сообщения
объединенного
института
ядерных
исследований
дубна

E17-88-743

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EFFECTS OF COULOMB CORRELATIONS ON ONE-PARTICLE SPECTRA OF PARAMAGNETIC TRANSITION METALS: NICKEL AND IRON

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1988

1. Introduction

A quarter of the century passed since the invention of the simplest possible model of many-body interactions in metals, the so-called Hubbard model [1]

$$H = \sum_{ij\sigma} t_{ij} a^{+}_{i\sigma} a_{j\sigma} + \frac{U}{2} \sum_{i\sigma} n_{i\sigma} n_{i-\sigma},$$

where $a^{+}_{i\sigma}$, $a_{i\sigma}$ are the creation and annihilation operators for an electron of spin σ in the Wannier state at the i-th lattice site, respectively, t_{ij} is the hopping integral, and U describes the electron-electron repulsion between two electrons of opposite spins occupying the same lattice site. In spite of its simplicity, all known difficulties of the many-body problem appear here and very few exact results exist in the Literature [2]. Many approximations have been developed to study the density of states - the physical characteristic, very useful in description of the whole class of phenomena in the physics of condensed matter (see e.g. [3-5]). It is well-known that for some transition metals the photoemission measurements show disagreement between some characteristics measured in experiment and calculated within the one-particle scheme. Particularly, in nickel there is a satellite structure about 6 eV below the Fermi level in the experimental density of states, which has received much attention in literature (see e.g. [6]). A widely accepted point of view is that the satellite structure can be shown to arise from many-body effects within the unfilled d -band (see, e.g., [7,8]). It should be noted here that the spectroscopic measurements for copper (a metal with filled d -band) especially angle-resolved photoemis-

sion spectra are in very good agreement with the predictions of the band structure calculations /9/. The important main difference between copper and nickel is the position of the Fermi level. In copper the d-band lies well below the Fermi level and in nickel d-band there are approximately 0.6 d holes per atom. In addition, the Fermi level lies within the energy region of large changes of the density of states.

In this paper we want to investigate the influence of the electron correlation effects on the one-particle spectra of metals with realistic band structures. A convenient frame for description of these processes is the degenerate Hubbard Hamiltonian with contact electron-electron interactions. The problem of electron correlations is usually treated in a T-matrix approximation /10,12,13/ or non-self-consistent second-order-perturbation theory (SOPT) /5,11/. The self-consistent SOPT description of the electron-electron correlation in transition metals with model uncorrelated densities of states (rectangular and semielliptic ones) is discussed in Ref./14/. Here we report on similar calculations performed for metals described by the canonical bcc, canonical fcc (d-bands) /15/ and true nickel density of states /16/. The self-energy operator is calculated within the local approximation of Treglia et al. /5/.

The organization of this paper is as follows. In section 2 we give a short recapitulation of the SOPT approach and describe the self-consistent procedure. In section 3 we discuss numerical results for the one-particle spectra, the spectral density of states and the self-energy curves calculated in a non-self-consistent (NSC) and self-consistent (SC) way.

2. Theory

Starting with the degenerate Hubbard Hamiltonian for d-band

$$H = H_0 + \frac{U}{2} \sum_{ii'\sigma\sigma'} (1-\delta_{ii'}\delta_{\sigma\sigma'}) n_{i\sigma} n_{i'\sigma'} \qquad (1)$$

where H_0 is the band Hamiltonian and U - effective on-site Coulomb interaction, one can write the self-energy in the first and second order in U/W (W being the bandwidth) in the form

$$\Sigma(\vec{k},E) = \frac{9}{10} U N_e + \frac{9U^2}{N^2} \sum_{\vec{p}\vec{q}} \frac{f_{\vec{p}+\vec{q}}(1-f_{\vec{q}})(1-f_{\vec{k}+\vec{p}}) + (1-f_{\vec{p}+\vec{q}}) f_{\vec{q}} f_{\vec{k}+\vec{p}}}{E^+ + \epsilon(\vec{p}+\vec{q}) - \epsilon(\vec{k}+\vec{p}) - \epsilon(\vec{q})} \qquad (2)$$

