Free magnetic polaron: A nonlinear Hamiltonian approach

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(Received 29 June 1994; revised manuscript received 10 January 1995)

We show that in the mean-field approximation the free energy of the free magnetic polaron can be formally written as the average value of a nonlinear Hamiltonian. As a nontrivial consequence, the *exact* wave function which minimizes this free energy is the eigenfunction of another nonlinear Hamiltonian, the potential of which is just the carrier-ion exchange interaction. Solutions for various space dimensions are given. They depend upon a unique dimensionless parameter and lead to an easy discussion of the free magnetic polaron stability in one, two, and three dimensions. Conditions for the magnetic fluctuations to play a significant role are also given.

I. INTRODUCTION

The problem of free and bound magnetic polarons in diluted magnetic semiconductors has been an attractive topic for theoretical and experimental investigations during the last decade. A coupled carrier-ion state can exist due to the exchange interaction between a carrier and the magnetic ions randomly distributed in the crystal. The theory of a bound magnetic polaron, where the carrier is already bound by a nonmagnetic potential, is now in good agreement with experiments,[1] while the theory of free magnetic polarons (FMP's) (or the magnetically self-trapped state) still presents difficulties.[2] In the mean-field approximation, the carrier-ion exchange energy depends upon the carrier spatial wave function only. The usual variational approach is to look for the free-energy minimum through a one-parameter trial function.[3] Thus the results obtained depend upon the shape of the trial function.

In this paper, we show, by formally writing the free energy as the average value of a nonlinear Hamiltonian, that the *exact* free-energy minimum is obtained for a wave function eigenstate of another nonlinear Hamiltonian.[4] The potential of this Schrödinger equation is found to be just the carrier-ion exchange interaction. The corresponding eigenstates are shown to depend upon a unique coupling parameter. This feature allows a very simple study of the FMP stability, through a combination of analytical and numerical calculations. By extending our theory to quasi-two-dimensional (2D) and quasi-1D systems (Sec. II), we demonstrate the strong dependence of this stability upon space dimension (Sec. III). In Sec. IV, we discuss the role of magnetic fluctuations on the FMP stability near threshold, with a special emphasis on the quasi-2D case.

II. THEORETICAL BACKGROUND

A. General properties of a nonlinear Hamiltonian

We start with general statements on nonlinear Hamiltonians. Such statements will be used in the FMP problem.

Let us consider a nonlinear Hamiltonian

$$\mathbf{H}=K+V(u)\ , \tag{1}$$

in which K is the kinetic energy and V a potential which depends on $\mathbf{r}$ through the probability $u(\mathbf{r})=\psi(\mathbf{r})\psi^*(\mathbf{r})$ of the particle to be located at $\mathbf{r}$, $\psi(\mathbf{r})$ being the particle normalized wave function (let us remark that the nonlinear potential V is a *local* one). The ψ's which minimize

$$F=\langle\psi|K+V(u)|\psi\rangle \tag{2}$$

are such that, for any $\delta\psi$,

$$\langle\delta\psi|H|\psi\rangle+\langle\psi|H|\delta\psi\rangle+\langle\psi|\delta V|\psi\rangle = E(\langle\delta\psi|\psi\rangle+\langle\psi|\delta\psi\rangle)\ , \tag{3}$$

the Lagrange multiplier E arising from the $\langle\psi|\psi\rangle=1$ condition. By noting that

$$\langle\psi|\delta V(u)|\psi\rangle=\langle\delta\psi|u\frac{dV(u)}{du}|\psi\rangle+\text{c.c.}\ , \tag{4}$$

we can rewrite Eq. (3) as

$$\langle\delta\psi|K+V(u)+u\frac{dV(u)}{du}-E|\psi\rangle+\text{c.c.}=0\ . \tag{5}$$

The above equation shows that the ψ's which minimize F are eigenstates of another nonlinear Hamiltonian

$$\mathbf{H}_1=K+W(u)\ , \tag{6}$$

the potential of which is related to V through

$$W(u)=\frac{d[uV(u)]}{du}\ . \tag{7}$$

For such nonlinear Hamiltonians, the $\mathbf{H}$ average value extrema and the $\mathbf{H}_1$ eigenvalues are related by

$$F=E-\langle\psi|W(u)-V(u)|\psi\rangle \tag{8}$$

(for ψ independent potentials, we do have $\mathbf{H}_1=\mathbf{H}$ and $F=E$, as expected).

B. Application to free magnetic polaron

We now turn to our problem: a carrier, electron, or hole, coupled to magnetic ions through exchange interaction. In the mean-field approximation (see Appendix A) the carrier action on a magnetic ion located at $\mathbf{r}$ is equivalent to a magnetic field H along the carrier spin direction, given by

$$g\mu_B H=\alpha u(\mathbf{r})/2 , \tag{9}$$

g being the gyromagnetic factor, μ_B the Bohr magneton, α the carrier-ion exchange integral, and $u(\mathbf{r})$ the probability of the carrier to be located at $\mathbf{r}$. In the presence of magnetic ions, the carrier free energy is given by

$$F=\langle\psi|K|\psi\rangle+F_s , \tag{10}$$

where F_s is the magnetic contribution to the free energy. Its density f_s is related to the semiconductor magnetization $M(H)$ through

$$M(H)=-\delta f_s/\delta H . \tag{11}$$

By formally setting

$$f_s\equiv uV(u) , \tag{12}$$

we can write F_s as

$$F_S=\int d\mathbf{r}\, f_s=\langle\psi|V(u)|\psi\rangle , \tag{13}$$

so that, using Sec. II A, the ψ's which minimize F are eigenstates of the Hamiltonian $\mathbf{H}_1=K+W(u)$, in which the potential $W(u)$ is related to the magnetization via

$$W(u)=\frac{d}{du}[uV(u)]=\frac{df_s}{dH}\frac{dH}{du}=-\frac{\alpha}{2g\mu_B}M\left[\frac{\alpha u}{2g\mu_B}\right] . \tag{14}$$

It is useful to stress that the magnetization $M(H)$ is simply related to W, and not to the V potential appearing in the free energy we want to minimize. V depends on W through Eq. (7). Before going further, it is also interesting to mention that the $\mathbf{H}_1$ eigenvalues have a direct physical meaning since the Zeeman energy just reads

$$E_z=-\int d\mathbf{r}\, HM(H)=\langle\psi|W(u)|\psi\rangle . \tag{15}$$

By expressing the magnetization as

$$M(H)=M_s w(\chi H/M_s) , \tag{16}$$

where M_s is the saturated magnetization and χ the magnetic susceptibility, we define a function $w(x)$ which tends to x for $x\to 0$ and to 1 for $x\to\infty$. Using Eq. (16), we get

$$W(u)=-U_s w(\lambda_3 d_s^3 u) , \tag{17}$$

where $U_s=\alpha M_s/2g\mu_B$ is the saturated exchange energy, $d_s=(\hbar^2/2mU_s)^{1/2}$ a length unit ($m=m^*m_0$ is the carrier effective mass), and $\lambda_3=\alpha\chi d_s^{-3}/2g\mu_B M_s$ a dimensionless coupling parameter. From this W, we deduce

$$V(u)=-U_s v(\lambda_3 d_s^3 u) , \tag{18}$$

where the functions v and w are related by Eq. (7), i.e.,

$$\begin{aligned} v(x)&=x^{-1}\int_0^x w(x')dx' \\ &=w(x)-x^{-1}\int_0^x[w(x)-w(x')]dx' . \end{aligned} \tag{19}$$

The above equation shows that $v(x)$ tends to $x/2$ in the small-x limit, and to $w(x)-x^{-1}A_\infty$ for large x, A_∞ being a constant defined as

$$A_\infty=\int_0^\infty\{1-w(x)\}dx . \tag{20}$$

Since $w(x)$ is an increasing function of x, from Eq. (19) we always have $v(x)<w(x)$. Equation (8) then gives $E<F$; as $F<0$ for a stable FMP (the free energy of a delocalized carrier being zero), we conclude that the reduced free energy $\varphi=-F/U_s$ and the reduced energy $\varepsilon=-E/U_s$ of a stable FMP verify

$$0<\varphi<\varepsilon . \tag{21}$$

We can note, moreover, that, as $w(x)$ varies from 0 to 1, $W>-U_s$. The kinetic energy $\langle\psi|K|\psi\rangle$ always being positive, we thus have $E>-U_s$, i.e., $\varepsilon<1$. Consequently, the $\mathbf{H}_1$ eigenstates which can lead to stable FMP's are such that $0<\varepsilon<1$.

