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Magnetic-impurity-induced modifications to ultrafast carrier dynamics in the ferromagnetic topological insulators Sb$_{2-x}$V$_x$Te$_3$

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Abstract

Quantum anomalous Hall effect (QAHE) is a key phenomenon for low power-consumption device applications owing to its dissipationless spin-polarized and quantized current in the absence of an external magnetic field. However, the recorded working temperature of the QAHE is still very low. Here we systematically investigate the magnetic dopants induced modifications from the view points of magnetic, structural and electronic properties and the ultrafast carrier dynamics in a series of V-doped Sb$_2$Te$_3$ samples of composition Sb$_{2-x}$V$_x$Te$_3$ with $x = 0, 0.015$ and $0.03$. Element specific x-ray magnetic circular dichroism signifies that the ferromagnetism of V-doped Sb$_2$Te$_3$ is governed by the $p-d$ hybridization between the host carrier and the magnetic dopant. Time- and angle-resolved photoemission spectroscopy excited with mid-infrared pulses has revealed that the V impurity induced states underlying the topological surface state (TSS) add scattering channels that significantly shorten the duration of transient surface electrons down to the 100 fs scale. This is in a sharp contrast to the prolonged duration reported for pristine samples though the TSS is located inside the bulk energy gap of the host in either magnetic or non-magnetic cases. It implies the presence of a mobility gap in the bulk energy gap region of the host material that would work toward the robust QAHE. Our findings shed light on the material design for low-energy-consuming device applications.

1. Introduction

Topological insulators (TIs) possess spin-polarized massless Dirac fermions at surfaces and interfaces protected by time-reversal symmetry [1–3]. As a hallmark of TIs, a spin-momentum locking of the topological surface state (TSS) stemming from a strong spin–orbit coupling leads to robustness against non-magnetic impurities or other external perturbations as long as time-reversal symmetry is preserved. Moreover, spin-momentum locking results in a suppression of elastic electron scattering which limits ballistic electron transport in topologically trivial materials. In 2D TIs, dissipationless spin-polarized edge currents have been experimentally observed by transport measurements [4]. Recent time- and angle-resolved photoemission spectroscopy (TARPES) experiments have shown that terahertz light pulses can drive electron currents with ballistic mean free paths of several hundred nanometers also at the surface of 3D TIs [5]. Once magnetic impurities are introduced to TIs, a gap opening may happen at the Dirac node of the TSS as a result of the interaction between the magnetic impurity and the TSS [6–11]. This can lead to an exotic quantum phase such as a quantum anomalous Hall...
(QAH) state, accompanying spin-polarized and quantized chiral edge currents in the absence of an external magnetic field [12]. It is, therefore, believed that the QAH effect (QAHE) is a key for low power-consumption device applications. For the realization of the QAH state utilizing TIs, there are mainly two criteria; one is to break time-reversal symmetry, which leads to a gapped Dirac state. The other is to tune the Fermi level ($E_F$) into the magnetically induced gap. The former criterion can be achieved by adopting two ways; proximity effect of ferromagnetic materials or establishing long-range magnetic ordering by doping [13–18]. Recently, QAH states were experimentally verified in Cr- or V-doped (Sb,Bi)$_2$Te$_3$ thin films at extremely low temperature (10 mK-order) [19, 20]. A small amount of 3d magnetic dopants causes a long-range ferromagnetic order in the TIs. Furthermore, in the host TIs (Sb$_{1−x}$Bi$_x$)$_2$Te$_3$, the carrier type can be widely tuned from p to n with an increase of x [21–23]. Especially, V-doped (Sb,Bi)$_2$Te$_3$ is a hard ferromagnet exhibiting a larger coercive field and higher Curie temperature compared to the Cr-doped cases but the working temperature of the QAHE is still very low although extensive efforts have been made to improve it by modulation doping or co-doping methods [24, 25].

