Interlayer Transport in the Highly Anisotropic Misfit-Layer Superconductor $(\mathrm{LaSe})_{1.14}(\mathrm{NbSe}_2)$

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The *interlayer* transport in a two-dimensional superconductor can reveal a peak in the temperature as well as the magnetic field dependence of the resistivity near the superconducting transition. The experiment was performed on the highly anisotropic misfit-layer superconductor $(\mathrm{LaSe})_{1.14}(\mathrm{NbSe}_2)$ with T_c of 1.2 K. The effect is interpreted within the tunneling mechanism of the charge transport across the Josephson-coupled layers via two parallel channels—the quasiparticles and the Cooper pairs. Similar behavior can be found in the high-T_c cuprates but there it is inevitably interfering with the anomalous normal state. The upper critical magnetic field can be obtained from the interlayer tunneling conductance.

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Anomalous transport properties of the high-T_c cuprates remain unexplained. In contrast to the metallic temperature dependence of the *intralayer* resistivity ρ_{ab}, the *interlayer* resistivity ρ_c can reveal a semiconducting behavior in some cases [1]. The anisotropy ratio can reach a value $\rho_c/\rho_{ab} = 10^5$ in the most two-dimensional system of $\mathrm{Bi_2Sr_2CaCu_2O_8}$. The transport in magnetic field is a puzzle as well. The superconducting transition of ρ_{ab} broadens considerably in a magnetic field due to the complicated behavior of the superconducting vortex matter [2] where even melting of a vortex lattice can be observed [3]. The interlayer resistivity ρ_c as a function of magnetic field B displays very nonconventional behavior: starting from the superconducting state at $T < T_c$ with increasing magnetic field after an onset of the resistivity, a peak appears followed by a smooth decrease to a constant ρ_c at higher magnetic fields [4]. When the magnetotransport measurement is performed above T_c negative magnetoresistance can be observed [5].

Several models have been proposed to explain the interlayer magnetotransport. Most of them treat the peak in $\rho_c(B)$ and the following decrease of the resistivity at higher fields as a consequence of the anomalous normal-state properties, mainly due to an existence of the pseudogap in the quasiparticle spectrum and/or superconducting fluctuations [6]. But, Gray and Kim [7] proposed a model where the peak is due to an interplay of two different conductance channels present in the superconducting state of the sample. The model assumes a highly anisotropic superconductor as a stack of weakly coupled internal Josephson junctions, and the interlayer transport is accomplished by tunneling of quasiparticles and Cooper pairs. Below the upper critical magnetic field due to the opening of the superconducting gap in the quasiparticle spectrum, ρ_c increases, but at a sufficiently small field the Cooper pair tunneling channel is opened and ρ_c decreases to zero. Recently, Morozov *et al.* [8] obtained evidence supporting this model in their magnetotransport data on $\mathrm{Bi_2Sr_2CaCu_2O_8}$. However, in cuprates this purely superconducting effect is inevitably complicated by the anomalous normal-state properties.

In the present Letter we address the problem of the *interlayer* transport in the misfit-layer superconductor $(\mathrm{LaSe})_{1.14}(\mathrm{NbSe}_2)$, a quasi-two-dimensional system without any nonconventional behavior in the normal state. We show that the interlayer transport in the superconducting state of this layered system involves the tunneling of quasiparticles and Cooper pairs.

$(\mathrm{LaSe})_{1.14}(\mathrm{NbSe}_2)$ is a low temperature superconductor with T_c around 1.2 K belonging to the family of the lamellar chalcogenides [9], where two slabs MX and $\mathrm{TX_2}$ are stacked in a certain sequence. Because of the different symmetry of the MX and $\mathrm{TX_2}$ layers a misfit results along one intralayer crystallographic axis even if along the perpendicular intralayer axis a perfect fit of both structures is achieved [9]. In the case of $(\mathrm{LaSe})_{1.14}(\mathrm{NbSe}_2)$ every intercalated LaSe layer with the thickness of about 0.6 nm is sandwiched by one 2H-$\mathrm{NbSe_2}$ layer with about the same thickness [10]. The sandwich unit is stabilized by the electron transfer from the LaSe to the $\mathrm{NbSe_2}$ slab resulting in the natural layered system of the insulating LaSe and (super)conducting $\mathrm{NbSe_2}$ sheets, where the conduction is accomplished by the Nb $4d_{z^2}$ orbitals [9,11].