When the magnetization can be described by a modified Brillouin function for 5/2 spins, characterized by the two parameters S_0 and T_0, it was shown in Ref. 5 that the dimensionless coupling constant reads

$$\lambda_3=\frac{7}{24\sqrt{2}}\frac{(N_0\alpha)^{5/2}}{E_0^{3/2}k_B(T+T_0)}(m^*xS_0)^{3/2} , \tag{22}$$

where N_0 is the density of cation sites, x the fraction occupied by magnetic ions, and $E_0=\hbar^2N_0^{2/3}/2m_0$; m_0 is the free-electron mass and m_0m^* the carrier effective mass.

C. Extension to quasi-2D and quasi-1D systems

In the case of confined geometries, $\mathbf{H}$ or $\mathbf{H}_1$ contains, in addition, the localization potential in the z or (x,y) directions. In the presence of a magnetic potential, the motions along (x,y,z) are coupled. For strongly confined systems, however, we can neglect this coupling and set $\psi\simeq\tilde{\psi}\varphi$, where $\varphi(z)$ or $\varphi(x,y)$ is the localized wave function of the well or the wire in the absence of magnetic interaction. This corresponds to assuming that the magnetic potential modifies the carrier motion along the free directions (x,y) or z only. The corresponding part of the wave function $\tilde{\psi}(x,y)$ or $\tilde{\psi}(z)$ is now the solution of a 2D or 1D nonlinear Schrödinger equation in which the effective magnetic potential $\tilde{W}(\tilde{u})$, with $\tilde{u}=\tilde{\psi}\tilde{\psi}^*$, has to be an appropriate average[5] of $W(\varphi\varphi^*\tilde{\psi}\tilde{\psi}^*)$ over the z or (x,y) directions. In order to have the same energy, we must have $\langle\psi|W|\psi\rangle=\langle\tilde{\psi}|\tilde{W}|\tilde{\psi}\rangle$, so that this average must be the integral of $\varphi\varphi^*W(\varphi\varphi^*\tilde{\psi}\tilde{\psi}^*)$ over the well or wire thickness. Using Eq. (17), we can thus write $\tilde{W}$ as

$$\tilde{W}(\tilde{u})=-U_s\tilde{w}(\lambda_\delta d_s^\delta\tilde{u}) , \tag{23}$$

where δ is the space dimension (2 for a quantum well and

1 for a quantum wire), and $\tilde{w}(x)$ an increasing function of x which varies as x for small x and tends to 1 for large x. The dimensionless parameter λ_δ can be obtained in terms of λ_3 by relating $\tilde{w}(x)$ to $w(x)$ in the small-x limit. We then find

$$\lambda_\delta d_s^\delta \tilde{\psi}\tilde{\psi}^* = \lambda_3 d_s^3 \int \varphi\varphi^*(\varphi\varphi^*\tilde{\psi}\tilde{\psi}^*) , \tag{24}$$

so that $\lambda_\delta d_s^{\delta-3}/\lambda_3$ is the integral of $(\varphi\varphi^*)^2$ over the well or wire variables.

By defining, for the same reason, the effective potential $\tilde{V}(\tilde{u})$ which enters the free energy as the integral of $\varphi\varphi^* V(\varphi\varphi^*\tilde{\psi}\tilde{\psi}^*)$ over the well or wire, it is easy to check that these two potentials are still related by $d(x\tilde{V}(x))/dx = \tilde{W}(x)$, so that $\tilde{V}$ can also be formally written as

$$\tilde{V}(\tilde{u}) = -U_s \tilde{v}(\lambda_\delta d_s^\delta \tilde{u}) , \tag{25}$$

where $\tilde{v}(x)$ tends to $x/2$ for small x and to 1 for large x. Using this similarity, we conclude that the 3D, quasi-2D, and quasi-1D cases are formally the same, provided that $\mathbf{r}$ is seen as a vector in $\delta=3$, 2, or 1 dimensions. In the following, we will simply call λ the dimensionless coupling parameter λ_δ and drop the tilda over the 2D or 1D quantities. As shown below, the FMP stability is uniquely controlled by this λ parameter.

III. DIMENSIONALITY EFFECTS

A. Reduced equation

We are now looking for the eigenstates of $\mathbf{H}_1$, i.e., the solutions of

$$[K - U_s w(\lambda d_s^\delta \psi\psi^*)]\psi(\mathbf{r}) = -U_s \varepsilon \psi(\mathbf{r}) . \tag{26}$$

We have seen that the ψ's which could lead to stable FMP's correspond to $0<\varepsilon<1$ only. By setting

$$\boldsymbol{\rho} = \varepsilon^{1/2}\mathbf{r}/d_s , \tag{27}$$

we have

$$K = -\frac{\hbar^2}{2m}\nabla_r^2 = -U_s\varepsilon\nabla_\rho^2 , \tag{28}$$

so that the $\mathbf{H}_1$ eigenstates $\psi(\mathbf{r})$ with negative eigenvalues can be written as

$$\psi(\mathbf{r}) = (\varepsilon d_s^{-\delta}/\lambda)^{1/2}\phi_\varepsilon(\boldsymbol{\rho}) , \tag{29}$$

where the ϕ_ε solution of

$$\nabla^2\phi_\varepsilon = [1-\varepsilon^{-1}w(\varepsilon\phi_\varepsilon\phi_\varepsilon^*)]\phi_\varepsilon \tag{30}$$

depends on ε only. It is useful to note that the condition $\langle\psi|\psi\rangle = 1$ does not imply $\langle\phi_\varepsilon|\phi_\varepsilon\rangle = 1$, but instead

$$\lambda = \varepsilon^{1-\delta/2}\langle\phi_\varepsilon|\phi_\varepsilon\rangle , \tag{31}$$

which is in fact the relation between the reduced energy ε and the coupling parameter λ.

In Appendix B, we derive the two following useful expressions for the reduced energy ε and the reduced free energy φ in the case of $1S$ states (which are expected to give the lowest free energy):

$$\varepsilon = \langle w\rangle + (\langle v\rangle - \langle w\rangle)\delta/2 , \tag{32}$$

$$\varphi = \langle v\rangle + (\langle v\rangle - \langle w\rangle)\delta/2 , \tag{33}$$

where $\langle f\rangle$ stands for the average value

$$\langle f\rangle = \frac{\langle\phi_\varepsilon|f(\varepsilon\phi_\varepsilon\phi_\varepsilon^*)|\phi_\varepsilon\rangle}{\langle\phi_\varepsilon|\phi_\varepsilon\rangle} . \tag{34}$$

B. Unsaturated limit

If ε is small enough for $\varepsilon\phi_\varepsilon\phi_\varepsilon^*$ to stay small, we can replace $w(x)$ and $v(x)$ by their linear terms x and $x/2$. Then $\langle w\rangle = 2\langle v\rangle$, and from Eqs. (32) and (33) we find that ε and φ are simply proportional:

$$\varphi = \varepsilon(2-\delta)/(4-\delta) . \tag{35}$$

In this limit, the λ dependence of ε reduces to a power law since, for small ε, Eq. (31) tends to

$$\lambda = \varepsilon^{1-\delta/2}\langle\phi_0|\phi_0\rangle , \tag{36}$$

where the ϕ_0 solution of

$$\nabla^2\phi_0 = \phi_0 - \phi_0^2\phi_0^* \tag{37}$$

does not depend on ε.