The microscopic origin of the ferromagnetism and the QAHE state has been extensively investigated by theoretical and experimental approaches [26–38]. Element-specific x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD) experiments revealed that the long-range ferromagnetic order in the magnetic TIs is caused by a magnetic coupling between $p$–$d$ electrons of the host and magnetic dopants because the clear XMCD signals were observed not only at V (Cr) sites but also at Sb sites [29, 33, 35, 36, 38]. In addition, first principles calculations, scanning tunneling microscopy/spectroscopy (STM/STS) and resonant photoemission spectroscopy identified the impurity states of vanadium dopants at or near the Dirac node [26, 30, 32, 36]. These might induce several modifications to the band structure [39–41], implying, for instance, the existence of a mobility gap, namely, the coexistence of the gap opening and the presence of impurity states at the Dirac node [30]. It is, therefore, very important to understand the interaction between the host and the magnetic dopants to raise the critical temperature of the QAHE. Nevertheless, the influence of the magnetic impurities to the TSSs remains unexplored. One reason is that experimental access to the TSS upon magnetic doping is missing. Since the mother crystal Sb$_2$Te$_3$ is naturally hole-doped, conventional angle-resolved photoemission spectroscopy (ARPES) cannot access to the TSS. The implementation of TAR-PES as pump-probe method is a powerful solution to visualize the entire TSS and to investigate the ultrafast carrier dynamics in the magnetic TIs.

In this article, we systematically investigate the magnetic dopants induced modifications to electronic structures from the view points of magnetic, structural and electronic properties and the ultrafast carrier dynamics in a series of Sb$_{1−x}$V$_x$Te$_3$ ($x = 0, 0.015, 0.03$) by a combination of XMCD, STM/STS and TAR-PES. The XMCD experiment has revealed that the host Te 5$p$ and Sb 5$p$ electrons carry magnetic moments that mediate the localized V 3$d$ spins. Their dichroic signals tell us that the Sb 5$p$ and Te 5$p$ spins are coupled parallel and anti-parallel to those of V 3$d$ spins, respectively. Furthermore, STM and its differential conductance spectroscopy (STS) resolved two independent V-induced impurities, which are energetically overlapped with the TSS. More importantly, when the doping amount of V was increased, the electronic recovery time for the nonequilibrated state was drastically shortened to <500 fs. The observed shortened duration indicates an increased impurity scattering path between the host and the magnetic dopants.

2. Experimental

Sb$_{1−x}$V$_x$Te$_3$ single crystals ($x = 0.015, 0.03$) were grown by the modified vertical Bridgman method with rotating heat field [42]. Ferromagnetic properties of the samples were confirmed by SQUID and the Curie temperatures ($T_C$) were found to be in the range of 5–20 K. The STM/STS experiments were performed at HiSOR using an LT-STM (Omicron Nano Technology GmbH) operated at 77 K in an ultrahigh vacuum. The XAS/XMCD experiments were performed at BL23SU of SPring-8 for V $L_{2,3}$, Sb and Te $M_{4,5}$ absorption edges at 10 K, below the Curie temperature [43]. The applied magnetic field was set to $±2$ T that was high enough to saturate the magnetization. The XAS and XMCD spectra were acquired in total-electron-yield mode in ultrahigh vacuum. The TAR-PES experiments were performed at Philips University of Marburg [44]. A femtosecond Ti:sapphire regenerative amplifier (Coherent RegA 9040) generated 40 fs laser pulses with central photon energy of 1.55 eV (800 nm). The output of the amplifier was split into two parts which were used to pump two traveling wave optical parametric amplifiers (OPA) operating in visible (VIS) and near-infrared (NIR) range, respectively. The output of the VIS–OPA was frequency doubled by a $β–$BaB$_2$O$_4$ crystal to generate the ultraviolet (UV) probe pulses. The mid-infrared (mid-IR) pump pulses were generated by difference frequency mixing of signal and idler beams of the NIR-OPA. For this experiment, we tuned the photon energies of the VIS and UV pulses to be 2.52 eV (80 fs, 130 nJ/pulse) and 5.03 eV (100 fs, 2 nJ/pulse), respectively. The photon energy of the mid-IR pump pulses was set to 0.33 eV (100 fs, 10 nJ/pulse). The p-polarized mid-IR pump and UV probe beams were focused onto the sample with a diameter of 100 μm. To determine the temporal overlap between pump and
probe pulses, we used short-living image-potential states (IPSSs), which are populated by the UV pulses and photoemitted by the mid-IR pulses. The repetition rate was set to 100 kHz. To see solely the magnetic impurity effect in the absence of the long-range ferromagnetic order, our TARPES experiment was carried out at 110 K (well above $T_c$ of the samples).

