Letter | Editors' Suggestion

Temperature dependence of the energy band gap in $ZrTe_5$: Implications for the topological phase

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Using Landau-level spectroscopy, we determine the temperature dependence of the energy band gap in zirconium pentatelluride ($ZrTe_5$). We find that the band gap reaches $E_g = (5 \pm 1)$ meV at low temperatures and increases monotonically when the temperature is raised. This implies that $ZrTe_5$ is a weak topological insulator, with noninverted ordering of electronic bands in the center of the Brillouin zone. Our magnetotransport experiments performed in parallel show that the resistivity anomaly in $ZrTe_5$ is not connected with the temperature dependence of the band gap.

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Zirconium pentatelluride ($ZrTe_5$) is a narrow-gap system widely explored in the past [1–4], often in the context of so-called resistivity anomaly [5–10]. More recently, theoretical studies of $ZrTe_5$ that were primarily focused on topological properties induced a renewed interest in this system. In particular, the $ZrTe_5$ monolayer was proposed to be a large-gap quantum spin Hall insulator [11]. Over the past few years, a number of experiments have been performed on bulk and thin layers of $ZrTe_5$ and have expanded our knowledge about this material considerably. The experimentally observed phenomena in $ZrTe_5$ comprise the magnetochiral effect [12,13], the giant planar Hall effect [14], the quasiquantized quantum Hall effect [15,16], pressure-induced superconductivity [17], or optical conductivity that is linear in photon energy [18,19].

This series of important experimental observations contrasts with our current understanding of low-energy excitations in $ZrTe_5$. In fact, no consensus has so far been established whether the electronic bands in bulk $ZrTe_5$ have normal or inverted ordering, i.e., whether this material is a strong or weak topological insulator (STI or WTI). *Ab initio* calculations favor the STI phase [20], but their prediction force may be limited in a system close to the topological phase transition. Experimentally, both STI and WTI phases have been reported; see Refs. [13,21–25] and [26–30], respectively, based on transport, ARPES, and optical data.

In this Letter, we explore, using THz/infrared magnetospectroscopy, the temperature dependence of the band gap in $ZrTe_5$ to test the topological phase of this material. We find that the gap is nonzero at low temperatures, $E_g = (5 \pm 1)$ meV at $T = 5$ K, and increases with T monotonically. Such behavior is consistent with $ZrTe_5$ being a WTI.

Our experiments were performed on bulk $ZrTe_5$, synthesized using self-flux growth [12]. The explored sample was attached using GE varnish to a metallic holder that was covered by a thin paper sheet and surrounded by the helium exchange gas. The temperature was controlled locally, using a heating coil, and measured by two Cernox thermometers placed at two different locations nearby the sample. A relatively large thickness of the sample (≈0.5 mm) was chosen to avoid effects related to the temperature-induced strain. Results obtained on this sample, presented here, were reproduced on several other specimens with different thicknesses and glued in different configurations.

We measured relative magnetoreflectivity, R_B/R_0, in the Faraday configuration, i.e., with the wave vector of light propagating parallel to the magnetic field aligned with the b crystallographic axis. The radiation of a globar was analyzed by the Bruker Vertex 80v Fourier-transform spectrometer and delivered via light-pipe optics to the sample located in a superconducting coil. A part of the reflected signal was deflected toward an external bolometer using a silicon beamsplitter. In addition, magnetotransport data were collected *in situ*, i.e., in parallel with magneto-optical measurements. To this end, electric contacts were created using graphite paste in a standard four-contact configuration.

An illustration of the collected magnetoreflectivity data is presented in Fig. 1. It comprises relative magnetoreflectivity, R_B/R_0, measured at $T = 80$ K, plotted (a) in the form of a stacked plot of spectra taken at selected values of B and (b) as a false-color map. The observed response is in line with preceding magneto-optical studies of $ZrTe_5$ [19,22,31,32]. It closely resembles the magneto-optical response of Landau-quantized electrons in a conical band [33]; nevertheless, the characteristic $\sqrt{B}$ dependence of inter-LL excitations is here only approximate. The observed transitions extrapolate to a nonzero energy in the limit of vanishing B, thus indicating a band gap in $ZrTe_5$.

