change interaction of ions in the planes is ferromagnetic), whereas the magnetic moments of ions in different planes are antiparallel (that is, the exchange interaction of ions belonging to different planes is antiferromagnetic).

We consider the experimental data obtained at temperatures above the transition point T_N. As is seen from Fig. 3, above the phase-transition point the nonlinear magnetic-moment relation $M(H)$ in weak fields, $H < 10$ kOe, disappears, and $M(H)$ is sufficiently well described by the expression $M(H) = \chi_\perp(T)H$. On increase of the magnetic field above 10 kOe, there appears a strong nonlinearity of the magnetic-moment dependence, characteristic of a "saturating" paramagnet; that is, when the magnetic moment $M(H)$ of the paramagnet is close to the magnetic moment of the magnetic ion at the prescribed temperature. The values of the magnetic susceptibility $\chi(T)$ above the phase-transition point T_N, presented in Fig. 5, were obtained by processing of the linear sections of the $M(H)$ relations of Fig. 3 at $H < 10$ kOe. By processing the magnetic-susceptibility data above the phase-transition point, one can obtain a Curie-Weiss law with the value $\Theta = 3 \pm 1$ K. The fact that the value of Θ was found to be positive supports the assumption that the $NaMnCl_3$ monocrystal is perhaps a quasi-two-dimensional antiferromagnet, in which the value of the ferromagnetic exchange interaction in the basal plane, perpendicular to the c axis, exceeds the value of the antiferromagnetic exchange interaction between the planes.

Thus the results of the research show that in antiferromagnetic $NaMnCl_3$ at $T = 4.2$ K, a second-order phase transition from the antiferromagnetic to the paramagnetic state occurs in magnetic fields $H_c = 15$ kOe. In the research, the magnetic-moment dependence $M(H)$ was investigated with the magnetic field H perpendicular and parallel to the high-order axis of the crystal, at various temperatures.

The authors are deeply and sincerely grateful to P. L. Kapitza for constant interest in the research, to A. S. Borovik-Romanov for direction of the research and discussion of the results, and to S. P. Kapitza and K. S. Aleksandrov for their interest in the research.

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2) The authors thank B. V. Beznosikov for providing the monocrystals for the investigation.

3) It is seen from Fig. 2 that when $H = H_c$, for given magnetic fields the $M(H)$ relation approaches a value of the magnetic moment somewhat different from $2M_0$; but with increase of H, there occurs an increase of the magnetic moment $M(H)$, and this gives a basis for supposing that in strong magnetic fields $M(H)$ asymptotically approaches the value $2M_0$.

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Translated by W. F. Brown, Jr.

Ferromagnetism of dilute PdNi alloys

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(Submitted 6 February 1978)
Zh. Eksp. Teor. Fiz. **75**, 585–598 (August 1978)

According to the experimental data, in dilute PdNi alloys there exist local moments associated with groups of two or three nearest-neighbor nickel atoms in the lattice. A theory which takes into account the polarization of the d-electrons on isolated nickel atoms due to the local moments, and the strong nonuniformity of the alloy, has made it possible to explain in a natural way the unique concentration dependences of the longitudinal susceptibility, magnetization, and Curie temperature of these alloys.

PACS numbers: 75.50.Cc, 75.10.–b, 75.30.Cr

INTRODUCTION

Alloys of palladium with nickel possess many distinctive properties, which make them an interesting object for study.

Unlike iron or cobalt atoms, a nickel atom in palladium does not possess a magnetic moment. Ferromagnetism has been reliably observed in alloys with a nickel concentration exceeding 2.3 at.%. Near this concentration the susceptibility of the alloy and also the electronic specific heat and electrical resistivity have a maximum.[1,2] At concentrations greater than 2.3 at.% the Curie temperature and saturation magnetic moment decrease rapidly with decreasing concentration.

It is customary, therefore, to suppose (see, e.g., Ref. 2) that a concentration phase transition from the paramagnetic to the ferromagnetic state occurs in the PdNi alloy at a nickel concentration $c \approx 2.3$ at.%. The point at which this transition occurs has been determined in theoretical papers[3,4] as the concentration at which the average (uniform) susceptibility becomes infinite.

However, in Ref. 5, in which the angular dependence of the cross section for diffuse scattering of neutrons in PdNi alloys with $c > 3$ at.% was measured, it had already been shown that the magnetization of the alloy is strongly nonuniform and that the concentration of magnetic centers in it is considerably smaller than the concentration of nickel atoms. The magnetic centers are surrounded by large clouds of polarized *d* electrons of palladium. The polarization in a cloud falls off with increasing distance r from the magnetic center like $r^{-1}e^{-r/R}$; the radius $R \approx 7$ Å. The authors of this paper put forward the hypothesis that the magnetic centers are pairs of nearest-neighbor nickel atoms in the lattice. Owing to the strong interaction of the nickel atoms in a pair, a local magnetic moment arises.

It was shown subsequently that magnetic centers also exist at nickel concentrations less than 2 at.% (Ref. 6). According to Ref. 6, clusters consisting of not less than three nearest-neighbor nickel atoms in the lattice possess magnetic moments. But in this case, as in alloys of the PdFe type, at $T = 0$ magnetic order should exist in the alloy at any nickel concentration.[7] Magnetic order arises in the system of local magnetic moments (groups of two or three nickel atoms); the indirect exchange interaction between them is due to correlated *d* electrons of palladium (and of isolated nickel atoms). Of course, at small values of c the concentration x of magnetic centers, proportional1) to c^3, may turn out to be so small that the magnetic-ordering temperature becomes practically equal to zero.

