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Room temperature Compton profiles of conduction electrons in α-Ga metal

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Abstract. Room temperature Compton profiles of momentum distribution of conduction electrons in α-Ga metal are calculated in band model. For this purpose, the conduction electron wave functions are determined in a temperature-dependent non-local model potential. The profiles calculated along the crystallographic directions, (100), (010), and (001) are found to be nearly isotropic. This conclusion is in reasonable agreement with experimental observations.

Keywords. Near isotropy; Compton profiles; α-gallium; room temperature.

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1. Introduction

It is well-known that gallium metal in both monoclinic and orthorhombic phase is not a simple metal. Experimental results on electrical resistivity [1] and theoretical results on Compton profiles [2] in orthorhombic α-Ga, for example, confirm this. Both resistivity and Compton profiles in the latter metal show strong anisotropic behaviour at low temperature. On the other hand, the Compton profile data obtained experimentally [3] for α-Ga and theoretically for β-Ga [4] show near-isotropic behaviour at room temperature. Experimental data on Compton profiles in β-Ga are not available at room temperature for comparison with theoretical results. Similarly, no attempt has been made heretofore to calculate the room temperature Compton profiles in α-Ga metal for comparison with available experimental data.

In the present work, the room temperature Compton profiles of electron-momentum distribution due to conduction electrons are calculated in band model. For this purpose, the conduction electron wave functions and their energies are determined in the model potential method using temperature-dependent non-local model potential. Contributions to model potential form factor from the effect of lattice expansion and lattice vibration have been evaluated. The profiles are calculated for three crystallographic directions, namely, (100), (010) and (001). The results show near isotropy of the Compton profiles in reasonable agreement with experimental observations [3].

The paper is divided into four sections. In §2, theory of Compton profiles is briefly outlined. Results and discussion are presented in §3. Conclusions are summarised in §4.

2. Theory

A detailed theory of Compton profiles of electron-momentum distribution in metal is given in ref. [4]. Here we have described it briefly. In impulse approximation [5], the profile along the crystallographic direction $\hat{k}$ is given by

$$J_{\hat{\kappa}}(q) = \frac{\Omega}{(2\pi)^3}\int \mathrm{d}^3 p \rho(\vec{p})\delta(q - \vec{p}\cdot\hat{\kappa}), \tag{1}$$

where Ω is the volume of the unit cell and $\rho(\vec{p})$ is the momentum probability distribution function of electrons. In case of conduction electron the latter is given by the Fourier transform of its wave function Ψ. In model potential method of energy band calculation, the wave function for the tth band is expressed [2] as

$$\Psi_t(\vec{g},\vec{r}) = \sum_{\vec{K}} C_t(\vec{g},\vec{K})[PW(\vec{g}+\vec{K},\vec{r}) - \sum_i \langle\phi_i(\vec{g})|PW(\vec{g}+\vec{K})\rangle\phi_i(\vec{g},\vec{r})], \tag{2}$$

where $\vec{g}$ is a wave vector in the Brillouin zone (BZ), $\vec{K}$ is a reciprocal lattice vector and other notations have their usual meaning [2]. The energies $E_t(\vec{g})$ and the coefficients C_t are obtained as solutions of the standard secular equation [2].

In constructing the secular equation, the temperature-dependent model potential has been evaluated as follows: The contribution to form factor from lattice expansion is evaluated using the atomic volume Ω appropriate to the temperature in question. The contribution from the effect of lattice vibration, on the other hand, has been included through Kasowski mechanism [6]. According to the latter, the form factors at temperature T are related to those at 0 K by

$$\langle\vec{g}+\vec{K} \mid V_m \mid \vec{g}\rangle_T = \mathrm{e}^{-W(K,T)}\langle\vec{g}+\vec{K} \mid V_m \mid \vec{g}\rangle_0, \tag{3}$$

where the exponential is the square-root of the standard [11] Debye–Waller factor.

The occupied conduction electron states whose Fourier transform contributes to $\rho(\vec{p})$ have been obtained by determining the Fermi energy. Using $\rho(\vec{p})$ in eq. (1) and carrying out some simplifications, it has been shown [2] that

$$J_{\hat{\kappa}}(q) = \frac{\mathrm{d}F_{\hat{\kappa}}(q)}{\mathrm{d}q}, \tag{4}$$

where the expression for the function $F_{\hat{\kappa}}(q)$ is given in ref. [2].

