3D-2D crossover in the naturally layered superconductor (LaSe)$_{1.14}$(NbSe$_2$)

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The temperature and angular dependencies of the resistive upper critical magnetic field $B_{c2}$ reveal a dimensional crossover of the superconducting state in the highly anisotropic misfit-layer single crystal of (LaSe)$_{1.14}$(NbSe$_2$) with the critical temperature $T_c$ of 1.23 K. The temperature dependence of the upper critical field $B_{c2}||ab(T)$ for a field orientation along the conducting $(ab)$-planes displays a characteristic upturn at 1.1 K and below this temperature the angular dependence of $B_{c2}$ has a cusp around the parallel field orientation. Both these typical features are observed for the first time in a naturally crystalline layered system.

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The discovery of the cuprate superconductors with a high critical temperature has lead to renewed interest in anisotropic or even low-dimensional superconductivity. Already 30 years ago Lawrence and Doniach [1] described the behavior of layered superconductors in a magnetic field based on a model system of stacked two-dimensional superconducting layers coupled together by Josephson tunneling between adjacent planes. In contrast to the isotropic case where the magnetic flux penetration occurs in the form of vortices of circular symmetry, in an anisotropic superconductor the vortex cores will be flattened in the interlayer c-direction for magnetic fields parallel to the layered structure $(ab)$-planes with $\xi_c < \xi_{ab}$ for the vortex core radii $\xi_c$ and $\xi_{ab}$, respectively, perpendicular and parallel to the planes. If $\xi_c$ is bigger than the distance between the adjacent superconducting layers the system is anisotropic but still three-dimensional with the upper critical magnetic fields for fields perpendicular and parallel to the planes determined by a product of the corresponding coherence lengths: $B_{c2}||c = \Phi_0/(2\pi\xi_{ab}^2)$ and $B_{c2}||ab = \Phi_0/(2\pi\xi_{ab}\xi_c)$, respectively, where $\Phi_0$ is the flux quantum. In some cases including the high-$T_c$ cuprates the critical fields can be very high and in extremely anisotropic superconductors $\xi_c$ could shrink with decreasing temperature below the value of the interlayer distance $s$. Then, the vortices are confined between the superconducting layers for fields applied parallel to the layers leading to a dimensional cross over in the flux penetration. Here, we present such a 3D-2D dimensional cross over in the upper critical field data of the highly layered single crystal of (LaSe)$_{1.44}$(NbSe$_2$). As shown in our previous paper [2] (LaSe)$_{1.44}$(NbSe$_2$) has been the first found system based on conventional superconductor behaving like the intrinsic Josephson junctions.

According to the simplest Lawrence-Doniach model the upper critical $B_{c2}||ab$ parallel to the $ab$-planes is predicted to diverge at the dimensional transition at a temperature $T^*$, where $\xi_c(T^*) = s/\sqrt{2}$. The real finite value of the upper critical field $B_{c2}||ab$ is caused by the finite superconducting layer thickness, Pauli paramagnetism and spin-orbit coupling effects. Klemm, Luther and Beasley [3] have extended the Lawrence-Doniach model to include these effects. They show that the divergence is removed but the dimensional crossover to a two-dimensional superconductivity is still characterized by a strong upward curvature of $B_{c2}||ab(T)$. Experiments on the intercalated layered compounds based on $2H$-TaS$_2$ [4] revealed strong upward curvature of the temperature dependence of the parallel upper critical fields accompanied by the temperature dependent critical field anisotropy $B_{c2}||ab/B_{c2}||c$ reaching maximum values of about 60. Similar results were obtained on the intercalated MoS$_2$ [5]. In experiments performed on artificially grown superconducting superlattices [6] the 3D-2D crossover is also well expressed by a kink in the temperature dependence of $B_{c2}||ab(T)$. The crossover to the two-dimensional behavior can be highlighted by a second effect. Namely, there will be a qualitative change of the angular dependence of the upper critical field $B_{c2}(\Theta)$. For the three-dimensional situation, the angular dependence $B_{c2}(\Theta)$ of the upper critical field interpolates between $B_{c2}||ab$ and $B_{c2}||c$ in the simple ellipsoidal form $(B_{c2}||ab(\Theta)\sin\Theta)^2 + (B_{c2}||c(\Theta)\cos\Theta)^2 = 1$ with a rounded maximum around $\Theta = 0$ (magnetic field parallel to the layers). In the 2D regime the superconducting layers are decoupled and can be treated as isolated thin films. For this case Tinkham [7] proposed the equation $|\sin\Theta| + |\cos\Theta| = 1$. In the 3D case the slope of $B_{c2}(\Theta)$ is zero around $\Theta = 0$ while in the 2D case it has a finite value making a cusp. This effect was also observed in superconducting superlattices below the 3D-2D transition. The situation remains unclear in the natural layered single crystals. In the case of high-$T_c$ cuprates the angular dependence of the irreversibility field has shown up the cusp-like form for the
most anisotropic system, Bi$_2$Sr$_1$CaCu$_2$O$_{x}$ [10]. Here, we present in the critical field data of the naturally layered system (LaSe)$_{1.14}$(NbSe$_2$) experimental evidence of both effects pointing to the 3D-2D transition: The upturn of the upper critical field parallel to the layers at about 1.1 K as well as the cusp-like behavior of the angular dependence of $B_{c2}$ below this temperature.