The indirect-exchange interaction potential $V(r)$ due to the strongly correlated electrons has ferromagnetic sign and decreases exponentially with the distance between the local moments, if this distance is not too great. At large distances the ferromagnetic potential is replaced by an oscillatory Ruderman-Kittel potential.[8] Therefore, in PdNi alloys a concentration phase transition is possible from a spin-glass state, when, over the average distances between magnetic centers, an oscillatory potential is dominant, to the ferromagnetic state, when the dominant potential over these distances is the ferromagnetic potential. Owing to the large range of the ferromagnetic part of the exchange potential it is evident that in PdNi alloys this transition occurs at a very small nickel concentration, less than 2 at.%. Henceforth we shall assume that the concentration of nickel atoms is above the critical concentration of the spin-glass – ferromagnetism transition and discard the oscillatory part of the potential.

Since, for $c \lesssim 2-3$ at.%, the average distance between local moments is considerably greater than the range of the exchange potential, the spatial fluctuations of the exchange interaction and magnetization are large. Therefore, the model of an almost uniform weak band ferromagnet that was postulated in Ref. 7 as the basis of a theory of the ferromagnetism of PdNi alloys is scarcely applicable to these alloys.

The principal difference between a PdNi alloy and alloys of the PdFe type is that in PdNi there are almost-magnetic nickel atoms near which the *d* electrons are correlated more strongly than in the matrix. Therefore, the average susceptibility of the alloy at small concentrations increases with increasing concentration of nickel. At the same time, the radius R of the polarized clouds around the magnetic centers also increases, and the number of isolated nickel atoms whose *d* electrons are appreciably polarized by the molecular field of a magnetic center increases rapidly. As a result, the average susceptibility of the alloy reaches a maximum at a certain nickel concentration c^*. The radius of the polarized cloud also reaches a maximum at concentrations close to c^*.

The Curie temperature T_C of the PdNi alloy, like that of alloys of the PdFe type,[8] increases exponentially rapidly with increase of $x^{1/3} \sim c$. If we take into account that for $c < c^*$ the radius R also increases with c, the increase of T_C will turn out to be still faster. Therefore, for $c < c^*$, T_C is extremely small, but, in a narrow range of concentrations $c \sim c^*$, it attains comparatively large values. The magnetic moment of the alloy also increases rapidly (more rapidly than c^3) with nickel concentration.

Thus, the model proposed in this paper, which takes into account both the formation of the local moments and the strong magnetic nonuniformity of the alloy, makes it possible to explain in a natural way the principal properties of PdNi alloys with $c \sim c^*$.

1. SUSCEPTIBILITY OF THE ALLOY. EFFECT OF LOCALIZED SPINS

We shall consider a strongly paramagnetic metal A (Pd) with impurities B (Ni), with impurity concentration $c \ll 1$. The Hubbard Hamiltonian of the strongly correlated *d* electrons of such an alloy has the form

$$\mathcal{H} = \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} a_{\mathbf{k}}^+ a_{\mathbf{k}} + I_A \sum_l a_{l\uparrow}^+ a_{l\uparrow} a_{l\downarrow}^+ a_{l\downarrow} + (I_B - I_A) \sum_l p_l a_{l\uparrow}^+ a_{l\uparrow} a_{l\downarrow}^+ a_{l\downarrow}. \qquad (1)$$

Here $\varepsilon_{\mathbf{k}}$ is the energy of the *d* electrons with momentum **k**; a^+ and a are electron creation and annihilation operators, respectively; I_A and I_B are the correlation energies of the *d* electrons on the matrix atoms (Pd) and impurity atoms (Ni), respectively, with $I_B > I_A$; $p_l = 1$ at sites l occupied by nickel and $p_l = 0$ at the other sites.

In the Hartree-Fock approximation the longitudinal magnetic susceptibility $\chi(ll')$ is determined by the following Dyson equation

$$\chi(ll')=\chi_0(r_{ll'})+2\Delta\sum_m \chi_0(r_{lm})p_m\chi(ll'), \tag{2}$$

where $\Delta=I_B-I_A$, and $\chi_0(r_{ll'})\equiv\chi_0(|\mathbf{r}_l-\mathbf{r}_{l'}|)$ is the susceptibility of pure palladium, which depends exponentially on r:

$$\chi_0(r)\sim r^{-1}e^{-r/R_0}. \tag{3}$$

The range $R_0\approx a/(1+\Gamma)\gg a$, where a is of the order of the lattice constant and Γ is the exchange scattering amplitude of the electrons. In palladium $1+\Gamma\sim 0.1$. The formula (3) is valid for $r\gg R_0$. At small r the susceptibility should tend to a certain constant: $\chi_0(r=0)\equiv X$. Iterating Eq. (2) and averaging each term over the distribution of impurities, we obtain a graphical series (see Fig. 1) determining the average susceptibility $\bar{\chi}(r_{ll'})$. Here, as usual, a solid line corresponds to the susceptibility χ_0 of the pure matrix, each dashed line to the perturbation energy Δ, and a cross with k outgoing lines to a polynomial $P_k(c)$.[9] Assuming that $c\ll 1$, in $P_k(c)$ we keep only the leading terms in c, i.e., we replace $P_k(c)$ by c. Taking into account the exponential dependence of the susceptibility $\chi_0(r)$ on r, it is easy to show that crossed graphs are smaller, in the parameter $cR/a\ll 1$, than uncrossed graphs with the same number of dashed lines, and, therefore, they can be discarded. As a result we obtain the following series for $\bar{\chi}(r_{ll'})$:

$$\bar{\chi}(r_{ll'})=\chi^0(r_{ll'})+ct_0\sum_m \chi_0(r_{lm})\chi_0(r_{ml'})$$
$$+c^2t_0\sum_{m\neq n}\chi_0(r_{lm})\chi_0(r_{mn})\chi_0(r_{nl'})+\ldots \tag{4}$$

Here, the single-impurity scattering matrix

$$t_0=2\Delta/(1-2\Delta X). \tag{5}$$

The local susceptibility X is almost independent of the correlation of the electrons; therefore, we shall neglect the effect of the impurities on it. Vanishing of the denominator in this expression means that a localized moment is formed on the impurity atom. For nickel atoms in palladium, $1-\Delta X\ll 1$, (Ref. 10), i.e., the isolated nickel atoms are almost magnetic.

