ELECTRONIC STRUCTURE OF DISORDERED CuPd ALLOYS BY POSITRON-ANNIHILATION 2D-ACAR*

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Electronic Structure of Disordered CuPd Alloys by Positron-Annihilation 2D-ACAR

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Abstract:

We report 2D-ACAR experiments and KKR CPA calculations on alpha-phase single-crystal $Cu_{1-x}Pd_x$ in the range $x \leq 0.25$. The flattening of the Fermi surface near [110] with increasing x predicted by theory is confirmed by our experimental results.

Two-dimensional angular correlation (2D-ACAR) is a powerful tool for studying the electronic structure of concentrated substitutionally-disordered alloys. This was strikingly demonstrated in the early Fermi surface measurement in CuZn alloys by Haghgooie and Berko [1]. Only with the help of extensive theoretical calculations, however, can the experimental spectra yield their full meaning. The Korringa-Kohn-Rostoker (KKR) coherent-potential-approximation [2] (CPA) has proven to be an extremely effective framework for calculating the electronic structure in random substitutional alloys, and in recent years this approach has been adapted [3-5] to calculate both the electron momentum density $\rho(p)$ and the two-photon momentum density $\rho_{2\gamma}(p)$. With such momentum-density calculations, features of the electronic structure throughout the occupied bands can be identified, not merely at the Fermi surfaces. Nevertheless, relatively few critical comparisons between predicted and measured 2D-ACAR spectra for alloys have been made to date. Such comparisons require either (i) a reconstruction [6] of $\rho_{2\gamma}(p)$ from several 2D projections on a single-crystal specimen, or (ii) the calculation of $\rho_{2\gamma}(p)$ [or $\rho(p)$] on a sufficiently dense grid to simulate the measured spectrum

$$N(p_x,p_y) = \int \rho_{2\gamma}(p_x,p_y,p_z)\, dp_z.$$

We present in this paper both experimental 2D-ACAR spectra and KKR-CPA calculations of momentum density for the alpha-phase $Cu_{1-x}Pd_x$ system, with x up to 0.25. The electronic structure of this system is unusual in that the centers of the component d-bands essentially coincide, whereas their widths differ substantially, which gives rise to pronounced off-diagonal disorder [7], in contrast to the diagonal disorder that is dominant in most transition-noble metal alloys. This paper will focus primarily on the issue of Fermi surface (FS) flattening near [110] for CuPd alloys; other aspects of the electronic structure of this system as manifested in 2D-ACAR measurements are discussed elsewhere [8]. KKR CPA calculations [9,10] of the FS predict a flattening near the

[110] direction with increasing x (up to about 0.40). Gyorffy and Stocks [10] have argued that a Kohn anomaly at the wavevector spanning parallel flat regions of the FS is responsible for the short-range-order diffuse-scattering peaks observed experimentally in these alloys [11,12]. Our results provide the first direct evidence that the FS of CuPd does indeed flatten in the vicinity of [110] with increasing x.

Measurements were made on single-crystal specimens (x=0.0, 0.10, 0.15, 0.20, and 0.25) oriented with [100] directions aligned perpendicular to the axis of the Anger cameras. Spectra with 2 x 10^6 counts and 0.34-mrad channel width were taken for all specimens; spectra with 1.5-2.0 x 10^7 counts and a channel width of 0.17 mrad were also taken for the x=0.0, 0.15, and 0.25 specimens. The overall resolution, based on the combined effects of room-temperature thermal broadening and an instrumental resolution of 0.34 mrad, was approximately 0.6 mrad.

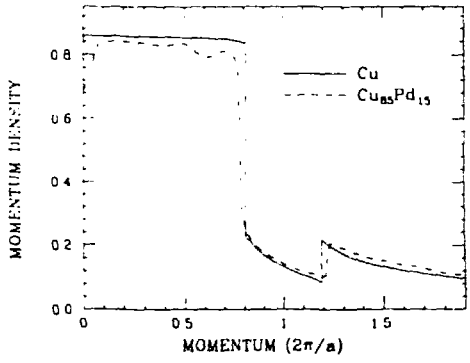

Figure 1:Momentum density ρ(p) in Cu (solid) and in $Cu_{0.85}Pd_{0.15}$ (dashed) along [100].