(LaSe)$_{1.14}$(NbSe$_2$) is a low temperature superconductor with $T_c$ around 1.2 K belonging to the family of the lamellar chalcogenides [13], where the two slabs, LaSe and NbSe$_2$, are stacked in a certain sequence. Due to the different symmetry of the LaSe and NbSe$_2$ layers a crystallographic misfit results along one intralayer axis whereas along the perpendicular intralayer axis a perfect fit of both structures is achieved [11]. In the case of (LaSe)$_{1.14}$(NbSe$_2$) every intercalated LaSe layer with the thickness of about 6 Å is sandwiched by one 2H-NbSe$_2$ layer with about the same thickness. The sandwich unit is stabilized by the electron transfer from the LaSe to the NbSe$_2$ slab resulting in the natural layered system of insulating LaSe and (super)conducting NbSe$_2$ sheets, where the conduction is accomplished by the Nb 4d$_x$-$2$ orbitals [11, 12].

A standard lock-in technique at 17 Hz was used to measure the magnetic field dependencies of the sample resistance in a four-probe configuration at different fixed temperatures down to 100 mK. The current and voltage was measured within the same $ab$-plane of the sample. The magnetoresistive transitions were measured at different angles between the $ab$-plane of the sample and the applied magnetic field keeping the current always orthogonal to the field. The angular resolution was better than 0.2 degree with the $\Theta = 0^\circ$ orientation defined from the $c$-plane of the sample and the $c$-plane of the sample. The magnetic field anisotropy

$$B_{c2} |_{c}(T)$$

with a linear decrease towards $T_c$ and a zero-temperature extrapolated $B_{c2}(0)$ equal to 1.2 Tesla instead of 0.4 T obtained at 0.95 $R_N$. An additional argument for the fact that the field value of $B_{c2}$ lies far beyond the main increase of the intralayer resistance has been found in our previous paper [3]. There we have shown that the interlayer transport in (LaSe)$_{1.14}$(NbSe$_2$) is accomplished by the tunneling of quasiparticles and Cooper pairs across the Josephson coupled layers. At higher magnetic fields the tunneling of quasiparticles dominates with a linear magnetic field dependence of the interlayer conductance until this conductance achieves the constant normal-state value at a certain magnetic field defining the value of $B_{c2} |_{c}(T)$. The resulting temperature dependence of the upper critical magnetic field is in a full agreement with the Werthamer prediction and gives $B_{c2} |_{c}(0)$ equal to 1.2 Tesla in agreement with the critical fluctuations model. Therefore, the estimation of the upper critical field from the intralayer resistance measurements in the present experiment inevitably interferes with the dissipative vortex processes and can be taken only as a lower bound of the real upper critical fields.

The upper critical field anisotropy

$$\gamma = B_{c2} |_{ab}/B_{c2} |_{c}$$

c as a function of the temperature is shown in the lower part of Fig. 2. Just below $T_c$ it starts at a value of $\sim$ 40 and rises rapidly to a maximum of $\sim$ 130 at around 1000 mK and it smoothly decreases down with a tendency to
saturation at the lowest temperatures where it achieves a value of $\sim 50$. The rapid rise of $B_{c2}\parallel ab/B_{c2}\parallel c$ below 1.2 K demonstrates the dominant effect of the upturn of $B_{c2}\parallel ab$ compared to the anomalous positive curvature of $B_{c2}\parallel c(T)$. Even assuming $B_{c2}\parallel c(T)$ to be linear between $T_c$ and 400 mK would not have an important effect on the overall temperature dependence of the anisotropy factor with a maximum at about 1.0 K, just the $\gamma$ values would be lowered in this temperature range. This supports a dimensional crossover around 1 K.

Just below $T_c$ the three-dimensional anisotropic Ginzburg-Landau limit is valid in any superconductor because the divergent temperature dependence of the coherence length close to $T_c$ will finally increase $\xi_c$ above the interlayer distance $s$ ($\xi_c > s$). In our interlayer transport measurements we found [2] that the real $B_{c2}\parallel c(1.2)$ K is about 0.2 T giving an in-plane coherence length $\xi_{ab} \sim 400$ Å. Taking into account the anisotropy factor at this temperature we can estimate the out-of-plane coherence length $\xi_c = \xi_{ab}/\gamma \approx 10$ Å. This value is comparable with the $c$-axis lattice constant ($\sim 12$ Å) what indicates a realistic starting condition for the 3D-2D dimensional transition since at lower temperatures $\xi_c$ becomes smaller than $c$ and the flux can be trapped in between the superconducting layers making the orbital pair-breaking effect ineffective.

