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Compton profiles of electron momentum distribution in β-Ga

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Abstract. We report for the first time the Compton profiles of electron momentum distribution in β-gallium calculated along the crystallographic directions (100), (110) and (111). The conduction electron states for this purpose are determined in the energy band calculations using model potential. The core states, on the other hand, are represented each by a single tight-binding function. The results show that the Compton profiles are nearly isotropic with very little directional dependence, which is suggestive of a free-electron-like distribution of the conduction electrons in this system. The latter conclusion is in close confirmity with similar conclusions drawn in augmented plane wave (APW) calculation of energy bands and the derived Knight-shift results in β-Ga.

Keywords. Compton profiles; β-gallium; electron momentum distribution.

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1. Introduction

The scattering of photons by electrons in a solid provides useful information as to the electron momentum distribution in these systems. Measurements of Compton profiles in scattering experiments are carried out using either x-ray [1,2] or γ-rays [3]. In free-electron-like metals [4], the Compton profiles are nearly isotropic whereas in transition metals [5, 6] there are significant deviations from the isotropy of the profiles.

There are various models using which Compton profiles are calculated. Two such early models are the free-atom-model and the renormalized-free-atom (RFA) model [3]. In the latter model atomic valence electron wave functions, truncated at the boundary of the Wigner–Seitz cell and renormalized over the cell volume, are used to describe the conduction electron states in the solid. While this model [3] is better than the free-atom-model, it is less realistic to describe the band states in metals. On the other hand, the model in which the conduction electron states are determined in band calculations is expected to yield more accurate profiles than the free-atom and RFA models. Calculations of Compton profiles using the band models [5, 6] are now available for a number of metals.

In the present work we have calculated Compton profiles in β-Ga using band model. There are two crystallographic phases of gallium metal. These are α-Ga and β-Ga. The former has orthorhombic structure while the latter crystallizes in monoclinic structure. In α-Ga, results of Compton profiles are available both from experiment [7] and theory [7]. The calculations [7] are, however, made using the less realistic RFA model [3].

In contrast to this, results for Compton profile in β-Ga, to our knowledge, are available neither from experiment nor from theory. This lack of result motivated us to take up β-Ga in the present work. There are also other motivations. In α-Ga, the results of Compton profile are found to be very nearly isotropic. We wanted to see how strong is the anisotropy of the Compton profiles in β-Ga. Further, it is well-known that electron correlation effects introduce a finite discontinuity with a high momentum tail at the free-electron Fermi momentum P_F in the electron momentum distribution. In taking up β-Ga, we wanted to study how strong is the eletron correlation effect in this system by calculating n_F, the number of conduction electrons with momentum larger than P_F. Lastly, the ready availability of band wave functions in β-Ga from our recent calculation [8] of electric field gradient in this system prompted us to take up β-Ga instead of α-Ga in the present investigation.

As to the various approximations used in the present calculation, we wish to state that apart from using a non-local model potential, we have made use of two other approximations namely, the impulse approximation [9] for the scattering of photons by electrons and the single tight-binding approximation for representing the core states in the solid.

In the impulse approximation [9, 10], it is assumed that the interaction of the photon with the electron until the latter is ejected is a constant potential. This approximation holds if the photon is of very short wavelength and the binding energy of the electron is much smaller than the photon-energy. In using X-ray photons, both these conditions are well satisfied for the conduction electrons. But for the core electrons, especially the innermost 1s electrons, it does not hold good. In spite of this, the impulse approximation has been used in the past to predict Compton profiles in reasonably good agreement with experiment.

In the single tight-binding approximation, each core state in solid is described by a single tight-binding function constructed from the corresponding atomic Hartree–Fock core wave function. This is a standard practice followed in many band calculations and the approximation is reasonably good as the core states in solid retain their atomic character to a large extent.

The paper is organized as follows. In § 2, we briefly describe the theory for the calculation of the Compton profiles. Section 3 summarizes the results of the present calculation and makes a comparative analysis with the available results in α-Ga. Conclusions are summarized in § 4.

