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Thickness dependence of the superconducting critical temperature in heavily doped Si:B epilayers

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We report on the superconducting properties of a series of heavily doped Si:B epilayers grown by gas immersion laser doping with boron content (n_B) ranging from $\sim 3 \times 10^{20} \text{ cm}^{-3}$ to $\sim 6 \times 10^{21} \text{ cm}^{-3}$ and thickness (d) varying between $\sim 20 \text{ nm}$ and $\sim 210 \text{ nm}$. We show that superconductivity is only observed for n_B values exceeding a threshold value ($n_{c,S}$) which scales as $n_{c,S} \propto 1/d$. The critical temperature (T_c) then rapidly increases with n_B , largely exceeding the theoretical values which can be estimated by introducing the electron-phonon coupling constant (λ_{e-ph}) deduced from *ab initio* calculations into the McMillan equation. Surprisingly $T_c(n_B, d)$ is fully determined by the boron dose ($n_B \times d$) and can be well approximated by a simple $T_c(n_B, d) \approx T_{c,0}[1 - A/(n_B \cdot d)]$ law, with $T_{c,0} \sim 750 \text{ mK}$ and $A \sim 8(\pm 1) \times 10^{15} \text{ cm}^{-2}$.

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I. INTRODUCTION

The discovery of superconductivity in Si:B¹ quickly followed that of diamond,² confirming that covalent semiconductors can be used as good starting materials to obtain new superconductors.³ In both cases, superconductivity develops in the partially unfilled valence band for boron concentrations $n_B \sim 10^{21} \text{ cm}^{-3}$ and most probably originates from a standard electron-phonon coupling mechanism. The critical temperature reaches $T_c \sim 0.6 \text{ K}$ in Si:B⁴ but rises up to $\sim 10 \text{ K}$ in C:B⁵ due to a better coupling potential in this lighter element (for a review see Ref. 6). However, if the boron concentration corresponding to the onset of superconductivity ($n_{c,S}$) coincides with that of the metal-insulator transition (MIT) in C:B ($n_{c,S} \sim n_{c,MIT} \sim 5 \times 10^{20} \text{ cm}^{-37}$), $n_{c,S}$ exceeds by several orders of magnitude $n_{c,MIT}$ in Si:B⁴ ($n_{c,S} \gg n_{c,MIT} \sim 5 \times 10^{18} \text{ cm}^{-3}$) and the origin of this threshold value still has to be clarified.

Moreover, it has been shown that T_c increases very rapidly for $n_B > n_{c,S}$ ($= n_{c,MIT}$) in C:B, roughly varying as $(n_B/n_{c,S} - 1)^{0.5}$. This unexpected variation imposed strong constraints on a possible variation of the screened Coulomb repulsion parameter (μ^*) which could become vanishingly small close to the MIT.⁷ However, a similar sharp increase is observed in Si:B (see below and Ref. 4) shedding some doubt on this initial interpretation. Furthermore, if the seminal measurements in Si:B¹ showed a very reasonable agreement between the experimental T_c values and those expected from *ab initio* calculations of the electron-phonon coupling constant (λ_{e-ph}), we will show here that, for boron contents on the order of 10^{21} cm^{-3} , the experimental T_c s largely exceed the values which can be estimated from theoretical calculations of λ_{e-ph} , hence rising the question of the origin of the superconductivity in Si:B thin films.

Thin films were initially considered as promising candidates for high temperature superconductivity due to a possible enhancement of the electron-phonon coupling constant at the sample surface.⁸ The importance of dimensionality has been further emphasized by the discovery of superconductivity at

the interface of insulating oxides,⁹ and the possible existence of high temperature superconductivity in single unit cell layers has been reported recently for FeSe deposited on SrTiO₃.¹⁰ However, other studies rather suggested a rapid suppression of T_c with the sample thickness (d) in this later system (for $d < 300 \text{ nm}$ ¹¹) and, except a few worth noting exceptions such as Al¹² or In,¹³ superconductivity is generally destroyed in reduced dimensionality.¹⁴⁻¹⁶ The *critical* thickness below which T_c is suppressed can then vary from several hundreds of nm down to a few lattice constants, and understanding the interplay between quantum coherence, disorder, and electronic interactions in those confined geometries remains a major challenge. However, despite decades of intensive studies, the question of *how* superconductivity is suppressed in thin films is still an issue.

