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INVAR-PROBLEM AND MARTENSITIC TRANSFORMATION

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Résumé. — Les alliages d’invar sont situés dans la zone frontière entre les alliages présentant la transformation martensitique et ceux qui ne la présentent pas (dans le système Fe-Ni). Les expériences y compris des mesures d’effet Mössbauer montrent que cette relation de phases apporte une inhomogénéité fondamentale dans ces alliages, d’où une diversité correspondante des états magnétiques ; ceci est en relation directe avec les propriétés anormales de ces alliages. Des structures de transition intermédiaires entre une transformation martensitique $\gamma = \alpha$ jouent un rôle fondamental dans le problème de l’invar.

Abstract. — The Invar alloys are situated in the boundary zone between the alloys showing the martensitic transformation and the alloys without it in the Fe-Ni system. The experiments, inclusive of the Mössbauer effect measurement, showed that this phase relation results in bringing an essential inhomogeneity into this type alloy, which causes the corresponding variety of magnetic states in it; this is closely related to the anomalous properties of these alloys. That is, some transitional structures between the martensitic transformations $\gamma = \alpha$ play a fundamental role in the Invar-problem.

1. The anomalous properties of the Invar alloys (the Fe-Ni alloys with about 30 to 50 % Ni), such as the enormous volume magnetostriction, the extremely low thermal expansion, the zero or positive temperature coefficient of elastic modulus, etc. are well known, and the origin of these anomalies has been followed up since Guillaume. Now the present author has conceived an idea that this must be closely related with the inhomogeneous state of these alloys rather than the homogeneous $\gamma$ phase.

It was already reported [1] that the irreversible alloys, such as the 33 % Ni alloy, can be transmuted into the reversible alloys with a temperature range of almost zero thermal expansion, by means of many thermal cycles between the martensitic transformations $\gamma \Rightarrow \alpha$. The anomalous properties are also obtainable by quenching from a state on the way of the martensitic transformation $\alpha \rightarrow \gamma$ or the two-phase transformation $\alpha \rightarrow \alpha + \gamma$. Examples of the anomalies in thermal expansion, Young's modulus of elasticity and volume magnetostriction obtained in such a way are shown in figure 1 (a), (b) and (c), with the thick lines, and in these cases, the thin lines show the thermal hysteresis in the annealed states, respectively. As for the reversible alloys, such as the 36 % Ni alloy, it is well known that cold-work or quenching is required for obtaining the better characteristics. We tried another method of annealing these alloys at so low a temperature for so long a period of time as had been done by Owen [2] when he constructed the two-phase region in their equilibrium phase diagram. Our purpose was, however, to obtain a distorted $\gamma$ structure developed prior to the $\alpha$ phase precipitation, and as the result, it was found that the Invar characteristics were improved by such a treatment. The above facts show that the Invar alloys, which are situated at the boundary zone from the irreversible alloys to the reversible ones, should include a more or less amount of the $\alpha$-embryo, i.e. something like unripe martensite or distorted $\gamma$ structure, developed on the halfway stages in the martensitic transformations $\gamma \Rightarrow \alpha$, and this will give rise to the anomalous properties.

Fig. 1. — The anomalous properties obtained by quenching the $\alpha$ phase alloys just after (a) heating up to 550 °C, (b) heating up to 527 °C, and (c) heating at 500 °C for 6 hours.

2. The magnetic nature of such an inhomogeneous states was looked into by means of the Mössbauer effect. It was found out that the absorption spectrum obtained from these alloys is better suitable for an assembly of many hyperfine fields rather than for one or a few hyperfine fields in the annealed specimens as well as the cold-worked ones. In
this calculation, the distribution of the volume belonging to the parts with different hyperfine fields in the specimen was assumed to be a Gaussian distribution, which was very crude but sufficient for the present purpose; the details are to be given in another paper. The temperature variation in the spectrum for the γ phase alloys was also characteristic, as the shape of spectrum passed very gradually from the six-peak pattern to the paramagnetic-like single peak over a wide temperature range with rising temperature, as shown in figure 2 for the 36 % Ni alloy.

The same effect appears, however, at temperatures far from the Curie point, too; an example is shown in figure 3 for the 32 % Ni alloy at room temperature, which is rather similar to the pattern at \(-60^\circ\)C in zero field. Such a large effect of the external field has a resemblance to the para-process observed in the neighborhood of the Curie point in usual ferromagnet, but now it ranges over a wider scope of temperature. Thus it follows that the structural inhomogeneity, which is ascribed to the various grades of distortion from the cubic equilibrium lattice, has the corresponding magnetic inhomogeneity, in which the energy of exchange interaction may be mostly smaller than that in the equilibrium structure. The Invar-type alloy should be approximately regarded as an assembly of different magnetic states with different Curie points.

3. Finally there remains a question why so stable inhomogeneity must be caused in the Invar alloys, which the present author ultimately ascribes to the following crystallographical relationship [4]: if a shear twin in a f. c. c. lattice, such as occurring on the \{111\} planes in the [211] direction, proceeds only halfway, the resulting configuration is very close to that of a b. c. c. lattice. Hereupon he assumes that addition of nickel to iron into the γ phase Invar alloys results in diminishing the height of potential barrier between the positions of adjacent atoms in the f. c. c. lattice, so that this small height of the potential barrier makes it possible to form various transitional structures. The above kind of shearing process should be accommodated by dislocations already available in the f. c. c. lattice, and such dislocations would be increased by cold-work, thermal cycles, etc. As a matter of fact, it
could be observed that these treatments caused a marked development of dislocation networks associated with a large increase in intensity of the peak (111) $\gamma$ in the X-ray diffraction pattern. To sum up, the Invar alloys have a synthetic nature of the two characters coming from the $\gamma$ and the $\alpha$ structures.

References