(i) For 1D, Eq. (37) can be solved analytically. By imposing $\langle\phi_0|\phi_0\rangle$ to be finite, we find only one solution (as is well known in soliton theory)

$$\phi_0(\mathbf{r}) = \sqrt{2}/ch(\mathbf{r}) . \tag{38}$$

For this ϕ_0, we have $\langle\phi_0|\phi_0\rangle = 4$, so that, from Eqs. (35) and (36),

$$\varepsilon = \lambda^2/16 = 3\varphi . \tag{39}$$

As this solution corresponds to a negative $F = -U_s\varphi$, we conclude that the FMP is stable in 1D for small coupling parameters, i.e., in the linear regime of $M(H)$.

(ii) for 3D, the numerical resolution of Eq. (37) gives a discrete set of finite $\langle\phi_0|\phi_0\rangle$'s, the lowest one being 18.7. However, as for $\delta=3$, Eqs. (35) and (36) give

$$\varepsilon = \langle\phi_0|\phi_0\rangle^2/\lambda^2 = -\varphi , \tag{40}$$

they all generate negative φ, i.e., positive F extrema. These extrema are in fact maxima; they correspond to potential barriers between fully delocalized states for which $F=0$ and small polarons. We thus conclude that no stable FMP exists in 3D within a linear $M(H)$.

(iii) For 2D, Eq. (37) also gives a discrete set of finite $\langle\phi_0|\phi_0\rangle$'s, the lowest one being 11.71. However for $\delta=2$, Eq. (35) gives $\varphi=0$ while Eq. (36) gives

$$\lambda = \langle\phi_0|\phi_0\rangle = \lambda_c \tag{41}$$

for any ε. This indeterminate situation disappears if we go beyond the $M(H)$ linear term. By inserting higher-order terms, i.e., by setting

$$w(x) \simeq x + a_n x^n , \tag{42}$$

we generate λ dependence of ε and φ close to λ_c: as the v potential associated to this w is

$$v(x)=x/2+a_n x^n/(n-1)\ , \tag{43}$$

we have

$$2v(x)-w(x)=a_n x^n(n-1)/(n+1)\ , \tag{44}$$

so that Eq. (33) for $\delta=2$ gives

$$\varphi=2\langle v\rangle-\langle w\rangle=-a_n\frac{n-1}{n+1}\frac{\langle\phi_\varepsilon|\phi_\varepsilon^{2n}|\phi_\varepsilon\rangle}{\langle\phi_\varepsilon|\phi_\varepsilon\rangle}\varepsilon^n\ , \tag{45}$$

in which, to lowest order in ε, we can replace ϕ_ε by ϕ_0. More precisely, for such $w(x)$, Eq. (30) reads

$$\nabla^2\phi_\varepsilon=[1-\varepsilon^{-1}\{\varepsilon\phi_\varepsilon\phi_\varepsilon^*+a_n(\varepsilon\phi_\varepsilon\phi_\varepsilon^*)^n\}]\phi_\varepsilon\ , \tag{46}$$

so that

$$\phi_\varepsilon\approx\phi_0+a_n\varepsilon^{n-1}G\ , \tag{47}$$

where ϕ_0 is solution of Eq. (37), and G verifies

$$\nabla^2G=G(1-3\phi_0\phi_0^*)-(\phi_0\phi_0^*)^n\phi_0\ . \tag{48}$$

We then have, to lowest order in ε,

$$\langle\phi_\varepsilon|\phi_\varepsilon\rangle=\langle\phi_0|\phi_0\rangle+2a_n\langle\phi_0|G\rangle\varepsilon^{n-1}\ , \tag{49}$$

so that, from Eq. (36), we get the ε dependence of λ:

$$\lambda\approx\lambda_c+2a_n\langle\phi_0|G\rangle\varepsilon^{n-1}\ . \tag{50}$$

As $w(x)\propto M(H)$, we usually have $n=3$ and $a_n<0$, while numerical calculations show that $\langle\phi_0|G\rangle$ is negative. Consequently, λ increases with ε as ε^2, while φ, given by Eq. (45), is positive and increases as ε^3. We thus conclude that a stable FMP exists in 2D for $\lambda>\lambda_c$. Close to this threshold, ε varies as $(\lambda-\lambda_c)^{1/2}$, while φ varies as $(\lambda-\lambda_c)^{3/2}$. We wish to stress that, in order to find this λ dependence, it is necessary to go beyond the linear regime, i.e., to include the $M(H)$ deviation from linearity.

C. Saturated limit

We have seen that the unsaturated limit corresponds to small w and v, i.e., $\varepsilon\gtrsim 0$. Conversely, the saturated limit corresponds to $w\simeq v\simeq 1$ in most of the FMP volume. Due to Eqs. (32) and (33), φ and ε are close to 1. More precisely, as $v<w$, ε is slightly less than 1 [while $\varphi<\varepsilon$ as usual due to Eq. (21)]. Let us note that w close to 1 imposes large $\varepsilon\phi_\varepsilon\phi_\varepsilon^*$. As $\varepsilon\lesssim 1$, this implies that $\phi_\varepsilon\phi_\varepsilon^*$ is large in the saturated region. The space extension L of this saturated region can be obtained from Eq. (30), which in the $w\simeq 1$ limit reads

$$\nabla^2\phi_\varepsilon=\phi_\varepsilon(1-\varepsilon^{-1})\ . \tag{51}$$

Its solution decreases exponentially over a length L which diverges when ε approaches 1 as

$$L\approx(1-\varepsilon)^{-1/2}\ . \tag{52}$$

This L deliminates the region of large ϕ_ε's. For such ϕ_ε, Eqs. (19) and (20) give

$$[\langle v\rangle-\langle w\rangle]\langle\phi_\varepsilon|\phi_\varepsilon\rangle\approx\langle\phi_\varepsilon|-A_\infty(\varepsilon\phi_\varepsilon\phi_\varepsilon^*)^{-1}|\phi_\varepsilon\rangle$$
$$\approx-A_\infty\int_0^L d^\delta\rho\sim L^\delta\ . \tag{53}$$

We then deduce from Eq. (32) that

$$(\varepsilon-1)\langle\phi_\varepsilon|\phi_\varepsilon\rangle=\langle\phi_\varepsilon|w(\varepsilon\phi_\varepsilon\phi_\varepsilon^*)-1|\phi_\varepsilon\rangle$$
$$-A_\infty\frac{\delta}{2}\langle\phi_\varepsilon|(\varepsilon\phi_\varepsilon\phi_\varepsilon^*)^{-1}|\phi_\varepsilon\rangle\ . \tag{54}$$

As $w(x)$ tends to 1 exponentially while Eq. (31) gives $\lambda\sim\langle\phi_\varepsilon|\phi_\varepsilon\rangle$ for $\varepsilon\simeq 1$, the above equation reduces to

$$\lambda(1-\varepsilon)\propto L^\delta,\ \text{i.e.,}\ \ \lambda\propto(1-\varepsilon)^{-(1+\delta/2)}\ . \tag{55}$$

We thus conclude that the saturated limit is reached for large λ, and that in this limit the reduced energy ε and free energy φ both approach 1 as $\lambda^{-2/(2+\delta)}$.