We have performed a detailed study of the influence of the thickness (d)—and boron concentration (n_B)—on the superconducting properties of heavily doped Si:B epilayers. We show here that, in Si:B, T_c decreases with d even though this system stays far from the superconductor-insulator transition. The influence of d remains visible up to the thickest samples, that is for d values well above the coherence length and/or the electronic mean free path. Even more surprisingly, we will show that T_c is fully determined by the boron *dose*, i.e., the $n_B \times d$ product and can be well approximated by a simple $T_c(n_B, d) \approx T_{c,0}[1 - A/(n_B \cdot d)]$ law, where $T_{c,0} \sim 750 \text{ mK}$ is *doping independent* and $A \sim 8(\pm 1) \times 10^{15} \text{ cm}^{-2}$. The measurements have been performed on boron doped silicon epilayers grown by gas immersion laser doping (see Sec. II A). The main results are presented in Sec. II B and compared to *ab initio* calculations in Sec. II C. Finally, Sec. III is devoted to the discussion of the results.

II. SAMPLE PREPARATION AND EXPERIMENTS

A. Sample preparation

In contrast to boron-doped superconductive diamond which can be grown using standard methods such as high

pressure high temperature techniques,² or microwave plasma chemical vapor deposition,¹⁷ Si:B has to be prepared by out-of-equilibrium techniques as $n_{c,S}$ exceeds the limit of solubility of boron into silicon ($\sim 10^{20} \text{ cm}^{-3}$). The growth technique used here is gas immersion laser doping (GILD) (for further details see Refs. 1 and 4). In this technique, a precursor gas BCl_3 is chemisorbed on a [001]-oriented silicon wafer which is subsequently melted using ultraviolet laser pulses. During each melting/solidification cycle, boron diffuses from the surface into molten silicon, mostly in substitutional sites. A large effort was put into creating homogeneous samples by reaching an ultrahigh vacuum ($P = 10^{-7} \text{ Pa}$) and improving the spatial homogeneity of the laser beam energy. The total amount of boron has hence been tuned by changing the number of laser shots (for a given melting time) leading to n_B values ranging from $3 \times 10^{20} \text{ cm}^{-3}$ (in the thickest layers) to $6 \times 10^{21} \text{ cm}^{-3}$ (in the thinnest layers), and the epilayer thickness has been modified by changing the melting duration from 18 ns ($d \sim 20 \text{ nm}$) to 122 ns ($d \sim 210 \text{ nm}$).

Well defined x-ray diffraction (XRD) peaks have been observed for the as-grown doped layers attesting for their good epitaxial quality. Indeed, as shown in Ref. 4, the crystalline structure of the layers is that of silicon, although distorted by the presence of smaller boron atom in substitution and the position of the diffraction peak increases linearly with the boron content (reaching a saturation limit around 36° for $d = 80 \text{ nm}$). The concentration of the epilayers has been verified by secondary ion emission mass spectroscopy (SIMS) confirming that the total amount of boron increases roughly linearly with the number of laser shots. Atom probe tomography confirmed the absence of boron aggregates in the epilayers.¹⁸