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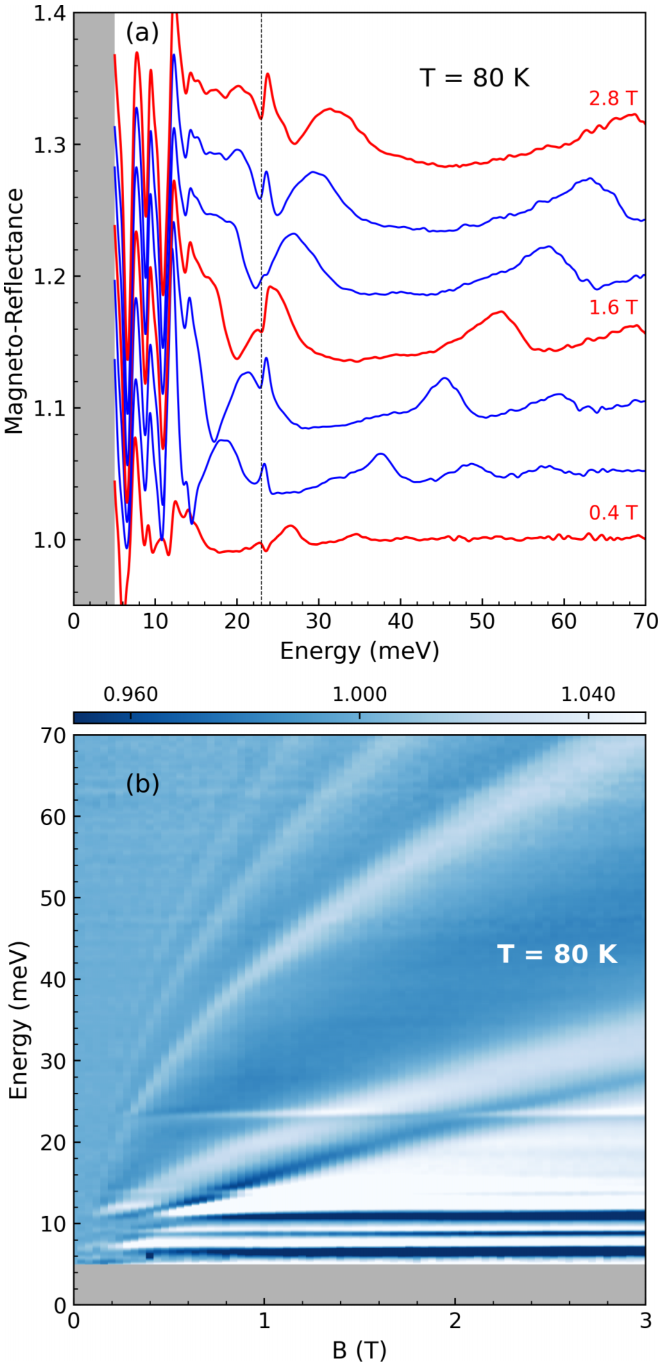

FIG. 1. (a) Stacked plot of relative magnetoreflectivity spectra, $R_B(\omega)/R_0(\omega)$, collected on $ZrTe_5$ at $T = 80$ K and plotted for selected values of B. (b) The false-color plot of R_B/R_0 plotted with a step of 40 mT. Weakly B-dependent features, nearly horizontal in the plot, are phonons, accompanied at low energies (below 10 meV) by an interference pattern. This appears since $ZrTe_5$ becomes transparent at low energies when a sufficiently strong magnetic field is applied. The vertical line in (a) at 23 meV shows the position of the most pronounced phonon mode in $ZrTe_5$ [18].

To extract the band gap, we have plotted and examined the first field-derivative of the collected spectra, $d/dB[R_B/R_0]$ [Figs. 2(a)–2(e)] and associated the corresponding maxima with energies of individual inter-LL resonances. This approach is applicable for absorption lines on a positive dielectric background. When additional dissipative processes appear in the background, the position of the resonance may be shifted from the inflection point of $R_B(\omega)/R_0(\omega)$ toward the maximum. This uncertainty increases the error bar of the below extracted/discussed values of the velocity parameter, but has a minor influence on the width of the band gap—the main target of our analysis.

