Why is the bandwidth of sodium observed to be narrower in photoemission experiments?

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Abstract

The experimentally predicted narrowing in the bandwidth of sodium is interpreted in terms of the non-local self-energy effect on quasi-particle energies of the electron liquid. The calculated self-energy correction is a monotonically increasing function of the wave number variable. The usual analysis of photo-emission experiments assumes the final state energies to be like those of the nearly-free-electron-like model and hence it incorrectly ascribes the non-local self-energy correction to the final state energies, thus leading to a seeming narrowing in the bandwidth.

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Sodium is the most suitable metal for testing the validity of electron liquid at metallic densities since it is almost free from band structure complications. Angle-resolved photo-emission experiments Plummer and his co-workers pointed out the possibility that the occupied energy-band of sodium might be remarkably narrowed in comparison with the usual value of nearly-free-electron theory. A number of authors attempted to interpret the band narrowing of sodium in terms of the occupied band using the GW approximation or its modifications which allow for vertex corrections after the manner of Hubbard. The definite answer to this problem has not yet been found in spite of all these efforts. In this letter we give a clear explanation of why the bandwidth of sodium is observed to be narrower in photo-emission experiments, based on the self-energy operator for the electron liquid. We think that the photo-emission experiments of sodium, if correctly interpreted, may support our assertion that the bandwidth of the electron liquid is somewhat broadened under the influence of electron-electron interactions.

Energy and momentum conservation in photo-emission experiments

\[ \hbar \omega = \varepsilon_f - \varepsilon_i \]

where \( \hbar \omega \) is the incoming photon energy; \( \varepsilon_i \) and \( \varepsilon_f \) are the initial and final state energies, respectively; the final state momentum \( p_f \) can differ from the initial state momentum by reciprocal lattice vectors \( G \) of solids. The occupied energy-band of sodium has been experimentally determined on the basic assumption that the final state energies may be identified on the nearly-free-electron-like model, i.e., \( \varepsilon_f = \hbar^2 (p+G)^2/2m \) or its simple modifications in the analysis of photo-emission experiments. In this paper we thoroughly consider sodium.
liquid perturbed by a very weak pseudo-potential rather than a non-interacting electron gas. In this case the initial and the final state energies are defined as the quasi-particle energies of the uniform electron liquid and hence the energy conservation relation is given

\[ h \omega = E(p+G) - E(p) \]

where \( E(p) \) denotes the quasi-particle energy of the electron liquid and differs from the free electron energy \( \varepsilon_p = \frac{h^2 p^2}{2m} \) by the wavenumber-dependent self-energy correction.

We shall begin with a detailed discussion on the wavenumber-dependence of the self-energy correction. The self-energy operator can be rigorously expressed as

\[ \Sigma_{\sigma}(p, \varepsilon \mid G) = \sum_{\sigma'} \frac{d\varepsilon'}{(2\pi)^3} \sum_{n} \frac{1}{2n - 1} I^{\sigma\sigma'}_{n}(p\varepsilon, p'\varepsilon'[G]) G^{\sigma'}_{p'}(p', \varepsilon') \]

where \( I^{\sigma\sigma'}_{n}(p\varepsilon, p'\varepsilon'[G]) \) denotes the contribution to the so-called particle-hole interaction from all the possible \( n \)-th order skeleton diagrams. Its functional derivative \( \frac{\delta G^{\sigma'}_{p'}(p', \varepsilon')}{\delta G(p, \varepsilon \mid G)} \) is the particle-hole irreducible interaction \( I^{\sigma\sigma'} \) above is superior to the standard expression composed from \( G, W \) and \( \Sigma \). It is highly symmetric form as a functional of \( G \) and hence suitable for including corrections up to higher orders far beyond the GW approximation. It is, however, formidably difficult to treat \( \Sigma(p, \varepsilon \mid G) \) as a functional of the renormalized \( G \) we approximately consider the self-energy operator as a functional of the renormalized \( G^0 \) and instead take the first iterative solution of Dyson’s equation \( G^0 \) in place of the self-consistent solution \( E(p) \) [8]. The summation over
expression for $\Sigma_\sigma(p, \epsilon \; [G^0])$ by the following coupling-constant-integ

$$\Sigma_\sigma(p, \epsilon \; [G^0]) = \frac{(e^2)^2}{2} \int d^2(\epsilon^2) \sum_\sigma \int \frac{dp'}{(2\pi)} d\epsilon' \Gamma^\sigma(p, p'; \epsilon, \epsilon' \; [G^0])G^\sigma(p', \epsilon')$$

(4)

$$\Gamma^\sigma(p, p'; \epsilon, \epsilon' \; [G^0]) = \sum_n I^{(n)}(p, p'; \epsilon, \epsilon' \; [G^0])$$

The replacement of $G$ with $G^0$ in the functional probably will be tolerable except for the description of the coupling between plasmons and multi-pair excitations. Such an accurate evaluation of the self-energy function around $p=p_F+q_c$ and $\epsilon=\epsilon_p+q_c$ where $q_c$ is the cut-off wavenumber. For the quantitative evaluation of many-body effects at metallic densities it is crucial that the form of $I^{\sigma'}(p, p'; \epsilon, \epsilon' \; [G])$ is sufficiently accurate as a functional of $G^0$. From the diagrammatic that the irreducible interaction $I^{\sigma'}(p, p'; \epsilon, \epsilon' \; [G])$ satisfies an integral equation diagrammatically represented in Fig. 5 in Ref. 6. Then, the irreducible interaction satisfy the integral equation in order that the resulting self-energy function.

