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Anisotropy in the momentum density of tantalum

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Abstract

The [1 0 0], [1 1 0] and [1 1 1] directional Compton profiles of tantalum have been measured, at an intermediate resolution, with 661.65 keV γ-radiation and a well-defined scattering vector. We have also computed the Compton profiles within the framework of pseudopotential using CRYSTAL03 code of Torino group. The measured anisotropy is found to be smaller to that predicted by our pseudopotential computations and also the available augmented plane wave (APW) calculations. A comparison of the absolute theoretical and the experimental profiles shows clearly for the first time the importance of anisotropic electron–electron correction effect in the third series transition metals. A quantitative comparison between the experimental and theoretical directional difference profiles reveals some accurate information about the Fermi surface topology of tantalum.

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Keywords: Compton scattering; Electron–electron correlation; Fermi surfaces topology; Electron momentum density; Band structure calculations

1. Introduction

Since last two decades, the Compton scattering method has been applied to a variety of materials with the main objectives of testing the accuracy of band structure models of the ground state electron momentum density $\rho(\mathbf{p})$. Within the criteria of impulse approximation, the Compton profile $J(p_z)$ is projection of $\rho(\mathbf{p})$ and can be deduced from the spectral distribution of Compton scattered photons [1]. The anisotropic part of $\rho(\mathbf{p})$ can be deduced by forming direction difference profiles $\Delta J(p_z) = J_{hkl}(p_z) - J_{h'k'l'}(p_z)$. Here, z-axis is along the scattering vector. It has practical advantage of eliminating many systematic errors like multiple scattering, bremsstrahlung background contribution, etc. Because of the technological importance of tantalum, the accurate determination of its electronic properties has long been a goal for the theorists and experimentalists (see, for example, [2–10]). Papanicolaou et al. [11,12] have reported electron momentum densities and directional Compton profiles of Ta using electron wave functions obtained from their self-consistent APW band structure calculations within the local density formalism. In these calculations, mass–velocity relativistic corrections have been included while the spin–orbit coupling has been neglected.

Previously, Compton scattering studies of Ta had been made on single crystals [13] using 60 keV γ-rays and on polycrystalline materials with 60 keV [14–16] and 661.65 keV γ-rays [17]. Due to extremely poor statistics of directional data reported by Chang et al. [13] viz. ±0.09 e/a.u. (one standard deviation $\pm\sigma$) at $p_z = 0$ of each profile, their data on single crystals were of no use and inadequate for probing subtle correlation effects and anisotropies in momentum densities, and Fermi surfaces topology. In case of data using 60 keV γ-rays [13–16], the extraction of the individual profile is more uncertain because of the failure of impulse approximation for the K, L-shell electrons and relatively poor resolution (0.60 a.u.) in comparison to that employing high-energy gamma-ray sources (like ^{137}Cs) or synchrotron radiations.

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It was therefore decided to make an accurate single crystal study of Ta at an intermediate resolution 0.38 a.u. using a high energy (661.65 keV) ^{137}Cs Compton spectrometer. To compare the anisotropies in the momentum densities, we have also computed the directional Compton profiles using pseudopotential (PP) with Hartree–Fock (HF) and PP with density functional (DF) approaches embodied in CRYSTAL03 code [18].

In the work reported below, the Compton profiles along [1 0 0], [1 1 0] and [1 1 1] directions of Ta have been measured in an extensive series of experiments. The absolute profiles and the difference profiles are compared with the available APW theory [12] and our CRYSTAL03-based computations. The experimental data are interpreted in terms of Fermi surface topology and energy bands of Ta.