B. Influence of the layer thickness and boron content on T_c

Four (in line) contact AC resistivity measurements ($\omega \sim 17 \text{ Hz}$) have been performed down to 30 mK in a dilution fridge. The change of the AC voltage with temperature has been recorded using a lock-in technique at fixed current. All measurements have been carried out in the Ohmic regime and we checked for any heating and/or nonlinearity effects by varying the current from 10 to 1 nA. As an example, the evolution of the superconducting transition with the doping content n_B is displayed in Fig. 1 for $d = 30 \text{ nm}$. As shown, in this case, the transition remains sharp and complete in all measured samples but no transition could be observed down to our lowest temperature below $n_{c,S} \sim 2 \times 10^{21} \text{ cm}^{-3}$. This threshold value decreases with increasing thickness down to $n_{c,S} \sim 2 \times 10^{20} \text{ cm}^{-3}$ for $d = 210 \text{ nm}$, but it is worth noting that, in the thickest samples, the transitions become incomplete for $n_B < 1 \times 10^{21} \text{ cm}^{-3}$ [see open symbols in Fig. 1(a) and corresponding light gray area in Fig. 1(b)] suggesting the existence of granular superconductivity in those layers. Note also that for low doping contents, the normal state conductivity (σ_n) increases linearly with n_B [see Fig. 1(b)] with a mobility $\mu = \sigma_n/en_B \sim 100 \text{ cm}^2 \text{ V/s}$ in very reasonable agreement with the value expected for those large doping contents.²⁰ However, as shown, σ_n decreases for $n_B > 2 \times 10^{21} \text{ cm}^{-3}$ and/or for $d < 50 \text{ nm}$ tending towards a constant value $\sim 5000 (\Omega \text{ cm})^{-1}$.²¹

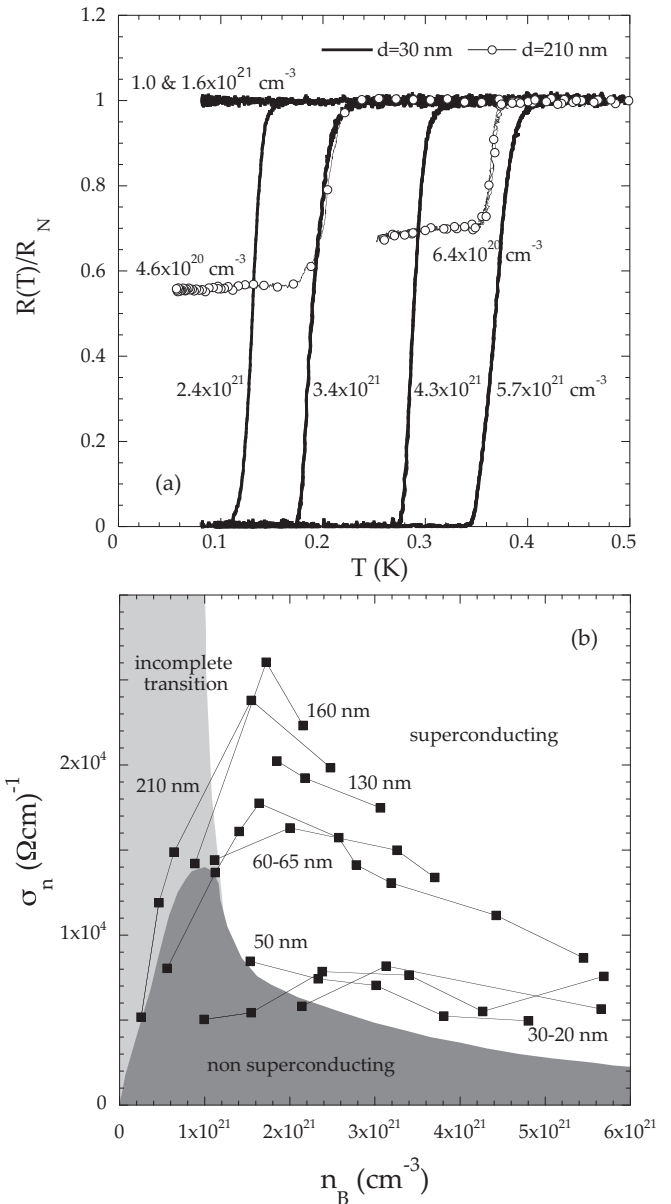


FIG. 1. (a) Temperature dependence of the normalized resistance (R_N being the resistivity at $T = 1 \text{ K}$) for the indicated doping contents in the $d = 30 \text{ nm}$ epilayer (thick lines) and the $d = 210 \text{ nm}$ (open circles). (b) normal state conductivity as a function of boron content for the indicated thickness values. The samples in the dark gray area do not present any superconducting transition down to 30 mK and the samples in the light gray area present partial transitions [see Fig. 1(a) for $d = 210 \text{ nm}$ as an example; see also Fig. 2].