To understand the observed response in the simplest possible manner, we neglect the relatively flat dispersion of electrons along the magnetic field (b axis in our case), see Ref. [19] and interpret our data in terms of excitations in the spectrum of two-dimensional Landau-quantized massive Dirac electrons: $E_n^{\pm} = \pm\sqrt{\Delta^2 + v^2 2e\hbar nB}$ where $n = 0, 1, 2 \ldots$. Here v stands for the velocity parameter averaged in the plane perpendicular to $\mathbf{B}$ (a-c plane) and $+(-)$ refers to the conduction (valence) band. The band gap corresponds to the value of $E_g = 2\Delta$.

The conventional selection rules for electric-dipole transitions, $n \rightarrow n \pm 1$, imply the existence of a series of interband inter-LL excitations: $E^+_{n+1(n)} - E^-_{n(n+1)} = \sqrt{\Delta^2 + v^2 2e\hbar Bn} + \sqrt{\Delta^2 + v^2 2e\hbar B(n+1)}$. In our data, such transitions are visible for $n = 0, 1, 2, 3$, and 4. In addition, cyclotron resonance (CR) absorption appears. In a weakly electron-doped sample, only the fundamental CR mode of electrons ($0^+ \rightarrow 1^+$) is active at low temperatures, at the energy of $\sqrt{\Delta^2 + v^2 2e\hbar B} - \Delta$. At elevated temperatures, the fundamental CR mode of holes ($1^- \rightarrow 0^-$) emerges as well, at the same photon energy. In contrast to a preceding study [34], we did not observe any series of excitations that would indicate another gap. The outlined massive Dirac model was used to fit the low-B part of the data (below 1 T); see Figs. 2(f)–2(j). This is due to Zeeman effect, which scales linearly with B and which starts to impact visibly the magneto-optical response of $ZrTe_5$ at higher magnetic fields; see Ref. [22,32].

Despite the apparent simplicity of the massive Dirac model, all observed transitions can be well-reproduced in the chosen range of low magnetic fields and also for all explored values of T [see Figs. 2(f)–2(j)]. Certain deviations appear in the region when transitions approach phonon lines [horizontal dotted lines in Figs. 2(f)–2(j)] which are excluded from the fitting procedure. This allows us to extract the temperature dependence of v and Δ which are the only adjustable parameters of the used model [see Figs. 2(k) and 2(l)]. The extracted values evolve smoothly and monotonically with temperature. The velocity parameter decreases slightly, which likely reflects only the unit cell increasing in size. This in turn implies a decrease in the overlap of atomic functions and, consequently, overall flattening of the band structure. The band gap gradually grows from the low-temperature value, $2\Delta = (5 \pm 1)$ meV, which is consistent with the preceding magneto-optical studies [19,22,32].

Interestingly, having established that the massive Dirac model is valid, one may infer the width of the band gap 2Δ, as well as its increase with T, directly from the data in Figs. 2(a)–2(e) without any fitting procedure. It simply corresponds to the separation between the lowest interband line ($0^- \rightarrow 1^+$ and $1^- \rightarrow 0^+$) and the fundamental CR mode ($0^+ \rightarrow 1^+$ and $1^- \rightarrow 0^-$). Other methods, presumably with

