

# Suppression of superconductivity by strong magnetic fields in PbTe/PbS heterostructures with a superconducting interface

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This is a comprehensive study of the effect of strong magnetic fields on superconductivity in PbTe/PbS heterostructures with semiconducting layers of different thicknesses. Metallic conductivity and superconductivity (critical temperature  $T_c \le 6.5$  K) in PbTe/PbS heterostructures are caused by inversion of bands along a continuous network of misfit dislocations that develops at the interfaces between semiconductor layers of sufficient thickness (d > 80 nm). With decreasing d the continuity of the superconducting interface is disrupted,  $T_c$  decreases, and the metallic conductivity changes to a semiconducting type. Disruption of the continuity of the superconducting type. Disruption of the continuity of the superconducting type. Disruption of the continuity of the superconducting transition (SIT) and has a significant influence on its features: a fan-like set of resistance curves R(T); intersection of the R(B) curves for fields perpendicular, as well as parallel, to the interface; and, negative magnetoresistance. A scaling analysis based on Fisher's theoretical model is carried out for these samples. No evidence of a SIT was observed in heterostructures with a perfect interface. It appears that the SIT effect is related to percolation phenomena characteristic of granular superconductors. © 2013 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4818629]

## Introduction

In this paper we present the results of an experimental study of the influence of strong magnetic fields on the superconducting state in nanostructures that form on the interface between semiconducting layers of epitaxial PbTe/PbS structures.<sup>1-6</sup> The possibility of a quantum superconductorinsulator phase transition (SIT)<sup>7</sup> in systems of this type is examined. Superconductor-insulator phase transitions have been under intense study recently and have been found experimentally in a number of low-dimension systems such as ultrathin amorphous films, granular films, and arrays of Josephson junctions. This phenomenon occurs when the internal parameters of the system (such as disorder or film thickness (D-SIT)) change<sup>8-12</sup> or under the influence of external interactions such as magnetic fields (M-SIT),9,11,13-27 electric fields, or transport currents.<sup>28-30</sup> Superconductorinsulator transitions have also been observed experimentally in HTSC compounds,<sup>31-34</sup> and even in one-dimensional, long nanowires.<sup>35</sup> The features of the SIT depend on the type of system and the experimental conditions.<sup>7,36</sup>

Here we study magnetic-field induced superconductorinsulator transitions. The main indicator of an M-SIT is a "fanlike" set of resistance curves R(T) at low temperatures.<sup>7</sup> In magnetic fields lower than a critical value  $B_c$  the resistance decreases with falling temperature. When  $B > B_c$  the picture changes to the opposite: with decreasing temperature the resistance increases and the R(T) curves go upward. Another characteristic sign of the SIT is that the magnetoresistance curves R(B) measured at different temperatures all intersect at a single point. The third distinctive feature of the SIT is the appearance of a negative magnetic resistance at high magnetic fields.<sup>7</sup>

The nature of the superconductor-insulator transition is still an open question. Its cause is more obvious in granulated films than in uniform disordered films, HTSC, and, even more so, in one-dimensional nanowires. In granular systems with small granules<sup>37,38</sup> and artificially prepared regular arrays of Josephson junctions,<sup>39-42</sup> the SIT can be explained by a competition between the inter-granule Josephson binding energy J and the charge Coulomb energy  $E_C$  of an individual grain. When  $E_c \gg J$  Coulomb blocking predominates. As a result, Cooper pairs become localized and the system transforms to an insulating state. If, on the other hand, the granules are larger, then Coulomb blocking is no longer effective. Then a SIT takes place because the Josephson junctions are disrupted by an external interaction (e.g., a magnetic field). Here single particle transport is blocked because of the need to overcome a potential barrier

695

comparable to the superconducting gap energy.<sup>7</sup> There are many theoretical papers on the SIT in arrays of submicronsized Josephson junctions.<sup>43–45</sup> In one of these, a critical temperature for the transition from a superconducting state to an insulating state has been determined taking quantum fluctuations into account.<sup>43</sup>

