Surface State-Dominated Photoconduction and THz Generation in Topological Bi₂Te₂Se Nanowires

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ABSTRACT: Topological insulators constitute a fascinating class of quantum materials with nontrivial, gapless states on the surface and insulating bulk states. By revealing the optoelectronic dynamics in the whole range from femto- to microseconds, we demonstrate that the long surface lifetime of Bi₂Te₂Se nanowires allows us to access the surface states by a pulsed photoconduction scheme and that there is a prevailing bolometric response of the surface states. The interplay of the surface and bulk states dynamics on different time scales gives rise to a surprising physical property of Bi₂Te₂Se nanowires: their pulsed photoconductance changes polarity as a function of laser power. Moreover, we show that single Bi₂Te₂Se nanowires can be used as THz generators for on-chip high-frequency circuits at room temperature. Our results open the avenue for single Bi₂Te₂Se nanowires as active modules in optoelectronic high-frequency and THz circuits.

KEYWORDS: Topological insulators, surface state dynamics, ultrafast photodetector, on-chip THz generation

Bismuth and antimony chalcogenides as well as their alloys are among the most intensely studied three-dimensional (3D) topological insulators (TIs).1–9 Their surface states show remarkable properties such as a helical Dirac dispersion and the suppression of backscattering of spin-polarized charge carriers. It is equally critical and difficult to address the surface states independently of the bulk states and to exploit them for electronic and optoelectronic applications. Recent experimental studies have shown that the surface currents can be optically accessed and read-out by a photocurrent measurement within the lifetime of the surface states.10,11 According to theory, the helicity of the exciting photons can drive in-plane currents within the spin-relaxation lifetime of the materials.12,13 The possibility to induce such polarization-driven currents has been confirmed for different 3D TIs like Bi₂Se₃,10,11 Sb₂Te₃,14 and BiSbTe.15 In this context, tuning of the helicity-dependent photocurrents in Bi₂Se₃ has been accomplished through changing the carrier concentration by electrostatic doping.16 It is noteworthy that, depending on the specific 3D TI compound and the underlying substrate,17 the generation of photothermoelectric currents can be the dominant effect.18,19 This may provide access to high performance photothermal-electric devices which exploit the acoustic phonon-mediated cooling of the Dirac fermions in a TI.20 Interestingly, circularly polarized light also allows for controlling exotic topological quantum phases such as transient Floquet–Bloch states.17

For most 3D TIs, the surface lifetime is limited to only a few picoseconds. Recent photoemission experiments on Bi₄Te₅Se (BTS), however, demonstrate that its surface states exhibit a long lifetime even up to microseconds, while its bulk states are relatively short-lived with a lifetime in the range of several tens of picoseconds.21 This difference has been attributed to a charge drift caused by a surface photovoltage. Moreover, BTS as a compensated TI has a relatively low bulk carrier concentration, thus favoring charge transport through its topological surface states.22 Such a property has facilitated the detection of spin-polarized currents generated in this compound.23 As another advantage, the topological surface states of BTS have been documented to be robust against chemical surface modification including oxidation under ambient conditions.24 Furthermore, BTS can be grown in the form of nanowires or ribbons, which are required for high-density device integration.25 This renders them into valuable functional parts into optoelectronic high-frequency and THz circuits.

We demonstrate that BTS nanowires can be integrated as functional parts into optoelectronic high-frequency circuits and that a pulsed laser excitation allows addressing the surface states of the nanowires even at room temperature.26,22 In particular, an on-chip THz time-domain photocurrent spectroscopy27–29 enables us to identify a photoconduction with a lifetime of several hundreds of picoseconds, which can be assigned to the surface states of the BTS nanowires. Our study addresses the

Received: October 14, 2016
Revised: January 12, 2017
Published: January 13, 2017
entire range of relevant time scales, i.e., starting with a femtosecond photoexcitation, including the ultrafast (picosecond) relaxation processes in the electron reservoirs of the surface and bulk states, and furthermore extending the insights toward the (microsecond) energy dissipation through the phonon bath of both the BTS nanowires and the circuits. Intriguingly, the specific surface state dynamics give rise to a photoconduction which switches signs as a function of laser power. Moreover, owing to the acceleration of photogenerated charge carriers in the depletion field of the metal contacts, the nanowires act as on-chip THz generators.

