Temperature and field dependence of magnetic anisotropy of alloys of nickel with palladium

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The anisotropy constant is investigated in nickel-palladium band ferromagnets in the concentration range 0–78.4 at.% Pd, at temperatures 77–T_c K, in fields up to 30 kOe. The temperature, concentration, and field dependences of the magnetic anisotropy constant indicate the existence of two contributions, of opposite signs, to the anisotropy constant, caused by doubly degenerate levels, which however are split under the influence of spin-orbit interaction, along the high-symmetry axes ΓL and ΓX of the Brillouin zone. The presence of two fundamental competing contributions to the anisotropy constant leads, in alloys with negative anisotropy at 0 K, to compensaton of the anisotropy at sóme temperature. The compensation temperatures are found as functions of the palladium content.

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1. INTRODUCTION

Calculations of the band structure of nickel have been made in a number of papers.[1-6] On the basis of this, its magnetic anisotropy has been calculated numerically.[7,8] It was calculated as the difference of energy of the electrons for magnetization in directions [111] and [100] of the crystal. The calculation showed that the largest contribution to the anisotropy comes from doubly degenerate levels, intersecting the Fermi level, of energy 3β and 5β, along the two high-symmetry axes ΓL and ΓX of the Brillouin zone; it was taken into account that these levels are split under the influence of spin-orbit interaction, the constant of which is $\xi = 7.5 \times 10^{-3}$ Ry. The amount of the splitting depends on the direction of the magnetization. The lowering of the energy along ΓL on splitting of the 3β levels is proportional to the quantity[8]

$$(l+m+n)^4+(l+m-n)^4+(l-m+n)^4+(-l+m+n)^4, \tag{1}$$

where l, m, n are the direction cosines of the magnetization vector. Along [100], this quantity is equal to 4.00; along [111], to 9.33. Thus 3β gives the [111] axis as the direction of easy magnetization. The contribution of the 5β levels along ΓX is proportional to

$$(3/4-l^2/2)^2+(3/4-m^2/2)^2+(3/4-n^2/2)^2. \tag{2}$$

For magnetization along [100] this expression is equal to 1.19, whereas along [111] it is equal to 1.02. Thus 5β gives the [100] axis as the direction of easy magnetization. All remaining directions of the Brillouin zone also gives a contribution to the anisotropy, similar to the contribution of the levels along ΓX,[8] but it has been found[7] that this contribution is small. Altogether the direction of easy magnetization in nickel remains the [111] axis.

Thus the magnetic anisotropy of nickel has two different contributions: a negative (in the sense of a direction of easy magnetization along [111]) contribution of the 3β levels along ΓL and a positive (with easy axis [100]) contribution of the levels along ΓX, and perhaps of all the other directions in the Brillouin zone. It should be mentioned that the special role of degenerate levels along high-symmetry axes in the magnetic anisotropy was first pointed out by Slonczewski.[9]

In connection with the facts set forth above, it is of interest to investigate the behavior of the magnetic anisotropy in alloys of nickel with palladium. Nickel and palladium are isoelectronic, both have the fcc lattice; they form a continuous series of solid solutions. In this system there is no formation of a superstructure with long-range order,[10,11] but a component of diffuse scattering of x rays, indicating a tendency toward short-range order, is observed.[11] Thus the system is nearly ideal, a rare case among the whole mass of binary systems. Along with this, there is in the nickel-palladium system an increase of the spin-orbit interaction parameter with increase of the palladium content, with consequent increase of the splitting of the doubly degenerate levels along ΓL and ΓX. The energy of the electrons depends not only on the amount of the splitting but also on the position of the Fermi level.

2. EXPERIMENTAL TECHNIQUE

The single crystals were grown by the Bridgman method. From them, spherical specimens, of diameter $4-6 \pm 0.005$ mm, were prepared. Orientation of the axes was accomplished by the x-ray method. The magnetic anisotropy was measured by the torque method, with accuracy 1.5%. At the measurement temperatures, the departure of the temperature from the prescribed value amounted to 1°; this departure was taken into account by introduction of a correction. The variation of the anisotropy constant with field was measured at the temperature of liquid nitrogen, and it may be assumed that the temperature was constant; other quantities pertaining to the apparatus meanwhile remained unchanged. The magnetic field was set and automatically maintained, with high accuracy, in an electromagnet from Bruker Instruments. Thus in the temperature and field measurements, the relative accuracy was an order of magnitude higher than the absolute. The anisotropy constant was measured at field 20 kOe in all the measurements except the field measurements.

