Thermal transport signatures of the excitonic transition and associated phonon softening in the layered chalcogenide Ta₂NiSe₅

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The quasi-one-dimensional layered compound Ta₂NiSe₅ has been proposed to undergo a transition to an excitonic insulator at $T_c = 326$ K. We found a clear anomaly at T_c in the in-plane thermal conductivities both parallel ($\parallel a$) and perpendicular ($\parallel c$) to the one-dimensional chains, κ_a and κ_c . While κ_a shows a rapid decrease below T_c , κ_c shows a pronounced V-shaped suppression centered at T_c . We argue that the decrease of κ_a represents the suppression of the quasiparticle contribution below T_c due to the excitonic transition. On the other hand, the V-shaped suppression of κ_c comes from the enhanced phonon scattering by soft phonons associated with the monoclinic transition with momentum $\mathbf{q} \parallel c$. The continued suppression of κ_c up to an extremely high temperature above T_c suggests the persistence of phonon softening likely coupled to electronic, presumably excitonic, fluctuations.

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The excitonic insulator (EI), in which bound electron-hole pairs form a condensate below a transition temperature T_c , had been proposed theoretically more than 50 years ago [1], but its realization in existing materials remains an active subject of investigation. The excitonic transition is anticipated in a nearly zero-gap semiconductor when the exciton binding energy E_b exceeds the one-electron gap E_g , or in a semimetal when the attractive interaction between electrons and holes is not screened out well. The realization in bulk materials has been discussed, for example, in TmSe_{0.45}Te_{0.55} [2] and 1T-TiSe₂ [3], but the evidence for excitonic condensation in these compounds has been far from convincing. The former is a $4f^{13}$ Mott-like system, not a band semiconductor/semimetal [4], and the latter remains metallic and may suffer from strong screening of electron-hole interactions even below the proposed excitonic transition temperature [5].

Very recently, Ta₂NiSe₅ has emerged as a leading candidate for an EI [6–18], which is a direct gap semiconductor/semimetal with Ta *5d* conduction bands and Ni *3d* valence bands [8,15,19]. It shows a transition to an insulator at $T_c = 326$ K, which was proposed to be an excitonic transition. The crystal structure of Ta₂NiSe₅ is layered, and as shown in Fig. 1(a), each layer consists of an array of Ta and Ni chains running along the *a* axis, which renders the system electronically quasi-one-dimensional and gives rise to

spatially isolated electron and hole chains. The one dimensionality is known to enhance the exciton binding energy [7], which makes Ta₂NiSe₅ an ideal platform for the search for an EI. Well below T_c , an optical gap of 0.16 eV develops, which is close to the exciton binding energy of 0.25 eV experimentally observed in the optical conductivity spectrum of the sister compound Ta₂NiS₅. Ta₂NiS₅ has a much larger gap of 0.6 eV and exhibits no clear signatures of an electronic phase transition in electrical transport and optical conductivity [8,9]. These observations strongly suggest the excitonic origin of the insulating gap in Ta₂NiSe₅ below T_c . As a function of the one-electron gap, E_g , controlled by pressure and chemical substitution, the transition temperature T_c shows a dome-shaped behavior centered at $E_g \approx 0$, corresponding to pure Ta₂NiSe₅ at ambient pressure [8]. This is consistent with what is expected for the canonical EI [1].

The aforementioned experimental results clearly point to the excitonic character of the transition. Nevertheless, it has become increasingly clear that the transition cannot be purely excitonic and that the actual situation is more complicated due to the electron-lattice coupling [19–21], which makes the physics of the putative excitonic transition even more challenging and attractive. The putative excitonic transition at $T_c = 326$ K is accompanied by a structural phase transition from an orthorhombic to a monoclinic structure [22,23]. In the monoclinic phase, the TaSe₂ chain-ladder unit distorts, such that the Ta chain and the surrounding Se ladders move along the *a* axis, but in opposite directions [24]. The movement of two neighboring TaSe₂ chain-ladder units surrounding the Ni chain is asymmetric with respect to the Ni chain. This shear-type distortion with antiferroelectric-type displacement of the Ta and the Se chains does not bring about any superstructure (i.e., $\mathbf{q} = 0$) but allows for the hybridization of Ta conduction bands and Ni valence bands around the Γ point [19]. The resultant hybridization gap could account for