D. General case

Figure 1 shows the λ dependences of ε and φ in 1D, 2D, and 3D as numerically calculated using Eqs. (30) and (31). We recover the unsaturated limit $\varepsilon\to 0$ and the saturated limit $\varepsilon\to 1$ obtained analytically in the previous sections.

(i) In 1D, we see that ε and φ increase monotonically from 0 to 1 with increasing λ: a positive φ, i.e., a stable FMP with negative F, exists for any value of the coupling parameter. Close to $\lambda=0$, we have shown that $\varepsilon\approx\lambda^2/16$ and $\varphi\approx\varepsilon/3$.

(ii) In 2D, we find that the coupling parameter must be larger than $\lambda_c=11.71$ for the FMP to be stable. When λ increases above λ_c, φ as well as ε increases from 0 to 1. Close to threshold, we have shown that they behave like $\varphi\sim(\lambda-\lambda_c)^{3/2}$ and $\varepsilon\sim(\lambda-\lambda_c)^{1/2}$.

(iii) The 3D case is more interesting. We have seen that both the $\varepsilon\to 0$ and $\varepsilon\to 1$ limits correspond to large λ. As a consequence, the $\varepsilon(\lambda)$ curve must have an infinite slope for some particular value λ^* of the coupling parameter. It is possible to show (see Appendix C) that this λ^* corresponds in fact to the lowest φ, and that, for this λ^* value, $\varphi(\lambda)$ exhibit a cusp with a positive slope.

As an example, let us consider the case of a dilute paramagnetic semiconductor with spins S. Its magnetization is described by a modified Brillouin function, so that the deduced $w(x)$ and $v(x)$ reads, using $y=3x/[2(S+1)]$,

$$w(x)=\frac{2S+1}{2S}\coth(2S+1)y-\frac{1}{2S}\coth y\ ,$$
$$v(x)=\frac{1}{2Sy}\ln\frac{\sinh(2S+1)y}{(2S+1)\sinh y}\ . \tag{56}$$

In this case, for $S=\frac{5}{2}$, numerical calculations give $\lambda^*\approx 97$. For $\lambda>\lambda^*$, the FMP free energy has two branches.

(a) The upper branch corresponds to a negative $F=-U_s\varphi$, i.e., a stable state if λ is larger than a value λ^{**} for which $\varphi=F=0$ (numerical calculation give $\lambda^{**}\approx 124$). Between λ^* and λ^{**}, F is positive, and the corresponding FMP is metastable. As the $F(\lambda)$ slope is finite for $F=0$, the transition to a stable FMP looks like a first-order phase transition.

(b) The lower branch is always unstable since it corresponds to a negative φ, i.e., a positive F. The FMP states

of this lower branch constitute in fact a potential barrier between the delocalized carrier state (having a zero free energy) and the stable (or metastable) FMP of the upper branch.

We can illustrate these regimes of stability in the following way. In the numerical process to obtain the FMP solutions for any ε, we get a set of eigenfunctions ψ_ε. We use that set as trial functions to minimize the FMP free energy for a value μ of the coupling constant. For that purpose, we introduce the quantity $F(\varepsilon,\mu)$ defined as (see Appendix C)

$$F(\varepsilon,\mu)=-U_s\zeta(\varepsilon,\mu)=\langle\psi_\varepsilon|K-U_s v(\mu d_s^3\psi_\varepsilon\psi_\varepsilon^*)|\psi_\varepsilon\rangle\ . \tag{57}$$

If we plot $F(\varepsilon,\mu)$ as a function of $E=-U_s\varepsilon$ (see the dotted lines in Fig. 2), the extrema of this graph occur for ε values such that $\lambda(\varepsilon)=\mu$, where $\lambda(\varepsilon)$ is the value of the coupling parameter associates with ε, as given by Eq. (31). These extrema are located on the $F(E)$ graph (the full line in Fig. 2), since the free energy of the FMP of energy E is $F=F(\varepsilon,\lambda(\varepsilon))$.

Moreover, we can illustrate the three regimes of stability, depending on the position of the $F(\varepsilon,\mu)$ extrema on the $F(E)$ graph.

(a) If the extremum is located on the OA part of the $F(E)$ curve, we see that this extremum is a maximum, thus corresponding to unstable FMP states.

(b) Conversely, we see that these extrema are minima when located on the AC part of the $F(E)$ curve; they correspond either to metastable FMP states when located on the AB part since $F>0$, or to stable FMP states when located on the BC part since $F<0$.

Such a behavior for 3D FMP's was already predicted by Ryabchenko and Semenov,[6] using Gaussian trial functions to minimize the free energy. The results presented here are in fact exact in the framework of the mean-field approximation. Some of our results were also obtained by Mauger and Mills.[7] In their study of FMP's in antiferromagnetic material (called ferrons), they reached conclusions similar to the ones we obtained in the unsaturated limit (Sec. III B) for 1D and 3D, in the case of their type-II states.

It is straightforward to extend the present calculations for FMP's to dilute magnetic semiconductors with Van Vleck ions.[8] In this case we have just to deduce $w(x)$ from a theoretical expression of the magnetization of a

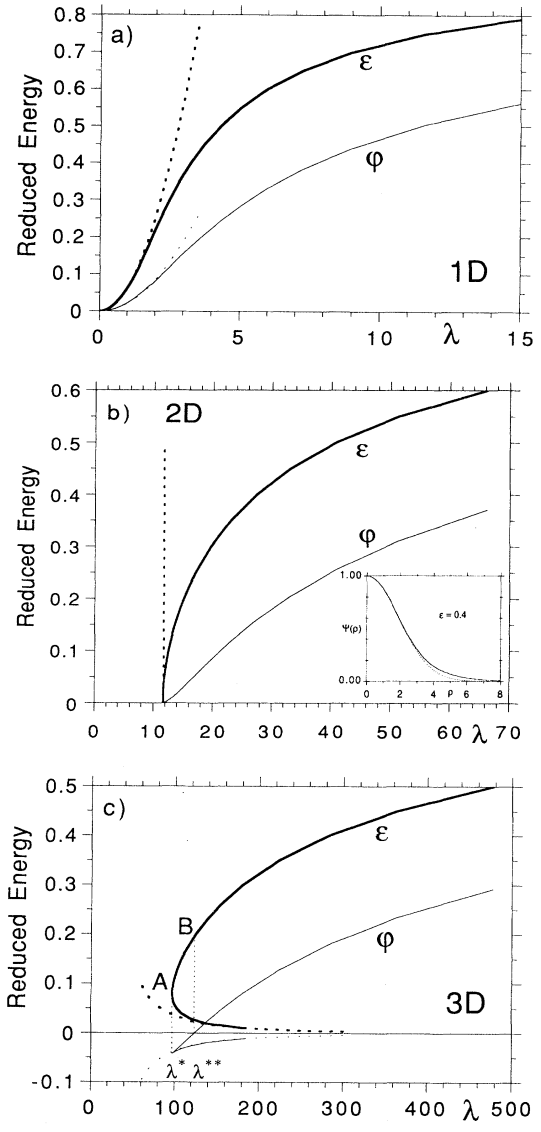

FIG. 1. Reduced energy ε and free energy φ of FMP as a function of the coupling parameter λ; the dotted curves correspond to the unsaturated limit. (a) 1D case, (b) 2D case; the inset shows the carrier wave function $\psi(\rho)$ for $\varepsilon=0.4$, with ρ in reduced units, the dotted curve being a Gaussian given for comparison. (c) 3D case; point B marks the limit between stable and metastable FMP states. Note the different λ scales for the three cases.