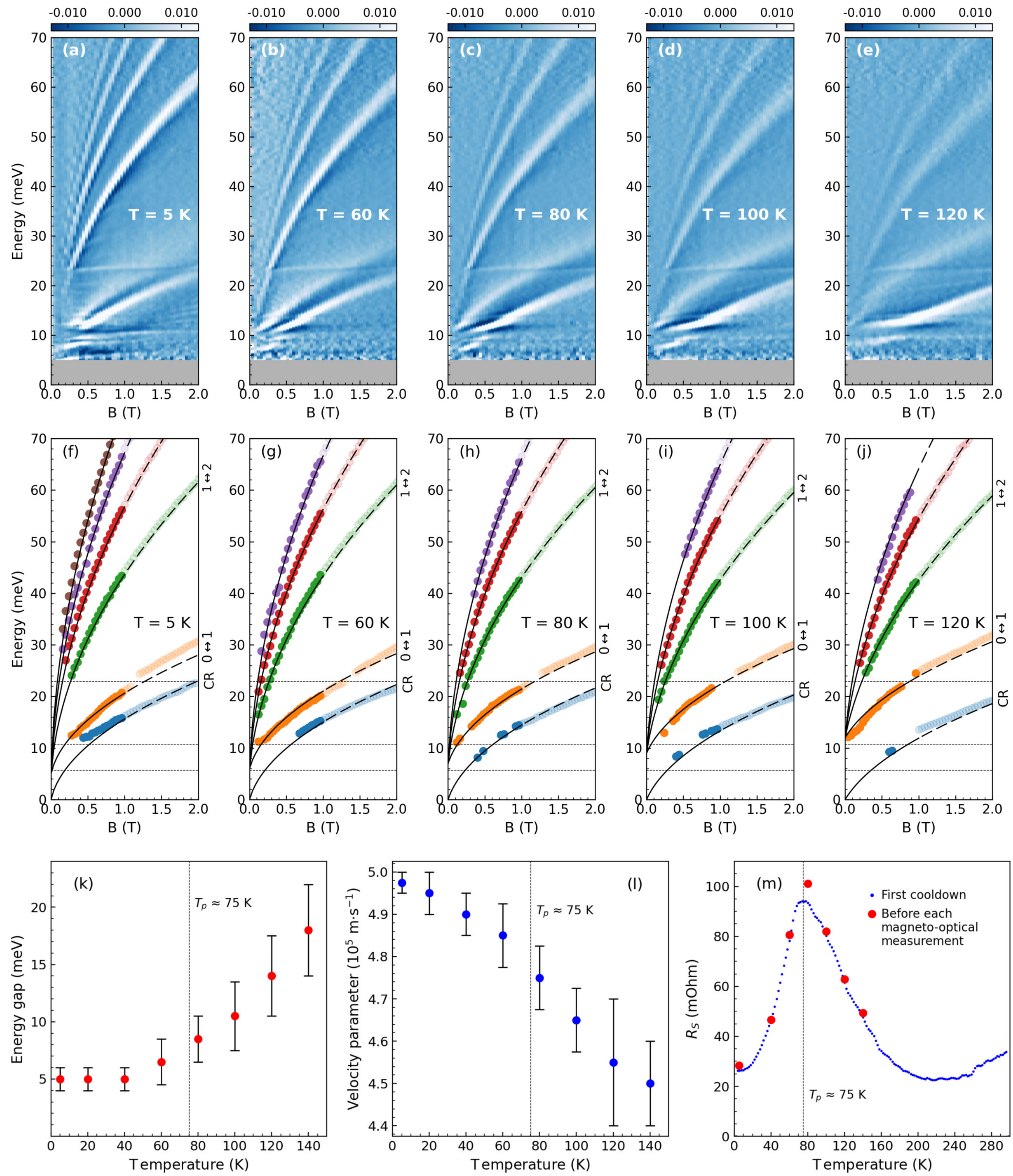

FIG. 2. (a)–(e) False-color plots of the first field-derivative of relative magnetoreflectivity, $d/dB[R_B/R_0]$, measured at the temperature of $T = 5, 60, 80, 100$ and 120 K. (f)–(j) The extracted positions of resonances at a given magnetic field and temperature. The solid lines represent the fits of interband inter-LL transitions for the data collected below 1 T. The dashed lines represent extrapolations of these fits to higher magnetic fields and energies. Dotted horizontal lines show energies of the three most pronounced phonon modes (at 6, 11, and 23 meV) observed in Refs. [18,19]. (k) The extracted energy band gap of $ZrTe_5$ as a function of temperature. (l) The experimentally determined temperature dependence of the effective velocity parameter. (m) The resistance anomaly in the explored $ZrTe_5$ sample, with the maximum of $R_S(T)$ around $T_p \approx 75$ K. The blue dots show the sample resistance during the first cooldown, the red circles before each run of the magneto-optical measurements at a given T. The temperature corresponding to the maximum in resistivity is marked by the vertical dashed lines in (k)–(m).

a smaller resolution in energy, such as ARPES or STM/STS techniques, provide us with the band gap larger by a factor of 3 or more [27–29,35].