Interpretation of the superconductor-insulator transition observed experimentally in uniform thin disordered films is much more complicated. As an example, for films with relatively low resistance per square the characteristic features of SIT can be explained by quantum mechanical corrections to the conductivity.<sup>46</sup> In this case, on the insulating side there will be a slight increase of no more than 10% in the resistance. In the case of a large increase in the resistance on the insulating side induced by a magnetic field, the superconducting-insulating transition in uniform thin disordered films is most often explained by Fisher's scaling theory<sup>47</sup> (a theory of duality between Cooper pairs and vortices). It is assumed that at T = 0 delocalized Cooper pairs and localized vortices exist below the transition for fields  $B < B_c$ (superconductor), and localized pairs with delocalized vortices above the transition  $B > B_c$  (insulator). The magnetic field and temperature dependence of the resistance per square follow the scaling law for phase transitions,

$$R(\delta, T) = R_c F(\delta x / T^{1/\nu z}), \tag{1}$$

where *F* is a constant introduced to maintain the dimensionality of the equation;  $\delta$  is a variable parameter that drives the phase transition, in this case, a magnetic field with  $\delta = |B - B_c|$ ; and, *vz* is the critical exponent. The model predicts that the critical resistance per square  $R_c$  should equal the universal quantum resistance  $R_Q = h/4e^2 = 6.5 \text{ k}\Omega$ . The scaling law proposed for the resistance by Fisher<sup>47</sup> is in good agreement with a variety of experimental data.<sup>11,13</sup> Nevertheless, many experiments have yielded a large scatter in the values for the critical resistance and the critical exponents vz.<sup>15,18,36,48</sup> Thus, one of the main predictions of Fisher's theory (a universal quantum resistance) is not observed experimentally in all systems during superconductor-insulator transitions.

A number of proofs of a percolation mechanism for SIT have been published, both experimental<sup>19,49</sup> and theoretical.<sup>50–52</sup> In addition, a numerical simulation<sup>53</sup> including quantum fluctuations in superconducting films with a fairly high level of disorder has shown that that ultrathin films decay (break up) into superconducting islands in an insulating matrix. Experimental evidence of this effect is also given there.<sup>53</sup> It may be assumed that the mechanism for SIT in uniform thin disordered films is similar to that observed for granular films. For example, a two-dimensional disordered ultrathin film of TiN at temperatures close to 0 K has been treated as an array of Josephson junctions, more precisely, granular structures, in which the granules can become superconducting domains at low temperatures; the grains, themselves, are separated by insulating regions that remain so even at ultralow temperatures. Thus, there is still no final universal theoretical model for SIT in low dimensional systems.

Here we report an experimental study of magnetic-field induced superconductor-insulator transitions in superconducting nanostructures formed at the interface between semiconducting layers of epitaxial PbTe/PbS heterostructures. These results provide qualitative support for a percolation mechanism for the superconductor-insulator transition in granular films.

Interest in research on semiconducting PbTe/PbS heterostructures arises from the possibility of creating superconducting nanostructures with different topologies in a controlled fashion at their interface. We have found<sup>3-6</sup> that superconductivity of the interface of A<sup>IV</sup>B<sup>VI</sup> heterostructures is related to an inversion of bands in the narrow-band semiconductors (PbTe, PbS, PbSe) owing to inhomogeneous elastic stresses along a network of misfit dislocations produced at the interface by relaxation of pseudomorphic epitaxial growth stresses. The period of the superconducting nanonetwork is equal to the period of the network of misfit dislocations and, depending on the combination of semiconductors, ranges from 3.3-40 nm. For PbTe/PbS heterostructures it equals 5.2 nm. Thus, by varying the heterostructure parameters, such as the thickness of the semiconductor layers and the number of these, we can create arrays of individual quantum dots with weak Josephson bonds, as well as continuous superconducting nanonetworks and quasi-three dimensional multilayer structures (superlattices). These superconducting nanostructures have properties inherent in 0-, 1-, 2-, and 3-dimensional systems. Thus, semiconducting PbTe/PbS heterostructures can serve as models for the study of the effects of localized superconductivity and for the creation of magnetic field-induced superconductor-insulator transitions in them. Up to now the effect of strong magnetic fields on these structures has not been studied.

#### Techniques for fabricating the samples and for making the transport measurements

The transport properties of more than ten two-layer PbTe/PbS heterostructures were studied. The thicknesses of the semiconducting layers that form the heterostructures discussed here are equal and range over  $d_{PbTe} = d_{PbS} = d$ = 50-100 nm. The lead chalcogenides, PbTe and PbS, are narrow band semiconductors (band gap width  $E_{g} < 0.3 \text{ eV}$  at 4.2 K). All of these heterostructures were fabricated by sequential condensation of the vapors of the corresponding semiconductors PbTe and PbS on a substrate heated to 520-570K in an oil-free vacuum of 10<sup>-6</sup> Torr. A freshly cleaved (001) surface of single crystal KCl was used as a substrate. The lowest layer on the substrate was always of PbS. The thicknesses d of the semiconductor layers and deposition rate were monitored in situ using a calibrated quartz resonator. The lead chalcogenides were thermally evaporated from tungsten boats. Only stoichiometric targets were used for preparing the samples.