In order to go over to the Fourier transform in (4) it is convenient to replace all intermediate susceptibilities $\chi_0(r_{lm})$, not containing the indices l and l', by $\chi_0(r_{lm})-X$. After this, in (4) we can sum over all sites without restriction, and for the Fourier transform $\chi(q)$ of the susceptibility we obtain

$$\chi(q)=\chi_0(q)/[1-\Sigma\chi_0(q)], \tag{6}$$

where

$$\Sigma=ct_0/(1+ct_0X). \tag{7}$$

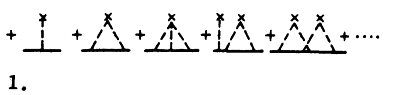

FIG. 1.

The expressions (6), (7) for $\chi(q)$ are internally consistent in the sense that they do not contain superfluous graphs,[11] and for $c\ll 1$ they describe the behavior of χ fairly satisfactorily. At a certain concentration $\bar{c}$ the denominator of (6) vanishes. However, the expression (6) is valid only at concentrations smaller than $\bar{c}$. The point is that, as c approaches $\bar{c}$ and $\chi(q)$ increases, the localized magnetic centers that arise on groups of three nearest-neighbor nickel atoms in the lattice begin to play an ever greater role. Because of the random disposition of the impurities, such groups of three are always present in the alloy. The polarization of the alloy due to the local moments leads to a decrease of the susceptibility. Therefore, the average susceptibility, as will be seen from the following, never becomes infinite. At small q it follows from (6), (7) that

$$\chi(q)=\chi_pa^{-2}(q^2+R^{-2})^{-1}, \tag{8}$$

where χ_p is the Pauli susceptibility,

$$R^2=R_0^2\frac{1+ct_0X}{1-ct_0(\chi_0-X)}, \tag{9}$$

and χ_0 is the uniform susceptibility of pure palladium, i.e., $\chi_0\equiv\chi_0(q=0)$.

As the concentration of nickel atoms increases, the radius of the polarized cloud around the magnetic atoms increases. The polarization of the d electrons leads to a decrease of the local susceptibility X and, correspondingly, to an increase of the denominator of the t-matrix (5). Since $1-2\Delta X\ll 1$, even a small decrease of X strongly decreases t. As a result, the effect of the nickel atoms on the susceptibility of the matrix is decreased. Therefore, with increase of nickel concentration the susceptibility and the radius of the polarized cloud pass through a maximum. Of course, the magnetic centers polarize the d electrons not only at those sites at which nickel atoms are situated but also on palladium atoms. But the polarization of the palladium d electrons is less important, since the correlation on the palladium sites is smaller than on the nickel sites. Evidence for the weak polarization of the palladium d electrons is the strong nonuniformity in the distribution of magnetic moment in the alloy, observed in Ref. 5. Therefore, in the following we shall take into account only the polarization of the d electrons on nickel atoms. Because of the strong magnetic nonuniformity of the alloy the degree of polarization of the nickel atoms, and, consequently, the single-impurity matrix t, depend on the configuration of the impurities about the localized spins. Therefore, the series, equivalent to (4), for the unaveraged susceptibility now has the form

$$\chi(ll')=\chi_0(r_{ll'})+\sum_m \chi_0(r_{lm})t_mp_m\chi_0(r_{ml'})$$
$$+\sum_{m\neq n}\chi_0(r_{lm})t_mp_m\chi_0(r_{mn})t_np_n\chi_0(r_{nl'})+\ldots \tag{10}$$

Here, t_m is the diagonal element of the T-matrix for a nickel impurity at the site m.

If the magnetization of the d electrons is small, i.e., $\mu_m\equiv|n_{m\uparrow}-n_{m\downarrow}|\ll n$, where $n_{m\sigma}$ is the concentration of electrons with spin σ at the point m and n is the total electron concentration, the correction to X that arises from the spin splitting of the d electrons is proportional to $(n_{m\uparrow}-n_{m\downarrow})^2$, so that

$$t_m=\frac{2\Delta}{1-2\Delta X+b(n_{m\uparrow}-n_{m\downarrow})^2n^{-2}}. \tag{11}$$

In Appendix I it is shown that $b=\frac{1}{2}$. The average susceptibility is obtained from (10) by averaging both over the configurations of nonmagnetic nickel impurities and over the configurations of localized spins:

$$\bar{\chi}(r_{ll'})=\chi_0(r_{ll'})+c\sum_m \chi_0(r_{lm})\langle t_m\rangle\chi_0(r_{ml'})$$
$$+c^2\sum_{m\neq n}\langle t_mt_n\rangle\chi_0(r_{lm})\chi_0(r_{mn})\chi_0(r_{nl'})+\ldots \tag{12}$$

The angular brackets denote the averaging over the configurations of localized spins, which, according to (11), reduces to averaging over the distribution of the magnetization μ_m of the d electrons at sites occupied by nickel atoms.

2. POLARIZATION OF THE ELECTRONS AROUND A MAGNETIC IMPURITY

If a magnetic impurity is placed at a point $\mathbf{r}_1$ in a metal, the polarization of the electrons at the point $\mathbf{r}$ is proportional to the susceptibility $\chi(\mathbf{r}_1,\mathbf{r})$:

$$\mu(\mathbf{r})=n_\uparrow(\mathbf{r})-n_\downarrow(\mathbf{r})\sim\chi(\mathbf{r}_1,\mathbf{r}). \tag{13}$$

According to (3), the polarization of the electrons around a magnetic impurity in pure palladium decreases exponentially, with a characteristic range R_0, with increase of the distance r from the impurity.