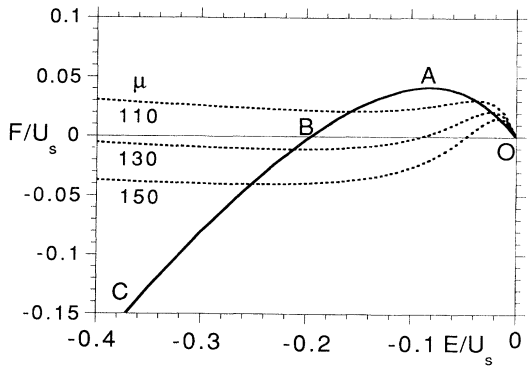

FIG. 2. In the 3D case, the full curve giving FMP free energy F/U_s as a function of its energy E/U_s shows different regimes: stability along CB, metastability along BA, and potential barrier along AO. The dotted curves give $F(\varepsilon,\mu)/U_s$ for three values of μ (110, 130, and 150). The extrema of F_μ are on the $F(E)$ curve; the OA part corresponds to maxima, while the AC part corresponds to minima.

Van Vleck paramagnet [or better, to use the experimental data for $M(H)$].

E. Practical application to diluted magnetic semiconductors

From Eq. (22), we can evaluate the coupling constant λ_3 for diluted magnetic semiconductors. The energy E_0, which depends only on N_0, is found equal to 0.23 eV. The product xS_0 has a maximum value of about 0.1; inserting these values into Eq. (22), we obtain

$$\lambda_3 \approx 600(N_0\alpha)^{5/2} m^{*3/2}/T^* , \tag{58}$$

with $N_0\alpha$ in eV, $T^* = T + T_0$ in K, and the effective mass m^* in m_0 unit. For the hole in $Cd_{0.9}Mn_{0.1}Te$, we get $\lambda_3 \sim 12$ at 2 K. In other diluted magnetic semiconductors, assuming larger values for $N_0\alpha$ (1.5 eV) with some correlated increase of T_0 and larger m^* (0.5), λ_3 could reach values close to 100, still smaller than $\lambda^{**} = 124$, the threshold for FMP stability in 3D. So we can conclude that FMP's are not stable in bulk diluted magnetic semiconductors.

Turning now to lower dimensions, let us recall that in Sec. II C, we assumed perfect carrier confinement. In 1D, FMP stability is always achieved. In 2D, from Eq. (24), we get the coupling parameter

$$\lambda_2 = 1.5\frac{d_s}{l_z}\lambda_3 , \tag{59}$$

where l_z is the well width, the length unit d_s being in the nm range. From the values of λ_3 discussed above, it is easy to achieve $\lambda_2 > \lambda_c = 11.7$ provided that the well width l_z is in the nm range. FMP stability could be achieved in *magnetic* quantum wells with strong carrier confinement.

IV. CONTRIBUTION TO FMP ENERGY FROM TRANSVERSE SPIN FLUCTUATIONS

A. General formulation

The effective field H introduced in Sec. II B corresponds only to the z component of the carrier-ion exchange interaction operator. This operator is indeed correct for Ising-type interactions; such interactions are found for holes in the ground-state band of hexagonal crystals or in the ground subband of 2D or 1D structures based on crystals with cubic lattices and degenerated valence-band edges. In the case of Heisenberg-type interactions, we must include, in addition, the contribution of the transverse spin fluctuations. It has been shown[9] that, far from saturation, these fluctuations induce an additional term in the magnetic free energy,[10] which now reads

$$\hat{F}_s = -\langle \psi | U_s \lambda d_s^{\delta} u/2 | \psi \rangle - k_B T \ln[1 + 2\langle \psi | U_s \lambda d_s^{\delta} u/2 | \psi \rangle / k_B T] , \tag{60}$$

where T is the temperature and $u = \psi\psi^*$ as before. We note that $\langle \psi | U_s \lambda d_s^{\delta} u | \psi \rangle$ is just the ε_p polaron energy shift of Ref. 9 and that $-U_s \lambda d_s^{\delta} u/2$ is the limit of $V(u)$ for small u. We propose to extend the validity of Eq. (60) beyond the linear regime of magnetization, by now writing $\hat{F}_s$ as

$$\hat{F}_s = \langle \psi | V(u) | \psi \rangle - k_B T \ln[1 - 2\langle \psi | V(u) | \psi \rangle / k_B T] . \tag{61}$$

This extension allows us to recover saturation in the small-T limit. $\hat{F}_s$ appears as a complicated function of ψ. However, we can note that, being a simple function of $\langle \psi | V(u) | \psi \rangle$, its variation reads

$$\partial \hat{F}_s = f \partial[\langle \psi | V(u) | \psi \rangle] , \tag{62}$$

where f is given by

$$f = 1 + \frac{2}{1 - 2\langle \psi | V(u) | \psi \rangle / k_B T} . \tag{63}$$

Then, following Sec. II A, the wave function which minimizes

$$\hat{F} = \langle \psi | K | \psi \rangle + \hat{F}_s \tag{64}$$

is still an eigenstate of a nonlinear Hamiltonian, this Hamiltonian now being

$$\hat{\mathbf{H}}_1 = K + fW(u) = K - fU_s w(\lambda d_s^{\delta} u) . \tag{65}$$

B. Solutions of $\hat{\mathbf{H}}_1$

We now show that the eigenfunctions of $\hat{\mathbf{H}}_1 \psi = \hat{E}\psi$ can be obtained from those of the reduced Eq. (30). From now on, we call the coupling parameter $\hat{\lambda}$ in order to help the comparison between the cases with and without fluctuations. From Eq. (63), we first note that f, being an average quantity, does not depend explicitly on $\mathbf{r}$. By making the transformations

$$\hat{E} = -U_s \hat{\varepsilon} \quad \text{and} \quad \boldsymbol{\rho} = \hat{\varepsilon}^{1/2}\mathbf{r}/d_s , \quad \text{so that} \quad K = -U_s \hat{\varepsilon} \nabla_{\rho}^2 , \tag{66}$$

and

$$\psi(\mathbf{r}) = \left[\frac{\hat{\varepsilon}}{f\hat{\lambda} d_s^{\delta}}\right]^{1/2} \phi(\boldsymbol{\rho}) , \tag{67}$$

it is easy to verify that an eigenfunctions ψ of $\hat{\mathbf{H}}_1$ corresponds to the $\phi = \phi_{\varepsilon}$ solution of Eq. (30), provided that

$$\hat{\varepsilon} = f\varepsilon . \tag{68}$$

The normalization of ψ now gives

$$\hat{\lambda} f^{\delta/2} = \varepsilon^{1-\delta/2} \langle \phi_{\varepsilon} | \phi_{\varepsilon} \rangle , \tag{69}$$

so that, from Eq. (36),

$$\hat{\lambda} = \lambda f^{-\delta/2} . \tag{70}$$

From Eqs. (32) and (33), we can compute $\langle v \rangle$ and $\langle w \rangle$ as functions of ε and φ, so that, using Eq. (63) f is given by

$$f = 1 + \frac{2}{1 + 2\tau^{-1}\langle v \rangle} = 1 + \frac{2}{1 + \tau^{-1}[\varepsilon\delta + \varphi(2-\delta)]} , \tag{71}$$

where $\tau=k_BT/U_s$ is the reduced temperature. By noting that

$$\langle\psi|K|\psi\rangle=-U_s(\hat{\varepsilon}-f\langle w\rangle)\ , \tag{72}$$

we obtain the free energy $\hat{F}$, Eq. (64), as

$$\hat{\varphi}=-\frac{\hat{F}}{U_s}=\varphi-\frac{\delta}{2}(f-1)(\varepsilon-\varphi)+\tau\ln[1+\tau^{-1}\{\varepsilon\delta+\varphi(2-\delta)\}]\ . \tag{73}$$

Equations (68), (70), (71), and (73) give the parameters $\hat{\varepsilon}$, $\hat{\lambda}$, and $\hat{\varphi}$ of a FMP in the presence of magnetic fluctuations in terms of the parameters ε, λ, and φ of the parent mean-field FMP state. In Figs. 3(a), 3(b), and 3(c), we plotted $\hat{\varphi}(\hat{\varepsilon})$ and $\hat{\lambda}(\hat{\varepsilon})$ for three space dimensions ($\delta=1$, 2, and 3) in the case of a reduced temperature $\tau=10^{-2}$. We have also plotted $\varphi(\varepsilon)$ and $\lambda(\varepsilon)$ (dotted lines) for comparison. We see that, in all cases, for a given value x of the reduced energy, $\hat{\varphi}(x)>\varphi(x)$ and $\hat{\lambda}(x)<\lambda(x)$. This means that the magnetic fluctuations always increase the FMP stability.

