PRESSURE DEPENDENCE OF THE ELECTRONIC STRUCTURE AND FERMI SURFACE OF PALLADIUM*

Shashikala G. Das

Argonne National Laboratory, Argonne, Illinois 60439

D.D. Koelling

Magnetic Theory Group, Department of Physics, Northwestern University,
Evanston, Illinois 60201

and

F.M. Mueller

Argonne National Laboratory, Argonne, Illinois 60439

We have considered the effects of hydrostatic pressure on the de Haas—van Alphen external cross-sectional areas of the Fermi surface of palladium. Our calculations for the density of states in palladium with extended and contracted lattice constants show that a change of 2 per cent in the lattice constant changes the density of states at Fermi energy by 10 per cent.

RECENTLY, interest in the electronic structure of pure palladium has been centered on its incipient ferromagnetic nature and the effects of spin fluctuations.[2] However, the importance of the electron—phonon interaction as compared to the electron—paramagnon interaction in the enhancement of the electronic specific heat[3] and the effective cyclotronic mass[4] is still unknown. Moreover, if the effects of spin—orbit coupling on the electronic states are included, then the electron—phonon interaction *does* couple to the exchange-enhanced RPA susceptibility.[5] Our purpose here is to stimulate further experimental interest in the effects of long-wave length phonons on the electronic properties of pure palladium so as to derive the effective electron—phonon coupling constant(s). Among the most direct experimental evidence would be those experiments involving the effects of pressure on the Fermi surface.

A priori we should anticipate that palladium would be a most favorable case on which to measure such effects since the Fermi energy falls at a place on the single-particle density of states curve where it has both large zeroth and first derivatives.[6] This is due to the presence of a saddle point in the purely *d*-like part of the 5th band just 1.89 mRy below the Fermi level and 90 per cent of the contribution to the total density of states comes from the *d*-like holes in Pd (4th and 5th band).[7] Moreover, the Fermi surface of palladium has three separate sheets, which have highly anisotropic effective masses.[4] Thus even small changes induced in the electronic structure of palladium by hydrostatic pressure would have large changes in the Fermi surface.

It has been only relatively recent that transition metal band structures have been available with sufficient
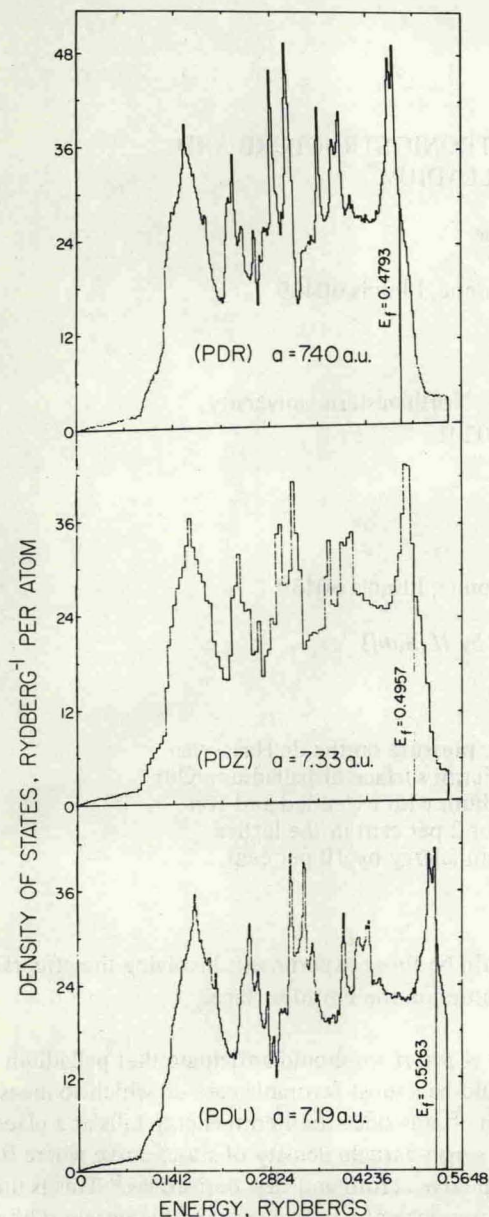
FIG. 1. The density of states for the compressed (*PDR*), normal (*PDZ*) and expanded (*PDU*) lattice of palladium. The energy scale has been shifted such that the bottom of the conduction bands are lined up. As is mentioned in the text, the density of states at the Fermi energy is a very sensitive function of the position of Fermi level because of the saddle point singularity just above the Fermi level

reliability and accuracy to make detailed comparisons with experimental results very meaningful.[6,7] A problematic feature of the early calculations was the

placement of the *d*-band complex relative to the plane wave background. Mattheiss[8] in 1965 using an approximation to self-consistency and charge neutrality gave the first systematic treatment of the first row of the transition metals. The essential feature of these calculations is: (1) the dominance of crystallographic structures in the determination of the form of the *d*-band complex; (2) the relative insensitivity of the electronic structure, other than *d*-band broadening and net shifts within a single crystal type. Based on this insight, Hodges, Ehrenreich and Lang,[9] (HEL) and simultaneously Mueller and Phillips[10] (CIS), proposed schemes which focussed on these results. The narrow *d*-bands were treated as five tight-binding states with a few disposable parameters (essentially the Slater Koster[11] Hamiltonian); the broad $s-p$ bands were treated as four orthogonalized plane waves (the main difference between HEL and CIS occurs here — HEL used a local pseudopotential approximation, where CIS uses a nonlocal pseudopotential), with further disposable parameters. Later on Pettifor[12] (and many others) derived from the first principles (starting from KKR formalism) the hybrid nearly free-electron tight-binding (H—NFE—TB) model Hamiltonian which was very similar to the one empirically derived by Mueller and Phillips and HEL. The model Hamiltonian approach is found to yield quite an accurate description of the band structure of the transition metals like palladium, platinum nickel, etc., at normal lattice constant.[7] Here we consider a more complicated case, namely, the analysis of the electronic structure of transition metal, palladium, using the model Hamiltonian approach, when the lattice constant is treated as a variable. In copper, such calculations by Faulkner, David and Joy[13] using the KKR scheme show remarkable agreement with pressure dependent de Haas—van Alphen data of O'Sullivan and Shirber.[14] This analysis which may be thought of as a form of 'Deformation Potential Theory', offers insight into electron—phonon interaction in the transition metals.

Relativistic augmented plane wave (RAPW) calculations were carried out for palladium based on the seven different lattice constants a_l listed in Table 1. The potentials used in these calculations were constructed as follows. A constant atomic configuration of $4d^{10} 5s^0$ was assumed. The muffin-tin constant was varied so as to maintain charge neutrality (via the Mattheiss scheme) for each choice of a_l. (Thus static electronic screening on these charge densities, exchange