3. Results and discussion

To reveal the element-specific magnetic properties, we have performed the XAS and XMCD experiment for the samples with $x = 0.015$ and 0.03. Figure 1(a) shows the XAS spectra acquired by using circularly polarized light at V $L_{2,3}$, Sb and Te $M_{4,5}$ absorption edges of Sb$_{1.985}$V$_{0.015}$Te$_3$ at 10 K. The external magnetic field was set to 2 T and applied along the perpendicular direction to the sample surface. At V $L_{2,3}$ absorption edges, a clear difference can be seen though the differences at Sb and Te $M_{4,5}$ absorption edges are very small. Figure 1(b) shows the XMCD spectrum using the same energy scale as used in figure 1(a). At the V $L_{2,3}$ absorption edges, clear and large dichroic signals are observed. We thus conclude that the magnetic moment is dominated by the 3d states of the V dopants. The complicated XMCD line shape at V $L_{2,3}$ can be described by the $2p^63d^4$ final state multiplet for $V^{3+}$ ion ($3d^2$) with a crystal field of 10Dq $= 1$ eV [45]. We have also observed XMCD signals at the Sb $M_{4,5}$ absorption edges. The observed Sb 5p magnetic moment at the non-magnetic element is most likely induced by the V magnetic dopant. This result is in good agreement with the previous XMCD studies for Cr- or V-doped compounds [29, 33, 35, 36, 38]. More importantly, very small but clear XMCD signals are observed at the Te $M_{4,5}$ absorption edges. The presence of XMCD at the Sb and Te $M_{4,5}$ edges tells us that the V 3d spins are coupled with the Sb and Te 5p spins of the host Sb$_2$Te$_3$. Figure 1(c) shows the XMCD intensities at the V $L_3$, Sb $M_3$ and Te $M_5$ edges as a function of the external magnetic field. The non-zero XMCD intensities are observed at zero field for all elements. These results signify that the magnetic coupling between the local magnetic 3d moment and the host 5p moment plays an important role in the forming of long-range ferromagnetic order. Here we get deeper insight into details of the underlying mechanism of magnetic coupling. It is noted that the sign of the XMCD signal at the Sb $M_3$ edge is opposite with respect to those at the V $L_3$ and Te $M_5$ edges. Having considered that the V $L_{2,3}$ edge corresponds to a 2p to 3d transition, the Sb and Te $M_{4,5}$ edges both correspond to a 3d to 5p transition, we can conclude that the V 3d and Sb 5p spins are aligned parallel while the V 3d and Te 5p spins are aligned anti-parallel. In figure 1(d), we also show the magnetic field dependent XMCD intensities of the sample with higher V doping ($x = 0.03$). The result is similar to the lower doping case (figure 1(c)) but the XMCD intensities and remanent magnetization at all absorption edges are larger for $x = 0.03$ than those for $x = 0.015$. This means that the magnetic moment of the V per atom is reduced upon decreasing V doping amount, which might be due to a reduced $T_c$ for $x = 0.015$ [13].