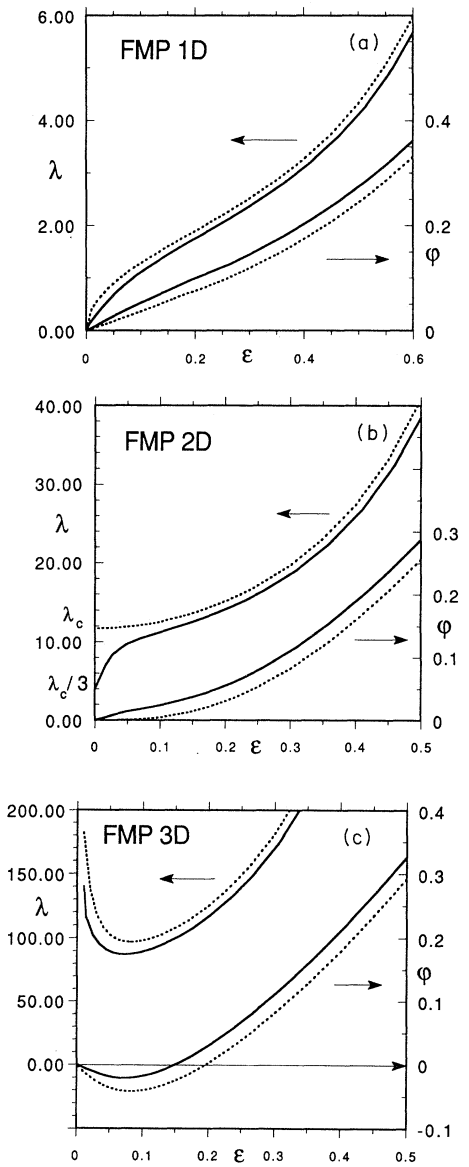

FIG. 3. $\hat{\lambda}$ and $\hat{\varphi}$ as a function of $\hat{\varepsilon}$ when magnetic fluctuations are included, for a reduced temperature $\tau=0.01$. The dotted curves give $\lambda(\varepsilon)$ and $\varphi(\varepsilon)$ in the mean-field approximation. (a) 1D and (b) 2D, with a threshold at $\lambda_c/3$. (c) 3D.

(a) For $T=0$, $f=1$, $\hat{\varepsilon}=\varepsilon$, and $\hat{\varphi}=\varphi$, we recover the results without fluctuations, as expected.

(b) For high-T, more precisely for small ε/τ, we have $f\approx 3$. If $\delta=1$ or 3, this leads to $\hat{\varepsilon}\approx 3\varepsilon$ and $\hat{\varphi}\approx 3\varphi$, so that the slope at the origin of the $\hat{\varphi}(\hat{\varepsilon})$ curve is not modified [even if it is hardly visible in Figs. 3(a) and 3(c)]. It is then easy to see that the spin fluctuations have little influence on FMP stability in 1D and 3D.

(i) In 1D, the FMP is already stable without fluctuations. Magnetic fluctuations transforms F into $9F$ in the $\lambda\to 0$ limit, and thus simply increase the FMP stability.

(ii) In 3D, the fluctuations have no significant influence on the parameters of *stable* FMP states, except for the height of the free-energy barrier, which is reduced since $F\to F/9$ in the large-λ limit.

The above changes of $\hat{\varepsilon}$ and $\hat{\varphi}$ simply correspond to transforming λ into $f\lambda$; this could be guessed directly from Eq. (65), since in the linear regime changing w into fw can be done by changing λ into $f\lambda$. (Let us stress that this simple transformation, useful in the linear regime, is different from the one given in Eqs. (66) and (67) which allowed us to find the solution outside the linear regime).

C. The 2D case

It is clear from Fig. 3(b) that the most important effect of spin fluctuations occurs in 2D. This is due to the fact that, in the $\varepsilon=0$ limit, a threshold at $\hat{\lambda}_c=\lambda_c/3$ is created, so that a domain of stability induced by spin fluctuations appears between $\lambda_c/3$ and λ_c. By developing Eqs. (68), (70), (71), and (73) for $\delta=2$ near the resulting threshold, we find that $\hat{\varepsilon}$ and $\hat{\varphi}$ behave as

$$\hat{\varepsilon}=\frac{9\tau}{4}(\hat{\lambda}/\hat{\lambda}_c-1),\quad \hat{\varphi}=\frac{9\tau}{8}(\hat{\lambda}/\hat{\lambda}_c-1)^2\ . \tag{74}$$

These dependences differ from those obtained in Sec. III B. The origin of this difference lies in the fact that $\hat{F}_s$, Eq. (61), is no longer an odd function of u.

Up to now, we considered T and λ as two independent parameters. Actually, for a given structure, λ is a function of T. We have shown in Sec. II B that λ is proportional to the magnetic susceptibility. For T larger than the spin glass temperature T_g, the magnetic susceptibility was found to follow a power law $\chi\simeq T^{-a}$, with $a=0.62$ characteristic of disordered magnetic systems.[11] This leads to

$$\lambda/\lambda_g=(T/T_g)^{-a}\ . \tag{75}$$

We restrict the following discussion to the $T>T_g$ domain. Adding relation (75) to Eqs. (68), (70), (71), and (73), we can obtain $\hat{\varepsilon}$ and $\hat{\varphi}$ as functions of T/T_g. Two

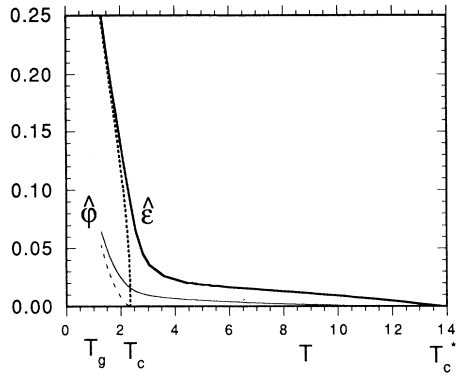

FIG. 4. Reduced energy $\hat{\varepsilon}$ and free energy $\hat{\varphi}$ for a 2D FMP in a magnetic quantum well as a function of T/T_g for $\lambda_g/\lambda_c=1.7$ and $U_s/k_BT_g=500$. The dotted curves show ε and φ in the mean-field approximation.

different cases may occur, depending on $\eta=\lambda_g/\lambda_c$.

For $\frac{1}{3}<\eta<1$, a weakly bound FMP induced by magnetic fluctuations is stable in the temperature range $T_g<T<T_c^*$, where $T_c^*/T_g=(3\eta)^{1/a}$. The polaron energy vanishes at T_c^*.

For $\eta>1$, we observe two regions of FMP stability (see Fig. 4).

(i) If $T_g<T<T_c$, where $T_c/T_g=\eta^{1/a}$, λ is larger than λ_c and a stable FMP already exists without fluctuations (see dotted curves) with a threshold at T_c.