Individual BTS nanowires are contacted by two coplanar, metallic striplines (Figure 1a). Such THz circuits allow the

### Figure 1.
(a) Sketch of a laser-excited Bi$_2$Te$_2$Se nanowire embedded in a metal/topological insulator/metal geometry. The blue, black, and yellow spheres represent bismuth, tellurium, and selenium atoms, respectively, in the crystal structure of the nanowire. The nanowires comprise several tens of such atom layers. The two contacts are made from Ti/Au, and they form a THz-stripline circuit. (b) Optical microscope image of a contacted Bi$_2$Te$_2$Se-nanowire with 400 nm width and 120 nm height. Figure 1c depicts a time-averaged illumination at $V_{sd} = 2$ V. (d) Photocurrent map for a continuous wave excitation at $V_{sd} = 1$ V, a photon energy of 1.54 eV, and $P_{laser} = 3.7$ mW. Metal contacts are indicated by dashed lines. (e) Equivalent map for a pulsed laser excitation and $P_{laser} = 9$ mW. The photocurrent map in panel d (e) is performed with a microscope objective with a numerical aperture of NA = 0.28 (0.42), and a spot size of 3 μm (∼1.5 μm). All measurements are at room temperature. All scale bars are 10 μm.

read-out of the ultrafast optoelectronic dynamics within the nanowires and also to measure the time-averaged photocurrent $I_{photo}$ at a bias voltage $V_{sd}$. A laser with a photon energy of 1.54 eV is focused normally incident onto the nanowires, such that polarization-driven photocurrents at the surface of the BTS nanowires can be neglected. $I_{photo}$ Figure 1b shows an optical microscope image of one of the connected BTS nanowires. This particular nanowire has a width of 400 nm and a height of 120 nm. Figure 1c depicts $I_{photo}$ vs the laser power $P_{laser}$ for a laser spot focused at the center of the BTS nanowire. Intriguingly, for a pulsed excitation scheme, $I_{photo}$ switches polarity at about 4 mW, while for a cw-excitation, $I_{photo}$ increases monotonically with $P_{laser}$. When the sample is scanned laterally by the help of piezoelectric positioners, the cw-excitation gives a homogeneous photoresponse at all positions on the BTS nanowire (Figure 1d). For the pulsed excitation, by contrast, $I_{photo}$ changes polarity at the perimeters of the BTS nanowire (Figure 1e). The polarity change is explainable by the effectively reduced excitation intensity, when the laser is focused at the nanowire edges, as compared to a center excitation. In turn, the power dependence of $I_{photo}$ in Figure 1c phenomenologically explains the lateral polarity change in the photocurrent map of Figure 1e. Microscopically, however, the polarity change reflects the different recombination dynamics of the surface and bulk states in the BTS nanowires, as discussed below.

For accessing the ultrafast optoelectronic dynamics, we use an on-chip THz-time domain photocurrent spectroscopy. To this end, a pulsed laser excites the charge carriers in the BTS nanowires (Figure 2a and cf. Supporting Information). This

### Figure 2.
(a) Optical microscope image of a Bi$_2$Te$_2$Se-nanowire, integrated in an ultrafast THz time domain photocurrent spectroscopy circuit. The pump laser pulse generates a photocurrent in the nanowire generating an ultrafast electromagnetic pulse in the THz striplines. It is sampled with a probe laser pulse as the signal $I_{sampling} (\Delta t)$, with $\Delta t$ the time-delay between both laser pulses. The scale bar is 30 μm. (b) $I_{sampling}$ vs $P_{laser}$ for $V_{sd} = 2$ V with a pump pulse focused onto the center of the nanowire. Red, blue, and black curves represent fit functions (see text for details). (c) The fit function’s offset increases linearly with increasing excitation power. (d) Integrated area of the blue and red fit functions in (b) vs $P_{laser}$. Both saturate for $P_{laser} \geq 4$ mW.
pump laser is the same as for the time-integrated photocurrent measurements with a pulsed laser excitation (Figure 1c and e). Since the contacts form striplines, the photocurrents give rise to electromagnetic transients in the metal striplines with a bandwidth of up to 2 THz.\textsuperscript{11} The transients run along the striplines, and they are detected on-chip by a time-delayed optical femtosecond probe pulse in combination with an Auston switch (Figure 2a).\textsuperscript{30} The current $I_{\text{ampling}}$ across the Auston switch samples the electromagnetic transients on the striplines as a function of the time delay $\Delta t$ between the two laser pulses. It is directly proportional to the ultrafast photocurrents in the BTS nanowires.\textsuperscript{30} As a consequence, the corresponding photocurrents can be measured with the THz bandwidth of the striplines. Figure 2b depicts $I_{\text{ampling}}$ of the nanowire as a function of $\Delta t$ for $V_{\text{sd}} = 2$ V and $P_{\text{pump}} = 7$ mW. When the pump pulse hits the BTS nanowire, the current increases promptly and then decays on a fast (red) and a slow (blue) time scale. In addition, we find a negative offset signal (gray). We fit the data by the following carrier-lifetime-limited model for ultrafast currents (black line).\textsuperscript{30}

