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Electronic structure of Ni_2TiAl: Theoretical aspects and Compton scattering measurement

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ABSTRACT

In this paper, we report electron momentum density of Ni_2TiAl alloy using an in-house 20 Ci ^{137}Cs (661.65 keV) Compton spectrometer. The experimental data have been analyzed in terms of energy bands and density of states computed using linear combination of atomic orbitals (LCAO) method. In the LCAO computations, we have considered local density approximation, generalized gradient approximation and recently developed second order generalized gradient approximation within the frame work of density functional theory. Anisotropies in theoretical Compton profiles along [1 0 0], [1 1 0] and [1 1 1] directions are also explained in terms of energy bands.

1. Introduction

Compton scattering is a well known technique to scrutinize the electronic properties of materials [1,2]. In Compton scattering technique one measures the double differential scattering cross-section, which is directly related to the Compton profile (CP), $J(p_z)$, of the studied material. The $J(p_z)$, is the one dimensional projection of electron momentum density $n(\boldsymbol{p})$ along the scattering vector (using z-axis) and is given by

$$J(p_z) = \iint n(\boldsymbol{p}) dp_x\, dp_y \propto \frac{d^2\sigma}{d\Omega\, d\omega_2} \quad (1)$$

where p_z is the component of electron momentum along the z-axis and ω_2 is the energy of photon after scattering. Eq. (1) is valid only within the impulse approximation (IA) [3], which dictates that the energy transferred to electron by the photon is very high and interaction between photon and electron is impulsive. Therefore, to a good approximation, within the IA a moving and bound electron can be considered as a free and at rest.

Ni_2TiAl belongs to a group of ternary intermetallics Heusler alloys which exhibits an ordered $L2_1$ crystal structure. Recently Heusler alloys have gained renewed interest due to their unique transport, electronic and magnetic properties [4,5]. Their electronic structure can range from metallic to semi-metallic or semiconducting behavior. To our knowledge there have been very few studies on the electronic properties of Ni_2TiAl. Boettinger et al. [6] have studied the disordered trapping in Ni_2TiAl at the solidification interface using pulse laser melting and rapid resolidification. The cohesive properties and electronic structures of Ni_2XAl (X=Ti, V, Zr, Nb, Hf and Ta) in both the $L2_1$ and the B2 phases have been determined by Lin and Freeman [7] using all-electron total energy self-consistent linear muffin-tin orbital within local density approximation (LDA). Rocha et al. [8] have measured the low temperature (3.2–10.3 K) specific heat and electronic structure of Ni_2TAl (T=Ti, Zr, Hf, V, Nb and Ta). Kellou et al. [9] have presented the structural, electronic and magnetic properties of X_2AlX' (X=Fe, Co, Ni and X'=Ti, Cr) using generalized gradient approximation (GGA) within the full potential linearized augmented plane wave method. To our knowledge, till date, there is no data on the electron momentum density of Ni_2TiAl. The aim of the present work was multifold:

(a) to measure the γ-ray based CP of Ni_2TiAl using ^{137}Cs Compton spectrometer, at the best possible resolution (b) to compute the theoretical CPs using density functional theory (DFT) and compare them with the experimental CP (c) to compute the energy bands and density of states using LCAO–DFT calculations and (d) to interpret the theoretical anisotropies in CPs in term of energy bands.

2. Sample preparation and experiment

Bulk polycrystalline sample of Ni_2TiAl was prepared by arc-melting of highly pure (4N, make: Aldrich, USA) metals namely, Ni, Ti and Al, in argon environment. During the sample preparation, oxygen contamination was avoided by evaporating the Ti inside the vacuum chamber before the arc-melting. To get the

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homogeneity in the sample, the melting process was repeated four times. Thereafter, the ingot was crushed and annealed at 800 °C for 5 days. During the melting process, weight-loss of the sample was less than 1/2%. Stoichiometric composition of the sample was re-confirmed using energy dispersive analysis of X-rays. The X-ray diffraction data confirmed the $L2_1$ phase (space group $Fm\bar{3}m$) of the sample.

To measure the CP, we have used an indigenous 20 Ci ^{137}Cs Compton spectrometer. The details of the spectrometer are reported earlier [10]. Only salient features of the experiment are mentioned here. For the Compton measurements, radiations of energy 661.65 keV were incident on sample and scattered photons, at $160 \pm 0.6^{\circ}$, were energy analyzed using HPGe detector (Canberra, USA made). To get uniform sample dimensions, the ingot was crushed to fine powder and a pellet of thickness 0.22 cm and diameter 2.0 cm was prepared. The scattered radiations from the sample were accumulated for a period of 360 h. The raw data were processed for several corrections namely, background, instrumental resolution, Compton scattering cross-section, multiple scattering correction, etc. [11,12]. For the measurement of background, the spectrometer was run for almost 4 days after removal of the sample from the sample holder. After the data reduction, the experimental profile was normalized to free atom CP area ($40.61e^{-}$) [13] in the momentum region p_z= 0–7 a.u. An overall resolution of spectrometer was 0.34 a.u. (Gaussian full width at half maxima (FWHM)). The stability of the system was checked using two weak calibrators, namely ^{57}Co and ^{133}Ba isotopes.