We shall examine how the nonmagnetic nickel impurities affect the polarization of the electrons.[2)] For this we shall investigate the dependence of the susceptibility $\chi(ll')$ of a PdNi alloy on the mutual disposition of the sites. We iterate Eq. (2) and consider a general term of this series:

$$\sum_{m,n,k\ldots s}\chi_0(r_{lm})p_m\chi_0(r_{mn})p_n\chi_0(r_{nk})p_k\ldots p_s\chi_0(r_{sl'}). \tag{14}$$

Let $r_{ll'}\gg R$. Since $\chi_0(r_{ij})$ depends exponentially on r_{ij} (to within a pre-exponential factor r_{ij}^{-1}, which is unimportant in the present case), the principal contribution to the sum (14) is given by those of its terms for which the length of the path from l to l', equal to $r_{lm}+r_{mn}+r_{nk}+\ldots+r_{sl'}$, exceeds the shortest distance $r_{ll'}$ by not more than R. The contribution of the other terms is exponentially small. It is easy to see that impurities satisfying this requirement are disposed within a cylinder of base radius of the order of $(Rr_{ll'})^{1/2}$ and length $r_{ll'}$ (Fig. 2). Consequently, the number of impurities in it is of the order of $Rr_{ll'}^2c\Omega^{-1}$, where Ω is the volume of the unit cell. If the distance $r_{ll'}$ from the localized spin to the point l' is small, so that $Rr_{ll'}^2c\Omega^{-1}\ll1$, in most cases only the shortest path from l to l', on which there is not a single nickel impurity, is optimal. Therefore, the susceptibility at the point l' is the same as in pure palladium. But if $Rr_{ll'}^2c\Omega^{-1}\gg1$, the number of optimal paths is large and the susceptibility at the point l' depends weakly on the actual disposition of the impurities, i.e., it coincides with the susceptibility averaged over the configuration of impurities $(\bar{\chi}(r))$.

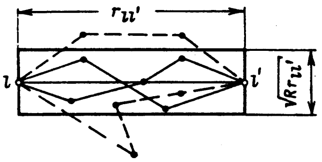

FIG. 2. Section of the cylinder inside which the optimal paths from the point l to the point l' are situated. The solid lines are optimal paths and the dashed lines nonoptimal paths, the circles indicate the positions l and l' of the impurities, and the points denote the positions of the impurities over which the summation in (14) is performed.

Thus, the polarization of the d electrons falls off with increasing distance from the localized center in accordance with the law

$$\mu(r)\sim\chi_0(r)\sim r^{-1}e^{-r/R_0},\quad R_0\ll r\ll(\Omega/cR_0)^{1/2},$$
$$\mu(r)\sim\bar{\chi}(r)\sim r^{-1}e^{-r/R},\quad r\gg(\Omega/cR)^{1/2}, \tag{15}$$

where R is determined by the expression (9). At not too large concentrations of localized spins, most of the isolated nickel atoms are situated far away from the nearest magnetic impurities, and, therefore, their magnetization is determined by the average susceptibility (15).

Assuming that $T\ll T_C$ or (for $T\gtrsim T_C$) that the alloy is in a strong magnetic field, aligning the localized spins, we have

$$\mu_m=IS\sum_l\bar{\chi}(r_{ml})\varphi_l, \tag{16}$$

where I is the integral of the exchange interaction of the d electrons with the localized spin S; $\varphi_m=1$ at sites occupied by a magnetic impurity, and $\varphi_m=0$ at the other sites.

Using the relation (15) it is not difficult to understand how the nickel impurities affect the indirect exchange interaction $V(r)$ of two localized spins. This interaction, as is well known, is proportional to the susceptibility of the d electrons. Consequently, the interaction between localized spins separated by a distance $r\ll(\Omega/cR)^{1/2}$ is the same as that for spins in pure palladium; i.e., $V(r)\sim r^{-1}e^{-r/R_0}$.

But if $r\gg(\Omega/cR)^{1/2}$, then $V(r)\sim\bar{\chi}(r)\sim r^{-1}e^{-r/R}$. Since $r\gg R$, even a small increase, due to the nickel impurities, of R as compared with R_0 strongly increases the interaction energy between the localized spins.

3. CONCENTRATION DEPENDENCE OF THE POLARIZED-CLOUD RADIUS, CURIE TEMPERATURE, AND MAGNETIZATION OF THE ALLOYS

We shall calculate the susceptibility (12) for large $r_{ll'}\gg R$. In this case, the nickel impurities that lie on optimal paths (Fig. 2) are located, in most cases, far apart; therefore, the molecular fields at these impurities are produced by different localized spins and can be assumed to be independent. Consequently,

$$\langle t_m t_n t_l \ldots \rangle = \langle t_m \rangle \langle t_n \rangle \langle t_l \rangle \ldots, \tag{17}$$

where $\langle t_m \rangle \equiv \langle t \rangle$, obviously, does not depend on the index m. Substituting (17) into (12), we obtain for $\bar{\chi}(r_{ll'})$ a series that differs from (4) only by the replacement of t_0 by $\langle t \rangle$, so that the radius is given by

$$R^2 = R_0^2 \frac{1 + c\langle t \rangle X}{1 - c\langle t \rangle (\chi_0 - X)}. \tag{18}$$

The quantity $\langle t \rangle$ itself depends on R, and, therefore, the expression (18) is an equation for R.

