University of Huddersfield Repository


Investigation of field-induced ferromagnetism in Pd–Ni–Fe–P metallic glass by x-ray magnetic circular dichroism

Original Citation


This version is available at http://eprints.hud.ac.uk/11988/

The University Repository is a digital collection of the research output of the University, available on Open Access. Copyright and Moral Rights for the items on this site are retained by the individual author and/or other copyright owners. Users may access full items free of charge; copies of full text items generally can be reproduced, displayed or performed and given to third parties in any format or medium for personal research or study, educational or not-for-profit purposes without prior permission or charge, provided:

- The authors, title and full bibliographic details is credited in any copy;
- A hyperlink and/or URL is included for the original metadata page; and
- The content is not changed in any way.

For more information, including our policy and submission procedure, please contact the Repository Team at: E.mailbox@hud.ac.uk.

http://eprints.hud.ac.uk/
Investigation of field-induced ferromagnetism in Pd–Ni–Fe–P metallic glass by x-ray magnetic circular dichroism


1Bragg Institute, Australian Nuclear Science and Technology Organisation, Lucas Heights, New South Wales 2234, Australia
2School of Physics, University of Western Australia, 35 Stirling Highway, Crawley, Western Australia 6009, Australia
3National Synchrotron Radiation Research Center, 101 Hsin-Ann Road, Hsinchu Science Park, Hsinchu 30076, Taiwan
4Institute of Materials Research, University of Salford, Salford M5 4WT, United Kingdom
5School of Chemistry, The University of Sydney, New South Wales 2006, Australia

(Received 3 October 2008; accepted 20 December 2008; published online 13 January 2009)

We have applied x-ray magnetic circular dichroism to investigate the field-induced ferromagnetism in Pd$_{40}$Ni$_{22.5}$Fe$_{17.5}$P$_{20}$ alloy. The experiment revealed that both Ni and Fe were in a divalent state and that the magnetic properties of the material were determined by the localized 3$d$ electrons of the transition metals. No clear evidence of Ruderman–Kittel–Kasuya–Yosida-type interaction among magnetic clusters was observed. It is believed that the detailed balance of fundamental spin-orbit and exchange interactions as a function of temperature and applied magnetic field determine the different magnetic properties of the alloy. © 2009 American Institute of Physics.

[DOI: 10.1063/1.3070528]

The metallic glass of the Pd$_{40}$(Ni$_{1-x}$Fe)$_{x}$P$_{20}$ alloy possesses interesting magnetic properties, including paramagnetic, superparamagnetic, and spin glass phases as a function of temperature and applied magnetic field. An interesting observation by Shen et al. was the existence of a field-induced ferromagnetic phase in amorphous Pd$_{40}$Ni$_{22.5}$Fe$_{17.5}$P$_{20}$. The formation mechanism of the field-induced ferromagnetic phase is not yet well understood. Hsiao et al. suggested that interaction among magnetic/chemical inhomogeneities through Ruderman–Kittel–Kasuya–Yosida (RKKY) coupling across the Pd-rich matrix may contribute to the complex magnetic properties of the alloy. Different cluster sizes of 30–40, 200, and 20 Å have been reported.

For Pd$_{40}$Ni$_{19.8}$P$_{20}$ the effective moment of nickel is 0.109 $\mu_B$/atom, which is double that of the Ni$_{80.2}$P$_{19.8}$ alloy as a result of the induced polarization of neighboring Pd atoms. For Pd$_{40}$Ni$_{40-x}$Fe$_x$P$_{20}$ ($x=5–17.5$) Shen et al. calculated an effective moment of 5.6–6.4 $\mu_B$/Fe atom, assuming that the magnetic moment of the Ni was 0. They suggested that the enhanced Fe moment was also due to a strong spin polarization of Pd-rich matrix. Although a classical interpretation of RKKY interaction, based on the system with unperturbed paramagnetic matrix, may not be applicable to the system containing Pd whose paramagnetic ground state may be strongly modified by the magnetic impurities and thus carrying induced magnetic moment, a similar type of coupling may be responsible for the formation of the field-induced ferromagnetism of the Pd$_{40}$Ni$_{22.5}$Fe$_{17.5}$P$_{20}$. We report on studies of individual magnetic moments of Fe, Ni, and Pd for the Pd$_{40}$Ni$_{22.5}$Fe$_{17.5}$P$_{20}$ alloy by x-ray magnetic circular dichroism (XMCD), aiming to identify the origin of the field-induced ferromagnetism.

