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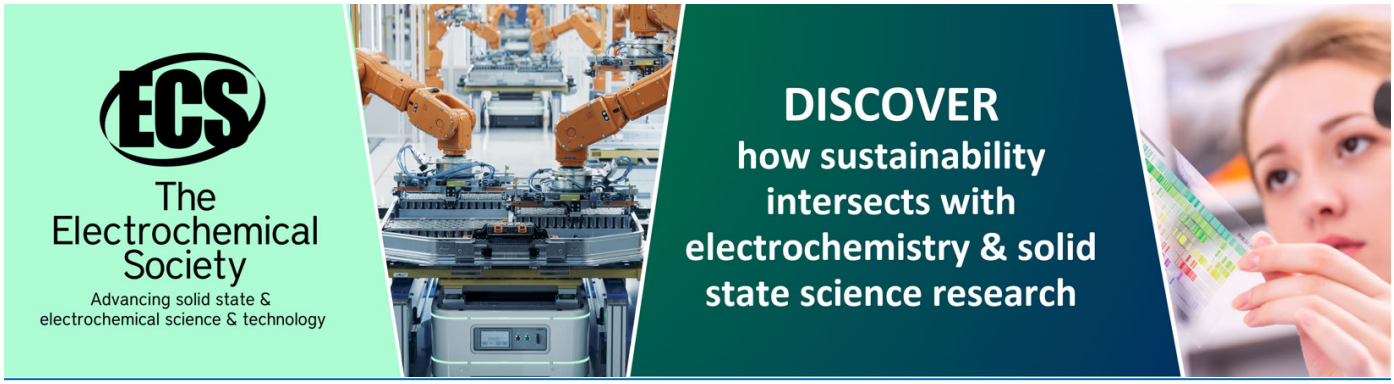

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Pressure dependence of magnetic states in Laves Phase $R\mathrm{Co}_2$ (R=Dy, Ho, and Er) compounds probed by XMCD

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Abstract. In order to probe pressure-induced modification in Co magnetic state in Laves phase $R\mathrm{Co}_2$ (R=Dy, Ho, and Er) compounds, we have measured X-ray magnetic circular dichroism (XMCD) under high pressure. The R L_2-edge XMCD indicates that Co magnetic moment M_{Co} gradually decreases as the pressure increases. The Co K-edge XMCD shows that the effect of R molecular field is significantly suppressed under high pressure. As a result, it is revealed that the pressure variation of M_{Co} is closely associated with R-Co interaction. The Co magnetic state is separately discussed from that of R partner.

1. Introduction

In the cubic Laves phase $R\mathrm{Co}_2$ (R= rare-earth element) compounds, magnetic phase transition is ascribed to instability of Co magnetic state, which is strongly affected by the internal molecular field provided by R moments [1,2]. To clarify experimentally this explanation, we have made X-ray magnetic circular dichroism (XMCD) in ErCo_2 under high pressure [3]. As a result, it has been revealed that the Co moments M_{Co} monotonically decrease with increasing pressure whereas the Er moments M_{Er} remain almost constant under the pressure up to 4.2 GPa, and that the applied pressure weakens the contribution of Er to the Co magnetic state. However, it is still not clear how the magnetic interaction between Co and R partner influences the Co magnetic state under pressure.

In order to probe pressure-induced transition of $R\mathrm{Co}_2$, XMCD is a powerful tool because of a capability of providing the information about the magnetic states with an element-selectivity. When the R partner element is replaced by heavier R, the magnetic properties show a systematic reduction, *e.g.* Curie temperature T_{C} [4], Co magnetic moment [2]. In the $R\mathrm{Co}_2$ series, DyCo_2, HoCo_2 and ErCo_2 show a first-order magnetic phase transition from paramagnetism to ferrimagnetism at T_{C}=137 K, 78 K, and 32 K under ambient pressure. As the applied pressure increases, T_{C} linearly decreases at low pressures and asymptotically approaches a temperature above critical pressure P_{c}. The magnetic phase transition also changes to a second-order above P_{c}, which goes down with the replacement of

R=Dy→Ho→Er [5,6]. Therefore, a systematic XMCD for these compounds under high pressure is crucial for the fuller understanding of the *R*-Co magnetic interaction in RCo_2.

In this study, we have made the XMCD measurement under high pressure in $DyCo_2$ and $HoCo_2$ as well as $ErCo_2$. The pressure variation is compared among the three compounds, and then the Co magnetic state is separately discussed from that of the *R* partner. This approach may be suitable for understanding the *R*(5*d*)-Co(3*d*) hybridization that plays an important role in itinerant electron magnetism.