The title compound was obtained by the direct reaction of the three constituents La/Nb/Se in the stoichiometric ratios [9]. Single crystals used are of typical dimensions $1 \times 0.8 \times 0.1\ \mathrm{mm}^3$ with $T_c = 1.23$ K. For the interlayer as well as intralayer resistivity measurements in the Montgomery configuration [12] four electrical contacts were prepared at the corners of the top side of the sample and another four contacts in symmetrical mirror positions at the corners of the bottom side. A standard lock-in technique at 17 Hz was used to measure the temperature and magnetic field dependence of the resistance. All measurements were

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performed with a magnetic field applied perpendicular to the planes—along the c axis of the samples. The field was generated by a superconducting solenoid placed in the Košice top-loading refrigerator working between 100 mK and 2 K.

The temperature dependence of both ρ_c and ρ_{ab} revealed a metallic behavior between 1.5 and 300 K with a saturation below 30 K. The residual resistivity ratio was about 4 and the anisotropy ratio ρ_c/ρ_{ab} calculated from the Montgomery configuration about 50 at 4 K.

In Fig. 1 the transition to the superconducting state is shown at zero magnetic field for the intralayer resistivity ρ_{ab} as well as for the c-axis resistivity ρ_c. The intralayer resistivity ρ_{ab} shows a conventional transition with a midpoint at $T_c = 1.23$ K and a width $\Delta T_c = 0.1$ K. This narrow single-phase transition represents the quality certificate of the sample. On the other hand, a very peculiar transition can be seen in the interlayer resistivity ρ_c: below 1.4 K the resistivity increases by about 3 times. Then, below 1.2 K, ρ_c drops down reaching the zero value at about 1.1 K. Even with the lowest current density (1 $\mathrm{mA/cm^2}$) along the c axis the zero resistivity of ρ_c is reached at slightly lower temperatures than for ρ_{ab}.

Figure 2 displays the full set of our intralayer as well as interlayer magnetotransport data measured at different temperatures from 100 mK up to 1.3 K. The intralayer magnetoresistivities (Fig. 2a) show conventional transitions to the superconducting state which are shifted to higher fields and broadened as the temperature is decreased. Above 1.4 K the normal state is already achieved in zero magnetic field and no magnetoresistance is observed any longer. The interlayer magnetotransport data are plotted in Fig. 2b. At all temperatures below 1.4 K the transition to the normal state (with no magnetoresistance) is preceded by the peak. Between 1.4 and 1.2 K the resistivity is nonzero at zero magnetic field, but the peak appears in the field dependence with an increasing amplitude. Below 1.2 K the magnetoresistivity starts from zero, the following peak is broadened, its amplitude decreases, and its position shifts to higher fields. The peak position in $\rho_c(B)$ is always found in the range of magnetic fields where the superconducting transition of the intralayer resistivity $\rho_{ab}(B)$ takes place at the respective temperature. Figure 2c displays the interlayer magnetotransport data recalculated in the conductance. One can see that in all curves below 1 K before reaching the normal state value a linear dependence on the applied magnetic field is achieved.

The effect of the measuring current density was also examined and the result can be seen in Fig. 3, where the interlayer resistivity ρ_c measured at 100 mK is shown for four different current densities. The high-field side of the peak in the resistivity is hardly affected unless a high current density (30 $\mathrm{mA/cm^2}$) is reached where heating affects

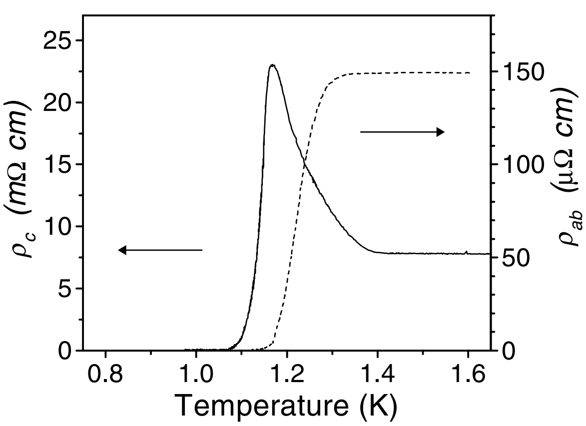

FIG. 1. Temperature dependence of the *intralayer* ρ_{ab} and *interlayer* resistivity ρ_c of $(LaSe)_{1.14}(NbSe_2)$.