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3. EXPERIMENTAL RESULTS AND DISCUSSION OF THEM

Figure 1 shows the temperature dependence of the anisotropy constant of nickel-palladium alloys containing 0–78.4 at.% Pd, in the disordered state (quenching from 900 °C in water). As is seen, the curves divide sharply into two groups. Up to 31.6 at.% Pd content, the alloys have a negative anisotropy constant; the corresponding curves run close together. Alloys with 51.4–78.4 at.% Pd have a positive constant. A characteristic feature is observed: all the alloys with negative anisotropy change the sign of the anisotropy constant at temperatures from 490 to 0 K (these limits correspond to pure nickel and to the alloy with 50 at.% Pd), as is evident from Fig. 2 and hereafter.

Figure 3 shows isotherms of the anisotropy constant. If we suppose that in the alloys, as in nickel, the anisotropy constant consists of two contributions with opposite signs, then the behavior of the isotherms can be explained as follows. In the initial part of the concentration (up to 20 at.% Pd), the negative part of the anisotropy increases, and then a rapid rise of the positive part leads to compensation at 47 at.% Pd at 77 K and then to a maximum at 64 at.% Pd. The subsequent drop of the constant is due to the rapid lowering of the exchange splitting. Thus the behavior of the isotherms justifies the assumption made regarding the existence of two competing contributions to the anisotropy of the alloys.

The rate of change of the constant with temperature, dK/dT, arouses interest. For the alloys with negative anisotropy, $dK/dT>0$; with positive, $dK/dT<0$. For both these groups of alloys, there are sharply expressed maxima of $|dK/dT|$, located close together; for the first group they are shifted closer to the ordinate axis by 0.2 T_c relative to the maxima for the second group (here T_c is the Curie point). The values of dK/dT and $K^{-1}dK/dT$ change sign at the anisotropy-compensation point. As a rule, the value of $|dK/dT|_{\max}$ for alloys of nickel with palladium is much larger than for iron, as is seen from Fig. 4. Hence it follows that all the alloys of nickel with palladium, as regards the type of anisotropy, belong to the nickel type.

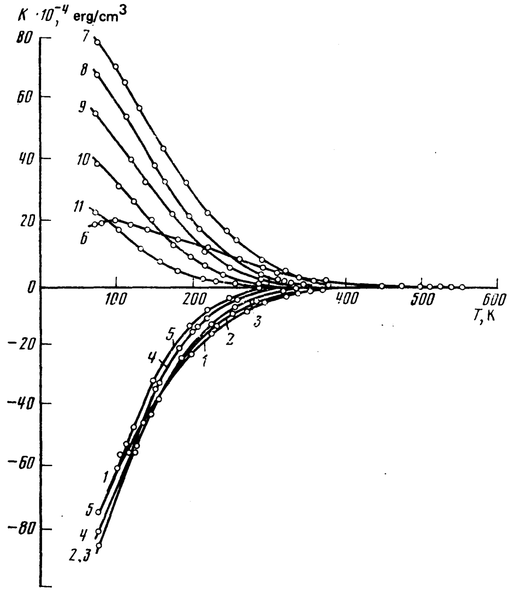

FIG. 1. Temperature dependence of the anisotropy constant of nickel-palladium alloys. 1, 0 at. % Pd; 2, 10.0; 3, 22.1; 4, 28.1; 5, 31.6; 6, 51.4; 7, 61.0; 8, 70.0; 9, 72.6; 10, 75.0; 11, 78.4 at. % Pd.

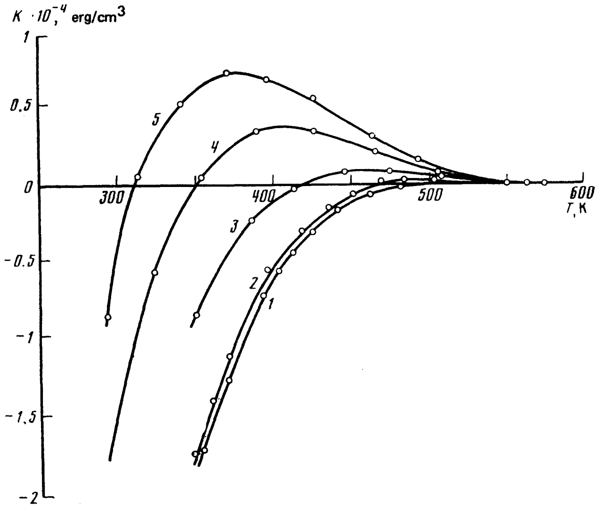

FIG. 2. Temperature dependence of the anisotropy constant with change of its sign. Notation the same as in Fig. 1.

This type of anisotropy is characterized as follows. As is well known, if we express the temperature dependence of the anisotropy constant in the form

$$K/K_0=(I_s/I_{s0})^n,$$

then for a Heisenberg ferromagnet in the low-temperature range the quantum theory gives $n=10$. Experiment shows that this is correct for iron but not for nickel, for which n reaches the value 70.[12] This indicates that nickel must be regarded as a band ferromagnet, in which the spin-wave contribution to the change of anisotropy with temperature is much smaller than the contribution of other mechanisms.

