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Free-electron-like Hall effect and deviations from free-electron behavior in Ca-Al amorphous alloys

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The Hall coefficients of Ca-Al amorphous alloys have been measured at 4.2 K over a wide range of compositions. It is shown that the magnitude of the Hall coefficients are close to the nearly-free-electron (NFE) prediction for low Ca concentrations but deviate significantly from the NFE values for Ca concentration greater than 45 at. %. The deviations from the free-electron values have previously been attributed to the effects of *s-d* hybridization, while a reduction in magnitude by Au doping has been argued to result from the side-jump effect.

The Hall coefficient has been investigated in quite a wide range of amorphous alloys[1–5] and can deviate significantly from the nearly-free-electron (NFE)-like behavior. Indeed, in some transition-metal-based alloys it can even be positive. The origin of these deviations in the Hall coefficient still remains a controversy. There are two effects that may influence the magnitude and sign of the Hall coefficient: *s-d* hybridization[6] and the side-jump mechanism.[7] Both the magnitude and sign of the Lorentz force contribution are affected by the *s-d* hybridization depending on the position of the Fermi energy relative to the *d* band. The side-jump effect leads to an extra contribution to the Hall effect arising from the spin-orbit interaction which again can be positive or negative depending on the position of the Fermi level in the *d* band. Ca-Al is a simple-metal-based amorphous alloy. Despite this, all previous measurements have shown significant deviations from the NFE-like behavior one might expect in a simple-metal alloy.[8,9] For all these measurements the concentration of Ca has been greater than ~50 at. %. In our previous paper[8] we argued that the deviations from nearly-free-electron-like behavior were the result of the Fermi level being pushed into the tail of Ca *d* band by Al increasing the effects of *s-d* hybridization. The side-jump contribution in this system is enhanced by increasing the strength of spin-orbit scattering and we show this using Au doping.[8] Both effects of *s-d* hybridization and the side-jump mechanism are expected to lead to a Hall coefficient that is not free-electron like. In this paper, we present an extension of the Hall-coefficient measurements of Ca-Al over a wider range of compositions and show that for concentration of Ca less than 45 at. %, the NFE model adequately describes the magnitude of the Hall coefficient. The measurements were made at 4.2 K on sputtered samples in magnetic fields up to 7 T. Experimental details have been outlined by Mayeya and Howson.[8]

Figure 1 shows the Hall coefficients of Ca-Al as a function of Ca concentration and the NFE calculation made by assuming a contribution of three free electrons per atom from Al and two from Ca. The Hall coefficients are negative over the entire composition range. Their magnitudes however differ significantly for low and high Ca content samples. In samples with a low Ca concentration, the magnitude of the Hall coefficients are close to the NFE. This can be explained from the fact that in this regime, the electronic structure is dominated by the aluminum *s* states and consequently the Hall coefficient takes on the free-electron value. In the case of Ca-rich samples the Hall coefficients deviate significantly from the free-electron value. It has been argued previously that this deviation comes from the effects of *s-d* hybridization due to the presence of Ca *d* states near the Fermi energy.[8]

Photoemission studies by Nagel *et al.*[10] for Ca-Al showed the existence of Ca *d* states at the Fermi level occurring in the tail of the Ca *d* band and thus there is a low density of *d* states at the Fermi level. Howson, Hickey, and Morgan[11] argued that increasing the concentration of Al up to 20 at. % in Ca-Al alloys pushes the Fermi level further into the Ca *d* band increasing the Ca *d* occupation. Recently, Morton *et al.*[12] used resonant photoemission to study Ca 3*d* states in Ca-Al amorphous alloys. Their results show that the resonance appears for

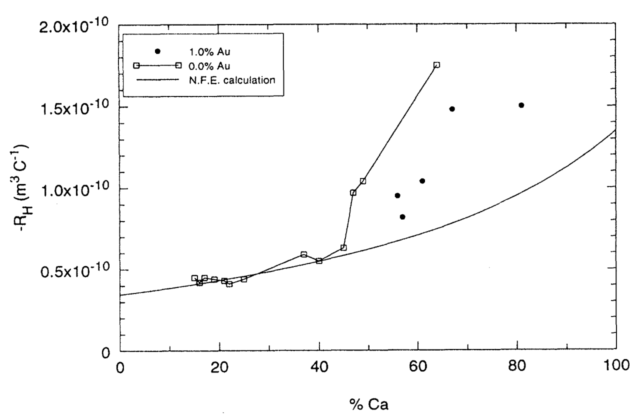

FIG. 1. Hall coefficients of Ca-Al amorphous alloys as a function of Ca concentration.

Ca concentration greater than ~46 at. % and this indicates a large increase in the density of *d* states near the Fermi level for concentration of Ca of greater than ~46 at. %. In our Hall-coefficient measurements, significant deviations from the free-electron values occurs at ~45 at. % Ca, which correlates with the sudden increase in this $3p \rightarrow 3d$ resonance from the photoemission studies. Thus it appears that the sudden onset of deviations from NFE behavior in Ca-Al may be related to a sudden increase in the density of *d*-like states near the Fermi level for Ca concentration greater than ~45 at. %.

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