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FIG. 1. (a) Crystal structure of Ta₂NiSe₅ showing the stack of layers along the *b* axis (left) and a single layer in the *a*-*c* plane consisting of an array of two chains of Ta octahedrally coordinated with Se and one chain of Ni in between, tetrahedrally coordinated with Se (right) [33]. (b) Thermal conductivities κ_a with heat current parallel to the chains ($\parallel a$; red) and κ_c with heat current perpendicular to the chains ($\parallel c$; blue) as a function of temperature. The dashed black line indicates $T_c = 326$ K. Inset: Enlarged view of κ_a and κ_c near T_c .

at least a part of the insulating gap and should be one of the important ingredients of the transition at 326 K. A symmetry analysis demonstrated that a finite excitonic order parameter must be accompanied by the monoclinic distortion and hence the admixture of the excitonic gap and the hybridization gap is unavoidable [25].

In this study, we measure the in-plane anisotropic thermal conductivities $\kappa_a(T)$ along the chains ($\parallel a$) and $\kappa_c(T)$ perpendicular to the chains ($\parallel c$) of Ta₂NiSe₅ single crystals in order to unveil the quasiparticle thermal conductivity of the putative EI and the phonon transport around the monoclinic transition. In EIs, quasiparticle transport analogous to Bardeen-Cooper-Schrieffer superconductors may be expected [1,26]. Thermal conductivity is an especially useful probe of the quasiparticle transport in an EI as the cancellation between the electron and the hole quasiparticles does not take place. In addition, thermal conductivity has a large phonon contribution, which is sensitive to the presence of soft phonons through phonon-

phonon scatterings [27,28] and therefore may give us hints to decipher the nature of the monoclinic lattice distortion.

We show that $\kappa_a(T)$ along the chains is rapidly suppressed below T_c and that it comes from the decrease in the number of quasiparticles (electrons and holes) below T_c , which is expected for a canonical EI [29]. In contrast, κ_c perpendicular to the chains shows a V-shaped suppression due to enhanced scattering by soft phonons associated with the orthorhombic-to-monoclinic transition with momentum $\mathbf{q}$ perpendicular to the chains ($\parallel c$). The strong suppression of κ_c persists up to more than 50 K above T_c , which suggests a robust phonon softening likely coupled to excitonic (and hybridization-gap) fluctuations.

Needle-shaped single crystals of Ta₂NiSe₅ with a typical size of 4 mm $\times$ 1 mm $\times$ 0.1 mm were grown by chemical vapor transport [8,30], with their longest dimension along the Ta/Ni chains (*a* axis) and their shortest dimension along the interlayer *b* axis. Rectangular-shaped samples with their long axis along the *a* and *c* axes were cut out from the single crystals. The electrical resistivity of the samples was measured by a conventional four-probe technique, while thermal conductivity measurements along the same axes were performed via a “heat-pipe” method [31,32]. The temperature gradients were detected with a differential type-E thermocouple. A transition was observed at $T_c = 326$ K as a clear kink in the electrical resistivity along the chain direction (ρ_a), as reported previously [8,30].

As shown in Fig. 1(b), $\kappa_a(T)$ along the chains ($\parallel a$) is a factor of 3–4 larger than κ_c perpendicular to the chains ($\parallel c$). With increasing temperature from 10 K, both κ_a and κ_c increase and show a peak around 30–40 K, followed by a gradual decrease that continues above room temperature. This behavior is common for a crystalline solid with phonon-dominated thermal conductivity [34,35].

