ThAsSe diamagnet: Evidence for a Kondo effect derived from structural two-level systems

Tomasz Cichorek
Max Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany
and Institute of Low Temperature and Structure Research, Polish Academy of Sciences, 50-950 Wroclaw, Poland

Hidekazu Aoki, Jeroen Custers, Philipp Gegenwart, and Frank Steglich
Max Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany

Zygmunt Henkie
Institute of Low Temperature and Structure Research, Polish Academy of Sciences, 50-950 Wroclaw, Poland

Eric D. Bauer and M. Brian Maple
Department of Physics and Institute for Pure and Applied Physical Sciences, University of California, San Diego, California 92093, USA
(Received 14 April 2003; published 8 October 2003)

We report a study of an anomalous scattering mechanism in the structurally disordered diamagnet ThAsSe. Below 20 K, its electrical resistance displays a logarithmic correction over one decade in temperature, which is affected by neither strong magnetic fields ($B \leqslant 17$ T) nor high hydrostatic pressures ($p \leqslant 1.88$ GPa). Dynamic disorder in the ThAsSe single crystals is reflected by, e.g., a quasilinear-in-T term of nonelectronic origin in the specific heat at $T \lesssim 1.7$ K. The $\ln T$ dependence of the resistance is interpreted as being due to the interaction of the conduction electrons with movable structural defects.

DOI: 10.1103/PhysRevB.68.144411 PACS number(s): 66.35.+a, 72.10.Fk, 72.15.Qm

I. INTRODUCTION

Within the broad spectrum of properties of amorphous and disordered solids, phenomena related to quantum tunneling take a special position. This is due to the fact that these processes may determine the low-energy physics of matter with some kind of disorder.[1] The simplest realization of a tunneling center is believed to be an atom that tunnels between two metastable states of the double-well potential. An ensemble of them, called two-level systems (TLS), typically displays a broad distribution in terms of the energy splitting and/or the tunneling probability. One of the fascinating aspects of the TLS is the appearance of an exotic Kondo effect; the Hamiltonian of the TLS interacting with a degenerate Fermi gas can be mapped to the Hamiltonian of the spin-$\frac{1}{2}$ Kondo problem.[2–4] At the strong-coupling fixed point, the spin of the electrons acts as a ''flavor'' variable and reflects the channel index in the two-channel Kondo effect. Thus, in this particular case, it is expected that the system would have a non-Fermi-liquid (nFL) ground state below the Kondo temperature T_K and above the Kondo temperature, a logarithmic temperature dependence of the electrical resistance, $R(T)$, should be observed.[3–6] The recent studies have shown that considerable T_K can be achieved only outside the tunneling regime, i.e., when the first excited state is above the barrier.[7] In such a case, however, the splitting is significantly larger and hence the nFL region is harder to reach.[8] In the TLS model the $\ln T$ singularity has its origin in the nonmagnetic interaction of the itinerant electrons with the defect centers. As a consequence, due to the electron-assisted hops, a movable particle cannot be localized in one of the two metastable states. This resembles the quenching of a magnetic moment of a single impurity by the conduction electrons in the classical spin Kondo problem. It is worth mentioning that Kondo himself investigated theoretically an interaction of conduction electrons with an atom jumping between two equivalent positions.[9]

The interest in two-level systems dates back to the early 1970s, when their influence on the low-temperature thermal properties in disordered solids was discovered.[10] Recently performed point-contact spectroscopy measurements on Cu nanoconstrictions revealed the anomalous zero-bias response that points to a coupling of the tunneling centers to the conduction electrons.[11,12] To date, however, there exists no convincing example for this electron-TLS interaction on a macroscopic scale. Experimental evidence for such a scattering mechanism in a bulk material might verify some of the aspects of the TLS Kondo model and open a new route to the nFL problem.

Historically, Cochrane *et al.*[13] were the first who raised the question whether structural disorder can lead to the existence of a low-energy degree of freedom to which the conduction electrons can couple. For the paramagnetic $Ni_{75}P_{25}$ alloy, they have found a $\ln T$ increase in $R(T)$ upon cooling that is unaltered by a magnetic field of 4.5 T. However, further studies on this and other related alloys, showed that their low-T properties are not equivalent to the ones expected for the TLS Kondo effect.[14,15] Two systems, the degenerate $Pb_{1-x}Ge_xTe$ semiconductor and Ni_xNb_{1-x} metallic glasses, are among the most interesting materials to investigate the TLS Kondo problem. In the former system the density of defect centers can be precisely controlled by the Ge concentration.[16] In the latter one, point-contact spectroscopy measurements have hinted at a nonmagnetic Kondo-type scattering.[17] Unfortunately, in both the cases, low-lying phase transitions interfere with the low-temperature $R(T)$ singularity, and hence prevent one from studying the TLS Kondo problem in more detail.

