation starts well before this weak-localization regime (12–16 Å), although it appears that superconductivity disappears ($T_c\rightarrow0$) at the end of this regime (12 Å). When films become thinner than 12 Å, the transport behavior begins to change. The sheet resistance of the 10- and 8.0-Å films can be fitted by the form $R=A+BT^{-\gamma}$, where $\gamma=0.1$ and 0.3, respectively; the logarithmic behavior may be regarded as the $\gamma\rightarrow0$ limit and an increase in γ can be interpreted as the beginning of strong localization. The 7.0- and 6.0-Å films can be represented by the form $R=R_0\times\exp(T_0/T)^\beta$, where $\beta=0.6$ and 0.8, respectively. The sheet resistance at room temperature separating the weak (logarithmic power or law) and the strong (exponential) dependence is around 10 kΩ, which is consistent with the data in Au-Pd films[22] and electron inversion layers.[23]

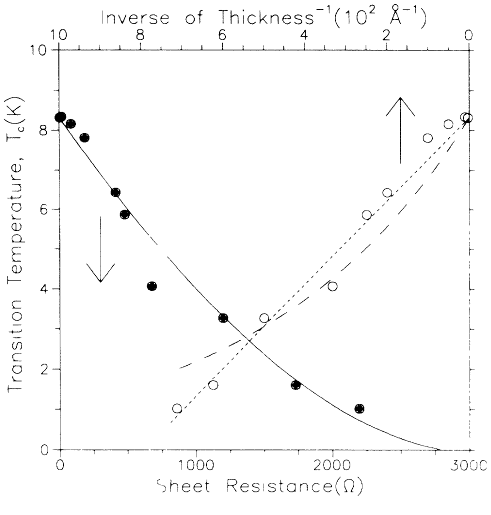

FIG. 3. Superconducting transition temperature as a function of the sheet resistance (solid circles) and of the inverse of the thickness (open circles). Solid line, Maekawa-Fukuyama theory with the parameters $g_1N(0)=-g'N(0)=0.6$, $\xi(0)=43$ Å (from H_{c2} measurement), and $l=4.3$ Å (interatomic distance); dotted line, linear least-squares fit; dashed line, exponential least-squares fit (Ref. 26).

As noted earlier in Fig. 2(a), severe suppression of T_c starts from approximately 100 Å, which is close to $2\xi(0)=86$ Å, where $\xi(0)$ is the extrapolated zero-temperature Ginzburg-Landau (GL) coherence length; $\xi(0)$ is determined from H_{c2} measurements on thick Mo-C films. Since $\xi(0)$ determines the range of the kernel in the self-consistent equation for $\Delta(\mathbf{r})$, naively we could regard T_c degradation as a surface-induced effect. Simonin[24] has examined the effect of including a surface term in the GL free energy (which is usually dropped) and found that it leads to a linear relation between T_c and the inverse of the thickness d^{-1}; the reported result is

$$T_c(d)=T_c(\infty)(1-d_c/d),$$

where $d_c=2a/NV(0)$, a is a lattice parameter, and $NV(0)$ is the BCS electron-phonon interaction strength. This linear relation fits our data quite well as seen from Fig. 3 and, if we take a as the lattice constant (~4 Å) and $NV(0)$ as 0.6,[25] the critical thickness d_c is 13 Å, which is in agreement with our result. Although admittedly phenomenological, this Ginzburg-Landau surface term provides a better description than the proximity effect[26] also seen in Fig. 3.

The dependence of T_c on the sheet resistance is also displayed in Fig. 3. Our data agree well with those on Mo-Ge films[6] (although we note that even though Ge and C have the same valence, they have radically different atomic radii). The initial linear portion of the $T_c(R_\square)$ curve for the Mo-Ge film system was interpreted by Graybeal[6] to be a two-dimensional localization effect. However, one difference with Mo-Ge films is that (as pointed out earlier) our Mo-C films show a $\ln T$ dependence only near the boundary of the superconductor-insulator transition (12–16 Å). A microscopic theory of the localization effects on T_c in 2D has been given by Maekawa and Fukuyama.[2] The decreasing slope of $T_c(R_\square)$ as $R_\square$ increases can be obtained in the limit that the strength of the repulsive Coulomb interaction g_1 is close to that of the attractive BCS interaction g' which is ignored in Refs. 2 and 6. This is plausible, however, since $g_1N(0)$ is of order unity and $|g'|N(0)$ is ~0.6.[25] Our data are in a reasonable agreement with the theory with $g_1N(0)=-g'N(0)=0.6$, implying that a T_c reduction is caused mainly by the depletion of the density of states rather than by an enhanced Coulomb interaction due to the diffusive motion of the electrons.

In conclusion, we have studied the T_c suppression and the transport behavior of thin Mo-C films as a function of thickness. The critical sheet resistance, separating the insulating and superconducting behavior at $T=0$, lies between 2.8 and 3.5 kΩ for this system, which is about one-half of the value reported for Bi and about one-third that of Pb.[8] It therefore appears that the sheet resistance separating the insulating and superconducting states at $T=0$ is a system-dependent quantity. Also Mo-C appears to be a rather unique candidate for studying the relation between the superconductor-insulator transition and the metal-insulator transition.

The authors acknowledge valuable experimental assistance by J. Q. Zheng and Z. Zhao. This work was supported by the Northwestern Materials Research Center (under Grant No. DMR-85-20280), the Office of Naval Research (under Grant No. N0014-88-K-0160), and the National Science Foundation (under Grant No. DMR-86-02857).

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