The expression (16) for the magnetization is analogous to the expression for the molecular field in a system of localized spins whose exchange interaction is $V(r) \sim \bar{\chi}(r)$. The distribution function of the molecular fields in such a system was studied in Ref. 12, in which it was assumed that $V(r) \sim e^{-r/R}$. Since at large distances $r \gg R$ the pre-exponential factor r^{-1} in $\bar{\chi}(r)$ is unimportant in comparison with the exponential dependence, we can represent $\bar{\chi}(r)$, like $V(r)$, in the form

$$\bar{\chi}(r) = X e^{-r/R}. \tag{19}$$

The distribution function of the magnetization μ_m in this approximation coincides with the molecular-field distribution function from Ref. 12, so that

$$\langle t \rangle = t_0 \int_0^\infty \frac{W(\tilde{\mu})\, d\tilde{\mu}}{1 + \alpha^2 \tilde{\mu}^2}. \tag{20}$$

Here,

$$\alpha \approx ISX/n(1 - 2\Delta X)^{1/2} \gg 1, \quad \tilde{\mu} = \mu/ISX.$$

The magnetization distribution function

$$W(\tilde{\mu}) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\lambda\tilde{\mu} - D(\lambda)}\, d\lambda, \tag{21}$$

where

$$D(\lambda) = i\nu\lambda \int_0^1 dx \ln^3 x e^{i\lambda x}; \tag{22}$$

here, $\nu = 4/3\pi N e^3 = \nu_0 (R/R_0)^3$, where N is the number of localized spins in unit volume.

Together with (20)–(22), the relation (18) is a self-consistent equation for the determination of the radius R. The integral (20) can be calculated analytically in two limiting cases.

1) The magnetization $\tilde{\mu}$ on a given nickel impurity is produced by one localized spin (the nearest to it). It follows from (16) and (19) that, in order that the magnetization at the given site not exceed $\tilde{\mu}$, the nearest localized spin should be situated at a distance not smaller than $r(\tilde{\mu}) = R \ln \tilde{\mu}^{-1}$. The number of spins in a layer of radius $r(\tilde{\mu})$ and thickness R, producing approximately the same magnetization $\tilde{\mu}$, is $4\pi r^2 R N = 3\nu \ln^2 \tilde{\mu}^{-1}$. If $3\nu \ln^2 \tilde{\mu}^{-1} \ll 1$, the magnetization at the given site is produced by one spin. Therefore, $W(\tilde{\mu})$ can be found by replacing the distance r in the Poisson nearest-neighbor distribution function $w(r) = 4\pi r^2 N \exp(-4/3\pi N r^3)$ by $r(\tilde{\mu})$, i.e.,

$$W(\tilde{\mu}) = \frac{3\nu}{\tilde{\mu}} \ln^2 \frac{1}{\tilde{\mu}} e^{-\nu \ln^3 \tilde{\mu}^{-1}}. \tag{23}$$

It follows from the foregoing that the applicability of this expression is bounded from below by the inequality $3\nu \ln^2 \tilde{\mu}^{-1} \ll 1$. In addition, since one localized spin cannot produce a magnetization greater than unity, the formula (23) is inapplicable for $\tilde{\mu} > 1$.

Substituting (23) into (20), putting the upper limit in (20) equal to unity in accordance with what has been said, and integrating by parts, we obtain, to within small terms of order α^{-1},

$$\langle t \rangle = t_0 \int_0^1 \frac{d\tilde{\mu}}{(1 + \alpha^2 \tilde{\mu}^2)} e^{-\nu \ln^3 \tilde{\mu}^{-1}}. \tag{24}$$

The principal contribution to this integral is given by the region $\tilde{\mu} \sim \alpha^{-1} \ll 1$. Since $\nu \ln^2 \tilde{\mu}^{-1} < 1$, in this region the function $\exp(-\nu \ln^3 \tilde{\mu}^{-1})$ varies very slowly with $\tilde{\mu}$ and can be taken outside the integral at the point $\tilde{\mu} = \alpha^{-1}$, so that

$$\langle t \rangle = t_0 e^{-\nu \ln^3 \alpha}. \tag{25}$$

This formula is applicable if $3\nu \ln^2 \alpha \ll 1$. Substituting (25) into (18) we obtain the following equation for $z \equiv R/R_0$:

$$z^2 = 1 + \frac{c t_0 \chi_0 \exp(-\nu_0 z^3 \ln^3 \alpha)}{1 - c t_0 (\chi_0 - X) \exp(-\nu_0 z^3 \ln^3 \alpha)}, \tag{26}$$

where $\nu_0 = 4\pi N R_0^3/3$. Since the polarization of the electrons in the given case is due entirely to one local spin, the orientation of the other spins is unimportant. Therefore, Eq. (26) is also applicable in the paramagnetic phase. The solution of this equation has a maximum at a nickel concentration c that can be determined from the condition

$$c \frac{d\nu_0}{dc} z^3 \ln^3 \alpha = 1. \tag{27}$$

In particular, if $x \sim c^3$, the relation (27) reduces to $\nu_0 z^3 \ln^3 \alpha = \frac{1}{3}$. Since $\alpha \gg 1$, the inequality $3\nu \ln^2 \alpha \ll 1$ is still fulfilled at the point of the maximum, i.e., Eq. (26) describes the dependence $R(c)$ in a broad range of concentrations, including the value of c at which $R(c)$ has the maximum.

2) At large concentrations of localized spins the inequality $3\nu \ln^2 \alpha \gg 1$ is valid, i.e., the magnetization $\tilde{\mu} \sim 1/\alpha$ at the given site is produced by several localized spins. Since in the region $\nu \ln^2 \tilde{\mu}^{-1} \gg 1$ the distribution function $W(\tilde{\mu})$ falls off with $\tilde{\mu}$ more rapidly than by a power law,[12] the principal contribution to the integral (20) in this case is given by values of $\tilde{\mu} \gg \alpha^{-1}$, so that in the denominator of the integrand in (20) we can discard unity in comparison with $\tilde{\mu}^2 \alpha^2$. The integral that remains is calculated in Appendix II.

