Effect of pressure on the Fermi surface of ferromagnetic nickel

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In this paper, we have studied the effect of pressure on the Fermi surface of nickel, using the linear-muffin-tin-orbital method in the atomic-sphere approximation. The pressure derivatives of extremal areas ($A^{-1}dA/dP$) are calculated with use of various exchange-correlation potentials. We find that the pressure derivatives of the smaller orbits are very sensitive to the choice of the exchange-correlation potential used. Our calculations are in agreement with the experimental data.

I. INTRODUCTION

Nickel has been a prototype metal for innumerable studies of various physical properties involving itinerant-electron ferromagnetism, d-band electronic structure, and transition-metal surfaces. The interpretation of pressure effects on both electron transport and crystallographic properties of metals usually requires some knowledge of the way in which the Fermi surface (FS) is affected by pressure. As a result there has been a concentrated effort to study the effect of pressure on the FS of metals both theoretically and experimentally. However, for the ferromagnetic metals there has been a relatively weaker impetus. We have seen that the pressure derivative $A^{-1}dA/dP$ calculated using the linear-muffin-tin-orbital (LMTO) method[1] agrees with the experimental data for the noble metals[2] and for the transition metals Pd and Pt.[3] The recent experimental studies of the effect of pressure on the FS of nickel were performed by Anderson *et al.*,[4] who obtained the pressure derivatives for the "neck" and "ellipsoid" orbits using the solid-helium high-pressure technique and by Vinokurova and co-workers,[5–7] who reported the results of measurement of the de Haas–van Alphen (dHvA) effect at pressures up to 1 kbar. Anderson *et al.*[4] measured the pressure derivative of the X-centered [001] hole-pocket cross-sectional area as positive, in contradiction to the negative value reported by Vinokurova and co-workers.[5–7] This was later explained by Gapotchenko, Itskevich, and Kulatov[8] as being due to the fact that pressure derivatives for the X-centered [001] hole pocket are small and less accurate than the measurements made by Vinokurova and co-workers.[5–7]

On the theoretical side, Anderson *et al.*[9] have studied the effect of a change in lattice spacing on the band structure of ferromagnetic nickel, using the augmented-plane-wave (APW) method. Their calculations for the change in the magneton number with pressure gave good agreement with the experimental data, while the pressure derivatives of the FS areas were not in good agreement with the data. In fact, the calculated pressure derivatives were within a factor of 3 of the experimental data. Vinokurova *et al.*[7] have calculated the band structure of ferromagnetic nickel using a model Hamiltonian technique and have obtained the pressure derivatives of two FS orbits. Since this calculation involves fitting parameters, we shall not discuss it any more.

Thus, there is only one *ab initio* band calculation concerning the effect of pressure on the FS of ferromagnetic nickel, and agreement with the experimental data is only qualitative. With the aim of ascertaining whether the agreement between theory and experiment can be improved, we have performed detailed calculations of the effect of pressure on the FS of ferromagnetic nickel, using different exchange-correlation potentials. The calculations are performed with use of the LMTO method.[1] Another reason for making these calculations is to compare the results of the APW and LMTO calculations. These calculations form a natural extension to our earlier calculations on the noble metals[2] and the paramagnetic transition metals Pd and Pt.[3]

In Sec. II we briefly give the method of calculation. Results and discussions are in Sec. III, and conclusions in Sec. IV.

II. METHOD OF CALCULATION

We have used the LMTO–atomic-sphere-approximation[1] method, including the combined corrections terms. Our calculations are done to self-consistency which we take to be that the change in potential parameters is in the fifth decimal place. The area-to-mass ratios of the computed surface in a plane normal to a direction (i.e., the magnetic field) were found by numerical integration of radii calculated at a fixed interval of rotation in the plane. In the past, authors have taken percentage error in the Fermi-surface area as a meaningful index of the success of the fit to the Fermi surface. We take the view that in a band calculation, since we are calculating energy eigenvalues, we would like to know the error in the eigenvalues. It would be, therefore, meaningful to talk of error in terms of the shift in the Fermi energy ΔE_F required to bring the calculated extremal area in agreement with the experimental area,

$$\Delta E_F = \frac{1}{\Pi}\frac{A_{\text{expt}} - A_{\text{calc}}}{m_b} ,$$

where m_b is the calculated band mass for the orbit. ΔE_F

is calculated for all the experimentally measured FS orbits and the extreme ΔE_F is a measure of the accuracy of the agreement with the experimental data.