At the transition temperature $T_c = 326$ K, we observe a clear anomaly both in $\kappa_a(T)$ and $\kappa_c(T)$. The anomaly at T_c , however, shows up in a contrasted manner between the two directions *a* and *c*, which can be seen more clearly in the enlarged plot around T_c in the inset of Fig. 1(b). For $\kappa_a(T)$ parallel to the chains, a shoulder structure with a well-defined kink at T_c is observed, followed by an abrupt drop below T_c . On further lowering of the temperature, $\kappa_a(T)$ recovers and begins increasing below ~ 315 K. In contrast, $\kappa_c(T)$ shows a discontinuous change of slope at T_c from a weak increase to a very rapid increase with decreasing temperature, which can be viewed as a V-shaped suppression from the $1/T$ -like increase expected for phonon-dominant, high-temperature thermal transport [34]. The contrast is even clearer in the temperature derivatives, $\partial\kappa_a/\partial T$ and $\partial\kappa_c/\partial T$ [30]. We argue that the contrasted behaviors of the anomaly at T_c between $\kappa_a(T)$ and $\kappa_c(T)$ come from the predominant electronic and phonon contributions in the temperature dependence of $\kappa_a(T)$ and $\kappa_c(T)$, respectively, near T_c , which allows us to discuss the behaviors of quasiparticles (electrons and holes) and phonons around T_c separately.

The measured thermal conductivity κ comprises contributions from electrons, $\kappa^{(e)}$, and from phonons, $\kappa^{(ph)}$. Around T_c , $\kappa^{(e)}$ can be estimated from the electrical conductivity σ using the Wiedemann-Franz (W-F) law. The W-F law is a realistic approximation around T_c , as large-angle electron-phonon

FIG. 2. Separation of electron and phonon contributions to the thermal conductivities κ_a with heat current parallel to the chains ($\parallel a$; red) and κ_c with heat current perpendicular to the chains ($\parallel c$; blue) in Fig. 1, using the W-F law. (a) The temperature-dependent electrical conductivities, $\sigma_a(T)$ (red) parallel to the chains and $\sigma_c(T)$ (blue) perpendicular to the chains. (b) The total, electron, and phonon thermal conductivities along the a axis, parallel to the chains, are denoted by κ_a (circles), $\kappa_a^{(e)}$ (broken), and $\kappa_a^{(ph)}$ (solid), respectively. (c) The total, electron, and phonon thermal conductivities along the c axis, perpendicular to the chains, are denoted by κ_c (circles), $\kappa_c^{(e)}$ (broken), and $\kappa_c^{(ph)}$ (solid), respectively.

scattering dominates at high temperature [34,35], especially in such a low carrier density system. Then $\kappa^{(ph)}$ can be estimated as the remnant $\kappa - \kappa^{(e)}$. As shown in Fig. 2(a), the electrical conductivity along the chains, σ_a , is an order of magnitude larger than that perpendicular to the chains, σ_c , reflecting the quasi-one-dimensional band structure of Ta_2NiSe_5 . In addition, the anomaly at T_c is much more pronounced in $\sigma_a(T)$ as compared with $\sigma_c(T)$. A much larger electronic contribution to the temperature dependence of $\kappa_a(T)$ than $\kappa_c(T)$ is thus expected.

As shown in Fig. 2(b), the electronic thermal conductivity along the chains $\kappa_a^{(e)}(T)$, estimated from σ_a , is only $\sim 5\%$ of the total $\kappa_a(T)$, but fully accounts for the shoulder structure in $\kappa_a(T)$ with the rapid decrease below T_c . On the other hand, the phonon thermal conductivity $\kappa_a^{(ph)}(T)$, estimated by subtracting $\kappa_a^{(e)}(T)$ from $\kappa_a(T)$, is surprisingly featureless and shows a monotonic increase through T_c , indicating that the

anomaly at T_c in $\kappa_a(T)$ is purely electronic in origin. The rapid decrease below T_c therefore reflects the decrease in the number of quasiparticles (electrons and holes), analogous to the prototypical behavior of thermal conductivity in a superconductor [36,37]. We therefore conclude that the quasiparticle transport of a putative EI is captured by thermal transport.

