# Spin waves in dilute ferromagnetic alloys of the PdFe type

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The propagation of spin waves in alloys of the type $Pd_{1-x}Fe_x$ is considered. For sufficiently low impurity concentrations such alloys are described by a model of randomly positioned magnetic centers with an exchange interaction falling off exponentially with distance. Using percolation-theory ideas it is shown that in this case the mass of the spin waves has the same exponential dependence on $x$ as the Curie temperature. The pre-exponential factor is also found. At high concentrations (but with $x \ll 1$), when the magnetization of the electrons of the matrix is substantial, this dependence becomes less steep. The results obtained are in satisfactory agreement with experiment.

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## INTRODUCTION

In dilute ferromagnetic alloys of the PdFe type the onset of magnetic order is due to the indirect exchange interaction of the localized impurity spins via the $d$-electrons of the matrix. As a result of the strong correlation of the electrons in the $d$-band of palladium the sign of the indirect interaction is ferromagnetic. If the magnetic-impurity concentration (per atom of the matrix) $c \ll 1 - I\rho(\varepsilon_F)$, where $I$ is the correlation energy and $\rho(\varepsilon_F)$ is the density of states of the $d$-electrons at the Fermi surface (for palladium, $1 - I\rho(\varepsilon_F) \approx 0.1$), the spin splitting of the $d$-band in the ferromagnetic state is small. In this case the magnetic properties of the alloy are well described by a model of randomly positioned spins, the exchange interaction between which varies with distance in accordance with the law

$$V(r) = V_0 \frac{R}{r} e^{-r/R}. \qquad (1)$$

The range $R$ of the potential is $R \approx a(1 - I\rho(\varepsilon_F))^{-1/2}$ ($a$ is the lattice constant). At low concentrations the range $R$ is much smaller than the mean distance $r_c \sim n_i^{-1/3}$ between impurities ($n_i$ is the impurity concentration in $cm^{-3}$).[1-3] Therefore, the exchange interaction between the spins depends strongly on the specific configuration. But if $c \gtrsim 1 - I\rho(\varepsilon_F)$, the spin splitting of the $d$-band of palladium is comparable with the width of the $d$-band, and the whole alloy is magnetized almost uniformly. Experiment shows[4] that uniform magnetization in PdFe sets in for $c \gtrsim 0.04$.

Because of the conservation law for the total spin, weakly-damped low-frequency spin waves with a quadratic dispersion law should exist for any degree of nonuniformity. However, the dependence of the mass of the spin waves on the impurity concentration is determined by the specific situation.

Spin waves in alloys of the PdFe type were first investigated theoretically in[5,6]. The authors of these papers, having confined themselves to the region of low concentrations $c \ll 1 - I\rho(\varepsilon_F)$, assumed at the same time that the molecular field acting on the localized spins was uniform. As a result they obtained an incorrect concentration dependence for the spin-wave mass, and also for the Curie temperature $T_c$. Unfortunately, these papers have been widely used in attempts to interpret experimental results (cf., e.g.,[7]).

In the present paper the concentration dependence of the spin waves is treated both in the region of low concentrations and in the region of high ($1 \gg c \gg 1 - I\rho(\varepsilon_F)$) concentrations. In the former case it is shown by means of percolation-theory ideas that the "stiffness" $D$ of the spin waves, equal to $\omega_q/q^2$, depends on the impurity concentration in accordance with the law $n_i^{-(1-\nu)/3} \exp(-\gamma/Rn_i^{1/3})$, where $\nu$ is the index of the correlation length $L$ in percolation theory[8,9] and $\gamma = 0.89$.[3] The spin excitations have the character of weakly-damped waves, if their wavelength $\lambda$ is large compared with the correlation length $L \approx r_c(r_c/R)^\nu \gg r_c$. This is connected with the fact that a hydrodynamic mode can exist only in the case when averaging of the exchange potential occurs over a wavelength. Since $R \ll r_c$, such averaging can occur only over distances larger than $r_c$ in the parameter $r_c/R$. Excitations with a characteristic length scale less than $L$ are localized owing to the strong fluctuations of the potential.

At high impurity concentrations, fluctuations of the magnetization are insignificant. This makes it possible to average the kernel in the equation for the Green function of the localized spins over the configurations of the impurities. In this case the stiffness $D$ depends on $c$ more weakly (nonexponentially) than at low impurity concentrations. The concentration dependence obtained for $D$ is in satisfactory agreement with the available experimental data.