These semiconductors have a NaCl-type crystal structure with a small misfit (8%) between the parameters of the unit cells, and during epitaxial growth of the heterostructures the pseudomorphic stresses relax through formation of a network of edge misfit dislocations at the interphase boundary (interface). When a critical thickness  $d_c$  of the upper layer PbTe layer is reached (for the PbS/PbTe system  $d_c = 1$  nm) the first isolated islands of a regular network of misfit dislocations show up at the interface.<sup>6,55</sup> Further increases in the thickness leads to coarsening of the islands and their subsequent merging into a continuous network of misfit dislocations. For larger thicknesses (80–100 nm) a continuous square network of edge misfit dislocations covers the entire interface, but some local defects can exist on it, possibly with an irregular periodicity.<sup>6</sup> The existence of networks of misfit dislocations on the interface was confirmed by "transillumination" electron microscopy (see Fig. 1 of Ref. 6).

The transport measurements were made in a helium cryostat equipped with a superconducting Oxford Instrument solenoid at temperatures of 1.4-300 K. The accuracy of the temperature determinations and the temperature stabilization were within  $10^{-3}$  K.

The resistance R was measured by the four-probe method. The samples were double Hall structures. The measurements were made with dc and alternating current (50 nA, 13 Hz). The direction of the transport current I is parallel to the plane of the sample with  $I \perp B$ . The upper critical magnetic fields  $B_{c2}$  were determined from the average of the resistive transitions at the point  $R = R_n/2$  ( $R_n$  is the residual resistance before the superconducting transition). The technique for the magnetotransport measurements is described in more detail elsewhere.<sup>4-6</sup>

### Experimental data and interpretation

In this paper we study the effect of strong magnetic fields on the superconducting properties of more than 10 bilayered PbTe/PbS heterostructures with different thicknesses  $d_{PbTe} = d_{PbS} = d$  of the semiconducting layers. We shall examine the data for three of the samples, whose basic parameters are listed in Table 1.

As noted in the Introduction, we have found<sup>3–6</sup> that the superconductivity of the interface of  $A^{IV}B^{VI}$  heterostructures is related to band inversion in narrow-band semiconductors (PbTe, PbS, PbSe) owing to periodic elastic stresses created by misfit dislocations near the interphase boundary. That is, a conducting metallic nanostructure which "repeats" the network of misfit dislocations develops at the interface. The period of this superconducting interface nanonetwork is 5.2 nm for PbTe/PbS nanostructures.<sup>3–6</sup>

During a comprehensive experimental investigation it was found that two-layer PbTe/PbS heterostructures can be divided nominally into 3 categories (although there is no sharp boundary between these categories).<sup>6</sup> The first category includes samples with semiconducting layer thicknesses  $d \ge 80$  nm. They have a metallic conductivity in the normal state. The ratio of the resistance at room temperature to the resistance before the onset of the superconducting transition,  $r=R_{300}/R_n$  varies from 2-8. The corresponding critical temperatures  $T_c$  lie in the interval 4.2–6.5 K. Sample C belongs to this category.

The second category includes samples with thicknesses of 50-70 nm. This category can be referred to as

intermediate. A sample in the normal state can exhibit both metallic conductivity and semiconductor behavior, but, regardless of the type of conductivity, at low temperatures it enters a superconducting state. The critical temperature ranges from 2.3–3.3 K and  $r = R_{300}/R_n$  ranges from 0.9–1.7. Samples A and B belong to this category.

The third category includes samples with  $d \le 50$  nm. The R(T) curves in the normal state for these samples are always characterized by a negative resistance coefficient dR/dT above  $T_c$ . The resistance per square  $R_{\Box}$  exceeds  $1.5 \,\mathrm{k\Omega}$  and r < 1. For these systems  $T_c$  is often below 1 K and they undergo an unending transition into the superconducting state down to the lowest temperatures at which the experiments were carried out (0.3 K), or they do not go into the superconducting state at all. Samples from the third category were not examined in the present experiments.