The critical temperature T_c has been determined as the temperature at which the resistivity reaches 90% of its normal state value and Fig. 2(a) displays the evolution of T_c as a function of n_B , for the indicated d values. As shown, T_c rises rapidly above some critical threshold value ($n_{c,S}$) which increases as d decreases. The evolution of $n_{c,S}$ with d is displayed in Fig. 2(b) clearly indicating that $n_{c,S}$ scales as $1/d$. Even more surprisingly the whole $T_c(n_B, d)$ curves can be rescaled on one single curve when plotting T_c as a function

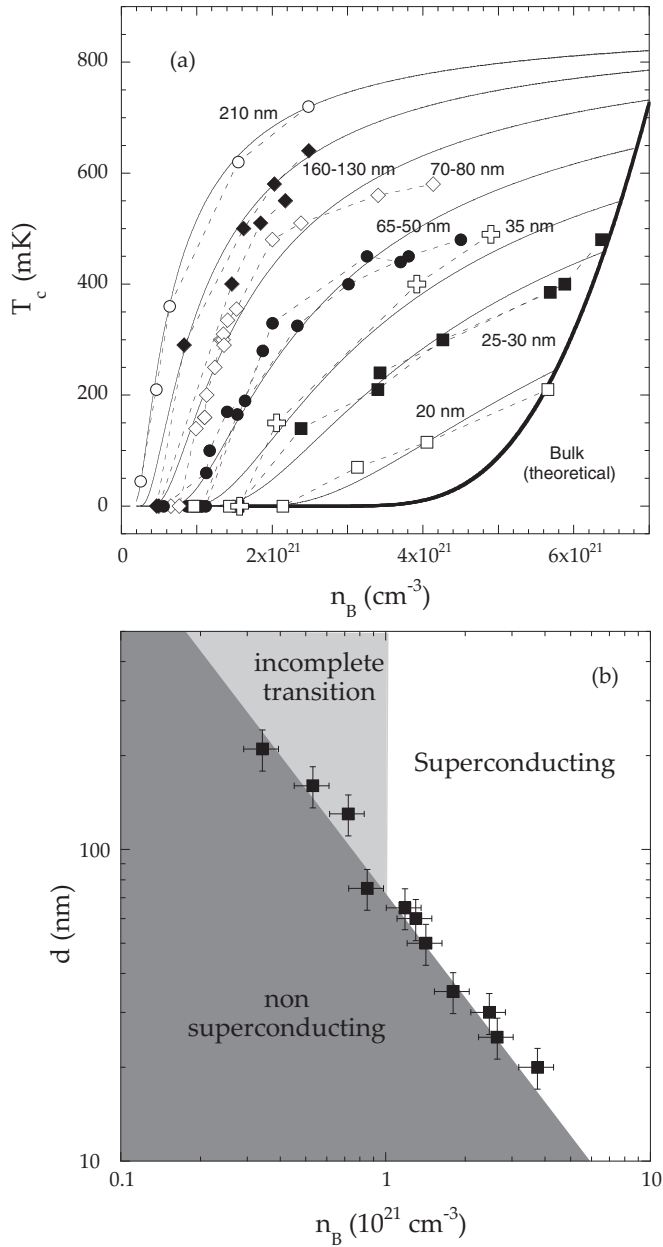


FIG. 2. (a) Doping dependence of the critical temperature of Si:B epilayers at the indicated values of the layer thickness. The thick solid line is the critical temperature expected from *ab initio* calculations of the electron-phonon coupling constant λ_{e-ph} (see Fig. 3). The thin lines correspond to the values expected from Eqs. (2)–(3) (see text for details). (b) Thickness dependence of the threshold value (black squares) for the onset of superconductivity ($n_{c,s}$). The samples in the dark gray area do not present any superconducting transition down to 30 mK and the samples in the light gray area present partial transitions (see also Fig. 1).

