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RAPID COMMUNICATION

# Realization of low-energy type-II Dirac fermions in $(Ir_{1-x}Pt_x)Te_2$ superconductors*

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Topological Dirac semimetals (DSMs) present a kind of topologically nontrivial quantum state of matter, which has massless Dirac fermions in the bulk and topologically protected states on certain surfaces. In superconducting DSMs, the effects of their nontrivial topology on superconducting pairing could realize topological superconductivity in the bulk or on the surface. As superconducting pairing takes place at the Fermi level $E_F$, to make the effects possible, the Dirac points should lie in the vicinity of $E_F$ so that the topological electronic states can participate in the superconducting paring. Here, we show using angle-resolved photoelectron spectroscopy that in a series of $(Ir_{1-x}Pt_x)Te_2$ compounds, the type-II Dirac points reside around $E_F$ in the superconducting region, in which the bulk superconductivity has a maximum $T_c$ of $\sim 3$ K. The realization of the coexistence of bulk superconductivity and low-energy Dirac fermions in $(Ir_{1-x}Pt_x)Te_2$ paves the way for studying the effects of the nontrivial topology in DSMs on the superconducting state.

**Keywords:** type-II Dirac semimetal, superconductor, topological superconducting, angle-resolved photoemission spectroscopy (ARPES), substitution

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The interplay between topological states and superconductivity is an important issue in condensed matter physics.[1,2] One consequence of the interplay is the generation of Majorana fermions at the boundary of a topological superconductor, which can be used for topological quantum computation. Tremendous efforts have been made to induce superconductivity in topological materials through various approaches, such as introducing carriers, applying external pressure, and proximity effect, or to search for superconductors with nontrivial topological electronic states. Superconductivity has been observed in doped topological insulators, such as Cu-intercalated $Bi_2Se_3$,[3] $Bi_2Te_3$ under pressure,[4] $Bi_2Se_3$ thin films on superconducting $NbSe_2$ substrate,[5] and In-doped SnTe.[6] Several superconductors such as *R*PtBi (*R*: rare earth elements),[7–10] $Bi_2Pd$,[11] and $FeTe_{0.55}Se_{0.45}$[12,13] have been revealed to have a nontrivial topology similar to that of topological insulators.

In addition to superconducting topological insulators, theory has proposed that in superconducting Dirac and Weyl semimetals, topological superconductivity can also be realized for the bulk or the surface due to their nontrivial topology.[14–16] Experimentally, the superconductivity was induced by point contact[17,18] or under high pressure[19] in the Dirac semimetal (DSM) $Cd_3As_2$. The point contact spectra of $Cd_3As_2$ show a zero-bias conductance peak, suggesting unconventional superconductivity around the point contact region.[17,18] Recent studies have reported that several transition metal dichalcogenide (TMD) compounds $PtTe_2$,[20] $PdTe_2$,[21,22] and $PtSe_2$[23] with the $CdI_2$-type 1T structure host type-II Dirac points in their electronic structures. As illustrated in Figs. 1(c) and 1(d), distinct from type-I DSMs like $Na_3Bi$[24,25] and $Cd_3As_2$,[26,27] type-II DSMs possess over-

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tilted Dirac cones so that the two Dirac bands have the same sign in their slope along one direction, in analogy to the type-II Weyl semimetals.[28–30] As the type-II Dirac points in these TMD compounds reside far below $E_F$, the Dirac fermions should have little contribution to the low-energy quasiparticle excitations at $E_F$. Very recently, Fei *et al.* reported that the energy position of the type-II Dirac points in $Ir_{1-x}Pt_xTe_2$ can be easily tuned by element substitution.[31] The Dirac points arrive at $E_F$ at $x \sim 0.3$, where a bulk superconductivity emerges with $T_c = 0.15$ K.