2. Experimental procedure

Sample of RCo_2 was prepared by arc-melting under an Ar atmosphere. Powdered sample was used for X-ray absorption spectroscopy (XAS) and XMCD measurements under high pressure. The spectral data were recorded using the helicity-reversal method in transmission mode on BL39XU of SPring-8. Pressure variation was measured on the same experimental conditions; at *T*=5 K and *H*=5 T. These circumstances have been achieved using a dual in-line tiny diamond-anvil-cell (DAC) to be inserted into a superconducting magnet. We have successfully made such measurement for two different samples on the same condition. Fluorinert was used as a pressure transmission medium. The applied pressure was estimated by the conventional ruby fluorescence method.

3. Results and Discussion

XAS and XMCD spectra at the *R* L_2-edge in RCo_2 (*R*=Dy, Ho, and Er) with various pressures are shown in Figure 1. White-line intensity progressively increases with increasing pressure, which is commonly observed for the series. This enhancement suggests that the 5*d* hole number is increased as the applied pressure is intensified, that is, the *R* 5*d* electronic state is gradually localized [3].

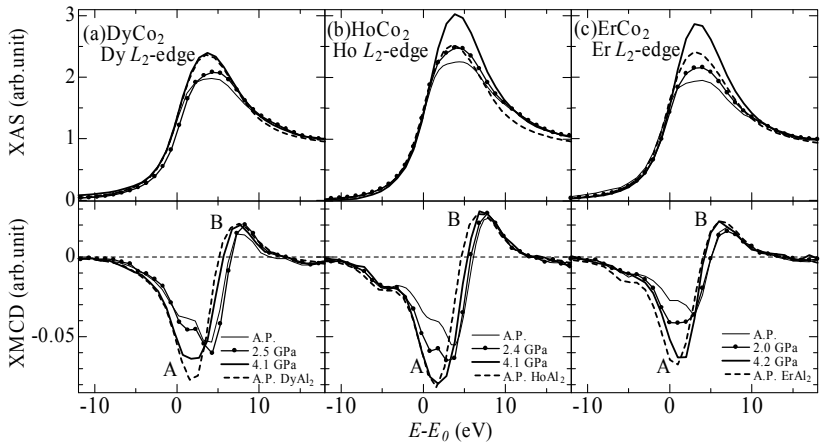

Figure 1. XAS and XMCD spectra at the *R* L_2-edge in RCo_2. The absorption edge E_0 is determined from the inflection point of the XAS spectrum.

The dichroic spectrum mainly consists of a negative broad peak **A** just above E_0 and a positive peak **B** at the higher energy side. As the pressure is increased, the peak **A** increases the intensity and the spectral profile is modified, whereas the peak **B** almost remains the initial profile. It should be emphasized that the dichroic spectrum is changed so as to reach that of the reference material RAl_2, in which only the *R* partner carries the magnetic moments. Since the peak **A** includes the *R*(5*d*)-Co(3*d*) hybridized component [7,8], the increase in the peak **A** may be interpreted as caused by pressure-induced suppression of M_{Co}. In the same way as the previous study using the Er L_2-edge [3], we assumed that the magnitude of M_{Co} is proportional to the difference between the XMCD spectra in RCo_2 and RAl_2. Thus estimated M_{Co} is shown as a function of pressure in Figure 2. As the

applied pressure is increased up to about 4 GPa, the magnitude M_{Co} in $HoCo_2$ decreases to ≈ 0.1. The reduction rate is relatively large and similar to the rate in $ErCo_2$ [3]. From extrapolation, M_{Co} in $HoCo_2$ would maintain up to about 5 GPa. In comparison with those, $DyCo_2$ shows a relatively small rate, and M_{Co} is about three times larger than that in $ErCo_2$ and $HoCo_2$ at 4 GPa. Therefore, the Co magnetic state in $DyCo_2$ is more stable against the applied pressure than the other compounds.

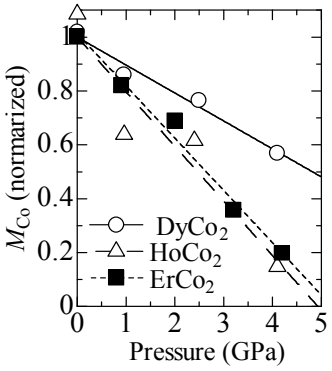

Figure 2. Co magnetic moment M_{Co} as a function of pressure. Magnitude of M_{Co} is normalized by the value at ambient pressure.