Our interest in the diamagnetic compound ThAsSe originates in recent studies on its U-based homologue. It has been shown that uncommon transport properties of the structurally disordered ferromagnet UAsSe may be consistently interpreted in terms of a TLS approach. This concerns, e.g., the disorder-dependent low-T upturn in $R(T)$ far below the ferromagnetic transition at around 110 K,[18] as well as changes in the thermoelectric power introduced by tiny variations of Se excess (less than 6%).[19] Thus, if the electron-TLS interaction is realized in UAsSe, some indication for it should be also observed in ThAsSe owing to the structural disorder in the anionic sublattice, as suggested by means of x-ray and scanning-electron-microscopy studies as well as ^{77}Se nuclear-magnetic-resonance measurements.[20–22] Moreover, the high-resolution transmission-electron-microscopy experiments point to the perfect occupation of the cationic sublattice of both systems without any hints at a disordered structure.[20]

This paper is organized as follows. In Sec. II, we briefly report some experimental details. Section III includes results of the electrical resistance and heat-capacity measurements performed on single crystalline ThAsSe under different conditions. At the beginning of Sec. IV, we discuss the low-temperature specific-heat data that provide clear evidence for a dynamic disorder in ThAsSe. In the main discussion, however, we analyze such experimental findings that differentiate the electron-assisted transition from other phenomena leading to a similar temperature variation of the resistance at ambient conditions. Section IV ends with an interpretation of our low-temperature $R(T)$ data for ThAsSe in terms of the TLS Kondo model. Finally, Sec. V gives our conclusions and an outlook.

II. EXPERIMENT

Single crystals of diamagnetic ThAsSe were grown by the chemical vapor transport method. Growing conditions resulted in platelike crystals with typical thickness of less than 1 mm and masses of a few mg. X-ray-diffraction measurements revealed the tetragonal structure (PbFCl-type, space group $P4/nmm$) with lattice parameters $a=4.084$ Å and $c=8.578$ Å. The crystallographic unit cell of ThAsSe with exposed ($ThAs_4Se_5$) polyhedrons, formed by the closest anion neighborhood of thorium, is shown in Fig. 1. More details concerning sample preparation as well as their crystallochemical analysis are described elsewhere.[20]

Due to the platelike shape of the crystals, the electrical resistance has been investigated in the basal plane only. Experiments were performed in zero and applied magnetic fields up to 17 T at temperatures as low as 0.02 K and under hydrostatic pressure up to 1.88 GPa. Electrical contacts were made by spot welding 25 μm gold wires to the crystals, by which self-heating is significantly reduced. To the same aim, the ac electrical current as low as 100 μA has been applied in the millikelvin temperature range. High pressure was achieved in a piston-cylinder cell utilizing fluorinert FC 77 as pressure transmitting medium. From the pressure dependence of the superconducting transition temperature of lead, measured by ac susceptibility, we estimated the absolute value of the hydrostatic pressure.

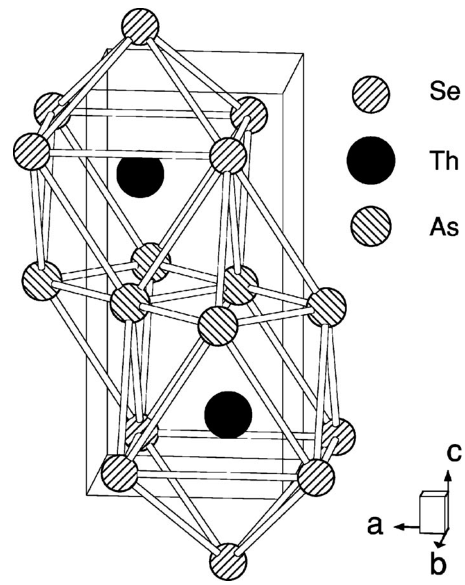

FIG. 1. Crystallographic unit cell of ThAsSe with exposed ($ThAs_4Se_5$) polyhedrons formed by the anion nearest neighbors of thorium.

The specific heat, in the temperature range 0.37–300 K, was determined with the aid of the thermal-relaxation technique utilizing two different commercial microcalorimeters (Oxford Instruments and Quantum Design). The dc magnetic susceptibility in fields applied along and perpendicular to the c axis was measured between 2 and 300 K using a superconducting quantum interference device magnetometer (Quantum Design).

III. RESULTS

In Fig. 2 we show the temperature dependence of the resistance normalized to its value at 300 K for several single crystals of ThAsSe. Whereas the resistivity ratio $RR=R(4\ \mathrm{K})/R(300\ \mathrm{K})$ of the crystals investigated by us is

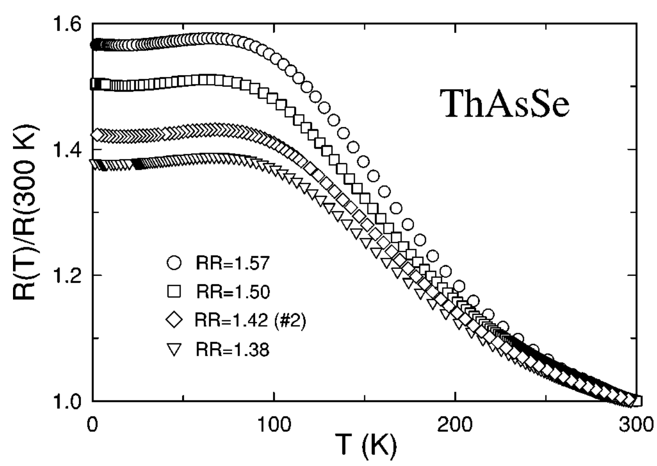

FIG. 2. The ab-plane electrical resistance normalized to its value at 300 K, $R(T)/R(300\ \mathrm{K})$, for different single crystals of ThAsSe as a function of temperature.

