Commensurate vs Incommensurate Charge Ordering near the Superconducting Dome in $Ir_{1-x}Pt_xTe_2$ Revealed by Resonant X-Ray Scattering

The electronic-structural modulations of $Ir_{1-x}Pt_xTe_2$ have been examined both by resonant elastic and inelastic X-ray scattering (REXS and RIXS) techniques. Charge-density-wave-like superstructures with wave vectors of Q = (1/5 0 −1/5), (1/8 0 −1/8), and (1/6 0 −1/6) are observed on the same sample of $IrTe_2$ at the lowest temperature. In contrast, superstructures around Q = (1/5 0 −1/5) are observed for doped samples. The superstructure reflections persist to higher Pt substitution than previously assumed. REXS and RIXS results indicate the importance of the Te 5*p* states rather than the Ir 5*d* states in the formation of the spatial modulation of these systems.

A charge-density-wave (CDW)-like structural transition was reported in the 5*d* transition-metal chalcogenide $IrTe_2$ at T_s ~ 280 K. This has attracted great interest because of the recent discovery of superconductivity in Pt- and Pd-substituted compounds [1–3]. With increasing Pt-substitution, the structural transition is suppressed, and a superconducting dome appears in the region of 0.04 ≤ x ≤ 0.12, indicating similar diagrams to those of other unconventional superconductors. Although numerous studies have followed these initial works, there is still no consensus on the mechanism of this structural transition. The phase transition of $IrTe_2$ is accompanied by the emergence of a superstructure lattice modulation, with wave vector $Q_{1/5}$ = (1/5 0 −1/5) as expressed in reciprocal lattice units in trigonal notation. The main elements are the Ir-Ir dimerization along the *a*-axis with period 5*a*, and the consequent distortion of the triangular Ir sublattice in the *a*-*b* plane, occurring together with a trigonal-to-triclinic symmetry reduction. Since in $IrTe_2$ the formal valence of Ir is +4, the Ir 5*d* electrons with t_{2g} configuration are the closest to the chemical potential, and they are thus expected to play a central role in the CDW. However, some previous studies indicated that the charge-transfer energy between Ir and Te is close to zero, and that the Te 5*p* states are also important for the low-energy physics, especially for their surface region. To revisit the superstructures in $Ir_{1-x}Pt_xTe_2$ and to clarify the relation between superconductivity and superstructural modulation, we studied the spatial ordering of electronic states by means of resonant elastic and inelastic X-ray scattering (REXS and RIXS) at both edges of Ir and Te.

Single-crystal samples of $Ir_{1-x}Pt_xTe_2$ (0.0 ≤ x ≤ 0.12) were prepared using a self-flux method. Resonant elastic X-ray scattering (REXS) at the Ir L_3 (2*p* to 5*d*) absorption edge in the hard X-ray region was performed

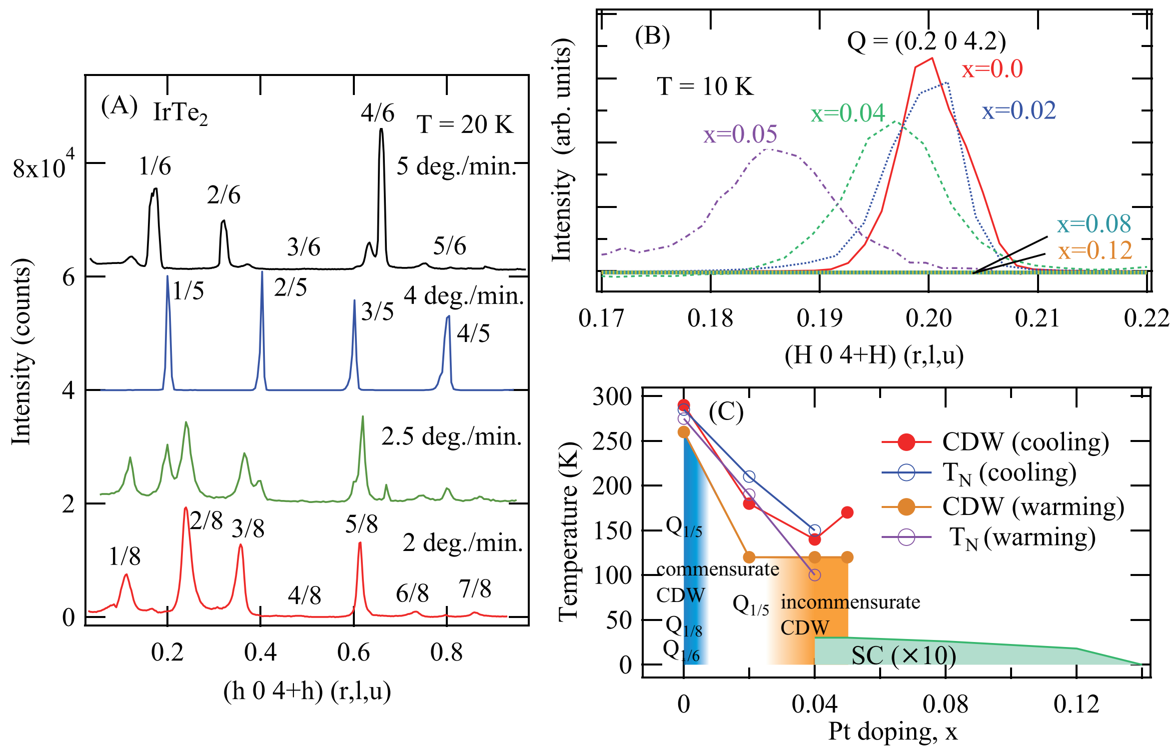

Figure 1: (A) Superstructure peaks for $IrTe_2$ at *T* = 10 K obtained at various cooling rates: 2, 2.5, 4, and 5 K/min. (B) Superstructure peaks of Q = (0.2 0 −4.2) for $Ir_{1-x}Pt_xTe_2$ (0.0 ≤ x ≤ 0.12). The cooling-warming rates were 4 K/min. (C) Phase diagram of $Ir_{1-x}Pt_xTe_2$.