In this work, by combining angle-resolved photoemission spectroscopy (ARPES), energy-dispersive x-ray spectroscopy (EDX), x-ray diffraction (XRD), and transport measurements, we systematically investigated the evolution of electronic structures and superconductivity of $(Ir_{1-x}Pt_x)Te_2$ with different doping levels. We found that the type-II Dirac points lie around $E_F$ in the superconducting region, in which the bulk superconductivity has a maximum $T_c$ of $\sim 3$ K, which is accessible in many experimental techniques, making it possible to study the physical properties of Dirac fermions in the superconducting state.

High quality $(Ir_{1-x}Pt_x)Te_2$ single crystals were synthesized by Te flux method. The chemical compositions of the single crystals for each doping level were analyzed by EDX in a Hitachi S-4800 at an accelerating voltage of 15 keV. Longitudinal resistivity was measured in an Oxford He-3. High resolution ARPES measurements were performed at the "Dreamline" beamline of the Shanghai Synchrotron Radiation Facility, the I05 beamline of the Diamond Light Source, and the 13U beamline of the National Synchrotron Radiation Laboratory at Hefei, with the energy and angular resolutions set at 15 meV and 0.2°, respectively. All the samples were cleaved *in situ* in vacuum condition better than $5 \times 10^{-11}$ Torr and measured between 15–20 K. The first-principles calculations were performed to calculate the electronic structure of the 1T-structure $IrTe_2$ with the projector augmented wave (PAW) method as implemented in VASP package.[32] The tight-binding Hamiltonians of semi-infinite samples are constructed by the maximally localized Wannier functions (MLWFs) for all the Ir d and Te p orbitals, which are generated from the first-principles calculations. The (100) surface Green's functions of the semi-infinite sample are obtained using an iterative method.

Our first-principles calculations show that the electronic structure of 1T-structure $IrTe_2$ is similar to that of $PdTe_2$, except that the type-II Dirac points lie at 0.18 eV above $E_F$ in $IrTe_2$ (Figs. 1(f) and 1(g)). This is because the number of valence electrons of $IrTe_2$ is one less than that of $PdTe_2$. The calculated three-dimensional (3D) constant energy surface at the energy of the Dirac point in Fig. 1(e) shows that a small electron pocket touches with a large hole pocket at one point, which is an indication of a type-II Dirac point.

Like in other DSMs such as $Na_3Bi$ and $Cd_3As_2$, the Dirac points in $IrTe_2$ are associated with an inverted band structure. The band inversion in $IrTe_2$ occurs at $A$ point around $E_F$. With the protection of threefold-rotation symmetry, the band crossing along $\Gamma$–$A$ is stabilized, forming the type-II Dirac points. It is thus expected that there are topological surface states (TSSs) in the inverted band gap. Our calculations confirm the existence of TSSs around $\tilde{Z}$ point on (100) surface, as shown in Figs. 1(h) and 1(i). When the Dirac points reside around $E_F$, the TSSs cross $E_F$, forming a closed Fermi surface (FS) or a pair of Fermi arcs connecting to the surface projection of the Dirac points.