For $\langle t \rangle$ we obtain

$$\langle t \rangle = \frac{t_0}{\alpha^2} \frac{\pi^{1/2} \exp(-2C_E)}{(6\nu)^{1/4}} \exp\left\{ \left(\frac{2}{3} \right)^{3/2} \frac{2}{\nu^{1/2}} \right\}, \tag{28}$$

where $C_E = 0.5772$ is Euler's constant. We recall that $\nu = \nu_0 (R/R_0)^3$. In the concentration region in which formula (28) is valid, the radius R decreases with increase of c.

If the magnetic impurities are groups of three nickel atoms, the average distance between them is $r_c \sim \Omega^{1/3}c^{-1} \gg (\Omega R^{-1}c^{-1})^{1/2}$. Consequently, the interaction energy of localized spins separated by a distance of the order of the average distance is equal to $V(r) \sim r^{-1}e^{-r/R}$ (see above). This means that the concentration dependence of T_C is determined by the relation[8]

$$T_c \sim N^{1/3} \exp\{-0.89/RN^{1/3}\}, \tag{29}$$

where the concentration of magnetic impurities is related to c in the following way[6]:

$$N \sim x = {}^1/_3 c[1-(1-c)^{12}-12c(1-c)^{18}]. \tag{30}$$

Since, for small c, not only N but also R increases with increase of c, T_C increases very rapidly with increasing concentration of nickel. On further increase of c the radius R begins to decrease, and the growth of T_C slows down.

The saturation moment M of the alloy is the sum of the moments of the localized spins and of the polarized clouds of d-band electrons that surround them. The moment of each cloud is $\sim \int \mu(r)d^3r$. Therefore, according to (15) we have

$$M \sim xR_0^2, \quad \rho > R\ln(R\rho/R_0^2), \quad \rho \sim (\Omega/cR)^{1/2}, \tag{31a}$$

$$M \sim xR\rho e^{-\rho/R}, \quad R \ll \rho < R\ln(R\rho/R_0^2), \tag{31b}$$

$$M \sim xR^3, \quad R \gg \rho. \tag{31c}$$

The last inequality is fulfilled when the number of nickel atoms in a volume of radius R is greater than unity, so that, for all r, the function $\mu(r) \sim \bar{\chi}(r) \sim r^{-1}e^{-r/R}$.

It follows from (31) that for small c the moment grows with nickel concentration like c^3; then the rate of growth increases, and, finally, at large c, when R decreases with increase of c, the moment increases with c more slowly than c^3.

4. THE UNIFORM SUSCEPTIBILITY

According to (10) the uniform longitudinal susceptibility of the alloy is

$$\chi = \frac{1}{\mathcal{N}}\sum_{ll'}\chi(ll') = \frac{1}{\mathcal{N}}\sum_{ll'}\chi^0(r_{ll'}) + \frac{1}{\mathcal{N}}\sum_{mll}\chi^0(r_{lm})t_m p_m \chi^0(r_{ml'})$$

$$+\frac{1}{\mathcal{N}}\sum_{mnll'}\chi^0(r_{lm})p_m t_m \chi^0(r_{lm})p_n t_n \chi^0(r_{nl'}), \tag{32}$$

where $\mathcal{N}$ is the number of sites in the lattice. Since $\chi^0(r)$ decreases exponentially with increase of r, in each term of the series (32), with exponential accuracy, the principal contribution is given by groups of impurities concentrated in small regions with linear dimensions of the order of R. The polarizations of the electrons on all the impurities of such a group almost coincide. Therefore, when averaging the series (32) over the configurations of the magnetic impurities we can replace averages of the type $\langle t_m t_n \ldots t_p\rangle$, containing f factors, by $\langle t_m^f\rangle$. Averaging (32) next over the configurations of the nonmagnetic impurities, we obtain

$$\chi = \left\langle \chi_0\left[1-\frac{ct_l}{1+ct_lX}\chi_0\right]^{-1}\right\rangle. \tag{33}$$

We recall that $\chi_0 = \chi_0(q=0)$.

The magnetization distribution function $W(\tilde{\mu})$, by means of which the averaging in (33) must be performed, depends now on the character of the range R_1 determining the decay of the polarization with the distance from those localized spins whose "sphere of influence" contains groups of closely spaced nickel impurities. In the calculation of R_1, as in the derivation of (33), all the t_l in each term of the series (12) can be assumed to be the same. Therefore, $R_1 \sim \sqrt{\chi}$, so that the relation (33) constitutes the following equation for χ:

$$\frac{\chi-\chi_0}{\chi_0} = \frac{ct_0\chi_0}{\alpha^2}\int_0^\infty d\tilde{\mu}\frac{W(\tilde{\mu})}{\beta+\tilde{\mu}^2}, \tag{34}$$

where $\beta = \alpha^{-2}[1-ct_0(\chi_0-X)]$. Denoting $\chi/\chi_0 \equiv y$, we obtain from (34)

$$y-1 = \frac{ct_0\chi_0}{1-ct_0(\chi_0-X)}\exp\{-\nu y^{3/2}\ln^3\beta^{-1/2}\} \tag{35}$$

for $\nu_0 y^{3/2}\ln^2\beta^{-1/2} \ll 1$, and

$$y-1 = \frac{ct_0\chi_0}{\alpha^2}\frac{\pi^{1/2}e^{-2C_E}}{(6\nu_0)^{1/4}y^{3/8}}\exp\left\{\left(\frac{2}{3}\right)^{3/2}\frac{2}{\nu_0^{1/4}y^{1/2}}\right\} \tag{36}$$

for $\nu_0 y^{3/2}\ln^2|\beta|^{-1/2} \gg 1$. It follows from (35) and (36) that $y(c)$ has a maximum at $\nu_0 y^{3/2}\ln^2\beta^{-1/2} \approx 1$. We note that Eq. (35), like (26), is also valid in the paramagnetic phase (or in the spin-glass state).