2. Theory

We start with the standard expression [5]

$$J(q) = \frac{\Omega}{(2\pi)^3} \int d^3p\, \rho(p)\, \delta(q - \mathbf{p}.\hat{k}) \tag{1}$$

for the scattering cross-section of a photon by the electrons in a metal, which holds good in impluse approximation [9, 10]. In eq. (1), Ω denotes the volume of the unit cell in the solid, $\mathbf{p}$ is the initial momentum of the electron, ρ is the momentum distribution function and $\hat{k}$ is the momentum transfer to the electron by the photon.

The momentum distribution function is constructed from the eigenfunctions of electrons in momentum space. The latter are related to co-ordinate eigenfunctions of the electron by a Fourier transform.

In the present work the co-ordinate eigenfunction of each core electron is represented by a single tight-binding function. The conduction electron wave functions, on the other hand, are obtained in the standard band calculation [8] using non-local model potential.

These wave functions are used in arriving at expressions for the Compton profiles of electron momentum distribution from core and conduction electrons. The details of derivation of these expressions are given in the work of Panigrahi and Mohapatra [6]. We simply quote those results here. The fact that the core states are completely filled and each one is represented by a single tight-binding function makes core contribution to $J_{\hat{k}}(q)$ isotropic, that is independent of $\hat{k}$ direction. It has been shown [6, 11] that the contribution to $J(q)$ per atom from the core electron is

$$J(q) = \frac{2}{\pi} \sum_{nl} (2l+1) \int_q^\infty I_{nl}^2(p) p \, dp, \tag{2}$$

where n, l are the principal and orbital quantum numbers of the atomic core state and I_{nl} stands for the integral

$$I_{nl}(p) = \int_0^\infty j_l(pr) \, p_{nl}(r) \, r dr. \tag{3}$$

In eq. (3), j_l is the spherical Bessel function of order l and p_{nl} is the radial part of the atomic core wave function which is normalized according to

$$\int_0^\infty p_{nl}^2(r) dr = 1. \tag{4}$$

The conduction electron contribution to $J_{\hat{k}}(q)$, on the other hand, is expressed [6] as

$$J_{\hat{k}}(q) = \frac{d F_{\hat{k}}(q)}{dq}, \tag{5}$$

where

$$\begin{aligned} F_{\hat{k}}(q) = & \sum_{\mathbf{g}(\text{irred})} w_g \sum_{\mathbf{K}} \left(\sum_{\alpha} \theta[q - (\mathbf{g}+\mathbf{K}) \cdot \alpha \hat{k}] \right) \left(\sum_t |C_t(\mathbf{g}+\mathbf{K})|^2 \right. \\ & - \frac{16\pi}{\Omega} S(\mathbf{K}) \sum_t C_t^*(\mathbf{g}+\mathbf{K}) \sum_{\mathbf{K}} C_t(\mathbf{g}+\mathbf{K}') \sum_{nl} (2l+1) \\ & \left. \times I_{nl}(|\mathbf{g}+\mathbf{K}|) \, I_{nl}(|\mathbf{g}+\mathbf{K}'|) \, P_l(\cos\gamma_{\mathbf{K}\mathbf{K}'}) \right) \\ & + \left(\frac{8\pi}{\Omega} \right)^2 \sum_{\mathbf{g}(\text{irred})} \sum_{\mathbf{K}_s} S^2(\mathbf{K}_s) \sum_{\alpha} \theta[q - (\mathbf{g}+\mathbf{K}_s) \cdot \alpha \hat{k}] \\ & \times \sum_t \left(\sum_{\mathbf{K}} C_t^*(\mathbf{g}+\mathbf{K}_s) \, S(\mathbf{K}) \right. \\ & \left. \times \sum_{nl} (2l+1) \, I_{nl}(|\mathbf{g}+\mathbf{K}|) \, I_{nl}(|\mathbf{g}+\mathbf{K}_s|) \, P_l(\cos\gamma_{\mathbf{K}\mathbf{K}_s})^2 \right). \end{aligned} \tag{6}$$

Here w_g is the weighting factor assigned to the sample g point in the irreducible part of the BZ, θ is the standard step function, P_l is the Legendre polynomial of order l, $\gamma_{\mathbf{KK}'}$ is the angle between the vectors $\mathbf{g}+\mathbf{K}$ and $\mathbf{g}+\mathbf{K}'$, α is the point-symmetry operators and $S(K)$ is the structure factor of the lattice.