3. Computational details

To interpret our experimental CPs, we have computed theoretical CPs, energy bands and DOS of Ni_2TiAl using CRYSTAL09 code [14,15] of Torino group. It is worth mentioning that CRYSTAL09 package includes various DFT schemes like LDA, GGA and second order GGA (SOGGA) within linear combination of atomic orbitals (LCAO). In the LCAO method, one electron crystalline orbitals are the linear combination of Bloch functions given as [14,15]

$$\Phi_i(\mathbf{r},\mathbf{k}) = \Sigma\Sigma a_{\mu i}(\boldsymbol{k})\Phi_\mu(\boldsymbol{r}-\boldsymbol{A}_\mu-\boldsymbol{g})\exp(i\boldsymbol{kg}) \quad (2)$$

The Bloch functions, which are the solutions of one electron equations, are built from local atoms by the linear combination of n_G individual Gaussian-type functions.

In the present calculations, the structure of Ni_2TiAl was taken as fcc, having the space group 225 ($Fm\bar{3}m$). Accordingly, the atomic positions were taken as: Ni at (¼, ¼, ¼) and ($^3/_4$, $^3/_4$, $^3/_4$); Ti at (½, ½, ½) and Al at (0, 0, 0). The value of lattice parameter a was taken as 5.854 Å, as derived from our XRD data. In the present computations, we have taken exchange potential prescribed by Dirac–Slater [16] for LDA and Wu–Cohen [17] for GGA. The correlation function suggested by Perdew–Burke–Ernzerhof [18] has been used for LDA, GGA and SOGGA. The all-electrons Gaussian basis sets for Ni, Ti and Al were taken from ⟨www.tcm.phy.cam.ac.uk/~mdt26/basis_sets⟩. The self consistent field calculations have been performed with 455$\boldsymbol{k}$ points in irreducible Brillouin zone. The tolerance on the total-energy convergence in the iterative solution of the Kohn–Sham equations was set to 10^{-6} Hartree. BROYDEN scheme [19] has been used to achieve a fast convergence.

4. Results and discussions

4.1. Energy bands and density of states

In Fig. 1((a) and (b)), we have presented the energy bands and DOS of Ni_2TiAl using DFT–GGA. Except some fine structure, overall shape of energy bands computed using DFT–LDA/SOGGA is almost similar to that of DFT–GGA, therefore we have not shown the bands and DOS using DFT–LDA/SOGGA computations. Except some fine structures, our DOS are found to be in reasonable agreement with the earlier data reported by Rocha et al. [8]. Crossovers of Fermi level (E_F) by some energy bands, confirm the metallic-like character of the alloy. Some important features deduced from energy bands and DOS (Fig. 1(a) and (b)) are summarized below:

1. The energy bands which are dispersed in the region −2.7 to −10 eV are mainly due to 3p and 3s electrons of Al. Since these bands are well below the E_F, it is concluded that the 3sp electrons of Al play a small role in deciding the electronic properties of Ni_2TiAl.

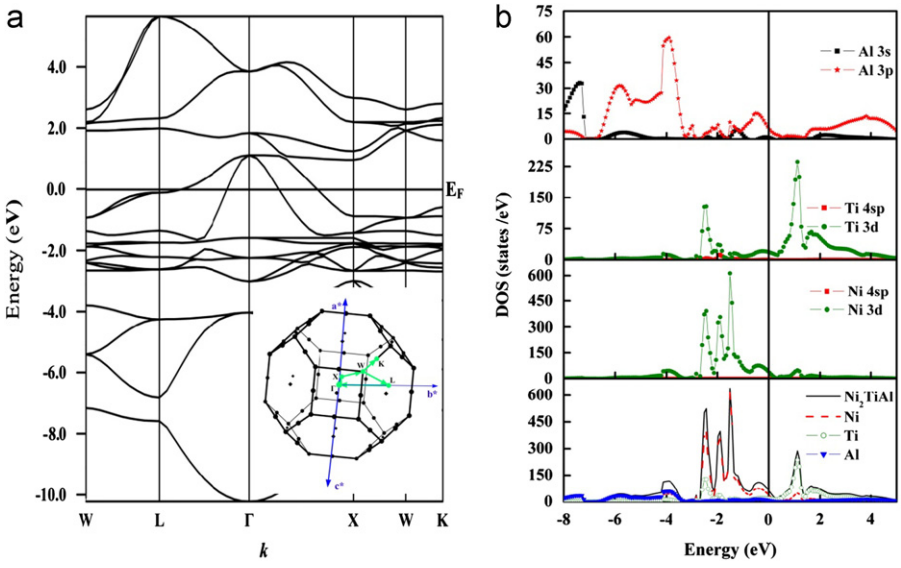

Fig. 1. (a) Energy bands of Ni_2TiAl along the high symmetry directions of the first Brillouin zone (BZ) using DFT–GGA scheme and (b) Partial and total DOS of Ni_2TiAl. The BZ corresponding to cubic (space group $Fm\bar{3}m$) structure is shown in inset of (a).