$$I_{\text{ampling}}(\Delta t) = I_{\text{fast}}[\exp(-\Delta t/\tau_{\text{fast}}) - \exp((\Delta t/\tau_{\text{fast}}))] + I_{\text{slow}}[\exp(-\Delta t/\tau_{\text{slow}}) - \exp((\Delta t/\tau_{\text{slow}}))] + I_{\text{offset}}$$\hspace{1cm}(1)

where $I_{\text{fast}}$ ($I_{\text{slow}}$) represents the amplitude of the fast (slow) decaying component. The time scale $\tau_{\text{fast}}$ ($\tau_{\text{slow}}$) is the corresponding lifetime of the current contribution. The rise time of the fast component $\tau_{\text{fast}}$ is related to the bandwidth of the used stripline circuit. The presence of the offset $I_{\text{offset}}$ indicates that there exists a photoresponse which prevails longer than the repetition time of the pulsed laser (76 MHz$^{-1}$ $\sim$ 13.2 ns). We note that the model of two carrier-lifetime-limited currents fits best our data, when we take the decay time of the fast decay (red) as the rise time of the slow one (blue). In other words, both dynamics are coupled to each other. This finding is consistent with the following microscopic model.

Generally, photo-generated charge carriers relax on different time scales in the bulk and surface states of 3D TIs.\textsuperscript{31} Directly after the photoexcitation, a nonequilibrium carrier density is established in the bulk bands. It relaxes to the corresponding band minima via electron–electron and electron–phonon scattering processes on a picosecond time scale.\textsuperscript{32} For BTS, the hot charge carriers then relax into the topological surface states. This redistribution process is reported to occur in the first tens of picoseconds.\textsuperscript{32} We find $\tau_{\text{fast}} = 46 \pm 6 \text{ ps}$ in our time-resolved photocurrent experiment. Accordingly, we interpret the fast current component (red in Figure 2b) to resemble the excitation of a bulk charge carrier density and $\tau_{\text{fast}}$ to describe the charge redistribution from the bulk to the surface states. The dynamics within the surface states occur on a slow time scale. In angle-resolved photoemission experiments,\textsuperscript{22} lifetimes are reported up to the $\mu$s regime for an equivalent excitation scheme and photon energy as in our optoelectronic experiment. We find $\tau_{\text{slow}} = 439 \pm 32 \text{ ps}$. In turn, we interpret the slow photocurrent component (blue) to relate to the population and subsequent depletion of the topological surface states. The nanowires are naturally $n$-doped, with an electron concentration on the order of $10^{19}$ cm$^{-3}$, as determined by Hall measurements (cf. Supporting Information). Importantly, the nanowire thickness above 60 nm exceeds the bulk phase relaxation length of BTS, thereby increasing the number of coherent charge transport channels via coupling between the surface and bulk states.\textsuperscript{52} This coupling explains why the longer lifetime ($\tau_{\text{slow}} = 439 \text{ ps}$) is significantly shorter than the lifetime on the order of $\mu$s, as determined from ARPES data on BTS whose Fermi level lies within the bulk band gap.\textsuperscript{52}

We now focus on the negative offset current (gray arrow in Figure 2b). It increases linearly without any saturation in the examined range of laser powers (Figure 2c). For comparison, Figure 2d shows the integrated area of the red and blue fit components from Figure 2b as a function of the pump power. After a linear increase, both components saturate at $\sim$4 mW. We interpret the saturation to represent a phase space filling at a high laser power. On the one hand, the lack of a saturation for the offset current indicates that this slow optoelectronic contribution is of a different kind than the faster ones. On the other hand, it also suggests for high laser powers that the negative offset overwhelms the saturated positive contributions, which explains the polarity change of the time-averaged $I_{\text{photo}}$ at $\sim$4 mW for a pulsed laser excitation (Figure 1c).