where the spin index σ is suppressed as we are dealing with the paramagnetic case. Here, $f_{\vec{k}}$ - is the Fermi distribution function for the state $|\vec{k}\rangle$ of the energy $\epsilon(\vec{k})$, N_e denotes the number of electrons per atom and indices $\vec{p}, \vec{q}$ run over the first Brillouin zone of the crystal. The self-energy given by Eq.(2) is very difficult to calculate because of the 6-dimensional integrals in $\vec{k}$-space. Fortunately, as it was shown in Ref./5/, the $\vec{k}$-dependence of the self-energy can be in the first approximation neglected and we find

$$\Sigma(\vec{k},E) \Rightarrow \Sigma(E) = \frac{9}{10} U N_e + 9U^2 \int_{-\infty}^{+\infty} \frac{d\omega_1 d\omega_2 d\omega_3}{E^+ + \omega_1 - \omega_2 - \omega_3} N(\omega_1,\omega_2,\omega_3) \cdot D(\omega_1) D(\omega_2) D(\omega_3), \qquad (3)$$

where $D(\omega)$ is the local density of states corresponding to the energy spectrum $\epsilon(\vec{k})$ of H_0. The function $N(\omega_1,\omega_2,\omega_3)$ combines all $f_{\vec{k}}$ factors appearing in Eq.(2).

The one-particle spectral density of states is given by the

imaginary part of the perturbed retarded Green function $G(\vec{k},E)$ and the quasiparticle local density of states is obtained by summing the imaginary part of $G(\vec{k},E)$ over wave-vectors $\vec{k}$. Since the self-energy is $\vec{k}$ -independent, the 3D summing over B.Z. may be replaced by integration over energy

$$D(E) = -\frac{1}{\pi} \mathrm{Im} \int_{-\infty}^{+\infty} \frac{D_0(E')}{E^+ - \Sigma(E) - E'} dE', \qquad (4)$$

where $D_0(E)$ is the local density of states corresponding to uncorrelated band structure $\epsilon(\vec{k})$. If μ is the chemical potential of the quasiparticles, then at the absolute zero of temperature we have $(\mu = E_F)$

$$\frac{N_e}{10} = \int_{-\infty}^{E_F} D(E)\, dE. \qquad (5)$$

To find $D(E)$ and E_F in a self-consistent way we start with the one-electron density of states $D_0(E)$ and calculate $\Sigma(E)$ from Eq.(3). Then, the quasiparticle local density of states and Fermi energy are found from Egs.(4) and (5) and used as input data for an iteration process carried out by Egs.(3),(4) and (5).

3. Numerical results and discussion

Recently, in Ref.[14], we have carried out the self-consistent calculations of the one-particle spectra of metals described by simple model uncorrelated DOS (rectangular and semielliptic). We have found that the additional structure on the one-particle spectra which can appear in the NSC approach is smoothed out by the self-consistent process. However, real metals reveal rather

complicated structure of DOS, so it is necessary to verify again to which extend the starting DOS is modified after including of the electron-electron interaction.

In this paper we have numerically solved equations (3)-(5) for the transition metals in the paramagnetic states using the canonical *bcc*, *fcc* (*d*-bands, only) and realistic nickel uncorrelated density of states as a starting ansatz. For the canonical *bcc* DOS we have used band filling N_e = 7.3 electrons/atom which corresponds to the iron case. For the canonical *fcc* DOS we have used N_e =9.4 electrons/atom. We have used a discrete energy mesh with step equal to 0.02 times half the bandwidth and as a check we repeated all the calculations for a step equal to 0.01 times half the bandwidth. No significant effect has been observed. During the iteration process we have checked variations in the value of the Fermi level $\Delta E_F^{(i)} = |E_F^{(i)} - E_F^{(i-1)}|$ and in the shape of $D(E)$, equation (4)