Figures 1–5 show the results of our experimental study of the effect of magnetic fields B on the superconducting properties of samples A, B, and C. The samples from the second category (the PbTe/PbS heterostructures with semiconducting layers of thickness 40 < d < 80 nm, i.e., samples A and B) have the most interesting behavior in strong magnetic fields. All the characteristic features of magnetic field induced superconductor-insulator transitions mentioned in the Introduction are observed for these samples (Figs. 1-4). It should be noted that the behavior of these samples depends substantially on the thickness of the semiconductor layers and, therefore, on the characteristics of the superconducting nanostructures at the interfaces. In particular, the two types of evolution of the R(T) curves characteristic of a magneticfield induced M-SIT as the magnetic field is varied are observed experimentally. The first type, illustrated in Fig. 1 (sample A), is characterized by the presence of a horizontal separatrix which clearly separates the R(T) curves with a superconducting transition that move downward with decreasing temperature from the R(T) curves with the increasing resistance characteristic of an insulator, i.e., which move upward with decreasing temperature. "Fanlike" curves of this type are regarded as "ideal" for observing M-SIT.<sup>7</sup> The behavior was observed in all the heterostructures with semiconductor layer thicknesses  $d_{PbS} = d_{PbTe}$ = 70 nm with a conducting network at the interface that has "weak" defects, which repeat the defects in the misfit dislocation network; see the electron microscope image of a network of misfit dislocations in Fig. 1(b) of Ref. 6.

For samples with "ideal fan" curves, i.e., with a horizontal separatrix, we always see another distinctive sign of the M-SIT—a single point at which the magnetic-field dependences of the resistance at different temperatures, R(B), for magnetic fields perpendicular and parallel to the interface (Fig. 2), intersect. Figure 2 shows that all the magnetic field curves corresponding to different temperatures intersect

TABLE 1. Parameters of the PbTe/PbS samples.

Sample	<i>d</i> , nm	$R_n, \mathbf{k}\Omega$	<i>T</i> <sub>c</sub> , K	$B_{c2\parallel}(0), \mathrm{T}$	B <sub>crll</sub> , T M-SIT	$B_{c2\perp}(0), \mathrm{T}$	$B_{cr\perp}$ , T, M-SIT
A	70	1.2	3.3	1.86	2.4	0.27	1.23
В	60	1.4	3.1	1.23	Not determined	0.17	Not determined
С	100	0.17	6.5	2.81	Nonexistent	1.02	Nonexistent

Note:  $d_{\rm PbTe} = d_{\rm PbS} = d$ .

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FIG. 1. R(T) for different values of the magnetic field (*B*, T) parallel to the interface; d = 70 nm.

precisely at a single point for which the parameters are the critical M-SIT resistance  $R_c = 1.126 \text{ k}\Omega$  and the critical parallel magnetic field for the M-SIT,  $B_{crll} = 2.4 \text{ T}$ . The magnetic field curves for a perpendicular magnetic field look exactly the same qualitatively, but the critical perpendicular magnetic field is  $B_{cr\perp} = 1.23 \text{ T}$ .

The existence of a horizontal separatrix opens up the possibility of a scaling procedure based on Eq. (1) in accordance with Fisher's theoretical model.<sup>47</sup> Strictly speaking, Fisher's model is applies only to a perpendicular magnetic field. Thus, for sample A this scaling with a magnetic field perpendicular to the interface is shown in the inset to Fig. 2. The critical parameters for the M-SIT are determined from the scaling curves: the perpendicular critical magnetic field for M-SIT  $B_{cr\perp} = 1.23$  T, which naturally coincides with the value for the single intersection point of the magnetic field curves, and the critical exponent vz = 3.7. The perpendicular M-SIT critical field for sample A is substantially higher than the upper perpendicular critical field  $B_{c2\perp}(0) = 0.17 \text{ T}$  found by extrapolating the linear segment of  $B_{c2\perp}(T)$  to T = 0. We note, however, that our value of  $B_{c2\perp}(0)$  may be inaccurate because there is only a small linear segment in the  $B_{c2\perp}(T)$ curve.6

Scaling was done for the resistance curves measured in a parallel magnetic field, when the concept of duality of the



FIG. 3. R(T) for different values of the magnetic field (*B*, T) parallel to the interface; d=60 nm.

Cooper pairs and vortices<sup>47</sup> is not applicable and the critical exponent has a different significance. The scaling dependences were used to determine the critical parameters for M-SIT with a parallel magnetic field:  $B_{crll} = 2.4$  T and critical exponent vz = 1.23. The parallel SIT critical field  $B_{crll} = 2.4$  T is also higher than the upper critical field  $B_{c2ll}(0) = 1.86$  T determined from the temperature dependence  $B_{cr2ll}(T)$ . A detailed comparison of the upper critical magnetic fields and the critical fields for the magnetic-field induced superconductor-insulator transition will be reported in a later paper.

It should be noted that for samples with "ideal fanlike" curves, i.e., with a horizontal separatrix, the resistance in high magnetic fields is roughly 10% higher than  $R_C$  in fields of about 6 T even for temperatures near 1.5 K, but not at ultralow temperatures as in the case of ultrathin disordered films.