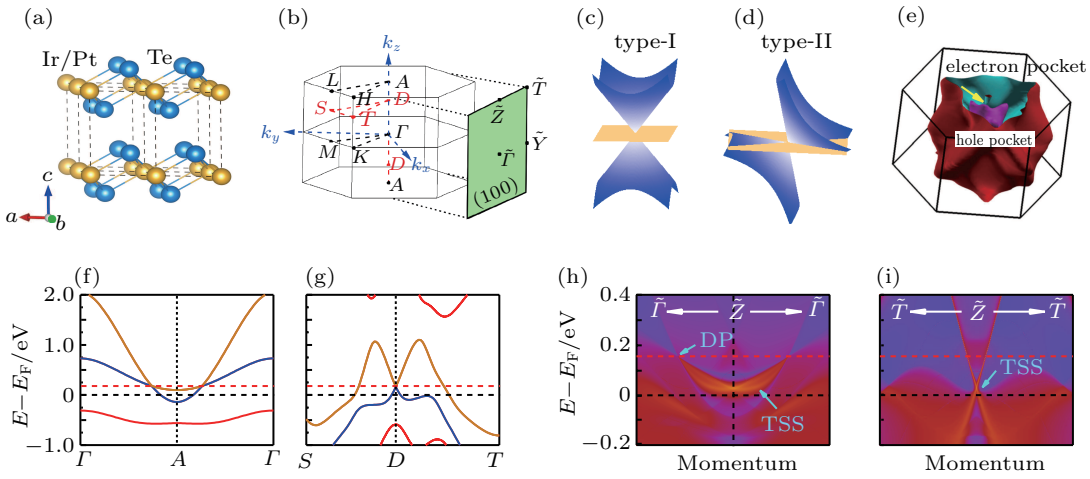



**Fig. 1.** Crystal structure and calculated electronic structures of $IrTe_2$. (a) Crystal structure. Orange and blue balls represent Ir and Te atoms, respectively. Black dashed lines indicate the unit cells. (b) Bulk BZ and projected (100) surface BZ. Red dots on $\Gamma$–$A$ line (labeled as $D$) mark the momentum locations of the Dirac points (DPs). (c) and (d) Schematic illustrations of type-I and type-II Dirac cones, respectively. (e) Calculated 3D Fermi contours at the energy of the Dirac points. (f) and (g) Calculated bands along $\Gamma$–$A$ and $S$–$D$–$T$, respectively, showing the band structures of the type-II Dirac points. (h) and (i) Projection of calculated band dispersions along $\tilde{Z}$–$\tilde{\Gamma}$ and $\tilde{Z}$–$\tilde{T}$, respectively, on (100) surface.

At room temperature, $IrTe_2$ is isostructural to the 1T-structure $PdTe_2$ (Fig. 1(a)) and a structural phase transition occurs at $\sim 250$ K.[33] By substituting Pt or Pd with Ir, the structural transition is rapidly suppressed, which is accompanied with the appearance of bulk superconductivity.[33] In the phase diagram of $(Ir_{1-x}Pt_x)Te_2$, $T_c$ reaches a maximum of $\sim 3$ K at $x = 0.05$ and gradually decreases as the Pt content is further increased.[33] We have synthesized a series of $(Ir_{1-x}Pt_x)Te_2$ single crystals with the nominal $x$ up to 0.5. Our resistivity measurements show bulk superconductivity for $x = 0.05$ and 0.1 (Fig. 2(a)). The EDX measurements confirm that the chemical compositions are consistent with the nominal ones (Fig. 2(b)). The variation of Ir and Pt contents is further confirmed by core level photoemission measurements in Fig. 2(d). The x-ray diffraction data recorded on the (001) plane in Fig. 2(c) show a reduction of the $c$ lattice as the Pt content increases. These results clearly indicate variation of the chemical compositions in the series of $(Ir_{1-x}Pt_x)Te_2$ samples that we have synthesized.