## 1. LOW CONCENTRATIONS

We shall consider a system of randomly positioned spins, the exchange interaction between which is described by a Heisenberg Hamiltonian with the potential

$$V(r) = V_0 e^{-r/R}.$$

If the mean distance between impurities $r_c \sim n_i^{-1/3} \gg R$, then, according to[1-3], the Curie temperature of such a system is determined, to within a pre-exponential factor, as follows:

$$T_c \approx V_0 \exp\left(-\frac{\gamma}{Rn_i^{1/3}}\right), \quad \gamma = 0.89 \pm 0.03. \qquad (3)$$

For $T \ll T_c$ spin waves with the quadratic dispersion law

$$\omega_q = Dq^2. \tag{4}$$

exist in this system. We shall find the dependence of $D$ on the concentration of localized spins.

The equation of motion for the transverse component of the spin positioned at the point $\mathbf{r}_m$ has the form

$$dS_m^+/dt = i\sum_n V_{mn}(S_m^+ - S_n^+), \tag{5}$$

where $V_{mn} = V(\mathbf{r}_m - \mathbf{r}_n)$ is determined by formula (2), and $S_m^+ = S_m^x + iS_m^y$. Moreover, as shown in[10], the static conductivity $\sigma$ in a system of randomly positioned centers in which the probability of an electron hop between centers is determined by formula (2) is related to the eigenvalues of the operator

$$\sum_n V_{mn}(\Phi_m - \Phi_n). \tag{6}$$

Namely, it is easy to see that $\sigma \sim n_i c_0$, where $c_0$ is the coefficient of $q^2$ in the expansion of the eigenvalue of the operator (6) in powers of $q$, the coefficients of the lower powers of $q$ being equal to zero. Comparing (5) and (6), we remark that $c_0$ coincides with $D$; consequently,

$$D \sim \sigma / n_i. \tag{7}$$

The analogous formula for the problem with nearest-neighbor interaction in a lattice was first obtained by Kirkpatrick.[11]

The dependence of $\sigma$ on the concentration is well known[12,13]:

$$\sigma \sim \frac{1}{r_c^{1+\nu}} \exp\left(-\frac{\gamma}{Rn_i^{1/3}}\right), \tag{8}$$

where $\nu$ is the correlation-length index. According to[9,14], $\nu = 0.8 - 0.9$. Thus,

$$D \sim n_i^{-(2-\nu)/3} \exp(-\gamma/Rn_i^{1/3}). \tag{9}$$

It is known[3] that in a system of randomly positioned centers in which all the centers are coupled and the distance $r$ between them does not exceed a certain distance $r_1$, an infinite connected cluster arises if $r_1 \geqslant r_0 = \gamma n_i^{-1/3}$. In the spin system that we are considering, "bonds" with length $r_0 \pm R$ correspond to an energy $E \approx \exp(-\gamma/Rn_i^{1/3})$. It can be seen from (9) that it is precisely this energy which determines the stiffness of the system with respect to propagation of spin waves. This circumstance seems entirely obvious if we regard the propagation of the wave as the hopping of a spin excitation from center to center, and regard $V_{mn}$ in (5) as the hopping probability.

Weakly-damped spin waves exist if their wavelength is greater than the characteristic distance determining the length-scale of spatial fluctuations in an infinite cluster with $r_1 = r_0 + R$. According to the scaling theory of[8,9]: for $|(r_1 - r_0)/r_0| \ll 1$ the characteristic distance is the correlation length $L \sim (r_1 - r_0)^{-\nu}$. Thus, the spin-wave wavelength $\lambda \gtrsim L_R \sim r_c(r_c/R)^\nu$.

Excitations with length $\lambda < L_R$ are localized. The maximum spin-wave energy is

$$\omega_m \sim DL_R^{-2} \sim (R/r_c)^{3\nu} \exp(-\gamma/Rn_i^{1/3}).$$

It is easy to see that for $T \approx \omega_m$ the deviation of the magnetization from saturation, associated with the excitation of spin waves, is small, i.e., $\omega_m \ll T_c$.

We note that if the exchange interaction is described by a potential differing from (2) by a pre-exponential power-law factor, i.e., if

$$V(r) = V_0 f(r) e^{-r/R},$$

where $f(r)$ is a power function of $r$, an additional factor $f(r_c)$ will appear in (9). In particular, for the potential (1), which is realized in alloys of the PdFe type, we have

$$D \sim n_i^{-(1-\nu)/3} \exp(-\gamma/Rn_i^{1/3}). \tag{10}$$

Since $\nu$ differs little from unity, this formula predicts an exponential dependence of $D$ on $n_i^{1/3}$, without a pre-exponential factor.