Figure 2(a) shows the topographic image of $x = 0.015$ (left) recorded by STM in the constant-current mode. We can see several different type of defects identified by comparing the topographic image with that of the pristine sample (figure 2(a, right)). Here, we can distinguish two possible V-induced defects as marked by dashed squares (defect 1, defect 2). The observed defects are in good agreement with a previous study which argues that the V dopants occupy the second and fourth Sb layers of the Te-Sb-Te-Sb-Te quintuple layer block [38]. Thus, the location of the magnetic dopant and the relative spin orientation of V-doped Sb$_2$Te$_3$ can be summarized as (Sb, V) $\uparrow$ $-$ (Te) $\downarrow$, as depicted in figure 2(c). In order to see the local density of states created by the V dopants,
we have also measured the differential conductance \( dI/dV \) as a function of sample bias voltage. Figure 2(b) shows the \( dI/dV \) (STS) spectra obtained at defect 1, defect 2, and on flat areas. The \( dI/dV \) spectrum on flat areas shows a steep rise below \( V \sim 0.1 \) V and above \( V \sim 0.25 \) V that correspond to the onset of the bulk valence band and the bulk conduction band (BCB), respectively. The minimum of the \( dI/dV \) curve is found at \( V \sim 0.15 \) V on the flat area, which coincides with the Dirac point of the TSS. We now know from these \( dI/dV \) features that the TSS is located within these bands and well above \( E_F \). The bulk band gap is estimated to be \( \sim 200 \) meV. When the \( dI/dV \) spectra are acquired on the defects, some additional features emerge within the bulk band gap. The impurity-induced local intensity maxima are seen at sample bias voltages of 0.2 V and 0.15 V at defects 1 and 2, respectively.

To investigate the magnetic impurities on the ultrafast carrier dynamics, we have performed TARPES measurements using a mid-IR-pump and UV-probe scheme. For this experiment, we have chosen a mid-IR pump energy of 0.33 eV which has been shown to generate a population of the TSS by a direct optical transition between its lower and upper branch across the Dirac point [44]. This is in sharp contrast to VIS or NIR pumping which results in a bulk states mediated indirect population [47]. Figure 4 shows the TARPES images recorded at several pump-probe delay times for \( x = 0 \) (a), 0.015 (b) and 0.03 (c) samples. Upon increasing \( x \), the Dirac point is slightly shifted to higher energies. The Dirac points for \( x = 0, 0.015 \) and 0.03 are located at 200, 210, and 220 meV above \( E_F \), respectively.
210 and 220 meV, respectively. The locus of the Dirac point of the pristine sample is consistent with previous reports \[48–51\]. Around \(t \sim 0\) ps, we observe TARPES signals of IPSs with quantum numbers \(n = 2\) and 3 which are excited by the UV pulses and probed by the mid-IR pulses. Excited electrons in the parabolic IPSs surface bands are short-lived with decay times of \(\sim 50\) fs \[52\]. We even observe quantum beats between these electronic states \[53\], indicating a clean, flat and well-ordered surface of high quality \[54\].

Figure 3. (a) Band dispersion of the sample with \(x = 0.015\) recorded at 2.52 eV pump energy. (b) \(dI/dV\) (STS) spectra for \(x = 0.015\) in the same energy scale as that for panel (a) obtained at defects 1 and 2 and on flat areas. Magnetic impurity-induced local densities of states are depicted by arrows.

Figure 4. Nonequilibrium carrier dynamics in the Sb\(_{2-x}\)V\(_x\)Te\(_3\) crystals. TARPES images recorded at various delay times for \(x = 0\) (a), 0.015 (b) and 0.03 (c). Time-evolution of the momentum-resolved intensities at \(E − E_F = 0.33\) eV for \(x = 0\) (d), 0.015 (e) and 0.3 (f). (g) Time- and V doping-dependent electronic energy \(U\) estimated for \(x = 0, 0.015\), and 0.03 (see text for more details). The solid line is a fit to the experimental data by the convolutions of a Gaussian function with exponential-decay curves.
remain at $t = 1.5$ ps not only in the Dirac band above $E_F$ but also at the bottom of the BCB (figure 4(a)). This is because the Dirac point acts as a bottleneck in phase space [49]. For $x = 0.015$, the excited electrons in the upper TSS and the BCB are mostly relaxed within 1 ps (figure 4(b)). Finally, we look into the dynamics of the sample $x = 0.03$ (figure 4(c)). In this case, the recovery time of the nonequilibrated state is the shortest among the three samples, although the shape of the band structure and the locus of the Dirac point are almost similar to those of the other specimens. At $t = 500$ fs, the relaxation is mostly accomplished. To our knowledge, this is the fastest relaxation among the $p$-type TIs. Note that we could not see a signature of population inversion because the mid-IR pump fluences are insufficient to populate a large amount of electrons to the upper TSS.

