Possibility of Pressure-Induced Superconductivity by Phonon Mechanism in Pd

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Abstract

Pressure dependencies of electronic structures and lattice dynamics of Pd which is close to ferromagnetic instability at ambient pressure are investigated on the basis of first-principles calculations. Particular attention is paid to phonon-mediated superconductivity: the transition temperature T_c without taking into account spin fluctuation effects are estimated to be $6.40 - 8.55$ K at ambient pressure and $0.58 - 1.44$ K at 50 GPa. Though spin fluctuation effects would vanish the T_c at 0 GPa, the T_c under pressures would survive due to the rapid decreasing of spin fluctuation effects with increasing pressure. We estimate the T_c at 50 GPa to be at most 0.71 K if we include spin fluctuation effects.

Key words: first-principles calculation; pressure-induced superconductivity; Pd

1. Introduction

Superconductivity in palladium (Pd) has not been observed down to 1.7 mK at ambient pressure[1]. The large Stoner enhancement factor results in large spin fluctuation, which is thought to be the reason for the absence of superconductivity in Pd. Pure Pd films, however, can be transformed into superconductors below a temperature, T_c=3.2K, by means of irradiation at low temperatures with He^+ ions[2]. The defects produced by irradiation would smear out the sharp peak in the density of states at the Fermi level, $N(E_f)$, and the Stoner enhancement factor would be reduced and hence spin fluctuation. The decrease in $N(E_f)$, however, would lead to a decrease in the electron-phonon coupling (BCS phonon parameter λ) and this would be detrimental to superconductivity. On the other hand some theoretical studies[3,4] show that in the irradiated Pd the decrease in spin fluctuation does not accompany the decrease in $N(E_f)$.

In this paper we evaluate the effects of pressure on the electronic band structures, the lattice dynamics and hence the superconducting transition temperature, T_c, in Pd. Possibility of pressure-induced superconductivity by phonon mechanism is discussed by taking into account spin fluctuation effects and its pressure dependencies.

2. Calculational procedure

We perform the first-principles calculations using the Full-Potential Linear Muffin-Tin Orbital (FP-LMTO) method[5]. The vibrational frequencies of lattice dynamics are calculated based on the liner-response theory[6]. We calculate the dynamical matrix as a function of wave vector for a set of irreducible q-points on a (8,8,8) reciprocal lattice grid. We use 3κ-*spd*-LMTO basis set with the one-center expansions performed inside the MT spheres up to $l_{max} = 6$. The induced charge densities and the screened potentials are represented inside the MT spheres by spherical harmonics up to $l_{max} = 6$ and in the interstitial region by plane waves with the cut off corresponding to the (16,16,16) fast-Fourier-transform grid in the unit cell of direct space.

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The transition temperature T_c without taking into account spin fluctuation effects are estimated by the Allen-Dynes formula[7]:

$$T_c = \frac{\omega_{\log}}{1.2}\exp\left(-\frac{1.04(1+\lambda)}{\lambda-\mu^*(1+0.62\lambda)}\right), \quad (1)$$

where λ denotes the dimensionless electron-phonon coupling constant, $\omega_{\log}$ the logarithmic-averaged phonon frequency and μ^* the effective screened Coulomb repulsion constant whose value is usually taken to be between 0.1 and 0.15.

3. Results and discussion

Before calculating lattice dynamics we first performed first-principles band structure calculations for fcc and bcc structure by adopting the FP-LMTO method with GGA correction. It has been confirmed that the fcc structure is energetically favorable than the bcc structure at least up to 50 GPa. Therefore we calculate the lattice dynamics only for fcc structure.

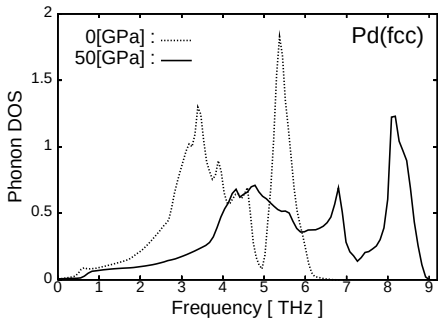

Fig. 1. The phonon density of states (DOS) of Pd at ambient pressure and 50 GPa.

Figure 1 shows the phonon density of states (DOS) calculated at ambient pressure and 50 GPa. Hardening of the phonon frequencies is clearly seen. Our dispersion curves at ambient pressure (not shown here) are in good agreement with the experimental results[8] and theoretical one[9]. In Table 1, the parameters for the Allen-Dynes formula and the superconducting transition temperature T_c calculated by the equation (1) are summarized. Our results show that T_c of Pd estimated by taking into account only the phonon-mediated mechanism are fairly high. As mentioned in Introduction, however, it is expected that large spin fluctuation suppresses T_c of Pd at ambient pressure down to 0 K.

Following Mazin *et al.*[10] we take into account the role of spin fluctuations by adopting the following modified Allen-Dynes formula:

$$T_c = \frac{\omega_{\log}}{1.2}\exp\left(-\frac{1.04(1+\lambda+\lambda_{sf})}{\lambda-\lambda_{sf}-\mu^*(1+0.62(\lambda+\lambda_{sf}))}\right),$$

$$\lambda_{sf} = \frac{\alpha}{1-UN(E_f)}. \quad (2)$$

Here λ_{sf} represents the effects of ferromagnetic spin fluctuation with U and α being the exchange parameter and some constant parameter.

Table 1
The values of $N(E_f)$, $\langle\omega^2\rangle^{1/2}$, $\omega_{\log}$, λ and T_c evaluated at ambient pressure and 50 GPa.

$P[GPa]$	0.0	50.0	
$N(E_f)[\frac{\text{states}}{\text{Ry}\cdot\text{cell}}]$	36.7442	27.4058	
$\langle\omega^2\rangle^{1/2}[K]$	182.4726	215.5134	
$\omega_{\log}[K]$	134.3628	113.3958	
λ	0.9459	0.5051	
$T_c[K]$	6.40	0.58	μ^*=0.15
	8.55	1.44	μ^*=0.1

Since Pd is believed to be very close to ferromagnetic instability, we tentatively assume the value of $UN(E_f)$ to be 0.96 - 0.98. Furthermore we determine the value of α so that T_c at ambient pressure is $\sim$0 K. Then we obtain α=0.0113 - 0.0226 for μ^*=0.1. If we assume that U and α are independent of pressure, T_c at 50 GPa is estimated to be 0.32 - 0.71 K ($UN(E_f)$ at 50 GPa is 0.716 - 0.731). The estimations of T_c at 50 GPa considerably depend on the parameters so that first-principles evaluation of U and α is necessary in order to give more quantitative and reliable discussion.

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