2. Experiment

For the present measurements, a single crystal of Ta (length 30 mm, dia. 12 mm) grown along [1 1 0] direction was procured from Metal Crystals and Oxides Ltd., UK. Other two principal directions [1 0 0] and [1 1 1] were marked on the cylindrical shaped single crystal and accordingly the crystal was cut for these directions using spark erosion method. These crystal pieces of about 12 mm dia. were properly grinded with abrasive paper, chemically itched and polished. The orientations of each polished surface was confirmed through their Laue diffraction patterns. A description of our first Indian 740 GBq (20 Ci) ^{137}Cs Compton spectrometer (resolution 0.38 a.u. FWHM) used in the present measurements has been reported elsewhere [19,20]. Only the salient features are given here. The spectrometer (Fig. 1(a)) is built in a steel chamber of dimension $1150 \times 350 \times 400$ mm^3 divided into two parts namely, sample chamber in the left-hand side and source chamber in the right-hand side. γ-Rays of energy 661.65 keV were scattered by the sample at an angle of $160 \pm 0.6°$. The spread in the scattering angle ($\Delta\theta$) was estimated using a Monte-Carlo simulation which simulates the path of incident and scattered photons through the sample. Our simulation for 99,999 photons as applicable to a sample size of 12.0 mm dia. and source collimator size of 8.0 mm dia. shows a Gaussian FWHM of 1.25°. This small spread was helpful in orienting the samples (vertically) so that the scattering vector was along the direction of the sample. To keep the almost identical conditions for scattering, the samples were screwed in a sample holder consisting of cylindrical ring of brass (dia. 23 mm). The diameter of incident beam at the sample position was kept to be about 10 mm. Compton profiles were accumulated over a total period of 837 h for the combined investigations of all the principal directions. For each orientation, an integrated intensity was found to be $2–2.5 \times 10^7$ counts in the Compton profile region. We refer to Fig. 1(b)–(d) for raw data of our measurements. The background spectrum was measured in the same manner but without any sample and subtracted from the Compton spectrum after scaling the

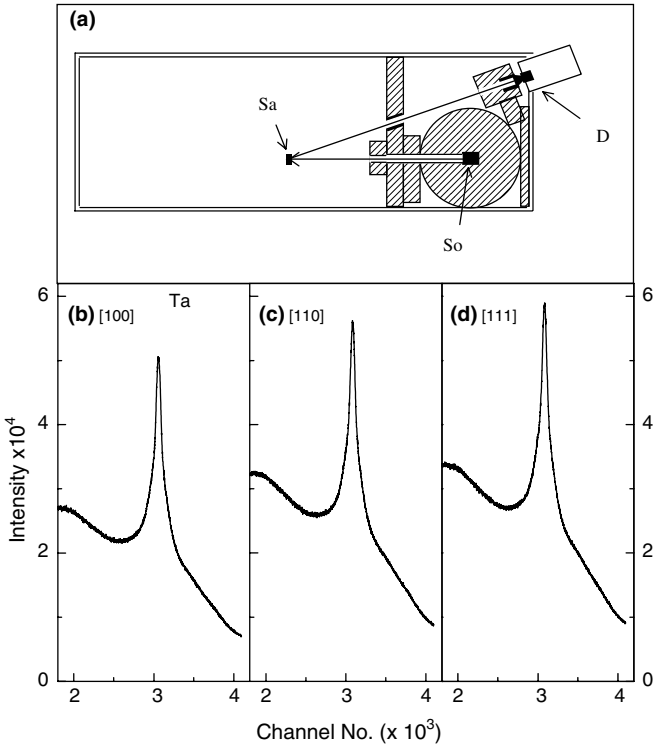

Fig. 1. (a) Layout of 20 Ci ^{137}Cs Compton spectrometer. The ^{137}Cs source (23 mm dia. and 36 mm length) is contained in a cylindrical lead block (220 mm height and 220 mm dia.) with rotatable collimator-cum-beam shutter (dia. 50 mm and length 140 mm). The distances between source (So)–sample (Sa) and sample (Sa)–detector (D) are 382 mm and 548 mm, respectively. The shaded region shows the lead shielding (50–70 mm). The lead shielding all around the steel chamber is not shown. (b) Energy distribution of photons from a ^{137}Cs source scattered at $160 \pm 0.6°$ from single crystal of Ta for [1 0 0] direction. In the low energy side, a systematic higher intensity is due to energy dependent HPGe detector efficiency. (c) Same as (b) except the direction, which is [1 1 0] and (d) same as (b) except the direction which is [1 1 1].

measurement time of the raw data and the corresponding background. The scattered radiations were energy analysed using a high purity Ge detector (Canberra, Model GL0210P) and associated electronics like spectroscopy amplifier (Canberra, Model 2022), analog-to-digital converter (Canberra, Model 8701) and 4096 channel analyzer (Canberra, Accuspec B). The Compton profiles were further corrected for several systematic corrections like instrument resolution (limited to stripping off the low energy tail), sample absorption, detector efficiency, Compton scattering cross-section and multiple scattering, etc. using the computer code of Warwick group [21,22]. Finally, the experimental profiles were normalised to have an area of 26.91 electrons, which is equal to that of the free atom profile [23] in the momentum range 0–7 a.u.

3. Theory

3.1. Pseudopotential calculations

Valence electrons Compton profiles for Ta (bcc) have been computed using the pseudopotential (PP) with