3. Results and discussions

The lattice parameters corresponding to room temperature [7] of α-Ga metal are: $a = 8.5406$ atomic units (a.u.), $b = 8.5495$ a.u., $c = 14.4787$ a.u., $u = 0.0785$, and $v = 0.1525$. Its primitive cell contains four atoms. The point-symmetry of orthorhombic structure [8]

is characterised by D_{2h} group. The model potential used in the present work is the one derived by Animalu and Heine [9]. However, their form factors [9], as listed in [10], refer to a temperature different from room temperature. So, in the present calculation the form factors have been determined afresh corresponding to the room temperature. As already mentioned in §2, the form factor consists of contributions from two sources, namely, lattice expansion and lattice vibration. The former has been evaluated using the atomic volume corresponding to the room temperature. The latter is evaluated in Kasowski formalism [6] using eq. (3). From the latter equation it is evident that to evaluate the vibrational contribution to the form factor at temperature T one needs to know the form factor at 0 K. In the present work, the latter has been evaluated using the atomic volume extrapolated [7] to 0 K. The Debye–Waller factor has been calculated from the standard expression derived in [11].

Energies and conduction electron wave functions have been determined at 972 representative $\vec{g}$ points suitably chosen in one-eighth irreducible part of BZ. For this purpose, the irreducible zone is divided into six equal parts by slicing its g_x axis by perpendicular planes. Each plane is further divided into 972 sub-parts consisting of 486 rectangles and 486 triangles. The centroids of rectangularpipeds and the triangular prisms formed between successive planes are chosen as the representative $\vec{g}$ points, at which energies and eigenfunctions have been determined. To each $\vec{g}$ point so chosen, a weighting factor $w_{\vec{g}}$ in proportion to the volume it represents is assigned. The total number of $\vec{g}$ points in the whole BZ at which energies and wave functions are available is thus 7776. The sum total of weight factors of 7776 $\vec{g}$ points is equal to 1, as it should.

At each representative $\vec{g}$ point, the model wave function is expanded in a plane wave basis consisting of 24 plane waves corresponding to the shortest $|\vec{g}+\vec{K}|$ lengths at that $\vec{g}$ point. With this choice of basis functions the energies of the occupied bands have converged to be within 0.001 Ryd. The conduction electron wave function ψ_t for the tth band is obtained by orthogonalising the corresponding model wave function to all the core states in the metal. For this purpose, each core state of the metal is represented by a single tight binding function formed out of the corresponding atomic core wave function. In the present work, the atomic core functions used are the Hartree–Fock wave functions of Clementi [12]. As ψ_t consists of a plane wave (PW) part and a tight-binding (TB) part, $F_{\hat{\kappa}}$ function consists of contributions resulting from their combinations, such as, PW–PW, TB–TB and the hybrid (namely, PW–TB and TB–PW). Expressions for these contributions are given in ref. [2]. The net $F_{\hat{\kappa}}$ functions have been determined in the present work for (100), (010) and (001) crystallographic directions in exactly the same way as in ref. [2]. The first derivatives of $F_{\hat{\kappa}}$ with respect to q are determined for each value of q chosen in a uniform mesh with mesh size 0.1 a.u. These derivatives directly give the profile function $J_{\hat{\kappa}}(q)$.

The choice of 972 $\vec{g}$ points in the irreducible part of BZ is primarily dictated by consideration of the convergence of $J_{\hat{\kappa}}(q)$ with respect to variation in the number of g points. The present calculation is repeated for four sets of representative $\vec{g}$ points, namely, 108, 216, 588 and 972. With the last set the profiles are found to be well-converged.

The results of $J_{\hat{\kappa}}(q)$ are summarized in table 1 for all the three directions. While the fifth column in the table lists J_{av}, the average values of data in columns 2–4, the experimental results [3], J_{LLM} of Lengeler, Lasser and Mair (LLM) are presented in the sixth column. The profile functions have also been plotted in figure 1. It is evident from table 1 and figure 1 that the profile function $J_{\hat{\kappa}}(q)$ from the present calculation has nearly the same

Table 1. Present results of Compton profiles of electron-momentum distribution of conduction electrons in α-Ga at room temperature. $J_{100}(q)$, $J_{010}(q)$, $J_{001}(q)$, denote the profiles along (100), (010) and (001) directions respectively. $J_{\rm av}$ denotes the average profile and $J_{\rm LLM}(q)$ denotes the experimental profile of ref. [3]. All the quantities are expressed in atomic units.

q	$J_{100}(q)$	$J_{010}(q)$	$J_{001}(q)$	$J_{\rm av}$	$J_{\rm LLM}(q)$
0.0	2.534	2.542	2.443	2.507	2.40±1.5%
0.1	2.470	2.536	2.406	2.470	2.31
0.2	2.332	2.295	2.301	2.309	2.17
0.3	2.149	2.120	2.145	2.138	2.05
0.4	1.899	2.016	2.021	1.979	1.84
0.5	1.601	1.739	1.841	1.727	1.60
0.6	1.262	1.417	1.425	1.368	1.27
0.7	0.830	0.933	0.908	0.890	0.94
0.8	0.398	0.384	0.473	0.418	0.56
0.9	0.140	0.087	0.205	0.144	0.21
1.0	0.063	0.053	0.096	0.070	0.15±10%
1.1	0.046	0.042	0.060	0.049	0.15
1.2	0.035	0.035	0.046	0.038	0.07

