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Electronic Structure of TaC using Compton Scattering Technique

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The first ever Compton profile of polycrystalline tantalum carbide has been measured using 137Cs isotope. The results are compared with theoretical ionic Compton profiles and pseudopotential approach within Hartree-Fock theory. We have also computed the density of states using Hartree-Fock theory. The computed density of states confirms the metallic like behaviour of this compound. It is seen that the theoretical profile with ionic arrangement Ta+2C2- gives a better agreement with the experimental Compton profile.

Key Words: Compton scattering, Electronic structure, Band structure calculation

INTRODUCTION

Since last three decades, Compton scattering method has been applied to a variety of materials to access properties that are directly related to electronic wave functions[1]. It gives valuable information about the valence electrons through an analysis of Compton line shape. Within the impulse approximation, Compton profile $J(p_z)$ is a double integrated momentum density $\rho(\mathbf{p})$ over a plane perpendicular to the scattering vector ($\mathbf{k}$). Transition metal carbides exhibit broad and amazing electronic and chemical properties. TaC has attracted much attention due to its advantages as a barrier layer between copper and silicon wafers for the interconnections in integrated circuits. In earlier theoretical and experimental work, many researchers have attempted to establish a relationship between the basic crystal structure and electronic properties. For example, the electronic structure of TaC is investigated by Lavrent'ev *et al.*[2] with a full multiple scattering approximation within self-consistent semi-relativistic potential.

Zang *et al.*[3] have predicted the valence electronic structure of TaC from the empirical electronic theory. Sahnoun *et al.*[4] have calculated the electronic and optical properties of TaC using full potential linearised augmented plane wave (FPLAPW) method within the local density approximation (LDA). However, to

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our knowledge, there has been no calculation and measurement of the electron momentum density of TaC.

EXPERIMENT

The experiment was performed using our ^{137}Cs Compton spectrometer[5]. The ^{137}Cs source emits photons of energy 661.65 keV. The incident radiation, scattered inelastically on the polycrystalline sample (thickness 0.35 cm. and diameter 2.5 cm.) through 160º was detected by HPGe detector. The overall momentum resolution of the spectrometer was 0.38 a.u. The sample was exposed for about 299 h and an integrated intensity of 23 x 10^7 counts in the Compton region was obtained.

To deduce the true Compton profile from the raw data, several data corrections namely background, detector efficiency, sample absorption, instrumental resolution, Compton cross-section, etc. were applied[6]. The multiple scattering contribution to the Compton profile was removed by using a Monte Carlo technique[7]. Thereafter, the profile was normalised to free atom Compton profile of area 29.864 electrons in the momentum region 0-7 a.u.[8]

THEORY

Linear combination of atomic orbitals (LCAO) method, as implemented in the program CRYSTAL03[9], has been used in this study. It also allows the computation of the crystalline orbitals within the Hartree-Fock (HF) approach.

In the present calculations, the space group and the lattice constant of TaC were Fm-3m and a= 4.466 Å, respectively. Using the standard truncation criteria of the CRYSTAL03 code, the self-consistent-field (SCF) calculations have been performed at 120 **k** points in the irreducible Brillouin zone. The basis sets for C were all electrons[10], while for Ta large core pseudopotential (PP) basis sets[11] have been adopted due to non-availability of the corresponding all electrons basis sets.

RESULTS AND DISCUSSION

(a) Compton profiles

The difference curves between theoretical convoluted Compton profiles (calculated using different ionic arrangements and PP-HF schemes) and our experimental Compton profile are presented in Fig. 1. The theoretical profiles calculated for different ionic arrangements like $Ta^{+}C^{-}$ and $Ta^{+2}C^{-2}$ are obtained by transferring one and two electrons, respectively, from 6s orbital of Ta to 2p orbital of C. The shape of the difference profiles show that the theoretical Compton profiles are higher than the experimental for momenta below 1 a.u. which is in contrast to experimental data in momentum region 1 to 3 a.u. Usually

this kind of difference between the experiment and theory is attributed to exchange and correlation effects, which are not well incorporated in the present theory. Using χ^2 fitting it is seen that ionic profile with arrangement $Ta^{+2}C^{-2}$ gives a better agreement with the experiment in comparison to the other theoretical profiles.

(b) Density of states

In Fig. 2, we present the total and projected density of states (DOS) of TaC using PP-HF scheme. According to the projected DOS, it is clear that the lowest energy structure is mainly composed of C 2s states. The next structure below the Fermi level primarily contains electrons from C 2p and Ta 5d states. Broad

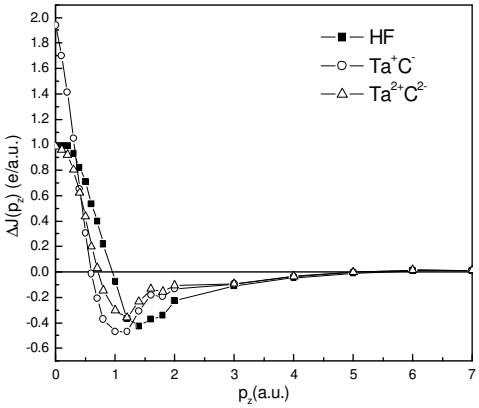

Fig. 1: Difference profiles (convoluted theory–experiment) of TaC.

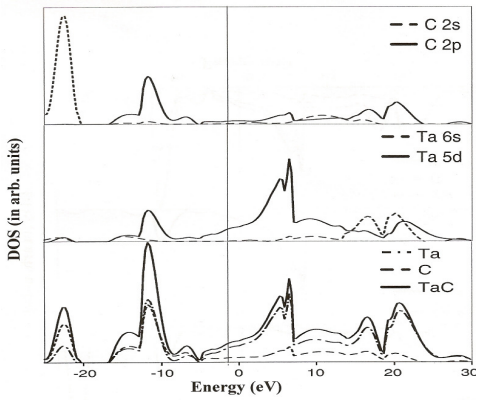

Fig. 2: Density of states (DOS) of TaC using PP-HF theory.

structures at about 5 eV above the Fermi level contains states arising from the Ta 5d states and to a lesser extends from the C 2p states. From the projected and total DOS, it is seen that the metallic contribution in TaC mainly arises from Ta 5d electrons.

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