A more complicated fanlike R(T) curve (Fig. 3, sample B) is observed experimentally in thinner samples with  $d_{PbS} = d_{PbTe} = 60$  nm. At intermediate fields B = 0.8 to 1.6 T the R(T) curves have two extremes: a minimum near the onset of the superconducting transition and a maximum at lower temperatures with a subsequent transition into the superconducting state.<sup>7</sup> The separatrix is inclined and nonlinear, so that Fisher's single-parameter scaling theory is not applicable, and a critical value of the resistance for the superconducting-insulator transition,  $R_C$ , cannot be determined.<sup>56</sup> The R(B) curves obtained for different temperatures also intersect, but, because of the



FIG. 2. R(B) for different temperatures in a magnetic field parallel to the interface; d = 70 nm. The inset shows the scaling relation  $R/R_n$  for a perpendicular magnetic field.



FIG. 4. R(B) for different temperatures in a magnetic field parallel to the interface; d = 60 nm.

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FIG. 5. R(B) for different temperatures in a magnetic field parallel to the interface; d = 100 nm.

inclination of the separatrix, a single intersection point does not exist. A resistance minimum is observed in systems with thicker dielectric layers. If their thickness is comparable to the coherence length, then superconductivity occurs in steps: first in the granules and then, as the temperature is lowered, a Josephson coupling develops between them. The negative value of dR/dT is explained by "freezing" of the thermally activated single particle transport along the semiconductor spacers between the superconducting granules.<sup>7,15</sup>

For heterostructures with thicknesses 40 < d < 80 nm, a third sign of SIT is observed in parallel magnetic fields: a negative magnetoresistance which shows up most distinctly in samples with d < 60 nm (Fig. 4, Sample B).

It has been assumed previously that the superconductorinsulator phase transition is characterized by a universal quantum resistance  $R_c = R_Q = h/4e^2 \approx 6.5 \text{ k}\Omega^{47}$  Now, relying on a large number of experiments, it is assumed that no universal resistance  $R_c$  exists for these systems.<sup>7</sup> No universal quantum resistance was observed in our experiments either. However, in our case the situation is made more complicated by the impossibility of determining exactly the critical resistance per square,  $R_c$ , because of the unclear geometry of the superconducting interface, which is made up of a multicoupled superconducting nanostructure with a variable thickness.<sup>5</sup> In addition, the critical resistance is reduced by the shunting effect of the semiconducting layers of PbTe and PbS, which have a finite resistance.<sup>6</sup> We also see no need to search for weak localization effects, because the increase in the resistance in a magnetic field on the insulating side of the SIT correlates clearly with the onset of the superconducting transition and because of the multiphase nature of the granular medium.

Figure 5 shows that no characteristic features of M-SIT were observed for heterostructures in the first category (d > 100 nm) with a continuous network of misfit dislocations and, therefore, a continuous<sup>6</sup> superconducting interface (sample C). Thus, it has been shown that the major condition for the occurrence of a magnetic-field induced SIT in PbS/PbTe heterostructures is an insular structure of the superconducting interface.

At the same time, there are many questions<sup>7,36</sup> regarding the realization of SIT. Why is the universal quantum resistance found in many experiments not a necessary attribute of SIT? Should the structure be uniform granular or is random percolation sufficient?<sup>52</sup> What shape and size<sup>57</sup> of the granules will ensure reliable suppression of quasiparticle transport between the granules of the superconductor? Is Coulomb blocking actually necessary? How does a spatial inhomogeneity of the order parameter develop in uniformly distributed films? Answers to these questions will require more detailed studies of the features of superconducting-insulating transitions at low temperatures.

#### Conclusions

The suppression by strong magnetic fields of superconductivity in the self-organized interfaces of superconducting nanostructures in PbTe/PbS heterostructures with semiconducting layers of different thicknesses has been studied experimentally.

For the first time, all the characteristic signs of magneticfield induced superconductor-insulator transitions have been observed in these structures. The structural features of the interface are found to have a significant effect on the conditions for development of M-SIT and on their characteristic features. SIT are observed in samples with a defect (island) structure of the superconducting interface, but do not occur in samples with defect-free, nearly ideal structures of the superconducting interface.

The mechanism for SIT in these objects is similar to the percolation mechanism in granular systems.

Thus, it has been shown that A<sup>IV</sup>B<sup>VI</sup> semiconducting heterostructures can serve as a model for studying the features of magnetic-field induced superconductor-insulator transitions, since the properties of the superconducting interface can be varied in a controlled manner in these materials.

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