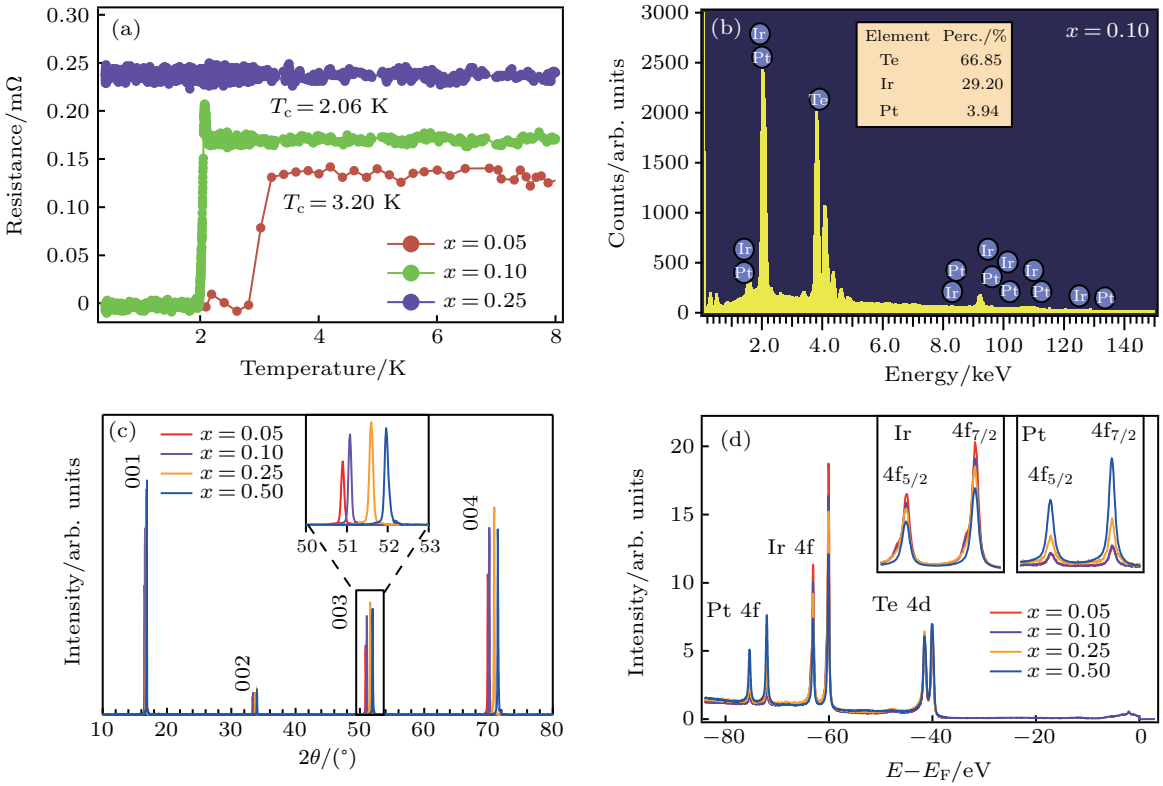



**Fig. 2.** Superconductivity and chemical compositions of $(Ir_{1-x}Pt_x)Te_2$. (a) Resistivity as a function of temperature for the samples with $x = 0.05$, 0.1, and 0.25. (b) Representative EDX spectrum of one sample with the nominal $x = 0.1$. The inset shows the percentages of Ir, Pt, and Te elements, which are consistent with the nominal composition. (c) XRD data recorded on the (001) plane of single crystals with $x = 0.05$, 0.1, 0.25, and 0.5. Inset shows that the (003) peak position gradually shifts to higher angles as the Pt content increases. (d) Shallow core level spectra of the samples with $x = 0.05$, 0.1, 0.25, and 0.5. The two insets show the evolution of intensities of the Ir 4f and Pt 4f peaks, respectively, with Pt substitution.

Since $IrTe_2$ and $PtTe_2$ have similar electronic structures, in which the Dirac points reside above and below $E_F$, respectively, the Dirac points can be tuned to $E_F$ in between them. To determine the composition that the Dirac points traverse $E_F$, we investigate the electronic structures of $(Ir_{1-x}Pt_x)Te_2$ with different doping levels using ARPES.