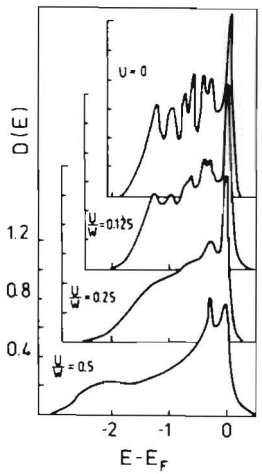
$$\Delta^{(i)} = \frac{1}{N} \left(\sum_{j=1}^{N} \left(D^{(i)}(E_j) - D^{(i-1)}(E_j) \right)^2 \right)^{1/2} \qquad (6)$$

where the index i counts the iterations and N is the number of points of the energy lying within the band limits of $D(E)$. To terminate the iteration process we have used the conditions: $\Delta E_F^{(i)} \leq$ half of the energy mesh and $\Delta^{(i)} \leq 10^{-4}$ where i is the number of the last iteration step. The self-consistency has been achieved after 5-15 iterations, mainly depending on the strength of the Coulomb interaction parameter U.

The results are displayed in figures 1-14. The origin of the energy scale is always set to the Fermi level and one half of the uncorrelated bandwidth $W/2$ is used as an energy unit.

In figures 1-3 we present the resulting one-particle spectra for the ratios U/W equal to 0, 1/8 and 1/4 for the canonical bcc uncorrelated DOS and for U/W equal to 0, 1/8, 1/4 and 1/2 for the canonical fcc and nickel starting DOS, respectively, calculated in the NSC way. In figures 4-6 we present the same characteristics as in figures 1-3 except for the SC approach. We can observe that for small correlation strength, $U/W = 1/8$, there are very small differences between the results obtained in the NSC and SC way. Furthermore, even details of the starting DOS are repeated on the resulting one-particle spectrum. For greater correlation strength, $U/W = 1/4$, general shape of uncorrelated DOS is unchanged but the bands become broader. For the correlation strength $U/W = 1/2$, the differences between the NSC and SC approaches are significant. The results obtained within the NSC approach reveal the satellite-like structure far below the Fermi energy level - see figures 2 and 3. Probably, this satellite structures appear for a strong correlation in the NSC approach regardless of the shape of the uncorrelated DOS - see also results for the rectangular and semielliptic uncorrelated DOS[5,14]. In the SC approach this satellite structure disappeared but still typical details of the uncorrelated DOS survived. In figures 7-12 we present the spectral density of states calculated for the iron (canonical bcc DOS) and nickel realistic starting DOS in the NSC and SC way for different values of the correlation strength. For one-electron states, the spectral density of states reduces to a set of delta functions peaked at the corresponding band energies but in the presence of the electron correlations these peaks are shifted and broadened. Here, we plotted

Fig. 1. The one-particle spectrum calculated for the canonical bcc uncorrelated DOS for different values of the electron correlation in NSC way. The band filling is 7.3 electrons/atom (iron).



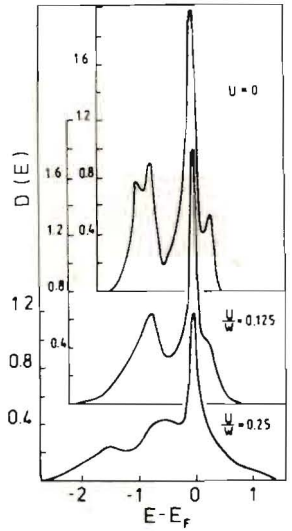


Fig. 2. As figure 1 except for canonical fcc $D_0(E)$. The band filling is 9.4 electrons/atom (nickel).

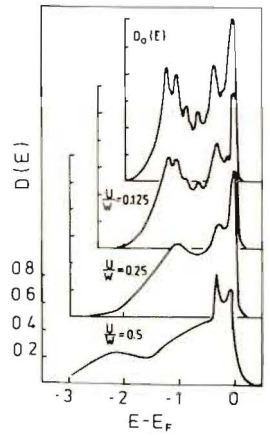


Fig. 3. As figure 2 except for nickel $D_0(E)$.

