Effect of hydrostatic pressure on the Fermi surface of Pd and Pt

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The pressure derivatives of a number of Fermi-surface orbits in Pd and Pt are calculated using the linear muffin-tin orbital method in the atomic-sphere approximation and including the combined correction terms. The calculations are performed with use of various exchange-correlation potentials. We compare our results with available experimental data and theoretical calculations.

I. INTRODUCTION

The pressure derivatives of the Fermi-surface (FS) orbits provide a crucial test of the accuracy of a band-structure calculation. In a previous paper we calculated the effect of hydrostatic pressure on four FS orbits in the noble metals and found good agreement with experimental data.[1] In fact, we had suggested that the pressure derivatives could be used to ascertain which exchange-correlation (XC) potential is appropriate for the noble metals. In this paper we present calculations for the pressure (hydrostatic) derivatives $A^{-1}dA/dP$ of FS orbits in the transition metals Pd and Pt which have a complicated FS geometry compared to the noble metals.

We have chosen Pd and Pt because for both these metals we have calculated the zero-pressure FS using the linear muffin-tin orbital (LMTO) method in the atomic-sphere approximation (ASA) and including the combined correction terms.[2,3] Our calculations indicate that the LMTO method does give a good description of the FS for both Pd and Pt. Hence, as a natural extension we have calculated $A^{-1}dA/dP$ for these metals. While there exist three sets of data[4–6] for Pd, there is only one[4] for Pt with which to compare our results. Moreover these works did not include all the FS orbits as we have done. We hope our work will generate more interest in this area. For Pt, ours is the first calculation of $A^{-1}dA/dP$.

II. METHOD OF CALCULATION

We have used the LMTO-ASA method including the combined correction terms.[7] As is well known, the LMTO is a fast *ab initio* method involving a diagonalization procedure and is well suited for FS work. The band-structure calculations are done self-consistently (which we take to be the convergence of the energy eigenvalues within 10^{-4} Ry). Starting with the parameters in Skriver's book, this requires half a dozen more iterations. We have used potential parameters for $l=0,1,2$ and the Brillouin-zone integrations are done with 240 **k** points.

Using these self-consistent parameters, we have calculated the FS orbital areas and masses using Stark's area-mass routine.[8] The areas of the computed surface in a plane normal to the direction of the magnetic field were found by numerical integration of the radii calculated at a fixed interval of rotation in that plane. The areas are calculated for 17 orbits in Pd and 11 in Pt. The orbit designations are given in Tables I and II. For a description of the orbits we refer the reader to two excellent articles on the de Haas–van Alphen effect (dHvA) in Pd (Ref. 9) and Pt.[10] Next the lattice is expanded by 0.1% and the self-consistent parameters recalculated. From the calculation of the FS areas $A^{-1}dA/dP$ can be obtained using compressibilities of 5.5×10^{-4} kbar^{-1} for Pd and 3.59×10^{-4} kbar^{-1} for Pt.[11] These calculations are done with the Slater $X\alpha$,[12] von Barth–Hedin (vBH),[13] von Barth–Hedin–Janak (vBHJ),[14] and the Vosko-Wilk-Nussair (VWN) (Ref. 15) exchange-correlation potentials. In the $X\alpha$ potential $\alpha=0.75$ for Pd (Ref. 2) and $\alpha=0.817$ for Pt (Ref. 3) because these gave a good fit to the zero-pressure FS.

III. RESULTS AND DISCUSSION

The pressure derivatives for the FS orbits in Pd and Pt using various XC potentials are given in Tables I and II. Consider first the case of Pd. There exist three sets of data obtained by Vuillemin and Bryant,[4] Skriver *et al.*,[5] and Joss and van der Mark.[6] While the first two measured $A^{-1}dA/dP$ under hydrostatic pressure, the last group deduced $A^{-1}dA/dP$ from their unaxial data. Hence the uncertainties are larger. All the data are consistent with each other. Skriver *et al.*[5] have calculated $A^{-1}dA/dP$ using the LMTO-ASA with the vBH XC potential (including combined correction terms). The differences (large in some orbits) could be due to the tetrahedron technique used by them for calculating pressure derivatives. However our $X\alpha$ and VWN XC potential results are in better agreement with theirs. We find that the pressure derivatives of the large FS orbits are insensitive to the XC potential used, while the small FS orbits are very sensitive. It would be tempting to decide on the appropriateness of any XC potential based on the experimental $A^{-1}dA/dP$. However no single XC potential yields a good agreement with all the FS orbits. We hope that with data available on more orbits the situation could be more encouraging.