Figure 3 shows the pressure variation of XAS and XMCD spectra at the Co *K*-edge. The Co *K*-edge XAS is characterized by a small peak **C** just above E_0 under ambient pressure. The peak **C** intensity is suppressed with increasing pressure, and the dichroic intensity is rapidly diminished. The peak **C** structure is interpreted as caused by 4*p*-3*d* hybridization on the nearest neighbour sites [3]. Therefore, the present observations indicate that the *p-d* hybridization is weakened by the applied pressure in spite of shrinkage of interatomic distance. This suppression is more enhanced in order the replacement of *R*= Dy→Ho→Er.

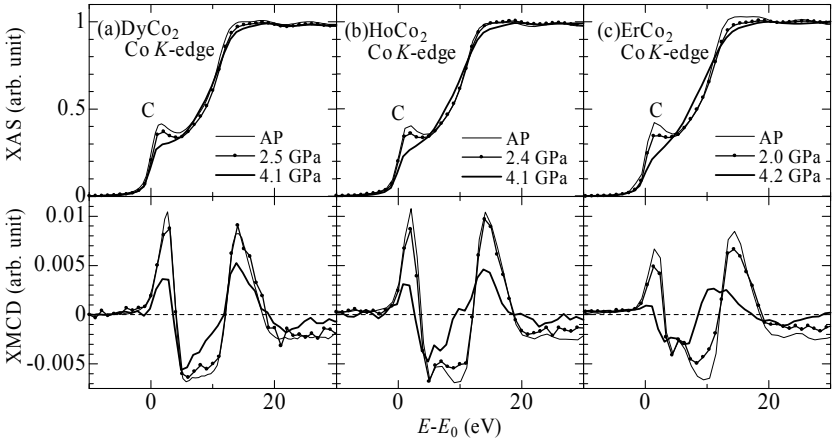

Figure 3. XAS and XMCD spectra at the Co *K*-edge in RCo_2.

The XMCD spectrum is characterized by the oscillatory profile mainly consisting of two positive peaks and a negative one. Because these spectral profiles differ from that in RCo_2 composed of non-magnetic *R* partner such as Y and Lu [9], the observed XMCD spectrum is strongly influenced by the molecular field provided by the heavy *R* partner [10]. As discussed for $ErCo_2$ in the previous paper [3], the Co *K*-edge dichroic spectrum can be classified into two regimes: the dichroic amplitude gradually decreases with a small change in spectral profile at low pressures (LP regime), and the

profile changes with a shift to the lower energy side at high pressures (HP regime). For the present observations, the oscillatory spectrum is changed for $HoCo_2$ and $ErCo_2$ under 4.2 GPa, so that an alteration form LP to HP regime occurs. The alteration in $ErCo_2$ is larger than that in $HoCo_2$. In the case of $DyCo_2$, the amplitude gradually decreases; however, the spectral shape is almost retained at 4.2 GPa. Hence, the Co magnetic state in $DyCo_2$ is still under the LP regime even at 4.2 GPa.

For the decrease in the dichroic amplitude in LP regime, it is considered that the molecular field from *R* partner is weakened due to less mixing between Co and *R* conduction electrons. Moreover, the change of spectral profile in HP regime is interpreted as the weak influence of the *R* moment. The present result indicates that the stability of the Co magnetic state is closely related to the pressure dependence of the *R* contribution. Therefore, the weak *R* contribution in HP regime gives rise to the steep decrease in M_{Co} for $HoCo_2$ and $ErCo_2$, while the more stable *R* contribution causes the moderate reduction in M_{Co} for $DyCo_2$ in LP regime.

4. Conclusion

We recorded the XMCD spectra at the *R* L_2-edge and Co *K*-edge in RCo_2 (*R*=Dy, Ho, and Er) under high pressure up to 4.2 GPa, and studied the relationship between M_{Co} and the molecular field from *R* partner. From the *R* L_2-edge XMCD, M_{Co} linearly decreases with increasing pressure, and the pressure-induced suppression of M_{Co} in $DyCo_2$ is smaller than those in $HoCo_2$ and $ErCo_2$. The Co *K*-edge XMCD shows that the effect of *R* molecular field on the Co magnetic state is also significantly suppressed under high pressure. The molecular field of Dy in $DyCo_2$ is a stronger effect on Co site compared with those in $HoCo_2$ and $ErCo_2$. As a result, it is revealed that the pressure-induced suppression of M_{Co} is strongly influenced by the *R*-Co magnetic interaction.

Acknowledgments

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