5. COMPARISON WITH EXPERIMENT

The equations for the radius R and uniform susceptibility χ contain three parameters: $t_0\chi_0$, t_0X, and α^2. In principle the parameter $t_0\chi_0$ can be determined if we know the susceptibility for very small c, since $\chi^{-1}d\chi/dc)_{c\to 0} = t_0\chi_0$. However, there are no reliable data at sufficiently small c. We shall take the value $t_0\chi_0 = 70$, obtained from an analysis of the experimental data in Ref. 4. According to Ref. 13, $\chi_0 \approx 7\chi_p$. Assuming that[4] $2\Delta\chi_p \approx 0.3$, we obtain $1-2\Delta X \approx 0.03$; $t_0X \approx 35$. The concentration dependence of the uniform susceptibility χ and radius R, obtained with the above values of the parameters for two values of α^2 ($\alpha^2 = 20$ and $\alpha^2 = 40$), is shown in Figs. 3 and 4. It can be seen that χ and R are not very sensitive to the value of the parameter α^2.

Figure 3 shows the experimental values of the susceptibility, obtained in Ref. 1 at $T < 2$ K and in a magnetic field greater than 20 kOe, when all the localized spins are aligned parallel to each other. The calculated susceptibility agrees well with the experimental susceptibility.

The experimental values of the ratio R/R_0 given in Fig. 4 were found from the data of Ref. 5, with $R_0 = 4.3$ Å from Ref. 8. The agreement between the theoretical and experimental results must be regarded as satisfactory, if we take into account the uncertainty in the determination of R associated with the fact that the

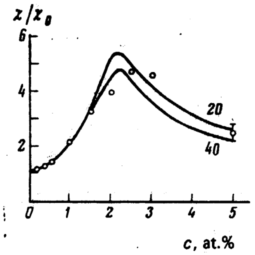

FIG. 3. Concentration dependence of the uniform longitudinal susceptibility. The values of the parameter α^2 are shown in the figure. The experimental points are taken from Ref. 1.

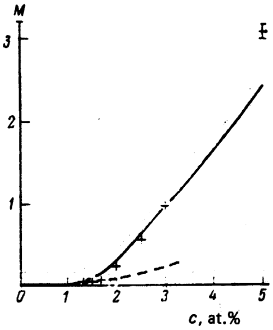

FIG. 5. Dependence of the saturation moment (in arbitrary units) on the nickel concentration. The solid curve is plotted from formula (31c) and the dashed curve from formula (31a). The experimental points are taken from Refs. 1 and 6; $\alpha^2=40$.

linear dimensions of a magnetic cluster are smaller than R by a factor of only 2, approximately. Changing the parameters slightly (e.g., increasing α^2 or t_0X), we can easily achieve better agreement.

Figure 5 shows the theoretical dependence of the saturation moment on the nickel concentration, calculated for $\alpha^2=40$. The solid curve is plotted from formula (31c) and the dashed curve from formula (31a). Almost the same dependences are obtained for $\alpha^2=20$. The experimental points, taken from Refs. 1 and 6, lie satisfactorily on the dashed curve for $c<2$ at.% and on the solid curve for $c \geqslant 2$ at.%. The crossover from one dependence to the other at $c \lesssim 2$ at.% seems to be entirely reasonable, if we take into account that $R \approx 8-9$ Å.

Figure 6 shows the theoretical dependence of T_C, calculated from formula (29) for values of R obtained from the solution of Eq. (20) ($\alpha^2=40$) and with $R_0=4.3$ Å. Since, generally speaking, Eq. (20) is applicable only when the spin orientations are parallel, the values of R found from (20) can differ appreciably from the true values at $T \approx T_C$ in the concentration region $3\nu \ln^2\alpha > 1$ ($c \gtrsim 0.03$). Therefore, the dependence of T_C on c obtained in this way has only qualitative meaning.

The Curie temperature of PdNi alloys has been measured in Ref. 14 for $c>2.5$ at.% and in Ref. 2 for $c>2.3$ at.%, the values of T_C obtained in the first of these papers being considerably higher than those obtained in the second. The character of the concentration dependence of T_C on c in Fig. 6 is close to the experimental dependence given in Ref. 2. There is, however, one substantial quantitative discrepancy. The steep rise of T_C on the theoretical curve begins at $c \approx 1.6$ at.%, whereas in Ref. 2 the rapid growth of T_C begins at $c \approx 2.3$ at.%. This discrepancy can be explained, first, by the fact that the values of R used for the calculation of T_C are too low, as already noted, when $c \gtrsim 0.03$. Moreover, it is not ruled out that for $c \lesssim 0.02$ the oscillatory part of the exchange potential already substantially decreases the energy of the exchange interaction of localized spins over the average distance, and, consequently, also decreases T_C.

APPENDIX I

We introduce the susceptibility

$$\chi_{\sigma\sigma'}(ll')=\frac{1}{2}\int_{-1/T}^{1/T}\langle T_\tau a_\sigma^+(l\tau)a_\sigma(l\tau)a_{\sigma'}^+(l'0)a_{\sigma'}(l'0)\rangle d\tau, \tag{I.1}$$

where $\sigma=\uparrow,\downarrow$. The longitudinal susceptibility is

$$\chi(ll')={}^1/_4[\chi_{\uparrow\uparrow}(ll')+\chi_{\downarrow\downarrow}(ll')-\chi_{\uparrow\downarrow}(ll')-\chi_{\downarrow\uparrow}(ll')]. \tag{I.2}$$

If in the palladium there is one nickel impurity, located at site 1, then

$$\chi_{\sigma\sigma'}(1l')=\overset{0}{\chi}_{\sigma\sigma'}(1l')-\Delta\sum_{\alpha\neq\beta}\chi_{\sigma\alpha}{}^0(11)\chi_{\beta\sigma'}(1l'). \tag{I.3}$$

The system (I.3) can be divided into two systems of equations for $\chi_{\uparrow\uparrow}(1l')$, $\chi_{\uparrow\downarrow}(1l')$ and $\chi_{\downarrow\downarrow}(1l')$, $\chi_{\downarrow\uparrow}(1l')$. Solving these, substituting the solutions into (I.2), and taking into account that $\chi^0_{\uparrow\downarrow}=\chi^0_{\downarrow\uparrow}$, we obtain

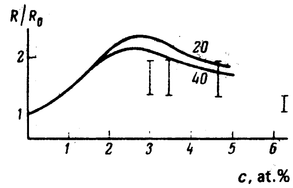

FIG. 4. Dependence of the radius of the polarized cloud around a localized spin on the nickel concentration. The values of the parameter α^2 are shown in the figure. The experimental results are from the data of Ref. 5.