In superconductors, the thermal conductivity sometimes does not show a rapid decrease right below T_c , the prototypical behavior, but instead shows an increase [38,39]. This can be ascribed to the suppression of scattering of electrons and phonons by electrons due to the opening of an electronic gap below T_c . In certain strongly correlated superconductors with anisotropic pairing, such as high- T_c cuprates [38], the enhancement of quasiparticle lifetime due to the suppression of strong electron-electron scattering overcomes the decrease of quasiparticle density and thereby results in an increase of κ right below T_c . In certain conventional electron-phonon superconductors, the increase of phonon mean-free path due to the suppression of strong scattering of phonons by electrons may also result in the enhancement of phonon thermal conductivity below T_c , which brings about the increase of κ right below T_c [39]. The observation of a rapid decrease in $\kappa_a(T)$ below T_c in Ta_2NiSe_5 thus indicates that the scattering of phonons and electrons with momentum parallel to the chains by electrons should be reasonably weak as compared with other dominant scattering mechanisms, such as phonon-phonon scattering for phonon transport.

As σ_c is one order of magnitude smaller and exhibits a much weaker structure at T_c than σ_a , the electronic thermal conductivity $\kappa_c^{(e)}(T)$ perpendicular to the chain direction is as small as $\sim 1\%$ of the total $\kappa_c(T)$ in magnitude and, more importantly, does not contribute appreciably to the kinklike anomaly at T_c in the temperature dependence of $\kappa_c(T)$, as seen in Fig. 2(c). In contrast to $\kappa_a(T)$ along the chains, $\kappa_c(T)$ around T_c is dominated by $\kappa_c^{(ph)}(T)$ and is almost purely phononic in origin. The comparison of $\kappa_c^{(ph)}(T)$ and $\kappa_a^{(ph)}(T)$ reveals a substantial anisotropy not only in electron transport but also in phonon transport between the directions parallel and perpendicular to the chains. As clearly seen in Fig. 3, while a $1/T$ -like monotonous decrease with featureless behavior around T_c is observed in $\kappa_a^{(ph)}(T)$ ($\parallel$ chain), a clear V-shaped suppression from the $1/T$ -like decrease centered at T_c is seen in $\kappa_c^{(ph)}(T)$ ($\perp$ chain). The suppression is broad in temperature range and appears to be asymmetric above and below T_c . While $\kappa_c^{(ph)}(T)$ shows a rapid increase and crossover to $1/T$ -like behavior with decreasing temperatures right below T_c , it is almost flat up to at least 400 K with increasing temperature from T_c , implying the persistence of strong suppression of out-of-chain phonon conduction over a wide temperature range above T_c .

Phonon thermal conductivity is generally expressed as $\kappa^{(ph)} = \frac{1}{3} C^{(ph)} v^{(ph)} l^{(ph)}$, where $C^{(ph)}$, $v^{(ph)}$, and $l^{(ph)}$ are the specific heat, the velocity, and the mean-free path of phonons [34], and specifically for Ta_2NiSe_5 many acoustic and optical phonon branches are expected to contribute to $\kappa^{(ph)}$ around T_c [20,24]. The temperature dependence of the total heat capacity C for Ta_2NiSe_5 around $T_c = 326$ K is rather small and the small anomaly at T_c is highly likely to be purely electronic in origin [8]. The temperature dependence

FIG. 3. Comparison of temperature-dependent thermal conductivities $\kappa(T)$ for Ta_2NiSe_5 ($\parallel a$ red; $\parallel c$ blue) with typical order-disorder-type ferroelectrics, NH_4Cl (reproduced from [40]), and KH_2PO_4 (reproduced from [41]). The solid black line is a guide line for the $1/T$ power law.