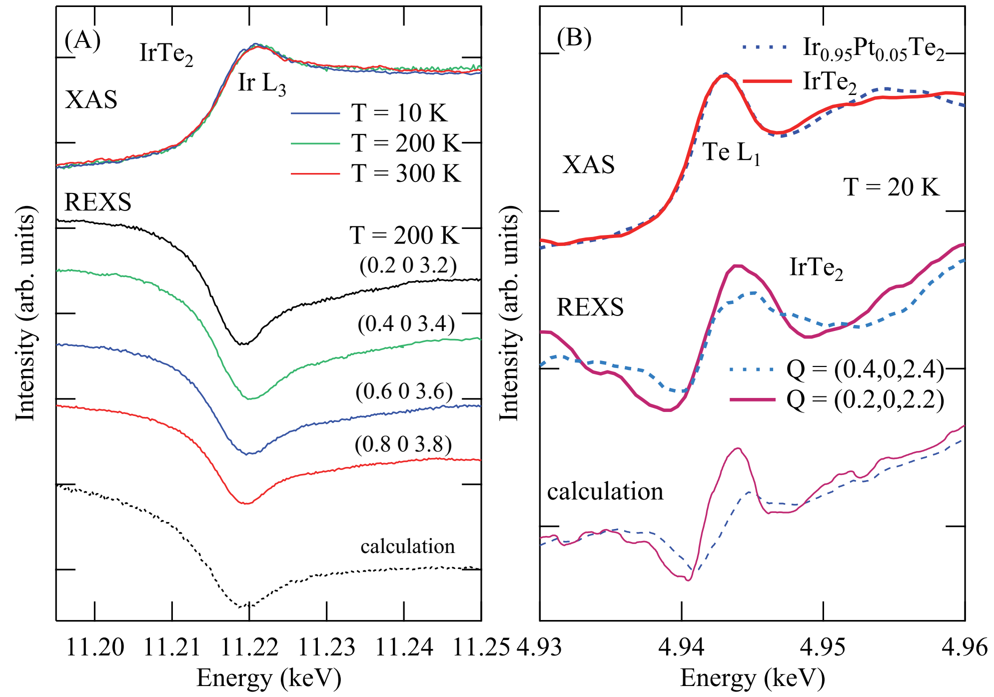

Figure 2: REXS and XAS spectra at the (A) Ir L_3 and (B) Te L_1 edges. The calculated spectra are also shown using the (A) lattice displacement for the Ir sites and (B) valence modulation model for the Te sites, respectively.

at the Photon Factory's BL-4C. REXS at the Te L_1 (2*s* to 5*p*) absorption edge was conducted at BL-22XU of SPring-8. On the other hand, resonant inelastic X-ray scattering (RIXS) at the Ir L_3 edges was carried out at BL-11XU of SPring-8. REXS at the Te $M_{4,5}$ (3*d* to 5*p*) edge in the soft X-ray region was performed at the REIXS beamline of the Canadian Light Source.

Figure 1(A) shows X-ray diffraction along (0 0 −4) to (1 0 −5) through the superstructure peaks for $IrTe_2$ (x = 0.0). The Q = (1/5 0 −1/5)-, (1/8 0 −1/8)-, and (1/6 0 −1/6)-type superstructures are observed for $IrTe_2$ at the lowest temperature of *T* = 10 K, whereas only the $Q_{1/5}$-type superstructures were reported in previous studies for these samples [4, 5]. The periods of the superstructures at low temperature strongly depend on the experimental protocol, or cooling rate. In contrast, incommensurate ordering peaks around Q = (1/5 0 −1/5) are observed for the doped samples of 0.02 ≤ x ≤ 0.05 at any cooling rate [Fig. 1(B)]. The superstructures coexist with the superconducting phase for $Ir_{1-x}Pt_xTe_2$ (x = 0.05) as shown in Fig. 1(C), suggesting that CDW persists to higher Pt substitution than previously thought. In addition, incommensuration of CDW is observed for x = 0.04 and 0.05 samples which coincide with the onset of superconductivity.

Surprisingly, the REXS and RIXS spectra for the Ir L_3 edge scarcely depend on the wavevectors, while the REXS line shape at the Te edges strongly depends on the Q position (Fig. 2), indicating that the spatial charge modulation only exists on the Te sites. Therefore, the charge modulation in the bulk regions resides in the Te orbitals rather than the Ir orbitals. The phase diagram re-examined in this work suggests that the CDW incommensurability in the Te sites will correlate with the emergence of superconducting states. $Ir_{1-x}Pt_xTe_2$ may harbor similar exotic phases for unconventional superconductors.

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K. Takubo[1], K. Yamamoto[1], Y. Hirata[1], H. Wadati[1], T. Mizokawa[2], R. Sutarto[3], F. He[3], K. Ishii[4], Y. Yamasaki[5], H. Nakao[6], Y. Murakami[6], G. Matsuo[7], H. Ishii[7], M. Kobayashi[7], K. Kudo[7] and M. Nohara[7] ([1]The Univ. of Tokyo, [2]Waseda Univ., [3]CLS, [4]QST, [5]NIMS, [6]KEK-IMSS-PF, [7]Okayama Univ.)