## 2. HIGH CONCENTRATIONS

In the region of impurity concentrations $c \gtrsim 1 - I\rho(\varepsilon_F) \ll 1$ the spin splitting of the $d$-band of palladium becomes important. Therefore, it is necessary to take the electron subsystem into account explicitly. Here we assume, as before, that $c \ll 1$.

As usual, we write the Hamiltonian of the system of localized spins and $d$-electrons in the form

$$\mathcal{H} = \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} a_{\mathbf{k}}^+ a_{\mathbf{k}} + \sum_l I\hat{n}_{l\uparrow}\hat{n}_{l\downarrow} - 2J\sum_l (\mathbf{S}_l \boldsymbol{\sigma}_l)\varphi_l, \tag{11}$$

where $a_{\mathbf{k}}$ and $a_{\mathbf{k}}^+$ are respectively $d$-electron annihilation and creation operators, $\varepsilon_{\mathbf{k}}$ is the $d$-electron energy, $\hat{n}_l$ is the operator for the number of electrons at site $l$, and $\sigma_l$ is the electron-spin operator; $\varphi_l = 1$ at sites occupied by a magnetic impurity and $\varphi_l = 0$ at all other sites. The ensemble average $\overline{\varphi}_l = c$.

We represent the Hamiltonian (11) in the form of a sum of two terms:

$$\mathcal{H} = \mathcal{H}_e + \mathcal{H}_S.$$

The electron Hamiltonian, describing the strongly correlated electrons magnetized by the iron spins, has the form

$$\mathcal{H}_e = \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} a_{\mathbf{k}}^+ a_{\mathbf{k}} + I\sum_l \hat{n}_{l\uparrow}\hat{n}_{l\downarrow} - 2JS\sum_l \varphi_l \sigma_l^z. \tag{12}$$

The Hamiltonian of the localized spins is

$$\mathcal{H}_S = -J\sum_l (S_l^+\sigma_l^- + S_l^-\sigma_l^+)\varphi_l + 2J\sum_l S_l^z \varphi_l \langle\sigma_l^z\rangle. \tag{13}$$

In the last term in (13) the longitudinal component $\sigma_l^z$ of the $d$-electron spin operator has been replaced by the thermodynamic average $\langle\sigma_l^z\rangle$, since we are considering temperatures $T \ll T_c$.

We first consider the Hamiltonian (13). We introduce the correlators of the localized and electron spins:

$$\mathscr{D}^{+-}(l, l', \tau-\tau')=\langle T_\tau S_l^+(\tau) S_{l'}^-(\tau')\rangle,$$
$$\chi^{+-}(l, l', \tau-\tau')=\langle T_\tau \sigma_l^+(\tau)\sigma_{l'}^-(\tau')\rangle. \tag{14}$$

In the zeroth approximation we discard the first term in (13). The remaining Hamiltonian

$$\mathscr{H}_{0S}=2J\sum_l \varphi_l\langle\sigma_l^z\rangle S_l^z \tag{15}$$

describes a system of localized spins in an effective field created by the $d$-electrons. The corresponding Green function has the form[15]

$$\mathscr{D}_0(l, l')=2\delta_{ll'}\varphi_l S/(\omega_l-i\nu_m), \tag{16}$$

where

$$\omega_l=2J\langle\sigma_l^z\rangle=J(\langle\hat{n}_{l\uparrow}\rangle-\langle\hat{n}_{l\downarrow}\rangle), \tag{17}$$

and $\nu_m=2\pi mT$, where $m$ is an integer.

We now take into account the interaction of the transverse components of S and $\sigma$. It is not difficult to see that, in lowest order in the parameter $J/\varepsilon_F$, the equation for $\mathscr{D}(l, l')$ has the following form:

$$(\omega_l-i\nu_m)\mathscr{D}(l, l')=\delta_{ll'}\varphi_l+J^2\varphi_l\sum_m \chi^{+-}(l, m)\mathscr{D}(m, l'), \tag{18}$$

in which the electron susceptibility $\chi^{+-}(l, m)$ should be calculated from (14) using the Hamiltonian (12).