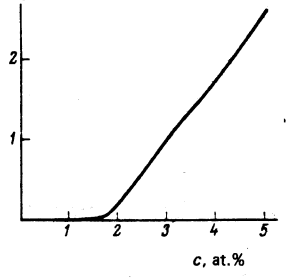

FIG. 6. Dependence of the Curie temperature (in arbitrary units) on the nickel content; $\alpha^2=40$.

$$\chi(1l')=\{\chi^0(1l')+{}^1\!/\!_4\Delta[(\chi_{\downarrow\downarrow}{}^0(1l')-\chi_{\uparrow\uparrow}{}^0(1l'))(\chi_{\uparrow\uparrow}{}^0(11)+\chi_{\uparrow\downarrow}{}^0(11))+(\chi_{\uparrow\uparrow}{}^0(1l')-\chi_{\uparrow\downarrow}{}^0(1l'))(\chi_{\downarrow\downarrow}{}^0(11)+\chi_{\uparrow\downarrow}{}^0(11)]\}[1+\Delta(\chi_{\downarrow\downarrow}{}^0(11)\chi_{\uparrow\uparrow}{}^0(11))^{1/2}+\Delta\chi_{\uparrow\downarrow}{}^0(11)]^{-1}[1+\Delta\chi_{\uparrow\downarrow}{}^0(11)-\Delta(\chi_{\downarrow\downarrow}{}^0(11)\chi_{\uparrow\uparrow}{}^0(11))^{1/2}]^{-1}. \qquad (I.4)$$

Assuming the splitting of the d-band of palladium to be weak, we can neglect the difference between $\chi^0_{\uparrow\uparrow}$ and $\chi^0_{\downarrow\downarrow}$ everywhere except in the last factor in this expression. Then,

$$\chi(1l')=\frac{\chi^0(1l')}{1-\Delta(\chi_{\downarrow\downarrow}{}^0(11)\chi_{\uparrow\uparrow}{}^0(11))^{1/2}+\Delta\chi_{\uparrow\downarrow}{}^0(11)} \qquad (I.5)$$

and in the same approximation,

$$\chi(ll')=\chi^0(ll')+\chi^0(l1)t_1\chi^0(1l'), \qquad (I.6)$$

where

$$t_1=\frac{2\Delta}{1-\Delta(\chi_{\downarrow\downarrow}{}^0(11)\chi_{\uparrow\uparrow}{}^0(11))^{1/2}+\Delta\chi_{\uparrow\downarrow}{}^0(11)}. \qquad (I.7)$$

Since the denominator in this expression is symmetric in the indices $\uparrow, \downarrow$, the correction due to the spin splitting at the point 1 is proportional to $(n_\uparrow(1)-n_\downarrow(1))$.[2] In the calculation of $\chi^0_{\sigma\sigma'}(11)=\Sigma_q\chi^0_{\sigma\sigma'}(q)$ we can confine ourselves to the free-electron model, since the principal contribution in the integral over q is given by large values of q and the electron correlation is unimportant. Simple calculations lead to the formula (11) with $b=\frac{1}{2}$.

APPENDIX II

We shall consider the integral

$$A=\int_0^\infty\frac{d\bar{\mu}W(\bar{\mu})}{\bar{\mu}^2} \qquad (II.1)$$

Using the representation

$$1/\bar{\mu}^2=\int_0^\infty e^{-\bar{\mu}t}t\,dt,$$

we obtain

$$A=\int_0^\infty dt\,t\int_0^\infty d\bar{\mu}W(\bar{\mu})e^{-\bar{\mu}t}. \qquad (II.2)$$

According to Ref. 12, the inner integral in (II.2) is equal to $e^{-D(it)}$, so that

$$A=\int_0^\infty dt\,te^{-D(it)}, \qquad (II.3)$$

Since $\nu\ll 1$, the principal contribution to (II.3) is given by $t\gg 1$. The asymptotic form of $D(it)$ at large values of t is (cf. Ref. 12)

$$D(it)=\nu\ln^3 t+3C_E\nu\ln^2 t+\ldots \qquad (II.4)$$

Substituting (II.4) into (II.3) (we put the lower limit in (II.3) equal to unity) and changing to the variable $x=\ln t$, we obtain

$$A=\int_0^\infty dx\exp\{2x-\nu x^3-3C_E\nu x^2\}. \qquad (II.5)$$

This integral is calculated by the method of steepest descents. Finally, we obtain

$$A=\frac{\pi^{1/2}\exp(-2C_E)}{(6\nu)^{1/4}}\exp\left\{\left(\frac{2}{3}\right)^{3/2}\frac{2}{\nu^{1/2}}\right\}. \qquad (II.6)$$

[1] Henceforth we shall assume for definiteness that nickel clusters containing not less than three nickel atoms possess a magnetic moment.

[2] A magnetic impurity in a PdNi alloy, as already noted, is a cluster of three nearest-neighbor nickel atoms in the lattice. In this case it is possible to speak of a point of spin localization only so long as the characteristic distances in the problem are large compared with the size of the cluster.

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Translated by P. J. Shepherd