In the following, we demonstrate that a bolometric effect in the BTS nanowires describes the slow offset current. Figure 3a depicts the two-terminal conductance $G$ of a BTS nanowire as a function of the bath temperature $T_{\text{bath}}$ without any laser excitation. The conductance decreases with increasing temperature with a corresponding bolometric coefficient $\Delta G/\Delta T_{\text{bath}} = -86 \pm 1 \text{ nS K}^{-1}$ at room temperature. The decrease is consistent with metallic surface states dominating the electronic transport properties without laser illumination. In a next step, we perform a finite element simulation of the laser-induced

![Figure 3](https://example.com/figure3.png)

Figure 3. (a) Two-terminal conductance $G$ without laser excitation vs temperature. (b) Schematic side view of the laser-excited Bi$_2$Te$_2$Se nanowire and the Al$_2$O$_3$ substrate. The assumed height and width of the nanowire are 120 and 400 nm, respectively. For the thermal conductivity and specific heat of the nanowire, we used the values for Bi$_2$Te$_3$ (1.5 W m$^{-1}$ K$^{-1}$ and 126 J mol$^{-1}$ K$^{-1}$))\textsuperscript{50–52} Only the heat transfer to the Al$_2$O$_3$ substrate and the metal contacts is taken into account. For Al$_2$O$_3$, we use the following values of the thermal conductivity and specific heat as reported for room temperature (20 W m$^{-1}$ K$^{-1}$ and 79 J mol$^{-1}$ K$^{-1}$)).\textsuperscript{53,54} (c) Top view depicts the simulated heat profile after the laser excitation at $P_{\text{laser}}$ of 10 mW. Lower panel shows the corresponding relative conductivity change. (d) Simulated current change across the whole Bi$_2$Te$_2$Se nanowire vs $P_{\text{laser}}$ for $V_{\text{sd}} = 2$ V.
heat profile and the corresponding bolometric current change in the BTS nanowires. Figure 3b sketches a side view of a nanowire on top of an Al₂O₃ substrate and electrical contacts on both ends. We simulate the laser spot as a constant heat source with a Gaussian profile and a diameter of 4 μm (with further simulation parameters given in the figure caption). The upper panel of Figure 3c presents the resulting heat profile along the nanowire for an excitation/heating power of 10 mW. The simulation suggests that heat dissipation occurs through the substrate on the time scale of a few nanoseconds (cf. Supporting Information). Moreover, the temperature increase is mainly localized within the laser spot (dashed circle) with a maximum temperature increase of about 100 K in the center. The lower panel shows the corresponding normalized conductivity change along the nanowire taking into account the experimentally determined bolometric coefficient. In turn, the local conductivity drops by more than 5% in the laser spot center due to heating. As the laser spot covers the full nanowire cross-section, all current paths are affected by the conductivity bottleneck. This is a major difference between a nanowire and a planar thin-film geometry. Figure 3d shows the calculated current change ΔI_{calc} across the BTS nanowire as a function of the excitation power for V_{sd} = 2 V. We find that the computed current linearly drops to a value of about −5 μA at 20 mW. This value is in very good agreement with our experimental findings of the time-averaged photocurrent values in Figure 1c. Consequently, we can explain the polarity change of the time-averaged photocurrent I_{photo} at a pulsed laser excitation as a bolometric decrease of the charge carrier mobility μ.