of $n_B \times d$ and, as shown in Fig. 3:

$$T_c(n_B, d) = f(n_B \times d) \sim T_{c,0} [1 - A/(n_B \cdot d)] \quad (1)$$

with $T_{c,0} \sim 750$ mK and $A \sim 8(\pm 1) \times 10^{15} \text{ cm}^{-2}$, clearly showing that the critical temperature of the films is fully determined by the boron dose $n_B \times d$. It is worth noting that we have also grown superconducting epilayers using a pulsed

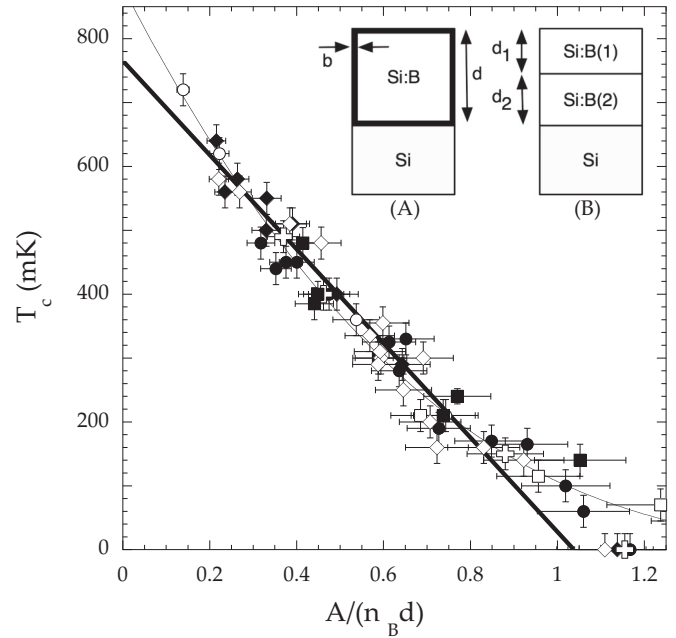


FIG. 3. Critical temperature as a function of $1/(n_B \cdot d)$ [n_B being the boron content and d the epilayer thickness, see Fig. 2(a) for symbols correspondences]. The thick line is a guide to the eyes suggesting a $T_c(n_B, d) = T_{c,0} [1 - A/(n_B \cdot d)]$ law with $T_{c,0} \sim 750$ mK and $A \sim 8(\pm 1) \times 10^{15} \text{ cm}^{-2}$ and the thin line would correspond to an electron-phonon coupling constant proportional to $1/n_B \cdot d$ (see text for details). Inset: sketches of possible geometries of the Si:B epilayers grown on Si substrates (see text for details).

laser induced epitaxial technique.¹⁹ In this case, silicon wafers were preimplanted with B ions and subsequently annealed in order to obtain the epitaxial recrystallization of the top layer. In this method, the implantation dose was fixed and n_B was varied by changing the melting depth (i.e., $n_B \times d$ was kept constant in the whole series). All measured samples¹⁹ then presented the same T_c value in agreement with the present data in which T_c is fully determined by the $n_B \times d$ product. Moreover, as expected we observed an increase of T_c by doubling the preimplantation dose (not shown in Ref. 19).

C. *Ab initio* calculations

On a theoretical point of view, the electron-phonon coupling constant λ_{e-ph} can be calculated in bulk Si:B crystals using a supercell model, namely a periodic distribution of dopants combined with a density functional theory (see Ref. 1 for details). At various doping rates, the cell volume is isotropically relaxed showing good agreement with the experimental Vegard's law (see Ref. 1). Figure 4 displays the λ_{e-ph} value previously reported for $n_B \sim 3.1 \times 10^{21} \text{ cm}^{-3}$ in Refs. 1 and 22 together with additional calculations for $n_B \sim 1.6 \times 10^{21}$, 2.2×10^{21} , and $6.2 \times 10^{21} \text{ cm}^{-3}$ using SiB_n units cells with $n = 31, 23$, and 7, respectively (solid circles). As shown those values lie slightly below those obtained in a virtual crystal approximation²³ (open circles), but both approaches suggest a rapid decrease of λ_{e-ph} for $n_B \rightarrow 0$ in striking contrast with the “experimental” values which can be deduced from T_c by