The quantities $J_{\hat{k}}(q)$ for any $\hat{k}$ satisfies [6] the relation

$$\int_0^\infty J_k(q)\,\mathrm{d}q = n, \tag{7}$$

where n is equal to half the number of electrons per atom. Equation (7) may be used as a check for the accuracy of the band as well as core contribution $J_{\hat{k}}(q)$.

3. Results and discussion

β-Ga crystallizes in monoclinic structure with two atoms per primitive cell. The lattice parameters available [12] at 248 K are $a = 2{\cdot}766$ Å, $b = 8{\cdot}053$ Å, $c = 3{\cdot}332$ Å and $\beta = 92°\,03'$. The band calculation [8] has been carried out using the non-local model potential of Appapillai and Williams [13]. These model potentials include correction from exchange and correlation effects. Besides, these have quite successfully explained a number of electronic properties of metals, including Knight shift, electric field gradients, density of states etc. In view of the success of these model potentials we have used them in the present calculation of electron momentum distribution in β-Ga. In constructing the band wave function, the atomic core functions used are those of Clementi [14]. In order to carry out the g sum in eq. (6), 176 sample g points are suitably chosen in 1/4th irreducible part of the BZ as described in previous work [8]. The energies of the occupied bands have converged within 0·001 Ryd with the choice of 23 plane waves of the shortest wave vector $|\mathbf{g}+\mathbf{K}|$ in the expansion of pseudo wave function. The results of the present band calculation [8] compare very well with those from APW calculation [15] in regard to shapes of bands, Fermi energy and density of state at Fermi energy.

We now discuss the result of $J_{\hat{k}}(q)$ as obtained in the present work. We begin with the contribution from the core electrons. The atomic core functions of Clementi [14] are used in evaluating the integral in eq. (3). The radial integrals are evaluated using the standard 5-point numerical integration procedure after generating the integrand at 201 mesh points, chosen in a logarithmic r scale, $r_n = r_1 \exp[-(n-1)h]$ with $r_1 = 0{\cdot}00097$ atomic units (a.u) and $h = 0{\cdot}05$ a.u. The farthest distance up to which the integration is carried out has the value $r_{201} \approx 21{\cdot}45618$ a.u.

As the core states are localized, the corresponding momentum distribution is delocalized. Hence, in order to satisfy eq. (7) for the core electrons, one needs to integrate $J(q)$ in eq. (7) over a large distance in the q scale. This is particularly true for $1s$ electrons whose contribution to $J(q)$ varies very slowly with q. Since impulse approximation [9] is not good for the $1s$ electrons, one may ignore them for the purpose of checking the result in eq. (7.) By ignoring the $1s$ electrons, the integrated result from eq. (7) turns out to be approximately 12·63 which is close to 13, the number of $2s$–$3d$ core electrons in half an atom of Ga. The remaining discrepancy is attributed to $2s$ and $2p$ electrons whose contributions to $J(q)$ vary so slowly that unless the integration in eq. (7) is carried over sufficiently long distance in q scale, their full contribution to the integral will not be obtained.

Now coming to the band contribution $J_{\hat{k}}(q)$, we note that the band states being delocalized, their momentum distributions are expected to be fairly localized. Using eq. (6) we have evaluated the function $F_{\hat{k}}(q)$ for the three crystallographic directions of $\hat{k}$, namely, (100), (110) and (111). In carrying out the sum in eq. (6) over **g** in the irreducible part of the BZ, we have used 176 sample **g** vectors. In order to study the dependence of the result on the number of sample points, the calculation was repeated at a smaller set of **g** vectors than 176. The result shows that the change due to this effect is less than 1%. The derivative of $F_{\hat{k}}(q)$ with respect to q, which has been evaluated numerically provides the corresponding $J_{\hat{k}}(q)$. In order to study the convergence of $F_{\hat{k}}(q)$ with respect to the number of reciprocal lattice vectors **K** and $\mathbf{K}_s$ occurring in eq. (6), we split the contribution F_k into two parts; one arising from the plane wave and hybrid (i.e. plane wave-tight binding combination) part of band wave function and the other from the tight-binding part. The first three lines in eq. (6) give the plane wave and hybrid contribution while the remaining terms give the tight-binding contribution. The former

Table 1. Present results of Compton profiles $J_{\hat{k}}(q)$ of β-Ga along the directions (100), (110) and (111). The results are expressed in atomic units.