In Fig. 3, we display the ARPES results of the $x = 0.5$ sample. The calculations indicate that the Dirac point is located on the high-symmetry line $\Gamma$–$A$ along the $k_z$ direction. The ARPES experiments were preformed on the (001) cleavage surface. To search for the theoretically predicted Dirac point, we have measured the band structures in the $k_x - k_y$ planes at different $k_z$ values by varying the photon energy $h\nu$ of incident lights, according to the formula $k_z = \sqrt{2m(h\nu - \phi + V_0)}/\hbar$, where $\phi$ and $V_0$ are the work function and inner potential, respectively. Figures 3(a) and 3(b) show the ARPES intensity plots and corresponding curvature intensity plots, respectively, along $\Gamma$–$K$ direction taken with different photon energies. There is a Dirac-like band crossing at $-0.62$ eV in all the cuts in Figs. 3(a) and 3(b). The dispersions of the crossing bands do not change as $h\nu$ varies, indicating their surface origin. Notably, there is another Dirac-like band crossing at $-0.27$ eV in the cut measured at $h\nu = 22$ eV (Figs. 3(a3) and 3(b3)). The crossing bands become separated in the other cuts measured with different $h\nu$ values, indicating that the observed band crossing at $-0.27$ eV is a 3D Dirac point. The upper branch of the Dirac bands slightly shifts downwards as $h\nu$ increases from 22 eV to 24 eV, but rapidly

moves upwards as $h\nu$ decreases from 22 eV to 20 eV. It seems that the ARPES intensity plot in Fig. 3(a1) at $h\nu = 20$ eV exhibits an outline of an electron-like band below $E_F$, whereas the maximum intensity appears at $E_F$. This suggests that the electron-like band is actually located above $E_F$ while the intensity below $E_F$ could arise from a $k_z$ broadening, which is not negligible due to short mean free length of excited photoelectrons. The lower branch of the Dirac bands evolves into a M-shaped band as $h\nu$ increases from 22 eV to 24 eV and a hole-like band as $h\nu$ decreases from 22 eV to 20 eV. As indicated by the guide lines in Fig. 3(a), on the $\Gamma$–$A$ line, the bands monotonically shift downwards as $h\nu$ increases from 20 eV to 24 eV, which is consistent with the band calculations in Fig. 3(c). This means that the two crossing bands have the same sign in the slope along the $k_z$ direction, confirming that the Dirac point belongs to the type-II class. In addition to the Dirac band, we observed some other bands that cross $E_F$ at $k_y \sim \pm 0.4$ Å$^{-1}$ in Figs. 3(a) and 3(b). These bands do not change as $h\nu$ varies, and are not consistent with the calculated bulk bands, indicating their surface origin. Figure 3(d) shows the stacking ARPES intensity maps at several constant energies measured at $h\nu = 22$ eV. There is a circular FS around the Brillouin zone (BZ) center at $E_F$. The FS shrinks into a point at $-0.27$ eV and then grows into a circle again as the energy is further reduced. These results exhibit that the Dirac cone is nearly isotropic in the $k_x$–$k_y$ plane. The Dirac point of the $x = 0.5$ sample lies at 0.27 eV below $E_F$, which is closer to $E_F$ as compared with that in $PtTe_2$, where the Dirac point lies at $\sim 1$ eV below $E_F$.[20]