of $C^{(\text{ph})}$ can thus be ignored in discussing the origin of the V-shaped suppression of $\kappa_c^{(\text{ph})}(T)$. The suppression of $\kappa_c^{(\text{ph})}(T)$ should originate instead from the reduction of $v^{(\text{ph})}$ and/or $l^{(\text{ph})}$ around T_c . As it occurs only for phonons with momentum $\mathbf{q} \parallel c$ and phonon scattering should dominate $\kappa_c^{(\text{ph})}(T)$ at high temperatures around T_c , it is natural to ascribe its origin to the reported softening of the acoustic shear-mode phonon with $\mathbf{q} \parallel c$, which corresponds to the monoclinic distortion of lattice frozen at the transition $T_c = 326$ K [24]. As pronounced softening occurs only for one specific acoustic phonon mode, the reduction of $v^{(\text{ph})}$ is unlikely to account for the large suppression of $\kappa_c^{(\text{ph})}(T)$ at T_c [Fig. 2(c)]. The primary origin of the suppression of $\kappa_c^{(\text{ph})}(T)$ is highly likely the increased scattering of phonons, namely, the reduction of $l^{(\text{ph})}$ by the soft acoustic shear-mode phonon. The strong suppression above T_c up to at least 400 K should then imply the robust softening of the shear mode acoustic phonon above T_c , which is indeed quite consistent with the reported phonon dispersion relationship of Ta_2NiSe_5 measured by inelastic x-ray scattering up to 400 K [24].

We should point out that the asymmetric V-shaped suppression of $\kappa_c^{(\text{ph})}(T)$, more robust above T_c , is analogous to those observed around the order-disorder-type ferroelectric transition in KH_2PO_4 and NH_4Cl , as shown in Fig. 3. The order-disorder-type ferroelectrics have extended ferroelectric fluctuations above T_c , where well-developed but fluctuating local dipoles give rise to soft local phonons and enhanced phonon scattering over a wide temperature range and hence suppress $\kappa(T)$ strongly over a wide temperature range above T_c [28,40–42]. It is tempting to infer that, analogous to the order-disorder-type ferroelectrics, large and local excitonic and/or hybridization gap ($\mathbf{q} = 0$ charge density wave) fluctuations may be present above T_c in Ta_2NiSe_5 . The electronic fluctuations are inherently and strongly coupled to the shear mode phonon, which may give rise to the extended phonon softening above T_c and may suppress $\kappa_c^{(\text{ph})}(T)$ over a wide temperature range. Such giant excitonic and/or hybridization gap fluctuations may be inferred from the temperature dependence of the bulk magnetic susceptibility $\chi(T)$ of Ta_2NiSe_5 [22], where $\chi(T)$ at temperatures well above T_c shows a pronounced downturn on lower temperatures toward T_c , indicative of such fluctuations.

In summary, we have discovered an extremely anisotropic behavior of the thermal conductivity around the putative excitonic transition in the layered chalcogenide Ta_2NiSe_5 . The distinct temperature-dependent in-plane $\kappa_a(T)$ and $\kappa_c(T)$, parallel and perpendicular to the Ta and Ni chains, respectively, allow us to unveil the behaviors of quasiparticles (electrons and holes) and phonons around the transition temperature $T_c = 326$ K. The rapid suppression in the number of quasiparticles below T_c , analogous to superconductors, is captured in $\kappa_a(T)$ due to the quasi-one-dimensional electronic structure and the $\mathbf{q} \parallel a$ phonons being insensitive to the transition. The soft shear-mode acoustic phonons with momentum $\mathbf{q} \parallel c$, which correspond to the monoclinic structural distortion below T_c , manifest themselves only in $\kappa_c(T)$ perpendicular to the chains as a pronounced suppression of dominant phonon thermal transport. The suppression of κ_c extends up to high temperatures well above T_c , indicating the persistence of phonon softening over a wide temperature range. This may suggest the presence of giant excitonic and/or hybridization gap fluctuations above T_c and the strong electron-lattice coupling for the monoclinic distortion in Ta_2NiSe_5 .

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