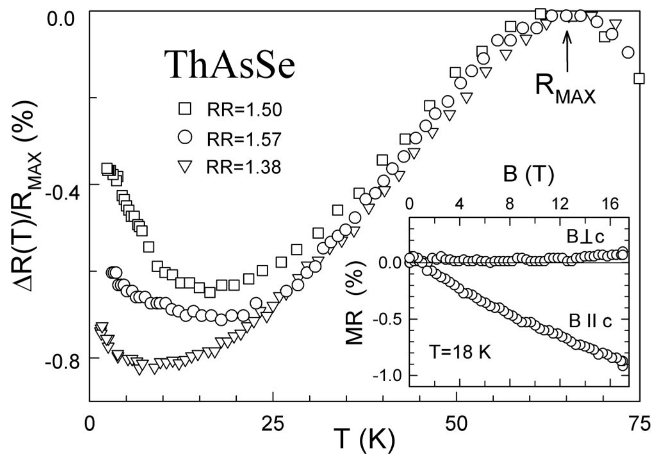

FIG. 3. The relative change of the low-T resistance normalized to its maximum value at around 65 K, $\Delta R(T)/R_{max}$, of different ThAsSe single crystals down to 2 K. The measurements have been done on the same specimens as in Fig. 2. Inset: The transverse magnetoresistance along the ab plane for the single crystal of ThAsSe with $RR=1.57$ at 18 K in magnetic fields applied along and perpendicular to the c axis.

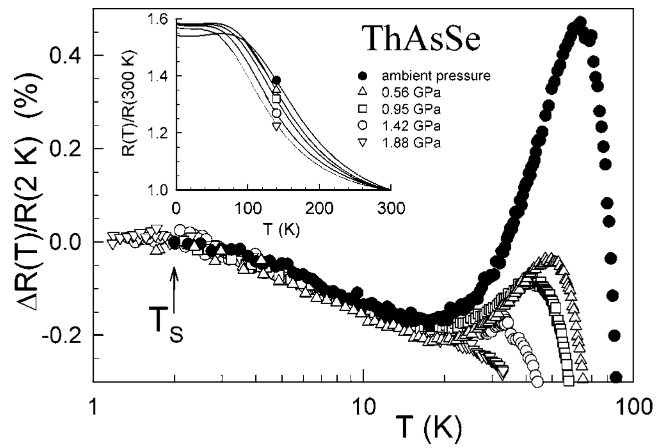

FIG. 4. Effect of hydrostatic pressure on the relative change of the low-T resistance normalized to the corresponding value at 2 K, $\Delta R(T)/R(2\ \mathrm{K})$, for the ThAsSe single crystal with $RR=1.54$. The arrow indicates the temperature of saturation T_S. Inset shows the ab-plane $R(T)/R(300\ \mathrm{K})$ dependence at $p \leqslant 1.88$ GPa in the entire temperature range.

much smaller than 2.25 reported in the literature and varies between 1.38 and 1.57, the overall $R(T)$ behavior is very similar.[23] (In the following, we will identify the samples by their RR values.) The negative temperature coefficient of the resistance holds down to $\simeq 65$ K. Upon further cooling $R(T)$ levels off. For the specimen with $RR=2.25$, an ab-plane room-temperature resistivity close to 220 $\mu\Omega$ cm was found.[23]

Figure 3 allows for a closer inspection of our low-temperature $R(T)$ results for ThAsSe. The data were normalized to the maximum resistance R_{max} at $\simeq 65$ K. Below around 20 K, the resistance increases again with decreasing temperature. We found that samples with almost the same RR value show considerable deviations in both the size and the temperature dependence of this low-T upturn. In the inset of Fig. 3 we plot the transverse magnetoresistance, MR(B) $=[R(B)\text{-}R(0)]/R(0)$, in fields applied both perpendicular and parallel to the c direction. The MR(B) data have been obtained at 18 K for the specimen with $RR=1.57$. For $B\perp c$, the resistance change due to an applied magnetic field of 17 T is extremely small, i.e., 0.1%. For $B\| c$, the magnetoresistance is negative and varies almost linearly with increasing field, reaching almost 1% at 17 T.

Although the low-T resistance is strongly sample dependent, for several specimens a distinct $-\ln T$ behavior was observed in a wide temperature window. At the lowest temperatures, the resistance is found to saturate. An example is shown in Fig. 4. There we have plotted the relative change of the resistance normalized to the corresponding value at 2 K, $\Delta R(T)/R(2\ \mathrm{K})$, for the single crystal of ThAsSe with $RR=1.54$. In addition, the influence of the hydrostatic pressure up to 1.88 GPa is displayed. As already reported in Ref. 24 and presented in the inset of Fig. 4, the overall $R(T)/R(300\ \mathrm{K})$ behavior slightly changes due to applied pressures. However, the RR ratio is hardly affected by $p \leqslant 1.88$ GPa. (A rapid increase of the ambient-pressure value of RR from 1.54 to 1.58 at $p=0.56$ GPa is, most likely, of an extrinsic nature, e.g., due to microcracks.) Several important observations are made at $T<100$ K: The application of pressure shifts the position of R_{max} towards lower temperatures; simultaneously, its value is significantly reduced. At $p=1.88$ GPa, the maximum in $R(T)$ disappears. Obviously, due to the suppression of the humplike anomaly, the $R(T,p)$ curves are different at intermediate temperatures. Below around 20 K, there are no differences between the various $R(T,p)$ curves as far as the $\Delta R(T)/R(2\ \mathrm{K})$ dependence is concerned. This holds for both the logarithmic rise over one decade in temperature and the pressure-independent temperature at which the saturation sets in, $T_S \simeq 2$ K.