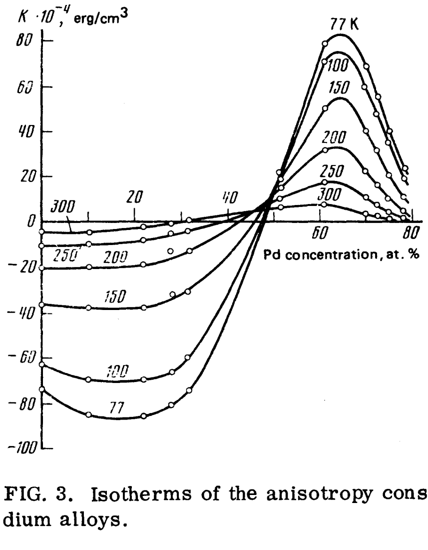

FIG. 3. Isotherms of the anisotropy constant of nickel-palladium alloys.

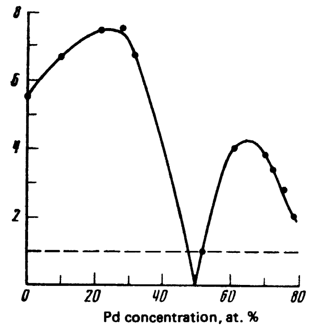

FIG. 4. Dependence of maximum $|dK/dT|$ on concentration. Dashed line, maximum $|dK/dT|$ for iron.

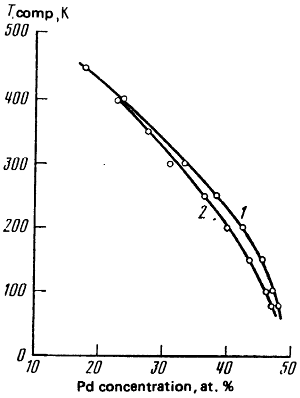

FIG. 5. Dependence of the anisotropy-compensation temperature on concentration: 1, quenched; 2, annealed.

The strong temperature dependence of the anisotropy constant of alloys of nickel has been attributed[8] to the temperature dependence of the Fermi level. In nickel, the Fermi level is raised by 0.013 Ry by a rise of temperature by 1000°. In alloys of nickel with palladium, a value of the shift close to this is to be expected.

In the alloys with negative anisotropy, a change of sign of the anisotropy constant occurs (see Fig. 2). This can be explained thus. Shift of the Fermi leads to a change of the relative position of the split levels ΓL and ΓX in the energy band bounded by the upper and lower Fermi levels. A rise of temperature lowers the negative anisotropy component more rapidly than the positive, except in the composition range 0–20 at.% Pd at low temperatures. This leads to compensation of the anisotropy. We determined the compensation temperatures, which are shown in Fig. 5. Here it is seen that apparently zero compensation point corresponds to 50 at.% Pd. Prolonged annealing of the alloys for 300 h, from 600 to 200 °C, led to no important change of the compensation curve in Fig. 5.

According to the quantum theory of magnetic anisotropy, the anisotropy constant increases with increase of the external field.[13] This is apparently valid also for band ferromagnets, since in nickel with palladium we observed a deviation from this rule, although the field dependence always remains exactly linear. For symmetric alloys in the nickel-palladium system, we obtained the following results:

28.1 at.% Pd, $K=-75.1\cdot10^4$ erg/cm^3, $dK/dH=-1.27$ erg/cm$^3\cdot$Oe,
51.4 at.% Pd, $K=21.0\cdot10^4$ erg/cm^3, $dK/dH=-0.67$ erg/cm$^3\cdot$Oe,
75.0 at.% Pd, $K=35.8\cdot10^4$ erg/cm^3, $dK/dH=0.67$ erg/cm$^3\cdot$Oe.

For prolonged annealing of the alloy with 51.4 at.% Pd, the field derivative of the anisotropy constant was unchanged as compared with its value in the quenched state.

From these data it is seen that in the two limiting alloys with 28.1 and 75.0 at.% Pd, the absolute value of the constant increases with increase of the field, whereas in the alloy with 51.4 at.% Pd it drops. The latter fact, it would seem, contradicts the quantum theory of anisotropy. But if we assume that the anisotropy contains two contributions of opposite sign, with field dependence close to that for the alloys with 28.1 and 75.0 at.% Pd, then everything becomes clear, and there is no contradiction with theory. The sum of the derivatives for the alloys with 28.1 and 75.0 at.% Pd, as is seen from the data presented, is -0.60 erg/cm^{3}Oe, which is close to the value of the derivative observed experimentally for the alloy with 51.4 at.% Pd. Thus the field dependence of the anisotropy also indicates the existence of two contributions to the anisotropy of alloys of nickel with palladium, as was found by numerical methods in pure nickel.

Our results correlate well with the data of an earlier paper.[14]

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