To further clarify the underlying optoelectronic dynamics, we express the optically induced change of the local conductivity σ as

\[ \Delta \sigma(P_{\text{laser}}) = \Delta n(P_{\text{laser}}) \mu + n e \Delta \mu(P_{\text{laser}}) \]

with Δμ the optically induced change of the mobility and e the electron charge. The expression Δn describes the optically induced change of the overall charge carrier density at the excitation spot. Equation 2 neglects the contact resistances of the experimental two-terminal (photo-) conductance. Nonetheless, it allows to explain the polarity change of I_{photo}. For a pulsed excitation and high P_{laser}, the second term in eq 2 dominates the time-averaged photocurrent (I_{photo} in Figure 1c). In other words, the laser heats up the BTS nanowire, and a prevailing bolometric contribution with Δμ(P_{laser}) < 0 in the surface states dominates, because the faster lifetime-limited photocurrents with Δn(P_{laser}) in the surface and bulk states have decreased for the majority of the repetition time of the laser (compare Figure 2b). At low P_{laser} however, the first term of eq 2 dominates. This explains the polarity change of I_{photo} vs P_{laser} for a pulsed excitation (Figure 1c). For the pulsed excitation, the instantaneous power is 10^5 higher than the average power. In other words, with a cw laser excitation, the maximum photon density per time is reduced by a factor of about 10^-3 compared to the 150 fs laser pulses with a repetition frequency of 76 MHz. Therefore, neither the surface nor the bulk states are pumped to saturation for the investigated range of P_{laser} (compare Figure 2d). In turn, the ratio of the photon-to-electron-conversion is higher than in the pulsed excitation regime. Consequently, for the cw laser excitation, an increased steady-state charge carrier density dominates the overall photocurrent also for a higher P_{laser} (Figure 1c). Moreover, we note that the repetition rate of the pulsed laser in the nanosecond regime is comparable to the heat dissipation rate of the substrate (for the latter compare the Supporting Information). In other words, a laser pulse excites certain charge carriers which are transported according to a reduced charge carrier mobility, and the reduction is caused by the impact of the laser pulse(s) before. With respect to eq 2, this could be expressed as an additional term in the form of Δn(P_{laser})eΔμ(P_{laser}). However, it is beyond the scope of the present study to determine the exact values of Δn and Δμ in the quasi-stationary limit of the pulsed excitation scheme.

So far, we discussed photoconductance phenomena driven by a finite bias voltage. Figure 4a shows I_{sampling} at zero bias (green) in comparison to V_{sd} = ±2 V (black). The data are manually offset for clarity. One can see that the photocurrent responses at positive and negative bias are mirrored with opposing sign, which is consistent with eq 2. At zero bias and zero time delay, however, an oscillatory signal clearly shows up. We note that this oscillatory signal also occurs at a finite bias. There, however, the oscillatory signal is superimposed by the already discussed slower optoelectronic dynamics. Figure 4b shows the oscillatory signal for |Δt| ≤ 10 ps at zero bias. The black line is a fit to the data consisting of a Gaussian shaped wave package. (c) Fourier transforms of the ultrafast oscillation vs frequency. (d) THz amplitude (green) and phase (black) as a function of the laser spot position along a Bi₂Te₂Se nanowire. Black dashed lines indicate the positions of the striplines. The amplitude features maxima in the vicinity of the metal contacts, whereas the phase of the oscillation changes from π to 0 when the laser is scanned from one contact to the other.
bandwidth of our stripline circuitry. Moreover, we observe that the THz generation depends on the position of the laser excitation with respect to the BTS nanowires. Figure 4d shows the THz amplitude (green) and phase (black) as a function of the laser spot position along the BTS nanowire, which is depicted in the optical microscope image of Figure 1b. Notably, this nanowire extends by a few micrometers beyond the right stripline. This elongated part of the nanowire shows no time-integrated (bias-dependent) photocurrent (compare Figure 1d and e). However, it emits THz radiation (Figure 4d). The position scan further reveals that the THz amplitude features maxima in the vicinity of the metal contacts, whereas the THz phase of the oscillation changes from $\pi$ to 0 when the laser beam is scanned from one contact to the other.

Generally, there are several THz generation mechanisms, which can induce a THz emission from a material after optical excitation. Among them are the effect of nonlinear optical rectification and photo-Dember effects. The latter rely on the ambipolar diffusion of electrons and holes due to distinct charge-carrier mobilities. Moreover, a THz radiation can be generated by ultrafast transient photocurrents, where optically excited charge carriers are accelerated in an intrinsic electric field at the material’s surface or in the depletion area caused by a Schottky barrier at a metal–semiconductor interface.