In our earlier work on transition metals palladium[10] and platinum,[11] we have studied (i) the effect of increasing the number of **k** points in Brillouin-zone summation and (ii) the effect of including angular momentum expansion up to $l=3$. We find that increasing the number of **k** points from 240 to 505 for Pd resulted in a change in extreme ΔE_F from 5.7 to 6.1 mRy and including angular momentum expansion from $l=0,1,2$ to $0,1,2,3$ resulted in a change in extreme ΔE_F from 4.0 to 4.9 mRy. Hence, the calculations are done self-consistently with potential parameters for $l=0,1,2$ and Brillouin-zone integrations are done with 240 **k** points. Using the self-consistent parameters, we have calculated the FS areas for various orbits for magnetic fields along the [001], [111], and [110] directions. The lattice is then expanded by 0.1% and the self-consistent parameters are recalculated. From these two self-consistent calculations at two different lattice spacings, and using the value of compressibility 5.38×10^{-4} for nickel,[12] we obtain $A^{-1}dA/dP$. The calculations are done with the von Barth–Hedin (vBH),[13] Vosko-Wilk-Nussair (VWN),[14] and Slater $X\alpha$-XC (Ref. 15) potentials. In the $X\alpha$-XC, α was taken to be 0.715 because this gave a good fit to the zero-pressure FS.[16]

III. RESULTS AND DISCUSSION

A. Fermi-surface areas at ambient pressure

We have calculated the extremal areas of the FS orbits for magnetic fields along the [001], [110], and [111] directions. These results are presented in Table I, together with the experimental results of Tsui[17] and Stark.[18] Also given are the results of Anderson *et al.*,[9] Prasad, Joshi, and Auluck,[19] and Wang and Callaway.[20] The first-principles calculation of Wang and Callaway gives qualitative agreement with the Fermi-surface data with an extreme error of 17 mRy. All *ab initio* calculations predict two sets of X-centered hole pockets, one arising from level X_5 and the other from level X_2, whereas experimentally only one set of hole pockets associated with level X_5 has been observed. Prasad, Joshi, and Auluck[19] have reported the energy-band structure calculated using a modified interpolation scheme and get good agreement

TABLE I. Cross-sectional areas of the Fermi surface (ΔE_F in units of millirydbergs).