In order to see the time-evolution of the photoemission intensities more clearly, figures 4(d)–(f) show the time- and momentum-resolved intensities extracted at $E-E_F = 0.33$ eV, where the population of the upper TSS is pronounced by the direct optical transition. We can clearly see that the lifetime of the population at $E-E_F = 0.33$ eV is systematically shortened from $\sim 3$ ps to $\sim 200$ fs as $x$ is increased. We also realize that the duration of the asymmetric populations at the upper TSS is shortened upon increasing $x$. Especially, in figure 4(f), the photoemission intensities at the left and right branches of the TSS are almost symmetric even at $\sim 0$ fs.

To quantify the magnetic impurity-induced modifications in the carrier dynamics, we estimate the following integral $U(t) \equiv \int_{\omega > 0} \omega I(\omega, t) d\omega$, which represents the electronic energy retained by the electrons above $E_F$. Here, $\omega = E - E_F$, and $I(\omega, t)$ is the angle-integrated photoemission intensity. In figure 4(g), we plot $U(t)$ for $x = 0, 0.015$, and 0.03. Upon the arrival of the pump pulse, $U(t)$ sharply rises and peaks at $\sim 0$ ps for all samples. The pump-induced changes at $x = 0$ persist even at $>3$ ps while the transient electron intensities at $x = 0.015$ and 0.03 are almost diminished within $\sim 1$ ps. The solid line is a fit to the experimental data by convolutions of a Gaussian function with exponential-decay curves. The estimated time constants for $x = 0, 0.015$, and 0.03 are $1520, 181$, and $44$ fs, respectively.

Finally, we discuss the reason for the faster decay observed in the V-doped specimens. Most probably it is attributed to interband scattering within the TSS via induced impurity states. As shown in figure 3, the local density of states of V overlaps with the TSS, thus, interband scattering from the TSS to the impurity states may be enhanced (figure 5(left)). The magnetic dopants are expected to enhance also the momentum scattering, in particular the scattering between $+k$ and $-k$ within the TSS. For non-magnetic TIs, backscattering between right and left branches of the TSS is basically prohibited (figure 5(upper right)) but should be tolerated in the presence of magnetic impurities (figure 5(lower right)). That is, impurity-assisted interband scattering can occur even in the lack of the long-range ferromagnetic order (well above $T_C$) due to the local magnetic moments of the V atoms. The fast relaxation of the asymmetric population observed for $x = 0.03$ (figure 4(f)) probably supports this scenario. Both these scattering mechanisms between the impurity and the host carrier would accelerate the carrier dynamics.

Let us also remark the relation between magnetic ordering and surface gap opening. In this study, we define the loci of the Dirac node and the V impurity induced states by a combination of the STS and TARPES (see figure 3(a)). Although our measurements are performed above the Curie temperature, this is likely to induce the dual nature of the magnetic TIs [30]. Namely, even if an opening of the gap is expected at the Dirac node below the Curie temperature, the impurity resonance states fill the gap at the same time (mobility gap scenario).
4. Conclusion

In conclusion, we have investigated the influence of magnetic dopants on magnetic, structural and electronic properties and the ultrafast carrier dynamics in a series of $Sb_2xV_xTe_3$ samples with $x = 0, 0.015, 0.03$ by a combination of XMCD, STM/STS and TARPES. We revealed that the ferromagnetism of $V$-doped $Sb_2Te_3$ is governed by the $p-d$ hybridization between the host carrier and the magnetic dopant. Furthermore, the impurity states underlying the TSS were found to add scattering paths that shorten the duration of transient TSS electrons significantly through a combination of the STS and TARPES using a mid-IR pumping scheme. Our findings might contribute to an improvement of the QAH properties by scrutinizing the interaction between host and magnetic dopants and pave a way for the material design for low-energy-consuming device applications.

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