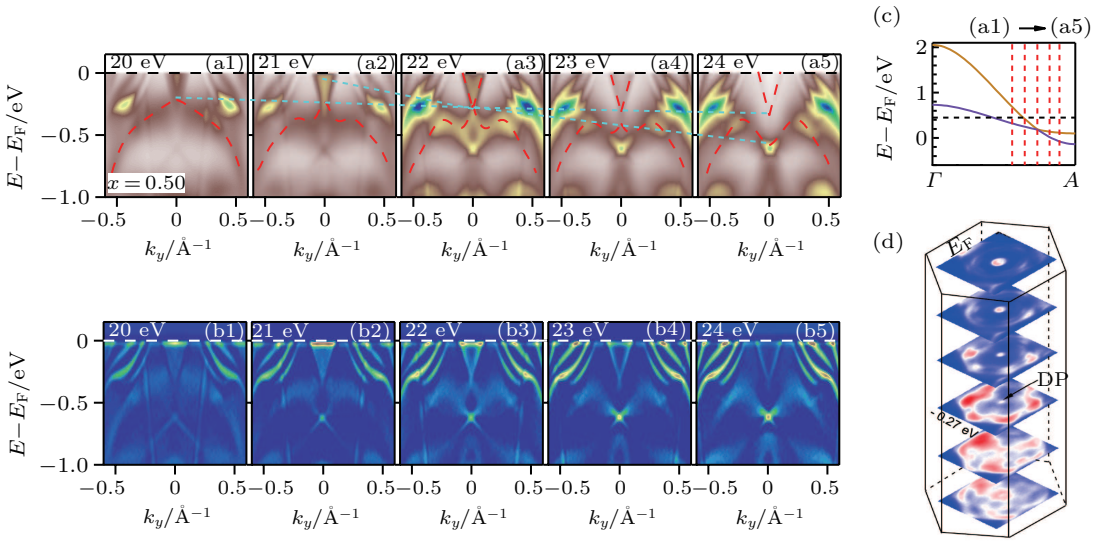



**Fig. 3.** Electronic structure of the $(Ir_{1-x}Pt_x)Te_2$ sample with $x = 0.5$. (a) ARPES intensity plots along the $\Gamma$–$K$ direction measured at (a1) $h\nu = 20$ eV, (a2) 21 eV, (a3) 22 eV, (a4) 23 eV, and (a5) 24 eV. (b) Corresponding curvature intensity plots. (c) Calculated band structure along $\Gamma$–$A$ of $IrTe_2$. We determine the chemical potential of $Ir_{0.5}Pt_{0.5}Te_2$ at 0.46 eV, as marked by the horizontal dashed line. The vertical dashed lines indicate the $k_z$ locations of the measured cuts in panels (a) and (b). The inner potential $V_0$ is set to be 13 eV and the lattice constant $c$ is determined to be 5.27 Å from the XRD data in Fig. 2(c). (d) Stacking plot of the ARPES intensity maps at several constant energies measured at $h\nu = 22$ eV, showing the nearly isotropic Dirac cone structure in the $k_x$–$k_y$ plane.

Next we focus on the evolution of band structures with the Pt substitution. For all the samples with different Pt contents, the measured band structures are almost consistent except for the energy positions relative to $E_F$. Figures 4(a)–4(d) show the band dispersions along $\Gamma$–$K$ direction measured at $h\nu = 22$ eV for the $(Ir_{1-x}Pt_x)Te_2$ samples with $x = 0.05$, 0.1, 0.25, and 0.5, respectively. The results clearly show that the Dirac point monotonically shifts upwards with the decrease of Pt content. In Fig. 4(g), we extract the Dirac bands from the ARPES data. Figure 4(h) plots the energy position of the Dirac point as a function of Pt content, from which we determine that the Dirac point passes through $E_F$ at $x \sim 0.1$. Figure 4(e) exhibits that two linearly dispersive bands for the $x = 0.1$ sample cross each other at $E_F$, leading to a point-like FS at the BZ center in Fig. 4(f). Figure 4(h) shows that the Dirac point does not shift linearly with the doping level. The energy shift becomes faster at higher Pt contents. The extrapolation based on the trend is consistent with the previous result in $PtTe_2$.[20] We note that if the calculated bands of $IrTe_2$ are rigidly shifted, the Dirac point would pass through $E_F$ around $x = 0.25$. This value is higher than that determined from our experimental results. This might be because the band calculations slightly overestimate the energy of the Dirac point in $IrTe_2$. The previous study in $PtTe_2$ shows similar inconsistency between calculation and experiment,[20] with a discrepancy of $\sim 0.1$ eV.