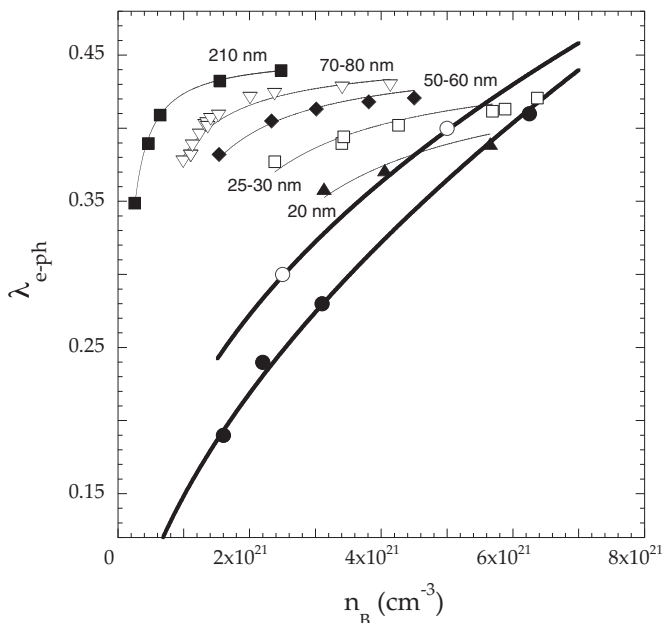


FIG. 4. Doping dependence of the electron-phonon coupling constant λ_{e-ph} deduced from *ab initio* calculations in the supercell (closed circles, present work) and virtual crystals (open circles, from Ref. 23) models. The other symbols correspond to the λ_{e-ph} values deduced from the experimental T_c values [see Fig. 2(a)] inverting the McMillan formula [Eq. (2)] with $\mu^* = 0.15$ (for the indicated epilayer thicknesses) and the thin lines are fit to the data using Eq. (2) (see text for details).

inverting the McMillan formula:

$$T_c = \Omega \times \exp[-(1 + \lambda_{e-ph})/(\lambda_{e-ph} - \mu^*(1 + \lambda_{e-ph}))], \quad (2)$$

where Ω is the characteristic phonon energy scale (~ 450 K²³). The values displayed in Fig. 4 correspond to $\mu^* = 0.15$ (standard value for metals) but it is important to note that a clear disagreement between theoretical and “experimental” values remains visible whatever the choice of μ^* . Correspondingly, introducing the λ_{e-ph} values obtained from our *ab initio* calculations into this bulk isotropic formula leads to exponentially small T_c s for low n_B values which should drop below our 30 mK experimental limit for $n_B < 4 \times 10^{21}$ cm⁻³ [thick line in Fig. 2(a)], in striking contrast with the measured values in the thickest samples.

III. DISCUSSION

As pointed out in the introduction, the influence of the sample thickness on the superconducting properties of thin films remains a puzzling issue. Even more surprisingly, in our Si:B films the thickness d actually enters through the $n_B \times d$ product as T_c is fully determined by the dose. Boundary effects, phase fluctuations, and/or a change of the electron-phonon coupling constant are the main scenarios which can be at the origin of a change in T_c . Those different scenarios are discussed below.

A. Boundary effects

It has first been suggested by Naugle *et al.*¹⁵ (see also Ref. 16) that the destruction of the superconducting state in metallic films may be described in terms of a modification of the boundary conditions for the Ginzburg-Landau order parameter (f). Indeed, introducing a surface energy term in the Ginzburg-Landau free energy leads to a decrease of the critical temperature as $T_c \sim T_{c,0}[1 - 2\xi_0^2/(bd)]$ ¹⁶ where $T_{c,0}$ is the critical temperature of the bulk sample and b a characteristic distance related to the boundary condition: $\vec{\nabla} f \cdot \vec{n}|_S = f/b|_S$ [see sketch (A) in Fig. 3]. As shown in Fig. 3 such a $1/d$ dependence is in very reasonable agreement with the experimental data. However, it is hard to understand why $T_{c,0}$ should be doping independent (what would this “bulk” phase of constant $T_c = 750$ mK be). Moreover, the critical thickness corresponding to the complete destruction of superconductivity $d_c = 2\xi_0^2/b$ is usually on the order of a few lattice constants^{15,16} so that superconductivity is expected to survive down to the nanometer scale (see also Ref. 24) whereas, in our case d_c can exceed ~ 200 nm and is scaling as $1/n_B$. We hence believe that boundary effects can not account for the observed dose dependence of T_c .

