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SUPERCONDUCTING TRANSITION TEMPERATURES OF ULTRATHIN AMORPHOUS FILMS (*)

C. G. GRANQVIST and T. CLAESON
Physics Dept., Chalmers University of Technology
Fack, S-402 20 Gothenburg, Sweden

Résumé. — La température de transition de supraconductivité des films amorphes de Bi et Ga, évaporés sur support refroidi, est diminuée par un facteur deux quand l'épaisseur est réduite de 50 à 3 nm.

La dépendance d'épaisseur est en accord avec un modèle basé sur l'effet de proximité, dans lequel le film consiste en une couche mince non supra-conductive mise en contact avec la couche supra-conductive restante.

Abstract. — The superconducting transition temperature of vapour-quenched, amorphous films of Bi and Ga is suppressed by a factor of two as the thickness is decreased from 50 to 3 nm. The thickness dependence agrees with a proximity type model in which the film consists of a thin, non-supersconducting, surface layer in contact with a remaining, bulk superconducting part.

We will report upon measurements of the superconducting transition temperature, $T_c$, of amorphous Bi and Ga films, that have been extended to the ultrathin limit. The results are in accord with a model explaining the depression of $T_c$ as due to the proximity between the bulk part of the superconductor and a normal surface layer.

It has long been known [1-6] that the $T_c$'s of ultrathin films can be appreciably depressed compared to the thick-limit transition temperatures ($T_{co}$). A large number of theories and ideas have been given to explain this effect. They include modifications of the phonon spectrum [7], fluctuations of the order parameter [4, 8-10], quantization of electronic levels in minute metallic particles [11], and substrate effects [4, 5]. Several of these models are discussed by Strongin et al. [4].

It is experimentally documented [5] that relatively thick vapour quenched Bi and Ga films (being amorphous [12] and superconducting [13]) display a $T_c$ depression ($T_{co} - T_c$) approximately proportional to the inverse film thickness ($t^{-1}$). The dependence was described in terms of a modification of the boundary condition for the superconducting order parameter [5]. This approach assumes that the film thickness is much larger than the superconducting coherence length ($t \gg \xi$).

The opposite limit ($t \ll \xi$) is valid for the experiments with ultrathin films of Pb and Bi by Strongin et al. [4]. They interpret their results in terms of a model where the superconducting interaction is lowered within a surface sheath (of thickness $b$). When $t \ll \xi$, one can simply use an effective interaction strength being a weighed average of the values in the bulk and the surface layer as suggested by Cooper [14].

Our $T_c$ measurements on Bi and Ga were done in the thickness range $3 < t < 50$ nm. Since $\xi$, for amorphous Bi has been estimated to be 7.2 nm [15] (and a similar value ought to be valid for Ga), neither of the extreme limits discussed above can be applied directly.

Our conceptual model is essentially the same one as used by Naugle et al. [5] and Strongin et al. [4]. The main part of the thin film is assumed to behave as a bulk superconductor with transition temperature $T_{co}$. At the film surfaces the electron density of states drops [16, 17], and we assume that superconductivity is lost in a layer of thickness $b$ (of the order of a few times the Thomas-Fermi screening length). A proximity effect then lowers the $T_c$ of the superconducting part and induces superconductivity in the normal one. The de Gennes-Werthamer theory of the superconducting proximity effect [18-20] is valid when the electron mean free path, $l$, is much smaller than $\xi_x$. This is certainly true for our amorphous films and we extract from that theory:

$$\ln \left( \frac{T_{co}}{T_c} \right) = \frac{2}{\xi_x^2} q_x \tan q_x (t - b)$$

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$$N_q \xi_x^2 q_x \tan q_x (t - b) = N_x \xi_x^2 q_x \tanh q_x b$$

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The subscript \( s(n) \) refers to the « superconducting » (« normal ») part of the film, \( q^{-1} \) is the « extrapolation length » of de Gennes [18], \( N \) is the electron density of states at the Fermi energy, and

\[
\chi(z) = \Psi\left(\frac{1}{2} + \frac{1}{2}z\right) - \Psi\left(\frac{1}{2}\right).
\]

where \( \Psi(z) \) is the digamma function [21].

The coherence length is given by

\[
\xi = \left(\hbar v_F / (\pi k_B T_c)\right)^{1/2}
\]

(3) where \( v_F \) is the Fermi velocity, \( k_B \) is Boltzmann’s constant, and \( h \) is (Planck’s constant)/2 \( \pi \).

The low temperature conductivity, \( \sigma \), and the coefficient of the electronic specific heat, \( \gamma \), are related by

\[
\frac{(\pi k_B/e)^2 \sigma/\gamma}{v_F l} = \frac{N}{\xi z}
\]

(4) Assuming that \( \sigma_n \approx \sigma_s \) and observing that \( \gamma \propto N \) we get:

\[
N_s \xi_s^2 = N_n \xi_n^2.
\]

(5)

Eq. (2) can now be written

\[
q_s \tan q_s (t - b) = c
\]

(6) where the constant \( c \) only depends upon the properties of the normal layer and not upon the film thickness.

Eq. (1) and (6) finally give us \( T_c \) as a function of \( t \) with only two parameters, \( b \) and \( c \). In the thin film limit, the result is the same as for the Cooper model [14].

The predicted values (with reasonable parameters) are compared with experimentally determined \( T_c(s) \) of thin, amorphous Bi and Ga films in figure 1. The agreement is good for films thicker than 5 nm, while the experimental \( T_c(s) \) drop below the theoretical curve for the thinnest films. This is, however, reasonable both on experimental and theoretical grounds.

Fig. 1. — \( (T_c - T_{co})/T_{co} \) vs. \( t^{-1} \) for amorphous Ga and Bi films. The curves are derived from a numerical solution of eq. (1) and (6) using the indicated values of \( T_{co}, b \) and \( c \). Our \( T_{co} \)'s agree very well with measurements on thick films by others [5, 13, 22]. We have used \( \xi_n = 7.2 \text{ nm} \) (applying to amorphous Bi [15]) also for Ga.

5 N Bi and Ga are evaporated from resistively heated Mo boats onto a liquid \(^3\)He cooled single quartz substrate (upon which a layer of Ge had previously been evaporated at room temperature) [23]. The deposition rate is about 0.1 nm/s. The thickness is measured by a quartz crystal oscillator micro-balance (resolution about 0.1 nm; absolute accuracy 10 to 15 %). The \( T_c(s) \) are measured resistively in situ with a current of 1 or 10 \( \mu \text{A} \). The midpoint of the normal-superconducting transition is taken to define \( T_c \).
References