Up to now we have nowhere assumed that there is substantial spin splitting of the $d$-band of the matrix. Therefore, Eq. (18) should also be applicable in the region of low concentrations $c \ll 1 - I\rho(\varepsilon_F)$. In this case, in the calculation of $\chi^{+-}(l, m)$, in (12) we can discard the last term, which gives rise to spin splitting of the $d$-band. Then $\chi^{+-}(l, m)$ is the well-known susceptibility of an almost ferromagnetic electron gas without impurities, and at large $r$ we have $\chi^{+-}(r) \sim r^{-1}e^{-r/R}$. On the other hand, it follows from (17) and (12) that

$$\omega_l=J\sum_n \chi^{zz}(l, n)\varphi_n.$$

Since, for small splitting of the $d$-band, $\chi^{zz}(l, n)=\chi^{+-}(l, n)$, Eq. (18) reduces to the equation for the Green function of localized spins interacting with potential $V(r) \sim r^{-1}e^{-r/R}$, and this equation (to within the factor $r^{-1}$ in $V(r)$) is equivalent to the equation of motion (5).

For $c \gtrsim 1 - I\rho(\varepsilon_F)$, the molecular field that arises from the localized spins and acts on the $d$-electrons of the matrix is sufficiently large for spin splitting of the $d$-band to become important. In this case, as already noted, the polarization of the $d$-electrons becomes almost uniform. Bearing this situation in mind, we can average over the distribution of impurities in (12) and (18). Going over then to the Fourier transform of the Green function in (18), we obtain the following equation for the spectrum of the electrons:

$$\omega=\omega_0-2J^2Sc\chi^{+-}(q, \omega), \tag{19}$$

where

$$\chi^{+-}(q, \omega)=\frac{\chi_0(q, \omega)}{1-I\chi_0(q, \omega)}, \tag{20}$$

$$\chi_0(q, \omega)=\sum_{\mathbf{k}}\frac{f_{\mathbf{k}\uparrow}-f_{\mathbf{k}+\mathbf{q}\downarrow}}{\varepsilon_{\mathbf{k}+\mathbf{q}\downarrow}-\varepsilon_{\mathbf{k}\uparrow}+\omega}, \tag{21}$$

$$\omega_0=J(n_\uparrow-n_\downarrow), \tag{22}$$

$n_\uparrow$ and $n_\downarrow$ are respectively the concentrations of electrons with spin parallel and antiparallel to the magnetization.

It is easy to see that $\omega_0 = 2J^2Sc\chi^{+-}(0, 0)$, so that (19) can be rewritten in the form

$$\omega=2J^2Sc(\chi^{+-}(0, 0)-\chi^{+-}(q, \omega)).$$

At low frequencies $\omega \ll \Delta = I(n_\uparrow - n_\downarrow) + 2JSc$ and small $q \ll \varepsilon_F/v$, where $v$ is the velocity of the electrons at the Fermi surface, we obtain, analogously to[16],

$$D=\frac{1}{6(n_\uparrow-n_\downarrow+2cS)}\left\{\sum_{\mathbf{k}}(f_{\mathbf{k}\uparrow}-f_{\mathbf{k}\downarrow})\nabla^2\varepsilon_{\mathbf{k}}-\frac{2}{\Delta}\sum_{\mathbf{k}}(f_{\mathbf{k}\uparrow}-f_{\mathbf{k}\downarrow})|\nabla\varepsilon_{\mathbf{k}}|^2\right\}. \tag{23}$$

The detailed character of the dependence of $D$ on the impurity concentration is essentially determined by the structure of the Fermi surface of the $d$-electrons. This dependence can be established analytically when

$$n_\downarrow/n_e \ll JSc/\varepsilon_F \ll 1, \tag{24}$$

where $\varepsilon_F$ is the Fermi energy and $n_e \approx n_\uparrow$ is the electron concentration. In this case, in (23) we can discard the terms proportional to $n_\downarrow$ and expand in powers of $c$. The following expression is obtained:

$$D=A+Bc, \tag{25}$$

where

$$A=\frac{1}{6n_e}\left\{\sum_k f_{k\uparrow}\nabla^2\varepsilon_k-\frac{2}{In_e}\sum_k f_{k\uparrow}|\nabla\varepsilon_k|^2\right\} \tag{26}$$

$$B=\frac{1}{6n_e}\left\{\frac{4JS}{(In)^2}\sum_k f_{k\uparrow}|\nabla\varepsilon_k|^2-12SA\right\}. \tag{27}$$

It is clear that $A < 0$. Otherwise, a large moment would arise for $c = 0$. The quantity $B > 0$, and the expression (25) has meaning only for $c$ such that $D > 0$.

For a quadratic spectrum ($\varepsilon_{\mathbf{k}} = k^2/2m$), the quantity

$$A=\frac{1}{2m}\left(1-1.27\frac{\varepsilon_F}{In_e}\right),$$

and, near the threshold of the onset of ferromagnetism ($In_e/\varepsilon_F = \frac{4}{3}$), $|A| \ll B$; i. e., for not too small $c$, the stiffness $D \sim c$.