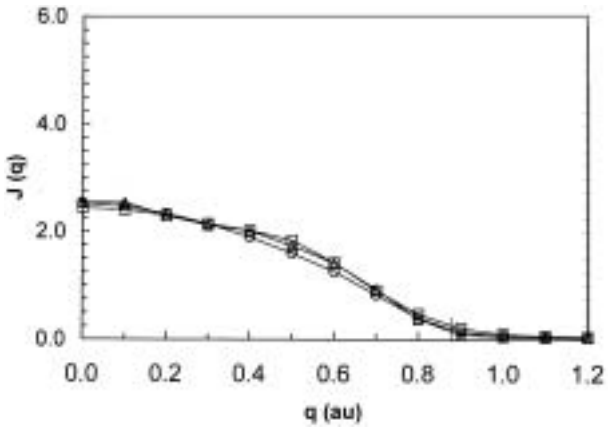

Figure 1. Variation of Compton profile $J(q)$ in α-Ga metal at room temperature. Solid lines are labelled by symbols ◯, △ and □ which represent band model results along (100), (010) and (001) directions respectively. The vertical line at $q = 0.88$ a.u. marks the position of Fermi momentum.

value along the three directions. That is, the profile is nearly isotropic. Experimental results for $J_{\hat{\kappa}}(q)$ at room temperature are available from the work of LLM [3] for the crystallographic directions, (100), (010) and (001). In their case [3] the results (column 6 of table 1) obtained are very nearly independent of the direction suggesting near-isotropy of Compton profiles. Thus, the present results, as far as near-isotropy is concerned, are in reasonably good agreement with experiment [3].

However, there is a quantitative difference between the present result and experiment for individual q. This may be seen from columns 5 and 6 of table 1. Up to $q = 0.6$ a.u., the values of J_{av} are higher than J_{LLM} and differ between 4.4% and 7.9% of the latter. For higher momentum, that is, for $q > 0.6$ a.u., the results of J_{av} are lower than J_{LLM} and the percentage of discrepancy in this region of momentum is somewhat large. This variation in behaviour may be explained as follows:

In the lower momentum (0.0–0.6 a.u.) region, the dominant contribution to $\rho(\vec{p})$ arises from ψ at large r. In model potential method of band calculation the wave function in large r region is determined more accurately than in the near-region. Hence $\rho(\vec{p})$ determined in the present method is more accurate for lower momentum than for higher momentum. This accounts for small discrepancies between J_{av} and J_{LLM} for low q and higher discrepancies for large q.

The accuracy of the present results has also been checked [2] by evaluating the integral

$$\int_{-\infty}^{\infty} J_{\hat{\kappa}}(q)\mathrm{d}q = N, \tag{5}$$

where N is the number of conduction electrons per atom. In the present work the calculated value for N is 3.07 for all the three crystallographic directions. This result is very close to 3, the actual number of conduction electrons per atom in gallium metal.

It may be remarked here that due to the combined effects of electron–electron interaction and periodic potential in the metal, one expects [13] a non-zero percentage of conduction electrons, n_{F} to have momentum larger than p_{F}, the Fermi momentum. In the free-electron system n_{F} is exactly zero. In the present work n_{F} is calculated by integrating $J_{\hat{\kappa}}$ between p_{F} and ∞ and is found to be about 2.86% (average) of the total number of conduction electrons. This is smaller than 6.7%, the result obtained by LLM [3] in the renormalised free atom (RFA) model. The discrepancy between the two may be attributed partly due to forced localisation of atomic functions in the RFA model by artificial truncation of 4s and 4p atomic functions of gallium atom at the Wigner–Seitz cell boundary. The localisation of electron in the r space amounts to delocalisation of its momentum in the momentum space. That is, $J_{\hat{\kappa}}(q)$ in RFA model is more stretched out in q-space than it is in the band model where no such artificial truncation of wave function is made. It is this effect which makes n_{F} in RFA model larger than it is in the band model.

The change of Compton profiles of α-Ga metal from anisotropy at low temperature [2] to near-isotropy at room temperature, as the present results suggest, may be understood in terms of weakening of model potential with increase of temperature. As temperature increases, there is a tendency that the strength of the model potential is reduced as may be seen from eq. (3). This, in turn, drives the conduction electron system in the metal towards free-electron-like distribution. The latter is known to have its Compton profile nearly isotropic.

4. Conclusions

The room temperature Compton profiles of electron-momentum distribution in α-Ga metal calculated in the band model is found nearly isotropic and in agreement with similar conclusions arrived at from experiment. The percentage of conduction electrons with momentum larger than p_{F} is about 2.86%.

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