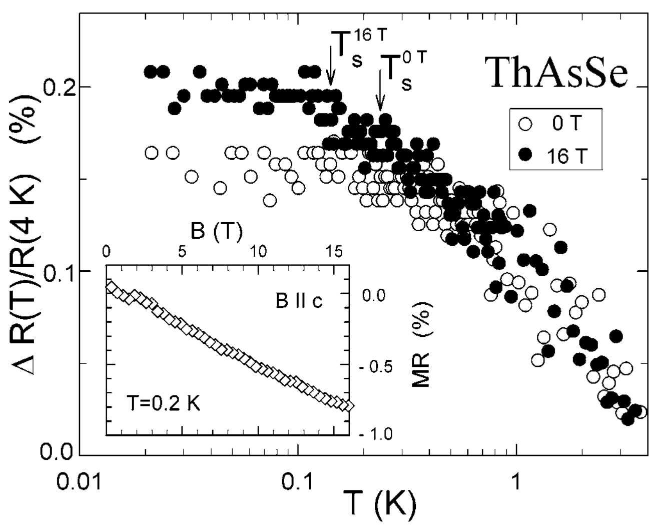

FIG. 5. The ab-plane $\Delta R(T)/R(4\ \mathrm{K})$ dependence for the ThAsSe single crystal with $RR=1.42$ (No. 1) between 0.02 and 4 K in zero field and 16 T. The saturation of the resistance upon cooling is indicated by the arrows. Inset: the transverse magnetoresistance at 0.2 K. The magnetic field was aligned parallel to the c axis.

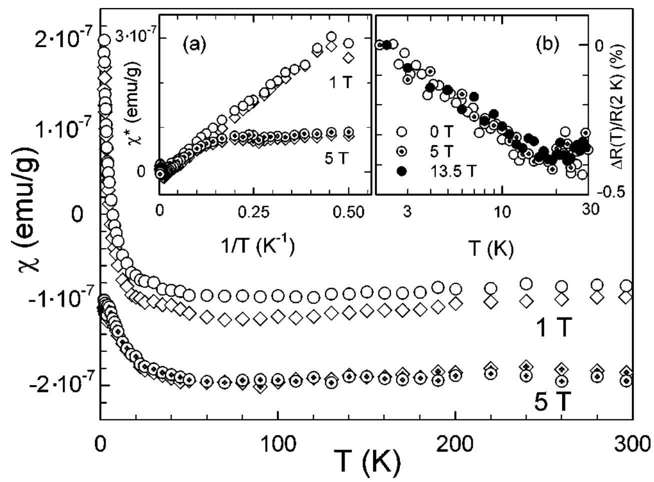

FIG. 6. The dc magnetic susceptibility for the ThAsSe single crystal with $RR=1.42$ (No. 2) measured in magnetic fields of 1 and 5 T applied perpendicular (circles) and parallel (diamonds) to the c axis. (a) The same results plotted as χ^* vs $1/T$, where χ^* denotes the low-T susceptibility, from which the average value of the diamagnetic component was subtracted. Note a pronounced saturation of $\chi(T)$ below around 8 K in a magnetic field of 5 T. (b) The low-T upturn for the same specimen shown on a semilogarithmic temperature scale. The results, plotted as $\Delta R(T)/R(2\text{ K})$, were obtained along the ab plane in varying magnetic fields applied along the c axis. We emphasize that there exists no field effect on the low-T upturn at $B \leqslant 13.5$ T.

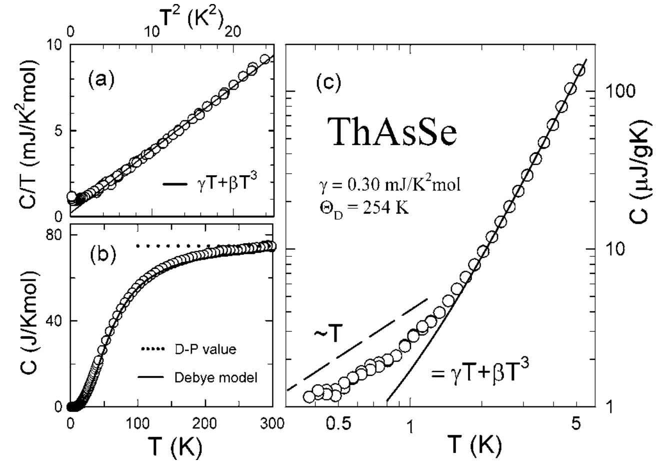

FIG. 7. (a) The low-T specific heat, as $C(T)/T$ vs T^2, of a diamagnetic ThAsSe between 0.37 and 5 K. The measurements have been done on a few single crystals giving a total mass of 30 mg. The solid line shows the $\gamma T+\beta T^3$ dependence with $\beta = 3\times 1944/\theta_D^3$ in units of J/K^4 mol. Fitting parameters were determined from the temperature range 1.7–5 K. (b) Molar heat capacity for ThAsSe up to 300 K. The solid line is the Debye approximation with $\theta_D = 254$ K and the dotted line denotes the Dulong and Petit value. (c) The low-temperature $C(T)$ data on a double-logarithmic scale. The solid line represents a fit with the function $\gamma T+\beta T^3$, the dashed line indicates a linear temperature dependence of $C(T)$.

The significant variation of T_S in ThAsSe is highlighted by the results presented in Fig. 5. There we show the $\Delta R(T)/R(4\text{ K})$ dependence for the single crystal with $RR=1.42$ (No. 1), obtained between 0.02 and 4 K in $B=0$ and 16 T. For this specimen, the resistance increases to around 0.2 K and saturates at the lowest temperatures. The $\Delta R(T)/R(4\text{ K})$ value slightly increases with B at $T \lesssim 0.2$ K. Furthermore, we state that the temperature T_S is somewhat lower for the $B=16$ T data than for zero-field ones. We believe that the self-heating effects are not responsible for the saturation of the resistance at temperatures as low as 0.2 K for the following reasons. First, no detectable change of T_S was found for the current being reduced by one order of magnitude. Second, if the saturation would be due to overheating or radioactivity, T_S should not be altered by magnetic fields.