	Core contribution	Band contribution		
q	$J(q)$	$J_{100}(q)$	$J_{110}(q)$	$J_{111}(q)$
0·0	3·620	2·599	2·617	2·609
0·1	3·618	2·342	2·328	2·350
0·2	3·614	2·801	2·813	2·808
0·3	3·606	2·198	2·202	2·200
0·4	3·592	2·011	2·012	2·011
0·5	3·573	1·483	1·483	1·478
0·6	3·548	1·122	1·124	1·117
0·7	3·517	1·013	1·015	1·011
0·8	3·479	0·536	0·544	0·538
0·9	3·433	0·132	0·142	0·138
1·0	3·381	0·010	0·017	0·018
1·2	3·253	0·023	0·026	0·029
1·4	3·097	0·019	0·017	0·019
1·6	2·921	0·021	0·012	0·013
1·8	2·726	0·019	0·009	0·009
2·0	2·527	0·007	0·008	0·007
2·2	2·325	0·002	0·007	0·006
2·4	2·131	0·003	0·005	0·005
2·6	1·943	0·004	0·004	0·004
2·8	1·769	0·006	0·003	0·003
3·0	1·610	0·006	0·001	0·002
3·5	1·271	0·000	0·000	0·000
4·0	1·019	0·001	0·000	0·000
4·5	0·829	0·000	0·000	0·000
5·0	0·687	0·000	0·000	0·000
6·0	0·496	0·000	0·000	0·000
7·0	0·373	0·000	0·000	0·000
7·2	0·353	0·000	0·000	0·000

contribution involving the delocalized plane wave is found well converged with 23 reciprocal lattice vectors **K**. But the tightbinding contribution which involves more localized wave functions than plane waves required 315 reciprocal lattice vectors $\mathbf{K}_s$ for the result to converge. In order to test the accuracy of the band contribution to $J_{\hat{k}}(q)$, we have used eq. (7). For all the three directions of $\hat{k}$, the integral in eq. (7) gives $n \approx 1{\cdot}499$, which is very close to exact result of 1·5 conduction electrons in half an atom of Ga.

We present our results of $J_k(q)$ in table 1 for the three directions, (100), (110) and (111). Since the core contribution in the present work is isotropic, it has the same value for all the three directions. It is evident from table 1 that the Compton profiles are very nearly isotropic. Therefore, in order to economise figure space, we have preferred to plot the results of $J_{\hat{k}}(q)$ for only one direction, namely, (100) and this is shown in figure 1. But to show the direction dependence, we have plotted the difference functions, $J_{110}(q)-J_{100}(q)$ and $J_{110}(q)-J_{111}(q)$ in figures 2 and 3 respectively. These figures also display near isotropy property of $J_{\hat{k}}(q)$. The maximum value of the difference is about 0·68% of $J(0)$ due to conduction electrons. This result is not surprising because the band structure results, such as, Fermi energy and density of state at Fermi energy in β-Ga suggest that the conduction electrons in this metal are mostly free-electron-like. The results from APW [15] calculation as well as the Knight-shift data [15] also lead one to this conclusion. The structures seen in the profiles of conduction electron contributions (figure 1) are similar to those noted in the work of Rath *et al* [5].

In a free-electron gas, n_F, the number of electrons with momentum larger than Fermi momentum P_F is exactly zero at absolute zero temperature. But for an interacting