Furthermore, the coupling constant integration which is responsible entering the original equation is indispensable for the qualitative the self-energy correction at metallic densities. This implies that the counted correctly. For the spin-parallel part of the irreducible interaction solution for the integral equation, which is given by eq.(3.13) in Ref. 6, w.
the spin-antiparallel part of the irreducible interaction we deal with it as a wavenumber independent constant for the moment because it is minor compared with the correlated part. The approximate solution for the irreducible interaction $I_{\sigma\sigma}$ substantially reducing the screening strength in the GW self-energy calculated self-energy correction in the present theory, i.e., $\text{Re } \Sigma$ above is plotted as a function of $p$ for $r_s=4.0$ approximately corresponding comparison the self-energy correction in the GW approximation is also order to sketch out the wavenumber dependent features of the self- here separate the wavenumber variable into two regions, namely, the region $0<p<p_F$ and the off Fermi sphere region where $p$ is not extremely far from $p_F$ In the Slater-approximation-like region, the dip of $\text{Re } \Sigma(p, \epsilon_p)$ [G the present theory is rather gradual but positive in contrast with that the off Fermi sphere region on the other hand, it approaches much more a function of $p$. The difference between the two calculations is enlarged in the region $p>p_F + q_c$ The dip may disappear in renormalized self-energy functional because plasmons with finite wavevectors have damping widths due to a coupling with multi-pair excitations. The systematic inclusion of vertex corrections up to higher orders is thus indispensable for the qualitative and quantitative evaluation of the self-energy correction not only in the Slater-approximation-like region but also in the off Fermi sphere
In Fig. 2 the energy-band of the electron liquid under the empty lattice potential is plotted in the direction [110]. For comparison the energy-band of the free electron gas is also plotted in the figure. The two curves are adjusted so as to agree at the Fermi level. The occupied band of the present theory is somewhat broadened in width and the unoccupied bands are, by a considerable amount, shifted to the high energy side in comparison with the free electron bands.

In the analysis of photo-emission experiments the final state energies have been conventionally assumed on the nearly-free-electron-like model, leaving the self-energy correction to the final states out of account. In other words, the usual assumption of the final state energies has been mistaken \( \epsilon_p + \text{Re} \Sigma(p, \epsilon_p [G^0]) = \text{Re} \Sigma(p+G_{110}, \epsilon_{p+G_{110}} [G^0]) \) for \( \text{E}(p) = \epsilon_p + \text{Re} \Sigma(p, \epsilon_p [G^0]) \). As is apparent from the figure, the difference \( \text{Re} \Sigma(p, \epsilon_p [G^0]) - \text{Re} \Sigma(0, 0 [G^0]) \) is much larger than the difference \( \text{Re} \Sigma(p+G_{110}, \epsilon_{p+G_{110}} [G^0]) - \text{Re} \Sigma(0, 0 [G^0]) \). From these calculations we conclude that the narrowing in the occupied bandwidth of sodium is a consequence of the wrong assumption of the final state energies. Note that the occupied bandwidth can be observed to be somewhat broader if as the final state energies one adopts the quasi-particle energies \( \epsilon_p + G_{110} \) in place of the free electron energies \( \epsilon_p + G_{110} \).

In order to make a direct comparison with experiment possible we shall here define the virtual energy-band with the dispersion \( \text{E}_{\text{vir}}(p) = \epsilon_p + \text{Re} \Sigma(p, \epsilon_p [G^0]) [G^0] + \text{Re} \Sigma(p+G_{110}, \epsilon_{p+G_{110}} [G^0]) \) which is the theoretical analogue of the experimental energy-band obtained from...
An excellent agreement between the experimental band [2] and the calculated virtual band can be seen from Fig. 3. Not only the magnitude of the narrowing in the bandwidth but also the overall features of the experimental energy-band are almost completely interpreted in terms of the non-local self-energy effect on the quasi-particle energies of the electron liquid around \( p = 0.2p_F \). In the virtual energy-band curve is ascribed to the fact \( \text{Re}\Sigma(p, \epsilon_p) - \text{Re}\Sigma(p + G_{110}, \epsilon_{p + G_{110}}) \) is an almost linearly decreasing function of wavevectors.

According to the present theory, the slope of the self-energy \( \Sigma \) gradually steepened with increasing \( r_s \) in the off Fermi sphere region over the entire range of metallic densities in the Slater-approximation the calculated virtual bandwidth corresponding to the experimental bandwidth density level of sodium, is much less narrowed for \( r_s = 2.0 \) and on the narrowed for \( r_s = 5.0 \). This is consistent with the experimental results of all potassium \( (r_s = 4.87) \) [10], though the quasi-particle energies of these by the crystal potential than those of sodium.

In conclusion, for the quantitative determination of the occupied energy-band of simple metals the usual assumption that the final state energies may be identified like the nearly-free-electron-like model requires reconsideration from a many-body theoretic point of view.
course applies to transition metals like nickel. In order to interpret photo-emission experiments of metals one must then rely on the Dyson’s equation which enables one to treat the non-local self-energy effect on quasi-particle states since the Kohn-Sham equation in its construction cannot describe such a non-local effect on both the initial and final states. It is highly desirable that the final state energies be absolutely identified by combined with photoelectron spectroscopy[11].

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Figure Captions

Fig. 1.
The self-energy correction in the present theory is plotted as a function of p for comparison the correction in the GW approximation is also plotted.

Fig. 2.
The energy-band of the electron liquid under the empty lattice potential [110]; bold line: present theory, thin line: free electron theory.

Fig. 3.
A comparison between the experimental energy-band and the calculated: line: virtual energy-band in the present theory, dotted line: free
triangles: experimental results in Ref. 2.