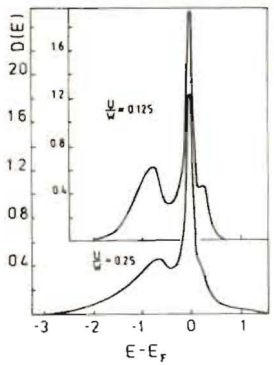


Fig. 4. As figure 1 except for calculations in SC way.

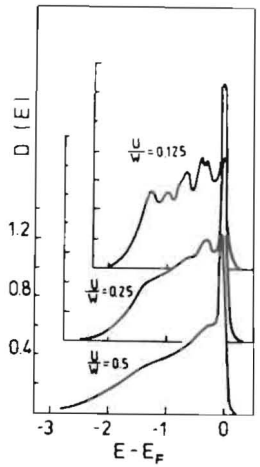


Fig. 5. As figure 2 except for calculations in SC way.

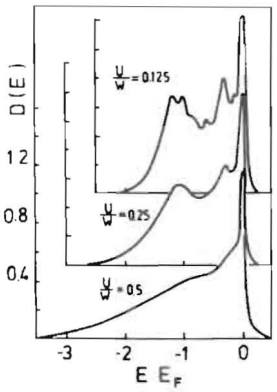


Fig. 6. As figure 3 except for calculations in SC way.

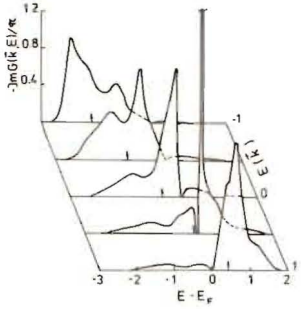


Fig. 7. One-particle spectral density of states for canonical $D_0(E)$ - iron case, calculated in NSC way, $U/W = 1/4$.

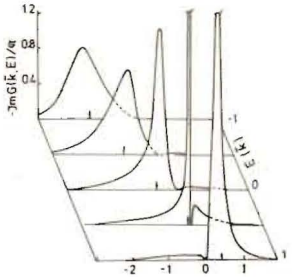


Fig. 8. As figure 7 except for calculations in SC way.

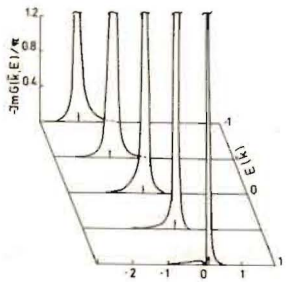


Fig. 9. As figure 7 except for nickel $D_0(E)$.

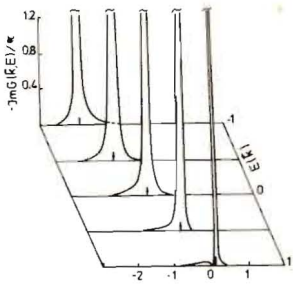


Fig. 10. As figure 9 except for calculations in SC way.

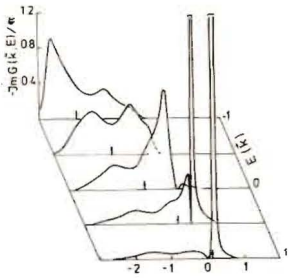


Fig. 11. As figure 9 except for $U/W = 1/2$.

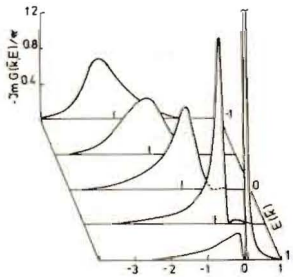


Fig. 12. As figure 11 except for calculations in SC way.