The substitution of Pt in $IrTe_2$ not only raises the chemical potential, making the type-II Dirac points approach $E_F$, but also introduces bulk superconductivity into the system. The maximum $T_c$ is up to $\sim 3$ K, which is accessible in many experimental techniques, making it possible to study the physical

properties of Dirac fermions in the superconducting state. The theoretical studies have proposed that topological superconductivity could be realized in type-I DSMs.[14,15] In contrast, there is no theoretical study on the superconducting properties of type-II DSMs. Moreover, as the Dirac points are close to $E_F$ in the superconducting $(Ir_{1-x}Pr_x)Te_2$, the associated topological surface bands may cross $E_F$, forming a pair of Fermi arcs or a closed FS. The proximity effect induces superconductivity in the TSSs below $T_c$, which could realize two-dimensional topological superconductivity on the surface of DSMs, in analogy to that in superconducting topological insulators. Our results suggest that the Pt-substituted $IrTe_2$ would be a promising material for studying the topological superconductivity of bulk and surface states in DSMs, and may find potential application in novel quantum devices such as quantum computers.

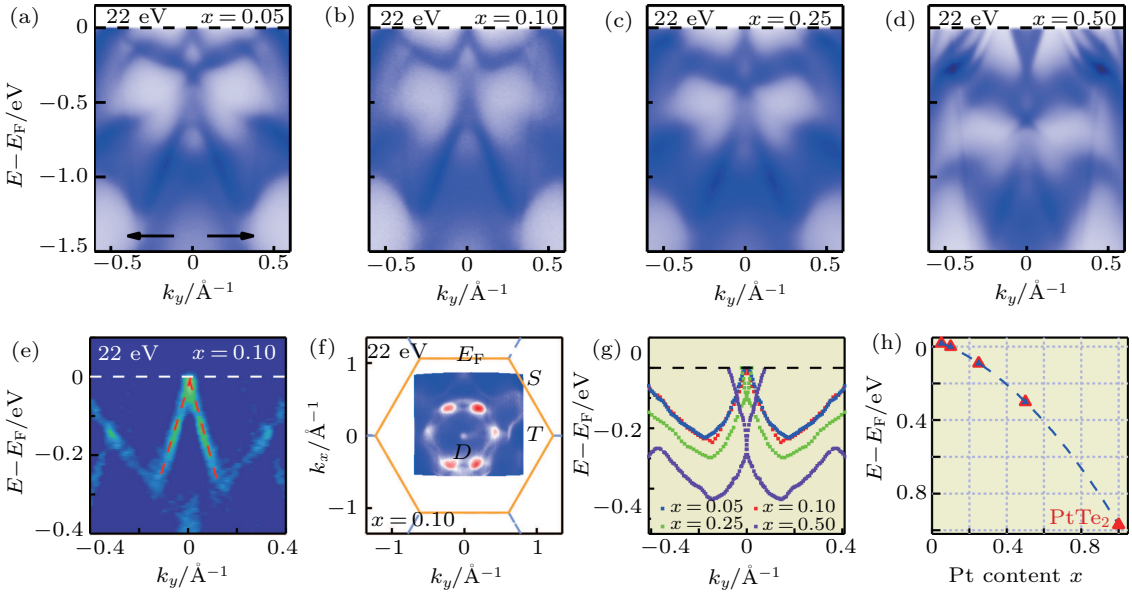



**Fig. 4.** Evolution of the Dirac point with Pt substitution. (a)–(d) Band dispersions along the $\Gamma$–$K$ direction measured at $h\nu = 22$ eV for the $(Ir_{1-x}Pt_x)Te_2$ samples with $x = 0.05$, 0.1, 0.25, and 0.5, respectively. (e) Second derivative intensity plot of the data in panel (b), showing the Dirac point lying at $E_F$ for the $x = 0.1$ sample. Dashed lines are guide to the eye, indicating the linear Dirac bands. (f) ARPES intensity map at $E_F$ measured at $h\nu = 22$ eV for the $x = 0.1$ sample, showing the point-like FS at the BZ center. (g) Extracted Dirac bands from the data in panels (a) to (d). (h) Energy of the Dirac point as a function of the Pt content. The energy for $PtTe_2$ is extracted from the previous result in Ref. [20].

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