(ii) If $T_c<T<T_c^*$, the FMP stability is induced by magnetic fluctuations. We see that, with our expression of $\hat{F}_s$ in Eq. (61), a smooth transition between the two regimes is obtained around T_c.

The parameters chosen in Fig. 4 correspond to a hypothetical $Cd_{0.9}Mn_{0.1}Te$ with a simple isotropic valence band (in order to have a Heisenberg carrier-ion interaction), confined in a quantum well of width 1.4 nm and a U_s value of about 50 meV.

Experimental verification of the existence of these FMP states induced by magnetic fluctuations might be difficult for at least two reasons: (a) We need a quasi-2D system with true Heisenberg carrier-ion interaction; this excludes zinc-blende materials. (b) The predicted FMP energy in this regime is small and could be masked by the weak localization induced by interface roughness, which also favors MP formation.[5]

V. CONCLUSION

In the FMP problem, we established a link between the free-energy minimization and the nonlinear Schrödinger equation whose eigenfunction of lowest energy is the *exact* carrier wave function. In the mean-field approximation, we have shown that the nonlinear potential of this Schrödinger equation is just the carrier-ion exchange interaction. Numerical solutions as a function of the polaron coupling parameter and space dimensionality suggest that the mean-field approximation is indeed valid in all cases where a sizable (hence measurable) FMP effect is predicted. We also show that, in the 2D case, magnetic fluctuations extend FMP stability toward a smaller coupling constant. Extension of these calculations in the presence of an external magnetic field should be quite easy.

However, we should keep in mind the distance between the present theoretical results and experimental data. Most of the latter involve exciton MP's, so a nontrivial extension of the theory to the exciton case is needed, even if we can argue that the hole takes the larger role in FMP formation, since it has a larger effective mass and a much larger exchange integral. In the 3D case, we have not taken into account the valence-band degeneracy in zinc-blende material; this causes a lot of uncertainty on the appropriate value of m^* to be used in the evaluation of λ_3 in Eq. (58). Another contribution to the carrier-ion exchange was neglected: the effect of a field gradient, here $\nabla(\psi^2)$, on small $j=0$ ion clusters.[12] Finally, the most important difference between our models and the experiment is the absence of any imperfections or defects. Existing data on MP's in bulk $Cd_{1-x}Mn_xTe$ already suggest the role of alloy density fluctuations as the primary cause of exciton localization, MP formation taking place later on.[13] In 2D structures, other imperfections related to the so-called interface roughness play an additional role in exciton localization, allowing MP formation in many different 2D heterostructures.[14] It is clear that any prediction of MP energy in real structures should worry about imperfections and disorder. Let us stress, however, that it is certainly useful to have a reliable theory of FMP stability in perfect systems.

APPENDIX A

The starting Hamiltonian for a carrier in a crystal containing N magnetic ions like Mn^{2+} is

$$\mathcal{H}=H_e(\mathbf{r})+\alpha\sum_{j=1}^{N}\mathbf{s}\cdot\mathbf{S}_j\delta(\mathbf{r}-\mathbf{R}_j)+\sum_{j>j'}J_{jj'}\mathbf{S}_j\cdot\mathbf{S}_{j'}\ , \tag{A1}$$

where $H_e(\mathbf{r})$ is the kinetic and potential energy of the carrier; the next term is the carrier-ion exchange interaction with an exchange integral α; $\mathbf{R}_j$'s are the positions of the magnetic ions, $\mathbf{S}_j$'s are their spins; and $\mathbf{s}$ is the carrier spin. The last term is the exchange spin-spin interaction.

Minimization of the free energy corresponding to the full Hamiltonian (A1) over the system wave function $\Psi(\mathbf{r},\sigma,\{\tau^j\})$ leads to the solution of FMP problem.

A first important assumption is to take the system wave function as a product

$$\Psi(\mathbf{r},\sigma,\{\tau^j\})=\psi_e(\mathbf{r})\chi_e(\sigma)\Psi_i(\{\tau^j\})\ , \tag{A2}$$

where $\psi_e(\mathbf{r})$ and $\chi_e(\sigma)$ are the carrier orbital and spin wave function, $\Psi_i(\{\tau^j\})$ being the wave function of the magnetic subsystem. This is justified if the carrier bandwidth is large with respect to $N_0\alpha$. Then we calculate the eigenvalues of (A1) in two stages:

(i) We average $\mathcal{H}$ over the carrier state

$$\tilde{\mathcal{H}}_i = \langle \psi_e \chi_e | \mathcal{H} | \chi_e \psi_e \rangle$$
$$= \langle \psi_e | H_e | \psi_e \rangle + \alpha \sum_{j=1}^{N} |\psi_e(\mathbf{R}_j)|^2 \mathbf{S}_j \cdot \langle \mathbf{s} \rangle + \sum_{j>j'} J_{jj'} \mathbf{S}_j \cdot \mathbf{S}_{j'} \ . \quad \text{(A3)}$$

We note that the second term of (A3) can be considered as an exchange field $\mathbf{H}^*$ acting on the ion spin $\mathbf{S}_j$:

$$\mathbf{H}^* = \alpha |\psi_e(\mathbf{R}_j)|^2 \langle \mathbf{s} \rangle / g\mu_B \ . \quad \text{(A4)}$$

Thus $\tilde{\mathcal{H}}_i$ does not depend on carrier variables $\mathbf{r}$ and $\mathbf{s}$; it depends on $\psi_e(\mathbf{R}_j)$ as parameters. So, if we can find the free energy $\mathcal{F}_i$ of the magnetic subsystem which is submitted to the effective field $\mathbf{H}^*$, the carrier wave function will minimize

$$\mathcal{F} = \langle H_e \rangle + \mathcal{F}_i \ . \quad \text{(A5)}$$

We then assume that the radius of the carrier wave functions is large enough to treat the magnetic medium as continuous, so that $\sum_j \ldots$ is changed into $\int \ldots n(\mathbf{r}) d^3\mathbf{r}$. This corresponds to neglecting the composition fluctuations and to considering the magnetic properties as local, depending on the average magnetic ion concentration $n(\mathbf{r})$.

In the last assumption, we neglect the carrier spin fluctuations. As a result, we can choose the z direction along $\langle \mathbf{s} \rangle$ (if the quantization axis is not imposed by other constrains). Then χ_e will be the eigenfunction of s_z and $|\langle s_z \rangle| = \frac{1}{2}$, so that

$$|H^*| = |\alpha/2| |\psi_e(\mathbf{r})|^2 / g\mu_B \ . \quad \text{(A6)}$$

H^* can thus be considered as a static nonhomogeneous magnetic field acting on the magnetic medium.