the spectral density of states for several values of $\epsilon(\vec{k})$ between bottom and top of the energy band. As one can see from figure 7 and 11, (the NSC approach and relatively strong electron correlation) the quasiparticles are not well defined. The situation for the results obtained in the SC way is quite different - figures 8 and 12. Now we have well defined peaks (although rather broad for energies near the bottom of the band) and one can obtain from the positions of these peaks the correlated band structure. Note that from figures 8 and 12 one can deduce the narrowing of the upper part of the one-particle spectrum. In figures 9 and 10 we present the spectral densities calculated for the realistic nickel starting DOS for small electron correlation. As expected, (see also figures 3 and 6 for the one-particle spectra) the differences in the shape of the spectral density of states calculated in the NSC and SC ways are of minor importance and have a well defined peaked structure centered at the corresponding uncorrelated band energy levels. In figures 13 and 14 we present the self-energy curves (real and imaginary parts) calculated for the nickel case in the NSC and SC way for two values of the electron correlation strength $U/W = 1/8$ and $1/2$. Especially for a stronger electron correlation the self-energy calculated in the SC way has a very delocalized character in comparison with the NSC results.

In conclusion, we can say there is no additional structure due to the Coulomb correlations on the one-particle spectrum arising from the canonical and realistic uncorrelated DOS. The details of the uncorrelated DOS survive on the one-particle spectrum for the strength of $U/W \leq 0.25$. For a stronger correlation

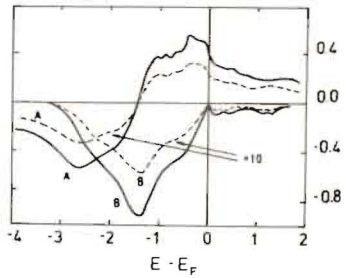


Fig. 13. The self-energy for nickel $D_0(E)$ calculated in NSC way. The full curves correspond to $U/W = 1/2$ and broken curves to $U/W = 1/8$. The real (imaginary) part is denoted by letter A(B). Note the change of scale for $U/W = 1/8$.

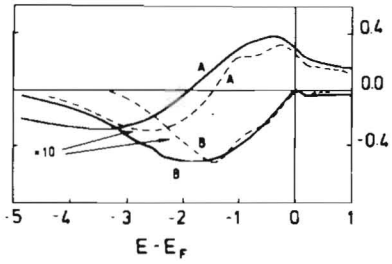


Fig. 14. As figure 13 except for calculations in SC way.

the resulting one-particle spectrum has rather structureless character.

Acknowledgements

The work of R.T. was partially supported by INTiBS under contract CPBR 15.6/45.

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Received by Publishing Department
on October 11, 1988.

Таранко Р., Таранко Э., Малек И. E17-88-743

Влияние кулоновских корреляций на одночастичные спектры парамагнитных металлов: никель и железо

В рамках модели Хаббарда рассмотрены электронные корреляционные эффекты в переходных парамагнитных металлах: никеле и железе, для описания которых использованы реалистические плотности состояний. При использовании самосогласованной теории возмущения во втором порядке по параметру разложения U/W (U - кулоновское взаимодействие, W - ширина зоны) показано, что рассчитанные одночастичные спектры проявляют типичное поведение для некоррелированной плотности состояния только для $U/W \leq 0,25$. Для более сильных кулоновских корреляций одночастичные спектры не проявляют почти никакой структуры и предсказываемый ранее (в несамосогласованном подходе) дополнительный пик исчезает.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

Сообщение Объединенного института ядерных исследований. Дубна 1988

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Effects of Coulomb Correlations on the One-Particle Spectra of Paramagnetic Transition Metals: Nickel and Iron

Electron correlation effects in transition metals are studied within the Hubbard model for realistic local density of states of paramagnetic nickel and iron. Using the self-consistent second-order-perturbation theory in U/W (where U is the Coulomb integral and W is the bandwidth), it is shown that the one-particle spectra reveal typical details of the uncorrelated density of states for $U/W \leq 0.25$, only. For stronger Coulomb correlations the resulting one-particle spectrum reveals rather structureless behaviour, without any additional satellite-like structures predicted by the non-self-consistent approach.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

Communication of the Joint Institute for Nuclear Research. Dubna 1988