Orbit	Orientation	Center	Band	Expt. value	Calc.[a]	Calc.[b]	Calc.[c]	Our calculation BH-XC	Our calculation VWN-XC	Our calculation $X\alpha$-XC
Neck	[111]	L	6↑	0.0071[d]	0.0071	0.0236		0.0095	0.0099	0.0119
					0.0	−30.9		(−4.7)	(−5.3)	(−9.1)
Neck	[110]	L	6↑	0.0102[d]		0.0402		0.0118	0.0124	0.0156
						−32.9		−2.6	−3.5	−7.3
Neck	[112]	L	6↑			0.0313		0.0104	0.0109	0.0134
Large square	[001]	Γ	6↑	1.15[e]	1.154	1.23	1.24	1.2079	1.2115	1.2818
					−0.1	−13.1	−6.4	−9.0	−9.6	−23.2
Pockets	[001]	X	3↓	0.0267[d]	0.0258	0.023	0.018	0.0246	0.0250	0.0262
					−0.4	−1.3	−2.1	−1.4	−1.3	−0.4
Pockets	[100]	X	3↓	0.0665[d]	0.0671	0.049	0.038	0.0536	0.0559	0.0610
					0.1	−3.9	−6.9	−1.6	−1.4	−0.8
Pockets	[110]	X	3↓	0.0585[d]		0.034		0.0445	0.0463	0.0506
						−4.6		−2.1	−1.9	−1.5
Pockets	[101]	X	3↓			0.028		0.0274	0.0281	0.0299
Pockets	[111]	X	3↓	0.0442[d]		0.028		0.0347	0.0359	0.0388
								−1.8	−1.6	−1.5
Pockets	[001]	X	4↓			0.050		0.0408	0.0309	0.0356
Pockets	[100]	X	4↓			0.086		0.1044	0.1074	0.1165
Pockets	[110]	X	4↓			0.070		0.0529	0.0542	0.0582
	[001]	Γ	5↓			2.04		2.1124	2.1037	2.0824
	[110]	Γ	5↓			1.59		1.5969	1.5865	1.5621
	[111]	Γ	5↓			2.08		2.1371	2.1224	2.0895
Small square	[001]	Γ	6↓	0.9[e]	0.903	0.80	0.84	0.8997	0.8968	0.8871
					0.1	9.2	2.2	0.0	0.3	1.1
	[110]	Γ	6↓			0.96		0.8977	0.8948	0.8853
	[111]	Γ	6↓			0.71		0.7725	0.7698	0.7671
Extreme ΔE_F (mRy)					0.5	42.1	9.1	9.0	9.9	24.3

[a]Reference 19.
[b]Reference 9.
[c]Reference 20.
[d]Reference 17.
[e]Reference 18.

with the experimental results on the large Γ-centered sheets as well as on the smaller FS sheets. They have reported an extreme ΔE_F of around 0.5 mRy, which is very low in comparison to the other calculations. Prasad, Joshi, and Auluck have missed a factor of 2 while calculating the extreme ΔE_F, so the corrected extreme ΔE_F should be 1.0 mRy. Earlier *ab initio* calculations do not give a complete calculation for all the FS areas. A detailed calculation of this type was presented by Anderson *et al.*,[9] who used the self-consistent spin-polarized augmented-plane-wave method with the vBH-XC potential. We have calculated ΔE_F for these FS areas using their band masses and find a large ΔE_F for the necks of the majority band and an overall extreme ΔE_F around 42 mRy.

A look at Table I shows that we obtain good agreement with the experimental data with an extreme ΔE_F of 9.0 mRy (vBH), 9.9 mRy (VWN-XC), and 24.3 mRy ($X\alpha$) with $\alpha=0.715$. We have varied the value of α in the $X\alpha$-XC potential to obtain the best agreement with the experimental data. Our calculations give a smaller ΔE_F in comparison with the calculation of Wang and Callaway[20] and the self-consistent calculation of Anderson *et al.*[9] Detailed discussions are given elsewhere.[16]

B. Pressure derivatives

The calculated pressure derivatives, along with the experimental data, are given in Table II. Consider first the majority-spinup orbits. For the Neck [111] and Neck [112] orbits, our pressure derivatives for VWN-XC and $X\alpha$-XC are in better agreement with the experimental results[4–7] compared with the calculation of Anderson *et al.*[9] The agreement with the vBH-XC potential is fair with experiment, as is agreement with the calculation of Anderson *et al.*[9] For the Neck [110] orbit experimental data is not available, while for the large square [001] neither experimental nor theoretical values are available. Our calculations for the different XC's are consistent with each other for these orbits.

Consider now the minority spindown orbits. The small X pockets [001] have a positive pressure derivative in agreement with the experimental results of Anderson *et al.*[4] as well as their calculations.[9] For the pressure derivative of the $X_{3\downarrow}$ pockets for magnetic fields along the [111] and [112] directions, our calculation for VWN-XC and $X\alpha$-XC shows better agreement with the measured values compared with the calculation of Anderson *et al.*[9] Our results for ellipsoids and small squares centered at Γ are consistent with the calculation of Anderson *et al.*[9] for all three XC potentials, while for the remaining pockets and ellipsoids centered at X, our calculations for VWN and $X\alpha$-XC, as well as for vBH-XC, are not in agreement with the calculations of Anderson *et al.*[9] This difference could be due to the APW method and the tetrahedron interpolation scheme used by them. Another reason may be the calculation of FS areas at ambient pressure. The ΔE_F for our calculation using the vBH-XC potential is 9.0 mRy while for the calculation of Anderson *et al.* with the vBH-XC potential it comes out to be 42.1 mRy. Such a large difference at ambient pressure could contribute to the difference between the two