In the inset of Fig. 5 we show the isothermal MR data ($T=0.2$ K) obtained in fields applied parallel to the c axis, with the current perpendicular to the field direction. Similarly to the MR(B) results at 18 K, the linear decrease of the resistance with increasing B does not exceed 1% in 16 T.

The dc magnetic susceptibility $\chi(T)$ of another single crystal with $RR=1.42$ (No. 2), measured in magnetic fields of 1 and 5 T, is plotted in Fig. 6. The $\chi(T)$ data are temperature-independent and negative in a wide temperature range as expected for a diamagnetic material. However, upon cooling to below 50 K, the susceptibility increases as $1/T$ [Fig. 6(a)] and then saturates below around 8 K in a magnetic field of 5 T. A saturation of $\chi(T)$ at 1 T seems to develop below $T=2.5$ K, too. This strongly field-dependent paramagnetic contribution is most probably due to a very small amount of magnetic impurities carrying around $5.4\times 10^{-4}\ \mu_B$/f.u. Assuming that this is due to U contamination, we estimate 0.09 mg of uranium per gram of ThAsSe, i.e., less than 0.01%. There are no significant distinctions between the $\chi_{ab}(T)$ and $\chi_c(T)$ data taken in the same field.

In Fig. 6(b) we show the relative change of the ab-plane resistance normalized to the corresponding 2 K value, $\Delta R(T)/R(2\text{ K})$, obtained on the same specimen in different magnetic fields. Note that there are no distinctions between the $\Delta R(T)/R(2\text{ K})$ dependencies upon applied fields $B \leqslant 13.5$ T. This holds for both the size and the $\ln T$ behavior of the low-temperature increase.

In a first attempt, no linear-in-T contribution to the specific heat could be resolved for any of the ThAsSe specimens investigated. Therefore, in spite of the strongly sample-dependent resistance, we decided to enlarge the total mass (up to 30 mg) by putting a few single crystals together into the cryostat. In Fig. 7(a) we show our specific-heat data, as $C(T)/T$ vs T^2, obtained between 0.37 and 5 K for such a quasi-single-crystalline sample of ThAsSe. The Sommerfeld coefficient γ close to 0.3 mJ/K^2 mol as well as the Debye temperature θ_D equal to 254 K were determined from the data in the range $1.7 \leqslant T \leqslant 5$ K. The very small electronic term allowed us to detect an additional contribution to $C(T)$ upon cooling below 1.7 K. This quasilinear-in-T term dominates the low-temperature heat capacity of ThAsSe, as displayed in Fig. 7(c) in a double-logarithmic plot.

The Debye temperature for ThAsSe is very close to $\theta_D = 252$ K estimated for a UAsSe single crystal with the Curie temperature $T_C = 101.5$ K that displays the most pronounced upturn in $R(T)$ far below the ferromagnetic transition.[25] This clearly points to a far-reaching analogy in collective vibrations of the crystallographic structure of both systems. The temperature dependence of the heat capacity of ThAsSe up to 300 K is presented in Fig. 7(b). The high-temperature $C(T)$ data are well described by the Debye formula taking a single empirical parameter, i.e., $\theta_D = 254$ K, only. At $T > \theta_D$, the Dulong and Petit value is reached. No phase-transition anomaly has been detected in the entire temperature range.

IV. DISCUSSION

So far, very little is known on the diamagnetic compound ThAsSe.[23,26,27] From optical reflectivity measurements at 300 K, an electron density $n = 3.6\times 10^{22}$ cm^{-3} was estimated.[26] On the other hand, Hall-effect results interpreted in the one-band approximation point to $n(300\ \mathrm{K}) = 8.4\times 10^{21}$ cm^{-3} as well as a linear decrease of $n(T)$ upon lowering the temperature down to around 100 K.[23] At $T \lesssim 100$ K, the electron density of ThAsSe approaches a constant value of 1.1×10^{21} cm^{-3}. Although the one-band approach to ThAsSe can be questioned (ThAsSe seems to be a compensated metal with multiband structure because its primitive cell includes two formula units), the statement of Ref. 23 concerning the temperature-independent carrier density below around 100 K remains unchanged.

Even though the anomalous behavior of $R(T)$ above 65 K has been attributed to the effect of charge suppression, no evidence for a change of the electronic structure could be found.[23] Thus, the negative temperature coefficient of the resistance in ThAsSe at 65 K $< T <$ 300 K remains unclear. Nevertheless, the strong sample-dependent RR ratio should be related to a variation of the As-Se composition, since the Th sublattice of the ThAsSe crystals investigated is free of structural defects.[20] Consequently, the high-temperature $R(T)$ data provide additional evidence for disorder in the As-Se sublattice. Note that a detailed crystallochemical analysis did not reveal any difference in the As-Se composition between various specimens.[20]