With increasing T, and above $T = 100$ K, in particular, the observed interband inter-LL transitions visibly weaken, see Figs. 2(a)–2(e) This is partly due to thermally induced broadening of LLs, partly when k_BT exceeds Δ, due to charge carriers excited thermally to LLs with higher indices. The latter reduces the transition strengths via the corresponding occupation factor. At low energies, the optical response also becomes modified by thermally excited carriers. With increasing T, the plasma edge forms gradually [19] and the simple one-electron picture is then no longer applicable. Instead, the observed CR mode starts to resemble, see Fig. 2(e), the upper CR branch in the classical magnetoplasma response [36]: $\hbar(\omega_c^2 + \omega_p^2)^{1/2}$, where ω_p and ω_c are the screened plasma frequency and bare single-electron cyclotron frequency, respectively.

Let us now discuss implications of our experimental findings. Theoretical studies [11,20,37] suggest that bulk $ZrTe_5$ is a system close to a topological phase transition, which may, in fact, be the reason why no consensus about the topological nature of $ZrTe_5$ has so far been achieved. Theoretically, only a relatively small increase in the unit-cell volume is sufficient to bring $ZrTe_5$ from the STI to WTI regime, i.e., from the inverted to the normal ordering of bands at the center of the Brillouin zone. Importantly, the band gap always closes and reopens during such a topological phase transition.

Experimentally, it was shown that the unit-cell volume in $ZrTe_5$ increases monotonically with temperature [2]. Thanks to this, the temperature dependence of the band gap was proposed [20,37] to be an unambiguous signature of the topological phase in $ZrTe_5$. In the STI regime, the band gap is first supposed to shrink with increasing T, then close completely, and finally reopen in the WTI regime. In contrast, a monotonic increase of the band gap with temperature is expected in the WTI regime. Only the latter scenario is consistent with our data [Fig. 2(g)], thus implying the WTI phase in $ZrTe_5$. Let us note that the volume of the unit cell changes considerably in the explored range of temperatures, approximately by 0.5% [2].

Additional magnetotransport measurements, performed along with the magnetoreflectivity experiments, allowed us to correlate the evolution of the band gap width with the well-known resistivity anomaly in $ZrTe_5$. This effect refers to a characteristic peak in resistivity appearing when the temperature increases, see, e.g., Refs. [5–10]. The temperature dependence of resistance of the explored sample measured during the first cooldown of the sample, $R_S(T)$, is plotted in Fig. 2(m) along with R_S values measured prior to the sweeps of our magnetoreflectivity measurements at a given fixed T. We speculate that the small discrepancies between the resistance values in the subsequent experiments, particularly visible at the peak value, could be due to a thermal-cycle-induced change in the contact resistance. This would modify the sourcing current, assumed to be constant while extracting resistance value. The observed profile, reaching maximum around $T_p \approx 75$ K, is perfectly in line with preceding experiments on samples with similar doping. In literature [24,38], the appearance of resistivity anomaly in $ZrTe_5$ was sometimes associated with the temperature-driven closure of the band gap around T_p. Our data do not support such a conclusion.

In summary, we have extracted the temperature dependence of the band gap in $ZrTe_5$ using Landau-level spectroscopy. We have found that the band gap increases monotonically from the low-temperature value of $E_g = (5 \pm 1)$ meV to nearly 20 meV at $T = 140$ K. The observed behavior is consistent with $ZrTe_5$ being a WTI.

Note added. Recently, we learned about another magneto-optical study of $ZrTe_5$ [39] presenting similar experimental results but suggesting a different interpretation.

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