TABLE II. Experimental and calculated values of $A^{-1}dA/dP(10^{-4}\,\text{kbar}^{-1})$.

Orbit	Orientation	Center	Band	Expt. value	Calc.[a]	Our calculation BH-XC	VWN-XC	$X\alpha$-XC
Neck	[111]	L	6↑	6.0[b] (8.0)[c]	2.6	11.0	8.1	7.9
Neck	[110]	L	6↑		1.5	9.7	6.8	5.1
Neck	[112]	L	6↑	6.6[c]	2.3	10.7	7.8	7.2
Large square	[001]	Γ	6↑			1.9	1.8	1.0
Pockets	[001]	X	3↓	1.0[b] (−0.8)[c]	2.6	7.3	2.6	2.8
Pockets	[100]	X	3↓		3.1	17.5	7.3	7.3
Pockets	[110]	X	3↓		2.6	16.8	7.1	6.5
Pockets	[101]	X	3↓		2.5	10.5	4.1	4.0
Pockets	[112]	X	3↓	1.5[c]		12.5	5.3	5.0
Pockets	[111]	X	3↓	6.6[c]	2.7	14.6	5.8	5.0
Ellipsoids	[001]	X	4↓		−0.4	−47.5	12.7	14.1
	[100]	X	4↓		1.4	12.3	5.2	6.4
	[110]	X	4↓		1.9	10.1	3.9	6.0
	[001]	Γ	5↓		4.4	5.6	4.7	4.9
	[110]	Γ	5↓		4.5	6.7	5.3	5.7
	[111]	Γ	5↓		4.5	6.7	5.1	5.7
Small square	[001]	Γ	6↓		4.3	5.3	4.5	4.7
	[110]	Γ	6↓		4.0	5.3	4.6	4.8
	[111]	Γ	6↓		4.3	5.3	4.5	4.7

[a]Reference 9.
[b]Reference 4.
[c]Reference 5.

calculations. An overall look at Table II shows that VWN-XC and $X\alpha$-XC with $\alpha=0.715$ gives the better agreement with measurements[4-7] in comparison to vBH-XC.

IV. CONCLUSIONS

In the de Hass–van Alphen experiments one measures the quasiparticle FS. Theoretically, we calculate the one-electron Kohn-Sham (KS) FS. The recent work on density-functional theory indicates that there is no reason for the two to be the same.[21] However, there is ample empirical evidence to demonstrate that the calculated KS FS agrees qualitatively with experiments, suggesting that the difference between the KS FS and quasiparticle FS is not very large. Thus, one can use the KS FS to compare with the experimental data.

Our calculations of $(1/A)dA/dP$ for the nickel suggest that the values given by the LMTO method are in good agreement with experiment. It shows that the LMTO method is capable of explaining the FS topology of the ferromagnetic transition metals as well as for paramagnetic metals. For nickel, there is a dearth of data. We hope that our calculations will generate more enthusiasm among the experimentalists to fill the gap. Our calculations show that $X\alpha$-XC gives the best agreement with the measured orbits. For the small X pocket $\langle 001 \rangle$, our calculated pressure derivative is positive, which is in agreement with the measurement of Anderson *et al.*[4] For the larger orbits, the pressure derivatives are insensitive to the choice of the XC potentials. This is consistent with our work with the noble metals and transition metals Pd and Pt. We find that particular attention must be paid to convergence, in spin-polarized calculations, to obtain the accuracy required for the computation of FS changes. Our calculations with the LMTO method show a partial improvement over the existing calculations.

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