In the following, we will focus only on the low-T properties of ThAsSe, i.e., on the features observed at temperatures where the electron density is constant.[23] We start with our low-T specific-heat results, which carry the key information on the nature of the disorder in ThAsSe: The nuclear magnetic heat capacity, C_N, as the cause for the additional contribution to $C(T)$ found below 1.7 K can be safely ignored. In fact, the temperature at which the $C(T)$ data start to deviate from the $\gamma T + \beta T^3$ dependence would be much higher than in any other nonmagnetic system displaying a nuclear-magnetic Schottky anomaly.[28] Furthermore, the deviations due to a nuclear Schottky anomaly in magnetically ordered USe and UAs are detectable only below 0.25 and 0.3 K, respectively, i.e., well below the low-temperature limit of our specific-heat measurements.[29] Finally, the nuclear-quadrupole specific heat is negligible in ThAsSe as well, despite a noncubic environment of all the nuclei. This is because of the very small nuclear electric quadrupole moment of the ^{75}As isotope (0.29×10^{-24} cm^2), the only component that possesses a nuclear spin larger than 1/2. A detailed analysis of the low-T dependence of $C(T)$ for ThAsSe requires more experimental work. Especially, specific heat and ultrasonic measurements on one sufficiently large single-crystalline specimen are needed. However, we do not expect to find, in these future experiments, significant deviations from our present results, concerning (i) the temperature below which the additional term occurs, $T \simeq 1.7$ K, as well as (ii) the size (≈ 1 μJ/gK at 0.5 K) and (iii) the quasilinear T dependence of the additional term. By the negligible C_N term, all these features should to be ascribed to dynamic disorder derived from two-level systems. Indeed, the results from Fig. 7(c) reveal striking similarity to the heat-capacity data for, e.g., vitreous SiO_2 — the textbook example of the TLS material.[1,10]

Next we turn to our low-temperature electrical resistance results, which reveal upon cooling an anomalous upturn in all the ThAsSe single crystals measured. The question that naturally arises here is: how far are the transport properties of ThAsSe modified by dynamic disorder? In other words, does the low-T upturn originate from the electron-TLS interaction or is it indicative of another phenomenon? To resolve the low-lying excitations in a ThAsSe electron system, we discuss different scattering mechanisms and interaction phenomena showing a similar temperature variation of the resistance. A pronounced $-\ln T$ dependence observed for several ThAsSe single crystals allows us to confine the discussion to only a few of such mechanisms.

Frequently, electron scattering on magnetic impurities, i.e., the magnetic Kondo effect, is the main reason for the $-\ln T$ dependence. Its irrelevance to the ThAsSe specimens investigated is already suggested by the extremely small number of magnetic impurities, as inferred from the dc magnetic susceptibility data. Nevertheless, in order to completely exclude the magnetic Kondo effect, we studied the influence of strong magnetic fields on the resistance of ThAsSe (cf. the insets of Figs. 3 and 5). Both isothermal MR(B) curves measured in fields parallel to the c direction exhibit very similar behaviors, although the temperature of the measurements differs by almost two orders of magnitude. Note that the MR(B) data were obtained near the maximum and the minimum of the low-T singularity. This clearly points at a nonmagnetic origin of the additional scattering. However, the most convincing proof that magnetic impurities are no cause of the low-T upturn in the electrical resistivity of ThAsSe is reflected by the $\chi(T)$ and $R(T)$ results on the same specimen, as presented in Fig. 6(a) and (b), respectively. While the dc magnetic susceptibility saturates already below 8 K in $B = 5$ T, the $-\ln T$ divergence in $R(T)$ is unchanged for $B \leqslant 13.5$ T. This fact proves that the applied magnetic field was sufficient to polarize the magnetic moments of impurities and hence to destroy a possible antiferromagnetic interaction of an isolated impurity spin with the surrounding conduction electrons. Therefore, a lack of the response of the $-\ln T$ correction to $B \leqslant 13.5$ T unambiguously excludes the magnetic Kondo effect in ThAsSe.

In spite of the layered PbFCl-type crystal structure of ThAsSe, there is no evidence for a strong anisotropy so far, as suggested by the dc magnetic susceptibility data (cf. Fig.

6). However, even if the quasi-two-dimensionality of the electron gas in ThAsSe would be important, the electron localization and the electron-electron interaction should be ruled out as a source of the logarithmic correction to the resistance for the following reasons.

(i) Since a magnetic field suppresses the localization effect, its magnetotransport response should be significant already in fields much smaller than the ones applied in our experiments.[30,31] Therefore, the slope of the $-\ln T$ singularity should distinctly vary with B, if it was due to quantum interference. This is in striking contrast to the behavior observed for ThAsSe; since its scattering cross section is unchanged by magnetic fields, as mentioned above.

(ii) A very small (0.05% per tesla) and linear decrease of the resistance upon increasing the magnetic field ($B \leqslant 17$ T) cannot be explained either by the quantum interference or by electron-electron interactions. In fact, because these are additive effects, a complex MR(B) behavior is expected. While in small fields the resistance decreases as B^2 due to the electron localization, in the high field limit the interaction effect dominates, leading to a positive magnetoresistance.

(iii) Additional proof of the irrelevance of the electron-electron interaction in ThAsSe is provided by the $R(T)$ results for two different samples presented in Figs. 4 and 5. Though for these two samples, T_S is varying by more than one order of magnitude, $R(T)$ is almost invariant for both strong magnetic field and high hydrostatic pressure. Indeed, the saturation of the resistance upon cooling is in striking disagreement with the theoretical models for electron-electron interaction, which predict a $-\ln T$ dependence also in the limit $T \rightarrow 0$.[30,31]

The $R(T,p)$ results allow to neglect Fermi surface effects as a cause for the low-T singularity in ThAsSe. While applied pressure does not alter the $-\ln T$ behavior, the hump-like anomaly at $T \simeq 65$ K (ambient pressure) is completely suppressed at $p = 1.88$ GPa. The way, in which p influences R_{max}, i.e., by shifting it to lower temperatures accompanied by a uniform reduction of its size, hints at a relation to the electronic structure. Furthermore, the different response to pressure of both singularities, in spite of their similar energy scales, indicates that they are of different origin. In particular, the pressure-independent low-T upturn in $R(T)$ cannot be related to either charge-density wave formation or structural rearrangement.[32] This is supported by the lack of nonlinearities of the voltage-current characteristics, as reported in Ref. 23. We wish to point out that no phase-transition anomaly has been resolved in our specific-heat measurements.