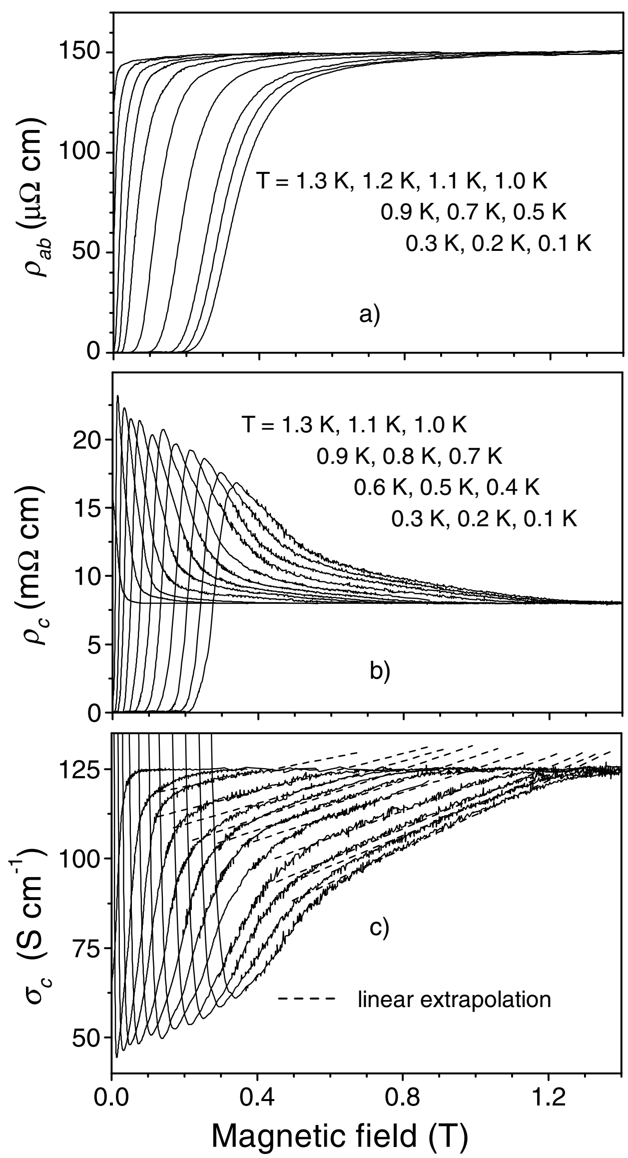

FIG. 2. (a) Intralayer and (b) interlayer magnetoresistive superconducting transitions at different temperatures. (c) Recalculated conductances from part (b).

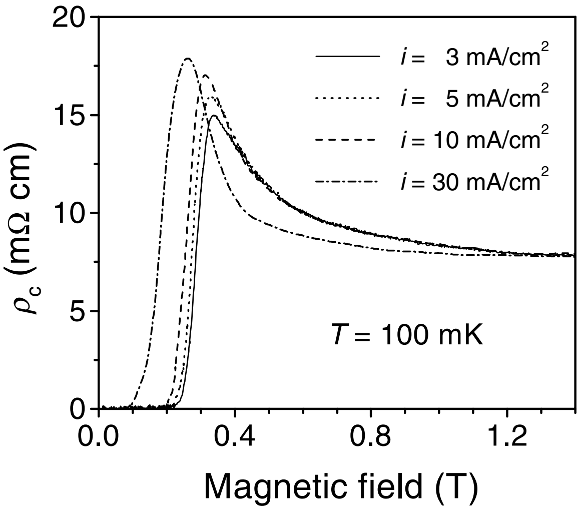

FIG. 3. Effect of the measuring current density on the interlayer magnetoresistive superconducting transition.

the superconductivity. But the low-field side of the peak reveals a strong sensitivity to the current density—with increasing current density the peak amplitude increases and its position is shifted to lower magnetic fields. Below 3 mA/cm^2 the current density does not influence the resulting magnetic field dependence of ρ_c. The same effect of the measuring current density has been observed in the interlayer magnetotransport at all temperatures below T_c as well as in the temperature dependence of ρ_c at zero magnetic field.

The asymmetric effect of the measuring current density on the peak in the interlayer resistivity indicates to different carrier-transport mechanisms below and above the peak. Moreover the metallic character of both intralayer and interlayer resistivities above the superconducting transition temperature indicates that the peak effect observed in the temperature as well as in magnetic field dependencies of the interlayer resistivity ρ_c is related only to the superconducting transition. Therefore, we will consider the two channel tunneling model with the quasiparticles and Cooper pairs passing across the layers.

By means of high magnetic field transport measurements for both perpendicular and parallel field orientation [13] we have shown that $(LaSe)_{1.14}(NbSe_2)$ behaves as a quasi-two-dimensional system below 1.1 K. This means that the superconducting coherence length ξ is very anisotropic with a value in the c-axis direction smaller than the total thickness of the insulating layer and the superconducting layer [14]. We note that the two-dimensional behavior of a layered system is more pronounced when the insulating layer thickness is bigger than the superconducting layer thickness [15]. As far as the conductance is due to the Nb orbitals [11] the latter thickness can be very small here, being restricted just to the atomic thickness of the Nb layer, and the two-dimensional regime of superconductivity would be enhanced.