B. Phase fluctuations

It has been suggested that the loss of superconductivity in 2D systems could occur below the mean field transition temperature ($T_{c,0}$) due to the unbinding of thermally generated vortex-anti-vortex pairs.²⁵ The 2D Berezinskii-Kosterlitz-Thouless critical temperature is then given by $k_B T_c = \Phi_0^2 d / 8\pi \mu_0 \lambda^2(T_c)$ where $\lambda(T)$ is the temperature dependent penetration depth. Taking $1/\lambda^2(T) \sim 1/\lambda^2(0)(1 - T/T_{c,0})$ with $\lambda^2(0) = m^*/\mu_0 n_B q^2$, one expects: $T_c \sim T_{c,0} \times [\alpha n_B d / (T_{c,0} + \alpha n_B d)]$ with $\alpha = \hbar^2 / 8\pi k_B m^*$ and the dose ($n_B \times d$) dependence of the critical temperature would find here a straightforward explanation. However, in our case ($\hbar^2 / 8\pi k_B m^*) \cdot (n_B d) \sim 10^6$ K and the reduction of T_c due to those phase fluctuations should be fully negligible in our case.

The influence of the sample thickness and/or carrier density on the superconducting properties has been studied into details in disordered systems¹⁴ (for a review see Ref. 26). Two classes of materials have emerged in this case: In systems such as MoGe, a.Bi, or TaN the reduction of T_c is most probably due to suppression of the Cooper pairing (see discussion below) but in a second category (a:InO, TiN) the loss of superconductivity is driven by the localization of Cooper pairs due to *disorder induced* phase fluctuations.²⁷ A strong resistance peak prior to the superconducting transition has also been reported recently in boron doped diamond films²⁸ and this peak has been interpreted as evidence for phase fluctuations between the different grains in those granular films hindering the development of long range superconductivity (so called bosonic insulator).

However, the suppression of superconductivity in disordered systems is usually observed for a Ioffe-Regel parameter $k_F l$ on the order of 1 (k_F being the Fermi wave vector and l the electronic mean free path) whereas, in our films, the conductivity values suggest that $l \sim 3$ nm,⁴ and $k_F \sim 3 \times 10^9$ m⁻¹ (for $n_B \sim 10^{21}$ cm⁻³, in a free electron approximation) so that $k_F l \sim 10$. Moreover, in contrast to

disordered films, the conductivity of our films *decreases* (or remain roughly constant for the thinnest samples) as the critical temperature *increases* [see Figs. 1(b) and 2(a)] and no sign of an insulating behavior was observed down to the smallest thickness (the normal state resistance remaining almost temperature independent up to several tens of K even in the nonsuperconducting samples). The proximity of a thickness induced superconductor-insulator transition can hence be excluded in our case.

C. Reduction of the electron-phonon coupling constant

A thickness dependent λ_{e-ph} value can be the hallmark that the film is constituted of two sublayers of thickness d_1 and d_2 ^{12,29} [see sketch (B) in Fig. 3]. Indeed, if $\xi_1 > d_1$, $\xi_2 > d_2$, and $l < \xi$ (ξ_i being the coherence length in the sublayer i) the “effective” coupling constant resulting from the proximity effects between the two sublayers is given by:³⁰ $\lambda_{\text{eff}} = (N_1^2 V_1 d_1 + N_2^2 V_2 d_2) / (N_1 d_1 + N_2 d_2)$, where N_i and V_i are the density of states and coupling potentials in sublayer i . Assuming that $N_1 \sim N_2$ one then expects

$$\lambda_{\text{eff}} = \lambda_1 [d - (1 - V_2/V_1)d_2] / d = \lambda_1 [1 - \tilde{d}/d]. \quad (3)$$