Calculations of the electron-momentum density ρ(p) were performed for $Cu_{1-x}Pd_x$ alloys with x=0.15 and 0.25 based on the KKR CPA [3] and for pure Cu using the KKR method [13]. In the case of pure Cu, $\rho_{2\gamma}(p)$ was also calculated. The Cu and Pd muffin-tin potentials employed here were taken from earlier work [9], where it is shown that these potentials yield good agreement with angle-resolved photo emission. The difference between $\rho_{2\gamma}(p)$ and ρ(p) is usually small for $p<p_F$. In Fig. 1, we show the calculated momentum densities ρ(p) for pure Cu and $Cu_{0.85}Pd_{0.15}$. We observe that in this system the Fermi surface (cf. the breaks near p ≈ 0.8 and 1.2 x 2π/a) remains sharp in the alloy. This is in contrast to the behavior, e.g., in CuGe alloys, where the FS breaks are broadened considerably [14].

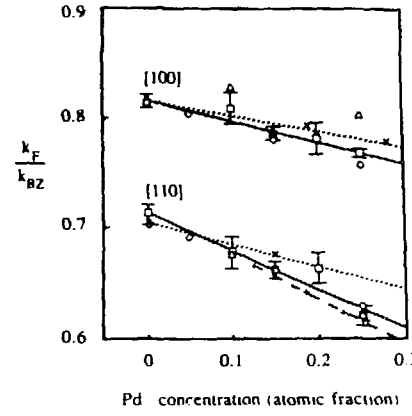

Figure 2: k_F/k_{BZ}, where k_{BZ} is the zone-boundary dimension, as a function of Pd concentration. Solid lines are fits to the FS radii obtained from the present experiments (squares). Fits to results of Hasegawa, et al (crosses and dotted lines, ref 15) and Ohshima and Watanabe (plusses and dashed line, ref. 11,12) also shown. Theoretical values: lozenges, present work and ref. 9; triangles, ref. 10.

We address now the problem of extracting FS dimensions from the measured 2D-ACAR spectra. The FS radii in various directions are in principle given by the positions of extrema in the derivative of $N(p_x,p_y)$. However, the positions of the extrema may be shifted from the true FS as a result of finite experimental resolution, and a more detailed analysis is therefore

required to obtain accurate values. To gain additional insight into this problem, we have performed several tests including (i) simulations of $N(p_x,p_y)$ based on the theoretical $\rho(p)$ convoluted with the experimental resolution function and (ii) applying a high-pass filter to the differentiated data. On the basis of these studies, we conclude that for the alloys considered here, the FS radius in the [110] direction can be reliably obtained by differentiation of $N(p_x,p_y)$, but a more elaborate analysis is required to extract the [100] FS radius. The [100] FS radius was obtained by fitting the experimental spectra $N(p_x,p_y)$ to a parametrized function as described in ref. 8. The values of k_F obtained in this manner were insensitive to the size of the (p_x,p_y) fitted region and to the assumed experimental resolution function. Our results for the [100] and [110] FS radii are plotted in Fig. 2 along with the KKR CPA calculations of Rao et al [9] and of Gyorffy and Stocks [10]. We note that both sets of theoretical calculations [9,10] are in excellent agreement with the present experimental results, particularly with respect to the concentration dependences. The slope of k_F/k_{BZ} is larger in the [110] direction than in the [100] direction, which indicates that the FS is flattening in the vicinity of [110] with increasing x. Such an interpretation is in accord with the FS radii deduced from diffuse scattering measurements [11,12]. We have also investigated the composition dependence of the FS of Cu within the rigid-band model [16] and by application of the KKR CPA for several transition and nontransition solutes. These calculations all exhibit a flattening FS in the vicinity of [110] with decreasing electron/atom ratio. In conclusion, the evidence of the present experimental data in conjunction with our theoretical calculations shows clearly that the FS flattening in CuPd alloys does indeed occur.

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