Magnetic-field-independent superconductivity of ultrathin Pb films on cleaved GaAs surface

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Abstract. We performed magnetotransport measurements on ultrathin Pb films deposited onto cleaved GaAs surface and observed two-dimensional superconductivity for an amorphous 2.2-Å-thick film, which is below one monolayer. The superconducting transition is almost independent of parallel magnetic field as high as 14 T. This means that the superconducting state has much larger critical magnetic field than Pauli paramagnetic limit. We consider two different mechanism relating Rashba spin splitting.

1. Introduction
Two-dimensional (2D) superconductivity in ultrathin films is attractive. The onset of two-dimensional superconductivity have intensely studied by tuning film thickness or applying magnetic fields etc. in granular or amorphous thin films [1–3], and recently in crystalline layers [4–10]. Markedly in even a single-atomic layer of Pb and In grown epitaxially on silicon substrate, and superconducting energy gaps were observed by scanning tunneling microscopy (STM) measurements [11]. Furthermore macroscopic zero resistance state in one-atomic layer was confirmed by electric transport measurements [12].

In this article, we examined the magnetic-field effect of two-dimensional superconductivity in amorphous ultrathin Pb films on cleaved GaAs surface. We used GaAs cleaved surface as a substrate and observed 2D superconductivity even for one monolayer (ML) notwithstanding amorphous films. While the superconducting state was fragile to the perpendicular magnetic field, it was robust to the parallel magnetic field as high as 14 T. The critical magnetic filed in parallel direction much larger than 14 T cannot be explained by Pauli paramagnetic limit.

2. Experimental Setups
The experimental setups were based on the technique of the previous works on InAs inversion layers [13, 14]. The substrate was a non-doped semi-insulating GaAs single crystal. It was cleaved at liquid helium temperatures in an ultrahigh vacuum chamber with 3He cryostat. Amorphous Pb films were made by quench-condensation. Magnetotransport data on the (110) cleaved surface were taken in a Hall bar geometry (4 mm × 0.35 mm) using the standard four probe lock-in technique at 13.1 Hz with two current electrodes and four voltage electrodes on non-cleaved surfaces prepared by deposition of gold at room temperature. The resistance of GaAs substrate was higher than 100 MΩ at 4.2 K. The sample was mounted on a rotatory stage to control the magnetic field direction precisely.
3. Experimental Results
Figure 1 shows the sheet resistance in the normal state and the critical temperature \((T_c)\) as a function of film thickness. For calibrating film thickness we used quartz oscillator assuming the bulk density of Pb. The critical temperature was defined as the value of the half of the sheet resistance in the normal state. We observed superconductivity from a thickness of 2.2 Å, which is below 1 ML. A thickness of 2.2 Å corresponds to a Pb atom coverage of \(7.2 \times 10^{14}\) cm\(^{-2}\), and it is smaller than \(8.9 \times 10^{14}\) cm\(^{-2}\) of the atomic density of GaAs (110) surface. In the 2.2-Å-thick film, \(T_c\) was 0.90 K and the sheet resistance in the normal state was 4.35 kΩ, which is below the quantum resistance for pairs, \(h/4e^2 = 6.45\) kΩ, the threshold value of Superconductor-Insulator transition [1].

![Figure 1](image)

**Figure 1.** Plot of the sheet resistance in normal state \((R_n)\) and the superconducting critical temperature \((T_c)\) versus film thickness for several runs. 1 ML corresponds to 2.7 Å.
Figure 2 shows the temperature dependence of the sheet resistance for different parallel magnetic field. The results are almost independent of parallel magnetic field. This is surprising because the upper critical filed in parallel direction to the 2D systems is normally limited by Pauli paramagnetic effect. Since at Pauli paramagnetic limit the Zeeman energy arising from spin splitting of Cooper pairs exceeds the superconducting condensation energy, Pauli paramagnetic limit is expressed as $H_P = 1.84T_c$ for an isotropic BCS superconductor [15]. In the 2.9-Å-thick film $T_c$ is 2.6 K and $H_P$ is 4.8 T. In the 3.4-Å-thick film $T_c$ is 3.3 K and $H_P$ is 6.1 T. Though Pauli paramagnetic limit calculated from the expression is much lower than 14 T, the 2D superconductivity is robust to parallel magnetic fields as high as 14 T.

![Figure 2.](image_url)  

**Figure 2.** The temperature dependence of the sheet resistance for different parallel magnetic field. The film thickness is 2.9 Å and 3.4 Å. $H_P$ is 4.8 T and 6.1 T.

### 4. Discussion

We consider two different mechanism to explain upper critical field far above Pauli paramagnetic limit.

The first mechanism is based on spin-orbit scattering. According to the theory including the effect of spin-orbit scattering [16,17], the upper critical field is given by solving the equation,

$$\ln \left( \frac{T}{T_c} \right) + \psi \left( \frac{1}{2} + \frac{3(\mu_B H_c)^2}{4\pi k_B T \hbar \tau_{SO}} \right) - \psi \left( \frac{1}{2} \right) = 0$$

where $\tau_{SO}$ is spin-orbit scattering time, $\mu_B$ is Bohr magnetron, $\gamma = 1.78$, and $\psi(z)$ is the digamma function. In the limit of $T \to 0$, the upper critical field is given by

$$H_{c2}(0) = \left( \frac{\pi k_B T_c \hbar \tau_{SO}^{-1}}{3\gamma \mu_B^2} \right)^{1/2} = 0.602 \left( \frac{\hbar \tau_{SO}^{-1}}{k_B T_c} \right)^{1/2} H_P.$$  

$$\text{(2)}$$
With strong spin-orbit scattering the upper critical magnetic field can exceed Pauli paramagnetic limit. The strong symmetry-breaking with respect to the direction perpendicular to the surface is expected to cause a large Rashba spin splitting [18]. On the spin-split Fermi surface, momentum scattering involves the spin relaxation via D’yakonov-Perel mechanism if the Rashba splitting is strong enough. It might lead to strong spin-orbit scattering and large critical magnetic field.

The second mechanism is based on Cooper pairing on spin-split Fermi surfaces caused by Rashba effect. The band splits into two spin-dependent subbands as shown in Figure 3. The pair wave function is a mixture of both singlet and triplet states as

\[
| \vec{k}, \uparrow \rangle - | \vec{k}, \downarrow \rangle = \frac{1}{2} (| + \vec{k}, \uparrow \rangle - | - \vec{k}, \downarrow \rangle) + \frac{1}{2} (| + \vec{k}, \uparrow \rangle + | - \vec{k}, \downarrow \rangle)
\]

at 0T [19]. In a magnetic field along the x axis, the centers of the normal state Fermi surfaces of two subbands are shifted in opposite y directions while the Fermi surfaces remain circular. If the superconducting state is formed on the basis of the new Fermi surfaces, the spin susceptibility is independent of the superconducting transition and the critical magnetic field can be very large.

**Figure 3.** Schematic of 2D free electron Fermi surfaces with the Rashba term. Fermi surfaces at \( B = 0 \) (dotted lines) shifts in opposite y directions (solid lines) by applying magnetic field along the x axis.

In summary, we have observed 2D superconductivity below 1 ML of Pb films made by quench condensation onto GaAs cleaved surfaces. The 2D superconducting state is almost independent of parallel magnetic field as high as 14 T. We consider two different mechanism to explain it. Rashba effect might have something to do with 2D superconducting state.

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