Our results for Pt are given in Table II. Unfortunately, Vuillemin and Bryant[4] have measured the pressure derivative only for the Γ-centered FS. All XC potentials yield good agreement with the data. The scarcity of data

TABLE I. $A^{-1}dA/dP$ (10^{-4} kbar^{-1}) with different XC potential for Pd.

Field direction \ $A^{-1}dA/dP$	using BH-XC	using vBHJ-XC	using VWN-XC	using $X\alpha$-XC $\alpha=0.75$	Calculation Ref. 5	Joss and van der Mark Ref. 6	Expt. values Ref. 5	Expt. values Ref. 4
L center								
$\langle 111\rangle LKW$	−20.3	−17.7	−20.2	−11.6	−3	−34.5±10		
$\langle 111\rangle$	−16.9	−15.3	−12.8	−9.4				
$\langle 110\rangle LK\Gamma$	−12.4	−13.1	−12.6	−9.3	−15.6			
$\langle 100\rangle$	−18.0	−16.6	−15.0	−12.5	−20.0		−17.0±2.0	
X center								
$\langle 010\rangle XW\Gamma$	2.9	2.9	4.6	7.7	6.8	7.0±0.8	6.1±0.7	
$\langle 110\rangle XU\Gamma$	5.8	5.7	8.4	7.3	6.9			
$\langle 100\rangle XWU$	5.4	5.4	8.0	4.6	4.2	10.5±2.0	10.5±0.7	
$\langle 110\rangle$	3.4	3.4	5.3	5.3	5.0			
$\langle 111\rangle$	3.0	3.0	4.8	4.8				
Γ center						3.9±1.0		
$\langle 111\rangle$	4.4	4.4	4.7	4.5		3.6±0.5		3.9±0.4
$\langle 100\rangle$	4.6	4.6	4.8	4.7	4.2	3.5±0.5	4.1±0.8	4.0±0.4
$\langle 110\rangle$	5.0	5.0	5.3	5.1				
open hole								
$\langle 100\rangle\alpha$	5.0	4.9	5.1	5.2				
$\langle 001\rangle\epsilon$	3.8	3.8	3.8	3.8				
$\langle 110\rangle\beta$	5.8	5.7	5.7	6.7				
$\langle 111\rangle\beta$	4.2	4.2	4.1	4.7				
$\langle 110\rangle\gamma$	7.3	6.4	6.4	4.7				

precludes us from making any more definitive conclusions.

IV. CONCLUSIONS

Our calculations of $A^{-1}dA/dP$ for Pd and Pt suggest that the LMTO-ASA method yields values that are in agreement with the minuscule experimental data. We find that the agreement is better for the Γ-centered FS sheet while it is not so good for the X-centered and L pockets in Pd. Our calculations show that the choice of XC potential does not influence the pressure derivatives of the larger FS orbits. The pressure derivatives of the smaller FS orbits are dependent on the XC potential used. Hence one could use the experimental data to determine the best XC potential. However, the lack of sufficient data precludes us from drawing any definitive conclusions. We hope our work will lead to more experiments on the measurement of $A^{-1}dA/dP$ in Pd and Pt.

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TABLE II. $A^{-1}dA/dP$ (10^{-4} kbar^{-1}) with different XC potential for Pt.

Field direction \ $A^{-1}dA/dP$	using vBH-XC	using vBHJ-XC	using VWN-XC	using $X\alpha$-XC $\alpha=0.815$	Expt. values Ref. 4
X center					
$\langle 010\rangle XW\Gamma$	11.9	10.9	32.3	52.0	
$\langle 110\rangle XU\Gamma$	11.8	11.1	32.0	51.8	
$\langle 100\rangle XWU$	10.1	9.3	27.8	47.8	
$\langle 110\rangle$	10.5	9.8	28.8	49.0	
$\langle 111\rangle$	10.1	9.4	28.0	48.0	
Γ center					
$\langle 111\rangle$	3.0	3.0	3.1	3.1	2.8±0.3
$\langle 100\rangle$	3.2	3.2	3.3	3.3	3.2±0.3
$\langle 110\rangle$	3.3	3.5	3.6	3.6	
Open hole					
$\langle 100\rangle\alpha$	3.3	3.2	3.4	3.7	
$\langle 001\rangle\epsilon$	2.6	2.6	2.7	2.7	
$\langle 110\rangle\beta$	4.6	4.3	4.8	5.2	

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