## 3. COMPARISON WITH THE RESULTS OF OTHER WORK

A theory of spin waves in alloys of the PdFe type was developed in the papers[5,6]. It was assumed that the spin splitting of the $d$-band of palladium is small. In this case, as already noted, the alloy is extremely nonuniform, and the nonuniformity determines all the features of its ferromagnetic properties. Nonetheless, in[5,6] averaging over the configuration of impurities was

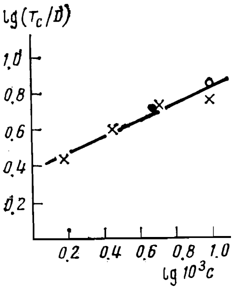

FIG. 1. Dependence of log $(T_c/D)$ on log$c$ for $Pd_{1-c}Fe_c$ alloys. The values of $D$ are taken from the papers[18] (+, ○) and[20] (●) and the values of $T_c$ from[21].

carried out at the very beginning. It turned out, therefore, that the principal contribution to $D$ was given by pairs of impurities with spacings $r\sim R\ll r_c$, even though such pairs obviously have no relation to the propagation of spin waves. As a consequence, the incorrect result $D\sim c$ was obtained.

In addition, the density of states of the spin waves was incorrectly calculated in[6]. It is obvious that the density of states of the spin waves, per unit volume, is

$$\rho_S(\omega)=\frac{1}{2\pi^2}k^2\frac{dk}{d\omega}=\frac{1}{4\pi^2}\frac{\sqrt{\omega}}{D^{3/2}}. \tag{28}$$

The expression obtained in[6] for the density of states contains an extra factor $c$. The source of the error made in[6] is best seen using the example, investigated in[17], of a system of randomly positioned spins in which the range of the exchange potential $R\gg r_c$. It was shown that to calculate the magnon spectrum it is sufficient to confine oneself to lowest order in $(r_c/R)^3$, while to calculate the density of states it is necessary to calculate higher approximations. As a result, the extra factor $c$ that arises in the Green function of the localized spins in first order in $(r_c/R)^3$ is cancelled.

An attempt to calculate $D$ for a strongly disordered system with $R\ll r_c$ was undertaken by us in[17]. The Green function of the localized spins was represented in the form of a series without a small parameter, and it postulated that the first term of the series, which gives $D\sim\exp(-\gamma_1/(R^3n)^{1/2})$, $\gamma_1=\text{const}$, correctly describes the concentration dependence of $D$. As can be seen from Sec. 1, this assumption was not justified.

The coefficient $D$ for PdFe, PdCo, and also PdMn alloys has been determined experimentally from measurements of the magnetization,[18] specific heat,[18,19] NMR[20] and electrical resistivity.[7] In Fig. 1, the experimental dependence of $T_c/D$ on the impurity concentration for low concentrations $c\leq 1\%$ is plotted on a double logarithmic scale. The values of $T_c$ are taken from[21] and the values of $D$ from[18–20].[1)] It can be seen that the experimental points are well fitted by a straight line; i.e., in agreement with the theory, $D$ and $T_c$ contain the same exponential factor, with $T_c/D\sim c^{0.45}$. The exponent here is close to the value 0.4 that follows from (3) and (9) with $\nu=0.8$, if we assume that $T_c$ for the potential (2) does not contain a concentration-dependent pre-exponential factor. The absence of such a factor seems reasonable from the following considerations.

The quantity $T_c$ can be estimated by means of the formula for the deviation of the magnetization from saturation:

$$\Delta M\sim\int\frac{\rho_S(\omega)}{e^{\omega/T}-1}d\omega,$$

by equating this deviation to the magnitude of the saturation magnetization.

As was shown in Sec. 1, for $\omega\ll\omega_m$ the function $\rho_S(\omega)$ is determined by the spin waves, i.e., $\rho_S(\omega)\sim\sqrt{\omega}$. On the other hand, at energies $\omega\gg\exp(-\gamma/Rn_i^{1/3})$ the density of states decreases with increase of $\omega$.[1] Since excitations with energies $\omega>\omega_m$ are localized over length scales less than $L$, it is natural to assume that $\rho_S(\omega)$ reaches a maximum at $\omega=\omega_m$. In this case we obtain for $T_c$ the expression (3).

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[1)] To obtain $D$ from the resistivity data the authors of the paper[7] had to make a number of poorly justified assumptions about the dependence of $J$ on the impurity concentration. Therefore, we do not use these data.

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