Finally, we mention that for none of the uranium dipnictides (the PbFCl-type structure) has a low-temperature singularity in the resistivity been reported so far, although in all these highly anisotropic antiferromagnets the cylindrical Fermi surfaces have been observed in magnetic quantum oscillations.[33] By contrast, all of these systems display an $R(T) \propto aT^2 (a>0)$ relation as expected for regular antiferromagnets without an anisotropy gap in their magnon spectra. The case of UBi_2 is of particular interest because the Brillouin zone in this system is not altered by the magnetic structure.[33] These same holds apparently true when we compare with UAsSe and ThAsSe. Since the ρ_c/ρ_{ab} ratio ~ 3 for UAsSe is approximately two orders of magnitude smaller than in $UBi_2(\rho_c/\rho_{ab} \sim 500)$, the quasi-two-dimensionality as a cause of the pronounced low-T upturn can be discarded for UAsSe and, obviously also for the nonmagnetic ThAsSe.[34]

As discussed above, a logarithmic correction to the electrical resistance in ThAsSe due to the spin Kondo effect as well as electron localization and electron-electron interaction can be ruled out. Taking into account dynamic disorder highlighted by our low-T heat-capacity results and further suggested by the crystallochemical analysis, the low-temperature resistance in ThAsSe is apparently dominated by the electron-TLS interaction. Though some details of the upturn were found to be strongly sample dependent, for several single crystals of ThAsSe the $-\ln T$ correction found over one decade in temperature appeared to be rather robust. A logarithmic singularity in the electrical resistance due to the structural two-level systems, $R_{TLS}(T)$, was predicted, although this interaction may lead to a rather complex temperature dependence of $R(T)$.[2–4] Remarkably, the TLS Kondo state develops when the Kondo temperature T_K exceeds the TLS splitting Δ_{TLS}. For the Kondo effect with dominant scattering on the TLS with larger splittings, the logarithmic resistance signature transforms into the Fermi-liquid saturation $R_{TLS}(T) \propto 1 - aT^2$ at $T < \Delta_{TLS}$. Most probably, such a case is realized in the ThAsSe single crystal with $RR = 1.54$ (cf. Fig. 4), for which the saturation holds already at $\simeq 2$ K. On the other hand, if $\Delta_{TLS} < T_K$, the saturation will be prevented by an $R_{TLS}(T) \propto 1 - aT^{1/2}$ relation, i.e., a two-channel Kondo effect and the resultant nFL behavior. At present, no signature for $R_{TLS}(T) \propto 1 - aT^{1/2}$ was found. Even though a search for the nFL properties in ThAsSe is outside the scope of this work, we suggest that the square-root temperature dependence may be realized in the specimens with a very low value of T_S. We note that the energy scale of the $-\ln T$ behavior fits well with the other experimental results on the mesoscopic samples ($T_K \sim 3-5$ K),[4,35] although no satisfactory theory exists.[7,8]

V. SUMMARY

In this paper we have discussed the low-temperature properties of the structurally disordered diamagnetic compound ThAsSe, for which anomalous low-lying excitations have been observed. A very small value of the electronic specific heat allowed us to detect a quasilinear-in-T term of nonelectronic origin in its low-T heat capacity. This experimental finding clearly proves the existence of the TLS centers in the ThAsSe single crystals investigated. By applying magnetic fields or hydrostatic pressure, we were able to discriminate between different mechanisms which could lead to a similar variation of the electrical resistance at ambient conditions. We propose that an unusual, low-temperature scattering in ThAsSe, being unchanged by either strong magnetic fields or high hydrostatic pressures, originates in the TLS interacting with conduction electrons. Furthermore, the pronounced $-\ln T$ dependence in $R(T)$ of ThAsSe suggests a macroscopic realization of the Kondo effect derived from structural two-level systems with a characteristic energy scale of the order of a few K for the Kondo temperature.

ThAsSe appears to be highly suited to study the interaction between the TLS and the conduction electrons for the following reasons: (i) the concentration of the dynamical scattering centers can be controlled by the As-Se chemical composition, (ii) its ground state is not affected by low-lying phase transitions, and (iii) high-quality single crystals can be studied, opening the possibility to determine a directional dependence of the electron-assisted hops. Additionally, the comparative experiments on diamagnetic ThAsSe and its ferromagnetic U-based homologue should reflect a dependence of the interaction between the conduction electrons and the TLS on the character of the former ones.

ACKNOWLEDGMENTS

The authors are grateful to A. Zawadowski for valuable discussions, which to a large extent stimulated this work. T. Cichorek wishes to acknowledge the Alexander von Humboldt Foundation for support. Part of this work done in Wroclaw was supported by the Polish Committee for Scientific Research, Grant No. KBN-2 P03B 062 18; 2000–2001.