APPENDIX B

As ϕ_ε is solution of Eq. (30), the integration by parts of $\langle \phi_\varepsilon | \phi_\varepsilon \rangle$ for $1S$ states, i.e., for $\phi_\varepsilon(\rho) = \phi_\varepsilon(\rho)$, gives

$$\langle \phi_\varepsilon | \phi_\varepsilon \rangle = \Omega_\delta \int \rho^{\delta-1} d\rho \, \phi_\varepsilon^* \phi_\varepsilon = -\frac{\Omega_\delta}{\delta} \int \rho^\delta d\rho \left[\frac{d\phi_\varepsilon^*}{d\rho} \phi_\varepsilon + \text{c.c.} \right]$$
$$= -\frac{1}{\delta} \int d^\delta \rho \, \rho \left[\frac{d\phi_\varepsilon^*}{d\rho} \{ \nabla^2 \phi_\varepsilon + \phi_\varepsilon \varepsilon^{-1} w(\varepsilon \phi_\varepsilon \phi_\varepsilon^*) \} + \text{c.c.} \right] , \quad \text{(B1)}$$

where Ω_δ is the solid angle in the δ dimension. By making ∇^2 in the first term of (B1) explicit, we find

$$\int d^\delta \rho \, \rho \left[\frac{d\phi_\varepsilon}{d\rho} \nabla^2 \phi_\varepsilon + \text{c.c.} \right] = \int d^\delta \rho \, \rho \left[\frac{d\phi_\varepsilon^*}{d\rho} \frac{1}{\rho^{\delta-1}} \frac{d}{d\rho} \left[\rho^{\delta-1} \frac{d\phi_\varepsilon}{d\rho} \right] + \text{c.c.} \right]$$
$$= \Omega_\delta \int \rho^{\delta-1} d\rho \frac{\rho}{\rho^{2\delta-2}} \frac{d}{d\rho} \left[\left[\rho^{\delta-1} \frac{d\phi_\varepsilon}{d\rho} \right] \left[\rho^{\delta-1} \frac{d\phi_\varepsilon^*}{d\rho} \right] \right] .$$

An integration by parts then gives

$$-(\delta-2)\Omega_\delta \int \frac{d\rho}{\rho^{\delta-1}} \left[\rho^{2\delta-2} \frac{d\phi_\varepsilon^*}{d\rho} \frac{d\phi_\varepsilon}{d\rho} \right] = -(\delta-2) \int d^\delta \rho \frac{d\phi_\varepsilon^*}{d\rho} \frac{d\phi_\varepsilon}{d\rho} = -(\delta-2) \left\langle \frac{d\phi_\varepsilon}{d\rho} \middle| \frac{d\phi_\varepsilon}{d\rho} \right\rangle . \quad \text{(B2)}$$

In the second term of Eq. (B1), we replace $w(u)$ by $d(uv(u))/du$ and integrate once again by parts. We get (with $u = \varepsilon \phi_\varepsilon \phi_\varepsilon^*$)

$$\int d^\delta \rho \, \rho \left[\frac{d\phi_\varepsilon^*}{d\rho} \phi_\varepsilon + \text{c.c.} \right] w(u) = \Omega_\delta \int \rho^{\delta-1} d\rho \, \rho \frac{1}{\varepsilon} \frac{du}{d\varepsilon} \frac{d\{uv(u)\}}{du}$$
$$= -\delta \Omega_\delta \int \rho^{\delta-1} d\rho \frac{1}{\varepsilon} \{uv(u)\} = -\delta \langle \phi_\varepsilon | v(u) | \phi_\varepsilon \rangle \ . \quad \text{(B3)}$$

Using Eqs. (B2), (B3) with Eq. (B1), we find

$$\langle \phi_\varepsilon | \phi_\varepsilon \rangle = \frac{2-\delta}{\delta} \left\langle \frac{d\phi_\varepsilon}{d\rho} \middle| \frac{d\phi_\varepsilon}{d\rho} \right\rangle + \langle \phi_\varepsilon | \varepsilon^{-1} v(u) | \phi_\varepsilon \rangle \ . \quad \text{(B4)}$$

The first term of Eq. (B4) can be obtained by calculating $\langle \phi_\varepsilon | \nabla^2 | \phi_\varepsilon \rangle$ in two ways:

$$\langle \phi_\varepsilon | \nabla^2 | \phi_\varepsilon \rangle = \Omega_\delta \int \rho^{\delta-1} d\rho \, \phi_\varepsilon^* \frac{1}{\rho^{\delta-1}} \frac{d}{d\rho} \left[\rho^{\delta-1} \frac{d\phi_\varepsilon}{d\rho} \right]$$
$$= -\int d^\delta \rho \frac{d\phi_\varepsilon^*}{d\rho} \frac{d\phi_\varepsilon}{d\rho} = -\left\langle \frac{d\phi_\varepsilon}{d\rho} \middle| \frac{d\phi_\varepsilon}{d\rho} \right\rangle . \quad \text{(B5)}$$

Using Eq. (30) verified by ϕ_ε, we also find

$$\langle\phi_\varepsilon|\nabla^2|\phi_\varepsilon\rangle=\langle\phi_\varepsilon|\phi_\varepsilon\rangle-\varepsilon^{-1}\langle\phi_\varepsilon|w(u)|\phi_\varepsilon\rangle\ , \tag{B6}$$

so that Eq. (B4) finally reads

$$\langle\phi_\varepsilon|\phi_\varepsilon\rangle=\frac{2-\delta}{\delta}[-\langle\phi_\varepsilon|\phi_\varepsilon\rangle+\varepsilon^{-1}\langle\phi_\varepsilon|w(u)|\phi_\varepsilon\rangle]+\varepsilon^{-1}\langle\phi_\varepsilon|v(u)|\phi_\varepsilon\rangle\ , \tag{B7}$$

which is nothing but Eq. (32).

Writing Eq. (10) in reduced units, we get

$$\varphi-\varepsilon=\langle\phi_\varepsilon|v(u)-w(u)|\phi_\varepsilon\rangle/\langle\phi_\varepsilon|\phi_\varepsilon\rangle\ . \tag{B8}$$

Replacing ε by its value from Eq. (B7), we find Eq. (33).

APPENDIX C

Let us consider the expression

$$\zeta(\varepsilon,\mu)=-\langle\psi_\varepsilon|K/U_s+v(\mu d_s^3\psi_\varepsilon\psi_\varepsilon^*)|\psi_\varepsilon\rangle\ ,$$

where ψ_ε, given by Eq. (26), is the eigenfunction for the reduced energy ε, the coupling constant being $\lambda(\varepsilon)$. We know that $\zeta(\varepsilon,\mu)$ should be an extremum for ε such that $\lambda(\varepsilon)=\mu$:

$$\partial\zeta(\varepsilon,\mu)/\partial\varepsilon=0 \quad \text{when } \mu=\lambda(\varepsilon)\ . \tag{C1}$$

Then $\zeta(\varepsilon,\lambda(\varepsilon))$ is nothing but the reduced free energy $\varphi(\varepsilon)$. We thus have

$$\frac{d\varphi}{d\varepsilon}=\left[\frac{\partial\zeta}{\partial\varepsilon}+\frac{\partial\zeta}{\partial\mu}\frac{d\lambda}{d\varepsilon}\right]_{\mu=\lambda(\varepsilon)}\ . \tag{C2}$$

By noting that

$$\partial\zeta(\varepsilon,\mu)/\partial\mu=\langle\psi_\varepsilon|d_s^3\psi_\varepsilon^2v'(\mu d_s^3\psi_\varepsilon^2)|\psi_\varepsilon\rangle\geq 0\ , \tag{C3}$$

we deduce from Eqs. (C1) and (C2) that $d\varphi/d\varepsilon=0$ imposes $d\lambda/d\varepsilon=0$. This proves that the minimum of $\varphi(\lambda)$ take place at λ^*, where the slope of $\varepsilon(\lambda)$ is infinite. By calculating

$$\frac{d\varphi}{d\lambda}=\left[\frac{\partial\zeta}{\partial\mu}+\frac{d\varepsilon}{d\lambda}\frac{\partial\zeta}{\partial\varepsilon}\right]_{\mu=\lambda(\varepsilon)}\ , \tag{C4}$$

we see that [except exactly for $\lambda=\lambda^*$, where the second term of (C4) is undetermined], the slope of $\varphi(\lambda)$, which reduces to $(\partial\zeta/\partial\mu)|_{\mu=\lambda(\varepsilon)}$, is always positive as shown in Eq. (C3), and tends toward $(\partial\zeta/\partial\mu)|_{\mu=\lambda^*,\varepsilon=\varepsilon^*}$ on its two branches. Hence the $\varphi(\lambda)$ curve exhibits a cusp at λ^*.

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