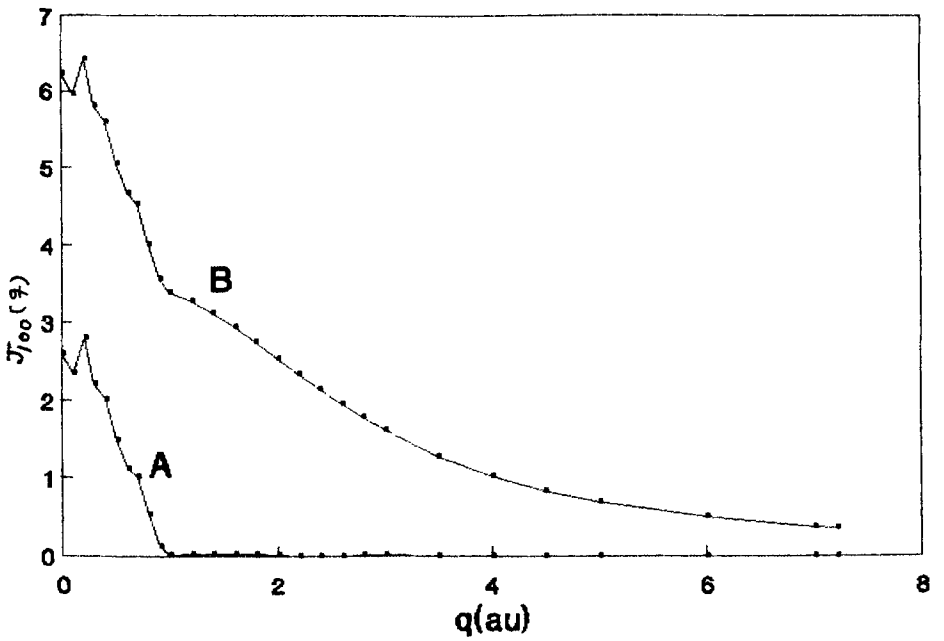

Figure 1. $J_k(q)$ for β-Ga with $\hat{k}$ along (100) direction: curve A, band contribution; curve B, total contribution (core + band).

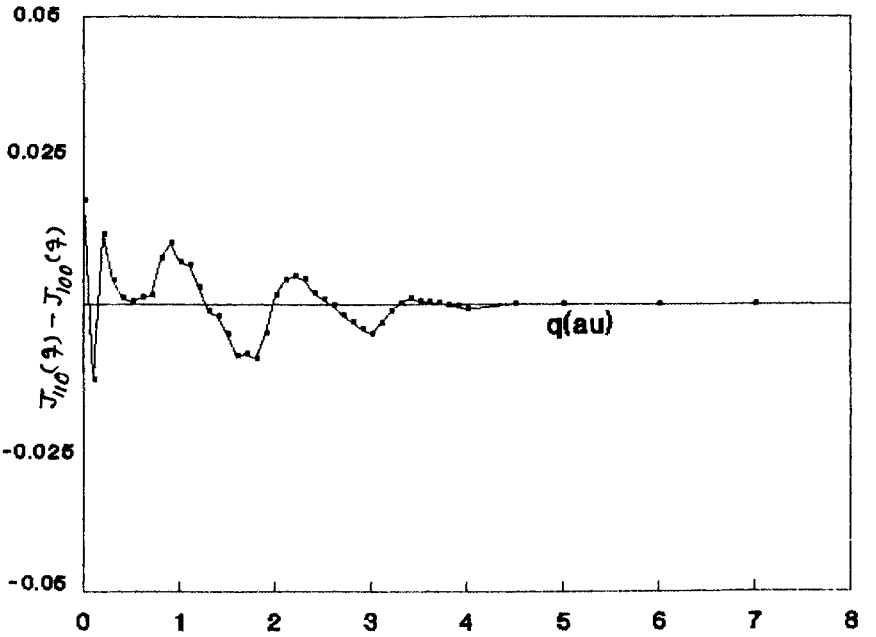

Figure 2. Difference profile $J_{110}(q)$–$J_{100}(q)$ in β-Ga.