Below the upper critical field $B_{c2}(T)$ a superconducting gap is opened in the superconducting sheets, which leads to an increase of the resistivity ρ_c at smaller fields as here the quasiparticle tunneling channel is carrying the current. At still smaller fields a minimal Josephson current can flow opening a second conductance channel which leads to a rapid decrease of the resistivity. The quasiparticle tunneling through internal Josephson junctions is obviously independent of the measuring current. But the Josephson tunneling of the Cooper pairs reveals a strong current dependence as is also observed in our experiment (Fig. 3). Similarly we can explain the temperature dependence of the interlayer resistivity ρ_c. Within this scenario also a slight shift in T_c's as measured by the intralayer and interlayer zero resistivities can be understood: T_c measured in the plane represents the thermodynamical value of the material while T_c measured across the planes is determined by the fact that a measurable Josephson current can flow across the whole sample.

Certain "critical" magnetic fields can be obtained from the magnetic field dependences of the resistivities $\rho_c(B)$ and $\rho_{ab}(B)$. For instance, the critical field obtained from the points when the intralayer resistivity achieves 95% of the normal state value is displayed in Fig. 4 by closed circles. One can see a saturation of the critical fields at the lowest temperatures what is typical for the upper critical magnetic field $B_{c2}(T)$ of the type-II superconductors [16], but at higher temperatures there is an anomalous positive curvature instead of the linear temperature dependence. The latter fact can be indicative of the presence of vortex melting. Taking account of the critical fluctuations near 100% of the transition to the normal state, we obtain a classical $B_{c2}(T)$ with a linear decrease up to T_c and the zero temperature extrapolated $B_{c2}(0)$ equal to 1.2 T [17].

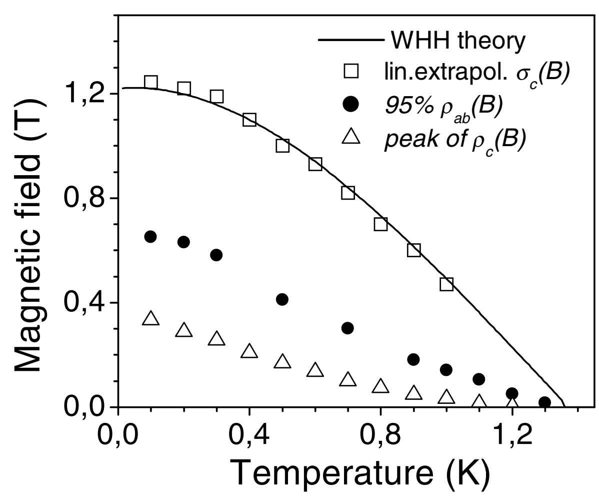

FIG. 4. Critical fields evaluated from different criteria of the superconducting transition in the intralayer and interlayer resistivities shown in Fig. 2. The full line shows the standard Werthamer-Helfland-Hohenberg temperature dependence of the upper critical field B_{c2}.

A very anomalous temperature dependence of the critical field is obtained when the peak in the interlayer resistivity versus field is taken as the upper critical field position (triangles in Fig. 4) as is sometimes done in the cuprates [18]. The only criterion giving the expected classical $B_{c2}(T)$ dependence is to use the magnetic field where the normal state is reached (squares in Fig. 4). These points can be practically obtained from the linear extrapolation of the interlayer conductance. It is another argument supporting the model where a peak in the interlayer resistivity is due to the interplay between the quasiparticle and Josephson tunneling across the layers. In our previous papers [19] we have shown that the quasiparticle tunneling in magnetic fields can give reliable information on the upper critical fields. The zero-bias tunneling conductance proportional to the averaged quasiparticle density of states (DOS) develops a linear magnetic field dependence near the upper critical field since the DOS is proportional to the number of vortex cores. In the case of the quasi-two-dimensional superconductor $(LaSe)_{1.14}(NbSe_2)$ the interlayer transport at higher fields where the Josephson component is suppressed is realized via quasiparticle tunneling and the respective interlayer conductance is then the zero-bias tunneling conductance of the stack of the junctions.

Finally, in the *interlayer* (magneto)transport measured on the quasi-two-dimensional low-T_c superconductor $(LaSe)_{1.14}(NbSe_2)$ we observed a peak effect in the superconducting transition as in the cuprates. This phenomenon is observed regularly in all of many samples we measured. The same effect has also been observed on another misfit-layer crystal of $(LaSe)_{1.14}(NbSe_2)_2$ with $T_c \approx 5.7$ K which will be presented elsewhere. We have found strong indications that the peak effect is related to the superconducting transition via the interplay of the quasiparticle and Cooper-pair tunneling mechanisms of the carrier transport across the layers. The observation of this effect in a layered superconductor with conventional normal-state transport is of importance for the interpretation of the anisotropic transport properties in the cuprate superconductors.

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