Below the 3D-2D transition the anisotropic Ginzburg-Landau limit is no longer valid and the interlayer coherence length $\xi_c$ can not be anymore calculated from the parallel upper critical field $B_{c2}\parallel ab$ since the superconducting layers are almost decoupled. For the single superconducting layer of the thickness $d$, the parallel upper critical field is given [13] as $B_{c2}\parallel ab = \Phi_0\sqrt{T/T_c}/(2\pi d\xi_{ab})$. In a more realistic model Deutscher and Entin-Wohlman [20] have shown that a layered superconductor consisting of alternating insulating layers of thickness $s$ and superconducting layers of thickness $d$ shows a dimensional crossover with an upturn in $B_{c2}\parallel ab(T)$ when the interlayer coherence distance $\xi_c$ is smaller than the sum of $s + d$. They have found that the two-dimensional behavior of a layered system is more pronounced when the insulating layer thickness is larger than the superconducting layer thickness. Being in the 2D regime, the temperature dependence of the parallel critical field can be directly compared with that of the square root of the perpendicular critical field. This is demonstrated by the open symbols in the Fig. 1 giving $\sqrt{B_{c2}\parallel c(T)}$ multiplied with a constant factor. At temperatures above 1 K one can see disagreement between the temperature dependence of $B_{c2}\parallel ab$ and of $\sqrt{B_{c2}\parallel c(T)}$ indicating the crossover to the three dimensional regime. Of coarse such a comparison is a significant oversimplification since in our case also the spin-orbit scattering plays an important role producing very high upper critical fields in the $ab$-plane.

Another strong argument for the 3D-2D transition of a superconductor can be obtained via studying the angular dependence of the upper critical field. In Fig. 3 we present the data for four different temperatures at 0.45, 0.58, 1, and 1.2 K. The data are compared with both models: the anisotropic 3D model of Lawrence and Doniach as well as the 2D model of Tinkham. Since for such high degree of anisotropy the models differ only for the field orientations very near to the $ab$-planes, the data are presented only at small range of angles $\Theta$. As is apparent the behavior is cusp-like for the temperatures $T = 0.45$ K and 0.58 K indicating the two-dimensional character of the superconducting state. At $T = 1$ K these two models are almost nondistinguishable due to the high anisotropy factor, but still more probably the system is two-dimensional as no round maximum could be observed in $B_{c2}(\Theta)$. At $T = 1.2$ K a cusp-like form near $\Theta = 0$ is changed to the round maximum indicating more three-dimensional behavior. The values of $B_{c2}\parallel ab$ diminish as temperature increases in a similar way as the data presented in Fig. 2 for a different crystal. Also the anisotropy factor behaves qualitatively as in the Fig. 2, i.e. it increases from the lowest temperature to the 3D-2D crossover and then it drops down in the 3D regime near $T_c$. We note that the value of the anisotropy factor differs from sample to sample [21], but the qualitative picture holds. Such a pronounced cusp-like behavior of $B_{c2}(\Theta)$ in the two-dimensional regime could be observed only on very tiny samples with a clear plane parallel geometry, while in samples with little wavy surfaces the angular dependence always revealed the round maximum which could be fitted by the 3D anisotropic formula.

Finally, in the strongly anisotropic superconductor $(LaSe)_{1.14}(NbSe_2)$ with $T_c = 1.23$ K we have found experimental evidence for the crossover from the three-dimensional to two-dimensional superconducting behavior. Namely, we observed a strong upturn in the temperature dependence of the parallel upper critical magnetic field at 1.1 K with, at lower temperatures, a temperature dependent critical field anisotropy and a cusp-like angular dependence of $B_{c2}$ around the parallel field orientation. All these typical features are observed for the first time in a naturally crystalline layered system.

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FIG. 1. Magnetoresistive superconducting transitions at different temperatures for the applied field perpendicular (upper part) and parallel to the superconducting planes.
FIG. 2. Upper part - resistive upper critical fields for $B_{||c}$ and $B_{||ab}$ are shown by closed symbols. $B_{c2||ab}$ calculated from $B_{c2||c}$ in 2D model - opened symbols. Lower part - the temperature dependence of the anisotropy factor.
FIG. 3. The angular dependence of the upper critical field at different temperatures. Solid line - the 2D Tinkham model, dashed line - the anisotropic 3D Lawrence-Doniach model.