If $V_2 > V_1$, \tilde{d} is negative and λ_{eff} is expected to increase for decreasing d and this scenario could account for the increase of T_c in very thin Al films¹² (assuming an enhanced electron-phonon coupling on a surface layer of thickness $d_2 \sim a$). On the contrary, if $V_2 < V_1$, T_c is expected to decrease with d , as observed in our Si:B films. As T_c is determined by the dose, \tilde{d} is expected to scale with $\propto 1/n_B$ and very reasonable fits to the “experimental” λ_{e-ph} values (thin lines in Fig. 4) can be obtained with $\lambda_1 \sim 0.45$ and $\tilde{d} \propto 1/n_B$ (see also thin line in Fig. 3). Introducing the misfit parameter $f = \Delta a/a$ (the lattice parameter a being smaller in the boron doped layer than in the pristine Si substrate) and using the fact that $f \sim -k n_B$ with $k \sim 10^{-23} \text{ cm}^3$,⁴ one obtains that $\tilde{d} \sim a/f$.

It is worth noting that dislocations are actually expected to appear in strained epilayers for thicknesses larger than $\sim a/f$ and \tilde{d} is hence on the order of the thickness of the pseudomorphic sublayer. The proliferation of dislocations for $d > \tilde{d}$ can have drastic consequences on λ_{e-ph} . Indeed, it has then been shown by Gurevich and Pashitskii³² that the strain associated to the presence of dislocations can lead to large fluctuations of T_c . It has also been suggested that this strain could be responsible for the increase of T_c observed in thin In films¹³ and that misfit dislocations

could even lead to the formation of a metallic inversion layer in semiconducting monochalcogenide heterostructures (PbTe/PbS, PbTe/PbSe, and PbTe/YbS). This inversion layer then turns out to be superconducting at an unexpectedly high critical temperature (up to ~ 6 K for the largest misfit values).³¹ Finally, note that it has been shown²² that negative strain leads to a very strong increase of λ in doped silicon. However, the fits to the data suggested a *doping independent* λ_1 value (~ 0.45 for $\mu^* \sim 0.15$) which is, as already pointed above in the case of $T_{c,0}$, extremely puzzling. This scenario obviously requires further investigations.

Finally note that weak localization corrections to the electron-phonon coupling constant can also lead to a decrease of T_c . Indeed even though the mean free path $l \ll d$, the system can be essentially 2D as far as the diffusion process is concerned when the diffusion length $L_T = (\hbar D/kT)^{0.5}$ reaches d ³³ (see also Ref. 34). As $D = v_F l/3 \sim 0.1 \text{ cm}^2/\text{s}$, one has $L_T \sim 100 \text{ nm} \sim d$ in our films (at $T = 1 \text{ K} \sim T_c$). Localization effects are then expected to give rise to an $\exp(-K_{2D})$ correction to the bulk $T_{c,0}$ value³³ with $K_{2D} \sim [\hbar/E_F \tau] \times [1/k_F d]$ (with $\tau \sim l/v_F$). Rewriting $K_{2D} \sim [1/\sigma_n d] \times [e^2/\pi h] \sim R_{\square}/[100 \text{ k}\Omega]$, one then expects an exponential decrease of T_c with R_{\square} (and $1/d$). However, as the scattering time is obviously dependent on n_B (and d , see Fig. 1) we did not observe any scaling of T_c on $R_{\square} \propto \tau/[n_B \times d]$ and $R_{\square} \sim 1 \Omega \ll 100 \text{ k}\Omega$ so that those localization corrections are expected to play only a minor role in our case.

IV. CONCLUSION

In summary, we have shown that the threshold concentration for the onset of superconductivity in heavily boron doped silicon epilayers $n_{c,s}$ increases as the thickness (d) of the film decreases. Surprisingly, T_c is fully determined by the dose $n_B \times d$ scaling as $T_c(n_B, d) \approx T_{c,0}[1 - A/(n_B \cdot d)]$. This decrease can not be attributed to either phase fluctuations, boundary, or localization effects, but strain effects associated to misfit dislocations could play a role.

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