[1] *Tunneling Systems in Amorphous and Crystalline Solids*, edited by P. Esquinazi (Springer-Verlag, Berlin, 1998).
[2] A. Zawadowski, Phys. Rev. Lett. **45**, 211 (1980).
[3] K. Vladar and A. Zawadowski, Phys. Rev. B **28**, 1564 (1983); **28**, 1582 (1983); **28**, 1596 (1983).
[4] D.L. Cox and A. Zawadowski, Adv. Phys. **47**, 599 (1998).
[5] P.D. Sacramento and P. Schlottmann, Phys. Rev. B **43**, 13 294 (1991).
[6] G. Zaránd, T. Costi, A. Jerez, and N. Andrei, Phys. Rev. B **65**, 134416 (2002).
[7] I.L. Aleiner, B.L. Altshuler, Y.M. Galperin, and T.A. Shutenko, Phys. Rev. Lett. **86**, 2629 (2001).
[8] L. Borda, A. Zawadowski, and G. Zaránd, cond-mat/0302334 (unpublished).
[9] J. Kondo, Physica B **84**, 40 (1976); **84**, 207 (1976).
[10] R.C. Zeller and R.O. Pohl, Phys. Rev. B **4**, 2029 (1971).
[11] D.C. Ralph and R.A. Buhrman, Phys. Rev. B **51**, 3554 (1995).
[12] J. von Delft, D.C. Ralph, R.A. Buhrman, A.W.W. Ludwig, and V. Ambegaokar, Ann. Phys. (N.Y.) **263**, 1 (1998).
[13] R.W. Cochrane, R. Harris, J.O. Strom-Olsen, and M.J. Zuckerman, Phys. Rev. Lett. **35**, 676 (1975).
[14] P. J. Cote and L. V. Meisel, *Metallic Glasses*, edited by H. J. Guntherodt and H. Beck (Springer, New York, 1981), pp. 141–166.
[15] J. L. Black, *Metallic Glasses*, edited by H.J. Güntherodt and H. Beck (Springer, New York, 1981), pp. 167–190.
[16] S. Takano, Y. Kumashiro, and K. Tsuji, J. Phys. Soc. Jpn. **53**, 4309 (1984).
[17] A. Halbritter, O.Yu. Kolesnychenko, G. Mihaly, O.I. Shklyarevskii, and H. van Kempen, Phys. Rev. B **61**, 5846 (2000).
[18] T. Cichorek, Z. Henkie, P. Gegenwart, M. Lang, A. Wojakowski, M. Dischner, and F. Steglich, J. Magn. Magn. Mater. **226–230**, 189 (2001).
[19] Z. Henkie, A. Wojakowski, R. Wawryk, T. Cichorek, and F. Steglich, Acta Phys. Pol. B **34**, 1323 (2003).
[20] Z. Henkie, A. Pietraszko, A. Wojakowski, L. Kępiński, and T. Cichorek, J. Alloys Compd. **317–318**, 52 (2001).
[21] Z. Henkie, R. Fabrowski, A. Wojakowski, and A. Zalewski, J. Magn. Magn. Mater. **140–144**, 1433 (1995).
[22] R. Michalak, T. Cichorek, A. Wojakowski, and Z. Henkie (unpublished); T. Cichorek, R. Michalak, F. Kromer, J. Müller, F. Steglich, A. Wojakowski, and Z. Henkie, Acta Phys. Pol. B **32**, 3399 (2001).
[23] J. Schoenes, W. Bacsa, and F. Hulliger, Solid State Commun. **68**, 287 (1988).
[24] T. Cichorek, E.D. Bauer, A. Wojakowski, Z. Henkie, M.B. Maple, and F. Steglich, Phys. Status Solidi B **236**, 351 (2003).
[25] T. Cichorek, Z. Henkie, A. Wojakowski, A. Pietraszko, P. Gegenwart, M. Lang, and F. Steglich, Solid State Commun. **121**, 647 (2002).
[26] R. Reim, J. Magn. Magn. Mater. **58**, 1 (1986).
[27] F. Hulliger, J. Less-Common Met. **16**, 113 (1968).
[28] F. Pobell, *Matter and Methods at Low Temperatures* (Springer-Verlag, Berlin, 1996).
[29] H. Rudigier, H.R. Ott, and O. Vogt, Phys. Rev. B **32**, 4584 (1985).
[30] B. L. Altshuler and A. G. Aronov, in *Electron-Electron Interactions in Disordered Systems*, edited by A. L. Efros and M. Pollak (North-Holland, Amsterdam, 1985).
[31] P.A. Lee and T.V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985).
[32] Thermally activated hopping should be also ruled out. This is because the applied pressure should change a tiny energy difference between the localized states and hence the magnitude of the hopping term. Besides, in the concept of Anderson localization, a $\ln T$ divergence is not expected but rather a $\ln R(T) \propto T^{-1/\nu}$ behavior with ν varying between 2 and 4.
[33] D. Aoki, P. Wiśniewski, K. Miyake, N. Watanabe, Y. Inada, R. Settai, E. Yamamoto, Y. Haga, and Y. Onuki, Philos. Mag. B **80**, 1517 (2000).
[34] An anisotropy in ThAsSe even smaller than that in UAsSe is inferred form primary measurements of the thermoelectric power. For example while for Th-based system $S_{ab}(300\ \mathrm{K}) = -5.5\ \mu\mathrm{V/K}$ and $S_c(300\ \mathrm{K}) = -3.5\ \mu\mathrm{V/K}$ were found, the corresponding values for the U-based counterpart account for $20\ \mu\mathrm{V/K}$ and $-20\ \mu\mathrm{V/K}$.
[35] F.G. Aliev, V.V. Moshchalkov, and Y. Bruynseraede, Phys. Rev. B **58**, 3625 (1998).