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MÖSSBAUER EFFECT ON Fe-Pd ALLOYS

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Abstract. - Mössbauer effect has been observed for disordered and ordered Fe_{1-x}Pd_x alloys. The internal field and the quadrupole and isomer shifts were obtained. The asymmetrical spectrum for 0.45 < x < 0.75 and the temperature dependence of isomer shifts for x = 0.32 were interpreted as a distribution of the quadrupole field and a second order Doppler effect respectively.

With respect to the invar effect and the martensite as well as order-disorder transformations for Fe_{1-x}Pd_x alloys, a number of studies has been reported mainly by us on the structural [1], magnetic [2-4] and thermal [5] properties. Particularly, we have found the martensite transformation concerning on a lattice change from fcc to fct at low temperature in x < 0.34. In this paper, from a microscopic standpoint, the Mössbauer effect on the whole ferromagnetic fcc phase will be presented to make clear roles of Fe atom in relation to the lattice and ordering transformations.

Disordered alloys, Fe_{1-x}Pd_x (0.30 ≤ x ≤ 0.80) were prepared by quenching into ice water from 1 100 °C, while ordered ones (0.50 ≤ x ≤ 0.75) by slow cooling with a rate of 50 °C per day from 1 000 °C. The fcc structure and the ordered CuAu and Cu3Au types were confirmed by X-ray diffraction for above samples. Mössbauer measurements were carried out in room temperature for all the samples and temperature variations were observed on x = 0.30 and x = 0.32 for the fcc-fct transformation.

In the observed spectra of disordered Fe_{1-x}Pd_x, the quadrupole shift is given to be zero and the isomer shift is not so large (0.01 to 0.03 mm/s) at room temperature. While a remarkable quadrupole shift, 0.2 mm/s, was observed in the ordered FePd. It is noted that, as shown in figure 1, the spectra for 0.45 ≤ x ≤ 0.75 show a remarkable asymmetric behavior for its width and height of the six absorption lines. One the other hand, an appreciable temperature variation of the central shift between 15 and 300 K was observed on x = 0.30 and 0.32, which will be explained below.

The spectra were analysed by the following usual expression.

\[ S(v) = \sum_{i=1}^{6} \int_0^{H_{\text{max}}} \frac{A_i P(H) \, dH}{1 + \left( (v - \alpha_i H - \delta_{\text{cs}} \pm eqQ) / \tau \right)^2} \]

(1)

where the notations are standard and \( \tau = 0.14 \) mm/s is used to fit the observed spectra. The distribution function of internal field, \( P(H) \), is calculated from a histogram method. The internal field \( H_{\text{in}}(x) \) in figure 2 is obtained from the overall splitting of the spectrum, which coincides with \( H = H_0 \) value at the peak of \( P(H) \). A linear relation of \( H_{\text{in}}(x) \) on \( x > 0.45 \) can be expressed by the Wertheim formula, but that for \( x < 0.45 \) is interpreted as a change of coefficients of the formula with the concentration. The half width, \( \Delta H(x) \), for the \( P(H) \) peak shows a parabolic behavior in figure 2, suggesting a binomial distribution of Fe

Fig. 2. - Concentration dependence of the internal field \( H_{\text{in}} \), the half with \( \Delta H \) of \( P(H) \) and the correlation term \( Q_m \) in the quadrupole effect for Fe_{1-x}Pd_x alloys.
atoms in the disordered state. For the ordered alloys, \( \Delta H \) takes 1/3 and 1/2 value for FePd and FePd\(_3\) respectively as compared with the disordered ones. In order to explain the asymmetric behavior shown above, the following expression is offered as a correlation between the internal field and the quadrupole shift,

\[
eq qQ = Q_0 + Q_m (H_0 - H) / \Delta H
\]

(2)

with \( H_0 - \Delta H \leq H \leq H_0 + \Delta H \), where \( H_0 \) is the internal field at the maximum of \( P(H) \) and \( Q_0 \) is an ordinal quadrupole term and \( Q_0 = 0 \) for the perfect disordered alloys. The term \( Q_m \) is attributable to a dipolar magnetic field due to a local ordering for the neighboring Fe atoms [7]. Thus, as shown in figure 2, \( Q_m \) shows a maximum at \( z \approx 0.65 \) corresponding to the maximum of the order-disorder transition point in the equilibrium phase diagram, where the local ordering remains appreciably.

As for the martensite transformation, fcc to fct, though some experiments revealed as an anomaly of the second order transition [1, 3, 4], the thermal expansion and the electrical resistivity did not show any anomaly at the transition. Then let us reconfirm this by means of the Mössbauer effect on the sample \( x = 0.32 \) with the transition at \( T_t = 188 \) K. The temperature dependence of the internal field, \( H_{\text{in}}(T) \), and the half width, \( \Delta H(T) \), are shown in figure 3, where a small depression for \( H_{\text{in}} \) and an inflection for \( \Delta H \) can be seen at \( T_t \).

On the other hand, the central shift of the spectra for \( x = 0.30 \) and 0.32 shows a remarkable temperature dependence, which usually consists of the isomer shift \( \delta_{\text{IS}} \) and the second order Doppler (SOD) shift, viz.

\[
\delta_{\text{IS}}(T) = \delta_{\text{IS}}(T) + \delta_{\text{SOD}}(T)
\]

(3)

For all Fe\(_{1-x}\)Pd\(_x\) alloys, the \( \delta_{\text{IS}} \) observed at room temperature (\( \delta_{\text{SOD}} = 0 \)) is very small, so its temperature variation can be neglected. While the SOD shift is a difference of SOD shift between the source kept at room temperature and the absorber:

\[
\delta_{\text{SOD}}(T) = \delta_{\text{SOD}}(R.T.) - \delta_{\text{SOD}}(T)
\]

(4)

where each \( \delta_{\text{SOD}} \) and \( \delta_{\text{SOD}} \) is expressed as follows by the Debye temperature \( \theta_D \), the averaged atomic mass \( m \) and the light velocity \( c \).

\[
\delta_{\text{SOD}} = 9k_B \theta_D / (16mc) +
\]

\[+(9k_B T (T / \theta_D)^3 / (2mc) ) \int_0^{T/\theta_D} \frac{e^x dx}{e^x - 1}
\]

(5)

Therefore, we can determine the SOD shift \( \delta_{\text{SOD}} \) of the absorber from the observed \( \delta_{\text{SOD}}(T) \), where the Debye temperature at 0 K is quoted from our specific heat measurement [5]. The remarkable anomaly at \( T_t \) in \( \delta_{\text{SOD}}(T) \), as shown in figure 3, means an effect of lattice softening due to the temperature variation of \( \theta_D \) for this martensite transition.

Fig. 3. – Temperature dependence of the internal field \( H_{\text{in}} \), the half width \( \Delta H \) and the second order doppler shift \( \delta_{\text{SOD}} \) for Fe\(_{0.68}\)Pd\(_{0.32}\).