For BTS, an optical rectification effect can only occur in the topological surface states, as the second-order nonlinear susceptibility does not vanish when the inversion symmetry is broken. We can exclude the optical rectification as the dominant THz generation process in the discussed BTS nanowires because we do not observe any dependence of the THz oscillation on the polarization of the excitation laser (cf. Supporting Information). Instead, the maxima of the THz amplitude close to the metal contacts in Figure 4d suggest that an acceleration of photogenerated charge carriers in the corresponding electric depletion region of a Schottky barrier explains the THz generation. Also, the observed spatial variation of the THz phase is consistent with this interpretation (bottom panel in Figure 4d), since the THz radiation couples to the asymmetric waveguide mode of our coplanar striplines. Notably, the phase observed at the elongated part of the nanowire corroborates this interpretation (cf. Supporting Information). In principle, a lateral photo-Dember effect is also consistent with the spatial variation of the THz phase at the contacts. For a lateral photo-Dember effect, however, the THz phase should change polarity at the very end of the elongated part of the BTS nanowire, which we do not detect. Furthermore, the electron and hole mobilities are rather similar in bismuth chalcogenides, which makes photo-Dember effects unlikely in these materials. In THz-far-field studies on planar, macroscopic 3D TIs, a THz generation was reported to be consistent with an optical rectification, with an intrinsic surface voltage, and with an homogeneous photo-Dember effect. Another ultrafast process reported is a photoinduced charge transfer along the Se-Bi bonds with frequency components even up to 40 THz. For all of these mechanisms, the THz oscillation would occur homogeneously along the BTS nanowires. In principle, we cannot exclude that these THz generation mechanisms are still contributing on a minor level to the observed THz signal although the THz generation caused by electric fields associated with a Schottky barrier dominates. In particular, our on-chip THz spectroscopy has a bandwidth of only up to 2 THz. The bandwidth is limited by the effective index of refraction and attenuation of the striplines.

The main limitation is given by the so-called geometric dispersion of the striplines, but there are also small contributions of the material dispersion and radiative losses at the highest frequencies. However, our spectroscopy allows to resolve the spatial variation of the THz generation even on the level of single contacted BTS nanowires, which is impossible in far-field studies so far. Moreover, our results demonstrate that BTS nanowires can trigger THz radiation in the near-field which then couples into on-chip THz circuits. In other words, the data in Figure 4b, c, and d depict the THz components which are propagating along the THz striplines before they are detected on-chip (at room temperature). Hence, single BTS nanowires can be functional parts of chip-based, optoelectronic high-frequency and THz circuits.

To conclude, our experiments reveal the optoelectronic dynamics and interplay of bulk and surface charge carrier distributions, as well as thermal processes in nanowires of the 3D topological insulator (TI) Bi$_2$Te$_2$Se (BTS). We demonstrate that the pulsed photoconductance of the BTS nanowires changes sign as a function of laser power. We succeed in separating the bulk and surface state contributions to the generated photoconduction. Among these, the component carried by the surface states has a lifetime of several hundreds of picoseconds, one order of magnitude longer than for the bulk states. Under pulsed laser excitation, the photoconduction is dominated by a bolometric decrease of the charge-carrier mobility in the metallic surface states as a consequence of a steady-state heating of the BTS. For a continuous wave laser excitation, the photoconduction turns out to be dominated by an overall increased charge carrier density. Moreover, ultrafast transient photocurrent generation occurring in the Schottky depletion field of the metal contacts renders individual BTS nanowires into a novel type of THz generators. Our findings may open novel perspectives for designing photodetectors, solar cells, THz sources, and high-frequency circuits based on topological insulator nanowires.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.6b04312.

BTS nanowire growth and charge carrier density, design of the THz stripline circuits, time-integrated photocurrent spectroscopy, details on the on-chip time-domain THz photocurrent spectroscopy, and polarization dependence of the THz signal (PDF)

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Notes

The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

We acknowledge financial support by the DFG-priority program 1666 on topological insulators and the ERC-grant NanoREAL.
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