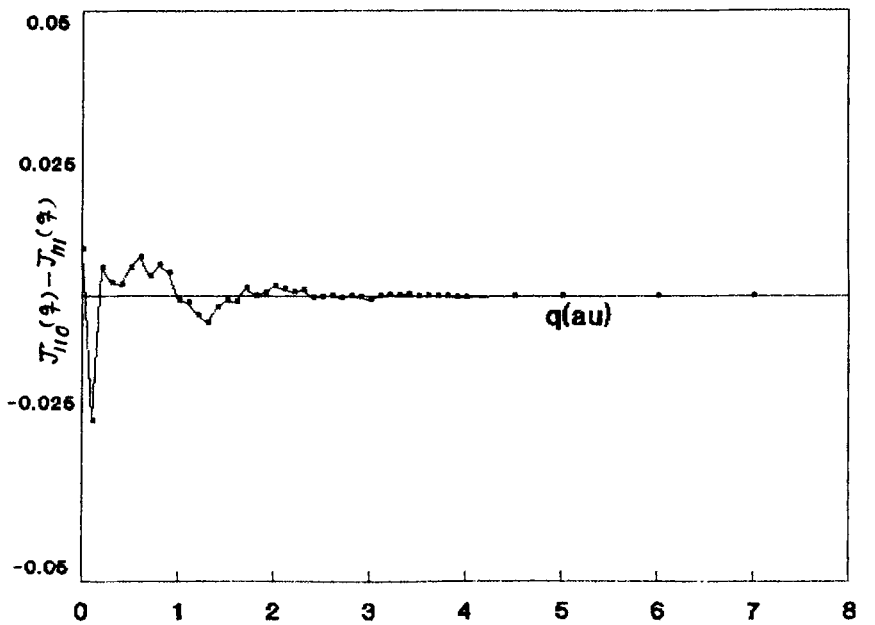

Figure 3. Difference profile $J_{110}(q)$–$J_{111}(q)$ in β-Ga.

electron system, it has been shown by Luttinger [16] that there are non-zero number of electrons with momentum larger than P_F. In the present work, P_F is found equal to 0·885 a.u. By setting the lower limit equal to P_F in (7), we have calculated n_F in β-Ga.

Table 2. Comparison of the present results in β-Ga of the conduction electron contribution to Compton profiles with those in α-Ga. All the results are expressed in atomic units. The data in α-Ga taken from ref. [7] are isotropic.

q	$J(q)$ in α-Ga	$J_{100}(q)$ in β-Ga
0·0	2·4 ± 1·5%	2·599
0·1	2·31	2·342
0·2	2·17	2·801
0·3	2·05	2·198
0·4	1·84	2·011
0·5	1·60	1·483
0·6	1·27	1·222
0·7	0·94	1·013
0·8	0·56	0·536
0·9	0·21	0·132
1·0	0·15 ± 10%	0·010
1·1	0·15	0·030
1·2	0·07	0·023
1·3	0·07	0·021
1·4	0·08	0·019
1·5	0·08 ± 20%	0·019
1·6	0·06	0·021
1·7	0·05	0·020
1·8	0·06	0·019
2·0	0·04	0·007
2·2	0·02	0·002
2·4	0·00	0·003

The result shows that there are about 2·2% such electrons. We remark that n_F so obtained includes both the band structure and electron correlation effects, the latter through the non-local model potential whose parameters are determined by fitting with the experiment.

Results of Compton profile from experiment or from other works are not available to compare with the present results. The present results in β-Ga are however compared with those available [7] in α-Ga. The calculated results [7] in α-Ga obtained in RFA model [3] agree well with experiment [7]. The latter [7] obtained for (100), (010) and (001) directions show near isotropy. The comparisons of the present result with those in α-Ga are made for two reasons. Firstly, to see if the present results are of the same order of magnitude as those in α-Ga. Secondly, to find if the present results are also very nearly isotropic as those in α-Ga. For this purpose we list the present results as well as those for α-Ga in table 2. The data in this table show that the present results are of the same order of magnitude as those in α-Ga and that they are nearly isotropic. Further we have also compared n_F in both the systems. While in α-Ga, about 6·7% of conduction electrons have momentum larger than P_F, it is about 2·2% in β-Ga. We note that any quantitative comparison of numbers in columns 2 and 3 of table 2 will not be meaningful as these are obtained in two different methods.

The higher value of n_F in α-Ga may be attributed to the renormalized conduction electron wave functions in RFA model [7]. Since the valence electron wave functions, 4*s* and 4*p* in α-Ga are truncated and renormalized within the Wigner Seitz cell in RFA model, it forces localization of the valence electron distribution. As a consequence, its momentum distribution becomes more delocalized than in the band model where no truncation of wave functions is made. A delocalized momentum distribution $J_\xi(q)$ is more stretched out in q-space than a localized distribution and is therefore expected to give a larger n_F as is obtained in α-Ga.

4. Conclusion

The results of Compton profiles in β-Ga are calculated for the first time in band model along the crystallographic direction (100), (110) and (111). The results show near isotropy suggesting the conduction electrons in this metal to be very nearly free electron-like, a conclusion in perfect agreement with band structure results from APW calculations and the Knight Shift data in β-Ga. About 2·2% of conduction electrons are found to have momentum larger than P_F.

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