1Electronic mail: dyu@ansto.gov.au.

The Pd$_{40}$Ni$_{22.5}$Fe$_{17.5}$P$_{20}$ ribbon, prepared by rf melt spinning, has been characterized using a superconducting quantum interface device (SQUID) magnetometer, and the same phase behavior as reported by Shen et al. has been confirmed. The XMCD experiment was conducted at the Dragon beam line at the National Synchrotron Radiation Research Centre in Taiwan. The sample temperature was 17 K and the applied magnetic field was 1 T, corresponding to the induced ferromagnetic phase of the alloy. The field was along the length direction ($y$-axis) in the surface plan of the sample. The propagation direction of the incident circular polarized light (about 80% polarization) was 30° with respect to the surface of the sample. The total electron yield mode was used for the measurements of x-ray absorption spectra (XAS). The XMCD spectra correspond to the difference of the spin-resolved XAS with two opposite magnetic field directions after proper background subtraction. The magnetic moments were derived using the XMCD sum rules. Oxidation of the top monolayers of the sample could not be excluded. However the measurement should mainly probe the bulk properties of the alloy as the probe depth of the total electron yield is of the order of 10 nm.

Figures 1 and 2 present the Ni and Fe 2p isotropic XAS and XMCD, respectively. Multiple structures have been observed as marked by capital letters from A to F in the figures. For Ni, two distinct peaks appear at both $L_2$ and $L_3$ edges, while three peaks can be identified from the corresponding Fe 2p XAS.

The double peak structures of the Ni 2p XAS have been predicated theoretically and observed experimentally in several compounds, for example, Ni dihalides and NiO, Ni oxalate, and NiFe$_2$O$_4$ (trevorite). The 17 eV splitting between peaks A and C corresponds to the $L_2$ and $L_3$ separations due to the large spin-orbit interaction of the core level. The splitting between peaks A and B is a measure of the 2p hole and 3$d$ electron exchange interaction. The mea-

0003-6951/2009/94(2)/022502/3/$23.00 94, 022502-1 © 2009 American Institute of Physics
Experimental observations for Pd$_{40}$Ni$_{22.5}$Fe$_{17.5}$P$_{20}$. For the iso-
for both initial and final states. The calculated spectra with
spin-orbit, exchange interactions, and interatomic screening
of the spin states involved in the transitions.\textsuperscript{13}

but with less pronounced spin-flip effects due to the nonpure
1.78

observed for trevorite and NiO, respectively.\textsuperscript{12} Similar to the
1.78

crystal field strength.\textsuperscript{9}

/H20849

is in good agreement with the calculated value of $B_{\text{spin}}=0.78$ for the high-spin ground state. Theory\textsuperscript{9} also pre-
predicted the triple peak structure at the $L_3$ edge. This provides
evidence that the localized 3$d$ electrons also dominate the
magnetic properties of the Fe in the Pd$_{40}$Ni$_{22.5}$Fe$_{17.5}$P$_{20}$ com-

The measured Ni 2$p$ XAS and XMCD spectra are con-
istent with the existing atomic multiplet calculations in a
crystal field with local octahedral symmetry for free Ni$^{2+}$ ion
with initial 3$d^8$ and final 2$p3d^6$ configurations.\textsuperscript{9,12} Coulomb,
spin-orbit, exchange interactions, and interatomic screening
and states mixing effects were included in the calculations
for both initial and final states. The calculated spectra with
crystal field strengths larger than 1 eV approached the ex-
perimental observations for Pd$_{40}$Ni$_{22.5}$Fe$_{17.5}$P$_{20}$. For the iso-
tropic branching ratios $B(i)$, defined as the integrated inten-
sity of the $L_i$ edge normalized to the integrated intensity of the
whole XAS,\textsuperscript{8} our measured value is 0.779 ± 0.009, which
is in good agreement with the calculated value of $B(i)=0.75$ corresponding to the high-spin ground state with 1 eV
crystal field strength.\textsuperscript{8}

The orbital-to-spin magnetic moments ratio ($L/S$) for
Ni from Pd$_{40}$Ni$_{22.5}$Fe$_{17.5}$P$_{20}$ is 0.33 ± 0.06, obtained using
XMCD sum rules. The value agrees well with the results of
0.27 ± 0.07 and 0.34 ± 0.11 from trevorite.\textsuperscript{12} This indicates

that the localized 3$d$ electrons determine the Ni magnetic
properties in both compounds, which are well described by
the atomic calculations.

For Fe, a similar atomic calculation has been reported
based on the transition of 3$d^7 → 2p3d^6$ of Fe$^{2+}\textsuperscript{9,14}$. The XAS
and XMCD spectra agree qualitatively with the present mea-
surement. Our experimental isotropic branching ratio of
0.788 ± 0.003 agrees very well with the calculated values of
$B(i)=0.78$ for the high-spin ground state. Theory\textsuperscript{9} also pre-
dicted the double peaks at the $L_3$ edge, which is approximately 40% of the Fe moment, which is substan-
tial.

The comparison of the XMCD signal for Fe, Ni, and Pd
is presented in Fig. 3. Iron is the dominant carrier of the
magnetic moment as evidenced by its about ten times larger
$L_3$ edge dichroism than that from the Ni $L_2$ edge. The XMCD
signal at the Pd $M_3$ edge is about ten times smaller than that
of Ni at $L_2$ edge. Since the sign of the Pd XMCD was not
observed to change when the polarization state of light was
changed, we conclude that there was no measurable XMCD
from Pd $M_3$ edge within the limits of the current experiment.
Due to the excitation range of the instrument, the XMCD at
the Pd $L$ edge has not yet been measured. The Pd $L$ edge may
show discernible XMCD signal.\textsuperscript{14} The failure to observe di-
chroism from the Pd $M_3$ edge does not totally exclude the
RKKY interaction through the Pd matrix. Further studies are
planned to measure XMCD at the Pd $L$ edge to investigate
the role of RKKY interaction in the formation of field-

\begin{table*}[h]
\centering
\begin{tabular}{cccccc}
\hline
Element & Fe & Ni & Fe & Ni \\
Orbital (L) & 0.0541 & 0.086 & 0.0579 & 0.05 \\
Spin (S) & 0.5240 & 1.98 & 0.1754 & 0.52 \\
$L/S$ & 0.0975 & 0.043 & 0.3301 & 0.096 \\
Total & 0.5751 & 2.066 & 0.233 & 0.57 \\
\hline
\end{tabular}
\caption{Magnetic moments for Fe and Ni; Pure element values are quoted from Refs. 8 and 13.}
\end{table*}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{Ni 2$p$ (a) XAS and (b) XMCD at $T=17$ K and $B=1$ T.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig2.png}
\caption{Fe 2$p$ (a) XAS and (b) XMCD at $T=17$ K and $B=1$ T.}
\end{figure}
induced ferromagnetism. Nevertheless our observations do show that the localized 3d electrons of Fe and Ni determine the overall magnetic properties of the alloy. The strong temperature and magnetic field dependence of the magnetic properties of the alloy may be originated from spin-orbit and exchange interactions which are of the order of $kT$.

In summary, individual magnetic moments from Ni and Fe in the Pd$_{40}$Ni$_{22.5}$Fe$_{17.5}$P$_{20}$ metallic glass have been determined by XMCD in the field-induced ferromagnetic phase. While Fe is the dominant carrier of magnetic moment, Ni contributes significantly to the total magnetic moment of the material. The enhancement of orbital magnetic moments of Ni and Fe makes a large contribution to the overall magnetic moments of the material. The experiment shows that the localized 3d electrons of Ni and Fe may determine the magnetic properties of the alloy, through the detailed balance of fundamental spin-orbit and exchange interactions as a function of temperature and applied magnetic field.

The support from the Australian Synchrotron Research Program is gratefully acknowledged. We thank Dr. Frank Klose for useful discussions and comments on the draft.